Utilization of Thorium in Power Reactors
UTILIZATION OF THORIUM IN POWER REACTORS
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REPUBLIC  REPUBLICS
UNION OF SOVIET SOCIALIST
REPUBLICS  UNITED ARAB REPUBLIC
UNITED KINGDOM OF GREAT
IRELAND  UNITED STATES OF AMERICA
VENEZUELA  VIET-NAM
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The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York, it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

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UTILIZATION OF THORIUM IN POWER REACTORS
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Thorium is receiving increasing attention as an important fertile material for the expanding nuclear power programmes around the world. The known reserves of low-cost thorium exceed those of uranium. The superior nuclear and physical properties of thorium-based fuels could lead to very low fuel-cycle costs and make thorium reactors economically attractive. In addition, the use of thorium in advanced converters and breeders would permit more efficient utilization of low-cost uranium reserves.

Already 12 reactors using thorium have been built; these include two commercial plants and a number of experimental facilities. Research and development work on the element is going on in nine countries, and soon seven more countries will be added to this number.

The International Atomic Energy Agency convened a Panel on the Utilization of Thorium in Power Reactors from 14 to 18 June 1965 in Vienna. Forty-five scientists from 14 countries and two international organizations took part in the Panel, which was asked to give an appraisal of the current status of thorium technology and make recommendations on a possible Agency programme of activities.

The proceedings of the Panel, which includes 23 survey papers and brief reviews on important aspects of thorium utilization prepared by well-known experts who acted as rapporteurs, is being published in response to keen interest. The papers and reviews, which stress the importance of utilizing thorium, deal especially with existing programmes in Member States, estimated thorium reserves; the status of work on thorium reactor physics, basic properties, fuel cycle technology, and promising reactor concepts, and the overall potential for thorium utilization as seen from the near-term and long term aspects. It is hoped that the publication will be of interest to engineers and scientists engaged in research and development work connected with the element.

The Agency is grateful to the authors of the survey papers, to the rapporteurs for preparing reviews, to all the participants for contributing to the discussions, and to the chairman for guiding the work of the Panel.
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SUMMARY REPORT AND RECOMMENDATIONS

I. TERMS OF REFERENCE

The terms of reference of the panel were as follows:

(a) To review the available information on various aspects of thorium utili-
zation, such as physics of thorium-bearing systems, fabrication and
reprocessing techniques for thorium fuels and their behaviour under
irradiation and reactor environments, economics of thorium fuel cycles
and the potential of various reactor concepts using thorium. While pro-
viding an appraisal of the present status of thorium technology for power
reactors, the panel would also indicate major gaps which remain to be
filled by further research and development in various areas.

(b) To indicate any possibilities for international collaboration in research
and development work relating to thorium utilization.

(c) To make a recommendation to the Director General regarding a possible
Agency programme of activities which should serve to promote the utili-
zation of thorium in power reactors as a part of the Agency's overall
programme in nuclear power.

II. SOME REASONS FOR THE IMPORTANCE OF UTILIZING THORIUM

Current interest in thorium reactors throughout the world stems largely
from the fact that the $^{233}\text{U}$ produced by neutron capture in thorium is a more
valuable fuel for thermal reactors than the plutonium that results from cap-
ture in $^{238}\text{U}$. Because of the high net neutron yield (eta) of $^{233}\text{U}$ in thermal
reactors relative to either $^{239}\text{Pu}$ or $^{235}\text{U}$, the thorium-$^{233}\text{U}$ fuel system has
higher conversion ratios and longer fuel life than is possible with $^{238}\text{U}$-based
fuel systems. Thorium reactors, therefore, are expected for long-term
purposes to have lower fuel cycle costs than many uranium converters using
natural or slightly enriched uranium or uranium with recycled plutonium.

Thorium reactors have an additional advantage in that in some of the
advanced thorium converters it may be possible to breed $^{233}\text{U}$ sufficiently
to achieve a self-sustaining recycle system whereby no supplementary input
of enriched material would be required. The establishment of thorium-
based power systems may make several countries with small domestic
sources of fissile material self-sufficient with regard to importation of
nuclear fuels.

The favourable neutronic properties of $^{233}\text{U}$ not only help in reducing
fuel cycle costs but also lead to more efficient utilization of low-cost uranium
reserves which necessarily form the basis of all nuclear power systems that
exploit only fission chain reactions. Thus the introduction of thorium con-
verters into nuclear power systems can delay the exhaustion of low-cost ore
and at least provide any time required for the development of commercially
competitive breeders.

In the long run, thermal breeders using thorium are also an attractive
alternative to the fast breeder line of development, primarily because of
the smaller amount of fissile inventory required per megawatt of electrical
capacity. This lower inventory makes it possible to establish a self-sustaining breeder industry (i.e. one which produces all of its fissile requirements) with a considerably smaller investment in mined uranium than is possible with fast breeders. Corresponding fuel investment costs are also lower and power costs less sensitive to the market for bred fuel. In addition, extensive resources of thorium exist in many countries, which are adequate to support large-scale nuclear power systems for many centuries. Because of these many advantages, the advanced thorium-converter thermal-breeder line of development represents an attractive approach to competitive long-term nuclear power.

III. EXISTING PROGRAMMES IN MEMBER STATES

The members of the panel gave a short review of the overall programmes in their respective countries relating to the utilization of thorium. The following summaries of the national activities have been prepared by the respective panel members concerned. The figures relating to thorium reserves in various countries were compiled by the rapporteurs for this topic, namely Dr. El Shazly and Professor Santos, on the basis of the information given at the panel and from available literature.

AUSTRALIA

The Australian Atomic Energy Commission believes that better utilization of nuclear raw materials and economic power costs can be achieved in a high-temperature gas-cooled reactor based on the thorium cycle. Both the $^{233}\text{U}-^{232}\text{Th}$ and plutonium-thorium cycles have been considered. The first cycle was preferred but the plutonium-thorium received more attention especially as it is found to be important for a small gas-cooled reactor. Studies have been carried out and presented which are related to homogeneous dispersions of thorium in beryllia assemblies fuelled by the three main fissile elements as aluminium alloys.

BRAZIL

Brazilian interest in thorium utilization stems primarily from its great reserves of thorium. At present the Brazilian programme on thorium includes:
(a) Intensification of efforts concerning assessment of thorium reserves
(b) Studies of thorium reactor concepts, starting from feasibility studies and comparative appraisals of various types, and leading to a choice of one of them for development
(c) Studies and experimental research on fabrication and reprocessing of thorium fuels
(d) Work on experimental devices for thorium reactors research.

Two types of heavy-water reactors using natural uranium and rather conventional technology have been considered. Parametric studies of a projected integrated consumption of nuclear fuels were made comparing dif-
ferent fuel cycles to minimize the consumption of uranium. A 12-month programme has been set up, including feasibility studies and laboratory research on fabrication and reprocessing of thorium fuel elements. Use of experimental facilities inside the country together with outside co-operation is being considered.

CANADA

Canada's interest in the use of thorium is based on the expectation that thorium-based fuels provide power at costs lower than those provided by natural uranium fuel in D$_2$O reactors when ore prices rise. According to Canadian authorities such D$_2$O reactors are expected to set the level of economic competition that would have to be met by any other reactors including breeders of any kind. By developing the thorium cycle in such neutron economical reactors any world shortage of uranium at an acceptable cost would be postponed for many centuries and effectively indefinitely as other scientific advances in nuclear processes are probable.

A reactor physics programme to study thorium fuels has been started at Chalk River. A programme of chemical and mass spectrometer measurements on irradiated thorium samples will be continued. Substitution experiments using $^{235}$U-$^{232}$Th fuel bundles are planned before the end of the year. In the meantime analytical studies will be pursued to assess the importance of thorium to the Canadian nuclear power programme. A preliminary survey indicates that the effect of thorium-based fuel on reactor design and capital costs may be appreciable and it is important that changes in design parameters be appreciated quickly so that reactors designed today can use thorium if it becomes economic during the latter part of their life.

Th$_2$O$_2$-UO$_2$ specimens have been irradiated with central melting to determine the effect of UO$_2$ content on the thermal conductivity. Others are being irradiated at power outputs up to about 750 W/cm of fuel to compare the sheath expansion and release of fission-product gases for stoichiometric Th$_2$O$_2$-2.2% UO$_2$ with those of pure UO$_2$. Initial results indicate that this composition has a slightly better performance than pure UO$_2$, generating the same power per unit length. The possibility of high burn-up irradiations (> 30 000 MWd/t of fuel) is being considered and a determination of the effective temperature of plasticity for Th$_2$O$_2$-UO$_2$ under irradiation may be made.

FRANCE

The Commissariat à l'énergie atomique has explored and exploited thorium deposits at Madagascar, where important reserves exist, especially in the form of uranothorite deposits. This led to establishing treatment and separation methods for these ores.

Some thorium slugs have been irradiated in Marcoule reactors for experimental purposes. Methods of extracting $^{233}$U from the irradiated thorium have been studied.

Although at present France has no short-term programme for thorium utilization in power reactors, this possibility is being considered with
interest. France has made a considerable effort to develop heavy water reactors as a second type of power reactor in its programme. Use of the thorium cycle may increase substantially its possibilities from the point of view of the utilization of natural uranium reserves. This may be achieved without developing a new type of reactor and by taking advantage of a technology already established, as soon as the need for better utilization of fuels is felt. At present, however, natural uranium cycles are simpler and more economic. Besides, there are still technological problems and uncertainties regarding the use of thorium.

Other concepts of interest for France would be the homogeneous and semi-homogeneous reactors (HTGR and fused-salt reactors).

FEDERAL REPUBLIC OF GERMANY

The gas-cooled pebble-bed converter reactor concept was taken up actively in the Federal Republic of Germany. This will use thorium as a fertile material. The first prototype is under construction, while development and research work is being carried out to prepare a project for a 300-MW(e) reactor of this type by an association (THTR) between EURATOM, BBK (Brown-Boveri, Krupp) and KFA (Kernforschungsanlage, Julich), which is financially supported by the Federal Government.

A theoretical study carried out by KFA on the evaluation of different thorium breeder systems indicated that the gas-cooled high-temperature reactors, the heavy-water-moderated reactors and molten-salt reactors are particularly promising, and a two and a half year programme has been started for experimental and theoretical studies on these systems. Furthermore, evaluation of the thorium cycle in a pressurized heavy-water 400 MW(e) reactor has been carried out since 1960 by Siemens-Schuckertwerke. A programme for calculation and experimental investigation of ThO₂-UO₂ lattices has been started.

In the field of fuel, thorium metal has been manufactured for the Karlsruhe FR-2 research reactor, and a production line for coated particles containing uranium and thorium carbides or oxides for gas-cooled high-temperature reactors is under construction. Finally, investigations are being carried out for the fabrication of UO₂-ThO₂ elements for heavy-water reactors.

INDIA

Monazite occurs in several areas in India. The processing of monazite to produce ThO₂ and thorium metal in the form of rods and pellets has been carried out on an industrial scale for the last few years. Some fifty thoria rods have been irradiated in the Canada-India reactor and are almost ready for processing.

Under an agreement of collaboration between India and Sweden, pellets of ThO₂ clad in aluminium will be supplied to the AB Atomenergi of Sweden. Joint experiments supported by theoretical studies are planned to investi-
gate the physics of $D_2O$-moderated reactors using thorium and mixed lattices of Pu or $^{235}U$-enriched $UO_2$ and $ThO_2$.

India is considering both thermal and fast thorium breeder concepts. Fast breeders can be used as a source of $^{233}U$ by diverting the surplus neutrons to $^{233}U$ production. Various alternatives are possible:

(a) To use fast reactors as plutonium burners to produce $^{233}U$
(b) To design fast reactors so as to be self-sufficient in plutonium and divert the excess neutrons to $^{233}U$ production. This, coupled with a good thorium thermal converter or a breeder of high specific power, will considerably reduce the consumption of uranium.

ITALY

The Italian programme on the thorium-uranium fuel cycle began in 1960 and there is a trend at present for the re-evaluation of the thorium-uranium cycle. The use of this cycle in Italian reactors is only a possibility at present, although it is believed to be more realistic to utilize the PCUT (Programme Ciclo Uranio Thorio) facility to rework thorium-uranium as oxide fuel from water reactors and carbide fuels from high-temperature gas-cooled reactors for foreign customers interested in this cycle.

Work carried out includes the modified sol-gel process for thorium-uranium production, and the head-end processing of carbide fuels by electrolytic disintegration and carbide particle fabrication by the sol-gel process under a contract connected to the OECD (Organisation for Economic Co-operation and Development) Dragon Project. Theoretical work has been done to evaluate the possible utilization of thorium in the existing water reactors and in the fog-cooled reactor.

JAPAN

Among reactor designs under study in Japan, particular interest is exhibited in advanced converters with high conversion ratios and potential for cheap energy cost. The evaluation of thorium behaviour in different types of advanced converters will be carried out during 1965 by the Japan Atomic Energy Research Institute while at the same time research on thorium fuel is being re-evaluated, to consider the possibility of utilizing thorium in advanced converters.

Research and development work on an aqueous homogeneous reactor and a semi-homogeneous reactor have been made for several years since 1957. Research has been, or is being, conducted on reactor-grade high-purity thorium ceramics, on trace-element determination in thorium and on the measurement of $\sigma_{\alpha}(E)$ of thorium in semi-homogeneous reactor critical assembly experiments. Reactor physics experiments will be made in TCA (light-water-moderated critical assembly) by inserting thirty thorium fuel rods to obtain design data on thorium utilization in the JPDR (Japan Power Demonstration Reactor), a boiling-water-type reactor.
NETHERLANDS

A part of the Netherlands nuclear energy programme is devoted to the thorium cycle. In this respect the possibilities of thermal breeders with the Th-233U cycle are being studied. The heavy-water aqueous homogeneous reactor is believed to be one of the best possibilities; suspensions for such a reactor are currently being investigated. A test facility called the KSTR (Kema Suspension Test Reactor) is under construction, which is not a prototype reactor but a research instrument for testing the suspensions at different power levels.

The suspension particles have been developed for the first time by using the sol-gel process. Research is being extended from oxides to carbides and from small particles to large particles for the high-temperature gas-cooled reactor (HTGR).

SWEDEN

A research programme has been started in co-operation with the Atomic Energy Establishment of India to investigate the potential of the Th-233U fuel cycle in the PHWR.

This programme includes development of calculation methods for the Th-233U fuel cycle, measurements to check the calculation methods and finally calculations pertaining to different schemes for using thorium in a PHWR.

According to the Indian-Swedish collaboration agreement, India is supplying 550 kg of ThO₂ fuel rods for measurements in the Studsvik zero-energy facilities. The thoria rods will be used together with rods of natural uranium, and 1.2 and 1.8% enriched uranium. By the end of 1965 a few 2% Pu-enriched UO₂ rods will also be available. In these experiments buckling, thermal neutron activations and conversion ratios will be studied.

UNITED ARAB REPUBLIC

Evaluation of the thorium resources of the United Arab Republic are being carried out. The latest work has proved the existence of large resources of ThO₂ contained especially in the placer deposits of the Nile origin. The raw sands are industrially treated to produce mineral concentrates and plans are under way to increase this production substantially.

As a preliminary step towards the utilization of thorium, pilot plants have been erected to produce monazite of 97-98% purity and thorium-uranium cake. The separation of thorium and uranium from the thorium-uranium cake produced has been initiated, utilizing an anion exchange process. It is planned to cover the range of the chemical and metallurgical treatment of thorium, and to test the use of thorium along with uranium in reactor programmes.
UNITED KINGDOM

Studies have been made of the use of thorium instead of fertile $^{238}$U in both slightly enriched thermal reactors and in fast reactors, and it seems that at present there is no advantage in using thorium in the reactors currently under development or in operation in the United Kingdom. The United Kingdom, however, sees a possible interest in thorium if a competitive thermal reactor can be developed.

The factors favouring such a step would be a lower capital cost and/or a lower fissile material inventory for a thermal reactor using thorium as compared to a fast reactor utilizing uranium. The high-temperature reactor is one type for which the thorium cycle might have special advantages, and the United Kingdom is supporting the development of this reactor type through the OECD Dragon Project.

UNION OF SOVIET SOCIALIST REPUBLICS

Research and design work on breeder reactors as well as development and construction of pilot and industrial atomic power stations using such reactors constitute major and immediate tasks in the field of atomic power in the USSR. A more rapid transition to the preferential use of fast-neutron power reactors is envisaged and a uranium-plutonium fuel cycle is being adopted as the initial basis for the development of these reactors.

The introduction of thorium in the fast-reactor fuel cycle without prejudicing the rate of power increase of atomic power stations is possible only when enough $^{239}$Pu has been accumulated in the uranium-plutonium cycle to supply fast reactors operating with a mixed cycle. The potential power doubling time of reactors operating with a mixed cycle approaches the doubling time of plutonium breeders. Research is being carried out on all these questions.

At present the fast reactor BN-350 with a power of 300-350 MW(e) is being built in the USSR and a 600 MW(e) reactor is being designed. A study for a 1000 MW(e) fast reactor with a breeding ratio of 1.6 to 1.7 is under way. The concept of a fast-neutron converter-breeder reactor using thorium and operating at various power levels is under study.

UNITED STATES OF AMERICA

The development programme in the USA for the economic use of thorium as a nuclear fuel spans the whole field of reactor technology. The programme, which is designed to put into operation several reactors which use thorium, is now being supplemented by a considerable effort devoted to the problems of fuel recycle. The apparent success of uranium-fuelled water reactors in achieving near-term economic goals allows greater emphasis in the thorium programme on achieving the longer-term goal of greater fuel utilization in thermal reactors which are near-breeders and breeders.
The high-temperature gas-cooled reactor and the basic seed-blanket reactor have been designed to achieve good fuel utilization using thorium. Furthermore, studies are under way on the use of thorium in a heavy-water-moderated organic-cooled reactor, and it is to be used in the molten-salt reactor experiment, which recently went critical at Oak Ridge.

Research and development work in the field of fuel materials for the thorium reactor systems is notably concerned with the development of economic methods for reprocessing the irradiated fuel and refabricating the fertile and fissionable materials into fuel elements suitable for pre-use in reactors. The thorium utilization programme at the Oak Ridge National Laboratory is expected to furnish many of the technical answers needed and to provide a focal point for the technical planning of an integrated programme for the development of the thorium fuel cycle.

EURATOM

Two approaches are being followed. The first consists of agreements such as Dragon and THTR, and is based on the high-temperature gas-cooled reactor concept. This type of reactor is particularly suited for thorium fuel. The second approach is based on the heavy-water-moderated organic-cooled reactor project (ORGEL) at C. C. A. Ispra, where thorium has been considered as potential fuel.

IV. THORIUM RESERVES

The world thorium reserves are given in Table I.

V. TECHNICAL STATUS OF RESEARCH AND FUTURE WORK

A. REACTOR PHYSICS*

Nuclear data

The panel reviewed the state of the nuclear data relevant to the thorium fuel cycle in thermal reactors. The fast-neutron data were not specifically discussed, although it was recognized that considerable uncertainties exist in certain areas.

Since thorium-based systems require a feed of fissile material, it is necessary to include all fissile nuclides, namely $^{235}$U, $^{235}$Pu, $^{239}$Pu and $^{241}$Pu, in the review. The main points emerging from discussions on the nuclear data are as follows:

1. The resonance parameters of $^{232}$Th are apparently adequately determined as a result of recent studies but further comparisons with experiments are needed for general verification.

* The rapporteurs for this topic were Messrs W. Gemmell, S. Jaye and J. Sanders.
The largest uncertainty in the conversion potential of the thorium cycle arises from uncertainty in the energy variation of the neutron yield (eta) of $^{233}$U, particularly in the resonance region from $1.0$ to $10^3$ eV.

While data on $^{233}$Pa, $^{234}$U, $^{235}$U and $^{239}$Pu are satisfactory for reactor assessments, better information on the resonance parameters is still required for detailed calculations.

The capture cross-section of $^{237}$Np is of importance in cases where $^{235}$U is recycled. "Segregated fuel" management schemes will eventually remove $^{236}$U and $^{237}$Np before saturation however, reducing their importance.

Cross-section data for $^{240}$Pu are satisfactory but some integral experiments are desirable to check the analytical treatment of the capture in this isotope in homogeneous and heterogeneous systems.

Data on the neutron yield (eta) of $^{241}$Pu as a function of energy from thermal up to $10^3$ eV is required, since events in this isotope are important in Pu-Th cycles. Better data on $^{242}$Pu are also desirable.

Further experimental verification of data on fission-product cross-sections and yields is needed, particularly for the large burn-ups (i.e. on fission per initial fissile atom and upwards) generally aimed at in the thorium cycle.

$^{232}$U build-up is important for the evaluation of chemical processing and refabrication methods, and better data on the $(n, 2n)$ cross-section of thorium and on the capture cross-sections of $^{231}$Pa and $^{232}$U may be needed.

Use of thorium in specific reactor types

The panel considered the physics of thorium fuels in the various reactor systems. It was recognized that it was not possible to divorce the discussion of the physics from the fuel cycle characteristics and economics. With this in mind the major physics considerations pertaining to the thorium cycle were identified as follows.

1. D$_2$O-moderated solid-fuel reactors

Very few lattice measurements are available to support the methods of calculation for thorium lattices, and the recipes developed for natural and low-enrichment uranium are used without direct verification, but with some assurance. If the thorium fuel cycle is pursued beyond the survey calculations presented at the panel, a number of well-defined integral experiments should be performed and analysed in detail using basic calculational methods.

The following general remarks were made on the substitution of thorium for $^{238}$U in the D$_2$O-moderated reactor.

(a) Because of the high value of eta for $^{233}$U, coupled to the potentially excellent neutron economy of D$_2$O-moderated reactors, breeding can be achieved in this system.

(b) The higher thermal capture cross-section of thorium leads to relatively higher fissile enrichment than natural uranium. However, the smaller
<table>
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resonance integral of thorium and the relative insensitivity of \( \eta \) of \( ^{233}\text{U} \) to the spectrum permits the use of a tighter lattice.

(c) As a consequence of the relatively high fuel inventory, high fuel ratings are desirable, despite resulting increase of neutron capture in \( ^{233}\text{Pa} \). Fuel elements suitable for high ratings may involve increased fabrication cost.

(d) While in near-natural uranium fuel the macroscopic cross-section change with irradiation is rather small, this may not be so with plutonium-thorium systems and consequently this consideration is likely to have implications on power peaking and fuel management schemes.

(e) Temperature coefficients in plutonium-thorium fuel will require the same detailed considerations as in the plutonium-uranium systems.

2. The fast-breeder reactor

The thorium fuel cycle and mixed fuel cycles can be used in a fast-breeder reactor and conversion ratios as high as 1.3 may be achieved. Since the fast fission cross-section in thorium is quite small and has a high threshold, the use of thorium rather than \( ^{238}\text{U} \) results in a negative sodium void coefficient in large cores. However, a negative void coefficient can also be attained in a fast reactor with \( ^{238}\text{U} \) either by degrading the spectrum or by using a high-leakage configuration or a gaseous coolant. It seems probable that \( ^{238}\text{U} \) can be used as safely as thorium and, with its superior neutron economy, might remain as the preferred fertile material in fast systems.

3. Graphite-moderated reactors

The thorium fuel cycle can lead to breeding in either a fluid fuel system or a solid fuel system. In either case the gaseous fission products may be stripped continuously from the fuel. Both systems can also meet the goal of a low fuel investment. Thus the thorium fuel cycle is well adapted to these reactors because of their excellent neutron economy and the dispersion of the fuel in the moderator, which allows very high specific powers. In the near term these reactors will probably be operated as converters, but the essential physics characteristics are similar to those of breeders. Extensive calculational methods are available for analysing these semi-homogeneous reactors, and satisfactory results can be achieved for reactor evaluations. However, further development is required for detailed design.

4. Aqueous-homogeneous reactor

Use of the thorium cycle in such systems as the high-specific-power \( \text{D}_2\text{O}-\text{suspension reactor} \) can lead to breeding. Fission products may be stripped continuously from the circulating fuel. There appear to be few reactor physics problems specific to this concept that cannot be adequately treated by existing calculational methods.
Continuing programmes

While noting the very much smaller amount of lattice data pertinent to the thorium cycle, no additional calculational problems have been identified that are not also found in uranium-plutonium systems. Provided, therefore, that the differential nuclear data are of satisfactory precision, the accuracy achieved in calculations should be similar to that in the uranium-plutonium cases on which there is now much experimental evidence available.

However, importance is attached to direct verification of conversion ratios in Th-\(^{233}\)U assemblies, and to the analysis of isotopic compositions of fuel irradiated to high burn-up in well-defined spectrum conditions.

The following experiments and plans were mentioned at the panel:

1. Swedish lattice studies on ThO\(_2\) rods, together with \(^{235}\)U and Pu-enriched rods;
2. Indian plants for the production of 1 kg \(^{233}\)U for experimental use;
3. Australian work on homogeneous Th-BeO assemblies with \(^{233}\)U, \(^{235}\)U and \(^{239}\)Pu fuels;
4. United Kingdom studies on near-homogeneous \(^{239}\)Pu-graphite assemblies;
5. German plans for measurement of integral parameters in D\(_2\)O-moderated ThO\(_2\) lattices;
6. Brookhaven (\(^{233}\)U-Th)O\(_2\) lattice experiments;
7. Canadian reactor physics programme on thorium fuels at Chalk River;
8. Japanese work on thorium reactor physics data, such as \(\sigma_a(E)\) with the semi-homogeneous critical assembly at JAERI.
9. United States is producing several hundred kilograms of \(^{233}\)U for critical experiments at General Atomics in support of HTGR and at Bettis Atomic Power Laboratory in support of the seed-blanket reactor.

Recommended programme for the IAEA

1. As part of its activities in the compilation of nuclear data on an international level, the IAEA should note that the following nuclides are of particular interest for the thorium cycle: \(^{231}\)Pa, \(^{233}\)Pa, \(^{232}\)U, \(^{233}\)U, \(^{237}\)Np, \(^{241}\)Pu and \(^{242}\)Pu, fission products and yields, both primary and secondary.
2. The Agency should compile data on thorium lattice experiments for use in testing calculational methods.
3. The Agency should support basic types of integral experiments (e.g. uniform dispersion) as verification of \(^{233}\)U data, by making \(^{233}\)U available to experimenters and by awarding research contracts to appropriate laboratories.
4. In view of the importance of obtaining isotopic analysis and reactivity measurements on very high burn-up Th-\(^{233}\)U under well-defined spectrum conditions, the Agency could support such work under its research contracts programme.
B. BASIC PROPERTIES OF THORIUM, AND OUT-OF-PILE AND IN-PILE PERFORMANCE OF THORIUM FUEL*

Irradiation behaviour of thorium-uranium alloys and compounds

The irradiation stability of the metal-clad oxide (ThO₂-UO₂) fuel system has been demonstrated. It is noted that the Indian Point Reactor has approximately 350 full power days of operation with no indication of fuel malfunctions. A fuel programme on metal-clad, bulk oxides is under way at ORNL and is concerned mainly with the evaluation of sol-gel derived ThO₂-UO₂ fuels and vibratory compaction for fuel rod fabrication. Experimental rods up to 36 in. in length fabricated in this manner have compared favourably with vibratory compacted rods containing arc-fused ThO₂-UO₂ and with rods containing pressed and sintered ThO₂-UO₂ pellet fuels. Burn-ups in excess of 80 000 MWD/t (Th+U) have been achieved at linear heat ratings up to 300-400 W/cm. Some tests have been run as high as 1000 W/cm to 22 000 MWD/t (Th+U) burn-ups with no evidence of swelling or central melting. Preliminary comparisons with similar UO₂ fuel rods indicate that higher power ratings are required to produce similar microstructural changes in the ThO₂-5% UO₂ fuels.

Concerning fuels for high-temperature gas-cooled reactors, small particles coated with pyrolytic carbon have been irradiated in loose beds and in graphite matrix elements. Spherical particles, with multi-layer coatings, behaved satisfactorily. The ratio of release to production rate (R/B) for ⁸⁸Kr of 10⁻⁵ to 10⁻⁷ has been achieved in loose bed tests for burn-ups over 200 000 MWD/t (Th+U). Coated particles of (Th, U)C₂ and (Th, U)O₂ compare favourably with similar UC₂ and UO₂ particles. The oxide particles are compatible with the intact pyrolytic carbon coatings but the data must be considered as preliminary.

A review of the limited experiments to date on thorium alloys irradiations indicated that such isotropic fuels are not subject to the "growth" phenomena of anisotropic uranium and that they do not exhibit the "cavitation swelling" reported for uranium metal fuels in the 400 to 600°C temperature range.

Concerning fuel for high-temperature gas-cooled reactors, there exists a reasonably well-established technology for the production of carbide and oxide microspheres. These are coated with pyrocarbon and/or silicon carbide for the purpose of controlling fission product release. These particles have successfully withstood irradiations at 1400°C and irradiations are now in progress at 1600°C. It would be possible to increase the conversion ratio of high-temperature gas-cooled reactors by disposing of (releasing) the fission products. This release seems to be more feasible for gaseous fission products than for metallic ones.

Properties of thorium, its alloys and its compounds

A preliminary analysis of creep data, high-temperature strength, and hot hardness data suggests that at temperatures above about 500°C thorium

* The rapporteurs for this topic were Messrs A. Boettcher, D. V. Ragone, and G. Schileo.
is significantly stronger than uranium. Since thorium, with its isotropic structure, may not be subject to such internal stresses present in irradiated uranium, its properties may not be degraded to the same extent in the temperature range of 400-550°C. However, the effect on creep strength of fission events during irradiation of thorium has yet to be established.

The data on physical properties of thorium oxide are quite extensive. The available data on mechanical properties are misleading, however, since these properties depend greatly on the fabrication of the specimen. Data on diffusion in thoria are completely lacking. Compatibility with many materials is excellent, either known from experiment or safely predictable. However, information on thoria containing uranium is very meagre. Despite the regular isomorphous replacement, the disorder introduced precludes prediction of properties by interpolation between those of the components; the scanty data on thermal conductivity prove this.

In contrast is the situation on the carbides, which are of interest only for application in nuclear energy. Information on these compounds is quite scanty but the rate at which new knowledge is being reported is quite high. Reflecting the nuclear interest, the information on mixed carbides with uranium is keeping pace with or even exceeding that on pure thorium carbides.

The compounds ThBe$_{13}$, ThB$_4$, ThN, ThSi, Th$_3$Si$_2$, ThS, and Th$_2$S$_3$ have been partially characterized for ceramic nuclear use.

Conclusions

Out-of-pile thermodynamic properties of thorium alloys, oxides and carbides are fairly well known. Out-of-pile transport properties are known to a lesser extent, especially their dependence on impurities or fabrication methods. Moreover, these transport properties are more markedly changed in a neutron field. More in-pile measurements are therefore required. This is particularly true for bulk monocarbides, alloys and oxides.

Some irradiation tests have been performed on thorium alloys and oxides. These results are encouraging but more irradiation information is required, especially in the case of the bulk monocarbide. Much more information is available on the behaviour of dicarbide in the form of coated microspheres.

Recommended programme for the IAEA

(1) In connection with the forthcoming Agency symposium on the thermodynamic properties of nuclear materials, it is recommended that the more important thermodynamic properties of thorium-based fuels be reviewed by an ad hoc committee generated at that symposium. The properties of interest include the heat capacities, the heats of formation, the free energies of formation, and other information concerned with chemical compatibility among reactor materials.

(2) A working group should be formed with the task of collecting and critically evaluating all the data available and currently being generated on thorium-based fuels. In particular, for metallic fuels, information on swelling data as a function of purity and fabrication method is required.
For oxide-based fuels, thermal conductivity and gas release data are needed. For the bulk monocarbide, for which there is almost no information, irradiation swelling and chemical compatibility should be investigated first. It should be the function in this working group not only to collect and evaluate the existing data but to stimulate the generation of additional data on well-characterized materials.

C. FUEL CYCLE TECHNOLOGY*

1. Fabrication methods

Direct fabrication techniques have been successfully developed for thorium-bearing fuels of various types, and there is a variety of techniques which can be used in the fabrication of any particular fuel. Generally, the fabrication processes of interest may be divided according to the type of fuel element to be fabricated. The principal ones of interest are those for (1) fuelled-graphite elements, (2) metal-clad oxide elements and (3) clad thorium metal or alloy elements.

a. Oxide metal-clad process

In general, the ceramic and metallurgical processes used for uranium dioxide can be applied to thoria-base fuel. These include pelletizing, swaging, extrusion techniques and vibratory compaction. Techniques of pelletizing are highly developed for thoria, for example, the cores of CETR and Elk River. Ceramic extrusion methods are to be practised in the PCUT facility and are also developed. The sol-gel process, coupled with vibratory compaction, has already shown great potential for cost reduction in the fabrication of non-recycle cores and promises even greater advantages for recycle fuel.

Methods of end-cap welding, inspection and assembly of fuel elements are well developed.

b. Fuelled-graphite processes

Many good fabrication techniques for spherical particles have been developed. The ability of fabricators to make fuel in reasonably large quantities has certainly been demonstrated and they are now aiming at cost reductions.

Fuel particle kernels can be made by several powder metallurgical techniques and by several sol-gel processes. Production experience has been obtained using the powder metallurgical techniques; however, the sol-gel processes have the potential for rapid and less expensive production.

Coatings of carbon or silicon carbides are deposited by pyrolysis of hydrocarbons or silicon-bearing gaseous compounds. Many studies of the properties and the nature of pyrolytic carbon have been made and this ma-

* The rapporteurs for this topic were Mr. A.L. Lotts, Dr. G. Orsenigo and Mr. J.T. Roberts.
terial can be reasonably well characterized. These coatings can be deposited in either fluidized beds or in rotating drum coaters. The fluidized bed coaters are more generally used. Rotating drum coaters are more complex mechanically but have the advantage of requiring no inert gas carriers for the purpose of particle levitation.

To date most of the results have been obtained on carbide-based fuels. Oxides are easier and less expensive to produce. If the particle coatings on the oxides remain intact, then the particles can be heated to temperatures much higher than those needed in reactors without any evidence of reaction between the oxide kernel and the carbon coatings.

c. Metal-clad thorium metal and alloys

Little experience with the fabrication of large quantities of thorium metals and alloys, either bare or extruded, exists. Hanford has co-extruded Th-2.5%U-1.0%Zr with Zircaloy-2. Experiments have shown that thorium metal is readily fabricated by casting, powder metallurgy, extrusion, rolling and other methods.

2. Reprocessing and refabrication

a. Introduction

Large centralized reprocessing and refabrication plants will be able to achieve quite low unit manufacturing costs and would appear to offer the most promising eventual solution for solid-fuelled reactors. This solution probably becomes effective when the industry becomes large enough that one plant services approximately 5000 MW(e) or more reactors. The high unit costs of small plants offer a serious economic obstacle to the development of advanced reactor concepts during their early growth phase if their fuel cannot be handled, or can be handled only at significantly reduced throughput rates, in existing plants.

On-site processing probably is necessary to the success of fluid-fuelled reactors and may well be an acceptable interim solution for advanced solid-fuel reactors. Though it is by no means proven for any type of reactor, at least for some reactor types there is promise that on-site reprocessing and refabrication can achieve acceptable economics during the period before a nuclear power industry is large enough to support a central plant. Some of the reasons for this include the following:

1. The cost of shipping irradiated fuel is avoided
2. The fuel cycle inventory outside the reactor is reduced by reducing time delays for cooling and transit
3. Standardization of fuel types has not yet been achieved and it may be premature to build a central plant before this is realized, even though one is sure of the overall growth of nuclear power industry.

For achieving maximum fuel utilization, both thorium and uranium-233 must eventually be recycled. Uranium recycle must begin fairly early, for economic reasons, though thorium recycle might be delayed to permit $^{228}\text{Th}$ decay. As the amount of recycle uranium and its $^{232}\text{U}$ level increase, re-
mote refabrication will become necessary anyway, at least in central plants, and the main economic justification for thorium decay-storage will be eliminated. Thus, immediate remote refabrication in central plants appears to be the long-term solution, though direct fabrication in glove boxes offers a practical solution for start-up of a thorium-fuelled reactor industry.

Integration of central reprocessing and remote-refabrication plants seems to be economically advisable, saving both time and money, as a result of sharing facilities and eliminating the need for shielded transportation of the fuel materials between sites. On-site reprocessing and refabrication facilities are, of course, integrated with each other as well as with the reactor, by definition.

Close-coupled processing, i.e. reprocessing and refabrication techniques based on minimum fission-product decontamination consistent with reactor physics requirements, is an intriguing concept, though it cannot yet be said to offer proven economic advantages.

The ORNL studies of reprocessing and refabrication costs indicate the economic advantages of large central plants. The Italian CNEN study of an on-site plant for a 300 MW(e) reactor is an example of the possible economic feasibility of this concept.

b. Reprocessing

For metal-clad thorium oxide or metal fuels reprocessing methods differ only in degree, not in kind, from those for the corresponding uranium fuels and the technology can be said to be well developed, though not actually demonstrated on irradiated thorium fuels in all cases. By comparison with uranium fuels, the thorium fuels typically require more powerful dissolvers, require a feed adjustment step, have lower solvent extraction throughput rates, have higher fissile enrichments and burn-ups, require allowance for $^{233}$Pa, $^{232}$U, $^{228}$Th and $^{234}$Th decay, and may generate wastes more expensive to handle. In a reprocessing plant designed to handle uranium fuels, these differences may involve a cost penalty of a factor of two or more for thorium fuels. On the other hand, in a plant designed to handle thorium fuels, only a modest extra cost should be involved. Actual checking out of the reprocessing methods on the proposed fuel types, both unirradiated and fully irradiated, would be desirable to eliminate uncertainties.

The reprocessing of carbide and graphite type fuels is not so well developed as the metal-clad oxide or metal types, but research programmes presently under way promise the eventual development of one or more acceptable head-end processes to permit subsequent decontamination by more-or-less conventional solvent extraction methods. Burn-leach and grind-leach head-end processes for HTGR-type fuels are receiving serious attention. Burning simplifies dissolution and feed adjustment, but the behaviour of fission products in the burning step cannot yet be said to have been proved as acceptable, though there are indications that this may not be a serious problem. Grinding itself may involve fewer technological problems than burning, though the losses of fuel to the graphite and the eventual disposal of the graphite yet remain to be proven acceptable. Electrolytic disintegration of fuel compacts may facilitate dissolution in a grind-leach
system. Physical separation of fuel compacts or particles from the un-fuelled graphite would simplify both burn-leach and grind-leach processing and would reduce shipping costs if this is performed at the reactor site. Fuel element differences, such as the presence or absence of a SiC coating on the fuel particles, might lead to burn-leach in one case and grind-leach in another. Hydrolysis and pyrohydrolysis as well as burning may be applicable with carbide fuels. All of these processes, both head-end and solvent extraction, for HTGR fuels require further development and eventual demonstration on high burn-up material.

The development of close-coupled reprocessing methods, involving process simplification at the expense of low decontamination factors (perhaps as low as 10 to 100), is still in an early stage. Much more work is needed to establish the validity of this approach to lower reprocessing costs. The build-up of impurities in the recycle system and the effect of impurities on sol-gel processing needs further study.

Most of the above-mentioned reprocessing methods are under active study and the suggested demonstrations on fuel-burning fuels can be expected during the next several years. There is, however, still room for research on new reprocessing methods for thorium fuels in an attempt to significantly reduce reprocessing costs.

A technologically-sound interim solution to the reprocessing plant start-up problem is to do no reprocessing during the early growth years of a new power reactor type, i.e. to store the fuel until the industry is large enough to support an economic-size reprocessing plant. This "throw-away" fuel cycle is economically less attractive for thorium reactors than for natural uranium reactors because of the typically much higher fissile content of the spent fuels and the desirability of beginning the $^{233}$U recycle early to gain its reactor physics advantage. Even so, it may be an acceptable economic solution, depending on inventory charges, government fuel buy-back guarantees, etc. and does not require further research and development before it can begin.

c. Refabrication

The technology for refabrication is in a very elementary stage.

(i) Mode of fabrication

One of the principal problems is the determination of the mode of refabrication of Th-$^{233}$U fuels. This problem is being, or has been, investigated through operation of pilot plants at Babcock and Wilcox Company, the ORNL Klorod Facility, the PCUT Facility and the ORNL Thorium-Uranium Recycle Facility.

The Babcock and Wilcox Company sees the need, first for developed concepts to completely decontaminate recycled uranium and thorium. During the industry growth period, complete fission product decontamination coupled with lightly-shielded fabrication facilities can meet fuel cycle requirements of thorium reactors with minimum capital investment in fabrication facilities.
Later, as the thorium economy demands, the greater investment in remote fabrication-reprocessing facilities can be justified.

The Babcock and Wilcox Company has built a pilot plant at the Nuclear Development Centre in Lynchburg, Virginia, to investigate the direct and rapid fabrication of $^{233}$U fuels and demonstrate the feasibility of such a concept. The pilot plant is completely encased in a system of glove boxes.

The first stage of the pilot plant is an ion-exchange system to strip $^{228}$Th and therefore reduce the gamma activity of the fuel. The second stage of the pilot plant includes the conversion of uranyl nitrate and thorium nitrates into a mixed-oxide power by the sol-gel process. The third stage of the pilot plant includes the vibratory compaction of the oxide powder into fuel rods. At this point the rods must be stored before being assembled into a complete fuel bundle. Because of the gamma activity the storage and assembly is done remotely underwater in the fourth stage of the pilot plant.

The pilot plant is now being operated with virgin thorium and depleted uranium, before operation, in a few months, with uranium-233.

ORNL Kilorod Facility. At ORNL, approximately 1100 fuel rods containing (3 wt.% $^{233}$U-97 wt.% Th)O$_2$ were produced in a semi-remote facility (4.5 in. steel) using the sol-gel process and vibratory compaction. The $^{233}$U contained 37 ppm $^{232}$U.

Approximately 900 of the Zircaloy-2 clad fuel rods were $\frac{1}{2}$ in. in diameter and 42 in. long, while 200 were 19 in. long. The fuel rods, which were fabricated at a rate of 10 per day, averaged 90% of theoretical density. The maximum permissible variation in density for a single rod was $\pm$ 2% of its average density. In the vibratory compaction process, almost 100% of the feed material was used; thus, the amount of material recycled within the process was very little.

The operation of the Kilorod facility has most certainly demonstrated the feasibility of economical semi-remote fabrication and encourages optimism concerning the prospect of completely remote fabrication. The experience gained in the design, construction and operation of the Kilorod facility has served as a part of the basis for ORNL programmes for fabrication cost evaluation and for remote fabrication of fuel.

PCUT Facility. CNEN is completing the construction of PCUT, which is a heavily shielded facility and can carry out integrated recycle processes. The equipment to be installed initially is based on a fabrication flow sheet, consisting of ceramic extrusion techniques, end-capping and assembly procedures. The equipment is presently being tested before installation in the facility.

ORNL has just started construction of the Thorium-Uranium Recycle Facility (TURF), which is also a heavily shielded facility. The facility consists of four process cells.

1. Mechanical processing cell for head-end processes and for reconstitution of fuel
2. The chemical cell which will be used for chemical processing and the purification of fuel
(3) The contaminated fabrication cell
(4) The clean fabrication cell.

Equipment is currently being designed and constructed for demonstration of remote fabrication processes in the TURF.

The construction of TURF is expected to be completed early in 1967. All of the oxide fabrication equipment has been conceptually designed, and a substantial portion of the detailed design has been done. The conceptual design of the equipment for fabrication of fuelled-graphite elements has just commenced.

(ii) Summary of process status

Oxide metal-clad fuel elements

A process for thorium-uranium oxide has been demonstrated in a semi-remote facility (Kilorad Facility at ORNL). Babcock and Wilcox Company plans to put in operation in about 3 months a glove box facility for $^{233}\text{U}$. The PCUT Facility and the ORNL TURF will demonstrate processes for remote fabrication of oxide-metal clad elements within several years.

Fuelled-graphite elements

The development of procedures for remote fabrication is in a very preliminary stage. ORNL has accomplished a conceptual design of equipment to be used in the remote fabrication of an HTGR fuel in the TURF. Also, it is conducting pilot scale experiments directed towards the development of economical methods of performing remotely key steps in the process. This work involves sol-gel techniques for making spherical oxide particles, methods for direct conversion of oxide particles to carbide, and methods of coating particles with pyrolytic carbon.

Thorium and thorium alloys

No consideration has been given to the remote fabrication of thorium and thorium alloys. However, it appears that some difficulties are involved in that metal working machinery is usually quite large in size, thereby raising questions of maintenance. Also, in certain procedures, pyrophoricity would be a problem, e.g. the steps in making thorium metal.

(iii) Refabrication costs

The design of fuel elements greatly affects the cost of fabricating fuel elements. For example, the designer of metal-clad elements must optimize the fuel diameter, cladding material, fuel element length, and number of enrichments to obtain minimal costs. Similar considerations must be given to the design of fuelled-graphite and other elements. Also, attempts should be made to reduce the number of steps which must be done remotely.

Data showing that the cost of refabrication will not be prohibitive were presented. Including all direct costs (capital, operating, and hardware), penalties range from 10 to 50%, depending upon the plant production rate,
number of operations that can be done outside the hot cell and the particular process. It was also shown that the effect of $^{232}$U concentration in fuel on the cost decreases with increase in plant capacity.

(iv) Work required

The accurate assessment of refabrication technology and costs must await the operation of the various pilot systems mentioned previously. Until these systems are evaluated, it will be difficult to specify what additional work should be done in this area.

3. Recommended programme for the IAEA

(1) Successful development of the fuel cycle technology is of crucial importance in the utilization of thorium. The Agency should, with the cooperation of various centres in each country or appropriate organization, accumulate and disseminate available information concerning fabrication, reprocessing and refabrication of thorium-based fuels. Each centre could send periodic reports of major activities and developments to IAEA Headquarters to be consolidated with other similar reports and then distributed.

(2) It is recommended that the IAEA contribute to processing fabrication development by dissemination of current information about the research programmes of Member States. One suggestion for implementation of this recommendation is that the Agency prepare suitable topical reports for general distribution by visits to important installations doing the research.

D. VARIOUS REACTOR CONCEPTS AND THEIR POTENTIAL*

The panel did not undertake a comparison of the relative merits of different reactor concepts but took note of the studies and analyses made in individual countries which are summarized below.

**Molten-salt reactor**

The 10-MW(e) molten-salt reactor experiment, a circulating fuel, graphite-moderated reactor in which the fuel is a molten mixture of $^{235}$U, $^7$Li, Be and Zr fluorides, achieved criticality at Oak Ridge on 1 June 1965. The molten salt enters the core at 650°C, flows through channels in the graphite moderator structure, and emerges at 663°C. No electricity is generated by the reactor, but the salt temperature is high enough that very favourable steam conditions could be achieved for a power cycle.

The reactors will be operated initially to provide a large-scale high-temperature test of the compatibility of fuel salt, graphite moderator, and the Hastelloy N reactor vessel and piping. Thorium salts are to be added

* The rapporteurs for this topic were Mr. R. Naudet, Mr. С Renme, Mr. M. W. Rosenthal, Prof. Dr. J.J. Went and Mr. J.A. Lane.
to the fuel salt later to demonstrate a technology which can be used to achieve breeding in a thermal reactor.

**Aqueous homogeneous suspension reactor**

The development of this reactor is continuing in the Netherlands. The problems of hydraulic nature, of erosion, of settling and of caking have been solved. With a small zero-energy reactor it was demonstrated that, without difficulties, the concentration of the suspension can be kept constant within the limits for a stable operation. A small test reactor (KSTR), in which power reactor conditions, such as fission density, temperature, pressure, water decomposition and flow conditions can be maintained, is in construction.

The main aim of this facility is to investigate the irradiation behaviour of the following suspensions:

(a) A mixed Th-U-oxide colloidal solution with particles of 100-200 Å
(b) A mixed Th-U oxide suspension with particles of 2-5 μm
(c) A coated particle with a UO₂ core and a ThO₂ coating.

**Pebble-bed reactor and other projects in the Federal Republic of Germany**

The 15-MW(e) BBC-Krupp pebble-bed reactor near Julich, called the AVR (Arbeitsgemeinschaft Versuchs-Reaktor GmbH) is expected to be loaded with the first fuel balls at the end of 1965. Like Dragon and Peach Bottom reactors it uses the coated particles of uranium and thorium carbides dispersed in a graphite matrix but the difference lies in the shape of the fuel elements which are spherical pebbles. While experience with AVR is being awaited, the THTR Association is going ahead with plans for a 300-MW(e) project of this type and a research and development programme has been initiated connected with pre-stressed concrete pressure vessel, carbon transport from hot to cold points and further improvement of the fuel.

In addition, feasibility studies have been initiated in Germany on three potential thorium breeder reactors which are as follows:

(a) A breeder version of the pebble-bed reactor will be studied using "fertile" and "fissile" balls in two zones
(b) A heavy-water reactor optimized for breeding will be considered, fabrication and re-processing methods together with irradiation experiments will receive primary attention
(c) The epithermal molten-salt reactor concept will be investigated, including external heat exchanger with molten lead as a coolant in direct contact with the salt.

**Dragon Project and related studies in the United Kingdom**

The Dragon Reactor Experiment was made critical for the first time in August 1964 and all the testing and commissioning of the reactor plant and the programme of zero-energy measurements has now been completed and the reactor should be at its design power of 20 MW(th) by September 1965. The reactor would then be run to test the coated particle fuel and to test the novel engineering design features of this helium-cooled high-temperature reactor, such as the leak tightness of the helium circuit, the gas bearing
circulators, the helium purification system, and the fuel handling equipment. So far no major difficulties have been encountered in the performance of the reactor plant.

Assessment studies are also being made for a 500 MW(e) high-temperature reactor power plant with the reactor core and heat exchangers in a pre-stressed concrete pressure vessel and using a high thermal efficiency steam cycle. Initially fission-product-retaining fuel is being considered, which may be directly cooled by the helium. Different fuel management schemes are being investigated with particular attention being paid to the economies of once-through fuel cycles, in which the spent fuel is stored until reprocessing plants are available, to see if these reactors can be operated initially in this manner. Subsequently the use of recycled fuel is envisaged to give lower fuel costs. The low capital cost, low inventory costs resulting from the high fuel ratings possible, and low make-up fuel costs resulting from the high conversion factors achievable are attractive features of these reactors.

**Heavy-water thorium reactors in France and Canada**

In France the possible use of thorium is being considered in connection with the heavy-water reactor system which is under rapid development in the country. The good neutron economy of heavy-water reactors makes them especially suitable as converters.

In considering reactors suitable for thorium utilization, one must not forget that there is an important difference between reactors specially designed for thorium use and requiring the development of a new technology and the ones based upon currently available technology, which could be adapted progressively to use the thorium cycle without important modifications. Heavy-water reactors which are already at an advanced stage of development should be favourably considered in this respect.

When speaking of "conservation of nuclear fuels" one should not think that uranium reserves are limited. Actually only low cost uranium reserves are limited. If one reduced the consumption of fuel per kilowatt-hour generated, one could afford to use even high cost reserves too. World uranium resources are very large if one can economically use high priced ores. Therefore it is not certain that fast breeder reactors are necessary.

Canada's reliance on heavy-water reactors and advanced position in their development leads to a viewpoint similar to that expressed by France. If uranium prices rise and processing costs fall the fuel cycle in the then existing reactors can be changed to take any advantage offered by thorium. Even on a world scale it appears, on this basis, that breeder reactors, fast or thermal, would stand or fall by their ability to meet the economic competition.

**Fast thorium reactor in the USSR**

In the Soviet Union the development of a fast thorium reactor is receiving attention because it is felt that the most efficient utilization of the nuclear fuel and the introduction of the uranium-238 and thorium in the fuel cycle could best be achieved by a fast neutron reactor of a converter type
of 600-800 MW(e), which would then gradually pass to a breeding regime of operation. The characteristic features of such a reactor appear to be quite satisfactory, also in case of a mixed fuel cycle containing thorium.

With an optimum power density in the reactor core (400-500 kW/l) corresponding to the shortest doubling time for an oxide fuel PuO$_2$-$^{233}$UO$_2$-$^{238}$UO$_2$ in the core and of thorium in the blanket, the doubling time would amount to 10 yr as compared to 7 yr for the PuO$_2$-UO$_2$ fuel cycle; this doubling time would be 20 yr for a pure $^{233}$U-Th cycle. (In the first case the breeding coefficient would amount to 1.33-1.4 as compared to 1.22-1.19 for the pure $^{233}$U-Th cycle).

It is therefore evident that the mixed fuel cycle opens new possibilities for utilization of thorium concurrently with the uranium-238. The advantages of such a cycle can be summarized as follows:

(a) High breeding coefficient (in view of the contribution of the uranium-238 and $\nu_{\text{eff}}$ for plutonium);
(b) Smaller critical mass (by some 100 kg);
(c) The rate of introduction of thorium exceeds by several times the rate of introduction of uranium;
(d) Introduction of thorium into the blanket reduced the protactinium poisoning and it practically does not affect the breeding coefficient;
(e) The activity of $^{232}$U and $^{233}$U is reduced by about 10 times as compared to utilization of thorium in thermal reactors or in the cores of fast reactors (in view of a reduction of the cross-section of $^{232}$Th(n,2n)$^{231}$Th reaction; moreover, in the fast reactor blanket the absorption cross-section (n,$\gamma$)$^{231}$Pa is 100 times less than in the thermal thorium reactors.)

A separate location of uranium-233 and of thorium reduces concentration of thorium-228 also as a result of elimination of uranium-233 and uranium-232 which is the parent of thorium-228.

**Comparative study of certain concepts**

A comparison was made in the United States of the design features, technical problems, economics, and fuel utilization of a number of 1000 MW(e) reactors which use thorium fuel and a 1000 MW(e) pressurized-water reactor using partially enriched uranium fuel$^1$. Included in the comparison with the PWR were a spectral-shift-control reactor, a pressure-tube heavy-water reactor$^2$ (cooled by pressurized heavy water), a movable-fuel seed-blanket, "converter-breeder" reactor, a high-temperature gas-cooled reactor, a one-region circulating-fuel molten-salt converter reactor, and a two-region molten-salt breeder reactor. The economic comparison was made for conditions postulated in the United States to exist on the average over the 30-yr life of reactors built in the 1970s ($17.6$/kg U$_3$O$_8$; $30$/kg separative work; 12%/yr fixed charges on the reactors; 10%/yr fuel and D$_2$O inventory charges). Fuel fabrication and processing costs were estimated for plants having capacities that would serve 15000 MW(e) of the concept considered. All the thorium reactors were assumed to be started up

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$^1$ A Comparative Evaluation of Advanced Converters, Rep. ORNL-3686 (Jan 1966)

$^2$ The reactor design was apparently optimized for a relatively lower efficiency as compared to plants already under construction
### COMPARATIVE STUDY OF CERTAIN REACTOR CONCEPTS

<table>
<thead>
<tr>
<th></th>
<th>PWR</th>
<th>SSCR</th>
<th>HWR</th>
<th>HTGR</th>
<th>SBR</th>
<th>MSCR</th>
<th>MSBR</th>
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<td><strong>Net electrical capacity, MW(e)</strong></td>
<td>1002</td>
<td>1000</td>
<td>1010</td>
<td>1008</td>
<td>1000</td>
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<td>31</td>
<td>31</td>
<td>26</td>
<td>44</td>
<td>31</td>
<td>44</td>
<td>44</td>
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<td><strong>Reactor inventory</strong></td>
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<tr>
<td>kg(fissile)/kW(e)</td>
<td>2.05</td>
<td>2.91</td>
<td>1.44</td>
<td>2.89</td>
<td>3.5</td>
<td>1.75</td>
<td>0.62</td>
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<td></td>
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<tr>
<td>MW(th)/kg fissile</td>
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<td>1.12</td>
<td>2.67</td>
<td>0.78</td>
<td>0.91</td>
<td>1.3</td>
<td>3.63</td>
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<td>Exposure MWd/t of fuel</td>
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<td>25400</td>
<td>29400</td>
<td>52300</td>
<td>12000</td>
<td>2300</td>
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<td>Fuel enrichment, wt. % fissile</td>
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<td>4.8</td>
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<td>3.1</td>
<td>1.4</td>
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<td><strong>Process capacity for 15 000 MW(e), t/yr</strong></td>
<td>670</td>
<td>550</td>
<td>590</td>
<td>190</td>
<td>1300</td>
<td>-</td>
<td>-</td>
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<td><strong>Fabrication cost, $/kg U+Th</strong></td>
<td>53</td>
<td>47</td>
<td>38</td>
<td>115</td>
<td>52</td>
<td>-</td>
<td>-</td>
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<tr>
<td><strong>Processing cost, $/kg U+Th</strong></td>
<td>31</td>
<td>39</td>
<td>40</td>
<td>105</td>
<td>23</td>
<td>2.7</td>
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<td><strong>Total shipping cost, $/kg U+Th</strong></td>
<td>4</td>
<td>8</td>
<td>6</td>
<td>26</td>
<td>5</td>
<td>-</td>
<td>-</td>
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<tr>
<td><strong>Final conversion ratio</strong></td>
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<td>0.75</td>
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<td>0.90</td>
<td>1.00</td>
<td>0.96</td>
<td>1.06</td>
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<td><strong>Plant capital cost, $/kW(e)</strong></td>
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<td>116</td>
<td>126</td>
<td>110</td>
<td>124</td>
<td>123a</td>
<td>128a</td>
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</table>

**Power cost, mill/kWh**

<table>
<thead>
<tr>
<th></th>
<th>Capital</th>
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<th>Fuel cycle</th>
<th>Heavy water</th>
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<tr>
<td></td>
<td>2.1</td>
<td>0.3</td>
<td>1.6</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>0.3</td>
<td>1.8</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>2.2</td>
<td>0.3</td>
<td>1.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.9</td>
<td>0.3</td>
<td>1.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.1</td>
<td>0.3</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.1</td>
<td>0.3</td>
<td>0.8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.2</td>
<td>0.3</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>4.0</td>
<td>4.2</td>
<td>4.4</td>
<td>3.5</td>
</tr>
</tbody>
</table>

- Excluding on-site processing facilities, for which charges are included under fuel cycle cost.

with $^{235}$U and the bred uranium recycled over a 30-yr life, but the plutonium from the PWR was assumed to be sold at $10/g fissile. The reactor designs were obtained from sponsors of the concepts in the United States, and with
the exception of the SBR, which was specifically designed to have a breeding ratio of 1.0 on $^{233}$U fuel, were generally optimized for lowest cost. Some of the results are given in Table II.

**Economic incentive for thorium reactors in the USA**

The economic incentive for thorium reactor development in the USA can be summarized as follows:

The continued construction of slightly enriched water reactors in the USA will lead to the exhaustion of low-cost uranium reserves (about one million tons in the USA and Canada) before the end of the century. In addition, further $^{235}$U separation plants would be required. The use of higher-cost uranium will increase power costs unless breeder reactors are developed which replace mining as a source of fissile material. The key to long-term economic power is to establish a self-sustaining breeder industry before running out of low-cost uranium and before new separation plants have to be built.

To meet this aim, sodium-cooled fast-breeder reactors would need to be commercially competitive by 1980, and this may not be achieved. Alternative approaches should therefore be considered. One approach is to build natural uranium heavy-water-moderated thermal reactors in the period 1975-1990 which would provide more plutonium for fast breeder reactors and avoid the need to build more separation plants. Fast breeder reactors with a doubling time of 10 yr introduced 10 yr later in 1990 would then limit the consumption of natural uranium to about one million tons.

Another approach is to develop advanced converter reactors fuelled with thorium which can reduce the requirements for uranium and separation plants and offer lower power costs than enriched water reactors. These advanced thorium-fuelled converter reactors could be developed into thermal breeder reactors later. Introducing advanced thermal converters in the period 1975-1990 followed by thermal breeders in 1990 would hold mined uranium requirements down to 600,000 tons and this amount of uranium is well within the available resources of low cost uranium. The corresponding present worth of saving in mining and separation plants would amount to 10 billion dollars which should provide an adequate economic incentive for the development of thorium reactors.

**Conclusion**

1. In view of the strong interest in breeder reactors for the long-term future, thermal-breeder reactors should be developed as well as fast-breeder reactors, for two reasons:
   (a) To find out which are the most economic types of breeder reactors for particular conditions
   (b) To exploit the lower fissile material inventory features of thermal breeder reactors in a rapidly growing power programme.
2. In parallel there should be emphasis on developing further those thorium converter reactors which do not require major advances in technology and which have good fuel utilization when operated under conditions which give low power costs.
Recommended programme for the IAEA

The Agency should closely follow the progress of various thorium reactor systems having good fuel utilization and promote their development by holding technical meetings. It would be best to organize a working group of technical experts from interested Member States to periodically review progress in thorium technology and utilization and thereby promote exchanges of information and possible international collaboration in thorium research.

VI. OVERALL POTENTIAL FOR NEAR-TERM AND LONG-TERM UTILIZATION OF THORIUM

The development of thorium reactors is still in an early stage relative to that of uranium-fuelled reactors; nevertheless, progress to date indicates a very encouraging outlook for the near-term utilization of thorium (see Table III). Highly significant in this regard has been the evolution of the technology of high-temperature gas-cooled reactors, and it appears that the installed capacity of these reactors might reach 1000 MW(e) within the next decade. Other advanced thorium converters, such as the heavy-water-moderated and seed-blanket types, are being considered as competitors to the uranium-plutonium converters for power and desalination applications. Differing opinions on the relative merits and prospects of these reactor types were expressed by panel members. Whereas the lack of fuel recycle technology necessary for the support of thorium reactors has been a barrier to the exploitation of these reactors, operation of the Kilorod facility and substantial economic analyses at Oak Ridge National Laboratory are encouraging and indicate that the penalty for remote refabrication may not be severe.

The long-term potential of thorium reactors is more difficult to assess. However, several promising types are under active development and a more accurate evaluation of their potential may be possible in the near future. Successful operation of the molten-salt reactor experiment, which recently achieved criticality at the Oak Ridge National Laboratory, would pave the way towards a vigorous thermal breeder development programme in the United States. In addition, the KEMA Suspension Reactor in the Netherlands should demonstrate the technical feasibility of thorium-uranium suspension reactors within about three years. Either of these types of thermal breeders if successfully developed would compete with fast breeders, particularly because of their lower fissile inventories, and could supply a significant fraction of world fissile needs for an indefinite period. Even after the breeder reactors have been developed fully there may be a useful role for advanced thorium converters which may be required to provide economically a power system with load factor flexibility, since the breeder reactors operate most economically with short doubling time and hence high load factors; also, if surplus fissile material were to become available from a well-established breeder industry, it could be used profitably in advanced thorium converters.

The development of both fast and thermal breeders involves very considerable advances in reactor technology relative to that necessary for current generation reactors. However, the introduction of advanced thorium
converters would permit a more gradual evolution in technology towards thermal breeders. This may result in reduced costs of development necessary to reach a self-sustaining breeder industry.

VII. POSSIBILITIES OF FURTHER CO-OPERATIVE EFFORTS IN THORIUM UTILIZATION

The panel generally endorsed the desirability of further international collaboration in research and development work connected with the utilization of thorium. However, it did not make any specific suggestions for co-operative effort among countries covering any particular areas of research. It asked the Secretariat to continue exploring various possibilities in this connection.

As a first step, the Agency should organize prompt dissemination and exchange of information about thorium technology among its Member States. The establishment of a working group to review periodically the progress and development in the utilization of thorium in power reactors would be useful for fostering closer co-operation among the interested countries.

It was noted that several international co-operative agreements were already in force. Dragon is a co-operative project of the Organisation for European Economic Co-operation (OEEC) and the United States has close relationship with EURATOM countries and the Dragon Project. Italy has a bilateral agreement with the United States covering reprocessing and re-fabrication of thorium fuel from the Elk River Reactor. Sweden and India have recently initiated a joint research effort.

However, there remains a need for further collaboration in the field, from which both advanced and developing countries with large reserves of thorium could benefit.

VIII. RECOMMENDATIONS OF THE PANEL

(1) The Agency should, with the co-operation of Member States, compile information on thorium resources.

(2) As part of its activities on nuclear data, the IAEA should compile neutron cross-section data on an international level, with particular emphasis on (a) $^{233}\text{U}$; (b) Fission products and yields, both primary and secondary; (c) $^{237}\text{Np}$; (d) $^{232}\text{U}$; (e) $^{231}\text{Pa}$; (f) $^{233}\text{Pa}$; (g) $^{241}\text{Pu}$; (h) $^{242}\text{Pu}$.

(3) The Agency should compile data on thorium lattice experiments for use in the design of reactors.

(4) The Agency should support a basic type of integral experiment (e.g. uniform dispersion) as verification of $^{233}\text{U}$ data by making $^{233}\text{U}$ available to experimenters and awarding research contracts to appropriate laboratories.

(5) It is most desirable to obtain isotopic analysis and reactivity measurements on very high burn-up $^{233}\text{U}$-Th under well-defined spectrum conditions and the Agency could support such work under its research contracts programme.
## TABLE III

THORIUM-FUELLED REACTORS AND CRITICAL ASSEMBLIES

<table>
<thead>
<tr>
<th>Name</th>
<th>Location</th>
<th>Year</th>
<th>Power</th>
<th>Fuel (^{a})</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>SRE</td>
<td>A1, USA</td>
<td>(1961)</td>
<td>20 MW</td>
<td>93% U(_{\text{metal}}) + Th</td>
<td>Graphite-Na</td>
</tr>
<tr>
<td>PDP</td>
<td>Du Pont, USA</td>
<td>(1953)</td>
<td>1 kW</td>
<td>variable</td>
<td>D(_{2})O</td>
</tr>
<tr>
<td>BORAX IV</td>
<td>NRTS, USA</td>
<td>(1958)</td>
<td>20 MW</td>
<td>90% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>BWR</td>
</tr>
<tr>
<td>ZENITH</td>
<td>Winfrith, UK</td>
<td>(1959)</td>
<td>200 W</td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>Graphite-gas</td>
</tr>
<tr>
<td>Peach Bottom</td>
<td>La Jolla, USA</td>
<td>(1960)</td>
<td></td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>Graphite</td>
</tr>
<tr>
<td>AHCF</td>
<td>JAERI, Japan</td>
<td>(1961)</td>
<td>100 W</td>
<td>20% U(<em>{2})SO(</em>{4}) + D(_{2})O (core)</td>
<td>D(_{2})O homogenous</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Th(<em>{2}) suspension in D(</em>{2})O (blanket reflector)</td>
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</tr>
<tr>
<td>CETR (Cnt. Ass.)</td>
<td>Santa Susano, USA</td>
<td>(1961)</td>
<td></td>
<td>235U + 233U + Th</td>
<td>AETR</td>
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<tr>
<td>CETR</td>
<td>Con Ed, USA</td>
<td>(1962)</td>
<td>585 MW</td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2}) (1st core only)</td>
<td>PWR</td>
</tr>
<tr>
<td>ERR</td>
<td>Elk River, USA</td>
<td>(1962)</td>
<td>22 MW(e)</td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>BWR</td>
</tr>
<tr>
<td>ROSPO</td>
<td>CENEN, Italy</td>
<td>(1963)</td>
<td></td>
<td>90% U(<em>{2})(</em>{O}) + Th blanket</td>
<td>Organic</td>
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<tr>
<td>SLE</td>
<td>Lynchburg, USA</td>
<td>(1963)</td>
<td></td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>D(_{2})O</td>
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<tr>
<td>THUD</td>
<td>Argonne, USA</td>
<td>(1963)</td>
<td></td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>D(_{2})O</td>
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<td>BR-1</td>
<td>Obninsk, USSR</td>
<td>(1963)</td>
<td>100 W</td>
<td>Th blanket</td>
<td>Fast breeder</td>
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<td>LPR (Cnt. Ass.)</td>
<td>Bettis Lab., USA</td>
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<td>Th(<em>{2}) + 233U(</em>{2})</td>
<td>Seed-blanket H(_{2})O</td>
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<tr>
<td>DRAGON</td>
<td>Winfrith, UK</td>
<td>(1964)</td>
<td>20 MW</td>
<td>93% U(<em>{2})(</em>{O}) + Th(_{2})</td>
<td>Graphite-He</td>
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### TABLE III (cont.)

<table>
<thead>
<tr>
<th>Name</th>
<th>Location</th>
<th>Year</th>
<th>Power</th>
<th>Fuel(^a)</th>
<th>Type</th>
</tr>
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<tr>
<td>KSTR</td>
<td>KEMA, Netherlands</td>
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<td>1000 kW</td>
<td>93% U(<em>{235})+Th(</em>{233})</td>
<td>Aqueous suspension</td>
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<td>Kilorröd Experiment</td>
<td>BNL, USA</td>
<td>(1964)</td>
<td></td>
<td>93% U(<em>{235})+Th(</em>{233})</td>
<td>H(_2)O</td>
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<tr>
<td>ZED-2</td>
<td>Chalk River, Canada</td>
<td>(1965)</td>
<td>200 W</td>
<td>93% U(<em>{235})+Th(</em>{233}) (first thorium loading)</td>
<td>D(_2)O</td>
</tr>
<tr>
<td>Peach Bottom HTGR</td>
<td>Peach Bottom, USA</td>
<td>(1965)</td>
<td>46 MW(e)</td>
<td>93% U(<em>{235})+Th(</em>{233})</td>
<td>Graphite</td>
</tr>
<tr>
<td>MSRE</td>
<td>Oak Ridge, USA</td>
<td>(1965)</td>
<td>10 MW</td>
<td>93% U(_{235})+Th (future)</td>
<td>MSR cooled</td>
</tr>
<tr>
<td>AVR</td>
<td>Julich, Germany</td>
<td>(1966)</td>
<td>15 MW(e)</td>
<td>93% U(<em>{235})+ThC(</em>{2})</td>
<td>Graphite-He</td>
</tr>
<tr>
<td>FSC</td>
<td>Colorado, USA</td>
<td>(1970)</td>
<td>330 MW(e)</td>
<td>93% U(<em>{235})+ThC(</em>{2})</td>
<td>Graphite-He</td>
</tr>
</tbody>
</table>

\(^a\) Number quoted in % refer to the \(^{235}\)U content of uranium.
(6) In connection with the forthcoming Agency symposium on the thermo-
dynamic properties of nuclear materials, it is recommended that the
more important thermodynamic properties of thorium-based fuels be
reviewed by an ad hoc committee generated at that symposium. The
properties of interest include the heat capacities, the heats of formation,
the free energies of formation, and other information concerned with
chemical compatibility among reactor materials.

(7) A consulting group should be formed with the task of collecting and criti-
cally evaluating all the data available and currently being generated on
thorium-based fuels. It should be the function in this group not only
to collect and evaluate the existing data but to stimulate the generation
of additional data on well-characterized materials.

(8) Successful development of the fuel cycle technology is of crucial im-
portance in the utilization of thorium. The Agency should, with the co-
operation of various centres in each country or appropriate organization,
accumulate and disseminate available information concerning fabrica-
tion, reprocessing and refabrication of thorium-based fuels. Each
centre could send periodic reports of major activities and developments
to IAEA Headquarters to be consolidated with other similar reports and
then distributed.

(9) It is recommended that the IAEA contribute to processing and fabri-
cation development by dissemination of current information about the
research programmes of Member States. One suggestion for implemen-
tation of this recommendation is that the Agency prepare suitable topical
reports for general distribution by visits to important installations doing
the research.

(10) The Agency should closely follow the progress of various thorium re-
actor systems having good fuel utilization and promote their develop-
ment by holding technical meetings. It would be best to organize a work-
ing group of technical experts from interested Member States to periodi-
cally review progress in thorium technology and utilization and thereby
promote prompt exchange of information and possible international
collaboration in thorium research.
PHYSICS ASPECTS OF DISPERSED FUEL REACTORS CONTAINING THORIA

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AUSTRALIA

1. INTRODUCTION

Nuclear power is well established. The development is now towards better utilization of the nuclear raw materials and economic power costs. The Australian Atomic Energy Commission believes that these aims can be achieved in a high-temperature gas-cooled reactor based on the thorium cycle.

With beryllia as moderator there are good prospects of a high initial conversion ratio and high-temperature operation. Australian work is aimed at a reactor possessing a long reactivity lifetime. Although breeding with thermal reactors has been shown to be possible, it is doubtful whether it could be achieved in practice. Studies of the properties of beryllia under irradiation indicate that its high thermal conductivity and good irradiation stability make it suitable for a homogeneous dispersed fuel reactor concept.

The $^{232}$U-$^{232}$Th cycle was preferred but owing to the difficulty of obtaining $^{233}$U, attention was given to the plutonium-thorium cycle, which had been shown [1] to be of importance in its own right for a small gas-cooled reactor. Both systems have received attention. The studies described in this paper relate to homogeneous dispersions of thorium in beryllia assemblies fuelled by the three main fissile elements in the form of aluminum alloys (30 wt. % fissile material).

An examination of the literature reveals many calculational surveys but little experimental work on homogeneous reactor systems containing thorium (or thorium). The most relevant work is that of Benson and Fox [2] and Mountford et al. [3].

Benson and Fox's measurements were concerned with the critical heights of three graphite-moderated systems having low thorium concentrations (i.e. approximately of the order of the fissile concentration). The Atomics International work relates to low moderator systems with thorium concentrations of the same magnitude as the moderator, which was generally graphite. The moderator ratios were less than 100, with a sole exception, i.e. $^{233}$U/Th/C = 1/25/800. These measurements were two-region critical configurations and yielded much information on the nuclear properties of the particular composition. The results were analysed largely on the basis of the 18-group cross-section data of Mills [4].

2. REACTOR STATICS

No experimental data existed which would allow the nuclear data available for BeO, thorium and the fissile materials ($^{235}$U, $^{233}$U and $^{239}$Pu) to be checked against some integral parameter, e.g. buckling or reactivity.
The present experiments were planned to provide measurements which would check the calculational method and data for assemblies containing appreciable quantities of resonance absorber, namely thorium. Some preliminary measurements on $^{235}$U-BeO assemblies acted as a check on the technique and on the results of Kloeverstrom et al. [5]. The two sets of measurements were in good agreement, considering the compositional and constructional differences.

The beryllia and thoria were mixed to provide a 5 wt.% dispersion of thoria in beryllia. Additives were necessary for the manufacturing stages, and these were later removed by heating at 1000°C. Various additives were tried. The general conclusion was that the better the additive from the pressing characteristic viewpoint, the poorer were the nuclear characteristics. Considerable time was spent to ensure that the reactivity effects of these impurities were small compared to other errors. The material was prepared in a hand press which represented a compromise between nuclear and metallurgical considerations. The dimensions of the compacts were 3 in×3 in×1 in and they were enclosed in aluminum trays 2 ft×2 ft×1.080 in. The top of the tray (0.080 in) had milled slots (2 ft×1.40 in×0.050 in) for holding fuel strips. The characteristics of the fuel strips are given in Table I.

The mean particle size of the thoria was 5 μm (maximum < 20 μm) and density variations did not exceed 5% of the mean density. The main impurity was iron at 150 ppm. Some trays were fitted with $\frac{1}{2}$-in diameter holes parallel to the length of the tray and suitably positioned to avoid harmonic effects, and all trays had two $\frac{1}{2}$-in diameter holes running perpendicular to the long axes. Each assembly consisted of 30 trays stacked together to form a 24 in×24 in×30 in rectangular parallelepiped with the holes running through the trays aligned to form two measuring channels in the vertical direction. Twelve slots were available per layer for the fuel and the various moderator-to-fuel ratios were obtained by changing the fuel pattern.

In some of the $^{233}$U-thorium-beryllia assemblies the above trays were interleaved with layers of beryllia tiles (6 in×6 in×1 in) to change the fertile-to-moderator ratio. Details of the assemblies, compositions and results are given in Tables II and III. The assemblies were built on a graphite plinth situated centrally above a reactor beam hole to provide a plane source (Fig. 1).

Measurements consisted of reaction rates from BF$_3$ and fission chambers (coatings of $^{233}$U, $^{235}$U and $^{239}$Pu) in the x-, y- and z-directions. From these scans, the extrapolated widths of the assemblies were obtained from the x, y results and the relaxation length from the vertical, or z, result.

In the $^{233}$U assemblies, we resorted to two-region exponential assemblies. With only 1 kg of $^{233}$U, the number of assemblies possible was restricted. Experiment showed that by replacing $^{235}$U with $^{233}$U, it was possible to obtain the same fission ratios in a central region of 1 ft×1 ft.
TABLE I

FUEL STRIP CHARACTERISTICS

<table>
<thead>
<tr>
<th>Material</th>
<th>Dimensions (in)</th>
<th>Fuel mass (g)</th>
<th>Fuel in aluminium (wt. %)</th>
<th>Principal isotope (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{233}$U</td>
<td>11.85 x 1.35 x 0.040</td>
<td>4.9</td>
<td>25</td>
<td>99</td>
</tr>
<tr>
<td>$^{239}$U</td>
<td>24 x 1.35 x 0.040</td>
<td>14.5</td>
<td>25</td>
<td>89</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>25 x 1.375 x 0.040</td>
<td>9.5</td>
<td>25</td>
<td>93</td>
</tr>
</tbody>
</table>

FIG. 1. A $^{233}$U-thorium-beryllia assembly
TABLE II

MEASURED BUCKLINGS AND FISSION RATIOS
FOR SUBCRITICAL ASSEMBLIES

<table>
<thead>
<tr>
<th>Composition</th>
<th>$B_m^2$ (m$^{-2}$)</th>
<th>$\delta_{f3}/\delta_{f5}$</th>
<th>$\delta_{f3}/\delta_{f5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$U-Th-BeO</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 8 1625</td>
<td>15 ± 0.4</td>
<td>1.98 ± 0.03</td>
<td>1.15 ± 0.02</td>
</tr>
<tr>
<td>1 12 2437</td>
<td>8 ± 0.15</td>
<td>1.79 ± 0.03</td>
<td>1.07 ± 0.02</td>
</tr>
<tr>
<td>1 24 4814</td>
<td>5.5 ± 0.1</td>
<td>1.71 ± 0.03</td>
<td>1.04 ± 0.02</td>
</tr>
<tr>
<td>1 36 7310</td>
<td>3.6 ± 0.1</td>
<td>1.60 ± 0.03</td>
<td>1.01 ± 0.02</td>
</tr>
<tr>
<td>$^{233}$U-Th-BeO</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 24 4800</td>
<td>5.5 ± 0.3</td>
<td>1.67 ± 0.03</td>
<td>1.14 ± 0.03</td>
</tr>
<tr>
<td>1 12 6747</td>
<td>15 ± 0.8</td>
<td>1.58 ± 0.03</td>
<td>1.00 ± 0.02</td>
</tr>
<tr>
<td>1 9 1800</td>
<td>20 ± 0.7</td>
<td>1.99 ± 0.04</td>
<td>1.21 ± 0.02</td>
</tr>
<tr>
<td>1 12 2402</td>
<td>18 ± 0.7</td>
<td>1.80 ± 0.03</td>
<td>1.14 ± 0.02</td>
</tr>
<tr>
<td>1 6 3374</td>
<td>27 ± 1.0</td>
<td>1.76 ± 0.03</td>
<td>1.09 ± 0.03</td>
</tr>
<tr>
<td>Pu-Th-BeO</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 9 1958</td>
<td>11.6 ± 0.7</td>
<td>2.39 ± 0.04</td>
<td>1.53 ± 0.03</td>
</tr>
<tr>
<td>1 13 2580</td>
<td>13.0 ± 0.7</td>
<td>2.07 ± 0.04</td>
<td>1.22 ± 0.03</td>
</tr>
<tr>
<td>Pu $^{233}$U-Th-BeO</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1/0 2 15 3000</td>
<td>13.5 ± 0.7</td>
<td>2.12 ± 0.04</td>
<td>1.27 ± 0.03</td>
</tr>
<tr>
<td>1/0 5 19 3800</td>
<td>13.8 ± 0.7</td>
<td>2.09 ± 0.04</td>
<td>1.26 ± 0.03</td>
</tr>
<tr>
<td>1/1 26 5220</td>
<td>15.8 ± 0.7</td>
<td>1.93 ± 0.04</td>
<td>1.24 ± 0.03</td>
</tr>
</tbody>
</table>
surrounded by $^{235}$U to make the size 2 ft $\times$ 2 ft $\times$ 30 in, as in a 2 ft $\times$ 2 ft $\times$ 30 in assembly containing only $^{235}$U. This is a useful extension, but requires care in that:

(i) The spectrum in the inner region must reach equilibrium, as in a one-region exponential, and

(ii) More sophisticated analysis is required.

Reaction rates measured perpendicular to the length of the fuel plates showed evidence of fine structure. The calculated value of this structure obtained from transport theory calculations was in good agreement with experiment, and when the data in this direction were corrected, a good fit to a cosine shape was obtained and the extrapolation length agreed with that obtained in the direction parallel to the fuel plates.

Extrapolation length parallel to fuel plates $= 2.00 \pm 0.13$ cm

Extrapolation length perpendicular to fuel plates $= 2.15 \pm 0.12$ cm

3. PARTICLE-SIZE STUDIES

The use of $^{10}$B and $^{240}$Pu as burnable poisons had been proposed for the achievement of long reactivity lifetimes. To this may be added the possibility of using thorium in particulate form. No experimental work had been done on the absorption or reactivity properties of thorium or thoria particles and this problem was studied both experimentally and theoretically.

Experimentally, the reactivity worth of cubes (3.5 cm side) of BeO containing dispersed ThO$_2$ particles was measured in a well-thermalized spectrum with $1/E$ tail for five particle sizes (5, 150, 350, 500 and 700 $\mu$m), and for two moderator ratios (BeO/Th = 100 and 500). At the same time and by the same method, the reactivity worth of ThO$_2$ (<5 $\mu$m) dispersed in BeO was measured as a function of moderator ratio. The results are shown in Figs. 2 and 3. The cubes consisted of hot pressed material and measurements were made with and without cadmium covers.

The measurement technique used was the replacement method where the difference in critical position of a control rod for the reactor with and without the specimen represented the reactivity worth of the sample.

The worth of ThO$_2$ as a function of particle size was taken with the samples under cadmium covers to avoid the thermal part of the absorption. The results given are those obtained after the subtraction of the worth of a beryllia sample.

The theoretical approach used [6] differs from that of Lane et al. [7] published while this work was in progress. It treats the particle as the fundamental and finds an equivalent cross-section for it such that homogeneous resonance absorption theory applies.

The effective resonance integral can thus be written

$$ I = \sigma_p \int \frac{\sigma_a}{\sigma + \sigma_p} \frac{dE}{E} $$
<table>
<thead>
<tr>
<th>No.</th>
<th>(^{235}\text{U})</th>
<th>(^{238}\text{U})</th>
<th>(\text{Al})</th>
<th>(\text{O})</th>
<th>(\text{Be})</th>
<th>(^{232}\text{U})</th>
<th>(^{239}\text{Pu})</th>
<th>(\text{Th})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-8-1625</td>
<td>(2.1 \times 10^{-5})</td>
<td>(2.3 \times 10^{-6})</td>
<td>(4.4 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-12-2437</td>
<td>(1.4 \times 10^{-5})</td>
<td>(1.5 \times 10^{-6})</td>
<td>(4.2 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-24-4874</td>
<td>(7.1 \times 10^{-6})</td>
<td>(7.6 \times 10^{-7})</td>
<td>(3.95 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-36-7310</td>
<td>(4.75 \times 10^{-6})</td>
<td>(5.1 \times 10^{-7})</td>
<td>(3.9 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-24-4800</td>
<td>-</td>
<td>-</td>
<td>(3.98 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(7.2 \times 10^{-6})</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
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<td>-</td>
<td>-</td>
<td>(2.25 \times 10^{-3})</td>
<td>(5.1 \times 10^{-2})</td>
<td>(5.1 \times 10^{-2})</td>
<td>(7.58 \times 10^{-4})</td>
<td>-</td>
<td>(8.9 \times 10^{-5})</td>
</tr>
<tr>
<td>1-9-1800</td>
<td>-</td>
<td>-</td>
<td>(4.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(1.9 \times 10^{-5})</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-12-2402</td>
<td>-</td>
<td>-</td>
<td>(4.3 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(1.4 \times 10^{-5})</td>
<td>-</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-6-3374</td>
<td>-</td>
<td>-</td>
<td>(2.6 \times 10^{-3})</td>
<td>(5.1 \times 10^{-2})</td>
<td>(5.12 \times 10^{-2})</td>
<td>(1.51 \times 10^{-5})</td>
<td>-</td>
<td>(8.9 \times 10^{-5})</td>
</tr>
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<td>1-9-1958</td>
<td>-</td>
<td>-</td>
<td>(4.5 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>(1.8 \times 10^{-5})</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1-13-2580</td>
<td>-</td>
<td>-</td>
<td>(4.3 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>-</td>
<td>(1.35 \times 10^{-5})</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1/0.2-15-3000</td>
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<td>-</td>
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<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
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<td>1/0.5-19-3800</td>
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<td>-</td>
<td>(2.6 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(4.5 \times 10^{-6})</td>
<td>(9.2 \times 10^{-6})</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
<tr>
<td>1/1-26-5220</td>
<td>-</td>
<td>-</td>
<td>(2.3 \times 10^{-3})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(3.5 \times 10^{-2})</td>
<td>(7.0 \times 10^{-6})</td>
<td>(6.7 \times 10^{-3})</td>
<td>(1.7 \times 10^{-4})</td>
</tr>
</tbody>
</table>
DISPERSED FUEL REACTORS CONTAINING THORIA

FIG. 2. Reactivity of ThO$_2$ as a function of ThO$_2$ particle size (BeO/ThO$_2$ = 100)

where $\sigma$, $\sigma_a$ = total and absorption cross-sections of the thorium atom, respectively

$$\sigma_p = \frac{\sigma_m \sigma}{\sigma_e}$$

$$= \frac{\sigma_m}{1 + aN D \sigma_m}$$
where

\[ 3/8 < a < 2/3 \]

\[ D = \text{particle diameter} \]

\[ \sigma_e = \text{effective cross-section of the thorium atom} \]

\[ \sigma_m = \text{scattering cross-section of the moderator per thorium atom} \]

\[ L = \text{chord length} \]

\[ N = \text{number of thorium atoms per cubic centimetre of the particle} \]

The limits on 'a' correspond to the cases where Nier is small and large, and where for spherical particles \[ a = 2D/3 \] and \[ a^2 = D^2/2. \]

Good agreement has been found between this theory, using \( a = \frac{1}{2} \), and experiment for the case of \( \sigma_m = 1000 \) b. The self-shielding for 50- and 100-\( \mu \)m particles is 0.97 and 0.95. The theory can easily be incorporated in multi-group cross-section data and is useful in burn-up analysis, since to first approximation \( N_0 \) is constant.

It is hoped to examine the effect of particle size on plutonium absorption, with particular respect to its \( ^{240} \text{Pu} \) content.

4. PARTICLE-SIZE EFFECTS

4.1. Dynamics

The plutonium-thorium-beryllia reactors studied have inconveniently large negative temperature coefficients of the order of \( 1.0 \times 10^{-4} \text{\Delta k/\text{deg C}} \). The use of 200-\( \mu \)m particles rather than a homogeneous system changes the coefficient by 10\%, although some uncertainties remain in the treatment of \( ^{240} \text{Pu} \). In addition, the temperature coefficient decreases during the core lifetime to about half its initial value, owing to plutonium burn-up.

Some preliminary analogue computer studies \[9\] have examined the influence of the fuel dispersion on transients. The model considered was a one-point reactor with one group of delayed neutrons containing a beryllium-based dispersed fuel of

(i) \( \text{PuO}_2\text{-ThO}_2 \) particles of 50-, 200- and 500-\( \mu \)m diameter, and

(ii) Separate \( \text{PuO}_2 \) particles (50- and 200-\( \mu \)m diameter) and \( \text{ThO}_2 \) particles.

The parameters recorded were power, mean fuel and moderator temperature, and reactivity, all as functions of time. Typical results are shown in Figs. 4 and 5 and the more important parameters in Table IV.

The 50-\( \mu \)m \( \text{PuO}_2\text{-ThO}_2 \) case behaves as a homogeneous core and as the particle size increases the maximum power and energy release decreases. However, the larger particles give rise to higher fuel/moderator temperature differences and are at least as important as the peak power in determining the stresses involved. The general conclusion after examining all the effects was that a 200-\( \mu \)m particle probably represented the best compromise. It should be noted that energy released after the peak power does not necessarily insert more negative reactivity because the rise in
TABLE IV

ENERGY RELEASED FROM THE ADDITION OF 1% $\frac{\Delta k}{k}$ IN 1 s STARTING AT POWER OF 10 kW

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PuO$_2$ particles</th>
<th>PuO$_2$-ThO$_2$ particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\frac{\Delta k}{k}) fuel</td>
<td>- 4.5 x 10^{-5}</td>
<td>- 1.1 x 10^{-5}</td>
</tr>
<tr>
<td>(\frac{\Delta k}{k}) moderator</td>
<td>- 9.5 x 10^{-5}</td>
<td>- 3.0 x 10^{-5}</td>
</tr>
<tr>
<td>Particle size (\text{mm})</td>
<td>200 50</td>
<td>50 200 500</td>
</tr>
<tr>
<td>Heat-transfer coeff (MW/degC)</td>
<td>1.5 x 10^2 2.4 x 10^6</td>
<td>3 x 10^4 2 x 10^3 3 x 10^2</td>
</tr>
<tr>
<td>Energy release (MW s)</td>
<td>8 x 10^3 1.2 x 10^3</td>
<td>1.2 x 10^6 9 x 10^3 7 x 10^5</td>
</tr>
</tbody>
</table>

Moderator temperature may not provide enough reactivity to compensate for the change in fuel temperature and underlines the importance of having the major part of the coefficient derived from the fuel.

4.2. Radiation damage

Hickman [10] has discussed this problem and showed that for a largely undamaged matrix, the dispersed phase must be limited to less than 30 vol.% for 200 \text{mm} particles on the assumption that fission-product recoil range is 10 \text{mm} in PuO$_2$ and UO$_2$, and 15 \text{mm} in BeO. The amount of fission products escaping from a 100-\text{mm} particle is \sim 10%.

The conclusions to be drawn are that particle size should be between 100 and 200 \text{mm}. An irradiation programme aimed at confirming this reasoning is in progress but no firm results are yet available.

4.3. Mechanical strength

Stress analysis of unirradiated dispersed fuel [11] in a beryllia matrix suggests that 30-\text{mm} cracks will propagate in a body whose strength is 15 000 lb/in$^2$ and that this should be an upper limit to the particle size.

It can thus be seen that the particle size of dispersed fuel arrangements is a compromise between various factors. For the present it is intended to retain the particle size in the region of 100-200 \text{mm}. 


5. DATA AND ANALYSIS

A digital computer (IBM 7040) became available recently and an attempt was made to analyse the experimental results using the codes DSN [12] and CRAM [13] and the multigroup data of Mills [4].

Mills' data was developed in 18-group form for intermediate systems in particular and was found unsatisfactory as it stood. Initial results produced much smaller critical systems than experiments showed. This resulted from the absence of moderator upscattering in groups 15-18 and was indicated by a large thermal peak in group 18. The experimental scattering law $S(\alpha, \beta)$ for BeO [14] was used to obtain group scattering cross-sections from PIXSE and LEAP. The given beryllia data were modified to accommodate this new data by keeping $\alpha_f$ constant. This resulted in critical sizes closer to those predicted by the experiments.

With this modification, an analysis of some simple fuel moderator assemblies was made. Agreement for the $^{235}$U-beryllia systems was good,
FIG. 5. Reactivity-time behaviour following the addition of 1% Δk/k in 1 sec (initial power 10 kW)

taking $\psi_{235} = 2.42$ rather than 2.45 while that for the $^{233}$U was unsatisfactory (see Table V).

Considerable effort has been devoted to examination of the $^{233}$U cross-section data, but with no useful outcome so far. Analysis is incomplete and awaits the development of codes capable of generating spectra and using these spectra as weighting functions in the derivation of multigroup cross-sections.

Thorium data are not included in Mills' results and we will shortly be constructing such data based mainly on Buckingham and Parker [15] and on the resonance parameters of Uttley and Jones [16]. The latter proved successful in analysis of the particle-size studies.

6. CONCLUSIONS

The work reported leaves most of the questions on the advantages of the thorium cycles unanswered. This is realized. However, the measurements...
TABLE V

COMPARISON OF CALCULATED AND EXPERIMENTAL $B_m^2$ FOR SIMPLE FUEL/MODERATOR EXPONENTIAL

<table>
<thead>
<tr>
<th>$^{235}$U/BeO</th>
<th>$B_m^2$ (m$^{-2}$) Calc.</th>
<th>$B_m^2$ (m$^{-2}$) Expt.</th>
<th>$^{233}$U/BeO</th>
<th>$B_m^2$ (m$^{-2}$) Calc.</th>
<th>$B_m^2$ (m$^{-2}$) Expt.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1460</td>
<td>60</td>
<td>54.9 ± 2</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2920</td>
<td>47</td>
<td>45 ± 1.5</td>
<td>3266</td>
<td>49</td>
<td>47 ± 1</td>
</tr>
<tr>
<td>5850</td>
<td>36</td>
<td>33 ± 1</td>
<td>4340</td>
<td>43</td>
<td>39 ± 1</td>
</tr>
<tr>
<td>8760</td>
<td>27</td>
<td>23 ± 1</td>
<td>6530</td>
<td>33.5</td>
<td>34 ± 1.5</td>
</tr>
</tbody>
</table>

do allow some investigation of the value of the data used in surveys, and future measurements of the conversion ratio of some of the thoria-loaded assemblies and of the activities associated with fuel burn-up and fission products will be made in two-region critical assemblies.

The Mills' cross-section set is not adequate for the more thermalized systems.

Not enough analysis has been made using the resonance data of Uttley but early indications suggest that it may be adequate.

ACKNOWLEDGEMENTS

Much of this work would not have been possible without the assistance of Messrs. Bardsley, Wright, and Silver of the Materials Division. The $^{233}$U work was made possible by the loan of $^{233}$U from the USAEC.

REFERENCES

DISPERSED FUEL REACTORS CONTAINING THORIA


1. INTRODUCTION

1.1. General

This paper presents preliminary results on possible advantages of thorium reactors in a nuclear power programme for South Central Brazil (Fig. 1). This programme is based on a more detailed study of power market requirements for the region. It represents part of a more extensive work that is being made on plutonium and thorium cycles and directed to the evaluation of fuel requirements for the Brazilian Nuclear Power Programme over a period of 60 yr. Projects over such a long-range period were necessary, since in most cases significant differences among various alternatives studied appeared only over an extended period, where equilibrium was met between plutonium or uranium-233 production and the fissile material requirements for the programme.

Growing uranium market restrictions could be expected in the West from the end of the 1970s, when, in particular, the present-known low-cost reserves in the USA and Europe would be exhausted [1]. At that time, countries that depend on the world market to meet their uranium requirements would be constrained "to buy all their uranium under unfavourable economical conditions, subject to political hazards" [1].

Uranium prospecting in Brazil is in its early stage; even if no significant uranium reserves are known at present, new discoveries could be expected. Whatever the results of present prospective work, it seems clear that Brazil must use its uranium reserves in the best way, directing its nuclear policy towards independence of foreign nuclear fuel supplies over a long range. The efficiency of such a policy could be evaluated by two fundamental parameters: the amount of basic natural uranium requirements and the time required to reach a self-sustained balanced economy on nuclear fuel.

1.2. Pu-$^{238}$U cycle

The first step in the study was an evaluation of various programmes to meet demands for the Pu-$^{238}$U cycle. The cases studied included utilization of graphite-gas, light-water or heavy-water start-up converter re-
actors, with "once-through" fuel burn-up, plutonium recycling in the same reactors or the use of plutonium in fast reactors (with or without excess plutonium recycling in converters).

The results are shown in Fig. 2. "Once-through" results are not discussed here. The results obtained show that such a programme is not worthwhile because of the enormous fuel consumption for any of the three reactor types referred to above. Plutonium recycling would result in a great saving of natural uranium, but the resulting total consumptions are still too large. In all cases it is necessary to go on to advanced breeders to keep uranium consumption at reasonable levels. Results obtained by the combination of converters and fast plutonium breeders seem compatible with a programme expected to use Brazilian fuel.

2. THORIUM CYCLE POSSIBILITIES

2.1. General

Because of Brazil's resources of thorium ore it is of particular interest to study the utilization of thorium in a power programme to verify if a
Studies were less detailed for the thorium cycle than for the plutonium, because the various reactor parameters involved are not easily found in the literature. Two start-up reactors were considered, both D$_2$O-moderated; the thorium breeders' doubling time was adopted as a parameter. In the thorium cycle such reactors have a thermal or near-thermal spectrum while in the plutonium cycle the spectrum is fast. Results are compared with those obtained for the plutonium cycle with natural-uranium heavy-water E1-4 type start-up reactors (which, in particular, leads to minimum uranium consumption in the plutonium cycle).
2.2. Reference reactors

For the thorium cycle, start-up reactors and equilibrium reactors (breeders) are of D₂O type. Two concepts of D₂O-thorium reactors were considered to initiate the thorium cycle: (i) Du Pont de Nemours concept, referred to here as DN [2] and (ii) the under-moderated D₂O reactor concept which is being studied at the Comissão Nacional de Energia Nuclear, referred to here as SM¹.

Doubling time of the breeder reactors was varied in the same way in both cases. There is, however, a difference between them: for the DN case, breeder reactors are the same as start-up reactors, with ²³³U recycling, while for the SM case breeder reactors are different from start-up reactors. In both cases highly enriched ²³⁵U fuel was assumed for start-up.

2.3. Parameters

Physical parameters used in each case were the following:

DN reactor concept²

(a) 0.800 t natural uranium/MW(e) necessary for each reactor to achieve equilibrium.
(b) Two hypothetical times (7 and 10 yr) were assumed for the reactor to reach equilibrium³.

SM reactor concept

(a) Fuel inventory: 0.500 t/MW(e)
(b) Natural uranium consumption: 0.160 t/MW(e) yr
(c) Diffusion plant tail enrichment: 0.30%
(d) ²³³U net production: 0.600 kg/MW(e) yr

The immobilization assumed for the plutonium breeder reactor was 4 kg Pu(f)/MW(e). For the thorium breeder reactor this was 2 kg ²³³U/MW(e) divided into 1.5 kg charge and 0.5 kg cooling, reprocessing and refabricating. This value can be justified if it is assumed that the lifetime of the reactor charge is 900 d, corresponding to 3 yr of reactor operation with a load factor of 80%. A 1-yr delay was assumed for a fuel cycle outside the reactor.

The specific power of thorium cycle reactors was 10 MW(e)/t (Th). A waiting time of 12 yr was admitted for thorium to permit decay of ²²⁸Th and its descendants, which results in a thorium immobilization of 0.4 t/MW(e). This hypothesis is pessimistic, and was deliberately used in view of obtaining maximum values to compare with Brazilian thorium reserves (upper curve, Fig. 6). Thorium requirements would be substantially decreased

¹ The studies are in a preliminary phase. The parameters used in this paper are subject to corrections as the studies progress.
² Other data on this type may be found in Ref. [2].
³ The reference quoted does not give any information about delay to reach equilibrium. Hypotheses assumed resulted from brief calculations.
if appropriate reprocessing and fabrication methods could be used; a thorium immobilization of 0.14 t/MW(e), which corresponds to an "out-of-reactor" time of 1 yr, has been considered also (lower curve, Fig. 6).

Technological development would depend on the conditions of the thorium market. Probably thorium consumption will be given at first by the upper curve and will approximate the lower one as the thorium requirements increase.

The following ranges were chosen for doubling time: (a) 5 to 15 yr for fast breeders of the plutonium cycle, and (b) 15 to 25 yr for thermal breeders of the thorium cycle. These two ranges are believed to be comparable. Values between 5 and 15 yr are commonly accepted for fast plutonium breeders. For thermal $^{233}$U breeders the values 15-25 yr are based on breeding possibilities that are being investigated in another study.

Preliminary curves on the dependence between doubling time, breeding factor and fuel immobilization are shown in Fig. 3. In these curves it is seen that 15-25 yr is a possible range in agreement with the characteristics of thorium-fuelled thermal reactors. The expression used for doubling time is valid if the total installed output as well as annual rate of reactor installation are high.

The immobilization comprises fuel charge and fuel outside the reactor. The breeding factor is defined as

$$b = 1 + \frac{C_f - C_i}{C_q}$$

where

- $C_f = \text{final concentration of fissile material in irradiated fuel}$
- $C_i = \text{initial concentration of fissile material}$
- $C_q = \text{fissile material burned during irradiation}$

2.4 Results

Accumulated uranium consumption is represented for each case (Fig. 4). Assumed doubling times limit the range of each hypothesis. During the first 25 yr consumption is not very different. After this, the range in each case is enlarged with the following tendency: the DN-type for the thorium cycle reactor is near to the most pessimistic hypothesis for the plutonium cycle, while the SM-type is near to the optimistic one.

Figure 5 shows the total power in the converter for the thorium cycle. Note in this respect the strong influence of equilibrium time, $T_e$.

Figure 6 represents thorium consumption. It is assumed to be the same for all cases.

3. CONCLUSIONS

The results summarized above lead to the following general conclusions:

1. The thorium cycle has good prospects in the Brazilian Nuclear Power Programme. Indeed, if start-up reactors are similar to the heavy-water
under-moderated, SM reference reactor, total natural uranium requirements and the extent of the uranium-dependent, start-up programme could be as low as in the most optimistic assumptions under the Pu-238U conditions (natural-uranium heavy-water start-up reactors, highly efficient plutonium fast breeders).

(2) The technology involved in the development of the SM concept is basically conventional natural-uranium heavy-water-reactors technology which is simpler than fast or other advanced reactors technology.

(3) This reactor concept offers Brazil the possibility to develop some original ideas on nuclear power plant technology, aiming at a national programme that could be independent of a foreign supply of nuclear fuel in a reasonably short time.

(4) The results obtained so far are very preliminary and must be confirmed by more intensive work on reactor physics and technology and on chemical and metallurgical problems in thorium fuel cycles, before an enlarged and more objective programme can be defined.

(5) In all cases, a coherent programme on research and development and
on nuclear power production, cannot be realized before domestic uranium and thorium resources are estimated to some degree of confidence.

Aware of these results, CNEN is actively considering the formulation of a 12-month programme to prepare a more objective long-range research and development programme on thorium reactors, including:

1. Assessment of known Brazilian thorium reserves and intensive uranium ore prospecting
2. Studies on thorium reactors, including feasibility studies on the SM concept, to be compared with those on other thorium reactors at present being developed in other countries
3. Studies and laboratory research on fabrication and reprocessing of thorium fuel elements
4. Experimental facilities for thorium reactor research (foreign experience and Brazilian possibilities).

ACKNOWLEDGEMENTS

We are grateful to Professor Borisas Cimbleris for his helpful discussions and suggestions during the preparation of the present paper, as well as the Brazilian Nuclear Energy Commission and Instituto de Pesquisas Radioativas for the permission to publish this paper.
FIG. 5. Installed power in natural uranium consumer reactors

FIG. 6. Thorium requirements
REFERENCES


BIBLIOGRAPHY


APPENDIX

BRAZILIAN THORIUM RESOURCES

Brazilian thorium occurrences are of two types: monazite sand beach placers and inland deposits.

\[
\begin{array}{l|c}
\text{Sample} & \text{Tho}_2\text{O}_2 \text{ t} \\
\hline
\text{Monazite sands} & \sim 15000 \\
\text{Pyrochlore of Araxa} & \sim 130000 \\
\text{Bastnasite of Morro do Ferro} & \sim 37000 \\
 & \sim 182000 \\
\end{array}
\]

Other occurrences not yet evaluated are São João del Rey, Serra Negra, Tapira, etc. [3].

The present-day exploration is restricted to monazite sands. This exploration is being done by Orquima, aided by the Government, and is directed toward rare earth salts and thorium oxide production.

The present production capacity is 300 t ThO₂/yr and the Comissão Nacional de Energia Nuclear stocks are about 1000 t. This number is quite sufficient for the first years of a power programme.
A PRELIMINARY ASSESSMENT OF THORIUM
AS A FUEL FOR THERMAL REACTORS

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ATOMIC ENERGY OF CANADA LIMITED, CHALK RIVER,
ONTARIO, CANADA

INTRODUCTION

Nuclear power is growing rapidly. In Canada nuclear power is generated by irradiating natural uranium as long as the fuel reactivity will allow. The spent fuel is stored. This fuel contains about 0.4% plutonium, of which about 69% is fissile. To indicate the 'real' worth (as opposed to the economic value) of this fuel in terms of familiar numbers we note that CANDU, a 200-MW(e) station produces about 90 kg of plutonium a year. About 70% of this plutonium, if separated, can be fissioned in a single further irradiation of about 3 n/kb. If the plutonium burn-up rate is 90 kg/yr it will produce (at a station efficiency of 30%) a further 60 MW(e) of power, i.e. 30% of the reactor power producing it. If this plutonium is used to prolong the irradiation of natural uranium to 3 n/kb the total power obtained by burning 90 kg/yr is 160 MW(e) greater than would be obtained by burning natural uranium alone. The importance of plutonium to the overall power programme will thus depend on how it is used.

The real problem of course is economic and other fuels may compete very effectively. In this report we compare various combinations of the fissile materials $^{235}$U, $^{233}$U, plutonium and the two fertile materials $^{238}$U and $^{232}$Th in thermal reactors having a neutron economy similar to that of CANDU. We assume that fuel burn-up is limited only by reactivity. The basis for this comparison is the fuel cost so that attention must be paid to factors affecting this cost such as the manufacturing cost, interest on fuel inventory and processing costs. The operating fuel cost depends on manufacturing cost and the burn-up. The inventory charges depend on the manufacturing cost and the fuel rating. The processing contribution to power cost depends on the concentration of fissile material in the fuel at the end of the irradiation and its value which is determined to some extent by its radioactivity and the consequent handling difficulties. The interrelation of these costs with reactor parameters is shown schematically in Fig. 1. (All costs in this report are in US dollars).

A single once-through fuel cycle followed by processing has been considered. The effect of further recycling is taken into account by ascribing a value to the irradiated fuel.

METHOD OF CALCULATION

Reactivity

Changes in fuel composition with irradiation are calculated in the burn-up programme BOT. A thermal reactor with cross-sections specified using
the Westcott formulation is assumed. For the uranium cases the spectrum parameters chosen have been based roughly on the spectrum in the Douglas Point CANDU reactor. For the thorium cases three $^{235}$U concentrations were run on LATREP for a 37-element rod design having roughly twice the fuel rating (MW/kg) of the Douglas Point Reactor fuel in order to form estimates of r-value and neutron temperature. The 2% and 2.5% enrichment results are plotted in Fig. 2 as a function of irradiation. BOT assumes a constant r-value so the value 0.09 was chosen as being representative of the major part of the irradiation. This value is perhaps a little low for the more highly enriched cases but will not affect the overall picture. The neutron temperature was taken to be 175°C and the standard flux $5 \times 10^{13}$ n/cm$^2$ s, although thorium has been considered in a range of fluxes for comparison.

Irradiation of fissile and fertile material is computed independently and normally these results can be added in any ratio to correspond to a given enrichment. However, when plutonium is used to extend the burn-up of natural uranium the $^{240}$Pu effective cross-section will depend on whether the plutonium enrichment is intimately mixed with natural uranium or is in the form of spikes. This occurs because of the large amount of resonance absorption in $^{240}$Pu and its dependence on the self-shielding occurring in this resonance. We have assumed that the enrichment is introduced in such a way that irradiations of fissile and fertile materials may be calculated independently.

The reactor in which this burn-up occurs is characterized by $\bar{\eta}$, a number which indicates how much greater than the total neutron absorption the total neutron production must be to provide for leakage, reactivity control and neutron losses in structural materials. From the calculated fuel properties for a given irradiation the enrichment required to make the reactor critical during this irradiation can be determined. Thus if a fuel is to be irradiated
FIG. 2. Average r-value and fuel temperature in W.B.L. 37 element clusters

to an irradiation \(\phi\) and the integrated neutron yield and absorption during this irradiation in the fertile materials are \(Y_s\) and \(A_s\) and in the fissile materials \(Y_f\) and \(A_f\), then

\[
\frac{Y_c + NY_f}{A_s + NA_f} = \eta
\]

is an equation which can be used to determine \(N\), the required enrichment. If the fertile and fissile materials are placed in different fluxes, their irradiation proceeds at different rates but the equation is still true providing the proper integrals are used. The BOT programme has been incorporated into a control programme which calculates the enrichment required for a given irradiation and makes an estimate of the fuel costs.

**Cost of fissile materials**

\(235\text{U}\)

The cost of \(235\text{U}\) depends on how it is to be used. As a starting point we use prices quoted in the United States of America for uranium hexafluoride from a diffusion plant. The cost per gram of \(235\text{U}\) is $12.03 if it is bought in the form of 93% enriched material. At lower enrichments the cost per gram of \(235\text{U}\) is lower but the cost of converting the hexafluoride to \(\text{UO}_2\) becomes more important so that the cost reaches a minimum of about $10.61/g at an enrichment of about 2.2% [2]. For enrichments lower than
this it pays to mix this material with Canadian natural uranium oxide. For higher enrichments material from the diffusion plant is cheaper. The resulting cost per gram of $^{235}$U as a function of enrichment is plotted in Fig. 3. For thorium fuels only mixtures of thorium and 93% $^{235}$U have been considered, although there might be an economic advantage in using lower enrichments.

$^{233}$U, plutonium

Since these fuels are obtained only by irradiating other materials their cost cannot be obtained without reference to the nuclear power complex which is producing them. This will vary from one country to another, it may include fast and thermal breeders, and it will certainly change with time as experience is gained with handling reactor fuels. There may be some degree of economic 'insulation' between different countries however, each country preferring to develop an integrated system of producing and using fissile materials internally rather than depending on a foreign source of fissile material to expand or change its power programme. Thus, 'costs' of fissile material may be expected to vary from one country to another and will reflect the line of reactor development chosen. In Canada it seems reasonable to estimate the relative worths of fissile materials by adding small amounts of each to natural uranium and comparing the resulting incremental burn-up.

The regular BOT programme without its control programme was used for this purpose. The first case run (for comparison purposes) was natural uranium in a reactor with an $\eta$ of 1.06. The burn-up obtained was 8400 thermal MWd/t so that this reactor is not very different from the Douglas Point reactor in neutron economy. Subsequent cases were run on BOT with
compositions of $1 \text{ } ^{235}\text{U} + 137.8 \text{ } ^{238}\text{U} + 0.1X$ where $X$ represents an atom of $^{233}\text{U}, \text{}^{235}\text{U}$ or plutonium of various isotopic compositions. These compositions are the result of irradiating natural uranium to various irradiations. The incremental burn-ups obtained are plotted in Fig. 4 as a function of the irradiation of the natural uranium from which the plutonium is extracted and compared to that obtained for the pure fissile isotopes $^{233}\text{U}$ and $^{235}\text{U}$. Two sets of curves are shown. The lower curve (curve (a)) which refers to 0.1 atom of plutonium has been divided by the fractional fissile content to determine the burn-up increment per fissile atom of plutonium, plotted in the same figure (curve (b)). The neutron spectrum in which the plutonium is fissioned was characterized by the parameters $T_n = 150^\circ \text{C}$ and $r = 0.07$ which one would expect to find in a well-thermalized reactor spectrum. Reasonable changes in this spectrum will have only a small effect on the result.

Also shown in Fig. 4 are the effects of (i) storing the uranium fuel for 10 yr before reprocessing and using it (curve (c)) and (ii) processing after a 5-yr storage and then storing for a further 5 yr before using it (curve (d)). In the first case the plutonium becomes poorer because of the loss of $^{241}\text{Pu}$ during the storage interval. In the second case, in addition to the loss of $^{241}\text{Pu}$, there is 5-yr worth of $^{241}\text{Am}$ in the fuel, adding to the parasitic absorption.

For $^{235}\text{U}$ and $^{233}\text{U}$ the incremental burn-ups are 2295 and 2756 thermal MWd/t. The incremental burn-up per fissile plutonium atom is very nearly constant at about 2300 MWd/t (see Fig. 4). Assuming as a first approximation that the relative worths of the three fissile materials are proportional to the incremental burn-up obtained and assigning a value of $\$12.03$ to pure $^{235}\text{U}$ we obtain:

\[
\begin{align*}
^{235}\text{U} & \text{ } \$12.03/\text{g} \\
^{233}\text{U} & \text{ } \$12.03 \times \frac{2756}{2295} \text{ } = \text{ } \$14.45/\text{g} \\
\text{Plutonium} & \text{ } \$12.03 \times \frac{2300}{2295} \text{ } = \text{ } \$12.06/\text{g \text{ of fissile plutonium}}.
\end{align*}
\]

The United States' guaranteed prices \[3\] per gram of $^{233}\text{U}$ and fissile plutonium are $\$14.00$ (less a penalty depending on the $^{232}\text{U}$ content) and $\$10.00$. In practice the plutonium should be somewhat less valuable than estimated above since usually some $^{241}\text{Pu}$ will be lost and some $^{241}\text{Am}$ will grow in during the interval between removal from one reactor and insertion in another. Plutonium is also somewhat more costly to handle than $^{235}\text{U}$ because of its toxicity and $\alpha$-activity. The difference between $\$12.06$ and $\$10.00$ may well reflect the American experience with the handling difficulties associated with plutonium. Similar considerations apply to $^{233}\text{U}$. The difficulties in handling $^{233}\text{U}$ arise partly from its $\alpha$-activity but mostly from the hard $\gamma$-rays from daughter products of $^{232}\text{U}$.

Fast neutrons produce a $^{231}\text{Th} \text{(n, } 2\text{n})^{231}\text{Th}$ reaction. Depending on the hardness of the reactor spectrum the equivalent thermal cross-section lies between 10 and 50 mb. $^{232}\text{Th}$ decays with a half-life of 25.6 h to $^{231}\text{Pa}$. The
cross-section for the reaction $^{231}\text{Pa} (n, \gamma) ^{232}\text{Pa}$ is about 200 b and $^{229}\text{Pa}$ decays with a half-life of 31.7 h to the comparatively stable but $\alpha$-active $^{232}\text{U}$. If the $^{232}\text{U}$ capture cross-section is taken to be 100 b and the short half-life of its precursors and the burn-out of $^{232}\text{Th}$ are neglected, the $^{232}\text{U}$ concentration at an irradiation $\omega$ is simply $^{232}\text{U} = 200 \left( 1 - e^{-\omega/10} \right)^2 \text{ ppm } ^{232}\text{Th}$. We have assumed here a relatively thermal spectrum where the (n, 2n) reaction has a cross-section of only 20 mb.

At irradiations which are likely to be of interest, i.e. in excess of $3 \text{n/kb}$ the total uranium concentration in thorium has reached an equilibrium value of about $1.4 \times 10^4 \text{ ppm}$. After processing to extract uranium from irradiated thorium the $^{232}\text{U}$ contamination of uranium will be $^{232}\text{U} = 14000 \left( 1 - e^{-\omega/10} \right)^2 \text{ ppm } ^{232}\text{U}$.

Table II lists the penalty due to handling difficulties for $^{233}\text{U}$ contaminated with $^{232}\text{U}$. As can be seen from Table I the $^{232}\text{U}$ concentration at irradiations of $4 \text{n/kb}$ is high enough so that the maximum handling penalty will be incurred. A higher cross-section for the $^{232}\text{Th} (n, 2n) ^{231}\text{Th}$ cross-section will lead to higher concentrations of $^{232}\text{U}$ and the maximum penalty will be incurred at lower irradiations. Anticipating results to some extent we have assumed the full penalty for $^{232}\text{U}$ contamination.

From these considerations we adopt the following price scales for the fissile materials. As raw materials in manufacturing thorium fuel elements; $^{233}\text{U}$, $^{235}\text{U}$ and fissile plutonium are assumed to cost $14.00, $12.00 and $12.00 per gram, respectively, as derived from their relative worths in improving the burn-up of natural uranium. In determining the credit to be allowed for spent fuel we deduct $2.00 from the above values for $^{233}\text{U}$ and plutonium to allow for handling difficulties, and use $12.00, $12.00 and $10.00.

Note that when a value is given to spent fuel, residual $^{233}\text{U}$ or $^{235}\text{U}$ in $^{238}\text{U}$ must be discounted since it will not be recovered in a chemical process.
TABLE I

232U CONCENTRATION AT VARIOUS IRRADIATION LEVELS

<table>
<thead>
<tr>
<th>Irradiation (n/kb)</th>
<th>232U in ppm of equilibrium uranium</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>130</td>
</tr>
<tr>
<td>2</td>
<td>460</td>
</tr>
<tr>
<td>3</td>
<td>940</td>
</tr>
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<td>4</td>
<td>1520</td>
</tr>
<tr>
<td>5</td>
<td>2170</td>
</tr>
<tr>
<td>6</td>
<td>2850</td>
</tr>
</tbody>
</table>

TABLE II

PENALTY FOR HANDLING CONTAMINATED 233U

<table>
<thead>
<tr>
<th>232U in ppm of uranium</th>
<th>Penalty (US $)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.40</td>
</tr>
<tr>
<td>20</td>
<td>0.60</td>
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<tr>
<td>45</td>
<td>0.80</td>
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<td>80</td>
<td>1.00</td>
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<tr>
<td>130</td>
<td>1.20</td>
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<tr>
<td>190</td>
<td>1.40</td>
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<tr>
<td>250</td>
<td>1.50</td>
</tr>
<tr>
<td>350</td>
<td>1.60</td>
</tr>
<tr>
<td>500</td>
<td>1.70</td>
</tr>
<tr>
<td>700</td>
<td>1.80</td>
</tr>
<tr>
<td>1000</td>
<td>1.90</td>
</tr>
<tr>
<td>1500 and above</td>
<td>2.00</td>
</tr>
</tbody>
</table>

In processing thorium fuel all uranium isotopes are recovered together with any plutonium which may have been used as enrichment.

Processing

The cost of processing irradiated uranium would at present be prohibitively high in Canada owing to the small throughput. In the United Kingdom where several reactors are operating to relatively low burn-ups the throughput
is sufficiently high to make processing economic and presumably this state of affairs will exist in Canada in the 1970s. The processing cost of $25/kg selected for this study is based on estimates of processing cost by Nuclear Fuel Services [4]. The United Kingdom processing may be even lower. An extra dollar was added for processing thorium since slight additional complications are expected in the chemistry of the extraction process. The processing costs for uranium and thorium were therefore taken to be $25.00/kg and $26.00/kg, respectively, though comparison cases were run with this reduced to zero.

Losses in processing have been neglected except that 98% of $^{233}\text{Pa}$ has been assumed to decay to $^{233}\text{U}$ before processing. $^{239}\text{Np}$ is assumed to decay completely to $^{239}\text{Pu}$ and decay of $^{241}\text{Pu}$ is neglected though this last effect could reduce the worth of the plutonium by $1/g$ if there is a 5-yr lapse between withdrawal of irradiated fuel and manufacturing new fuel and a further 5-yr lapse before irradiation of this fuel begins (Fig. 4).

**Manufacturing costs**

The method used for estimating manufacturing costs is not accurate but serves to point out one of the major problems likely to be encountered when using thorium as a fuel.

Estimates of manufacturing costs have been made as a function of pellet diameter for natural cluster-type fuel bundles of UO$_2$ similar to those in the Douglas Point reactor [5]. These estimates may be expressed roughly in the form

$$A + B \frac{S}{M}$$

where the volume term $A$ probably arises mainly from the cost of UO$_2$ and the $S/M$ term mainly from Zircaloy. The constants in this expression are:

$$A = \$22.22/\text{kg U}$$

$$B = \$0.102/\text{cm}^2$$

where $S/M$ is expressed in $\text{cm}^2/\text{kg}$.

The fuel rating also depends on $S/M$. When the heat rating of the fuel is limited by the heat transfer coefficient the heat which may be taken from the fuel is proportional to the surface area while the amount of fuel is proportional to $M$ so that the fuel rating is proportional to $S/M$.

Thus we express the cost of fabricating natural uranium fuel as

$$F_U = 22.22 + \frac{25.93 \times \text{MW/kg}}{\text{MW/kg for CANDU}} \frac{\$}{\text{kg}}$$

for UO$_2$ fuel bundles.

For ThO$_2$ bundles with a somewhat lower density we write

$$F_T = \frac{10.5}{9.5} \left( 22.22 + \frac{25.93 \times \text{MW/kg}}{\text{MW/kg for CANDU}} \times \frac{9.5}{10.5} \right)$$
More explicitly

\[
F_U = 22.22 + 1546 \times \frac{\text{MW}(\text{th})}{\text{kg}}
\]

\[
F_T = 24.56 + 1546 \times \frac{\text{MW}(\text{th})}{\text{kg}}
\]

For enriched fuel the cost of the enrichment is just added to \(F_U\) or \(F_T\) neglecting any complication which might be required to handle enriched material in the manufacturing plant. The fuel ratings used in these formulae are averaged over the fuel irradiation.

The precise form of these fabrication costs, especially that for thorium, has a considerable effect on the final power cost and should be considered in more detail. The difficulty in the case of thorium is the large thermal cross-section which necessitates a high enrichment to achieve criticality. Thus the same weight of thorium fuel in the same flux as uranium fuel would have at least twice the heat rating with correspondingly higher fabrication costs. This point is discussed later.

**Inventory charges**

One final cost contribution which has been included is the interest charge on fuel inventory. In the case of uranium fuels interest charges have been made on the basis of the cost of fabricated natural uranium even in cases where enrichment is considered. The reason for this is that a reactor of the type considered (\(\eta = 1.06\)) loaded initially with natural uranium would be well over critical so it would not be reasonable to charge interest against the cost of enriched fuel. For the thorium cases interest has been charged against the fuel composition which is required to make the reactor critical. This treatment of the inventory charges is only approximate and a detailed fuel management study is required to determine these charges more accurately. In addition, other capital charges will depend on the fuel rating and enrichment and a proper treatment of the capital contributions to power cost will involve a study of the reactor design as well as fuel management problems. In this preliminary study a simple treatment of fuel inventory is sufficient to indicate where differences between fuelling with thorium-based fuels and natural uranium will arise.

**Nuclear data**

Nuclear data in this calculation are based on the 1960 Westcott compilation [6] with revisions to the \(^{235}\text{U}\) and \(^{239}\text{Pu}\) data summarized by Critoph [7]. A recent estimate of \(\nu\) \((^{241}\text{Pu})\) by Westcott \((\nu = 2.966)\) has been used but the capture and fission cross-sections are taken to be proportional (as a function of energy) as in the 1960 Westcott compilation.

**Results**

Figure 5 compares \(^{232}\text{Th}\) and \(^{238}\text{U}\) using \(^{235}\text{U}\) as the fissile material. Curve (a) shows the result of enriching natural uranium to prolong fuel life.
At a flux of \(5 \times 10^{13}\) n/cm\(^2\) s the fuel rating for natural uranium is 0.0167 thermal MW/kg giving a manufacturing cost of $48.00/kg (U). This fuel is similar to that for the Douglas Point reactor. As \(^{235}\text{U}\) is added both manufacturing cost and burn-up increase. The increase in manufacturing cost is primarily from the cost of the added fissile material. The change in average fuel rating as enrichment is added is small, so the fabrication costs change very little, as can be seen by comparing curves (a) and (b). Curve (a) includes the manufacturing cost and shows how the burn-up depends on fuel costs. In curve (b) fabrication costs have been omitted leaving only the cost of refined natural \(\text{UO}_2\) ($21.50/kg (U) and extra \(^{235}\text{U}\) required. Curves (c) and (d) are corresponding results for \(^{232}\text{Th}\) and \(^{235}\text{U}\). Here thorium dioxide has been assumed to cost $21.50/kg of Th. Evidently a large capital investment is required to provide a given burn-up using thorium as a fertile material. However, the incremental fuel supply cost which is inversely proportional to the slopes of the curves at any point is much better for \(^{232}\text{Th}\) than \(^{238}\text{U}\). This is because of the superior nuclear properties of \(^{233}\text{U}\). However, with thorium the flux is important and curve (e) shows the effect of flux on burn-up. Here fabrication costs have been omitted so curve (e) is to be compared with curve (d).

The fuel supply cost is inversely proportional to the slope of a line drawn from the origin to any point on the curves. Thus at any point where the slope is greater than that of a line joining the point to the origin, fuel supply costs will be reduced by enriching further.

Figure 6 shows how costs are modified when credit for spent fuel is taken into account. Curves (b) and (d) are the result of subtracting the net value of this spent fuel from the cost of the manufactured fuel. With the processing costs used here it does not pay to process natural uranium fuel until it has reached a burn-up of 7000 MWd/t, i.e. processing costs more than the extracted plutonium is worth. Beyond this point the burn-up per dollar invested increases. For \(^{232}\text{Th} - ^{235}\text{U}\) fuel the difference due to processing (curves (c) and (d)) is much more striking since the concentration of
The comparison is still not complete. There will be a considerable time lag between providing manufactured fuel for the reactor and recovering fissile material from spent fuel. During this period interest must be paid on the inventory of fuel in the reactor. The conventional way of treating this problem is to capitalize the cost of fuel required to make the reactor critical (plus, possibly, a small inventory to allow a few months' operation). The fuel supplied to the reactor to maintain reactivity is an operating cost. This we call the fuel supply cost. Thus, interest is paid on the fuel required to make the reactor critical and the total fuel cost includes this interest and the difference between fuel supply cost and spent fuel credit. At equilibrium, spent fuel will be discharged as new fuel is provided. Since this fuel inventory is a non-depreciating item in the capital cost, low interest rates are appropriate. For these studies we have used 5%.

These points are brought out more clearly in Figs. 7-14 where we have plotted fuel costs directly. Except where noted these costs include inventory charges and credit for spent fuel. The curves in these figures all have the same general form. The fuel required to achieve criticality with thorium is very expensive compared to natural uranium because of the high initial concentration of fissile material. In addition fabrication costs are higher than for natural uranium because of the high heat ratings which must be achieved to keep inventory charges low. Thus for low ultimate burn-ups the fuelling costs for a thorium-fuelled reactor are prohibitively high. However the incremental burn-up per additional dollar invested is large so that the fuel cost decreases rapidly with increasing burn-up. Eventually an accumulation of fission products and capture production poisons will cause the cost to rise again. Figure 7 indicates the relative contributions of fuel supply cost, inventory charges and spent fuel credit to the total fuel costs.

$^{233}_{\text{U}}$ in the fuel is much higher and $^{233}_{\text{U}}$ is more valuable than the plutonium in $^{238}_{\text{U}}$.
FIG. 7. Spent fuel credit and inventory charges

FIG. 8. Thorium with various fissile materials
Use of various fissile materials

For the purposes of this general survey it has been assumed that any fissile material introduced will be irradiated in the same spectrum and for the same length of time as the fertile material. For $^{235}$U and $^{233}$Th used as enrichment with thorium, this is probably a reasonable assumption to make since these fissile materials would very likely be intimately mixed with $^{238}$U and $^{232}$Th using $^{235}$U as the fissile material. Clearly, inventory charges and spent fuel credit are major contributors to the total fuel cost for $^{232}$Th.
FIG. 11. Effect of flux without rating dependence

FIG. 12. Processing costs
thorium. The $\bar{\eta}$-values for $^{235}$U and $^{233}$U fall to 1.06 after about 4.0 and 4.7 $\mathrm{n}/\mathrm{kb}$ respectively and so would have received about their optimum irradiation when the thorium fuel was removed for processing. Plutonium (or Pu* as it will be called, being plutonium which has been extracted from natural uranium at a burn-up of about 8400 MWd/t) has an $\bar{\eta}$-value of 1.06 at 1.7 $\mathrm{n}/\mathrm{kb}$ so it would be unreasonable to irradiate it much further since it
would be acting as a parasitic absorber and producing little power. A better way of using Pu* with thorium might be in the form of spikes. With bi-directional fuelling these spikes could be pushed through the reactor faster than thorium bundles. The main difficulty here would be that initially all the power would come from the plutonium slugs and finally it would mostly come from thorium. This problem has not been considered in this report and Pu* has been treated in the same way as 235U and 233U, i.e. as if in the form of uniform enrichment.

In Fig. 15 the burn-up as a function of irradiation which would be obtained with each fissile material is plotted together with the contribution from thorium alone. The amount of fissile material is just sufficient to provide the irradiation indicated in Table III, considering, for example, an irradiation of 4 n/kb. The better fissile materials provide more burn-up of the fertile material.
Total fuel costs for thorium with different fissile materials are plotted in Fig. 8 as a function of burn-up. Curves (a) and (b) with $^{235}\text{U}$ and $^{233}\text{U}$ as enrichment show similar costs. The Pu* curve (c) shows the consequences of using Pu* as a uniform enrichment where even at lower burn-ups ($\bar{\eta} (\text{Pu}*) > 1.06$) the cost is high compared with $^{235}\text{U}$ enrichment. This is due partly to the higher fabrication costs necessitated by the high fission cross-section of $^{239}\text{Pu}$ and partly to the difference in price at which plutonium is bought and allowed for as a credit in spent fuel. At higher irradiations parasitic absorption in fission products becomes more important. While there is also a difference in cost and credit value of $^{233}\text{U}$ this is compensated by a better $\bar{\eta}$-value of $^{233}\text{U}$ so that the fuel costs are very similar for $^{233}\text{U}$ and $^{235}\text{U}$.

Results for natural uranium enriched with fissile material are shown in Fig. 9. The behaviour is basically the same as with thorium but on a different scale. Natural uranium contains sufficient 'free' $^{235}\text{U}$ for burn-ups of the order of 9000 MWD/t to be possible without further enrichment, so we do not have the problem of providing large quantities of enrichment to start up a reactor. Inventory charges are thus low and the total fuel costs are lower than those in Fig. 8. However, plutonium is not so good a fissile material as $^{233}\text{U}$, so that with $^{238}\text{U}$ instead of $^{232}\text{Th}$ as the fertile material minimum fuel costs are obtained at much lower burn-ups (~14 000 MWD/t). $^{235}\text{U}$ compares favourably with $^{233}\text{U}$ when used to enrich natural uranium. This is primarily owing to the low cost of $^{235}\text{U}$ for these low enrichments (Fig. 2).

Variation of flux

Figure 10 shows the dependence of the fuel cost on flux for thorium and natural uranium enriched with $^{235}\text{U}$. Several factors contribute to this variation.

(a) Xenon absorption increases with flux, reducing the effective $\bar{\eta}$ of the fuel. This is a small effect.

(b) For the same reason $^{239}\text{Np}$ hold-up in $^{238}\text{U}$ and $^{235}\text{Pa}$ hold-up in $^{232}\text{Th}$ become more important as the flux is raised. The $^{239}\text{Np}$ effect is relatively small because of its short half-life (2.35 d compared to 27 d for $^{233}\text{Pa}$).

(c) In a high flux less fuel will be required to produce a given power output, although it will of course need to be changed more often. Therefore interest charges on the fuel inventory will be lower. In a thorium reactor containing very expensive fuel the inventory charges contribute a bigger percentage to the total fuel cost than is the case with uranium so there may be a greater incentive to go to higher fluxes even at the expense of increased absorption in $^{233}\text{Pa}$.

(d) The simplified formulation adopted for the fuel fabrication costs in this report is such that the advantage of increasing the flux to lower the inventory charges is outweighed by the extra fabrication cost incurred. If the flux is doubled to halve the inventory charges the heat rating of the fuel must also be doubled. This calls for very thin fuel pencils with a consequent high fabrication cost. Even with a flux of $5 \times 10^{13} \text{n/cm}^2 \text{s}$ as in the Douglas Point reactor, the heat rating with a thorium fuel would be at least double that for uranium and a 37-element rod design would probably be required.
(e) If the flux in a reactor could be doubled, twice the power output would be obtained from the same reactor and the resultant savings in capital costs might be considerable. With natural uranium fuel, fabrication costs are a large proportion of the total fuel cost and so it is not worthwhile to increase the flux in the Douglas Point reactor above \(5 \times 10^{13} \text{n/cm}^2 \text{s}\) in an attempt to reduce capital costs. The cost of fabricating fuel with the necessary high heat rating would be prohibitively high. With a thorium fuel in which the fabrication cost represents a smaller fraction of the total fuel cost it may pay to increase the flux to save on capital costs.

The magnitude of the effects (a) and (b) can be seen in Fig. 16 which shows the ultimate irradiation in \(\text{n/kb}\) as a function of \(^{235}\text{U}\) enrichment. At zero irradiation in the thorium cases, an increase of enrichment from 1.780% to 1.783% is sufficient to overcome the extra xenon absorption penalty incurred when the flux is increased from \(5 \times 10^{13}\) to \(10^{14} \text{n/cm}^2 \text{s}\). The subsequent divergence of the curves is caused by the \(^{233}\text{Pa}\) hold-up.

![Graph](image.png)

**FIG. 16.** Burn-up versus irradiation for thorium and enrichment

The dependence of total fuel cost on flux is caused primarily by the increase in manufacturing cost with increase in flux as can be seen by comparing Fig. 10 with Fig. 11 where the effect of fuel rating on the manufacturing cost has been omitted.
Processing costs

At present in Canada no fuel processing is carried out on a commercial scale and processing costs in the United States are high enough so that it is barely worthwhile to process spent natural uranium. At $25/kg the cost of extraction of Pu at 4 g/kg is $6.25/g. The fissile content is 69% so the cost of extraction is about $9/g of fissile material compared to the value of $10/g which we have assigned to it. In the thorium fuel cases 14 g/kg of fuel of $^{233}$U are obtained for a processing charge of $26/kg. The extraction cost of $^{233}$U is thus only $2/g compared to the value of $12/g we have assigned to it. Thus processing will be an essential part of any thorium fuel cycle with the cost of processing contributing only a small fraction to the total fuel costs. Since the processing costs represent a much higher fraction of the cost of a uranium-plutonium fuel cycle, a reduction in the cost of processing could give the uranium-plutonium fuel cycle a big advantage. Figure 12 indicates the limiting size of this effect by showing fuel costs for thorium and natural uranium enriched with $^{235}$U. Separate curves compare results when the processing costs $26/kg and $25/kg are assumed, and when these costs are zero. For natural uranium this represents a saving of ~50%, whereas for thorium irradiated to 40 000 MWD/t the saving is about 10%. As more reactors come into operation and the quantity of fuel to be processed increases, the processing cost will decrease significantly [4]. Spent fuel will be rejected from the Douglas Point reactor at a rate of 1 t/12 d and the $25/kg cost is based on a throughput of 1 t/d. It is not likely therefore that a processing plant operating at 80% utilization would be built in Canada until the installed capacity of nuclear reactors is at least ten times that of CANDU, or 7000 MW(th).

Other input parameters

(a) Interest rate
A rate of 5% has been used throughout. This could be rather low and if so would bias the results in favour of the thorium fuel. An increase from 5% to 7% in interest rate increases the fuel cost for natural uranium by 0.028 mill/kWh but a similar increase with thorium fuel adds 0.073 mill/kWh.

(b) Station efficiency
A net station efficiency of 30% has been assumed. This is a typical value for the conversion of thermal to electric power in a large station. Any variation from 30% would merely introduce the same proportionality factor into both uranium and thorium fuel costs.

(c) Plant utilization
A station has been assumed to be on power for 7000 h/yr (80%). The effect of this is to increase the effective interest rate from 5% to 6\%\%\%\%. It is therefore more important for a thorium reactor than for a uranium reactor to operate for the maximum possible number of full power days in any given period.

(d) Neutron economy
A higher $\bar{\eta}$ value indicates a reactor of poorer neutron economy. The value 1.06 which has been used throughout this study is a reasonable number.
to take for a large D$_2$O cooled and moderated system. The exact value chosen has a large effect on the attainable burn-up for natural uranium. An increase in $\bar{\eta}$ from 1.06 to 1.08 reduces the burn-up from 8400 to 6450 MWd/t. This reduction in attainable burn-up of 23% is reflected in a correspondingly increased fuel cost. Figure 13 shows the total fuel cost curves for natural uranium and thorium, both enriched with $^{235}$U, with $\bar{\eta}$-values of 1.06 and 1.08. The penalty incurred by increasing $\bar{\eta}$ from 1.06 to 1.08 in natural uranium is about 0.3 mill/kWh compared with about 0.1 mill/kWh in enriched thorium or enriched uranium.

(e) Plutonium price

The $12 (including handling charges) cost of plutonium assumed for most cases assumes a certain degree of economic coupling with the nuclear power programme in the United States. Another equally valid viewpoint would consider that plutonium inventories built up in Canada are similar to the tailings of a working mine and thus of no value until it becomes economic to use them.

By the time a significant inventory of plutonium has been accumulated in Canada, processing costs may well be low enough to provide fissile plutonium at $\$6/g. To assess the impact of this low cost, fuelling costs have been calculated for Pu$^{232}$Th fuel assuming a supply cost of $\$5 per fissile atom and a credit of $\$4 for plutonium contained in the spent fuel. The results plotted in Fig. 14 compare the low fuel costs attainable with those obtained under similar conditions, assuming $\$12.00/g of fissile plutonium. With this choice for the cost of plutonium Fig. 14 does not tell the whole story. In addition to giving low fuel costs, the value of the fuel inventory will increase during the life of the reactor. The use of plutonium and thorium in reactors looks interesting and should be investigated further.

SUMMARY AND CONCLUSIONS

In these exploratory calculations cost estimates are not precise. Nevertheless some general conclusions can be made which do not depend on the precise costing methods used.

The high thermal cross-section of thorium requires a large amount of fissile material (or a small amount of thorium) to make a thorium-fuelled reactor critical (Fig. 17). This makes the fuel expensive and fuel costs with no credit for spent fuel are high (Fig. 7). However, the high cross-section also leads to a high concentration of $^{233}$U at the end of the irradiation, and processing to recover this uranium is economic with present day processing costs. The effect of the spent fuel credit is to lower reactor fuelling costs so that they are comparable with a natural uranium reactor (Fig. 7). Lower processing costs will reduce fuel costs still more but would benefit natural uranium fuel costs more than thorium fuel costs (Fig. 12).

A thorium-based fuel is very much more expensive than natural uranium because it must contain more fissile material; this fissile material is more expensive than that in natural uranium and fuel ratings are likely to be higher than for natural uranium. High fuel ratings reduce inventory charges which are an appreciable fraction of the fuel costs (Fig. 7). Natural uranium is so cheap that, in spite of the large contribution of fabrication cost to the manu-
FIG. 17. Thorium burn-up as a function of enrichment

Fabrication cost (Fig. 5), inventory charges are a relatively minor contribution to the fuel cost. For thorium fuels we have the reverse situation. Here the fabrication charges are a much smaller fraction of the manufacturing cost but the fuel itself is so expensive that inventory charges are high. Inventory charges may be reduced by increasing the fuel rating. However, increasing the fuel rating increases the manufacturing cost. For cluster-type fuel bundles the effect of rating on manufacturing cost is so large that an increase in fuel rating (and reactor flux) leads to higher fuel costs (Fig. 10). The increase in manufacturing cost more than compensates for the reduction in inventory charges. The contribution of fabrication charges to reactor fuel costs can be seen by comparing Fig. 10 with Fig. 11 where the fabrication costs are fixed at $48/kg(U). There is thus a large incentive to increase fuel ratings cheaply for thorium-based fuels, and different types of fuel channels and fuel management schemes must be investigated.

From Fig. 4 the relative worths of $^{233}$U, $^{235}$U and plutonium were determined and an "indifference" value ascribed to $^{233}$U and plutonium based on $12/g of 238$U. This seems to be unrealistic, at least in Canada, where large amounts of relatively cheap natural uranium are available. While the calculations performed here are not exhaustive, they indicate that it is not worthwhile to irradiate natural uranium much beyond 14 000 MWD/t in CANDU-type reactors. This sort of burn-up may be obtained using an enrichment less than 2.2% (Fig. 15) and according to Fig. 3, $^{235}$U enrichments in this range may be obtained for $10.61/g. The costs used in this report have been based on guaranteed prices quoted by the USAEC. In fact these prices may be high as far as indicating their worth in extending the burn-up of natural uranium in Canada is concerned (Fig. 9). In any case a complete study of the use of Pu*-Th fuels in CANDU-type reactors must take into account heavier isotopes such as $^{241}$Am and $^{243}$Am which are potentially valuable materials and may affect the credit allowed for spent fuel appreciably. Nuclear data for the
heavier nuclides are not accurate and an effort to improve this knowledge would be useful.

There is no doubt that thorium-based fuels will be irradiated to much higher burn-ups than natural or slightly enriched uranium. However, the gain beyond about 40 000 MWd/t is small and for this sort of burn-up fuel swelling is probably quite manageable. Some development work in this area will, however, be necessary.

Fuel costs for thorium-based fuels appear to be very nearly competitive with fuel costs from natural uranium CANDU-type reactors, even with the present low price of natural uranium. The effect of thorium-based fuels on reactor design and capital cost may be appreciable and for this reason more detailed studies are of immediate importance.

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QUELQUES ASPECTS DES CYCLES THORIUM
DANS LES REACTEURS A EAU LOURDE

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I. INTRODUCTION

On a cherché dans ce qui suit à préciser au moyen de calculs simplifiés quelques propriétés des cycles thorium, afin d'en déduire des remarques générales sur leur économie. L'étude est limitée aux cycles amorcés et entretenus à l'aide d'uranium-235. Elle concerne plus spécialement les réacteurs à eau lourde qui sont particulièrement bien adaptés à l'utilisation du thorium; on peut d'ailleurs considérer que cette possibilité constitue à long terme un atout important pour ces réacteurs.

Il est bien connu que dans un réacteur thermique le cycle $^{233}$U-Th avec recyclage de $^{233}$U formé est plus avantageux du point de vue utilisation des noyaux fissiles que le cycle $^{235}$U- $^{238}$U avec recyclage du $^{239}$Pu formé. Ceci résulte de ce que le $\eta$ de l'uranium-233 est plus élevé que celui du plutonium-239 et que, compte tenu des descendants, il se dégrade moins vite avec l'irradiation.

Lorsqu'on peut réduire suffisamment les pertes stériles de neutrons dans le réacteur (fuites et captures dans le modérateur ou les matériaux de structure) on peut même aller jusqu'au «breeding», c'est-à-dire ne plus faire d'appoint de noyaux fissiles une fois le cycle amorcé. Cela nécessite cependant qu'on raccourcisse suffisamment la durée d'irradiation pour diminuer l'empoisonnement par les produits de fission. Du point de vue économique il n'est pas forcément avantageux d'aller jusqu'au «breeding»: il y a un compromis à faire entre la consommation de noyaux fissiles neufs et la fréquence des retraitements. Tant que l'uranium n'est pas excessivement cher, on peut préférer consentir un appoint plus grand de noyaux fissiles pour viser des taux de combustion plus élevés.

Il est intéressant d'examiner ce compromis, mais il est clair que de toute manière le meilleur «convertisseur» est celui qui gaspille le moins les neutrons. Toute économie sur les pertes stériles permet de gagner à la fois en consommation de noyaux fissiles et en prolongation de «burn-up», et c'est pourquoi les réacteurs à eau lourde sont les mieux placés à ce point de vue. Les petits calculs qui suivent sont destinés en particulier à préciser ces questions.

II. CALCULS

On considère d'abord le premier passage dans le réacteur et on étudie comment varie le «burn-up» en fonction des conditions initiales; ensuite on étudie les recyclages.
1. Premier passage dans le réacteur

a) Hypothèses simplificatrices

Afin de mettre plus clairement en évidence l'effet des paramètres, on a introduit les simplifications suivantes:
- On suppose que l'uranium-235 ajouté au thorium est enrichi à 100%, ce qui évite d'introduire la chaîne des plutoniums.
- On admet que les sections efficaces effectives ne varient pas au cours de l'irradiation et sont les mêmes dans les diverses comparaisons.
- On admet que le flux dans le combustible reste le même au cours de l'irradiation; c'est en principe le flux moyen dans la pille.
- On définit la relation entre le taux de combustion moyen et la réactivité disponible, en considérant un réacteur en équilibre d'irradiation avec circulation «idéale», c'est-à-dire qu'on écrit la condition critique pour une composition qui est la moyenne par rapport au flux intégré de toutes les compositions entre l'entrée et la sortie du combustible.
- Enfin les variations avec l'irradiation des paramètres neutroniques autres que $\eta$ sont traitées de façon sommaire. On a également négligé quelques réactions secondaires.

b) Paramètres considérés

Le combustible entrant est supposé constitué d'un mélange de thorium-232 et d'uranium-235; un premier paramètre est la teneur $\gamma$ en $^{235}\text{U}$. Un deuxième paramètre est le facteur de conversion qui dépend de $\gamma$ et de la section effective de captures du thorium, c'est-à-dire du facteur antitrappe $p$. Soit $k-1$ l'excédent de réactivité du réseau neuf; lorsqu'on s'est donné $\gamma$ et $p$, $k$ dépend des captures parasites et des fuites. Posons $A = \gamma p/k = (1 + B^2 L^2) (1 + B^2 L^2) / \gamma f$ ; la donnée de trois des paramètres $\gamma$ $p$ $k$ entraîne celle du quatrième. Enfin, à cause de la décroissance radioactive du protactinium il faut se donner un flux moyen $\varphi$; lorsque $\varphi$ et $\gamma$ sont choisis, la puissance spécifique initiale $w$ est fixée; étant donné son importance pratique, il vaut mieux choisir $w$ comme paramètre indépendant plutôt que $\varphi$. Finalement on considère quatre paramètres indépendants.

Par convention on a inclus les captures du xénon et du samarium non pas dans $\eta$ mais dans $f$, donc dans $A$. On a pris comme valeur de référence $A = 1,10$, ce qui correspond à un «bon» réacteur à eau lourde, c'est-à-dire à un réacteur dans lequel les captures de matériaux de structure et les fuites sont maintenues à un niveau assez bas. L'enrichissement $e$ du cas de référence a été choisi de manière que, pour cette valeur de $A$ et pour un $p$ qui soit par exemple de l'ordre de 0,86, c'est-à-dire un rapport de modération convenable, on trouve un taux de combustion de l'ordre de 25 000 MWj/t; ceci conduit à une teneur initiale en uranium-235 voisine de 2%, et à un $k$ d'environ 1,035. Enfin on a pris comme puissance spécifique moyenne de référence 15 MW/t, ce qui est une valeur acceptable du point de vue économique. A partir de ce cas-type, on a effectué des variations indépendantes des paramètres (voir tableau I).

Dans les cinq cas 2 à 6, on fait varier un des quatre paramètres $e$ $p$ $k$ $A$, deux restant constants, le dernier s'en déduisant. On maintient constants
successivement les couples $eA$, $pA$, $kA$, $ek$, $ep$; on s'arrange en outre pour qu'il n'y ait en tout que deux valeurs de $e$, deux valeurs de $k$, deux valeurs de $A$; trois valeurs de $p$. Dans le cas 7 on fait varier seulement $W$. Ces modifications sont faites de manière à mettre en évidence les effets suivants:

- Il y a trois manières d'augmenter la réactivité initiale: en gagnant sur $p$ (donc en diminuant le coefficient de conversion), en gagnant sur les captures des structures ou les fuites (sans toucher le coefficient de conversion), et en enrichissant davantage. Ces trois modifications sont étudiées successivement en comparant au cas 1 respectivement les cas 2, 6 et 3.

- Le supplément d'enrichissement peut être utilisé, soit pour accroître la réactivité initiale, soit pour détériorer volontairement $p$ (en économisant de l'eau lourde, et en améliorant le facteur de conversion): ces deux effets sont étudiés dans les cas 3 et 4 comparés à 1.

- Les cas 4 et 5 permettent de comparer l'effet d'un accroissement du coefficient de conversion, selon qu'il est obtenu par enrichissement ou par gain sur les captures parasites.

- En comparant les cas 5 et 6 on voit qu'on retire d'un gain sur les captures stériles, suivant qu'on investit ce gain dans le facteur de conversion ou en réactivité disponible pour l'irradiation.

Enfin en comparant les cas 1 et 7 on met en évidence l'effet d'une augmentation de la puissance spécifique moyenne, c'est-à-dire de la variation du flux moyen indépendamment de celle liée à l'enrichissement.

c) Résultats

Les calculs ont été faits en utilisant les sections efficaces effectives (température des neutrons 100°C; $R$ Westcott 0,15) (voir tableau II).

Les produits de fission sont introduits au moyen de trois pseudo-produits, suivant l'ajustement de Walker. Les équations de transformation des noyaux lourds permettent de calculer des compositions en noyaux et d'en déduire les variations de $\eta$ et des sections effectives macroscopiques. La variation de $A$ est calculée approximativement en tenant compte des variations de
TABLEAU II

SECTIONS EFFICACES

<table>
<thead>
<tr>
<th></th>
<th>$^{233}$U</th>
<th>$^{234}$U</th>
<th>$^{235}$U</th>
<th>$^{238}$U</th>
<th>$^{232}$Th</th>
<th>$^{239}$Pa</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\theta_a$</td>
<td>712</td>
<td>219</td>
<td>672</td>
<td>54</td>
<td>7,56</td>
<td>150</td>
</tr>
<tr>
<td>$\nu\sigma_f$</td>
<td>1610</td>
<td>1375</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$\theta_a$ et $\nu\sigma_f$; comme celles-ci sont faibles, l'effet est presque négligeable, ce qui justifie un traitement sommaire indépendant de la géométrie détaillée du réseau. On cherche le taux de combustion tel que $k$ défini par $\eta p/A$ sur la composition moyenne soit égal à l'unité. Les résultats sont donnés au tableau III.

TABLEAU III

TAUX DE COMBUSTION

<table>
<thead>
<tr>
<th>Cas</th>
<th>$\tau$ (MWj/t)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>24 700</td>
</tr>
<tr>
<td>2</td>
<td>29 900</td>
</tr>
<tr>
<td>3</td>
<td>32 200</td>
</tr>
<tr>
<td>4</td>
<td>27 000</td>
</tr>
<tr>
<td>5</td>
<td>32 000</td>
</tr>
<tr>
<td>6</td>
<td>37 100</td>
</tr>
<tr>
<td>7</td>
<td>19 800</td>
</tr>
</tbody>
</table>

d) Commentaires

Pour retrouver un $\eta$ initial comparable à celui de l'uranium naturel, il faut une teneur en $^{235}$U qui soit environ 2,8 fois plus élevée; en effet la section de capture du thorium est 2,8 fois plus grande que celle de l'uranium-238; pour une même géométrie, la capture du combustible est donc à peu près multipliée par ce rapport. Il se trouve que pour un même excédent de réactivité initial et un même $p$, le taux de combustion que l'on peut atteindre est lui aussi près de trois fois plus grand; le $\eta$ se dégrade en effet beaucoup moins vite avec l'irradiation.

La composition du combustible à 25 000 MWj/t dans le cas de référence est montrée au tableau IV.
LE THORIUM DANS LES REACTEURS À D₂O

TABLEAU IV

COMPOSITION DU COMBUSTIBLE

<table>
<thead>
<tr>
<th></th>
<th>²³⁵U</th>
<th>²³⁴U</th>
<th>²³³U</th>
<th>²³⁴U</th>
<th>²³²Th</th>
<th>²³⁵Pa</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1,240%</td>
<td>0,259%</td>
<td>0,380%</td>
<td>0,131%</td>
<td>95,3%</td>
<td>0,060%</td>
</tr>
</tbody>
</table>

Le $\eta$ du mélange thorium isotopes de l'uranium est à ce moment de 1,309 contre 1,318 dans le combustible neuf; la baisse est donc très faible, la plus grande partie de la perte de réactivité est due à l'accumulation des produits de fission. On remarque par ailleurs qu'au cours de l'irradiation, il y a eu à peu près le même nombre de fissions dans l'uranium-233 et dans l'uranium-235.

La réactivité baisse d'abord très rapidement à cause de la formation de protactinium, puis, une fois celui-ci saturé, beaucoup plus lentement. La lenteur de cette décroissance fait qu'il suffit d'un faible gain sur le bilan initial pour gagner énormément en MWj/t; ceci apparaît dans la comparaison des cas 1 et 6 pour lesquels le facteur de conversion est le même; un gain de 1450 pcm sur A suffit pour prolonger le taux de combustion de 50%, soit 12 400 MWj/t. D'où le très gros intérêt d'un excellent bilan neutronique.

On gagne moins si le supplément de réactivité est obtenu par enrichissement; la comparaison 1–3 montre qu'avec le même $\Delta k$ initial, on ne gagne que 7500 MWj/t. Le supplément est cependant considérable, si l'on remarque qu'il n'a nécessité que 0,82 g/kg d'uranium-235 supplémentaire, soit un rendement de 9 MWj/g (alors que la destruction pure et simple d'un gramme de $^{235}$U ne produit que 0,8 MWj); l'augmentation de la durée de la conversion est plus avantageuse que sa légère détérioration.

Une autre manière de jouer sur le coefficient de conversion consiste à modifier p. Si l'on fait varier simultanément $e$ et $p$ de manière à maintenir $k$, la comparaison des cas 1 et 4 montre que l'effet $p$ est prépondérant; c'est ce qui explique l'ordre des rapidités de décroissance de la réactivité; le meilleur cas est 5, puis 4, puis 1 (ou 6), 3 et enfin 2.

On voit néanmoins que vouloir gagner sur le coefficient de conversion en réduisant la réactivité disponible a pour effet de diminuer le «burn-up»: on ne récupère pas intégralement les neutrons investis dans le thorium. Ceci apparaît aussi bien lorsqu'on compare 4 par rapport à 3, ou 5 par rapport à 6, que 1 par rapport à 2; dans tous les cas le fait d'avoir détérioré $p$ de 1450 pcm entraîne une perte d'environ 5200 MWj/t (comparés aux 12 400 MWj/t mentionnés plus haut, on voit néanmoins, comme il se doit, qu'il vaut mieux perdre des pcm dans $p$ que dans les captures stériles).

On pourrait penser que la détérioration volontaire de $p$, si elle fait perdre en «burn-up», toutes choses égales d'ailleurs, est avantageuse pour les recyclages, en permettant de retrouver davantage de noyaux fissiles dans le combustible sortant; c'est vrai. Mais faisons la comparaison en raisonnant au même «burn-up»; en tenant compte des résultats 1, 3 et 4, on trouve que pour obtenir un même taux de combustion avec un $p$ inférieur de 1450 pcm,
Il faut ajouter au départ 0,57 g/kg de $^{235}\text{U}$; on constate alors que le supplé-
ment $^{238}\text{U} + ^{235}\text{U}$ dans le produit sortant est du même ordre de grandeur. On
ne peut donc pas dire que l'opération soit payante; cet investissement supplé-
mentaire est inutile.

Enfin lorsqu'on augmente la puissance spécifique de 15 à 20 MW/t,
l'accroissement de l'empoisonnement protactinium se traduit par une perte
de réactivité de 560 pcm qui équivaut, toutes choses égales d'ailleurs,
à une diminution de «burn-up» de 4900 MWj/t, ou encore nécessaire, pour le
même taux de combustion, un enrichissement supplémentaire de 0,54 g/kg
de $^{235}\text{U}$.

e) Effet protactinium

Le retard à la formation de l'uranium-233 à partir du protactinium cons-
titue une perte dans le bilan neutronique, non seulement du fait de la capture
du protactinium, mais surtout à cause du manque à gagner sur l'uranium
fissile. Si par exemple dans le cas 1, on sort de la pile le combustible
irradié à 1 n/kb et qu'on le replace lorsque 95% du protactinium s'est dés-
intégré, le $\eta$ passe de 1,2909 à 1,3134 soit un gain de 1740 pcm. Ce gain
va évidemment se résorber au fur et à mesure que le protactinium se sature
à nouveau, mais si l'on faisait l'opération un certain nombre de fois, par
exemple en mettant à profit les remaniements de combustibles (shuffling),
on pourrait théoriquement augmenter la réactivité moyenne.

On constate cependant que le gain est chaque fois rapidement résorbé;
compte tenu de l'immobilisation de combustible supplémentaire, il est
douteux que ces opérations soient payantes.

D'une manière générale, en jouant sur les variations de flux dans la
pile et sur les procédures de remaniement de combustible, on peut gagner
ou perdre un peu en réactivité par rapport au calcul à flux constant; cepen-
dant, étant donné la période de désintégration du protactinium (27 j) com-
parée aux durées de séjour en pile (plusieurs années), ces effets ne peuvent
être que marginaux.

2. Recyclages

a) Deuxième passage

Supposons qu'on utilise pour un nouveau cycle le combustible irradié
à 25 000 MWj/t dans les conditions de référence, après l'avoir débarrassé
de ses produits de fissions; on ne conserve que les isotopes de l'uranium
et du thorium, et on suppose qu'au moment du retraitement 95% du $^{238}\text{Pa}$ s'est
désintégré, et qu'il y a 1% de perte au retraitement des noyaux fissiles.
L'excédent initial de réactivité $k$ calculé dans les mêmes conditions qu'au-
paravant est de 1,042 au lieu de 1,035. Il est donc possible de fonctionner
à nouveau sans appoint de matière fissile.

Cependant l'évolution de la réactivité est moins favorable que la pre-
mière fois car on est plus proche du régime d'équilibre. On constate que
si on ne rajoute pas de noyaux fissiles, bien que le $k$ initial soit plus grand,
le «burn-up» est plus faible; mais il suffit d'ajouter 1 g/kg de $^{235}\text{U}$ pour re-
trouver un «burn-up» de 25 000 MWj/t. Le combustible initial comporte alors 1,28% de $^{233}$U et 0,48% de $^{235}$U.

Au terme de ce deuxième séjour en pile, la composition du combustible est montrée au tableau V.

**TABLEAU V**

**COMPOSITION DU COMBUSTIBLE**

<table>
<thead>
<tr>
<th></th>
<th>$^{233}$U</th>
<th>$^{234}$U</th>
<th>$^{235}$U</th>
<th>$^{236}$U</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1,449%</td>
<td>0,286%</td>
<td>0,148%</td>
<td>0,307%</td>
</tr>
</tbody>
</table>

Par rapport au 1er passage, il y a donc 0,209% de $^{233}$U en plus, et 0,242% de $^{235}$U en moins; la composition a peu évolué. Néanmoins le cycle n'est pas encore stabilisé: $^{233}$U est proche de sa valeur d'équilibre, mais $^{234}$U continuant à croître, la valeur de $^{235}$U en fin de cycle tend ensuite à remonter.

b) **Equilibre**

On peut raisonner directement sur les teneurs d'équilibre. Désignons par $N_2$, $N_3$, $N_4$, $N_5$, les nombres de noyaux respectivement de $^{232}$Th, $^{233}$Pa, $^{233}$U, $^{234}$U, $^{235}$U et par $\sigma_2$, $\sigma_3$, $\sigma_4$, $\sigma_5$, les sections d'absorption effectives correspondantes. Soit $\lambda$ la constante radioactive de $^{233}$Pa. Supposons que $N_3$, $N_4$ et $N_5$ aient atteint leurs valeurs d'équilibre (il s'agit d'un pseudo-équilibre puisque $N_2$ continue à décroître); on a les relations de conservation suivantes (en désignant par $\alpha_3$ le rapport entre la section de capture radiative et la section d'absorption totale de $^{233}$U):

- pour $^{233}$Pa $N_2\sigma_2\varphi = N_3(\sigma_3\varphi + \lambda)$
- pour $^{233}$U $N_3\lambda = N_3\sigma_3\varphi$
- pour $^{234}$U $N_3\sigma_3 + N_3\alpha_3 \sigma_3 = N_4\sigma_4$
- pour $^{235}$U $N_4\sigma_4 = N_5\sigma_5$

Calculons un $\eta^*$ relatif aux cinq noyaux mentionnés, c'est-à-dire, excluant les produits de fission ainsi que $^{236}$U (qui est en général loin de la saturation), mais incluant les captures résonnantes du thorium. Posant $t = \sigma_3' \varphi / \lambda$, on trouve

$$\eta^* = \frac{\eta_3 + \eta_5(\alpha_3 + t)}{2 + 2\alpha_3 + 4t}$$

c) **Discussion**

Cette formule permet d'apprécier immédiatement l'effet du flux, puisque celui-ci apparaît dans le coefficient $t$. 
En second lieu on en tire une condition pour que le «breeding» soit possible; il faut que $\eta^*$ ainsi calculé soit supérieur au paramètre $A$ considéré plus haut. En effet, supposons que les teneurs d'équilibre étant atteintes, on recycle le combustible en récupérant intégralement les noyaux fissiles après que tout le protactinium soit décomposé. Au bout de quelque temps le protactinium qui s'est à nouveau formé a atteint sa valeur de saturation, et $N_3$ est revenu à sa valeur d'équilibre. Si l'on néglige la capture de $^{238}\text{U}$ ainsi que celle des autres noyaux provenant des réactions secondaires, l'excédent de réactivité, indépendamment des produits de fission, est $\eta^*/A$; en effet les captures résonnantes du thorium sont incluse dans $\eta^*$. Les produits de fission s'accumulant, la pile peut fonctionner jusqu'à ce que leur capture moyenne compense cet excédent.

On remarque que cette condition est indépendante du facteur $p$: si l'on augmente les captures résonnantes du thorium, c'est-à-dire $\sigma_2$, les nombres de noyaux $N_3$, $N_3^1$, $N_4$ et $N_5$ rapportés à $N_2$ sont augmentés dans la même proportion, et $\eta^*$ est le même.

Si l'on veut accroître le taux de combustion, on doit rajouter des noyaux de $^{235}\text{U}$, et ce nombre supplémentaire décroit ensuite exponentiellement avec l'irradiation; le supplément de réactivité apporté par le nombre moyen de noyaux supplémentaires permet d'accumuler un plus grand nombre de produits de fission.

Ce raisonnement montre que la relation entre le «burn-up» et l'appoint de matières fissiles est pratiquement indépendante — à des effets du second ordre près — du facteur $p$. Certains auteurs pensent que dans le cas du cycle thorium, il y a intérêt à sous-modérer pour réduire la consommation de noyaux fissiles. C'est une erreur, qui provient de la confusion entre deux problèmes: si l'on détériore volontairement $p$ et qu'on ne fasse rien d'autre, on réduit nécessairement le taux de combustion, et le gain de matière fissile que l'on observe n'est que la contrepartie de cette réduction. Si l'on raisonne au contraire à même taux de combustion, il faut faire un appoint initial plus grand, et la consommation de matière fissile est pratiquement la même dans le cycle stabilisé; on a vu plus haut qu'elle est pratiquement la même aussi pour le premier passage. La seule conséquence d'une détérioration de $p$ est donc au contraire une immobilisation plus grande de matière fissile. Du point de vue économique, on pourra faire un arbitrage entre cette immobilisation et celle de l'eau lourde: il y a normalement un optimum. Il faut bien séparer ce problème de celui du taux de combustion optimal.

Pour ce dernier problème, la relation entre le taux de combustion et l'appoint de $^{235}\text{U}$ dépend des deux paramètres $A$ et $\varphi$. Comme une augmentation de $\varphi$ équivaut à une augmentation de $A$, l'équivalence étant obtenue en écrivant que $\eta^*/A$ conserve la même valeur, on peut raisonner sur le seul paramètre $A$. Il doit être entendu alors qu'une augmentation de puissance spécifique grève $A$ au même titre qu'un accroissement de la capture des matériaux de structure.

On a fait quelques calculs en considérant des combustibles dont la teneur initiale est déduite des équations d'équilibre avec un appoint variable de $^{235}\text{U}$. Les résultats sont les suivants (voir tableau VI): on donne, en fonction de $A$, le nombre de g/kg de $^{235}\text{U}$ qu'il faut ajouter pour obtenir un taux de combustion donné.
TABLEAU VI

NOMBRE DE g/kg DE $^{235}$U AJOUTÉ POUR OBTENIR UN TAUX DE COMBUSTION DONNE

<table>
<thead>
<tr>
<th>MWj/t</th>
<th>$\lambda=1,08$</th>
<th>$\lambda=1,10$</th>
<th>$\lambda=1,12$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10000</td>
<td>-</td>
<td>0,1</td>
<td>1,35</td>
</tr>
<tr>
<td>20000</td>
<td>0</td>
<td>1,45</td>
<td>2,95</td>
</tr>
<tr>
<td>30000</td>
<td>1,3</td>
<td>3,05</td>
<td>4,8</td>
</tr>
<tr>
<td>40000</td>
<td>2,9</td>
<td>5,0</td>
<td>7,1</td>
</tr>
</tbody>
</table>

Ces résultats sont un peu moins favorables que ceux indiqués plus haut pour les premiers cycles, parce qu'on a utilisé des valeurs plus récentes des produits de fission, en tenant compte en particulier des captures résonnantes; les résultats sont très sensibles aux hypothèses faites à ce sujet, puisque ce sont les produits de fission qui déterminent la chute de réactivité. De toute manière nous avons fait des hypothèses simplificatrices, les chiffres seraient un peu modifiés si l'on tenait compte de la présence d'uranium-238 à côté de l'isotope 235, et d'une description plus détaillée du spectre et des caractéristiques de la cellule; ils n'ont donc qu'une valeur indicative.

L'examen de ce tableau conduit aux mêmes conclusions que plus haut: on remarque le très fort rendement en énergie du gramme de $^{235}$U ajouté: son rôle est en effet moins de fournir de l'énergie par lui-même, que de permettre à la conversion du thorium de se poursuivre plus longtemps, en apportant des neutrons supplémentaires, et tirer ainsi de l'énergie de l'uranium-233 formé. Tout se passe comme si l'on retirait 6, 8, 10... fissions du gramme de $^{235}$U, ce nombre tendant vers l'infini lorsqu'on se rapproche du cycle auto-entretenu. On remarque d'autre part l'influence considérable du paramètre $\lambda$: les pcm coûtent extrêmement cher dans ce cycle de combustible.

Pour que $\lambda$ soit de l'ordre de 1,10, compte tenu de l'empoisonnement xénon et samarium, et des fuites (2 à 2,5% pour un réacteur de grande puissance unitaire), il faut que les captures du modérateur, éventuellement du refroidisseur, des gaines, tubes et matériaux de structure ne dépassent pas ensemble 4000 à 4500 pcm. Si l'on désire porter la puissance spécifique de 15 à 30 MW/t, ce chiffre doit être réduit considérablement. $\lambda=1,10$ est donc déjà une bonne valeur, et on peut considérer que 1,08 est une limite pratique pour un réacteur à eau lourde, cette valeur caractérisant un bilan neutronique absolument excellent. Si au contraire on considérait par exemple un réacteur à eau légère, il est certain qu'on ne pourrait pas descendre au-dessous de 1,12 à cause des captures du modérateur.

III REMARQUES SUR L'ECONOMIE DES CYCLES

Du point de vue économique, il faut distinguer l'immobilisation de combustible dans la première charge, et le coût de renouvellement.
Les procédures d’amorçage du cycle sont nécessairement très complexes. La teneur en uranium-235 du premier chargement peut être un peu plus faible que celle définie plus haut, puisqu’il suffit que le réacteur soit critique \((k = 1)\); mais il faut ensuite dégager la réactivité nécessaire pour compenser la détérioration du bilan neutronique. Par exemple si l’on adopte dès le départ une procédure de rechargement continu, on est amené à retraiter du combustible peu irradié, et à rajouter au fur et à mesure les quantités nécessaires de \(^{235}\text{U}\). Pratiquement, on cherche à éviter de retraiter prématurément le combustible, en utilisant des procédures plus compliquées: enrichissements différenciés, shuffling, poison soluble, etc. Mais on peut admettre en première approximation que la correspondance entre l’appoint de matière fissile et le «burn-up» moyen est du même ordre que dans le cas du régime «idéal», sauf, bien entendu, si l’on investit une partie des neutrons pour obtenir un aplatissement de la répartition des puissances.

On ne peut entrer ici dans le détail de cette période transitoire, mais il faut remarquer qu’elle s’étend sur une période très longue: si les puis- sances spécifiques sont de l’ordre de 15 à 20 MW/t, et les taux de combustion de 25 000 à 30 000 MWj/t, les temps de séjour moyens sont de l’ordre de cinq à six ans. Il faut remarquer également que l’investissement initial en \(^{235}\text{U}\) pèse lourd par rapport à la consommation ultérieure. Il peut être d’ailleurs une raison qui incite à augmenter fortement la puissance spécifique, malgré la pénalisation qui en résulte.

Examinons maintenant, du point de vue du coût de renouvellement du combustible, le compromis entre l’appoint de noyaux fissiles et le «burn-up». Si l’on allonge la durée d’irradiation, il faut fournir un peu plus de \(^{235}\text{U}\), mais on amortit sur un nombre plus grand de kWh les frais de fabrication et de retraitement. Si le thorium est recyclé, sa contribution dans le coût de l’énergie est extrêmement faible, puisqu’on tire 6500 à 7000 kWh de la destruction d’un seul gramme; ce chiffre est d’ailleurs indépendant du compromis choisi. Si le thorium n’est pas recyclé, il faut ajouter son prix aux frais de fabrication, par unité de poids. Il y a donc en définitive un seul paramètre économique pour définir l’optimum lorsque A est donné: c’est le rapport entre l’ensemble des coûts de fabrication et de retraitement par kilogramme de combustible, et le prix du gramme de \(^{235}\text{U}\). Lorsque ce rapport augmente, l’optimum est déplacé vers des taux de combustion plus élevés; lorsqu’au contraire l’uranium devient plus cher, on cherche à économiser sur la consommation de matière fissile.

A titre d’exemple, on peut utiliser les chiffres donnés plus haut dans deux hypothèses extrêmes qui n’ont bien entendu qu’une valeur indicative. Admettons que le prix du gramme de \(^{235}\text{U}\) soit successivement de 12 $ puis de 50 $. La première valeur est celle du barème AEC actuel pour de l’uranium fortement enrichi; ce barème est basé sur un prix de l’uranium naturel de 8 $/lb de U\(_3\)O\(_8\); la deuxième valeur correspond en gros à un prix dix fois supérieur de l’uranium (80 $/lb) avec des frais d’enrichissement («toll enrichment») du même ordre. On suppose d’autre part que l’ensemble des frais de fabrication et de retraitement s’élève à 100 $/kg. Le coût de renouvellement du combustible en mill/kWh, pour un rendement de 30%, se décompose alors de la manière montrée au tableau VII dans l’hypothèse \(A = 1,10\).
LE THORIUM DANS LES REACTEURS A D₂O

TABLEAU VII
COUT DE RENOUVELLEMENT
DU COMBUSTIBLE
POUR UN RENDEMENT DE 30%
(A = 1,10)

<table>
<thead>
<tr>
<th>MWj/t</th>
<th>12 $/g</th>
<th>50 $/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 000</td>
<td>1,39 + 0,02 = 1,41 mill/kWh</td>
<td>1,39 + 0,07 = 1,46 mill/kWh</td>
</tr>
<tr>
<td>20 000</td>
<td>0,70 + 0,12 = 0,82 mill/kWh</td>
<td>0,70 + 0,50 = 1,20 mill/kWh</td>
</tr>
<tr>
<td>30 000</td>
<td>0,46 + 0,17 = 0,63 mill/kWh</td>
<td>0,46 + 0,71 = 1,17 mill/kWh</td>
</tr>
<tr>
<td>40 000</td>
<td>0,35 + 0,21 = 0,56 mill/kWh</td>
<td>0,35 + 0,87 = 1,22 mill/kWh</td>
</tr>
</tbody>
</table>

Dans l'hypothèse d'un prix de matière fissile très bas on voit qu'on a intérêt à rechercher des taux de combustion aussi élevés que possible; on sera limité moins pour des raisons économiques que pour des raisons technologiques ou pratiques. Mais on voit que même si la matière fissile coûte beaucoup plus cher, et même si le «breeding» est possible, on n'a pas forçément intérêt à le rechercher; l'optimum économique peut avoir lieu pour un taux de combustion déjà relativement élevé, avec appoint d'une petite quantité de ²³⁵U.

On remarque d'autre part que même dans l'hypothèse d'un coût d'uranium extrêmement élevé, 80 $/lb de U₃O₈, le coût de renouvellement du combustible reste très bas, à cause de l'utilisation extrêmement poussée de cet uranium. Si l'on va à 30 000 MWj/t, tout se passe comme si l'on tirait 12 fissions de chaque noyau ajouté. Le cycle thorium permet donc d'être très peu sensible à un renchérissement de l'uranium, et c'est pourquoi ses perspectives d'utilisation sont intéressantes.

Néanmoins, cet intérêt dépend beaucoup du bilan neutronique, comme le montre la comparaison du tableau VIII dans l'hypothèse 50 $/g.

C'est pourquoi cet avantage potentiel des cycles thorium existe surtout pour les réacteurs à eau lourde, et spécialement pour ceux qui ont un excellent bilan neutronique.

IV CONCLUSIONS

L'évolution, au cours de l'irradiation, des caractéristiques neutroniques des combustibles formés de thorium et d'isotopes fissiles de l'uranium se présente de façon très favorable, de sorte qu'il est possible d'obtenir dans un réacteur à eau lourde des taux de combustion élevés avec un apport minimum de matière fissile. On peut théoriquement, dans un réacteur où les pertes stériles de neutrons sont réduites au minimum, aller jusqu'au «breeding», c'est-à-dire ne plus faire d'apport de ²³⁵U ou même produire ²³³U, une fois le cycle amorcé.

Cependant, on n'a pas intérêt en général à rechercher cette possibilité: il existe dans tous les cas un compromis entre la consommation de matière
fissile, et l'amortissement sur une plus ou moins grande quantité d'énergie des frais de fabrication et de retraitement du combustible. Le rendement en énergie des grammes de $^{235}$U ajoutés est extrêmement grand, de sorte que même si les matières premières coûtent très cher, on a presque toujours avantage à viser des taux de combustion élevés.

Il n'est pas particulièrement avantageux d'accroître volontairement les captures résonnantes du thorium, par exemple par sous-modération: le compromis dont il vient d'être question dépend très peu de la valeur du facteur antitrappe. Détériorer $p$ conduit en réalité à accroître l'investissement de matière fissile; il s'agit là d'un compromis de nature différente entre l'immobilisation d'uranium fissile et celle d'eau lourde, qui ne devrait pas conduire à des géométries de cœur très différentes de celles des réacteurs à uranium naturel.

La perte en réactivité due aux flux élevés du fait du protactinium est importante, mais des puissances spécifiques moyennes de 15 à 30 MW/t sont parfaitement envisageables sans pénalisation excessive.

La faible contribution du coût des matières premières dans le coût de renouvellement du combustible fait que celui-ci est très peu sensible au prix de l'uranium. Parce qu'on peut tirer huit à douze fissions de chaque noyau de $^{235}$U ajouté, on peut accepter des prix d'uranium dix ou vingt fois supérieurs aux prix actuels (par exemple 80 ou 100 $/lb de $U_3O_8$) sans que le coût de renouvellement dépasse par exemple 1,5 mill/kWh. C'est là l'intérêt essentiel des cycles thorium que de fournir une solution simple à l'éventuel épuisement des ressources d'uranium à bas prix. Les réserves d'uranium dans le monde sont en effet énormes si l'on accepte des prix très élevés.

Il faut cependant souligner l'importance de l'investissement de matière fissile dans le chargement initial.

Le cycle de combustible est extrêmement sensible au bilan neutronique. Spécialement lorsque l'uranium est cher, il suffit d'accroître d'une faible quantité des captures stériles de neutrons pour que le coût de renouvellement devienne élevé. C'est pourquoi l'avantage essentiel des cycles thorium qui vient d'être souligné valorise énormément les réacteurs à eau lourde, et parmi eux, ceux pour lesquels le choix du refroidisseur, de la conception
mécanique, ou des matériaux de structure permet un bilan neutronique très favorable.

Un attrait important des convertisseurs thermiques à thorium est que la technologie pourrait être presque identiquement la même que celle des réacteurs à uranium naturel. On pourrait même concevoir qu'on remplace les combustibles dans le même réacteur. Même si l'on estime que les cycles thorium, par suite des difficultés ou incertitudes technologiques qu'ils comportent encore, sont moins intéressants pour l'immédiat que les cycles à uranium naturel ou très légèrement enrichi, il est important de savoir que cette possibilité existe.

C'est là un avantage potentiel essentiel des réacteurs à eau lourde, qui apparaissent ainsi particulièrement bien adaptés aux perspectives économiques de l'énergie nucléaire non seulement à moyen terme, mais également à plus long terme.

REMERCIEMENTS

La plupart des calculs cités dans ce rapport ont été exécutés en 1961 par M. de Salvo Brito de la Commission nationale de l'énergie nucléaire du Brésil lors de son stage à Saclay. Nous lui adressons nos remerciements pour sa collaboration.
THORIUM FUEL PROGRAMMES IN THE FEDERAL REPUBLIC OF GERMANY

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INTRODUCTION

The atomic programme in the Federal Republic of Germany shows a substantial interest in the utilization of thorium in power reactors. In the early years of atomic energy development in the Federal Republic of Germany, work was mainly concentrated on the development and construction of normal uranium reactors and activities in the thorium field were limited to special projects. Development work for plutonium breeders started a number of years earlier than for thorium breeders.

1. THORIUM-UTILIZING REACTORS

1.1. Gas-cooled high-temperature reactors

The first thorium-utilizing reactor concept taken up actively was the pebble-bed reactor proposed by R. Schulten. The AVR group (Arbeitsgemeinschaft Versuchsreaktor GmbH), composed of a number of German utilities, ordered a first prototype reactor which is being built by Brown, Boveri/Krupp (BBK) in Jülich. This reactor will use thorium as a fertile material and aims at a high conversion rate and high final values. Whilst the reactor is still under construction, an association between EURATOM, BBK and KFA (Kernforschungsanlage, Jülich), which receives strong financial support from the German Federal Ministry for Scientific Research, has been established. The partners of this association (Thorium High-Temperature Reactor, THTR) are doing development and research work and have made a complete project for a medium-sized (200-300 MW(e)) reactor of this kind. The project will be completed at the end of 1967; it is intended to build, in subsequent years, a larger, advanced prototype. The AVR reactor, which is to run critical at the beginning of 1966, will give important experience which is to be evaluated for the THTR project.

1.2. Heavy-water reactors

Since the year 1960 the Siemens-Schuckertwerke, Erlangen, has made studies on the use of thorium in heavy-water-moderated reactors. Main efforts were burn-up calculations and the economic evaluation of a thorium cycle in a pressurized heavy-water reactor of 400 MW(e). A programme for the calculation of ThO\textsubscript{2}-UO\textsubscript{2} lattices on the basis of the ZEBRA programme is being developed.
1.3. Thorium-breeder studies

The Kernforschungsanlage Jülich has made in the past two years a theoretical evaluation of different thorium-breeder systems. The result showed that the gas-cooled high-temperature reactor, the heavy-water-moderated reactor and the molten-salt reactor are especially promising. For this reason a $2\frac{1}{2}$-yr programme (to end in 1967) has been started, which foresees experimental and theoretical studies for these three systems.

To develop the pebble-bed reactor in the direction of a breeder, it is planned to investigate the possibilities of a two-zone core, in which the fissionable and the fertile material would be contained in graphite balls, which form two zones within the vessel. It is expected that by adequate loading mechanisms separate burn-up and breeder zones can be maintained without a special inner structure in the vessel. A study will be made for the optimization of the corresponding fuel-cycle, which foresees a compromise between high breeding gain and low fuel cycle cost. Special problems are the cooling system for the two zones and a quick burn-up measurement of the single fuel and breeder elements.

The second study will investigate the potential of a heavy-water-moderated breeder, including a comparison of different heavy-water reactor systems for this purpose. Lattice experiments with different $\text{ThO}_2-\text{UO}_2$ fuel elements are planned, including a number of measurements of corresponding thermal neutron spectra and conversion rates. The main work for this reactor system will be the development of special fuel elements and investigations concerning reprocessing possibilities.

The studies concerning the molten-salt reactor will be concentrated on the investigation of epithermal systems of this type including a variation of the MOSEL concept of P. Kasten, presented at the Geneva Conference 1964. It is foreseen to do this work in close contact with Oak Ridge National Laboratory, USA, as the experience with thermal systems of this type will be of high value for the epithermal concept, too.

The three studies will be made from the very beginning in close cooperation with three German companies, namely, BBK (pebble bed), Siemens-Schuckertwerke ($D_2O$) and INTERATOM (molten salt).

2. THORIUM FUELS AND FUEL ELEMENTS

From nuclear-pure thorium oxide the German company NUKEM produced years ago, by reduction with metallic calcium in the presence of $\text{CaCl}_2$, nuclear-pure thorium metal for the research reactor FR-2 at Karlsruhe. This company, in cooperation with the Siemens-Schuckertwerke has begun development work for the production of mixed thorium-uranium oxide pellets for use in heavy-water thorium reactors. For the gas-cooled pebble-bed reactor the production of coated particles containing uranium and thorium carbide or oxides is being prepared. For this purpose the results of the Dragon development work, given to NUKEM, were of great value. For the production of uranium and thorium kernels for the coated particles, the sol-gel process is being studied.
For heavy-water reactors the Siemens-Schuckertwerke together with NUKEM are investigating different fabrication methods for UO$_2$-ThO$_2$ elements.

The KFA, Jülich, is beginning work for the development of fuel elements for pebble-bed breeders comprising mainly irradiation and post-irradiation examination of different fuel elements. Special emphasis is put on the development of such graphite elements, which can be loaded and unloaded with loosely coated particles. Similar irradiation experiments are planned for heavy-water elements.

For the epithermal molten-salt concept, studies have been prepared concerning possible changes of the constituents of salts as they have been developed in Oak Ridge for thermal systems.

3. REPROCESSING

For thorium-breeder systems economic reprocessing methods are of vital interest. Whilst for heavy-water breeders there is comparatively much technical experience and for molten-salt reactors extensive studies have been made by ORNL, for high-temperature gas-cooled reactors in general, and specifically for breeders, corresponding experience is still very limited. As the cost of reprocessing and the following refabrication of fuel elements is decisive for breeder concepts it is foreseen that a number of German companies, in close co-operation with KFA, Jülich, will make investigations which will, at the end of 1967, give an estimate of the technical possibilities and costs for the different fuel elements. At the KFA, Jülich, work has been started for developing special head ends for pebble-bed fuel elements. In addition fluorination of uranium carbide using SF$_6$ and reprocessing in molten salts are being investigated. A special study is being planned concerning the separation of protactinium.

With the exception of the gas-cooled pebble-bed converter the work on thorium-fuelled reactors in the Federal Republic of Germany is still in a very early state. It is hoped that, until the end of 1967, the studies of different thorium-breeder concepts will lead to results of sufficient technical and economic promise, at least for a part of the systems investigated, to continue the efforts by constructing first prototypes.
THORIUM IN PEBBLE-BED REACTORS

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INTRODUCTION

Thorium fuel cycles will be used in the AVR (Arbeitsgemeinschaft Versuchsreaktor, GmbH) reactor constructed at Jülich and in the THTR (Thorium High-Temperature Reactor) which will be designed and developed under a EURATOM contract. Mobile spherical fuel elements are used in these reactors. A continuous fuelling facilitates optimum conversion. Reactivity reserve by additional fissionable material which would cause neutron losses by control rods is not required, and a favourable burn-up factor is to be expected. The fissionable material inventory can be small, a fact which not only influences the capital cost, but makes it possible to maintain a high moderating ratio for optimizing the fuel cycle. During the residence time, the fuel elements will move through the reactor several times, by continuous shuffling. By this means, uniform burn-up will be obtained.

REACTOR LAYOUT

Typical design data for this reactor type are given in Table I.

The arrangement of the pebble-bed reactor results in a relatively low core density of ~1.0 g/cm³ and consequently in a higher neutron leakage which is, however, unimportant for large reactors. The heat transfer in the pebble bed is good. The gas speed in the reactors is relatively low. The blower power is therefore relatively small.

The flattening of the power distribution is a special problem with mobile fuel elements. By checking the temperature profile of the gas leaving the core the charging process can be controlled via different inlet tubes distributed radially by varying the ratio of fuel elements and graphite balls. The fuel concentration can be adjusted in such a way that the resulting temperature profile is sufficiently flattened from a technical point of view. If a mixture of fuel pebbles and graphite balls is used, hot spots may occur which can, however, be tolerated since the elements are temperature-resistant.

Continuous fuelling of the reactor described before is possible with a testing system with which the burn-up and the content of fissionable material and fission products after irradiation in the reactor can be measured. In this way it can be decided whether the irradiated fuel elements are to be re-used in the reactor or discharged and removed.

In the temperature range of the HTR, H₂O, CO, CO₂ and other contaminations of the gas circulation system react, by means of which the graphite in the reactor core is burned and carbon is transported from the reactor
to the cold parts of the loop. The experience gained up to now in experiments indicates that the reactor operation will not be seriously affected if the concentration of the free and chemically bound oxygen is below 20 ppm. The decisive factor for the selection of the gas purity is, above all, the mass transfer of carbon from the hot parts of the reactor to the cooler parts of the steam generator by changing the Boudouard equilibrium to avoid too short a life of the fuel elements and excessive dust production of the reactor system.

FUEL ELEMENT TYPES

The main design data are given in Table II.

Several types of fuel elements are being developed. They can be divided into two groups: one is characterized by a machined electro-graphite shell of the elements, the other by moulded spheres which are pressed from mixtures of graphite powders, coated particles and binders (synthetic elements). The elements with electro-graphite shell are loosely filled with the fuel particles (elements with annular or spherical gaps) or the particles are fixed in the inside of the electro-graphite shell in a thin spherical layer (wallpaper type).

All fuel element types provide a fuel-free graphite shell surrounding the part which contains the coated particles. In those elements in which the particles are not homogeneously distributed in the centre, the internal core is pure graphite moderator.

A graphite sphere with a spherical gap inside is promising. The shell thickness can be 5-10 mm. The strength of this sphere does not depend upon the shell alone because the shell is supported by bridges to the inner sphere, which divide the gap into several cavities which are all connected
TABLE II
PARAMETER FOR FUEL-ELEMENT SPHERES

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel content (g)</td>
<td>0.4-1.0</td>
</tr>
<tr>
<td>Thorium content (g)</td>
<td>8-15</td>
</tr>
<tr>
<td>Max. power per ball (kW(th))</td>
<td>2.5-3.0</td>
</tr>
<tr>
<td>Max. surface temperature (°C)</td>
<td>1000</td>
</tr>
<tr>
<td>Max. fuel temperature (°C)</td>
<td>1350</td>
</tr>
<tr>
<td>Release rate, (^{133}\text{Xe})</td>
<td>(10^{-4})</td>
</tr>
</tbody>
</table>

The gaps are produced by incorporating pieces of collapsible material during the moulding of the spheres. After moulding, the plastic material is volatilized and the production process finished by carbonizing and graphitizing the moulded spheres. With a gap of 2 mm width and a shell of 8-mm thickness, about 25% of the shell can be supported by bridges if a 9-cm\(^3\) gap volume for a loose particle is required.

Hollow-sphere fuel elements have a thin shell of about 10-mm thickness. The thickness of the shell is determined by a compromise between the requirements for heat transfer and strength. The coated particles are arranged in a thin layer of about 1-2 mm adjacent to the internal wall of the hollow spheres; to do this the hollow spheres are filled with a slurry of coated particles, graphite powder, binder and solvent. Subsequently the spheres are tumbled in a special machine to produce an even layer of particles. After complete drying of the layer, the inside of the sphere is filled with natural graphite powder (without any binder) in a special device. After that, the plug is screwed in and fixed with some binder. Then the binder of the particle layer and of the screw is cracked by heating the sphere to 1000°C in argon and to 1500°C in vacuum.

The so-called synthetic fuel elements have two zones: a shell of 2-8 mm, free of fuel particles, and the centre part containing fuel particles. The same graphite matrix is used for the shell and for the centre. First the prefabricated graphite pressing mixture is mixed with the coated fuel particles and then pre-pressed under low pressure to produce the kernel. Afterwards, the shell is pressed around the kernel. Both operations are performed in rubber insert dies. The graphite matrix of the synthetic fuel element differs from that of the electro-graphite fuel element mainly because of the absence of a final heat treatment. In synthetic fuel elements the heat treatment cannot be raised above 1700°C because uranium or thorium diffusion begins through the coating of the particles. Therefore the graphite body must be formed from a mixture of graphite powder and binder. Since the binder in this material is only carbonized and not graphitized, it is desirable to use as little binder as possible. Natural graphite powder is used as grist, together with some artificial graphite powder and sometimes a small percentage of carbon black.
For a pebble-bed reactor several fuel management schemes are possible. Fuel cycles where fissile and fertile elements with different lifetimes are separated in the reactor seem attractive. In this case, the lifetime of the fertile elements with higher fabrication and reprocessing cost is more independent from reactivity requirements. The requirement of reactivity can be met by changing the fissile elements.

The once-through cycle with separated fissile and fertile elements, although certainly not attractive from the point of view of conservation of energy resources, leads to acceptable fuel cycle cost. In addition this cycle at present seems to be feasible without reprocessing. In this case it is useful to start the fertile elements with the equilibrium fissile concentration.

If reprocessing in the fuel cycle is included, the fertile elements can be used first in outer blanket. Afterwards, they should be breeding elements in the reactor core. The bred fissile elements taken out by reprocessing will be manufactured for fuel elements. In this management scheme the most fissile is burned in the fuel elements, which can be reprocessed more often. By this means, an essential part of fission product poisoning can be avoided, which can lead to a higher conversion factor.

There are some advantages to using thorium in the HTR. The high specific power, which is only limited by the protactinium poisoning permits low-fuel inventory. This may be an important factor in the conservation of resources. It also allows attractive low-fuel capital cost. The conversion factor of these systems is for economic reasons a function of cost for reprocessing and refabrication. It is expected to increase the conversion factor step by step, if better and cheaper methods can be developed and more reactors of the HTR type are installed.

At present, economic reasons seem to prevent the use of the HTR breeder. However, there are some indications that the HTR could reach conditions similar to fast breeders as far as fuel consumption in the next 50 yr is concerned. Furthermore, HTRs seem to be capable of using economically the more expensive, practically unlimited resources of ore.
1. INTRODUCTION

The heavy-water-moderated reactors, both of pressure tube design and pressure vessel design are being studied in India. The pressure tube design reactors of the CANDU type have the advantage of cold moderator and bidirectional fuelling. The pressure vessel reactors of the PHWR (Pressurized, Heavy-Water Reactor) type developed in Sweden use high-temperature moderator, but no pressure tubes. It is rather difficult to do fuel shuffling in them. These reactors are based on PWR technology. The PHWR-type reactors have an added advantage. It is possible to change the fuel-to-moderator volume ratio from core to core and this may be useful in plutonium and thorium recycles.

Plutonium recycle and the gradual introduction of thorium are being studied for both types of reactor in India. Necessary computer programmes are being developed. This report discusses the computer programmes DUMLAC, OLBUS and OLBUSTAP which have been developed to study this problem. A few preliminary results using Pu-Th fuel in a PHWR- and CANDU-type reactor, obtained from these programmes, are discussed in section 5 of this paper.

2. DUMLAC

The DUMLAC (Deuterium-Moderated Lattice Calculations) programme, written for CDC-3600, calculates the lattice parameters of heavy-water-moderated lattices for clean as well as poisoned and irradiated conditions. The lattice recipe used in this code is very similar to that used in Chalk River and Sweden. It is based on the four-factor formula and two-group leakage. A brief description of the recipe follows. Reference [1] gives full details of the programme.

2.1. Materials and geometry

The programme can handle either uranium or thorium fuel in the form of metal or oxide. The enrichment can be of $^{233}$U, $^{235}$U or plutonium. Different coolants, such as D$_2$O, organic coolants or H$_2$O, can be used. Similarly, sheathing can be of any material.

The geometry of the cell is always assumed to be cylindrical in the calculations but the input pitch can be either square or hexagonal. The fuel can be in the form of open lattices or clusters of rods. Tubular fuel elements cannot be handled at the present moment. Surrounding the fuel there
can be up to a maximum of six annular regions. This is followed by moderator D$_2$O with slight amounts of H$_2$O in it.

2.2. Fast fission factor, $\epsilon$

This is calculated using the Fleishman and Soodak method \[2\]. The cross-sections for uranium are taken from Booth \[3\], and those for thorium from Driggers \[4\]. For open lattices, a correction for $\epsilon$ is applied to take into account the fast fissions by neutrons escaping from the neighbouring rods.

2.3. Resonance escape probability

Critoph's \[5\] formalism is used to calculate the resonance escape probability. The resonance integral is calculated from Hellstrand's formulae \[6\]. For open lattices an approximate formula is used for the Dancoff correction factor to take into account the shielding by the neighbouring rods.

2.4. Thermal neutron process

The neutron spectrum is to be given by the Westcott model designated by two parameters, namely, neutron temperature $T_n$ and epithermal index $r$.

The neutron temperature in the fuel region may be calculated by using Bighem's empirical formula \[7\] for these clusters which are nearly the same as the CANDU-type clusters. $T_n$ may also be given as an input. The epithermal index $r$ is given as input or it can be calculated by a simple formula.

The thermal utilization factor is calculated using McKay's \[8\] recipe and is based on Kushneriuk's method, which takes into account the thermal sources inside the cluster and in the annular regions.

2.5. Migration area

The fast leakage is calculated by calculating the slowing down area using the simple homogenized formula. The effect of inelastic scattering in the fuel is taken into account in the age calculation. The diffusion area is calculated by using the usual thermal flux, volume-weighted absorption cross-sections and only volume-weighted transport cross-sections.

2.6. Long-term irradiation calculations

The simple burn-up equations are solved by using the Runge-Kutta method. The programme can automatically choose the optimum length of integration step.

The cross-sections of all the nuclei are assumed to be Westcott effective cross-sections except for $^{232}$Th, $^{238}$U for which the epithermal absorption is taken into account through their respective resonance escape probabilities.
For $^{240}\text{Pu}$ the $S_4$ value is calculated using the effective surface of the fuel and the concentration of $^{240}\text{Pu}$. The formula is:

$$S_4 = \frac{1}{\sigma_0} \sqrt{\frac{4}{\pi} \frac{T_{NF}}{T_0}} \text{RI} \sqrt{\frac{\rho}{\sigma_p + C}}$$

$$\sigma_p = \sigma_p + \frac{S_{\text{eff}}}{4V_F N_0}$$

where $\sigma_0$ is the 2200-m cross-section of $^{240}\text{Pu}$, $T_{NF}$ is the neutron temperature in the fuel $T_0 = 293^\circ\text{K}$, $\text{RI}$ is the infinite dilution resonance integral of $^{240}\text{Pu}$, $\sigma_p$ is the potential scattering cross-section of the fuel per $^{240}\text{Pu}$ atom, $S_{\text{eff}}$ and $V_F$ are the effective surface and volume of the fuel, respectively, and $N_0$ is the number of $^{240}\text{Pu}$ atoms per unit volume of fuel. $C$ is a constant.

The fission products are treated by the usual Canadian method, by breaking them into three groups, namely, (i) $^{135}\text{Xe}$ (ii) other high cross-section fission products like Sm and (iii) three pseudo-fission products.

3. PROGRAMMES OLBUS AND OLBUSTAP

The programmes Open Lattice Burn-up Studies (OLBUS) and Open Lattice Burn-up Studies with Thorium and Plutonium (OLBUSTAP) have also been written in FORTRAN language for CDC-3600. The programmes are intended for point reactivity studies of burn-up cycle with various options. They could be used to compute lattice parameters as a function of irradiation or just to find them under clean or saturated poisoned conditions at zero irradiation. To find the build-up or destruction of isotopes a set of coupled differential equations describing the phenomenon of individual isotopes is solved numerically by the Runge-Kutta method. The cross-sections, based on Westcott formalism, are computed at every stage with the isotopic concentrations present at that stage and this set of cross-sections is used in the standard recipe to compute the various lattice parameters, namely, $\epsilon$, $p$, $L_I^2$, $\eta$, $f$, $L_L^2$, $k_{\text{in}}$, $k_{\text{eff}}$ and $B_m^2$.

In the case of burn-up cycle study a table is printed out after each irradiation interval showing the instantaneous and average values of all isotopes (real or pseudo) and xenon and samarium absorption rates per $^{238}\text{U}$ atom and the values of material buckling and other lattice parameters under two sub-headings, namely, instantaneous lattice parameters and average lattice parameters. The average lattice parameters are obtained by averaging the parameters from zero irradiation to the values obtained at the irradiation under consideration. The details of the programmes, usage procedures and equations are described in Refs [9, 10]

4. COMPARISON WITH EXPERIMENTS

Very few experimental results are available using thorium fuel. Argonne National Laboratory has conducted a series of experiments using $^{235}\text{U}$ en-
TABLE I

COMPARISON OF CALCULATED LATTICE PARAMETERS OBTAINED BY OLBUSTAP WITH THE EXPERIMENTAL RESULTS OBTAINED IN THE USA [1] FOR $^{235}$U-ENRICHED ThO$_2$ OPEN-TYPE LATTICES

<table>
<thead>
<tr>
<th>Lattice description</th>
<th>Rod (a)</th>
<th>Rod (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Pellet diameter (cm)</td>
<td>0.594</td>
<td>0.66</td>
</tr>
<tr>
<td>2. Clad rod diameter (cm)</td>
<td>0.785</td>
<td>0.787</td>
</tr>
<tr>
<td>3. Cladding ID/OD Al (cm)</td>
<td>0.613/0.785</td>
<td>0.715/0.787</td>
</tr>
<tr>
<td>4. Fuel density (g/cm$^3$)</td>
<td>8.434</td>
<td>8.2</td>
</tr>
<tr>
<td>5. $^{235}$U enrichment (%)</td>
<td>4</td>
<td>6.67</td>
</tr>
<tr>
<td>6. H$_2$O in D$_2$O (mol.%)</td>
<td>1.5</td>
<td>2.8</td>
</tr>
<tr>
<td>7. Moderator temperature (°C)</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>8. a$_0$=0.9525 cm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Lattice parameters

<table>
<thead>
<tr>
<th>Case</th>
<th>Hexagonal pitch</th>
<th>$V_m/V_f$</th>
<th>$k_\infty$</th>
<th>r</th>
<th>Calculated $B_{m1}^2$ (m$^{-2}$)</th>
<th>Calculated $B_{m1}^2$ (r fixed)</th>
<th>Measured $B_{m1}^2$ (m$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rod (a)</td>
<td>2 $a_0$</td>
<td>9.66</td>
<td>1.3103</td>
<td>0.33</td>
<td>19.61</td>
<td>21.00</td>
<td>21.43</td>
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<tr>
<td></td>
<td>3 $a_0$</td>
<td>23.83</td>
<td>1.4426</td>
<td>0.145</td>
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<td>24.73</td>
<td>24.12</td>
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<tr>
<td></td>
<td>4 $a_0$</td>
<td>43.69</td>
<td>1.4753</td>
<td>0.084</td>
<td>21.796</td>
<td>21.60</td>
<td>21.08</td>
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<tr>
<td></td>
<td>(r=0.14)</td>
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<tr>
<td>Rod (b)</td>
<td>2 $a_0$</td>
<td>7.94</td>
<td>1.4174</td>
<td>0.54</td>
<td>30.24</td>
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<td>32.52</td>
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<td>3 $a_0$</td>
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<td>4 $a_0$</td>
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<td>1.6205</td>
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<td>34.51</td>
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<td>(r=0.25)</td>
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</tbody>
</table>

It is seen from Table I that there is fairly good agreement for larger pitches, while for the smallest pitch the agreement is not very good, especially for the higher enrichment. The reason for this may be the limitations of the simple recipe for the spectrum, which is based on the Westcott formalism, used in the calculation. It is seen that the epithermal index r is rather poor.
## Table II

**Comparison of the Calculated Lattice Parameters Obtained by DUMLAC with the Experimental Results Obtained in USA [12] for 235U-Enriched ThO₂ Clusters**

**Lattice Description**

1. Rod diameter (cm) = 0.587
2. Cladding diameter ID/OD, Al (cm) = 0.610/0.808
3. Centre-to-centre distance between the rods (cm) = 0.9525
4. Calandria tube ID/OD, Al (cm) = 6.191/6.509
5. Density of fuel (g/cm³) = 8.655
6. Enrichment $^{235}$U atom/$^{232}$Th atom = 0.04
7. H₂O in D₂O (mol.%) = 1.5
8. Temperature (°C) = 20

**Lattice Parameters**

<table>
<thead>
<tr>
<th>No. of rods in the cluster</th>
<th>Square lattice pitch (cm)</th>
<th>$k_\infty$</th>
<th>$L_2^2$ (cm²)</th>
<th>$L_2^2$ (cm²)</th>
<th>$B_m^2$ (m⁻²)</th>
<th>Experimental buckling (m⁻²)</th>
<th>$k_{eff}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>19</td>
<td>10.8</td>
<td>1.414</td>
<td>48.6</td>
<td>135.3</td>
<td>20.92</td>
<td>19.70</td>
<td>1.018</td>
</tr>
<tr>
<td>19</td>
<td>13.0</td>
<td>1.432</td>
<td>74.3</td>
<td>129.8</td>
<td>19.41</td>
<td>17.63</td>
<td>1.031</td>
</tr>
<tr>
<td>25</td>
<td>10.8</td>
<td>1.407</td>
<td>38.8</td>
<td>139.0</td>
<td>21.49</td>
<td>20.0</td>
<td>1.022</td>
</tr>
<tr>
<td>25</td>
<td>13.0</td>
<td>1.429</td>
<td>60.3</td>
<td>132.05</td>
<td>20.59</td>
<td>19.44</td>
<td>1.018</td>
</tr>
<tr>
<td>31</td>
<td>10.8</td>
<td>1.368</td>
<td>31.9</td>
<td>178.3</td>
<td>16.76</td>
<td>20.0</td>
<td>0.943</td>
</tr>
<tr>
<td>31</td>
<td>13.0</td>
<td>1.415</td>
<td>49.0</td>
<td>155.5</td>
<td>18.96</td>
<td>19.82</td>
<td>0.986</td>
</tr>
</tbody>
</table>

*a The experimental buckling may be in error because the extrapolation distances have not been calculated properly.

large and this itself suggests that the Westcott recipe is not good. Another reason for the disagreement may be that a proper Dancoff correction in the calculation of resonance escape probability has not been applied. However if one uses a constant r-value for each enrichment (which is the same as a constant η-value) the agreement is better. This is also shown in Table I.

Table II shows buckling and other values for the clustered rod case, using the DUMLAC code. More disagreement is observed here, perhaps
because the experimental values of the buckling were not accurately known; the values were derived from the analysis of single-rod cases [11] where approximate reflector savings were used. In the experiment only the critical height was measured and no flux plots were made for measuring the reflector savings. DUMLAC was used to check UO$_2$ clusters and gave fair results ($k_{\text{eff}}$ between 0.99 and 1.01).

5. THORIUM-PLUTONIUM CYCLE

The computer programmes were used to study the thorium-plutonium cycle in two types of heavy-water-moderated reactor, namely, the PHWR developed in Sweden [13] and the CANDU-type reactor.

For the purposes of this study a reactor with $B^2_a = 1 \text{ m}^2$ has been considered. This corresponds to a reflected core volume of 150 $\text{m}^3$, active core volume of 95 $\text{m}^3$ and is equivalent to a 200-MW(e) CANDU-type reactor in size. The preliminary study does not take into account the core flux distributions and it is assumed that the reflector savings are equal to reflector thickness.

5.1. PHWR case

Table III gives the results for the PHWR case. Using the OLBUSTAP code, two types of fuelling were considered. One is the fixed core case and the other is the ideal core case which may nearly correspond to the bi-directional fuelling of the CANDU type. These two cases give the lower and upper limit to the discharge irradiation. The discharge irradiation, given for the fixed core case is obtained from the point reactivity curve, which may be 10% higher than the actual fixed core irradiation. For the ideal case the discharge irradiation is that obtained from average parameters.

The isotopic composition of the initial feed of Pu enrichment is nearly that obtained from the discharged plutonium of natural uranium PHWR with plutonium recycling in equilibrium condition. This plutonium contains only about 50% of fissile plutonium. If we had considered the plutonium discharged from the CANDU-type reactor (which may contain about 65 to 70% of fissile plutonium) larger irradiations would have been obtained.

The effect of $V_m/V_f$ and of the average specific power of the core has also been studied. It is seen that the effect of specific power on irradiation is not much, which may be because the flux levels are already low (of the order of $10^{13}$ n/cm$^2$ sec). The larger $V_m/V_f (=30)$ gives larger irradiations by burning more plutonium, while the discharged uranium is highest for a $V_m/V_f = 24$.

5.2. 200-MW(e) CANDU-type reactor

Fuelling of a 200-MW(e) CANDU-type reactor with self-generated plutonium and ThO$_2$ has been considered in Table IV. One observes from Table IV that an enrichment of about 2% plutonium would be adequate to attain the same discharge irradiation in ThO$_2$ channels as found in UO$_2$ channels. If $^{233}\text{U}$ and the fissile component of plutonium are taken on an equal footing,
TABLE III

<table>
<thead>
<tr>
<th>( V_m )</th>
<th>( V_f )</th>
<th>FIXED CORE</th>
<th>IDEAL CORE</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Discharge irradiation (MWd/t Th)</td>
<td>Fissile PuO(_2) (kg/t ThO(_2))</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
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<td>Average specific power (kW/kg)</td>
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</tbody>
</table>
TABLE IV

THORIUM FUEL CYCLE IN A 200-MW(e) CANDU-TYPE REACTOR

| Initial feed of total Pu atoms to Th atoms (%) | 1.75 | 2.0 | 2.5 |
| Initial feed of fissile (\textsuperscript{239}Pu + \textsuperscript{241}Pu) atoms to Th atoms (%) | 1.19 | 1.36 | 1.70 |
| Initial $B_{th}$ (m$^{-1}$) | 3.95 | 5.52 | 7.98 |
| Initial $\langle \delta_{cell}/\delta_e \rangle_{ave}$ | 3.02 | 3.24 | 3.68 |
| Ratio of power in a ThO$_2$ channel to that in a UO$_2$ channel for same cell flux as in normal CANDU | 1.48 | 1.51 | 1.57 |
| Discharge irradiation (MWD/t Th) | 4550 | 8775 | 17500 |
| \textsuperscript{239}Pu + \textsuperscript{241}Pu) atoms to Th atoms in discharged fuel (%) | 0.68 | 0.52 | 0.30 |
| \textsuperscript{232}U + \textsuperscript{233}Pa) atoms to Th atoms in discharged fuel (%) | 0.40 | 0.65 | 1.03 |
| Total fissile atoms to Th atoms recovered from discharged fuel (%) | 1.08 | 1.17 | 1.33 |
| Fissile atoms consumed to Th atoms (%) | 0.11 | 0.19 | 0.37 |
| Expected ThO$_2$ channels out of 308\textsuperscript{a} in equilibrium | 242 | 196 | 146 |
| Expected ThO$_2$ core\textsuperscript{a} (%) | 79 | 64 | 48 |

\textsuperscript{a} Under the assumption of no fissile material loss in reprocessing.

one could have about 60% of the core fuelled by ThO$_2$ in equilibrium. The ThO$_2$ core would be supported by its self-bred fissile isotopes and fissile isotopes produced in the rest of the core. By decreasing the discharge irradiation of ThO$_2$ fuel to 4500 MWD/t Th it is possible to increase the loading of ThO$_2$ in the core to 80% in equilibrium condition. It is expected that the equilibrium condition could be achieved in 10-12 yr.

The power in plutonium-enriched ThO$_2$ channels is about 50% higher than that in a UO$_2$ channel for the same cell flux as in normal CANDU. This difference would decrease when more and more $^{233}$U replaces plutonium enrichment.

The preliminary results presented in Table IV are based on the study of first recycle of plutonium with thorium and therefore may not be exact. Further work is continuing on this subject.

6. SUMMARY

A simple recipe to calculate the lattice parameters of heavy-water-moderated lattices has been used to compare the experimental results of $^{235}$U-enriched thorium lattices. As expected, the agreement is not very good, although it is not bad. The results have shown that the recipe has to be improved especially in the calculation of the neutron spectrum. More
experiments using thorium fuel are also needed to improve the calculational methods.

The preliminary studies using plutonium-enriched thorium as fuel show that the discharge irradiation obtained in PHWR-type and CANDU-type reactors is comparable for the same type of fuelling, although the CANDU type seems to be slightly better. Further studies are needed to show this exactly.

REFERENCES

[10] SUBRAMANIAN, T.A., GOPALAN, R., OLBUSTAP, a program to calculate the lattice parameters of heavy water moderated open type of lattices, to be published as an Atomic Energy Establishment Trombay report
PLANNING FOR A NUCLEAR POWER PROGRAMME

The nuclear power has now been established as an alternative to fossil fuels. But to be able to replace fossil fuels as a source of energy, the nuclear power must be installed at a rate which would be commensurate with the overall growth of demand for power. Thus if one assumes that the demand for power doubles every ten years, the nuclear component which is bound to be small in the beginning, must double with a doubling time of three to five years. This would allow a smooth transition from fossil energy to nuclear energy before the scarcity of fossil fuels sets in. The three-year period may turn out to be too small and will perhaps strain the economy because of the conventional fossil fuel-power stations. It is therefore felt that a five-year doubling period for the nuclear component will be a reasonable estimate.

Thus with an initial ratio of 24:1 for conventional sources of power to nuclear power and the above-mentioned values for the doubling time of the nuclear component, the overall energy pattern will be as shown in Table I. Time t = 0 would roughly correspond to year 1971-72 for India. It can be seen that the transition will take about 50 yr, which is quite reasonable.

Any scheme of fuel cycling based on thorium, natural uranium, or breeders must be capable of meeting the fissile material requirements of such a programme and after 50 yr be capable of growing with a doubling time of 10 yr to keep pace with the demand for power. One must also realize that such a transition is going to take place all over the world at about the same time and hence will affect the availability and prices of materials on a global basis.

Various views have been expressed as to the possible methods of meeting this challenge. Dr. Lewis [1] has suggested a fuel cycle based on the Th-233U cycle and CANDU-type reactors. According to him, abundant supplies of natural uranium will be available from sea and rocks, but only at higher and higher prices. But even at a price as high as $250/kg U, reasonable fuelling costs can be realized for thorium reactors started on 235U separated from natural uranium. He has shown that this type of cycle needs less uranium, per initial thermal megawatt of installed power, than the CANDU-type reactors which burn only natural uranium or a fast-breeder reactor started on separated 235U. Dr. Lewis has taken into account in his survey the inventory and the burn-up requirements of one megawatt of installed capacity over its lifetime.

On the other hand, a vigorous research and development programme in the field of fast breeders is under way in the United Kingdom, United States of America, EURATOM and USSR and a fuel cycle scheme based on
### Table 1

**DEMAND FOR POWER**

<table>
<thead>
<tr>
<th>Time (yr)</th>
<th>Total power ($10^6$ kW(e))</th>
<th>Nuclear power ($10^6$ kW(e))</th>
<th>Conventional (thermal and hydro) ($10^6$ kW(e))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>25.0</td>
<td>1.0</td>
<td>24.0</td>
</tr>
<tr>
<td>5</td>
<td>35.4</td>
<td>2.0</td>
<td>33.4</td>
</tr>
<tr>
<td>10</td>
<td>50.0</td>
<td>4.0</td>
<td>46.0</td>
</tr>
<tr>
<td>15</td>
<td>70.8</td>
<td>8.0</td>
<td>62.8</td>
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<tr>
<td>20</td>
<td>100.0</td>
<td>16.0</td>
<td>84.0</td>
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<td>25</td>
<td>141.6</td>
<td>32.0</td>
<td>109.6</td>
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<tr>
<td>30</td>
<td>200.0</td>
<td>64.0</td>
<td>136.0</td>
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<tr>
<td>35</td>
<td>283.2</td>
<td>128.0</td>
<td>155.0</td>
</tr>
<tr>
<td>40</td>
<td>400.0</td>
<td>256.0</td>
<td>144.0</td>
</tr>
<tr>
<td>45</td>
<td>566.4</td>
<td>512.0</td>
<td>54.5</td>
</tr>
</tbody>
</table>

A combination of fast and thermal reactors appears to be taking shape. It therefore becomes necessary to examine critically all the available information about the performance and the future potential of the various alternatives. The guiding principles should be

1. **Minimum requirement of primary nuclear fuel, i.e. natural uranium.**
2. **Maximum utilization of secondary fuels like thorium.**
3. **Capital layout for the programme.**

The following alternatives have been considered.

(a) The first alternative, or Scheme 1, would be the use of CANDU-type reactors alone to burn natural uranium. This alternative would be justified if an adequate amount of natural uranium could be found locally or imported and if the cost of power so-produced would be competitive.

(b) The other alternative would be to have a combination of first-generation reactors which would burn natural or slightly enriched uranium to produce plutonium and to use the plutonium thus produced in the fast breeders. It appears that the capital layout on the reactor proper plus the fuel inventory for the reactor, would be comparable for slightly enriched PWR- or BWR-type reactors and natural uranium CANDU types. Hence, for the sake of comparisons of the various alternatives, first-generation thermal reactors have been assumed to be CANDU types, which make the best use of natural uranium. The fast breeders could be run with the fertile material feed of
depleted uranium or thorium or both. Thus one would have the following three variations of Scheme 2.

Scheme 2(a) which has a feed of depleted uranium alone

Scheme 2(b) which has a feed of thorium alone, plutonium being supplied from the first-generation reactors and

Scheme 2(c) which would be self-sufficient in plutonium and divert surplus neutrons towards $^{233}\text{U}$ production which could be used to start Th-$^{233}\text{U}$ reactors.

(c) The third alternative, designated as Scheme 3, would envisage starting of Th-$^{233}\text{U}$ reactors on the basis of separated $^{235}\text{U}$. This envisages the availability of a diffusion plant or facilities for toll enrichment.

The diffusion plant approach does seem to need a closer examination. A diffusion plant can be considered for producing slightly enriched uranium if it could be proved that total capital investment per kW(e) of first-generation power reactors for PWR-or BWR-type reactors would be lower than that for CANDU-type reactors. Cohen [2] has estimated the capital costs of large diffusion plants which would produce slightly enriched uranium for light-water reactors at $\$11$/kW(e). The figure refers to the time when all the output of a diffusion plant is used up as feed for the reactors. One would get a figure of $\sim$ $\$30$ as the investment in a diffusion plant capable of installing 1 kW(e) every year. This can be compared with an investment of $\sim$ $\$150$ to $\$200$ in a $\text{D}_2\text{O}$ plant for installing 1 kW(e) every year in CANDU-type reactors. Diffusion plants on the other hand are themselves consumers of significant power and give poor utilization of uranium since recovery of $^{235}\text{U}$ will be only about 65%.

For each one of the alternatives, the total uranium required as inventory and feed for the nuclear power programme envisaged earlier (Table I) has been estimated. A mathematical model has been followed and can be explained by means of simple simultaneous differential equations. (See appendix.) The most important parameters on the choice of which the ultimate conclusions are based are also given in Appendix B.

The figures for the fast breeders are the result of a careful examination of the four 1000 MW(e) design studies [3, 4, 5, 6] carried out by different companies in the United States of America, under a USAEC contract.

The figures for first-generation reactors are based on the CANDU design. The figures for the third-generation thermal reactors are based on extrapolations of CANDU. Thus the whole set reflects a conservative approach based on the appraisal of the present state of nuclear technology. It is possible that if reactors like molten salt or a $\text{D}_2\text{O}$-moderated aqueous slurry breeder succeed, a major revision in the evaluation of Scheme 3 may be required. But it has been decided to follow a conservative approach.

This leads to the results given in Tables II and III. Table II gives the pattern of installed capacities in various types of reactors according to each scheme. Table III gives the uranium requirements for each of the alternatives. These are also illustrated by Figs. 1, 2 and 3.
### TABLE II

**INSTALLED CAPACITY IN DIFFERENT TYPES OF REACTORS**

*(in million kW(e))*

<table>
<thead>
<tr>
<th>Time (yr)</th>
<th>Schemes 1 and 3</th>
<th>Scheme 2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Thermal, CANDU-type</td>
<td>First-generation CANDU-type + second-generation fast breeders + third-generation</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Th-(^{233})U thermals</td>
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<tr>
<td></td>
<td></td>
<td>2(a)  (\text{Pu-thermal})</td>
</tr>
<tr>
<td>0</td>
<td>1.0</td>
<td>N(_{\text{th}}^1)</td>
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<tr>
<td>5</td>
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<td>N(_{\text{th}}^1)</td>
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<td>10</td>
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<td>N(_{\text{th}}^1)</td>
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<td>25</td>
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<tr>
<td>30</td>
<td>64.0</td>
<td>N(_{\text{th}}^1)</td>
</tr>
<tr>
<td>35</td>
<td>128.0</td>
<td>N(_{\text{th}}^1)</td>
</tr>
<tr>
<td>40</td>
<td>256.0</td>
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</tr>
<tr>
<td>45</td>
<td>512.0</td>
<td>N(_{\text{th}}^1)</td>
</tr>
</tbody>
</table>

N\(_{\text{th}}^1\): first-generation - thermal reactors  
N\(_{\text{f}}^2\): second-generation - fast reactors  
N\(_{\text{th}}^3\): third-generation - thermal reactors
TABLE III
TOTAL URANIUM REQUIREMENTS
\((10^3 \text{ t})\)

<table>
<thead>
<tr>
<th>Time (yr)</th>
<th>Scheme 1</th>
<th>Scheme 2</th>
<th>Scheme 3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(a)</td>
<td>(b)</td>
<td>(c)</td>
</tr>
<tr>
<td>0</td>
<td>0.185</td>
<td>0.185</td>
<td>0.185</td>
</tr>
<tr>
<td>5</td>
<td>0.86</td>
<td>0.803</td>
<td>0.814</td>
</tr>
<tr>
<td>10</td>
<td>2.49</td>
<td>2.15</td>
<td>2.20</td>
</tr>
<tr>
<td>15</td>
<td>5.76</td>
<td>4.70</td>
<td>4.82</td>
</tr>
<tr>
<td>20</td>
<td>12.31</td>
<td>9.50</td>
<td>9.98</td>
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<td>25</td>
<td>25.42</td>
<td>19.2</td>
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</tr>
<tr>
<td>30</td>
<td>51.64</td>
<td>38.2</td>
<td>40.22</td>
</tr>
<tr>
<td>35</td>
<td>104.0</td>
<td>75.6</td>
<td>80.22</td>
</tr>
<tr>
<td>40</td>
<td>201.9</td>
<td>149.7</td>
<td>159.8</td>
</tr>
<tr>
<td>45</td>
<td>418.7</td>
<td>297.1</td>
<td>318.4</td>
</tr>
</tbody>
</table>

CONCLUSIONS

If the transition from fossil fuels to nuclear power is to be attained within the time schedule mentioned, a large amount of uranium would be required. The total requirements would far exceed the Indian uranium reserves, so a country like India with limited uranium reserves will have to import uranium.

Utilization of thorium will not materially alter the situation; for example, Scheme 3, which is a direct thorium cycle based on separated \(^{235}\text{U}\), needs as much uranium as Scheme 1 which does not use any thorium. Scheme 2, which employs fast breeders, reduces the uranium requirements by about 30%, but here again Scheme 2(b) or 2(c) needs roughly the same amount of uranium as 2(a) and so it is the use of fast breeders and not thorium which helps in this reduction.

The important factor for second- and third-generation reactors is the specific power and not breeding, e.g. in Scheme 3, 80% of the uranium required is accounted by inventory. Similarly, for Scheme 2, the installed capacity of fast breeders and hence \(^{233}\text{U}\) production largely depends on plutonium feed from first-generation reactors and not on self-bred plutonium, due to relatively large doubling times, resulting from the present inventories and breeding ratios. The same is not true however, for the first-generation reactors. Here, because of low inventory of uranium per kW(e) installed (i.e. 0.185 t/kW(e)), 75% of the uranium is needed to feed the reactors (0.108 t/MW(e) yr at 80% load factor).
Table II also clearly indicates that the major load will be shared by first-generation CANDU-type reactors and the fast breeders in the end will carry only about 35% of the load. The share of fast breeders will increase with time, but in the next two or three decades, the first-generation reactors will dominate the scene.

This pattern can change only if some radically improved designs of the fast breeders or thorium-233U reactors become available. If for Th-235U thermal reactors a specific power of 1 MW(e)/kg and a breeding ratio of 1.05 can be attained, uranium requirements would be reduced to $75.3 \times 10^3$ t for the first 40 yr compared to $199 \times 10^3$ t given in Table III. Similarly, if a specific power of 0.33 MW(e)/kg (Pu) and a breeding ratio of 1.5 can be achieved for fast breeders, uranium requirements for Scheme 2(b) will be reduced to $111 \times 10^3$ t for the same period. Hence it is necessary that more attention be devoted to the high performance reactors for thorium to make a significant impact on the uranium requirements.
FIG. 2. Details of Schemes 2(b) and 2(c)

FIG. 3. Requirements of natural uranium
A - Uranium requirement for Scheme 1
B - Uranium requirement for Scheme 3
C and D - The same for 2(a), 2(b) and 2(c)
REFERENCES


APPENDIX A

NOMENCLATURE

- $P_1$: First-generation installed capacity
- $P_2$: Second-generation installed capacity, usually fast
- $P_3$: Third-generation installed capacity, usually thermal
- $B$: Burn-up (tonnes of natural uranium per MW(e) yr)
- $I$: Inventory (tonnes of natural uranium or kilograms of fissile material per MW(e))
- $BG$: Breeding gain (kg/MW(e) yr)
- $F$: Feed (kg/MW(e) yr) for the fast reactors
- $BR$: Breeding ratio
- $LF$: Load factor

1. CANDU-TYPE REACTORS

Inventory of uranium per MW(e) installed = 0.185 t of natural uranium.
Thermal rating = 18 MW/t.

Feed rate of uranium = 0.108 t/yr per MW(e), based on a burn-up of 9000 MWd/t, and load factor of 80%.
The feeding starts 1.5 yr after start-up and irradiated uranium yields 2.65 kg of fissile plutonium.
Time delay in recovering plutonium is 1 yr.

2. FAST-BREEDER REACTORS

Inventory of fissile material = 4.0 kg/MW(e) for (Pu+238U) reactor
Inventory of fissile material = 4.4 kg/MW(e) for (Pu+Th) reactor
Inventory of fissile material = 4.2 kg/MW(e) for (Scheme 2(c)) reactors
Breeding ratio for Scheme 2(a) = 1.4
Breeding ratio for Scheme 2(b) = 1.25
Breeding ratio for Scheme 2(c) = 1.33
kg of fissile material burnt per MW(e) yr at 80% LF for 2(a) = 0.8
kg of fissile material burnt per MW(e) yr at 80% LF for 2(b) = 0.84
kg of fissile material burnt per MW(e) yr at 80% LF for 2(c) = 0.80

3. THERMAL REACTORS: Th-232 U AND Th-233 U

Inventory = 2.7 kg/MW(e) with 233U fuel
Net burn-up = 0.1 kg/MW(e) yr at 80% LF
Inventory = 3.0 kg/MW(e) with 233U fuel
Net burn-up = 0.12 kg/MW(e) yr at 80% LF with 235 U fuel
4. DIFFUSION PLANTS

$^{235}$U recovery = 65%

This assumes a concentration of about 0.25 at.% of $^{235}$U in the waste uranium. This could be attained when the price of uranium would rise very much above the present prices. With $40/kg of natural uranium, the optimum discard fraction is about 0.35 at.% of $^{235}$U.

APPENDIX B

SCHEME 1

Total uranium required = Inventory + Feed = $P(t) \times B + \int_{1.5}^{t} P(t-1.5) \times B \, dt$

SCHEME 2

(a) $P_1 + P_2 = P(t)$, according to Table 1

$$\frac{dP_2}{dt} = \left[ P_2(t-2.5) \times B \times P_1(t-2.5) \times B \times kg(Pu)/t(U \, feed) \right] / I_2$$

If $P_1$ and $P_2$ are known, uranium requirements can be obtained by integrating over the whole period.

(b) $P_1 + P_2 + P_3 = P(t)$

$$\frac{dP_3}{dt} = -P_2(t-1.5) \times B / I_2 + \left[ P_1(t-2.5) \times B \times kg(Pu)/t(U) \right] / I_2$$

$$I_3 \frac{dP_3}{dt} = P_2(t-2.5) \times F \times B R \times P_3(t-1.5) \times 0.1$$

If $P_1$ is known, uranium requirements can be found as in Scheme 1

SCHEME 3

Total uranium required =

$$P(t) \times Sp. \, inventory/yield \, of \, ^{235}U \, per \, t \, (nat. \, U \, feed)$$

$$+ \int_{1.5}^{t} P(t-1.5) \times 0.12 \times dt/yield \, of \, ^{235}U \, per \, t \, (nat. \, U)$$

For all the schemes, fuelling is assumed to start after a 1.5-yr and 1-yr delay, respectively, in reprocessing and refabricating the fuel.
INTRODUCTION

In India, apart from the 380-MW(e) Tarapur nuclear power station, four 200-MW(e) CANDU-type reactors may be built by 1971. These power reactors will produce substantial quantities of plutonium which may be used for exploiting India's vast thorium reserves in either thermal or fast reactors.

The process of selection of the most suitable system for utilization of plutonium or thorium is a difficult one, for it depends on a number of factors such as unit energy cost, capital cost, conservation of fissile resources, national fissile reserves, load demand and availability of technology, which could not be predicted with certainty.

In recent years a number of studies [1-4] on fuel cycles in thermal and fast reactors has been made. Different studies have emphasized different aspects of the problem and consequently have arrived at quite contrary conclusions.

It is our aim to study this problem objectively and with improved and reliable methods of calculations.

THERMAL REACTOR STUDIES

Heavy-water-moderated reactors, both of pressure tube design (CANDU type) and pressure vessel design (PHWR type) are being studied in India. The CANDU-type reactors have the advantage of cold moderator and bi-directional fuelling. They have been studied extensively in the United States of America [5] from the point of view of thorium utilization. The pressure vessel reactors of PHWR type developed in Sweden use high-temperature moderator, but no pressure tubes. It is rather difficult to do on-load fuel shuffling in them. However, it is possible to change fuel-to-moderator volume ratio from core to core in them and this may be useful in plutonium and thorium recycles. Higher specific powers could also be attained in these reactors.

Plutonium recycle and gradual introduction of thorium are being studied for both these types of reactor. The results of the preliminary study have been discussed in Ref. [6]. The problems on which work is continuing are

(a) Plutonium and $^{233}$U recycle with thorium in a 200-MW(e) CANDU-type reactor assuming that there is no external source of fissile material. In the equilibrium condition part of the loading contains natural $\text{UO}_2$. The plutonium produced from it supplements $^{234}$U
produced in ThO₂ fuel. The effect of zoning the core is also being considered.

(b) Seed and blanket concept in pressure tube type reactors.
(c) Similar studies for a 400-MW(e) reactor.
(d) Plutonium and ²³³U recycle with thorium in a PHWR-type reactor. In the equilibrium condition the loading will consist of ²³³U, plutonium and ThO₂ only.
(e) Effect of zoning in PHWR core.

To study these problems lattice burn-up study programmes DLMLAC [7], OLBUS [8] and OLBUSTAP [9] have been made. Two-group diffusion theory [10] and multigroup transport theory [11] programmes have been completed. As discussed in the accompanying paper [6] for under-moderated lattices, further work is being done. Work is also continuing on multi-purpose optimization programmes for these two types of reactors.

Measurement of mixed bucklings and other lattice parameters with natural UO₂ and ThO₂ pencils of rod diameter 1.43 cm have been planned on the experimental side. Experiments with plutonium-enriched UO₂ have been planned also in ZERLINA.

FAST REACTOR STUDIES

Fast reactors based on thorium fuel are likely to be much less subject to the undesirable positive sodium void effects which are still a problem in large Pu-²³⁸U-fuelled fast reactors. To get a better picture of the situation, studies have been made on the reactivity coefficients caused by sodium loss and Doppler effect in large ²³³U-Th-fuelled fast reactors. The results are encouraging and it appears that one can go to very large core sizes of the type required for desalination of sea water based on ²³³U-Th fuel. These results are discussed in Ref. [12]. Conceptual design studies for smaller fast reactors using ²³³U-Th fuel will be made in due course.

Plutonium recycle with thorium in fast reactors is also being studied. The different aspects like nuclear considerations, technical feasibility and the economic potential of this fuel cycle are proposed to be analysed in greater detail. The basic physics data required for such an analysis is being collected. The problem of compiling reliable multigroup cross-sections for thorium is receiving particular attention. The composition and temperature-dependent effective group cross-sections in the resonance region for this element are being evaluated using the latest resonance parameters to study the reactivity coefficients more accurately.

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INTRODUCTION

The selection of a suitable fuel for a very large fast breeder reactor of the type mentioned in connection with the desalination of sea water depends very much on the nuclear properties of the fuel from the point of view of the reactivity coefficients. It has been observed that large sodium-cooled fast reactors based on Pu-238 fuel are subject to positive sodium void reactivity effects. Thorium-233 fuelled reactors are much less subject to the undesirable positive sodium void effects because of their nuclear properties. The fission cross-section of 233U varies more rapidly with energy than does that of 239Pu, and thorium has a much smaller fission cross-section and a higher threshold energy for fission than 238U. As a consequence, the reactivity gain caused by the spectral effect is much smaller with 233U-Th as fuel than that with Pu-238 as fuel. A parametric study of the reactivity coefficients caused by sodium loss and Doppler effect was therefore undertaken and the latest nuclear parameters were collected for this purpose.

The importance of taking into account the interference interaction between the resonances of the fertile and fissile isotopes in the study of Doppler effect in fast reactors has been realized and extensive studies of this effect have been made for typical 239Pu-238U systems. A similar study for the 239Pu-232Th system is of interest when plutonium is to be recycled with thorium in fast reactors. This problem has been studied for some typical core sizes and compositions.

CROSS-SECTION PREPARATION

A 22-group cross-section set [1] was prepared by using the first 13 groups of the YOM (Yiftah, Okrent, Moldauer) set [2] and adding 9 groups for the lower energies. Below group 11 (25 keV), the capture and fission cross-sections of heavy elements are composition-dependent because of the resonance self-shielding effects.

Resolved resonance parameters for 232Th, up to 3 keV obtained at Columbia University [3], were used to evaluate the effective resonance integrals in the narrow resonance approximation using the RE-270 programme of Argonne National Laboratory. Temperature-dependent effective group capture cross-sections for thorium below group 11, covering the unresolved and resolved resonance regions and taking the p-wave contribution also into account in the unresolved resonance region, were obtained. These are discussed in greater detail and tabulated in Ref. [4].

The experimental values of the capture and fission cross-sections for 233U are available only in a limited range of energies. Detailed investigations
to obtain shielded cross-sections for $^{233}\text{U}$ were not made in view of the absence of reliable resonance parameters for this element because of the problem of multi-level effects and also because its contribution to the Doppler effect is probably negligible. The 22-group cross-section set was prepared by using the same values as the YOM set [2] for the first 13 groups and by making some adjustments in the capture and fission cross-sections to obtain reasonable values of $\alpha$ in the lower energy groups. The cross-sections for $^{234}\text{U}$ and fission products from $^{233}\text{U}$ fissions were also prepared to study their effect on the reactivity coefficients. All those cross-section sets are also given in Ref. [4].

For studies involving Pu-Th systems, the temperature-dependent effective group cross-sections in the resonance region, which are influenced by the interference and overlap effects, were evaluated separately for thorium and $^{239}\text{Pu}$ over a number of combinations of the potential scattering cross-sections per thorium atom $\sigma_{p\theta}$ and those per plutonium atom $\sigma_{p\rho}$, to represent a range of Th-to-Pu atom ratios and core compositions for metal and oxide fuels. A simplified method in which each type of interference interaction is treated separately in the calculation, developed by Hwang [5,6], was employed to evaluate the cross-sections. The details of these calculations and the group cross-sections for $^{239}\text{Pu}$ and $^{232}\text{Th}$ in different cases have been tabulated in Ref. [7].

$^{234}\text{U}$-THORIUM-FUELED FAST REACTORS

The parametric studies of the reactivity coefficients were studied in large core volumes corresponding to 6000, 8000 and 15 000 litres. Both metal and oxide fuels were considered, with core volume fractions of 70% sodium, 15% stainless steel and 15% fuel. For comparison purposes one case with volume fractions of 50% sodium, 20% stainless steel and 30% fuel was studied.

The spectral and capture components of the sodium void coefficient are calculated by the perturbation theory method using the cross-sections derived from the ELMOE code; the total sodium void coefficient is evaluated using the REX code. The methods of calculation are similar to those used in Ref. [8]. The results are presented in Table I. It is seen that the spectral and capture components increase with decreasing enrichment, and the addition of $^{234}\text{U}$ and fission products makes them positive. Also they become more positive when evaluated at higher temperature. It is interesting to note that the total sodium void coefficient remains negative, even in the 15 000-litre core, under all conditions.

The Doppler effect which may provide the only reliable and prompt acting negative reactivity feedback towards the safety of a fast reactor, results, for the most part, from neutrons with energies below about 10 keV and therefore its effect is more pronounced in the systems with softer spectrum. The spectrum is softer in the $^{233}\text{U}$-Th metal-fuelled cores because of the lower density of the fuel and higher sodium volume fraction (70%) used in these calculations. The ELMOE programme was used for these calculations and the results are given in Table II. It can be seen from the table that the values of $\Delta k/\Delta T$ obtained for metal-fuelled systems are large, comparable
## TABLE I

COMPONENTS OF SODIUM VOID EFFECT IN $^{233}$U-Th-FUELLED SPHERICAL CORES

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Na (%)</th>
<th>Volume (litres)</th>
<th>$^{232}$Th + $^{234}$U ratio (approx.)</th>
<th>% δk Full-Half</th>
<th>% δk Half - No.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Total</td>
<td>Spectral and capture</td>
</tr>
<tr>
<td>Metal Clean core</td>
<td>70</td>
<td>6 000</td>
<td>6.7</td>
<td>-2.1</td>
<td>-3.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8 000</td>
<td>7.1</td>
<td>-1.7</td>
<td>-2.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>15 000</td>
<td>8.9</td>
<td>-3.7</td>
<td>-1.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oxide Clean core</td>
<td>70</td>
<td>6 000</td>
<td>5.5</td>
<td>-6.8</td>
<td>-3.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>15 000</td>
<td>7.5</td>
<td>-6.0</td>
<td>-3.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-6.8</td>
<td>-3.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-6.8</td>
<td>-3.5</td>
</tr>
<tr>
<td>Metal + $^{234}$U + F. P.</td>
<td>70</td>
<td>15 000</td>
<td>8.0</td>
<td>+0.2</td>
<td>-0.3</td>
</tr>
<tr>
<td>Oxide + $^{234}$U + F. P.</td>
<td>70</td>
<td>15 000</td>
<td>8.0</td>
<td>+0.1</td>
<td>+0.4</td>
</tr>
<tr>
<td>Metal Clean core</td>
<td>50</td>
<td>15 000</td>
<td>11.8</td>
<td>-0.6</td>
<td>+1.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-1.7</td>
<td>+1.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-0.9</td>
<td>+1.3</td>
</tr>
<tr>
<td>Metal (F. P. + $^{234}$U) T = 1500°K</td>
<td>70</td>
<td>15 000</td>
<td>8.1</td>
<td>-1.3</td>
<td>+1.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>-2.8</td>
<td>-5.3</td>
</tr>
<tr>
<td>Metal (F. P. + $^{234}$U) T = 1500°K</td>
<td>50</td>
<td>15 000</td>
<td>11.7</td>
<td>+0.7</td>
<td>+1.8</td>
</tr>
</tbody>
</table>

Note: For the cases in which the total coefficient is evaluated, the blanket is 54 cm thick and has a composition: 20 vol. % $^{232}$Th, 15 vol. % stainless steel and 65 vol. % sodium, and the reflector is 30 cm thick with a composition: 40 vol. % sodium and 60 vol. % stainless steel. Wherever present, atoms of $^{234}$U/atoms of $^{233}$U = 0.4 and fission products (F. P.) correspond to 5 at. % of fuel.
### Table II

**DOPPLER REACTIVITY COEFFICIENTS IN 233U-Th-FUELLED CORES**

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Th/233U</th>
<th>Sodium</th>
<th>- Δk/ΔT x 10^5 for temperatures of:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>300 - 750°K</td>
</tr>
<tr>
<td>Metal</td>
<td>6.0</td>
<td>Yes</td>
<td>1.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>0.09</td>
</tr>
<tr>
<td></td>
<td>6.8</td>
<td>Yes</td>
<td>1.56</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>0.09</td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>Yes</td>
<td>2.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>0.16</td>
</tr>
<tr>
<td>Oxide</td>
<td>4.4</td>
<td>Yes</td>
<td>2.178</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>0.533</td>
</tr>
<tr>
<td></td>
<td>5.5</td>
<td>Yes</td>
<td>2.956</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>0.800</td>
</tr>
<tr>
<td></td>
<td>6.5</td>
<td>Yes</td>
<td>3.822</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No</td>
<td>1.156</td>
</tr>
</tbody>
</table>

In magnitude to those that can be obtained in 239Pu-238U oxide-fuelled systems. It is also seen that this large Doppler coefficient drops to a low value when the sodium is removed from the core. This can be attributed to the sharp decrease in the low-energy spectrum in the core when sodium is expelled. The Doppler coefficients are larger for the oxide-fuelled cores. It appears therefore from the results of reactivity coefficients that 233U-Th-fuelled fast reactors may not pose many safety problems even in very large cores.

Based on these encouraging results, a preliminary conceptual study was made on a 15 000-litre thorium-233U-fuelled, sodium-cooled fast reactor. The core was cylindrical with L/D = 0.31 and had the following volume fractions: 15% fuel (metal or carbide), 5% BeO, 65% sodium and 15% structural materials (8% stainless steel + 5.6% V + 1.4% Ti). The radial blanket had the composition: 65% thorium, 15% stainless steel and 20% sodium, the axial blanket had the same composition as the core (with 233U absent). Both had a thickness of 45 cm each. A reflector with 60% stainless steel and 40% sodium was also considered having a thickness of 30 cm. The 5% BeO was added to the core to overcome the decrease in Doppler coefficient in the absence of sodium. The parameters, such as critical mass, breeding ratio and reactivity coefficients, were evaluated for the "clean" core as well as for the core with equilibrium composition. The same 22-group cross-section set prepared for the preceding studies was employed and the multigroup dif-
### TABLE III

**PERFORMANCE CHARACTERISTICS OF A $^{233}$U-Th-FUELLED FAST REACTOR**

<table>
<thead>
<tr>
<th>Core type</th>
<th>$^{233}$U</th>
<th>Critical mass (kg)</th>
<th>Breeding ratio</th>
<th>Sodium void effect, $-\Delta k$ (%)</th>
<th>Doppler coefficient$^a$, $-\Delta k/\Delta T \times 10^5$ for:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>IBR</td>
<td>TBR</td>
<td></td>
</tr>
<tr>
<td>Clean core Metal fuel</td>
<td>8.29</td>
<td>1757</td>
<td>0.707</td>
<td>1.088</td>
<td>3.3</td>
</tr>
<tr>
<td>Clean core Carbide fuel</td>
<td>8.09</td>
<td>1531</td>
<td>0.660</td>
<td>1.022</td>
<td>3.75</td>
</tr>
<tr>
<td>$^{234}$U + $^{235}$U + F.P. added Metal fuel</td>
<td>7.68</td>
<td>1881</td>
<td>0.735</td>
<td>1.196</td>
<td>1.29</td>
</tr>
<tr>
<td>$^{234}$U + $^{235}$U + F.P. added Carbide fuel</td>
<td>7.16</td>
<td>1707</td>
<td>0.687</td>
<td>1.124</td>
<td>1.58</td>
</tr>
<tr>
<td>Clean core No BeO Carbide fuel</td>
<td>8.43</td>
<td>1967</td>
<td>0.734</td>
<td>1.136</td>
<td>2.87</td>
</tr>
<tr>
<td>$^{234}$U + $^{235}$U + F.P. added No BeO Carbide fuel</td>
<td>8.00</td>
<td>2065</td>
<td>0.770</td>
<td>1.259</td>
<td>0.89</td>
</tr>
</tbody>
</table>

*Note:* Results are for an equivalent spherical core of volume 9000 litres. The blanket in all cases has the composition of the radial blanket. The sodium void effect is computed by removing 50% of the sodium originally present in the core.

$^a$ Revised values are smaller by 15 - 20%.

---

Fusion theory code REX was used for the calculations. Part of the results are presented in Table III. More details about these studies are given in Ref. [9].

The breeding ratios are close to unity and the maximum value obtained is 1.259 for the core with equilibrium composition and without the presence of BeO which was replaced by the fuel. The sodium coefficients are negative in all cases and the Doppler coefficients are also highly negative. One can go to very large sizes of fast power reactors using $^{233}$U-Th as fuel and achieve the advantages of coupling such large power reactors to the desalination plants for obtaining not only power but also pure water from the sea.
The effect of the interference interactions on the reactivity coefficients in $^{239}$Pu-$^{232}$Th-fuelled fast reactors was studied for three core volumes, namely, 1500, 4000 and 8000 litres. The cross-sections of $^{239}$Pu and $^{232}$Th used for calculations were obtained for the different cases as discussed earlier.

The Doppler coefficient calculations were performed with the ELMOE code, using the cross-sections with isolated resonance approximation and with the interference interaction effects included separately. The results of these calculations are given in Table IV. The interference effect makes the Doppler coefficients more negative. The percentage change in the overall $\delta k$ caused by interference interactions in Pu-Th systems seems to be small (4-10%) when compared to similar effects in $^{238}$U-$^{239}$Pu systems.

The spectral and capture components of the sodium void coefficient when half the sodium is removed from the core were evaluated using both cross-section sets for the 4000-litre core. The other cases were evaluated with the cross-sections including the interference effects. The methods employed for calculations were the same as those used for Th-$^{233}$U systems and the results are presented in Table IV. It is observed that the spectral plus capture components of sodium void coefficients become more positive when the interference effects are considered. The values tabulated are only the spectral plus capture components of the sodium void coefficient and do not include the leakage component which is negative. The effect of $^{241}$Pu addition to these systems has not been studied here, but its presence is expected to make the coefficients less negative. This study reveals the importance of taking the interference effects of resonances into account in predicting the reactivity coefficients in $^{239}$Pu-Th fuelled fast systems.

ACKNOWLEDGEMENTS

The author is grateful to Dr. H. H. Hummel and Dr. D. Okrent of the Argonne National Laboratory (ANL) for their suggestions and keen interest in this work and wishes to thank the United States Atomic Energy Commission and the Institute for Nuclear Science and Engineering of ANL for providing the facilities for this work during his stay at Argonne, Ill. Extensive assistance from the staff of the Applied Mathematics Division of ANL in the machine computations is also gratefully acknowledged.
<table>
<thead>
<tr>
<th>Core description</th>
<th>Approximation</th>
<th>Th/Pu</th>
<th>Doppler coefficients, $-\Delta k/\Delta T \times 10^5$ for $300 - 750^\circ K$</th>
<th>750 - 1500$^\circ K$</th>
<th>Sodium void effect (Spectral plus capture components) % $\Delta k$ Full-Half $(1500^\circ K)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4000 litres metal fuel 70 vol. % Na + 15% fuel + 15% stainless steel</td>
<td>Iso Res.</td>
<td>4</td>
<td>0.8891 0.3581</td>
<td></td>
<td>+0.8691</td>
</tr>
<tr>
<td></td>
<td>Interf. Interac.</td>
<td>4</td>
<td>0.9484 0.4001</td>
<td></td>
<td>+1.0674</td>
</tr>
<tr>
<td>400 litres oxide fuel 70 vol. % Na + 15% fuel + 15% stainless steel</td>
<td>Iso Res.</td>
<td>3</td>
<td>1.8851 0.9148</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Interf. Interac.</td>
<td>3</td>
<td>2.0602 1.0476</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1500 litres metal fuel 50 vol. % Na + 32% fuel + 15% stainless steel</td>
<td>Iso Res.</td>
<td>6</td>
<td>0.3522 0.1290</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>Interf. Interac.</td>
<td>6</td>
<td>0.3673 0.1350</td>
<td></td>
<td>+0.9219</td>
</tr>
<tr>
<td>Core description</td>
<td>Approximation</td>
<td>$\text{Th}_\text{Pu}$</td>
<td>Doppler coefficients, $-\Delta k/\Delta T \times 10^5$ for:</td>
<td>Sodium void effect (Spectral plus capture components) % $\delta k$ Full-Half (1500°K)</td>
<td></td>
</tr>
<tr>
<td>------------------------------------------------------</td>
<td>---------------</td>
<td>------------------------</td>
<td>-------------------------------------------------------------</td>
<td>-------------------------------------------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>1500 litres oxide fuel 50 vol % Na+ 32% fuel + 15% stainless steel</td>
<td>Interf.</td>
<td>4 5</td>
<td>17408 0.7133</td>
<td>+0.9816</td>
<td></td>
</tr>
<tr>
<td>8000 litres metal fuel 70 vol % Na+ 15% fuel + 15% stainless steel</td>
<td>Iso Res</td>
<td>5 8</td>
<td>19835 0.8132</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Interf. Interac</td>
<td>5 8</td>
<td>21157 0.8874</td>
<td>+2.6605</td>
<td></td>
</tr>
<tr>
<td>8000 litres oxide fuel 70 vol. % Na+ 15% fuel + 15% stainless steel</td>
<td>Interf. Interac</td>
<td>4 4</td>
<td>3663 1.625</td>
<td>+2.1848</td>
<td></td>
</tr>
</tbody>
</table>
REFERENCES

INTRODUCTION

The Italian programme on the thorium-uranium fuel cycle began in 1960. The first information on the status of the research and development work on the preliminary design of the so-called PCUT (Programme Ciclo Uranio Thoro) facility was available on June 1961 [1]. Additional research and development work and the situation of the programme at the end of the detailed design of the plant were made known at the end of 1962 [2]. A complete review of the programme covering the research and development work in support of the PCUT plant, the description of the plant design and the status of construction and purchase of equipment, as well as some reactor studies and an evaluation of the fuel cycle for an integrated 300-MW(e) power system, was presented in September 1963 [3].

Since 1963, the programme suffered from the general CNEN crisis, which involved both programmes and organization and caused a two years' delay. Now the new five-year plan, recently approved by the Italian Parliament, ensures sufficient funds for the completion of the construction of the Rotondella site and the running of the plant. Late in 1963 and during the first months of 1964, the PCUT programme was submitted, together with all the CNEN programmes for reactor development, for a general check by a joint CNEN-ENEL (Comitato Nazionale per l'Energia Nucleare - Ente Nazionale per l'Energia Elettrica) Commission.

The conclusion of their work was that there is no incentive for the application of the thorium-uranium cycle to boiling water and pressurized water reactors of the present generation in Italy. The programme was consequently on the point of being cancelled; it could survive only if the characteristics of the plant design, which is flexible enough to allow fuel cycling tests with industrial meaning both on the thorium-uranium and uranium-plutonium cycles, were considered.

The most recent evidence indicates that there is a trend for the re-evaluation of the thorium-uranium cycle, but no one at the present time can anticipate any reactor experiment based on thorium-uranium fuel in Italy. The use of the thorium-uranium fuel cycle in Italian reactor experiments is therefore mentioned here merely as a possibility.

Much more realistic appears to be the future utilization of the PCUT facility to re-work thorium-uranium fuels, both in the form of oxide fuels from water reactors and in the form of carbide fuels from high-temperature, gas-cooled reactors, on the behalf of foreign customers interested in the development of these cycles. CNEN is prepared to accept any suggestion in this field and to plan the operation of the plant accordingly.
THE PCUT FACILITY

The construction status of the PCUT facility is as follows.

Civil work

The construction of the process building was nearly completed by Feb. 1965. The office wing is completed except for the internal finishings. The storage pool is completed and lined with stainless steel. Miscellaneous stainless steel linings are being laid down. The auxiliary service building structures are also completed. The waste treatment building and vault construction started on March 1965. As far as the centre facilities are concerned, the centre workshops and warehouse have already been built, the cafeteria, the entrance and the electrical sub-station are under construction.

Chemical equipment

All the process vessels, with a very few exceptions, have already been built and tested. Hot cell vessels are now being installed in the racks; cold make-up area vessels have been delivered to the site ready for installation.

The rack piping is under construction. The chemical drains have been imbedded in concrete. Dissolution system centrifuge and mixed oxalate centrifuges are ready to be installed in the hot cell rack or in the warm cell.

The construction of mixer-settlers has been initiated. Process instrumentation bids have been examined and the orders are ready to be placed. Pool water treatment equipment (strainers, filters, demineralizers, pumps) has been ordered.

Mechanical equipment

The material handling equipment (50-ton shipping cask crane, 10-ton corridor crane, 2-ton cold make-up area crane) is being installed in the process building. The rack removal system and the steel hot cell shielding panels, the shielding doors and the shielding windows are also being installed. The cell plugs are already in place. The shielding casks have been built and inspected. The following pieces of the warm cell equipment have already been built: calcining and sintering furnaces, pellet inspection device (Densitron), pellet loader, helium leak tester furnace, assembly machine.

Orders have been placed for the extruder, cut-off mechanism, end plug welder (these items are to be installed in the warm cell) and for the fuel pin chopper.

Analytical facility and laboratories

The sampling gallery and the hot analytical facility are being ordered from a selected firm. Shielding windows are being installed, temporarily, on the mock-up of the analytical facility. As far as the laboratory equipment is concerned, the longest delivery item, i.e. the mass spectrometer, is being ordered from a selected manufacturer.
Heating and ventilation system

The bids for the heating and ventilation system and for the stack have been examined and the order is being placed.

Utilities

Orders have already been placed for process air and instrument air compressors and for the emergency compressors. The demineralized water equipment is also being ordered.

RESEARCH AND DEVELOPMENT

Research and development in support of the PCUT facility

Limited experimental work was carried out in support of the future PCUT-Elk River fuel operation. It includes,

(a) Experiments on various types of catalysts for the uranium (VI)-uranium (IV) reduction,
(b) Construction of dummy elements to check the mechanical performance of the modified Elk River fuel element design,
(c) Bench tests of the first remote fabrication machines, pellet loader and assembler,
(d) Analytical methods checked for the various sampling solutions and for purity tests on the mixed oxide powders.

Sol-gel process

CNEN chemical laboratory at Casaccia developed a modified sol-gel process which consists of the following main steps: denitrification of the thorium uranyl nitrate by contacting their solution with an aliphatic amine, reduction of the thorium-uranium(VI) sol to the thorium-uranium(IV) status, gelation of the sol by final denitrification as well as by dehydration.

Research on the thorium-uranium carbides

This work is carried out in connection with a CNEN-Dragon Project contract on both the head-end processing of carbide fuels via electrolytical disintegration and the carbide particle fabrication via sol-gel [4].

FUTURE PROGRAMME OF USE OF THE PCUT FACILITY

The planning of the PCUT facility operation is just starting after the decision to go on with the programme.

Elk River fuel experience

The experience with the Elk River fuel should require about two years of operation after the cold start up.
This experience could be usefully combined with the reprocessing of other UO₂-ThO₂ irradiated fuels which would supply a $^{233}$U-$^{235}$U mixture for the uranium make-up to reconstitute the refabricated Elk River fuel. In such a case, the plant would run first with the modified flow-sheet which includes the uranium-thorium partition.

_Possibilities of using the plant for other experiences with thorium-uranium oxide fuels_

This possibility is merely a matter of conjecture pending the definition of CNEN reactor programmes. The following possibilities could be mentioned here:

(a) A version of the heavy-water-moderated reactor which could take into consideration the thorium-uranium cycle,
(b) Use of existing Italian light-water power reactors for some experience with thorium-uranium oxide elements,
(c) Running the plant as a production service for reactors operating on the thorium-uranium cycle abroad.

_Studies on modifications of the PCUT plant to accommodate HTGCR fuels_

As mentioned previously, a research and development work is in progress on behalf of the Dragon Project. This work will supply flow-sheet information and technical data for the assessment which the Dragon Project is conducting jointly with UKAEA and CNEN on fuel cycling.

A parallel work is now being made by CNEN as a preliminary design for the additional provisions required by the PCUT facility to accommodate an integrated process of reprocessing and remote fabrication of graphite carbide type fuels.

The present capacity of the PCUT separation facility is enough for about 3750 MW(th) at 150 000 MWd/t burn-up.

Handling of graphite-carbide fuel requires modifications in the receiving area and an additional hot cell for the head-end treatment. The most suitable head-end process and the most consistent with the PCUT dissolution system would be the combustion of the graphite fuel or a combination of electrolytical disintegration, which allows the recovery of an enriched fraction of the fuel particles and the separation of the bulk of graphite, and combustion.

Remote fuel fabrication requires the substitution of the co-precipitation and extrusion equipment with a sol-gel line, which would be based on the CNEN route for sol-gel (i.e. denitrification by amines, U(VI) to U(IV) reduction and gelation by subtraction of the residual nitrate ions with an amine-alcohol system). A question might arise about the possibility to fit all the fuel element construction equipment into the PCUT fabrication warm cell. Enlargement of the warm cell along its longitudinal axis is possible since the plant design already envisages this future expansion.
Conversion for handling U-Pu fuels

This study is related to the plutonium programme included in the new five-year plan of CNEN. The complete experience of plutonium fuel cycling is not envisaged within the framework of this five-year plan.

A rough evaluation of the expenditure required to convert the plant to the U-Pu cycle was made, taking into consideration several cases from the point of view of the type of fuel elements (geometries, enrichment), the process followed to make mixed oxides and the philosophy for the reprocessing and reconstitution operation (complete decontamination followed by direct fabrication in other facilities or low decontamination followed by remote fabrication in the PCUT warm cell).

The costs range between about $500,000 for fabricating oxide fuels very similar in shape to the Elk River or SENN reactor fuels with the present concepts of the PCUT plant and $1,350,000 in the cases of complete substitution of fabricating equipment and addition of a second and third cycle of decontamination.

Owing to its characteristics, the PCUT plant should be more suitable for low decontamination and remote fabrication of plutonium-bearing fuels.

FUEL CYCLE COSTS

Based on the expected PCUT plant experience, reprocessing and fabrication costs were scaled up to a reprocessing and remote fabrication plant integrated with a 300-MW(e) boiling-water or pressurized-water reactor. This study gave as a result a reprocessing and fabrication cost of $145/kg of stainless-steel-clad mixed-oxide fuel or $160/kg of Zircaloy clad fuel. This figure was used in the CNEN evaluation of the fuel cycle cost in the case that the thorium-uranium cycle would be applied to pressurized- or boiling-water reactors in a range of net power between 300 and 400 MW(e).

The actual figure of the PCUT direct operating cost was also used for the evaluation of the SENN (150-MW(e) boiling-water reactor) fuel cycle, if converted to the thorium-uranium cycle. All these costs were compared with the fuel cycle costs of reactors of the same type loaded with slightly enriched uranium. Operating data of such reactors, specific costs, and the resulting fuel cycle costs are summarized in Tables I and II. The calculation was based on the following hypothesis:
(a) Rate of interest on the capital: 7% annual
(b) Fuel lease: 4.75% annual on the initial value
(c) Plant life: 20 yr
(d) Time for the fuel fabrication: 1 yr
(e) Time for cooling, transport, reprocessing and conversion of irradiated slightly enriched uranium fuel: 9 months
(f) Fuel stock, none in all the cases
(g) In the case of fuel lease for the SENN reactor working on the thorium-uranium fuel cycle, the fuel consumption for each cycle has been considered equal to the value of the required 235U make-up
### COMPARISON OF OPERATING DATA AND COSTS FOR BOILING-WATER REACTORS

<table>
<thead>
<tr>
<th>OPERATING DATA</th>
<th>BWR (General Electric)</th>
<th>BWR (Allis-Chalmers)</th>
<th>BWR (SENN)</th>
<th>BWR (SENN)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel cycle</td>
<td>slightly enriched U</td>
<td>U-Th</td>
<td>slightly enriched U</td>
<td>U-Th</td>
</tr>
<tr>
<td>Net power (MW(e))</td>
<td>313</td>
<td>305</td>
<td>150</td>
<td>150</td>
</tr>
<tr>
<td>Thermal power (MW(th))</td>
<td>1008</td>
<td>884</td>
<td>506</td>
<td>506</td>
</tr>
<tr>
<td>U or U + Th inventory (kg)</td>
<td>68500</td>
<td>30000</td>
<td>46000</td>
<td>46000</td>
</tr>
<tr>
<td>Average burn-up (Mwd/t)</td>
<td>15000</td>
<td>14000</td>
<td>20000</td>
<td>20000</td>
</tr>
<tr>
<td>Loading</td>
<td>batch 1/3</td>
<td>batch 1/3</td>
<td>batch 1/3</td>
<td>batch 1/3</td>
</tr>
<tr>
<td>Cladding material</td>
<td>Zircaloy-2</td>
<td>Zircaloy-2</td>
<td>Zircaloy</td>
<td>Zircaloy</td>
</tr>
<tr>
<td>Specific power (kW/kg)</td>
<td>14</td>
<td>29</td>
<td>11</td>
<td>11</td>
</tr>
<tr>
<td>Life-time of cycle (d)</td>
<td>400</td>
<td>180</td>
<td>450</td>
<td>450</td>
</tr>
<tr>
<td>Initial enrichment (235U wt.%) 1st third</td>
<td>2.02</td>
<td>3.5</td>
<td>2.6</td>
<td>2.5</td>
</tr>
<tr>
<td>Final enrichment (235U wt.%)</td>
<td>0.82</td>
<td>-</td>
<td>1.1</td>
<td>-</td>
</tr>
<tr>
<td>U make-up, 90% enriched (kg)</td>
<td>-</td>
<td>60 (eq.)</td>
<td>-</td>
<td>33 (eq.)</td>
</tr>
<tr>
<td>Pu produced (kg)</td>
<td>150.6 (eq.)</td>
<td>-</td>
<td>60 (eq.)</td>
<td>-</td>
</tr>
</tbody>
</table>

### FUEL COST ITEMS

| Direct fabrication cost ($/kg) | 120 | 120 | 120 | 120 |
| Fuel (new) transport cost ($/kg) | 5   | 5   | 5   | 5   |
| Reprocessing and remote fabrication cost ($/kg) | 180 | 180 | 180 | 180 | 210 |
| Reprocessing and conversion cost ($/kg) | 35 | - | 35 | - |
| Irradiated or refabricated fuel transport cost, insurance included ($/kg) | 25 | - | 20 | 15 |
| Plutonium credit ($/kg) | 7000 | - | 7000 | - |
| $^{233}$U + $^{235}$U credit ($/kg) | - | 12000 | - | 12000 |
| Annual interest (%) | 7 | 7 | 7.00\(^a\) | 7.00\(^a\) |
| | | | 4.75\(^b\) | 4.75\(^b\) |

**FUEL CYCLE COST**

| Fuel consumption and interests (mill/kWh) | 1.19 | 1.02 | 1.83\(^a\) | 1.11\(^a\) |
| Fabrication or, in the case of Th-U, reprocessing and relabification (mill/kWh) | 1.58 | 1.79 | 1.37 | 1.92 |
| Transport, reprocessing and conversion (in case of U) or transport (in case of Th-U) (mill/kWh) | 0.57 | - | 0.41 | 0.19 |
| **TOTAL FUEL CYCLE COST (mill/kWh)** | 3.34 | 2.81 | 3.61\(^a\) | 3.22\(^a\) |

\(^a\) Private ownership

\(^b\) Lease
TABLE II

COMPARISON OF OPERATING DATA AND COSTS FOR PRESSURIZED-WATER REACTORS

<table>
<thead>
<tr>
<th>OPERATING DATA</th>
<th>PWR (Westinghouse)</th>
<th>PWR (B &amp; W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel cycle</td>
<td>slightly enriched U</td>
<td>U-Th</td>
</tr>
<tr>
<td>Net power (MW(e))</td>
<td>360</td>
<td>405</td>
</tr>
<tr>
<td>Thermal power (MW(th))</td>
<td>1200</td>
<td>1355</td>
</tr>
<tr>
<td>U or U + Th inventory (kg)</td>
<td>54 000</td>
<td>56 000</td>
</tr>
<tr>
<td>Average burn-up (MWd/t)</td>
<td>24 000</td>
<td>29 000</td>
</tr>
<tr>
<td>Loading</td>
<td>batch 1/3</td>
<td>batch 1/3</td>
</tr>
<tr>
<td>Cladding material</td>
<td>Zircaloy-2</td>
<td></td>
</tr>
<tr>
<td>Specific power (kW/kg)</td>
<td>22</td>
<td>24</td>
</tr>
<tr>
<td>Initial enrichment ((^{235})U wt. %)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1st third</td>
<td>3.2</td>
<td>3.4</td>
</tr>
<tr>
<td>2nd third</td>
<td>3.6</td>
<td>3.4</td>
</tr>
<tr>
<td>3rd third</td>
<td>4.0</td>
<td>3.4</td>
</tr>
<tr>
<td>4th third</td>
<td>3.0 (eq.)</td>
<td></td>
</tr>
<tr>
<td>Final enrichment ((^{235})U wt. %)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1st cycle</td>
<td>1.70</td>
<td></td>
</tr>
<tr>
<td>2nd cycle</td>
<td>1.61</td>
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<tr>
<td>3rd cycle</td>
<td>1.83</td>
<td></td>
</tr>
<tr>
<td>4th cycle</td>
<td>1.11 (eq.)</td>
<td></td>
</tr>
<tr>
<td>U make-up, 90% enriched (kg)</td>
<td>---</td>
<td>160 (eq.)</td>
</tr>
<tr>
<td>Pu produced (kg)</td>
<td>164</td>
<td>---</td>
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FUEL COST ITEMS

<table>
<thead>
<tr>
<th>Cost Item</th>
<th>PWR (Westinghouse)</th>
<th>PWR (B &amp; W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direct fabrication cost ($/kg)</td>
<td>120</td>
<td>120</td>
</tr>
<tr>
<td>Fuel (new) transport cost ($/kg)</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Reprocessing and remote refabrication ($/kg)</td>
<td>-</td>
<td>180</td>
</tr>
<tr>
<td>Reprocessing and conversion ($/kg)</td>
<td>35</td>
<td>---</td>
</tr>
<tr>
<td>Irradiated or refabricated fuel transport cost, insurance included ($/kg)</td>
<td>25</td>
<td>---</td>
</tr>
<tr>
<td>Plutonium credit ($/kg)</td>
<td>7000</td>
<td>---</td>
</tr>
<tr>
<td>(^{231})U + (^{235})U credit ($/kg)</td>
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<td>12 000</td>
</tr>
<tr>
<td>Annual interest (%)</td>
<td>7</td>
<td>7</td>
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TABLE II (cont.)

<table>
<thead>
<tr>
<th></th>
<th>PWR (Westinghouse)</th>
<th>PWR (B &amp; W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FUEL CYCLE COST</td>
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<td></td>
</tr>
<tr>
<td>Fuel consumption and interest (mill/kWh)</td>
<td>1.48</td>
<td>1.28</td>
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<tr>
<td>Fabrication or, in case of U-Th, reprocessing and refabrication (mill/kWh)</td>
<td>1.01</td>
<td>1.09</td>
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<tr>
<td>Transport, reprocessing and conversion (in case of U) or transport (in case of U-Th) (mill/kWh)</td>
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</tr>
<tr>
<td>TOTAL FUEL CYCLE COST (mill/kWh)</td>
<td>2.85</td>
<td>2.37</td>
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</table>

(h) For all the examined reactors working on the thorium cycle, the first reloading envisages a fresh fuel fabrication outside the integrated plant, except the case of the Allis-Chalmers reactor type design which envisages two fresh reloadings. The fresh reloadings form, in the case of the thorium-uranium cycle, together with the first core, the minimum fuel inventory.

A similar evaluation was carried out independently by ENEL on more or less the same cases, with some simplifying assumptions. Its conclusions are less favourable to the uranium-thorium fuel cycle. For instance, the fuel cycle cost on the SENN reactor is said to be 2.30 - 2.74 mill/kWh if operating the reactor on the slightly enriched uranium cycle, and 2.61 - 3.05 mill/kWh if operating the reactor on the thorium cycle.

Both evaluations agree in that only a small difference exists between the costs of the two cycles. They could be questionable according to the values of the parameter assumed and to the calculational method.

REFERENCES

STATUS OF THE EXPENDITURE OF THE PCUT PLANT ON JUNE 1965

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3. Interest during construction | --- | 490,000

Total capital cost | 9,886,250
At the end of 1956 the Japan Atomic Energy Research Institute became interested in thorium utilization in power reactors. At that time the Japan Atomic Energy Commission pointed out, in a long-range power reactor development programme, that research and development on a breeder should be made in Japan; the aqueous homogeneous reactor with thorium was considered for development as a thermal breeder by the Japan Atomic Energy Research Institute mainly because of its inherent merits, such as its high breeding ratio and the low fuel inventory expected.

In 1957 the Japan Atomic Energy Research Institute proposed the semi-homogeneous reactor concept, in which fissile and fertile materials in the form of powdered oxide or carbide are dispersed in the moderator graphite and which attains higher coolant gas temperature because of the composition of its core materials. This type of reactor might be very promising with the thorium cycle.

In the above two programmes, basic and supporting research and engineering development work were carried out; critical assemblies of both types were constructed to study and check theoretical analysis, reactor design, reactor kinetics, and so on. Engineering experiments, such as those on fuel preparation, flow and heat transfer of slurry or boiling slurry, etc. for the aqueous homogeneous reactor, and on fuel manufacturing, fission-fragment behaviour in the graphite, helium purification problem, etc. for the semi-homogeneous reactor were also made.

On the other hand, chemical and spectrographic analysis methods on the trace elements in thorium were investigated for four years and the information was compiled by the Thorium Analysis Committee, Japan Society of Newer Metals in 1963.

These two programmes were stopped one or two years ago for many reasons; this might be considered unfortunate. However, some basic studies, such as measurement of $\sigma_a(E)$ of thorium in the semi-homogeneous reactor critical assembly, construction of the gas loop in the JRR-2(CP-5 type research reactor), properties of thorium carbide and so on are now being carried out.

In 1963 the Japan Atomic Energy Commission selected the heavy-water-moderated power reactor concept for the Japanese power reactor project, survey work and 300-MW(e) plant design study of four types of different coolants were performed to evaluate each type of reactor and to identify clearly the
THORIUM FUEL RESEARCH IN JAPAN

problems to be resolved. Recently, the Japan Atomic Energy Commission re-evaluated the reactor concept from the standpoint of the advanced converter, which has a higher conversion ratio and a possibility of attaining the thermal breeding for which thorium may always be considered. The type of advanced converter to be developed in Japan may be selected by the Japan Atomic Energy Commission in the near future. The evaluation of thorium in the advanced converters is to be made this year by the Japan Atomic Energy Research Institute and at the same time research on thorium fuel will be re-evaluated from this standpoint.

1. SEMI-HOMOGENEOUS REACTOR

After the basic concept of the semi-homogeneous reactor was proposed, research and development work was started. Many of these studies were made with uranium dioxide; however, these results might be applicable to a thorium system in this type of reactor. Development of thorium utilization in power reactors would benefit from technological data available from other reactor types. Given below is an outline of the general ideas of our research for this project.

a. Critical assembly (Fig. 1)

The semi-homogeneous reactor critical assembly was planned to obtain reactor physics data on semi-homogeneous enriched uranium- (and thorium-) graphite systems, and thereby contribute to the design of this type of reactor; it went critical in Jan. 1961. With this assembly, detailed data on criticality of many core loadings, neutron and power distribution, deviation of the resonance neutron spectra from the 1/E distribution, etc. were measured and obtained. At the same time, the theoretical analysis and calculation method were also developed.

The pulsed neutron source was set in the assembly and the reactor kinetics was studied. The reactivity worth of control rod and fuel element, $\beta_{eff}/1$ (changed by only about 20% though the ratio of U/C and the critical mass were greatly changed), etc. were measured; theoretical analysis and measurement methods were also developed.

b. Fuel

The manufacture of the fuel was studied by optical, electrical and X-ray microscopes and by autoradiographs, etc. to get the desired uniform dispersion of uranium in the fuel; one of the best methods of uniformly mixing $\text{UO}_2$ and graphite particles was established.

The reaction between $\text{ThO}_2$ and graphite, as well as the physical properties of $\text{ThC}_2$, were studied. The manufacture of thorium metal-graphite and thorium carbide-graphite fuels was studied and the properties of their pellets, such as thermal conductivity, density, thermal expansion coefficient, etc. were measured. The fuel assembly samples of the semi-homogeneous reactor are shown in Fig. 2.
FIG. 1. Semi-homogeneous reactor critical assembly

FIG. 2. Semi-homogeneous reactor fuel element sample
The behaviour of fission fragments in the graphite was studied with the samples irradiated in the JRR-1 (water boiler type research reactor); it was observed that their behaviour varies considerably according to the kinds of graphite, and the volatile fission fragments diffuse more rapidly in natural graphite than in the artificial one.

The grain-size uranium oxides or carbides were first considered to be of the order of 5 µm or less, so that with a small mixture ratio between the graphite and $^{235}U$ or $^{233}U$, most of the fission products are trapped and retained in the graphite, thereby permitting the fuel to be reprocessed by a simple method (mechanical separation). However, the reprocessing study of this problem revealed that this idea could not be accomplished.

c. Design

Design and survey studies on the semi-homogeneous reactor were stressed for the thorium cycle rather than uranium-plutonium cycle, and a 31-MW(th) (11-MW(e)) plant design was evaluated.

d. Miscellaneous

As regards the helium purification system, the active charcoal, alumina and molecular sieve were tested or studied to remove the volatile fission fragments.

Low-permeabilité graphite ($10^{-6}$ cm²/sec) was manufactured by using furan or tar. The properties of another graphite, known as glass carbon, which has low gas permeability and high strength, were studied. Non-destructive test methods on graphite were developed to check the defect of graphite and strength; it is of interest to estimate the graphite strength from the modulus of elasticity by measuring the sonic speed in the graphite. The alloy 36 Ni-Fe was considered best for brazing material for the graphite.

2. AQUEOUS HOMOGENEOUS REACTOR

In the early stages of the research programme, development of a two-region type reactor was planned but the unhappy results of the HRT affected this reactor programme. Effort was then shifted to a one-region boiling slurry reactor. Outlines of the research are as follows:

a. Reactor physics

The two-region type critical facility (Figs. 3 and 4) was constructed to obtain reactor physics data on a heavy-water homogeneous system. The facility became critical with the heavy-water reflector in June 1961 and with the ThO₂ slurry blanket in August 1963.

The critical experiments and theoretical analyses were performed over a wide range of $^{235}U$ concentration in the core and ThO₂ concentration in the blanket. The theoretical effective multiplication factor is decreased by treating the leakage of fast neutrons from the core rigorously, and is increased
by using spatial-dependent effective cross-sections. These treatments are justified by the agreement found between the calculated and measured values for the cadmium ratio and the thermal flux distribution. The disagreement seen in the effective multiplication factor between the theoretical and experimental values may be attributed to uncertainty in the resonance integral of $^{235}\text{U}$. The discrepancy in the effective multiplication factors does not exceed 1%, when the accepted value of $\nu I_f - 1$ is reduced to a smaller value. The resonance capture integral of $^{232}\text{Th}$ also was measured by the activation method.

b. Properties of thorium oxide and preparation of its microspheres

Microspherical particles of $\text{ThO}_2$ and $\text{ThO}_2-\text{UO}_2$ were considered preferable for the slurry reactor. The method adopted to obtain the $\text{ThO}_2$ microsphere is as follows: Thorium hydroxide slurry containing a small
Aqueous homogeneous reactor critical assembly

amount of thorium nitrate is heated in the water bath for several hours and a yellow sol is made. The spherical thorium hydroxides are then made by spraying this sol through a nozzel into isopropyl alcohol. The ThO₂ microspheres (Fig. 5) are obtained by heating them to a temperature greater than 260°C. Integrity of the microsphere was studied and was found to be adequate in out-of-pile test.

ThO₂ particles flocculate when suspended in distilled water and the addition of NaOH to the suspension promotes the flocculation. The sedimentation rate of commercial ThO₂ particles in water was observed to change markedly from $10^{-1} - 10^{-2}$ cm/s to $10^{-3} - 10^{-4}$ cm/s with about 7% ThO₂ concentration. At lower concentrations it was found that floc is 50 - 60 μm in diameter and the void fraction in the floc is 0.90 - 0.95.

c. Flow and heat transfer of slurry

The mock-up and test loops (Fig. 6) were constructed to test the technical feasibility of the boiling slurry concept and to get supporting data for designing the reactor. The preliminary test showed that the flow and boiling region could stay stable, and few particles escaped from the free surface.

The heat transfer of the slurry and the pressure drop of the three phases (steam-water-particles) were also studied. It was found that the heat transfer coefficient to the high concentrated slurries should be proportional
FIG. 5. Thorium oxide microsphere

FIG. 6. Mock-up test loop for slurry flow experiment
3. PRESENT STATUS

$\sigma_a(E)$ of thorium deviates a little from the $1/v$ shape mainly because of the negative resonance level, which affects the temperature coefficient of the reactor; it will be measured and studied in the semi-homogeneous reactor critical assembly soon.

The experiments on reactor physics (reactivity worth, cross-section check, etc., on thorium) will be made in TCA (light-water-moderated critical assembly) by inserting about 30 thorium fuel rods, so that reactor physics design data on thorium utilization in the JPDR (BWR type) may be obtained.

As thorium is important for the advanced converters because of its inherent properties, an evaluation of thorium in different types of advanced converters, such as the relation between burn-up and fuel composition, effect of fuel loading scheme and fuel type on burn-up or reactivity, etc., will be made this year.

Because of the importance of thorium for power reactors, especially for the advanced converters, engineering research on thorium fuel is being re-evaluated at the Japan Atomic Energy Research Institute, and will be re-organized in the near future.
INTRODUCTION

The importance of breeder reactors for future large-scale nuclear energy production is generally recognized [1]. With the thorium cycle, thermal as well as fast breeders are possible. In thermal breeders the investment of fissile material per installed kilowatt can be much lower than in fast breeders.

For thermal breeders, homogeneous heavy-water reactors can achieve the highest breeding ratios, especially if a continuous removal of $^{135}$Xe is possible. The consequence of an aqueous homogeneous reactor system is the introduction of a liquid fuel with its merits and disadvantages.

1. HOMOGENEOUS LIQUID FUEL REACTORS IN RELATION TO THE THORIUM CYCLE

The highest conversion factors for thermal reactors, using the thorium cycle, can be reached in two-region homogeneous reactors with $^{233}$U in the core and $^{232}$Th in the blanket.

In a one-region reactor the capture of thermal and epithermal neutrons in $^{233}$Pa reduces the conversion factor.

However, many technological and constructional arguments favour a one-region reactor. The difficulties of both types are briefly listed below.

(a) Difficulties related to a two-region reactor

(1) Two different and separate fuel circulation systems are required.
(2) Two different chemical reprocessing cycles are required, one for the purification of the fissile material in the core and one for the separation of $^{233}$U from the fertile material in the blanket.
(3) A "neutron window" with a low neutron absorption between the core and the blanket is necessary.
(4) The fissile fuel concentration in the core must be very low (about 10 g/l). To keep this concentration and its distribution constant only a solution form seems practical. However, possible phase separations at high temperatures might cause difficulties.
(5) The fertile material concentration in the blanket must be very high (about 1000 g/l). For such a high concentration only a suspension is possible, because of the limited solubility of thorium and uranium salts in water. These highly concentrated suspensions have difficult rheological properties.
(b) Difficulties related to a one-region reactor

(1) A reduction of the conversion factor must be accepted owing to the presence of $^{233}\text{Pa}$ in the core at a high neutron density.

(2) The necessary concentration of fissile and fertile material in the core (about 400 g/l) again requires a suspension as in the case of the blanket of a two-region reactor. As the concentration in the one-region reactor is lower by a factor of two to three, compared with the blanket concentration mentioned above, the rheological properties cause no more serious difficulties. However these suspension particles are present in a high fission density field. The radiation damage of the specific suspension particles under consideration must be kept within reasonable limits, so that either in situ recovery or a reprocessing will be acceptable.

Apart from the difficulties mentioned above for both types of reactor two other main problems exist. First, the complete leak tightness of the fuel and fertile circulation systems must be guaranteed. Secondly, because of the large amount of radioactivity circulating in the reactor systems, remote maintenance is a strict requirement.

On the other hand it should be realized that the application of a liquid fuel, either a solution or a suspension, simplifies very much the fuel fabrication, so that remote fabrication is possible and low fabrication costs are expected. Furthermore a frequent reprocessing of a thermal thorium breeder reactor is necessary. With a liquid fuel and a remote fuel fabrication facility, a simple, fast and only partial reprocessing is acceptable without long cooling times.

Another point of interest in homogeneous liquid fuel reactors is that, although the $^{232}\text{Th} - ^{233}\text{U}$ cycle is of primary importance, from a neutron physics point of view no objection exists against a start-up of the system with a $^{232}\text{Th} - ^{235}\text{U}$ or even a $^{232}\text{Th} - ^{241}\text{Pu}$ fuel. In both cases the initial conversion ratio will be lower than unity. The fuel which is homogeneously distributed in the liquid permits a simple adjustment of the criticality by adding additional fissile material during the first fuel cycle. As the temperature coefficient can be adjusted to a prompt negative value in an undermoderated reactor, because of the prompt thermal expansion of the water, neither the specific epithermal neutron captures nor the change in the yield of the delayed neutrons are important for the kinetic behaviour of the reactor.

With respect to point b.(1) of this section one special remark is of interest. The loss of $^{233}\text{Pa}$ is proportional to the neutron capture in $^{233}\text{Pa}$ and, as far as it is related to the thermal neutron flux, it is also proportional to the power density in the core vessel. In two respects, however, the situation is more favourable in an aqueous homogeneous reactor than in a thermal heterogeneous reactor. First, flux peaking will not be present in a homogeneous reactor, although the buckling related to the neutron leakage remains. Secondly, for heat extraction purposes a part of the circulating liquid fuel will be outside the core vessel. By optimizing a conceptional design of a 600 MW(th) power reactor it was found that the suspension volume outside the core vessel should be about equal to the volume of the core vessel proper. The average neutron flux to which $^{233}\text{Pa}$ is subjected is therefore also about half of the average neutron flux in the core.
For such a large single-region heavy-water-moderated reactor fuelled with a $^{233}\text{UO}_2-^{232}\text{ThO}_2$ suspension, the reaction rate $^{233}\text{Pa} \to ^{234}\text{U}$ (relative to the reaction rate $^{232}\text{Th} \to ^{233}\text{Pa}$) has been calculated as a function of the neutron flux for a moderator temperature of 180°C. The results are given in Table I. In column 2 the relative reaction rate $^{233}\text{Pa} \to ^{234}\text{U}$ is given as a function of the thermal flux (Maxwellian distribution). For the absorption cross-section of $^{233}\text{Pa}$ at 2200 m/s a value of 100 b is taken. Column 3 gives the relative reaction rates when the epithermal neutrons are taken into account also. For the resonance integral of $^{233}\text{Pa}$ the value of 1230 b is taken [2], with $\phi_r (\ln E)/\phi_{th} = 1/12$. It may be concluded from Table I that the influence of the epithermal neutrons is relatively large, and that the loss in conversion ratio at a power density of 40 kW/l is less than 5%, since the suspension is only part time in the core vessel. The total investment in such a reactor including the external heat exchanger is only 0.25 g $^{233}\text{U}$/kW.

Columns 4 and 5 show power density and breeding ratio, respectively. It may be seen that the breeding ratio is larger than unity for flux values up to $10^{14}$ n/cm$^2$ s, corresponding to a power density in the core of 37 kW/l. It should be mentioned, however, that the calculations have been made without taking into account neutron leakage and poisoning by fission products.

### Table I

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2. GENERAL OUTLINE OF THE FUEL TEST FACILITY

The primary object of the aqueous homogeneous suspension reactor project is the development of a liquid suspension that will be an acceptable fuel for a power breeder or near breeder reactor.

Apart from production methods of suspension particles, to be discussed in a separate paper, and the fundamental research of their properties and behaviour (section 3), much physical research (section 4) and technological development work (section 5) were needed. For these reasons a near-critical low-temperature suspension reactor was constructed and operated for a long time.
Moreover, five high-temperature and high-pressure loops have been used and are still in use for learning more about the behaviour of the suspension in a circulating system, for developing suitable components, for hydraulic and erosion research. In Fig. 1 one of these loops is shown. Apart from these high-temperature loops, several smaller specific loops, often for lower temperatures, have been used as well.

However, it is an essential part of the research and development programme to have a facility to test the different types of suspensions to be considered under real power reactor circumstances.

Apart from capsule irradiation experiments which have been continuing already for several years, a high-temperature, high-pressure test loop for suspensions is under construction with a part in the circuit of such a geometry that it can reach nuclear criticality [3, 4].

This test facility is called the KEMA Suspension Test Reactor (KSTR)\(^1\), a schematic flow sheet of which is given in Fig. 2. The critical part of the loop should be made as small as possible to reduce the critical mass to a minimum and to reduce the total power production to a minimum for a given power density in the critical part. The maximum power density, and that means also the heat extraction rate, should be equal to the maximum power density of interest in a nuclear power station. This power density has already been discussed in section 2 and is fixed at about 40 kW/l.

To reduce the critical mass further, light water instead of heavy water will be used, as for such a small core vessel (about 30 cm in diameter) the moderating properties of light water are more important than the low neutron absorption in heavy water.

A further measure essential for a small reactor is an efficient reflector. The actual reflector of 15 cm BeO and 35 cm graphite is at the same time a good moderator. This is important since the thermal flux is flattened by this moderation. That means that also the energy production distribution is flattened to the ratio of maximum-to-average thermal flux of 1.2.

As the reactor will be operated at temperatures up to 250°C, an over-pressure of 20 atm abs. of hydrogen is applied to prevent boiling. Since the neutron absorption by the core vessel should be reasonably low, the reflector is also pressurized to keep the wall thickness of the core vessel small. However, under high power conditions there is quite a large heat dissipation in the reflector, consequently a reflector cooling system (Fig. 1, II) is necessary.

The reflector and the core vessel are located in a steel pressure vessel having a wall thickness of 5 cm. The pressure vessel is designed to withstand 60 atm abs. of overpressure and thus will contain all radioactivity in case of leakage or rupture of the core vessel.

The nearly spherical core vessel has conical inlet and outlet sections on the vertical axis. The suspension flows from the bottom to the top. At the inlet of the core, a special device is provided to give the liquid a slight rotation to stabilize the flow in the vessel. In addition the flow velocity along the wall is increased to prevent settling of particles on the wall in the lower part of the core vessel (cf. section 5.4).

\(^1\) Although in principle an in-pile loop in a research reactor could be considered too, the complicated structure of the installation outside the in-pile part makes this last proposition impractical.
FIG. 1. High-pressure high-temperature test loop for UO₂-ThO₂ suspensions
As a consequence of the fissioning process in the core vessel, water is partly decomposed into hydrogen and oxygen. By injecting excess hydrogen at the top outlet of the core vessel the build-up of an explosive mixture can be prevented.

In addition, by injecting gas into the suspension liquid the gaseous fission products (Xe and Kr) can be stripped and separated from the liquid in a gas-liquid separator (Fig. 1: 3). After removing the gases from the liquid stream they are fed to the gas circulating system (Fig. 1, III). The hold-up of $^{133}$Xe in this system reduces the xenon contents and, therefore, the xenon poisoning in the primary circuit by a factor of ten.

The primary cooling system (Fig. 1, IV) which removes the fission heat during power operation, is also provided with heating elements to supply heat to the main circuit when the reactor is operated in a subcritical state.

The main reactor system is provided with a dump vessel (Fig. 1: 6) of sufficient capacity to store the total liquid contents of the main system. The dump vessel is isolated from the main system by normally closed valves, which can be opened in case of an emergency to permit the entire contents of the system to drain into the vessel in less than one minute. The dump vessel can be cooled to remove the decay heat of fission products. To prevent local overheating in this vessel, the suspension will be kept in motion as long as necessary by a specially developed stirrer. The main circuit is also provided with a concentration regulating system (Fig. 1, V).
Finally there is a liquid purification system (Fig. 1, VI) to study the removal of fission and corrosion products from the suspension. This system is based on the principle of recoil separation of fission products from the fuel particles into the surrounding fluid. The efficiency of the recoil process is very high for particles of 5 µm, as has been demonstrated in the zero-energy reactor [5].

Although in this paper a safety discussion is not necessary, it should be mentioned that this reactor will have a double containment required by the fact that about $10^6$ Ci can circulate in the system. In Fig. 3 the inner gas-tight and pressure-tight containment, under construction, can be seen. Parts I and V of the KSTR will be placed in one containment, parts III and VI in a second one and part II in a third one. The whole installation, including also part IV, is placed in the reactor hall, which is leak tight and gas controlled.

3. **FUEL**

3.1. **Introduction**

Aqueous suspensions of fissile and fertile oxide material have distinct advantages over solutions as a nuclear fuel for homogeneous breeder reactors:

(a) The fuel concentration can be considerably higher than in a salt solution. This is of special interest for the fertile component.
(b) By applying the principle of the recoil separation a continuous removal of fission fragments is feasible under certain conditions.

(c) The corrosiveness of the liquid is very low compared with that of the acidic fuel salt solutions.

(d) For fuel material with a size above 1 μm, a fast change of concentration in the reactor is possible by applying hydroclones.

On the other hand a number of specific problems arises, which are discussed in this paper in some detail.

The aim of the fuel test facility at Arnhem is to study the applicability of aqueous suspensions as a fuel for homogeneous reactors and to define clearly their limitations, whether they are of a technical, an economical or a more fundamental nature.

Consequently, the research and development should not be restricted to one specific type of fuel which appears to be most attractive on first sight, but a wide range of fuel types should be taken into account. Because the Th-U ratio is fixed for the KSTR, the different types of fuel are, in the first instance, fuels of different particle size only. It must however be pointed out that the Th-U ratio, which is about 6 for the KSTR, would be about 60 for a power reactor of the same type and for the same fuel concentration. The influence of the variation of this Th-U ratio will therefore be studied separately from the KSTR, as the uranium content might have a bearing on the irradiation stability of the suspension particles.

There are several reasons why the fuel research and development for the KSTR at KEMA started with a UO$_2$-ThO$_2$ mixed crystal fuel of spherical particles with a diameter of 5 μm. Near-critical measurements at low temperature with a UO$_2$ fuel of 10 μm demonstrated that the range of fission fragments in UO$_2$ (and expectedly also for UO$_2$-ThO$_2$) lies between 4.5 and 8 μm, depending on the mass number of the fragment [5]. This implies that from a fuel particle with a diameter of 5 μm virtually all the fission fragments escape by recoil, which in principle makes them accessible for continuous removal. In striving for a high conversion ratio, this removal of fission fragments is very important.

The penetration, however, of fission fragments through the fuel surface causes at the same time considerable damage, which destroys the initial particle at a fast rate. Although a major part of the research in this field is concentrated on this problem to find a remedy from a detailed understanding of the phenomenon, other types of fuel are being studied with increasing interest. As a first estimate two additional fuels were chosen, which represent at least in many respects the extremes for this type of reactor.

The first one is a fuel suspension containing very fine fuel particles with a size of the same order as that of the debris observed in irradiated fuel suspensions (200–500 Å). The system is more or less a colloidal solution of high concentration, which may, however, change under actual reactor conditions. It represents then the final equilibrium stage of disintegration by irradiation and growth by physico-chemical processes. Production and reprocessing of such a fuel could be cheap.

The second fuel consists of a suspension of coarse fuel particles, somewhere around 30 μm. The fission fragment penetration will decrease very
strongly, compared with the 5-μm fuel. It can even be decreased to zero by encapsulating the fissile material as a small sphere inside a concentric layer of the fertile component, which prevents the escape of fission fragments. This means, at the same time, that the recoil effect for the removal of fission fragments has become ineffective.

Together with the 5-μm fuel the choice presented above seems to cover as a first estimate the full range of fuel sizes which might be applicable in an aqueous homogenous suspension reactor.

3.2. Fuel research and development

A large number of problems related to the choice of the three types of fuel material (fine, 5 μm, coarse) has been studied or is still being investigated, some of a quite general nature, others specific for the application of one of the fuels. A short survey of the most interesting aspects is presented here.

(a) Fuel preparation

Until more information is available from our experiments it is supposed that the actual fine fuel requires no new preparation method. Fine fuel for purposes of research and development is produced by applying techniques used in the sol-gel process. This process was developed for the first time at Arnhem, to produce the 5-μm UO₂-ThO₂ spheres.

The coarse fuel can also be produced with the sol-gel process if it consists of mixed crystals. For the coated particles other procedures are still under development. In this last case a diffusion barrier between the UO₂ and the ThO₂ is required to prevent the formation of mixed crystals during sintering.

(b) Colloid chemical stability

The colloid chemical phenomena, especially the degree of colloid chemical stability and the mechanism of flocculation, may have a strong bearing on the reactor behaviour. No method for studying those phenomena at increased temperatures (up to 250°C) was known at the time. A new technique was therefore developed [4], which yields more reliable results even at room temperature than the methods well-established in colloid chemistry. The principle of the method is to measure continuously and in a very sensitive way the changes in concentration of a settling suspension as a function of time at various levels. From these data an apparent size distribution of the powder can be derived. The deviation of this apparent size distribution from the true one can be interpreted quantitatively in terms of degree of colloid chemical stability. The concentration measuring device consists of an X-ray tube and a detector which determines the absorption of the X-rays by the sample tube. Autoclaves can be used as sample tubes, at temperatures up to 250°C. Moreover the same apparatus can be used as an automatic and

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1 See the 'Sol-gel process for thorium fuel production' by Th. van der Plas and H S G. Slooten, this report.
extremely fast measuring device for particle-size distributions far down into the sub-micron range.

(c) Rheology

Closely related to the foregoing is the question of how the suspension behaves as a liquid. Attempts to measure its viscosity at 250°C in a capillary viscometer were still unsuccessful up till now. Technological measurements to determine the viscosity from pressure drops in loops have started. Generally the degree of colloid chemical stability has a strong influence on the viscosity of concentrated systems. This influence depends on the particle or the agglomerate size. From very different aspects of the research (fuel preparation, flocculation, hydrolysis, hydrodynamic experiments) it could be shown that the maximum agglomerate size depends on the shear rate in the liquid, and hardly on the initial size of the particles. This would mean that the different fuels will behave similarly in this respect. With the low viscosity of water at 250°C a small agglomerate size is expected in a flocculated system, which may be in the neighbourhood of 10 μm.

(d) Hydrodynamics

The influence of the rheology on the hydrodynamic behaviour of the suspension and the erosiveness of the very hard UO₂–ThO₂ fuel particles are discussed in section 4.

(e) Removal of fission fragments

The removal of fission fragments by recoil can in principle be applied to the fuel of 5 μm. At the concentration envisaged re-entrance of fragments into neighbouring particles hardly occurs in a dispersed system. The gaseous fission products are largely stripped from the system by a gas stream. This is especially important for the 135Xe. To collect the hydrolysable fission fragments and to prevent them from deposition onto the fuel itself a scavenger is needed [5]. The necessity to separate the fuel and the scavenger in a hydroclone requires a finely divided solid. Carbon, the initial choice, is unattractive for this purpose because of its fast oxidation. Fine ThO₂ seems more promising.

For the fine fuel the recoil principle still applies, but re-entrance of fragments increases and separation between fuel and scavenger is no longer possible. The addition of a scavenger therefore has no effect, but still the major part of the 135Xe can be removed, which certainly is a most attractive feature of the system. However, a more frequent reprocessing of the fuel seems to be required than in the case of non-damaged 5-μm fuel. With coarse fuel the removal of fission fragments in the reactor cannot be considered.

3.3. Caking

The introduction of fines into the reactor might enhance cake formation, which, however, must be avoided. The phenomenon of caking is very com-
plicated and not yet clearly understood. The two stages which are required to form a hard cake are: deposition of solids and intergrowth of the deposited material. The deposition is influenced by the colloid chemical conditions, but seems to depend mostly on the flow characteristics. At low velocities the boundary layer is too thick for the moving liquid to reach the wall and therefore deposition hardly occurs. On the other hand, at very high velocities, and thus high shear rates, a deposited particle can be torn away from the wall when its size is large enough and the sticking chance decreases. In the intermediate region lasting deposition may occur. Thereafter the formation of a hard cake by intergrowth will certainly depend on the presence of reactive or slightly soluble components (e.g. hydroxides or hydrated oxides). Research in this field has started both fundamentally and technologically. Experiments have shown that the fuel particles do not reach the wall of a 5-cm tube below the velocity of 9 m/s at 250°C. Therefore a flow velocity of 5 m/s has been chosen as the design value for the KSTR. One should be well aware of the fact that the final answer with regard to caking can only be gained from the experiments with the KSTR. In loops no true hard cakes have been found as yet.

3.4. Concentration control

The fuel concentration must be controlled in the KSTR in a reproducible way. Above a size of one micron, hydroclones are applicable. For the fines, however, an additional method is required, because the equilibrium size distribution will be unknown until it has been reached in the KSTR. The liquid with colloidal fines in it behaves quite similarly to a solution, and evaporation-dilution in a by-pass system was chosen to control the concentration. However, the system is no true solution, and it is therefore subject to settling. Consequently stirring or continuous circulation is required. Complicated as this seems for the KSTR, it certainly will be no serious drawback for a power reactor.

3.5. Irradiation stability

The irradiation stability of the fuel is, of course, of primary importance. The experiments were limited to the 5-μm fuel for evident reasons [6]. In large outline the results seem discouraging for the 5-μm fuel, because the general conclusion appears to be that the serious damage to the fuel particle surface is provoked by the penetration of the fission fragments. The phenomenon as a whole consists of two mechanisms in sequence, as could be shown by experiments. The primary effect of the damage caused by the penetrating fission fragment is still the main subject of investigation. It might be that this primary damage is only caused by a certain minor fraction of fragments which meet specific conditions. A solution of this problem must therefore not be excluded. In actual fuel application for a suspension reactor the secondary effects which determine the deposition or the dispersion of the fuel fragments are very important. Irradiations under colloid chemically unstable conditions revealed that the debris are redeposited on the fuel particles whereas in a stable system peptization of the fuel frag-
ments occurs. For irradiation at low temperatures the redeposited material can easily be etched off. The general line of approach chosen for this part of the research is to try to reveal the critical mechanism of the damage. Thereafter a solution may be found or this limitation of the applicability of suspension fuel must clearly be defined. Capsule irradiations and, to a greater extent, in-pile loop experiments are important. It must, however, be clearly understood that actual reactor conditions, including its hydrodynamics, cannot be reached and that experiments in the KSTR can provide the only dependable answer to the question of irradiation stability. In accordance with this conviction the KSTR forms an essential part of the research towards the possible development of a power reactor.

No experiments were yet carried out with the other fuels. Capsule experiments with fine fuel will provide no essential information, because the only interesting aspect will be its equilibrium size distribution. This will expectedly depend on the rate of disintegration (the specific power density) and the rate of growth (temperature and flow characteristics of the liquid).

3.6. Fuel research in the test facility.

It has been repeatedly pointed out that not the KSTR in itself but the results which can be obtained with that instrument are the aim of the project. It is impossible to collect the data required for the construction of an actual reactor without the fuel research in the KSTR. A number of important questions must be answered by this research.

(a) How does a reactor behave with a system that is homogeneous, but which can become inhomogeneous by settling of fuel or by gas evolution? Much experience will have to be collected in handling suspensions during operation.

(b) The fuel itself might change through disintegration, particle or agglomerate growth or through caking. Research with the KSTR will have to provide the final answer to the question of the limitations of the fuel itself.

(c) The important question of whether continuous removal of fission fragments is possible, in what form (gas or solid) and to what extent can only be studied in the KSTR, because in this facility true reactor conditions are reached including the flow characteristics that certainly influence the fission products removal.

(d) The smaller the fuel particles are, the larger the water decomposition, which expectedly will not be compensated by gamma recombination or by hydrogen overpressure. The rate of gas formation in the core might limit the maximum specific power density. With the KSTR the water decomposition will be studied as a function of power level, temperature and fuel particle size.

Because of the requirement to study these essential questions, a number of provisions and measuring devices will be included in the KSTR which for the greater part would never be incorporated into a power reactor of this type. Research with the KSTR has to reveal the chances of suspension fuels for nuclear reactors and at the same time will provide technological information for the construction of a power reactor in case a favourable answer is obtained with respect to the fuel behaviour.
4. HYDRODYNAMIC PROBLEMS ASSOCIATED WITH THE TRANSPORT OF FINE SUSPENDED HEAVY PARTICLES IN A CIRCULATING LIQUID

4.1. Introduction

When circulating a suspension in a pipe at a high temperature settling of particles and erosion of the walls may occur. These two aspects have long been considered the main disadvantage associated with the concept of an aqueous suspension fuel. The ability to operate a suspension fuelled reactor would largely depend on the prevention of settling and erosion, as far as technology is concerned. At the beginning of our investigations very little was known theoretically about the fluid dynamics of a mixture of discrete particles in a turbulent stream or in boundary layer motion. An extensive programme of tests was carried out in which suspensions have been circulated for many thousands of hours at high temperatures in high pressure loops. The main objectives of these experiments have been to learn how to design the equipment and choose the velocities so that settling does not occur and erosion is very limited.

4.2. Erosion

At the start of the work it was realized that the main velocity, \( U \), in the pipe must have a large influence on the phenomena. To study this influence a slowly converging tube was used and the erosion of the wall was observed by visual inspection. It was found that no appreciable erosion occurred if the velocity was below 9 m/s. The location where the erosion was first observed did not change, even after prolonged runs. So far no generally accepted theory exists that describes the observed phenomena satisfactorily. Erosion can only be caused by particles which still possess sufficient momentum when they reach the wall. Particles that have been deposited on the wall could be removed from it by virtue of the existing shear stresses in the viscous sub-layer near the wall. It is thus considered essential that the particle is transported across the viscous sub-layer in a projectile fashion, by virtue of some transverse velocity imparted to it. The velocity fluctuations of the liquid are the main source of this mechanism. These fluctuations depend largely upon the main velocity of the liquid (with a given geometry), and to some extent on the concentration. The phenomenon is of course very complicated. The properties of the wall enter into the picture and also other influences than purely hydrodynamic properties will interfere with the transport in the boundary layer. Several authors have found that the velocity of deposition of particles in gases is proportional to \( U^n \) (\( n > 3 \)). Our observations in liquids indicate that there is indeed a very strong dependence on \( U \).

As the terminal velocity of settling particles varies with the square of its diameter this factor should also enter into the equation. An influence of particle size in the small range studied (5–10 \( \mu \)m) was not found. This might be an indication that the trajectory of the particles is not very different from that of the surrounding liquid and that liquid fluctuations penetrate very deep into the viscous sub-layer, carrying the particles with them.
As a result of the experiments mentioned before, the main velocity in our pipes (diam = 4.6 cm) has been chosen to be 5 m/s. Test loops operated for many thousands of hours have shown no appreciable erosion. An inspection of the bends gave the same result. The problem of erosion appears more urgent in the case of the flow of suspensions in pumps where the liquid is circulating in a narrow gap between a stationary diffuser and inner rotating impeller. Here secondary flow patterns and vortices occur which enhance still more the transport of particles to the walls.

To minimize erosion the design of existing pumps had to be adapted very carefully. The result was that the stainless steel impeller of such a pump has lost only a few grams of material after several thousand hours of operation at 250°C and a concentration of about 400 g/l. By the nature of the flow in the annular gap between the impeller and the diffuser it is impossible to prevent erosion completely. Further progress can only be made by choosing materials more resistant to erosion.

4.3. Settling

To prevent gravitational settling it is sufficient to provide a high convective component of the velocity. In that case the gravitational component can be neglected. In flow through pipes this is easily accomplished. A suspension at 250°C appears to be flocculated. The settling velocity of these flocs measured in a settling tube was about 2.5 cm/s. The effective diameter of such flocs in turbulent flow in a pipe may be smaller but is not yet known. For turbulent flow of a Newtonian fluid it has been shown experimentally that the time-smoothed velocities are given roughly by the relation:

\[
\frac{\bar{\nu}}{v_{\text{max}}} = \left(1 - \frac{r}{R}\right)^{1/7}
\]

where \(R\) is the radius of the pipe and \(r\) the distance measured from the centreline, \(v_{\text{max}}\) is 1.25 times the mean velocity in the pipe.

If this relation could be used to estimate the velocity very near to the wall it is found that the velocity at a distance of 23 \(\mu\)m from the wall is 233 cm/s if the pipe radius is 2.3 cm. This is nearly 100 times as large as the settling velocity for a 10-\(\mu\)m particle, and should be sufficient to prevent settling even in a horizontal pipe. This has been confirmed by our experiments. So far no phenomena have been observed that indicate any settling at the chosen velocity of 5 m/s.

4.4. Reactor vessel

The danger of settling is much greater in the reactor core vessel where the suspension enters from below with a velocity of 500 cm/s, and then moves through expanding cross-sections of the nearly spherical shaped vessel (see Fig. 4) until the area-averaged mean velocity decreases to about 10 cm/s.

The danger of settling is, of course, great in the lower part of the reactor vessel. After extensive researches a flow distributing device has been
FIG. 4. Reactor vessel of the KSTR: temperature field at two different flow rates. A is relative residence time of the liquid.

constructed that consists of concentrical cones equipped with vanes. A large portion of the liquid is directed to the outer concentric ring of this inlet distributor to achieve a velocity of 100 cm/s at a few centimetres distance from the wall of the core vessel. To stabilize this flow the incoming liquid is forced into a slight rotation. It could be shown that the ratio of the rotational forces to the inertia forces, applied with this device, was responsible for the flow pattern that was observed. By increasing this ratio the flow pattern changed from a marked recirculation in the central part of the vessel into a pattern of less marked (though persisting) recirculation with a very large degree of instability that provoked a good amount of lateral mixing. Increasing the rotational forces finally resulted in a type of flow that is best characterized as a central jet with lateral recirculation.

Considering now the aspects of the recirculation another aspect enters into the discussion of the flow pattern: the residence time of the liquid. During its passage through the core vessel the liquid is heated. As the buckling of the neutron flux is very small the temperature rise above the inlet temperature should be proportional to the residence time of the liquid.
The observed flow pattern with recirculation should therefore give rise to a temperature profile with a maximum in the lower central part of the core vessel. The solution to this problem was to combine the high velocity near the wall with such a degree of rotation that a sufficient amount of lateral mixing would be provided. This should increase the effective lateral heat transfer from the centre of the vessel in the direction of the wall and thus prevent local overheating of the liquid. In a detailed study [7] residence times of the liquid have been measured between the inlet and several hundreds of points in the core vessel. The result is given in Fig. 4 as mean relative residence times. The residence time of the vessel as a whole, given by its volume divided by the flow, is set equal to 1.0.

It could be demonstrated that the maximum increase of temperature to be expected above the total temperature difference over the core would be about 30%. At a flow rate of 9.2 litres/s with $T_s - T_c = 30^\circ{C}$ and a heat production of 40 kW(th)/l this would be equal to $9^\circ{C}$.

As mentioned before, the flow pattern is quite sensitive to changes in the ratio of rotational to inertia forces. With a fixed geometry of cones and vanes, this profile could change with variable velocity at the inlet pipe of the vessel. The measurement of the residence times was therefore repeated at different flow rates. It was found that the profile was quite insensitive to changes of the inlet velocity in the range studied (2-12 litres/s). This compares well with data derived from the concentration measurements carried out with the zero energy assembly as discussed in the next section.

4.5. Nuclear stability

To investigate the nuclear stability of a circulating suspension system a zero-energy reactor was constructed and operated in a wide range of reactivities ($0.5 < k < 0.998$) [8]. The dimensions of the assembly are comparable with the dimensions of the main system of the test facility, however, no gas injection is used so that no gas separator is needed. The temperature of the suspension can be regulated with an accuracy of $\pm 0.05^\circ{C}$ in the temperature range 20-80°C.

For practical reasons it was necessary to start with a pure $\text{UO}_2$ suspension in light water. The suspension fuel was enriched in $^{235}\text{U}$ (up to 20%) to arrive at the same volume concentration of suspension particles as would be required in a large power reactor.

At the moment the assembly is operated with the suspension to be used later on in the KSTR. This means a suspension of mixed ThO$_2$-$\text{UO}_2$ particles containing 13% $^{235}\text{UO}_2$ and with a particle-size distribution of about 5 $\mu$m.

The colloid chemical properties of the suspensions, which determine whether the particles can flocculate or not, are determined by the pH of the liquid.

The homogeneity of the concentration distribution of fuel particles was checked by measuring the neutron flux distributions (thermal and epithermal) inside the core vessel of the near critical assembly.

Experimental results are given in Fig. 5. The neutron flux distributions in the core appeared to be symmetrical, the peak-to-average factor for the thermal neutron flux ($\phi_{th}$) being 1.20. For a small reactor this is very flat.
The constancy of the overall concentration and its distribution in the reactor vessel was demonstrated by the fact that the average value of the neutron flux as measured in the reflector by BF$_3$ counters was constant within the measuring accuracy of ± 1% for several hours (source multiplication 34.8, flow rate 9.7 m$^3$/h). This indicated that the average concentration and its distribution within the core were constant within 0.1% over a long period. In other words no sticking or uncontrolled release of fuel particles occurred within this limit (0.1% is equal to about 6 g of UO$_2$).

When the flow rate in the reactor system was changed it was found that at very low flow velocities the concentration in the core vessel depended on the flow rate. The upper curve of Fig. 6 gives the result for an unstable suspension. The concentration at the maximum flow rate of 24 m$^3$/h is arbitrarily taken as 100. From this curve it can be seen that the apparent concentration in the core vessel increases up to 12% at a flow rate of 1.75 m$^3$/h. The increase in concentration in the core vessel can be explained by the fact that at a flow rate of 1.75 m$^3$/h the average vertical velocity of the liquid in the cylindrical part of the core vessel (8 mm/s) is comparable with the settling velocity of agglomerates (flocs) of suspension particles. These flocs could be formed in the low velocity region of the circuit, since the suspension was unstable and there was enough time available for a certain degree of flocculation.

With a stable suspension in which no flocs can be formed, the increase in concentration is much lower, namely less than 1% (Fig. 6, lower curve).

In both cases there is a large region of flow rates, in which the concentration and consequently the reactivity is independent of the flow rate.  

3 In the KSTR a normal flow rate of 33 m$^3$/h will be used (~9 litres/s). Consequently the region where the concentration in the core is independent of flow rate is sufficiently large.
The apparent concentration of $UO_2$ in the core vessel as a function of flow rate $Q$. The concentration at $Q = 24 \text{ m}^3/\text{h}$ is arbitrarily taken as 100. The curve for a pH of 6.8 represents a flocculated suspension and the curve at 9.1 a stable one.

The temperature coefficient of reactivity has been measured in the range 20–80°C. However, with an unstable suspension practically no change in multiplication was found in this temperature range when heating up slowly (40°C/h). This may again be explained by the fact that flocs of fuel particles could be formed in the low velocity region of the circuit (core vessel). The settling velocity of these flocs increased with temperature because of the decrease in the viscosity of the water with increasing temperature. The negative temperature coefficient could thus be balanced by the increase of concentration within the reactor vessel (hold-up) caused by the increase in settling velocity.

The negative temperature coefficient was not balanced by the increase in settling velocity of the completely dispersed particles when using a stable suspension. Between 21 and 70°C a temperature coefficient of $-2.3 \times 10^{-4} \text{ Δk/k degC}$ was measured.

When an unstable suspension was heated up rapidly (200°C/h) a negative temperature coefficient was measured between 21 and 40°C, namely, $-1.2 \times 10^{-4} \text{ Δk/k degC}$. Now the flocs had apparently not enough time to make use of the increased settling velocity. It may be concluded that at a sudden increase of temperature the negative temperature coefficient will be completely effective, even when an unstable suspension is used.

5. TECHNOLOGY

The equipment to be used for the transport and handling of suspensions at high temperatures under reactor circumstances has many conventional aspects that need not be mentioned here. The nuclear requirements however give rise to problems that can only be solved after extensive testing under
circumstances that are as near to reactor conditions as possible. To mention some of them: (1) absolute leak tightness is required (less than $10^{-7}$ n/cm$^3$ s of gas) for flanges and valves, (2) absolute liability of all moving parts (impellers in pumps, stirrers in storage vessels, valves) with a minimum of erosion to limit poisoning of the reactor fuel, (3) perfect functioning of devices used to control the concentration (hydroclones, evaporators and separating cyclones) and (4) perfect functioning of the gas injection above the reactor core to dilute the oxygen formed by radiolysis followed by a recombination of the gases (gas-jet-pump, injection nozzle and catalytic recombiner). Many devices already known in industry had to be redeveloped to meet our requirements, since geometric sizes are restricted by criticality considerations.

To limit undue risks of very expensive high-temperature tests an extensive test programme is usually carried out at low temperatures. The next step is then to test single components in smaller high-temperature loops. The final stage consists of a combined test of more than one component in a high-temperature assembly that is almost identical to the reactor system. Here prolonged tests will be carried out for many thousands of hours to establish the performance characteristics of the equipment. So far most of the equipment to be used with the 5-μm fuel is in the last stage of the testing programme. Some components that will be used with the low particle size fuel (1μm) are still in the earlier stages of investigation. Here a programme has been started to investigate the behaviour of the suspension of sub-micron particles at high temperature as these suspensions are expected to show a distinct non-Newtonian behaviour. However, most of the equipment designed for the 5-μm particles can be used for the finer particles as well.

ACKNOWLEDGEMENTS

The Netherlands homogeneous suspension reactor project is executed in the KEMA laboratories in Arnhem in close co-operation with EURATOM and the Reactor Centrum Nederland (RCN). The report presented to the panel is the result of the efforts of the total team working in this project and not only of the authors who have written this report.

REFERENCES

In recent years the interest in sol-gel processes for the production of thorium-bearing fuels has increased considerably. On the one hand this rising interest is a consequence of the recognition of the importance of thorium as a potential nuclear fuel, while on the other, the large amount of work being done on fuel production processes making use of a sol-gel transformation has demonstrated the many possibilities of this type of process. Moreover, the increasing interest in the application of coated particles in reactor fuels has exerted a stimulating influence.

### TABLE I

**FUEL SPECIFICATIONS**

<table>
<thead>
<tr>
<th></th>
<th>Power reactor</th>
<th>KSTR</th>
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</thead>
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<tr>
<td>Composition</td>
<td>ThO$_2$-1.5% UO$_2$</td>
<td>ThO$_2$-15% UO$_2$</td>
</tr>
<tr>
<td>Size (μm)</td>
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<td>&gt; 98</td>
</tr>
<tr>
<td>Surface area (m$^2$/g)</td>
<td>&lt; 0.2</td>
<td>&lt; 0.2</td>
</tr>
</tbody>
</table>

At KEMA Nuclear Laboratories work on sol-gel started in 1959\(^1\) as part of the Homogeneous Suspension Reactor Development Programme [1]. The envisaged specifications for the fuel of a single-zone power reactor of the aqueous suspension type are given in Table I. In the same table the requirements for one of the fuels to be used in KSTR [2] are listed. The chemical composition in both cases follows from the optimum 235U or 233U content dictated by nuclear considerations when the volume concentration of the suspension is fixed at approximately 4% for hydraulic reasons. The upper limit of the particle size is determined by the fact that the diameter of the fuel particles should be smaller than the range of most of the fission fragments if the recoil effect is to be effective [3]. The lower limit follows from the condition that it must be possible to use hydroclones to change the concentration in the reactor. The spherical shape and smooth surface of the particles are dictated by the necessity to minimize the erosive properties of the circulating suspension.

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\(^1\) French Patent 1274653, priority date of filing, 16 Sep 1959.
In particular the last condition, namely the spherical shape of the particles, led to the choice of a sol-gel process for their preparation. Experience in the catalyst-industry had clearly shown that production of spherical particles can be achieved in this way. Furthermore the pseudo-morphic characteristics exhibited by most thorium compounds made it probable that if the first solid product in the production process had a spherical shape this shape would be retained throughout the rest of the process. These suppositions were borne out when the preparation process was developed.

The process in use for the production of particles in the 5-μm range is divided into five steps and can be outlined as follows: The first step of the process involves the preparation of a sol of hydrated ThO₂ by peptizing a thoroughly de-ionized Th(OH)₄ precipitate in Th(NO₃)₄, UO₂(NO₃)₂ or HNO₃ solution. Subsequently this sol is dispersed in an organic medium to form an emulsion. The choice of the dispersion method depends on the drop-size distribution and thus on the ultimate particle-size distribution that is required in the end product. In the case of 5-μm particles the use of a vibrational stirrer gave the best results. The resulting size distributions were very narrow. The sol drops of the emulsion are solidified by gelation brought about by the addition of ammonia. After dehydration by boiling in the organic medium that is also used in the emulsification and gelation steps the particles are washed, dried and fired to obtain a sufficiently low surface area.

Development work and experience accumulated by other workers, notably at Oak Ridge National Laboratory (ORNL) in the United States of America have shown that processes of the type described above are extremely flexible.

In the present process the composition of the oxide products is varied from pure ThO₂ to ThO₂ with 22 at. % UO₂ when the appropriate amount of UO₂(NO₃)₂ instead of Th(NO₃)₄ is used in the preparation of the sol. Furthermore the process is not limited to oxide particles since by the incorporation of a suitable amount of carbon in the sol a dried product can be obtained that is easily convertible to carbide. It is very probable that the incorporation of other elements like Zr, Al, etc. will not give rise to large difficulties.

With regard to the particle size of the product it has been shown that by adaption of the dispersion method and the gelation procedure, powders with a mean size from 2 μm to 2 mm can be prepared.

The flexibility of sol-gel type processes gives the possibility of producing a rather wide range of fuel types by this method. Thus the application of sol-gel to the production of the following fuels can be envisaged:

1. Fuel for a homogeneous suspension reactor.
2. Coated particles for dispersion in graphite or BeO for high-temperature gas-cooled reactor fuel.
3. Particles for the production of fuel dispersions in metals.
4. Particles for the production of loosely packed or vibro-compacted fuel elements.

Apart from this wide range of possible applications sol-gel processes are adaptable to remote handling operations which is an essential feature in connection with the handling of reprocessed thorium fuel.
It should not be concluded however from this rough description that the same process can be used for all products desired. Important changes in details may be necessary. A case in point is the production of large spheres (> 100 \mu m). It was found that the method in which ammonia is supplied from the outside to the sol droplet is no longer applicable here. More and more failures are met with, when the diameter of the droplets increases. This can be easily explained qualitatively.

It is observed that a membrane of gelled material forms around the drop and this membrane acts not only as a barrier to a further penetration of ammonia, but also gives rise to osmotic phenomena that can lead either to rupture or to wrinkling of the membrane.

The production of larger spheres was only possible by the incorporation of an ammonia donor in the sol, which may eventually give off ammonia slowly at room temperature but more rapidly at a higher temperature. In this way an increase of temperature can be used to trigger the gelation that now proceeds at equal rate throughout the whole drop. Satisfactory oxide and oxide plus carbon particles have been prepared in this way.

Together with this variation in the chemistry of the process a change had to be made in the technology. It proved to be extremely difficult to prepare sol dispersions of drops of equal size in the 200–2000 \mu m range by means of stirring. On a laboratory scale, a solution is found in the simple formation of drops from a capillary. Although even moderate flow-rates through the capillary lead to surprisingly high production rates, it would seem that the development of a more rapid method is desirable if production in the tonne range is envisaged.

Another difficulty has been found in the production of oxide spheres. The nitrates of thorium and uranium are the raw materials now readily available. If this nitrate remains behind in the gelled material the particles are invariably shattered because of a sudden gas production from the nitrate when the particles are dried at temperatures around 120°C.

A washing step is necessary to remove the nitrate from the freshly formed gel. It was found that on washing, phenomena occur that are similar to those outlined above for the external gelation. These difficulties can be overcome by a careful adjustment of the composition of the washing liquid.

It can be concluded from experience that the sol–gel process is adaptable, with only a small development effort, to changes in the requirements for the product. This flexibility is also inherent in the powder metallurgical route, at present its most important competitor in the case of production of coated particles. However, it would seem that the sol–gel process is more easily adapted to conditions of remote handling and a decision in favour of one of the competing routes will depend to a large extent on the particular fuel cycle chosen.

Data on the cost of the sol–gel process when used on a large scale with non-radioactive raw materials can perhaps be derived from the manufacture of catalysts for the oil industry. They will require considerable modification when radioactive raw materials are to be used. This subject was discussed at the EURATOM Symposium on Fuel Cycles of High-Temperature
Gas-Cooled Reactors. The information pertained particularly to the case of carbon-coated oxide or carbide particles; it is not repeated here but it is perhaps valid to conclude that the sol-gel process always has an advantage, small in the case of non-active materials and increasing with increasing radioactivity of the raw material.

Little recycle of material should be necessary when the sol-gel process is carried out well. Unfired gel material can easily be redispersed and recycled.

Methods for the reprocessing of thorium-uranium mixtures by the solvent extraction route are well developed, thanks to the ORNL work on the Thorex process, as well as the Italian work. The main difficulty arises in the head end processes for fuel coated with pyrolytic carbon or dispersed in a graphite matrix. Several institutes are at present engaged on this problem.

Other methods have hardly been considered for thorium fuel. The fluoride volatility method would seem to fail here because of the non-volatility of ThF₄. The potentialities of other methods have as yet to be assessed.

It is seen that significant advances have been made in the production of fuel according to the sol-gel process. The behaviour of these materials under irradiation, promising according to the first experiments, has to be studied more extensively. This work also is carried out actively in several places. Little work seems to be done at present on the fundamental properties of sols and gels. The results obtained at ORNL in this field are not yet fully known.

From our own experience greater effort in this field seems desirable. At the moment every sol prepared seems to be a little different from the preceding one. While this presents no major difficulty, it is troublesome and prevents attainment of the highest efficiency. Part of these difficulties might be overcome by rigorous standardization of procedures of course, but it seems desirable to know exactly what variables are to be controlled.

The first results of a modest effort in this field at KEMA were presented at the EURATOM Symposium. It is hoped that this work can be continued and be of some help to the rapidly advancing technology of the sol-gel process.

REFERENCES


2 Held in Brussels, 10-11 June 1965.
INTRODUCTION

Sweden has great resources of uranium but no thorium. Therefore the Swedish reactor development has been concentrated on uranium-fuelled reactors.

However, some preliminary work on thorium has been done. Hellstrand's measurements of the resonance integral of U and UO₂ are well known. The same methods have been used to measure the resonance integrals of thorium and ThO₂. The results are published in Refs. [1] and [2].

\[ \text{RI}_{\text{Th}} = \left( 3.3 + 16.1 \sqrt{\frac{S}{M}} \right) \pm 5\% \quad 0.14 < \frac{S}{M} < 0.87 \text{ cm}^2/\text{g} \]

\[ \text{RI}_{\text{ThO}_2} = \left( 5.0 + 15.6 \sqrt{\frac{S}{M}} \right) \pm 5\% \quad 0.15 < \frac{S}{M} < 0.65 \text{ cm}^2/\text{g} \]

POWER REACTORS

Ågesta and Marviken

The Swedish reactor programme has been concentrated on pressure vessel reactors moderated and cooled by D₂O. The first power reactor Ågesta with a thermal power of 65 MW started to deliver power in 1964. Ågesta is fuelled with natural UO₂ and cooled by pressurized D₂O. The fuel element consists of clusters of 19 fuel rods.

After Ågesta, reactors have been developed along two different lines. One type is represented by Marviken which is a boiling water reactor with the possibility of nuclear superheating. The fuel elements consist of clusters of 30 slightly enriched UO₂ rods. Refuelling and shuffling of fuel is made on power with the use of an internal refuelling machine. Marviken is now under construction.

PHWR

The other development line is represented by the PHWR (Pressurized Heavy-Water Reactor). In the work on this reactor there has been an aim to make the reactor as simple as possible. Reloading is made at shutdown. The core is different from most other heavy-water-moderated reactors, since the fuel is arranged uniformly over the core instead of being concentrated in clusters. The uniform core gives very good heat transfer and control.
characteristics. In practical cases, the reactivity of a uniform core is slightly less than for a corresponding clustered core. The decrease in burn-up is negligible for enriched cores.

PHWR is favourable for the Th-\(^{233}\)U fuel cycle for the following reasons:

1. Low parasitic absorption from the absence of pressure or flow tubes.
2. The excellent heat transfer properties of ThO\(_2\) can be fully utilized since burn-out is not a limiting factor.
3. There is no incentive to cluster the fuel since fast fissions are negligible and resonance absorption is of minor importance.
4. The disadvantage of discontinuous refuelling is small for a core where the reactivity varies slowly, and less control poisons are needed.
5. The flexible core design allows almost any moderator-to-fuel volume ratio and fuel rod diameter to be used.

CO-OPERATION WITH INDIA

In the spring of 1964 negotiations were started with the Indian Atomic Energy Establishment (AEET) regarding the possible construction of a PHWR in India. Because of the large Indian resources of thorium, AEET was especially interested in the potential of the Th-\(^{233}\)U fuel cycle in PHWR.

Therefore a joint Indian-Swedish research programme was started to study this fuel cycle in PHWR. This programme includes development of calculational methods, measurements to check the calculations and finally calculations of different possibilities to use thorium in PHWR.

**Calculational methods**

The first calculations\(^{[3]}\) were made with the Swedish BURNUP programme. This programme is based on the four factor formula with a number of improvements and has been used for design calculations on Ågesta, Marviken and PHWR. This has functioned well for the above-mentioned reactors which are well moderated and use natural or slightly enriched uranium, and where it exists a great number of measurements on similar lattices. It was regarded doubtful if this calculational scheme could be applied for calculations on the Th-\(^{233}\)U fuel cycle. The "Westcott" thermal cross-sections which are used in BURNUP can hardly be used, as the r-value tends to be very high for interesting thorium lattices.

We have also investigated the possibility of using the English programme Methuselah. Some comparisons were made with the Argonne National Laboratory (USA) measurements\(^{[4]}\). This programme uses four energy groups for pressure vessel reactors. The group division which originates from the USA light-water programme appeared suitable to us, but the programme is slow to run on the Swedish IBM-7090 installation. Therefore it was decided to make a special programme. This programme, CAROL (calculation of rod lattices), is presently being tested. The programme uses the same group division as Methuselah, and it uses partly the same cross-sections, but some improvements have been made. FLURIG, a Swedish collision theory programme, is used for the flux calculations, a more accurate routine is
used for the calculation of the Dancoff correction, and a more efficient routine is used for the burn-up calculation. The calculation time for one burn-up step on an IBM-7044 is about 1 s.

For the macroscopic burn-up and flux distribution we intend to use a programme called DIDIDEE. This programme uses two-group, two-dimensional diffusion theory to calculate the reactivity and flux distribution as a function of time. The two-group parameters as a function of burn-up will be furnished by CAROL. DIDIDEE can also automatically treat fuel shuffling. The programme is working on a Ferranti Mercury computer. A revised version is being programmed in Fortran IV for IBM-7044.

**Measurements**

According to the Indian-Swedish collaboration agreement, the Indian AEET is supplying 550 kg of ThO₂ fuel rods for measurements in the Studsvik zero-energy facilities. The thorium rods will be used together with rods of natural uranium and of 1.2 and 1.8% enriched uranium.

By the end of 1965 we shall also have a few Pu-enriched rods available. We intend to make measurements of bucklings, conversion ratios etc. at room temperature in the R0 critical facility and in the ZEBRA exponential assembly, and at temperatures up to 220°C in the pressurized exponential assembly TZ [5].

A study has been made of how to make useful measurements by mixing the UO₂ and the ThO₂ rods.

**REFERENCES**

ВОСПРОИЗВОДСТВО ГОРЮЧЕГО В ПРОМЕЖУТОЧНОМ ЭНЕРГЕТИЧЕСКОМ УРАН-ТОРИЕВОМ РЕАКТОРЕ (УТР) С ВОДНЫМ ЗАМЕДЛИТЕЛЕМ

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ГОСУДАРСТВЕННЫЙ КОМИТЕТ ПО ИСПОЛЬЗОВАНИЮ АТОМНОЙ ЭНЕРГИИ СССР
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1. ВВЕДЕНИЕ

Значение воспроизводства ядерного горючего для развития энергетики будущего неоднократно подчеркивалось различными учеными как в СССР, так и за рубежом (см. например [1, 2]). Создание реакторов-бридеров, которые производят больше ядерного горючего, чем потребляют, позволяет реализовать практически неисчерпаемые энергетические ресурсы — большие количества урана-238 и тория-232, содержащиеся в горных породах земной коры. Требования, предъявляемые к конструкции таких систем, в основном определяются экономикой реакторов-бридеров, которые должны производить электроэнергию и ядерное горючее для собственных нужд, в связи с чем необходимы достаточно высокие коэффициенты воспроизводства и хорошие теплотехнические характеристики.

Воспроизводство в $^{233}$U-Th промежуточных реакторах с обычной водой в качестве замедлителя впервые рассмотрено в обзорном докладе И.В. Курчатова "Некоторые вопросы развития атомной энергетики в СССР", сделанном в Харуэлле (Англия) в 1956 году. Доклад содержит краткое описание реакторов и указания на ряд преимуществ таких уран-ториевых реакторов, изучавшихся с 1953 года в ИАЭ под руководством С.М. Фейнберга. Однако на первой стадии исследования, ввиду отсутствия достаточно точных методов расчета и необходимых ядерно-физических констант, были получены лишь предварительные оценки некоторых параметров уран-ториевых реакторов-бридеров.

Данная работа является продолжением и развитием этих первоначальных исследований. Она охватывает гораздо больший круг вопросов, связанных с воспроизводством в уран-ториевых реакторах-бридерах (УТР), и использует более точные и надежные расчетные методы. Большое внимание уделяется выбору ядерно-физических констант.

В работе рассматривается двухцелевой реактор-бридер (УТР), который имеет форму цилиндра с центральной активной зоной, содержащей $^{233}$U, $^{232}$Th, $^{235}$U и конструкционные материалы: Ni, Fe и Cr, и кольцеобразную зону воспроизводства, в состав которой входит $^{232}$Th, $^{233}$U, Fe, Ni и Cr. Были рассмотрены варианты с различными концентрациями воды и конструкционных материалов в активной зоне и зоне воспроизводства. При этом отношение $\rho_K / \rho_{233}$ ($\rho_K$ — суммарная ядерная концентрация Fe, Cr и Ni; $\rho_{233}$ — ядерная концентрация $^{233}$U) изменялось в пределах: от 25 до 6. Отношение
\[ \frac{\rho_n}{\rho_{233}} (\rho_n - \text{ядерная концентрация водорода}) \text{ менялось от 5,0 до 30,0. В зоне воспроизводства } \frac{\rho_n}{\rho_{Th}} (\rho_{Th} - \text{ядерная концентрация } Th^{232}) \text{ изменялось от 1,3 до 0,3, а } \frac{\rho_n}{\rho_{Th}} \text{ изменилось от 0,54 до 0,92.}

Ввиду малого количества избыточных нейтронов, которые могут использоваться для воспроизводства \( (\nu_{233}^{Th} - 1 = 1,3^*) \), нужно ожидать, что в энергетическом U^{233} – Th^{232} реакторе, содержащем значительные количества конструкционных материалов, трудно будет добиться заметного превышения коэффициента воспроизводства над 1.

Поэтому необходимо было выполнить большое количество достаточно надежных расчетов с целью выявления систем с максимальными коэффициентами воспроизводства. Исследовалась зависимость коэффициента воспроизводства от ряда параметров: жесткости спектра, количества вредных поглотителей, размеров реактора и, что особенно важно, от используемых в расчетах ядерно-физических констант.

2. МЕТОДИКА РАСЧЕТА

Для определения коэффициента воспроизводства и других важных нейтронно-физических характеристик уран-ториевых систем использовалась 12 групповая программа расчета гомогенного реактора с водородосодержащим замедлителем, составленная на электронную машину [3]. В качестве уравнений реактора использовалось \( P_0 \) – приближение кинетического уравнения и однососточное уравнение диффузии тепловых нейтронов. Предполагалось, что замедление нейтронов на всех компонентах реактора, кроме водорода, осуществляется по возрастной схеме. Хотя программа являлась одномерной, она позволяла рассчитывать реактор, имеющий форму конечного цилиндра, благодаря введению в групповые константы поправок на утечу нейтронов через торцы цилиндра.

Коэффициент воспроизводства (KB) вычислялся по следующей формуле:

\[
KB = \frac{I_{Th}^{\text{c}}}{I_{c}^{\text{c}}} + \nu I_{f}^{\text{c}} \cdot \frac{\kappa_{\text{эф}} - 1}{\kappa_{\text{эф}}} \tag{1}
\]

Здесь \( \kappa_{\text{эф}} \) – эффективный коэффициент размножения системы, \( I_{c}^{1} \) – полное число нейтронов всех энергий, поглощаемых i-ой компонентой данной зоны во всем объеме зоны V за 1 сек, равное:

\[
I_{c}^{1} = \int d\vec{r} \int_{E_{c}}^{E_{c}} \Sigma_{c}^{1}(E) \Phi(E, \vec{r}) dE \tag{2}
\]

\( \nu \) – эффективное число вторичных нейтронов, приходящихся на 1 акт деления U^{233}, \( I_{f} \) – полное число делений U^{233} в объеме активной зоны за 1 сек, равное:

\[
I_{f} = \int d\vec{r} \int_{E_{a3}}^{E_{c}} \Sigma_{f}(E) \Phi(E, \vec{r}) dE \tag{3}
\]

\( \Sigma_{c}^{1}(E) \) и \( \Sigma_{f}(E) \) – макроскопические сечения поглощения и деления, соответственно, \( \Phi(E, \vec{r}) \) – поток нейтронов в данной точке \( \vec{r} \), отнесенный к единичному интервалу энергии.

* \( \nu_{233}^{Th} \) – эффективное число вторичных нейтронов, приходящихся на один поглощенный в U^{233} нейтрон.
Большой интерес представляло также определение коэффициентов использования нейтронов в активной зоне и зоне воспроизводства (отражатель):

$$\theta_{a3} = \frac{I_{233}^{233}}{I_{a3}} \quad \text{и} \quad \theta_{отр} = \frac{I_{Th}^{233}}{I_{отр}}$$ (4)

(В первом случае суммирование производится по всем компонентам активной зоны, во втором случае – по всем компонентам зоны воспроизводства).

Уже указывалось, что величина коэффициента воспроизводства очевидно будет близка к 1. Поэтому даже не слишком большие ошибки при расчете КВ могут привести к неправильным выводам о возможности осуществления расширенного или простого воспроизводства в данной системе.

Методика расчета критических размеров и пространственных распределений плотности делений проверялась сравнением с результатами экспериментов, выполненных на водяных промежуточных критсборках [3]. Сравнение показало, что расчет удовлетворительно согласуется с экспериментом.

Кроме того, использование других методик (например, многогрупповой программы Марчука [4]) не дало заметных разногласий с полученными результатами.

Некоторым подтверждением надежности выполненных расчетов являлось также сравнение с опубликованными данными, касающимися воспроизводства в U^{233} – Th системах [5, 6, 7].

Однако необходимо отметить, что в упомянутых работах исследуются системы, довольно сильно отличающиеся от изученного в настоящей работе реактора. В большинстве из описанных реакторов в активной зоне содержится торий (что приводит к гораздо менее выгодным условиям с точки зрения вредных потерь в U^{233}), в то время как в рассматриваемом реакторе УТР торий содержится только в зоне воспроизводства. В ряде работ не учитываются вредные поглощения в конструкционных материалах, утечки из реактора и т.п. Реакторы-бредеры на U^{233} с обычной водой, рассмотренные в работах [2, 5 и 6] являются тепловыми либо довольно близкими к тепловым, в то время как реактор УТР имеет довольно жесткий спектр. Наиболее близким к рассмотренной в настоящей работе системе является реактор АЕТР[7]. Однако и здесь имеется ряд различий: в замедлителе (АЕТР – на графите, УТР – на H_2O) в размерах реактора, спектрах нейтронов, качестве конструкционных материалов и т.д.

3. КРАТКИЙ ОБЗОР ЯДЕРНО-ФИЗИЧЕСКИХ КОНСТАНТ

Очень важную роль при расчетах коэффициента воспроизводства играет выбор ядерно-физических констант [8, 9 и 10]. Поскольку величина коэффициента воспроизводства в значительной степени определяется характером зависимости $\nu_{эф}^{233}$ от энергии, важно иметь достаточно достоверные сведения об этом параметре во всей области энергий. Для "базовых" физических расчетов использовались величины $\nu_{эф}^{233}$ (E), полученные в 1951–1955 гг. из экспериментальных данных Спивака П.Е. и т.д. [11]. Согласно этим данным, $\nu_{эф}^{233}$ равно 2,27 и практически не зависит от энергии вплоть до не-
ВОСПРОИЗВОДСТВО ГОРЮЧЕГО В УТР

Однако после 1955 г. появились экспериментальные и теоретические работы, которые указывают на существование ряда "провалов" $\nu_{233}^{233}$ (E) на резонансах 1,3 эв; 1,7 эв; 2,32 эв; 4,7 эв; 6,79 эв и др. (самое большое падение $\nu_{233}^{233}$ — до величины 1,2 — наблюдается на резонансе 2,32 эв). Поэтому были поставлены под сомнение все прежние оценки коэффициента воспроизводства. Чтобы устранить эти сомнения был выполнен анализ большого количества существующих в настоящее время данных по $\nu_{233}^{233}$ (E) и выяснено влияние возможных изменений этого параметра на коэффициент воспроизводства.

Наиболее ясна область 0,025—8 эв, где имеется большое число точных и согласующихся между собой данных, например измерения Фаллуда и др. [12], Смита и Мейгелби [13], Ж. де Сосора [14], теоретическая работа Эриха Фогта [15] и др.

Гораздо хуже обстоит дело в области промежуточных энергий — от 8 эв до 20 кэв, где ощущается острый недостаток в надежных экспериментальных данных (опубликованные величины $\nu_{233}$ часто противоречат друг другу).

В области высоких энергий, где имеется ряд достаточно точных данных, положение снова несколько улучшается. Вполне надежные современные измерения Дайвена [16] дают значения $\nu_{233}$ в интервале 20 кэв — 100 кэв. Выше 1 Мэв получена зависимость $\nu_{233}^{233}$ (E) [18] ($\alpha$ в этой области мала и $\nu_{233}^{233} \approx \nu_{233}$).

Рис. 1. Сравнение "старых" значений $\nu_{233}$ (E) с новыми

На рис. 1 сравниваются "старые" значения $\nu_{233}^{233}$ (E) [11] с новыми данными [12] — [16]. Тонкая структура зависимости сечений от энергии, полученная в упомянутых работах, была усреднена в области резонансов по ин-
тервалу энергии ~ 1-2 эв и по спектру нейтронов, следующему закону Ферми, и при \( \Phi(E) = \text{const.} \) Сравнение показывает, что в некоторых областях энергий (тепловой и 0,5 - 10 Мэв) \( \nu_{\text{ф}} \) новое больше \( \nu_{\text{ф старого}} \), в области же 1-60 эв, благодаря "провалам" \( \nu_{\text{ф}} \), старые значения превышают новые.

На рис.1 видно, что хотя в действительности \( \nu_{\text{ф}} \) испытывает ряд сильных "провалов" на резонансах, однако эти "провалы" очень узки и усреднение по конечным интервалам энергий приводит к сравнительно небольшому падению \( \nu_{\text{ф}} \) (до величины 2,0 - 2,2).

Ниже будет показано, что для спектра УТР замена старых значений \( \nu_{\text{ф233}}(E) \) на новые не приводит к изменению коэффициента воспроизводства.

Остановимся кратко на новых данных, касающихся других ядерно-физических констант, используемых при расчетах коэффициента воспроизводства.

В настоящее время опубликован ряд работ, посвященных измерениям и теоретическим оценкам сечений захвата нейтронов в конструкционных материалах (Ni, Fe и Cr) [17-20], продуктах деления [21] в Th\textsuperscript{232} [20, 22], а также в Pa\textsuperscript{233} [23, 24]. По сравнению с этими новыми данными сечения, использованные в настоящей работе, являются более "пессимистическими" (так например, завышены сечения поглощения нейтронов протактинием-233, Fe, Cr, Ni и др.). Это позволяет рассчитывать на некоторый "запас" в величине коэффициента воспроизводства, полученной в настоящей работе.

4. КОЭФФИЦИЕНТ ВОСПРОИЗВОДСТВА В U\textsuperscript{233} - Th СИСТЕМАХ И ЕГО ЗАВИСИМОСТЬ ОТ РАЗЛИЧНЫХ ПАРАМЕТРОВ: КОЛИЧЕСТВА ВРЕДНЫХ ПОГЛОТИТЕЛЕЙ, ЖЕСТКОСТИ СПЕКТРА И ДР.

Для того, чтобы исследовать возможные пути увеличения коэффициента воспроизводства в U\textsuperscript{233} - Th водяном реакторе запишем выражение для коэффициента воспроизводства в виде:

\[
KB = \theta_{\text{отр}} \left( \bar{\nu}_{\text{ф233}} - 1 - \frac{1 - \theta_{a\lambda}}{\theta_{a\lambda}} - B_{r 233} \right)
\]

Здесь \( \theta_{\text{отр}} \) и \( \theta_{a\lambda} \) - коэффициенты использования нейтронов в Th\textsuperscript{232} и U\textsuperscript{233}, соответственно; \( \bar{\nu}_{\text{ф233}} \) - эффективное число вторичных нейтронов, приходящихся на 1 поглощенный в U\textsuperscript{233} нейтрон, усредненное по спектру нейтронов во всем объеме активной зоны; \( B_r \) - доля нейтронов (от общего количества нейтронов, рождающихся во всем объеме активной зоны), вылетающих за пределы реактора.

Таким образом, задача определения максимальных величин коэффициентов воспроизводства сводится к отысканию способов увеличения \( \bar{\nu}_{\text{ф233}} \) и коэффициентов использования нейтронов в активной зоне и зоне воспроизводства, а также способов уменьшения утечки из реактора.

На рис.2 дана кривая зависимости коэффициента воспроизводства от толщины ториевого отражателя \( \mathcal{L} \). Кривая показывает, что при толщине ториевого отражателя 40 - 50 см наступает насыщение и, следовательно, дальнейшее увеличение толщины зоны воспроизводства не имеет смысла. Это подтверждается расчетом утечки нейтронов через боковую поверхность реактора: при толщине зоны воспроизводства \( \mathcal{L} = 50 \) см доля нейтронов,
Рис. 2 Зависимость KB от толщины зоны воспроизводства а

вылетающих за пределы реактора через его боковую поверхность (от общего числа рождающихся в активной зоне нейтронов), составляет всего 0,01%.
Если высота активной зоны достаточно велика (100 см и более), то утечка через торцы реактора также будет небольшой.
Таким образом, увеличивая толщину зоны воспроизводства ~ до 50 см и высоту активной зоны ~ до 100 см можно свести утечку из реактора к очень малой величине, так что она не будет практически влиять на коэффициент воспроизводства.

Увеличения коэффициентов использования нейтронов в активной зоне и зоне воспроизводства можно достичь путем изменения спектров нейтронов и количеств вредных поглотителей в реакторе.

Сдвиг спектра в активной зоне и зоне воспроизводства можно осуществить с помощью изменения количества замедлителя (H₂O).
Было рассмотрено изменение отношения концентрации водорода к концентрации U²³³ - ρₙ/ρ₂³³ в пределах: от ρₙ/ρ₂³³ = 5,0 до ρₙ/ρ₂³³ = 30,0.
В таблице 1 показано, каким образом изменяются относительные эффективные сечения поглощения (отнесенные к сечению поглощения для U²³³) различных компонент активной зоны - a²³³φ/a²³³φ - при уменьшении отношения ρₙ/ρ₂³³.

Эффективное сечение i-го элемента определялось усреднением по спектру нейтронов в различных точках данной зоны:

\[
\sigma = \frac{\int_v d\vec{r} \int_{\vec{E}} \sigma_i(E,\vec{E}) \Phi(E,\vec{E}) d\vec{E}}{\int_v d\vec{r} \int_{\vec{E}} \Phi(E,\vec{E}) d\vec{E}}
\]  

В 5-й строке таблицы 1 приведены для сравнения отношения тепловых сечений, соответствующих скорости нейтронов 2200 м/сек. В столбцах
ОТНОСИТЕЛЬНЫЕ ЭФФЕКТИВНЫЕ СЕЧЕНИЯ ПОГЛОЩЕНИЯ РАЗЛИЧНЫХ КОМПОНЕНТОВ АКТИВНОЙ ЗОНЫ

<table>
<thead>
<tr>
<th>( \rho_n/\rho_{233} )</th>
<th>( \sigma_{\text{Fe}}^{\text{pF}}/\sigma_{\text{Fe}}^{\text{pF}} )</th>
<th>( \sigma_{\text{Ni}}^{\text{pF}}/\sigma_{\text{Ni}}^{\text{pF}} )</th>
<th>( \sigma_{\text{Cr}}^{\text{pF}}/\sigma_{\text{Cr}}^{\text{pF}} )</th>
<th>( \sigma_{\text{H}}^{\text{pF}}/\sigma_{\text{H}}^{\text{pF}} )</th>
<th>( \sigma_{\text{O}}^{\text{pF}}/\sigma_{\text{O}}^{\text{pF}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>0.00178</td>
<td>0.00337</td>
<td>0.00195</td>
<td>0.000146</td>
<td>0.00115</td>
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<tr>
<td>10.2</td>
<td>0.00181</td>
<td>0.00337</td>
<td>0.00202</td>
<td>0.000178</td>
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<tr>
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<td>0.00381</td>
<td>0.00238</td>
<td>0.000217</td>
<td>0.000443</td>
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<tr>
<td>30</td>
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<td>0.00430</td>
<td>0.00271</td>
<td>0.000232</td>
<td>0.000319</td>
</tr>
</tbody>
</table>

Тепловые нейтроны \((v = 2200 \text{ м/сек})\)

0.00436 | 0.00793 | 0.0050 | 0.00057 | 0

Таблица 2

СООТНОШЕНИЕ МЕЖДУ ТЕПЛОВЫМИ СЕЧЕНИЯМИ И СЕЧЕНИЯМИ В НАДТЕПЛОВОЙ ОБЛАСТИ

<table>
<thead>
<tr>
<th>( \sigma_{\text{a}}^{\text{pF}} ) (2200 м/сек)</th>
<th>Th</th>
<th>Ni</th>
<th>Fe</th>
<th>Cr</th>
<th>H</th>
<th>O</th>
</tr>
</thead>
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<td>580</td>
<td>7,56</td>
<td>4,6</td>
<td>2,53</td>
<td>2,9</td>
<td>0,33</td>
<td>0</td>
</tr>
<tr>
<td>1200</td>
<td>85,0</td>
<td>4,0</td>
<td>2,3</td>
<td>1,9</td>
<td>0,16</td>
<td>0,052</td>
</tr>
</tbody>
</table>

2, 3, 4, 5 и 6 приводятся отношения эффективных сечений для Fe, Ni, Cr, водорода и кислорода, соответственно. Качественный характер соотношения между тепловыми сечениями и сечениями в надтепловой области показан в таблице 2, где приведены тепловые сечения (при скорости нейтронов 2200 м/сек) и резонансные интегралы (от \( E_{\text{Cd}} \)) для различных компонент реактора.

Ясно, что у водорода сечение поглощения падает с энергией гораздо быстрее, чем у \( U^{233} \), поэтому отношение эффективных сечений поглощения водорода и \( U^{233} \) \( (\sigma_{\text{H}}^{\text{pF}}/\sigma_{\text{H}}^{\text{pF}}) \) уменьшается на 37% при изменении \( \rho_n/\rho_{233} \) от 30 до 5 (см. таблицу 1). Для хroma этот эффект составляет 28%, для железа — 23% и для никеля — 22%. Что касается кислорода, то здесь картина обратная: с ростом жесткости спектра отношительное эффективное сечение поглощения на кислороде \( (\sigma_{\text{H}}^{\text{pF}}/\sigma_{\text{H}}^{\text{pF}}) \) увеличивается в 3,63 раза. Это связано с сильным ростом сечения поглощения нейтронов в кислороде при высоких энергиях (реакция \( n,\alpha \) в области 3,6 - 5,3 Мэв).

Аналогичная картина изменения отношений эффективных сечений наблюдается при увеличении жесткости спектра в зоне воспроизводства.

Заметим, что при сокращении количества воды в реакторе коэффициенты использования нейтронов \( \theta_{\text{взв}} \) и \( \theta_{\text{тр}} \) увеличиваются под действием двух факторов: уменьшения отношения эффективных сечений (см. табл. 1) и уменьшения концентраций вредных поглотителей - водорода и кислорода. Для кислорода эффект уменьшения концентрации перекрывает эффект увеличения эффективного сечения поглощения.
В табл. 3 приведены коэффициенты использования нейтронов в активной зоне $\theta_a$, $\rho^{233}$ и коэффициенты воспроизводства, соответствующие различным отношениям $\rho_n/\rho_{233}$.

Цифры, представленные в табл. 3, указывают на относительно слабую зависимость коэффициента воспроизводства и $\theta_a$ от жесткости спектра, а величина среднего количества вторичных нейтронов для $U^{233} - \bar{p}_{233}$ вообще остается практически постоянной при изменении $\rho_n/\rho_{233}$.

Для того, чтобы выяснить причину таких небольших изменений коэффициента воспроизводства, рассмотрим спектр нейтронов в уран-ториевом реакторе УТР.

Таблица 3

<table>
<thead>
<tr>
<th>$\rho_n/\rho_{233}$</th>
<th>$\theta_a$</th>
<th>$\bar{p}_{233}$</th>
<th>КВ</th>
</tr>
</thead>
<tbody>
<tr>
<td>5,0</td>
<td>0,942</td>
<td>2,284</td>
<td>1,127</td>
</tr>
<tr>
<td>10,2</td>
<td>0,940</td>
<td>2,278</td>
<td>1,110</td>
</tr>
<tr>
<td>20,0</td>
<td>0,931</td>
<td>2,274</td>
<td>1,083</td>
</tr>
<tr>
<td>30,0</td>
<td>0,921</td>
<td>2,273</td>
<td>1,063</td>
</tr>
</tbody>
</table>

Рис.3 Спектр нейтронов $\Phi(\nu)$ в различных точках реактора (для невыгоревшего варианта с $\rho_n/\rho_{233} = 20$) и $\Phi(\nu)$ для выгоревшего варианта

На рис.3 изображен поток нейтронов $\Phi(\nu, \tau)$, отнесенный к единичному интервалу летаргий, как функция летаргии "$\tau". На оси летаргий обозначены также энергии Е, соответствующие различным летаргиям.

Для сравнения, на рис.3 представлены функции $\Phi(\nu, \tau)$ в двух точках реактора – в центре активной зоны и в зоне воспроизводства на расстоянии 5 см от активной зоны. $\Phi(\nu, \tau)$ на рис.3 нормированы так, что площади, ограниченные этими кривыми, равны.
Распределение плотности делений в реакторе: $F(u,r)$, равное произведению $\Sigma_{f}^{233}(u) \cdot \Phi(u,r)$ — есть число делений, происходящих за 1 сек в 1 см$^3$ в данной точке г, которые лежат в единичном интервале летаргий. На рис.1 изображены функции $F(u,r)$ в центре активной зоны, соответствующие $\rho_{n}/\rho_{233} = 5,0$ и $\rho_{n}/\rho_{233} = 10,2$. На рис.1 видно, что плотность деления $F(u,r)$ имеет два максимума — один в области энергий $3 - 0,5$ Мэв, другой — в области $1 - 100$ эв. Первый максимум соответствует максимуму кривой $\Phi(u)$ (рис. 3) в области высоких энергий (спектр нейтронов деления); наличие второго максимума вызвано тем, что в энергетическом интервале $1 - 100$ эв лежат резонансы сечений деления урана-233.

Характеристикой рассмотренных спектров, соответствующих различным отношениям $\rho_{n}/\rho_{233}$, является доля делений, которые происходят в тепловой области энергий ниже границы 0,4 эв. В варианте с самым жестким спектром ($\rho_{n}/\rho_{233} = 5,0$) ниже 0,4 эв происходит всего 2% делений, в варианте с $\rho_{n}/\rho_{233} = 10,2 - 7$% делений, наконец в варианте с $\rho_{n}/\rho_{233} = 20,0 - 19$% делений.

В табл. 4 приводятся величины суммарных плотностей делений $\int_{u_{1}}^{u_{2}} F(u)du$ в различных энергетических группах, а также интервалы летаргий $\Delta u_{j}$ и энергий $\Delta E_{j}$, которые соответствуют энергетическому разбиению на группы, принятому в настоящих расчетах. $\int_{u_{1}}^{u_{2}} F(u)du$ определены в центре активной зоны для 3-х случаев, отличающихся количеством воды в активной зоне: 1) $\rho_{n}/\rho_{233} = 5,0$; 2) $\rho_{n}/\rho_{233} = 10,2$ и 3) $\rho_{n}/\rho_{233} = 20,0$.

Величина $\int_{u_{1}}^{u_{2}} F(u)du$ в табл.4 нормированы таким образом, что $\int_{u_{1}}^{u_{2}} F(u)du$ - полное число делений во всех группах - равно 1 в центре активной зоны.

Анализируя различные спектральные характеристики реактора УТР, можно прийти к выводу, что эффективная энергетическая область, в которой преимущественно происходят ядерные реакции, играющие основную роль в процессе воспроизводства, лежит в довольно широких пределах.

Этим фактом, очевидно, можно объяснить слабую зависимость относительных эффективных сечений $\sigma_{\phi}^{233}$, а, следовательно, и коэффициентов $\theta$ и $\mathbf{K}$ от жесткости спектра.

Как уже указывалось (см. табл. 3), $v_{\phi}^{233}$, усредненное по спектру в активной зоне, практически не зависит от $\rho_{n}/\rho_{233}$. Отчасти это вызвано "размазанностью" спектра, а, кроме того, связано с предположением, что $v_{\phi}^{233}(E) \equiv \text{const}$ во всем интервале энергий, за исключением области выше ~20 кэв. Однако на самом деле, как указывают более поздние работы (см. рис.1), $v_{\phi}^{233}$ заметно меняется с энергией как в области низких энергий (1 - 100 эв), так и в области высоких энергий, где наблюдается сильный рост $v_{\phi}^{233}$. Использование этих современных данных несомненно "усилит" зависимость $v_{\phi}^{233}$, а следовательно, и коэффициента воспроизводства от жесткости спектра.

На рис.1 ясно видно, что если раньше изменение вида функции плотности делений $F(u)$ не могло привести к заметным изменениям усредненной по спектру величине $\tilde{v}_{\phi}^{233}$ (поскольку $v_{\phi}^{233}$ практически не зависело от энергии), то $\tilde{v}_{\phi}^{233}$, полученное на основе современных величин, будет гораздо сильнее зависеть от спектра нейтронов. Действительно, при уменьшении отношения $\rho_{n}/\rho_{233}$ спектр "перемещается" из области низких значений $v_{\phi}^{233}$ (1 - 100 эв) в область высоких энергий, где наблюдается сильный рост $v_{\phi}^{233}$.

Необходимо было выяснить, не вызовет ли наличие "провалов" в величине $v_{\phi}^{233}$ уменьшения коэффициента воспроизводства. Сравнение величин
<table>
<thead>
<tr>
<th>№ группы</th>
<th>1</th>
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<th>3</th>
<th>4</th>
<th>5</th>
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<tr>
<td>ΔE₁, эв</td>
<td>10 \cdot 10^6 - 6,065 \cdot 10^6</td>
<td>6,065 \cdot 10^6 - 3,679 \cdot 10^6</td>
<td>3,679 \cdot 10^6 - 1,353 \cdot 10^6</td>
<td>1,353 \cdot 10^6 - 0,498 \cdot 10^6</td>
<td>0,498 \cdot 10^6 - 0,0248 \cdot 10^6</td>
<td>24,8 \cdot 10^3</td>
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<tr>
<td>ΔU₁</td>
<td>0,5</td>
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<td>1</td>
<td>1</td>
<td>3</td>
<td>4</td>
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<tr>
<td>ρₙ/ρ₂₃₃</td>
<td>= 5</td>
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<tr>
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<td>0,0559</td>
<td>0,0690</td>
</tr>
<tr>
<td>ρₙ/ρ₂₃₃</td>
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<td>0,0078</td>
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<table>
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<tbody>
<tr>
<td>ΔE₁, эв</td>
<td>454 - 61,4</td>
<td>61,4 - 3,06</td>
<td>3,06 - 1,13</td>
<td>1,13 - 0,414</td>
<td>0,414 - 0,0265</td>
<td>Тепловая</td>
</tr>
<tr>
<td>ΔU₁</td>
<td>2</td>
<td>3</td>
<td>1</td>
<td>1</td>
<td>2,75</td>
<td></td>
</tr>
<tr>
<td>ρₙ/ρ₂₃₃</td>
<td>= 5</td>
<td>0,1634</td>
<td>0,2426</td>
<td>0,0376</td>
<td>0,0169</td>
<td>0,0176</td>
</tr>
<tr>
<td>ρₙ/ρ₂₃₃</td>
<td>= 10,2</td>
<td>0,1450</td>
<td>0,3277</td>
<td>0,0827</td>
<td>0,0394</td>
<td>0,0559</td>
</tr>
<tr>
<td>ρₙ/ρ₂₃₃</td>
<td>= 20</td>
<td>0,1048</td>
<td>0,3239</td>
<td>0,1248</td>
<td>0,0614</td>
<td>0,1185</td>
</tr>
</tbody>
</table>
Изменение коэффициента воспроизводства в процессе работы реактора

При изучении изменения коэффициента воспроизводства в процессе работы реактора, кроме основных эффектов - выгорания $^{233}\text{U}$ и накопления осколков деления в активной зоне, учитывались следующие процессы:
1) вредное поглощение нейтронов в $^{234}\text{U}$, $^{235}\text{U}$ и $^{236}\text{U}$;
2) деление $^{234}\text{U}$ и $^{236}\text{U}$ в области высоких энергий;
3) деление и $^{235}\text{U}$ во всей области энергий.

В зоне воспроизводства исследовалось накопление $^{233}\text{U}$ и $^{233}\text{Pa}$. Решились соответствующие уравнения выгорания, причем на 1-ой стадии расчетов предполагалось, что выгорание равномерно распределено по объемам активной зоны и отражателя. В то время как коэффициент неравномерности в активной зоне невелик, в зоне воспроизводства накопление $^{233}\text{U}$ и $^{233}\text{Pa}$ будет происходить очень неравномерно. Однако в реакторе УТР предполагается производить перестановки горючего по радиусу, что приведет к более равномерному выгоранию.

Точное изучение неравномерного выгорания с перемешиванием горючего по радиусу потребует специальных расчетов.

В табл. 5 представлены результаты оценок изменения коэффициента воспроизводства при 25% глубине выгорания. При этом отдельно определен вклад каждого изотопа в коэффициент воспроизводства ($\Delta K$). Расчеты производились для случая, когда $\rho_1/\rho_{233} = 10,2$ (в начале кампании).

В табл. 6 приводятся отношения эффективных сечений различных изотопов, накапливающихся в активной зоне, к эффективному сечению поглощения для $^{233}\text{U}$ ($\sigma_1/\sigma_{233} = \sigma_2/\sigma_{233}$), определяемые по формуле (6).

Усреднение сечений производилось по спектру выгоревшей активной зоны при 25% глубине выгорания (см. рис. 3).

В табл. 6 приведены также резонансные интегралы $I_{\text{Эд}}$, тепловые сечения (при энергии 0,0253 эв) и отношения тепловых сечений $[\sigma_1/\sigma_{233}]$ (0,0253 эв).

Как видно из табл. 5, наибольший (отрицательный) вклад в коэффициент воспроизводства дает поглощение в продуктах деления. Поглощение и деление в $^{236}\text{U}$ настолько мало, что им можно пренебречь.
ВОСПРОИЗВОДСТВО ГОРЮЧЕГО В УТР

Таблица 5
РЕЗУЛЬТАТЫ ОЦЕНКИ ИЗМЕНЕНИЯ КОЭФФИЦИЕНТА ВОСПРОИЗВОДСТВА
25%-НОЙ ГЛУБИНЕ ВЫГОРАНИЯ

<table>
<thead>
<tr>
<th></th>
<th>( \Delta \text{КВ/КВ}_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Продукты деления</strong></td>
<td></td>
</tr>
<tr>
<td>( U^{234} )</td>
<td></td>
</tr>
<tr>
<td>Поглощение</td>
<td>-0,06</td>
</tr>
<tr>
<td>Деление</td>
<td>+0,7 ( \times 10^{-2} )</td>
</tr>
<tr>
<td>( U^{235} )</td>
<td></td>
</tr>
<tr>
<td>Поглощение</td>
<td>-0,41 ( \times 10^{-2} )</td>
</tr>
<tr>
<td>Деление</td>
<td>+0,76 ( \times 10^{-2} )</td>
</tr>
<tr>
<td>( U^{236} )</td>
<td></td>
</tr>
<tr>
<td>Поглощение</td>
<td>-0,29 ( \times 10^{-4} )</td>
</tr>
<tr>
<td>Деление</td>
<td>+0,12 ( \times 10^{-4} )</td>
</tr>
</tbody>
</table>

Таблица 6
РЕЗОНАНСНЫЕ ИНТЕГРАЛЫ, ТЕПЛОВЫЕ СЕЧЕНИЯ И ИХ ОТНОШЕНИЯ

<table>
<thead>
<tr>
<th></th>
<th>( U^{233} )</th>
<th>( U^{234} )</th>
<th>( U^{235} )</th>
<th>( U^{236} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \sigma(0,0253 \text{ эв}), \text{ барн} )</td>
<td>50</td>
<td>578</td>
<td>525</td>
<td>53</td>
</tr>
<tr>
<td>( \frac{I_{\text{фсд}}}{\text{барн}} )</td>
<td>230</td>
<td>1200</td>
<td>900</td>
<td>300</td>
</tr>
<tr>
<td>( \sigma_{233}^{233} )</td>
<td>0,131</td>
<td>1</td>
<td>0,890</td>
<td>0,112</td>
</tr>
<tr>
<td>( \left[ \frac{\sigma_1}{\sigma_2} \right]_{0,025} )</td>
<td>0,03</td>
<td>1</td>
<td>0,908</td>
<td>0,092</td>
</tr>
</tbody>
</table>

Суммарный эффект от всех процессов во всех изотопах (кроме осколков деления), накапливающихся в активной зоне, составляет \( \sim -0,02 \).

Аналогичным образом исследовался процесс изменения изотопного состава в зоне воспроизводства. Определялось влияние \( \text{Pa}^{233} \) и \( U^{233} \), накапливающихся в ториевых блоках, на коэффициент воспроизводства. \( \text{Pa}^{233} \) имеет большой резонансный интеграл захвата нейтронов (1200 барн) и это, естественно, вызывало опасения, что в реакторе УТР, имеющем очень жесткий спектр нейтронов, накопление протактиния приведет к значительному снижению коэффициента воспроизводства. Однако оказалось, что эффект протактиния относительно невелик: \( \Delta \text{КВ}_{\text{Pa}} = -0,015 \). Это объясняется тем, что в нашей схеме протактиний образуется только в зоне воспроизводства, где потоки промежуточных нейтронов гораздо меньше, чем в активной зоне.

Положительный вклад \( U^{233} \), накапливающегося в зоне воспроизводства, в КВ оказался весьма близким по абсолютной величине к эффекту протактиния: \( \Delta \text{КВ}_{233} \approx +0,013 \).

Таким образом, если учесть все процессы, связанные с изменением изотопного состава в процессе работы уран-ториевого реактора, средний за
кампанию коэффициент воспроизводства окажется равным ~1,11 (при 25% глубине выгорания \(\text{U}^{233}\) в активной зоне). При этом начальный и конечный коэффициенты воспроизводства равны соответственно: 1,15 и 1,06.

ЗАКЛЮЧЕНИЕ

Данная работа содержит нейтронно-физический расчет \(\text{U}^{233} - \text{Th}^{232}\) промежуточного энергетического двухзонного реактора-бридера на обычной воде. Основная часть работы посвящена исследованию зависимости коэффициента воспроизводства от спектра нейтронов, размеров отражателя (зоны воспроизводства), количества вредных поглотителей, а также от используемых ядерно-физических констант (главным образом от \(\nu^{233}_e\)).

Дан обзор опубликованной к настоящему времени ядерно-физической информации, касающейся элементов, используемых в реакторе УТР. Кратко рассмотрены вопросы выгорания горючего.

Оказалось, что коэффициент воспроизводства сравнительно слабо зависит от жесткости спектра. Это можно объяснить тем, что эффективная область энергий, в которой "работает" реактор УТР, довольно широка. Зависимость KB от спектра нейтронов усиливается, если воспользоваться новыми значениями \(\nu^{233}_e\) (E), согласно которым \(\nu^{233}_e\) заметно изменяется с энергией, начиная с 1 эв (рис.1).

Расчеты, выполненные для различных составов активной зоны и зоны воспроизводства, показали, что в реакторе УТР может быть получен коэффициент воспроизводства в начале кампании ~1,15. KB в конце кампании, при 25% глубине выгорания \(\text{U}^{233}\) активной зоны, оказался равным ~1,06, а средний за кампанию KB составляет ~1,11.

Таким образом, есть основания полагать, что в реакторе типа УТР коэффициент воспроизводства превысит единицу, даже если учесть некотоный запас на потери в химическом и металлургическом производстве (отравление ксеноном-135 и самарием-149 мало вследствие жесткости спектра).

Для того, чтобы с большей определенностью решить вопрос о возможности расширенного воспроизводства в уран-ториевых реакторах типа УТР, необходимо в первую очередь иметь измеренные с высокой точностью энергетические зависимости сечений поглощения нейтронов в \(\text{Th}^{232}, \text{Ra}^{233}\), конструкционных материалах, выших изотопах урана, а также \(\nu^{233}_e\) (E) в промежуточной и быстрой областях энергий.

Усовершенствование расчетной методики на данном этапе вряд ли имеет смысл, так как основная неопределенность при вычислении коэффициента воспроизводства заложена в сечениях для \(\text{U}^{233}, \text{Th}^{232}\), конструкционных материалах и др.

Получить необходимые экспериментальные данные можно либо путем непосредственных измерений сечений и \(\nu^{233}_e\), либо с помощью интегральных экспериментов на критических сборках.

Помимо высокого коэффициента воспроизводства, для экономически выгодной ядерной энергетики необходимо иметь высокую удельную теплоющую мощность на единицу веса ядерного горючего. В уран-ториевом реакторе-бридере рассмотренной конструкции с замедлителем и теплоносителем из обычной воды теплонапряженность единицы объема активной зоны можно довести до 1000 ~ 2000 квт на 1 л при теплонапряженности на единицу веса ядерного горючего 2000 ~ 5000 квт на 1 кг.
В заключение заметим, что общие закономерности, полученные в данной работе, такие как характер изменения коэффициента воспроизводства при изменении жесткости спектра, количества конструкционных материалов, толщины отражателя и т.п., могут быть полезны при выборе оптимальных вариантов реактора типа УТР.

Автор приносит глубокую благодарность профессору С.М. Фейнбергу, под научным руководством которого производилось изучение воспроизводства горючего в уран-ториевом реакторе УТР.

Некоторые проработки и расчеты реактора УТР проводились В.А. Чеботаревым, а численная методика расчета реактора в Р_1 — приближении была разработана Н.Я. Лященко, которым автор выражает искреннюю признательность.

ЛИТЕРАТУРА

[1] КУРЧАТОВ И. В. Некоторые вопросы развития атомной энергетики в СССР. Доклад в Харуэлле (Англия), 1956
THORIUM RESOURCES IN THE UNITED ARAB REPUBLIC AND THEIR POSSIBLE UTILIZATION

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INTRODUCTION

The beach deposits on the Mediterranean coast near the mouth of the Rosetta branch of the Nile have attracted attention for a considerable period of time, especially as they contain lenticular strips of very dark black sands. The rich sands have been exploited on a small industrial scale since 1932, thus producing fractions of ilmenite, magnetite, zircon, monazite, garnet and rutile in a factory near Alexandria. Black sands have also been noticed in the Mediterranean Sea bottom close off the coast between Rosetta and Damietta. The not so well known black sand deposits on the Mediterranean coast to the east of the Damietta branch of the Nile have lately been investigated by the United Arab Republic Atomic Energy Establishment, and they have been found to contain large reserves of monazite. The aerial radiometric work carried out by the Establishment helped in the delineation of the surface extensions of the above deposits which have been carried by the River Nile, laid down in the Mediterranean Sea, and transported to its shores by water currents, winds, waves and other agents.

Other monazite-bearing, but less important black sands have lately been discovered on the Mediterranean Sea coast of Sinai. Present information on these deposits has been gained by the aerial radiometric work of the Atomic Energy Establishment, and the studies of the Geological Survey and Mineral Research Department and Cairo University. Views differ as to the origin of these deposits, however, the author of the present work believes that they may be largely contributed by the local drainage of Sinai, and to some extent by the Nile sediments.

Placer deposits of Quaternary age have been encountered at the mouths of the valleys in the Eastern Desert and Sinai, especially those draining the basement rocks and discharging into the Red Sea and its gulfs. Fossil black sands containing monazite and other radioactive materials are known in several localities in the sedimentary column especially in geological formations of sandy facies, however, because monazite is more accessible in the unconsolidated and better exposed beach placers, no great efforts have been spent to study such occurrences in detail.

Finally, prospecting work carried out by the Atomic Energy Establishment led to the discovery of uranium and thorium-uranium occurrences in the basement rocks of the Eastern Desert. Investigations are being carried out to evaluate them, however, the thorium would only be recovered in such deposits as a by-product of uranium exploitation.
For the purpose of the present work only the following types of deposits will be dealt with, the first two in brief, and the third which is the most important thorium resource, in more detail:

1. Thorium-uranium mineralizations in the basement rocks.
3. Monazite-bearing beach placers on the Mediterranean Sea coast derived from the Nile and localized between Damietta and Port Said, East Rosetta and West Rosetta.

THORIUM-URANIUM MINERALIZATIONS IN THE BASEMENT ROCKS

Potential resources of thorium and uranium have recently been discovered by the UAR Atomic Energy Establishment in the Basement rocks of the Eastern Desert, and are being investigated to estimate their reserves. Of these, the deposits of Wadi Gemal [1] and Abu Garadi [2] were first discovered by carborne radiometric methods, while the mineralization of Um Safi was first found by aerial radiometric prospecting [3].

At Wadi Gemal thorium and uranium occurs dispersed in a psammitic gneiss formation, where uranothorite is the main radioactive mineral present. Thorium-uranium ratios range from 1 to 15 although they vary normally between 1 and 4; the highest values so far recorded in the deposit are thorium, 0.66% and uranium, 0.33%.

At Abu Garadi the radioactive mineralization occurs in fracture zones traversing late orogenic granite, these usually strike north-south and are almost vertical. The main radioactive mineral recorded is uranothorite, with a thorium-uranium ratio of 4 in the unaltered mineral, but which ratio may increase to 9 if the mineral is altered. Furthermore, secondary radioactive minerals have been encountered in the oxidation zone, which include thorogummite, uranophane and autunite. Thorium may reach 0.17% and uranium 0.05% in samples from Abu Garadi where the thorium-uranium ratio varies between 1.7 and 7.8 [4, 5]. Moreover, thorium-uranium mineralization occurs at Um Safi in a highly altered and fractured leucite mass, where thorium in the analysed samples has ranged from 0.04 - 0.2% [4].

MONAZITE-BEARING PLACERS ON THE MEDITERRANEAN COAST OF SINAI

Beach and dune deposits extending on the Sinai Mediterranean coast from Bardawil eastwards have been mineralogically examined by Shukri and Philip [6]. They concluded that the deposits in question are contributed by the Nile sediments carried towards the east by long shore currents with additions of local material brought about by torrents coming from the south especially those passing through Wadi El Arish. The addition of local material is evidenced by a higher amphibolepyroxene ratio in comparison to the deposits contributed solely by the Nile. Furthermore, the deposits of the El Arish area show an increase in the relative frequency of zircon, garnet, epidote and staurolite.
El Far [7] reported that the beach sand deposits in the Sinai coastal strip are found as black sand bars, grey mantle and submantle, and grey dunes. The black sand bars are 0.5 to +40 m in width, 1 to +50 cm in thickness and they were estimated to contain 1 342 460 t in 1958. The black sands are particularly concentrated between El Arish and Katib El Kals. The grey mantle and submantle are less concentrated deposits formed of alternate dark and light bands; they are 100 to +200 m in width, 1.2 m thick and they are estimated to contain 3 705 040 t. Finally, the grey dunes are estimated to contain 17 100 t in the area between El Arish and El Massaid.

### TABLE I

**DISTRIBUTION OF ECONOMIC MINERALS AND MONAZITE IN SAMPLES FROM THE BEACH SAND DEPOSITS OF SINAI MEDITERRANEAN COASTAL STRIP**

<table>
<thead>
<tr>
<th>Locality</th>
<th>Sample No.</th>
<th>Total economic minerals (%)</th>
<th>Monazite (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ras El Hattab</td>
<td>1</td>
<td>70.97</td>
<td>2.23</td>
</tr>
<tr>
<td>Ras El Hattab</td>
<td>2</td>
<td>85.6</td>
<td>1.4</td>
</tr>
<tr>
<td>West El Arish</td>
<td>3</td>
<td>25.31</td>
<td>1.74</td>
</tr>
<tr>
<td>El Arish</td>
<td>1</td>
<td>58.13</td>
<td>-</td>
</tr>
<tr>
<td>El Arish</td>
<td>2</td>
<td>47.52</td>
<td>0.01</td>
</tr>
<tr>
<td>El Arish</td>
<td>4</td>
<td>68.96</td>
<td>1.05</td>
</tr>
<tr>
<td>El Arish</td>
<td>5</td>
<td>61.23</td>
<td>0.12</td>
</tr>
<tr>
<td>El Arish</td>
<td>6</td>
<td>33.25</td>
<td>0.20</td>
</tr>
<tr>
<td>El Arish</td>
<td>7</td>
<td>19.60</td>
<td>0.02</td>
</tr>
<tr>
<td>Bardawil</td>
<td>1</td>
<td>56.68</td>
<td>-</td>
</tr>
<tr>
<td>Bardawil</td>
<td>3</td>
<td>20.09</td>
<td>0.15</td>
</tr>
<tr>
<td>Bardawil</td>
<td>5</td>
<td>12.52</td>
<td>0.03</td>
</tr>
<tr>
<td>Rafah</td>
<td>1</td>
<td>15.0</td>
<td>0.13</td>
</tr>
<tr>
<td>Rafah</td>
<td>2</td>
<td>40.07</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Mineralogical analysis of samples from the Sinai Mediterranean coastal strip has been carried out mainly by A. Ghoneim, and one sample has been examined by G. Saleeb; the total economic heavy minerals and monazite content are given in Table I, the heavy minerals being represented by ilmenite, magnetite, haematite, zircon, monazite, rutile and garnet. The monazite in the samples ranges from 2.23% to 0%; the highest values are encountered near El Arish and at Ras El Hattab.
MONAZITE-BEARING BEACH PLACERS DERIVED FROM THE NILE

General

The River Nile flows from Central Africa and the Abyssinian Plateau carrying with it broken material from the Precambrian basement rocks, volcanic flows and sediments. The average discharges of the Nile and its tributaries have been given by Hurst [8] for 1912-47 as 83 100 million m$^3$ for the main Nile at Aswan. If it is assumed that the Nile at Aswan receives seven units, one would be contributed by Atbara, four by the Blue Nile, and two units by the White Nile at Khartoum (one from the Sobat and one from the White Nile below the swamps).

Kuenen [9], however, estimated that the Nile carries 100 million m$^3$ of sediments to the Mediterranean annually, of which 15-45% are sand, the rest being clay and silt. Assuming that the sand fraction contains 1% heavy minerals and the specific gravity of the sand fraction is 1.5, the amount of heavy minerals discharged by the Nile to the Mediterranean annually is estimated to range from 225 000 to 675 000 t.

At present the Nile, after bifurcating north of Cairo, discharges its waters to the Mediterranean through the Damietta and Rosetta branches which embrace the Nile Delta. It is believed by Ball [10] that the Nile Valley in its present general form has been established by the regional uplift at the end of the Middle Miocene and the subsequent erosion. The Mediterranean coastal zone is rather flat at present, and after passing through gradual rising for 20 000 yr from the Middle Pleistocene to the Upper Pleistocene-Paleolithic, it underwent slow continental subsidence in the Nile Delta area at a rate of 1.4 mm/yr which seems to be continuing still [10].

The sediments carried by the Nile after discharging into the Mediterranean are piling up mainly to the north of the Delta, towards the east and some towards the west as evidenced by the depth contours of the floor of the Mediterranean (Fig. 1). The water currents in the Mediterranean in this part are mainly directed to the east (Fig. 1), although a back current exists locally in Abu Qir Bay. The prevailing winds at Damietta from April to December are directed NW-SE and accordingly the waves produced thereof hit the beach obliquely. In general, the natural forces are in favour of sorting, concentration and sedimentation of the heavy minerals in favourable wide stretches along the beach, especially in the following areas: (1) Damietta eastwards towards Port Said where the maximum concentration is encountered at a distance of 10 to 11 km from the Nile mouth at Damietta, (2) Rosetta eastwards towards Burullus, and (3) Rosetta westwards towards Abu Qir and this is believed to be mainly affected by the back currents in Abu Qir Bay.

The minerals carried by the Nile and its tributaries have been given by Shukri [11], in particular the distribution and derivation of iron ores, amphiboles, pyroxenes, sillimanite, epidote and garnet. The present writer and his collaborators divided the minerals of the beach placer deposits according to their specific gravity and their behaviour in the sedimentation processes into three groups, namely (1) minerals with relatively high specific gravity (more than 3.85) including mainly the economic minerals il-
menite, magnetite, zircon, monazite rutile, garnet and other uncommon minerals including spinel etc., (2) minerals with moderate specific gravity (3.84 to 2.95) with amphiboles, pyroxenes, biotite, epidote, staurolite, sillimanite, olivine, tourmaline, sphene etc. (these behave in sedimentation in approximately the same manner as the heavy minerals) and (3) minerals with relatively small specific gravity (less than 2.95) including quartz, feldspars, glauconite, calcite, etc. which act in an opposite way to the heavy minerals. In a recent study by Zaki [12] the major valuable minerals in the heavy fractions of the black sands of Damietta and Rosetta are ilmenites and magnetites, while the minor constituents present in appreciable amounts include pseudo-brookite, rutile, haematite, chromite, cassiterite, zircon and monazite. Minute quantities of gold grains have been observed by Zaki and Hammoud [13]; the latter managed to separate the gold grains on a laboratory scale. Moreover, Zaghloul [14] identified uranothorite grains in monazite and zircon fractions of Rosetta sands. The normal industrial fractions separated from the black sands of Rosetta are ilmenite, magnetite, zircon, monazite, rutile and garnet.

Physical and mechanical properties of monazite

The beach sands on the Mediterranean are almost flat sediments with alternate dark and light coloured bands. Their position in relation to other local mineral deposits has been given by El Shazly [15]. The maximum depth to which the black sands have been tested is 20 m, although examining their sedimentation conditions will lead to the assumption that they extend further in depth. The size of mineral grains is usually fine to very fine, especially
for the heavy minerals. In Damietta samples the coarse fraction (+0.500 mm) is 0 to 0.240%, the medium fraction (+0.250 mm) is 0.113 to 4.862%, the fine fraction (+0.125 mm) is 40.266 to 89.425%, and the very fine fraction (+0.064 mm) is 6.745 to 59.552% [16]. The black sands at Rosetta east are slightly coarser than those of Damietta, the median diameter for Rosetta east being 0.163 [17] and for Damietta being 0.144 [16]; however, the difference is small. A very high degree of sorting is also characteristic for the black sands and the sorting coefficient determined for Damietta and Rosetta samples is 1.27. The deposition of monazite in relation to the other heavy minerals is governed to a great extent by its relatively higher specific gravity (5.1) and its roundness which is more pronounced than the other heavy minerals. Thus will explain the relatively high concentration of monazite in the rich sands not far from the mouths of the Nile; monazite is deposited somewhat quicker than most of the other heavy minerals and it is transported for a relatively shorter distance along the beach (see Table II).

**TABLE II**

**SOME PROPERTIES OF MONAZITE FROM ROSETTA BLACK SANDS**

<table>
<thead>
<tr>
<th>Relative roundness</th>
<th>Percentage</th>
<th>Length classes</th>
<th>Percentage</th>
<th>Elongation L/B</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Angular</td>
<td>0.5</td>
<td>0.2185 to 0.184 mm</td>
<td>1.0</td>
<td>1.0 to 1.5</td>
<td>69.25</td>
</tr>
<tr>
<td>Sub-angular</td>
<td>0.75</td>
<td>0.184 to 0.1495 mm</td>
<td>10.5</td>
<td>1.5 to 2.0</td>
<td>25.25</td>
</tr>
<tr>
<td>Sub-rounded</td>
<td>7.5</td>
<td>0.1495 to 0.115 mm</td>
<td>53.5</td>
<td>2.0 to 2.5</td>
<td>5.0</td>
</tr>
<tr>
<td>Rounded</td>
<td>21.5</td>
<td>0.115 to 0.0805 mm</td>
<td>34.5</td>
<td>2.5 to 3.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Well rounded</td>
<td>69.75</td>
<td>0.0805 to 0.046 mm</td>
<td>0.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The average refractive index (n) for monazite from Rosetta is 1.796 for the pale yellow, and it is slightly lower for the brownish altered grains. Pleochroism is very weak for monazite and its optic axial angle is +13°. X-ray diffraction examination shows that the mineral in question is partly present in a metamict state [18].

Colour variations in monazite from Rosetta are given as follows: lemon yellow 59%, honey yellow 25%, colourless 6%, pale green 5.5%, brown 2.5% and dark yellow 2.0% [19]. The monazite is very well sorted with an estimated sorting coefficient of 1.10, and it is very-fine to fine grained with a median diameter of 0.125 mm. Monazite grains studied by Kamel Hassan [19] from Rosetta are normally well rounded to rounded; they are the smallest and least variable grains in length among the heavy minerals and they may contain zoned zircon inclusions. Egg-shaped, tablet-like or barrel-shaped grains are of common occurrence; moreover, euhedral crystals with rounded corners are encountered as well as some grains with hexagonal, octagonal or quadrilateral outlines.
Thorium and uranium content of monazite

The ThO₂ content of monazite for a Rosetta sample examined under the microscope is 5.92% while the U₃O₈ content is 0.45% [18]. Lower percentages of thorium and uranium reported in previous analyses of monazite from the same locality are definitely caused by the presence of other minerals.

### TABLE III
CHEMICAL AND SPECTROGRAPHIC ANALYSIS OF A MONAZITE SAMPLE FROM ROSETTA BLACK SANDS

<table>
<thead>
<tr>
<th>Chemical analysis (%)</th>
<th>Composition of rare earth elements separated from monazite (at.%)</th>
<th>Trace elements (g/t)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ThO₂ 5.92</td>
<td></td>
<td>Al 500</td>
</tr>
<tr>
<td>Ce₂O₃ 26.80</td>
<td></td>
<td>Be 50</td>
</tr>
<tr>
<td>La₂O₃ 13.70</td>
<td></td>
<td>Mg 700</td>
</tr>
<tr>
<td>Pr₂O₃ 3.77</td>
<td></td>
<td>Mn 40</td>
</tr>
<tr>
<td>Nd₂O₃ 11.93</td>
<td>La 21.9</td>
<td>Nb 30</td>
</tr>
<tr>
<td>Sm₂O₃ 2.74</td>
<td>Ce 42.9</td>
<td>Na 10</td>
</tr>
<tr>
<td>Gd₂O₃ 1.44</td>
<td>Pr 5.9</td>
<td>Sn 50</td>
</tr>
<tr>
<td>Y₂O₃ 2.26</td>
<td>Nd 19.2</td>
<td>Zr 80</td>
</tr>
<tr>
<td>U₂O₈ 0.46</td>
<td>Sm 4.4</td>
<td></td>
</tr>
<tr>
<td>TiO₂ 0.22</td>
<td>Gd 2.4</td>
<td></td>
</tr>
<tr>
<td>Fe₂O₃ 0.44</td>
<td>Y 3.3</td>
<td></td>
</tr>
<tr>
<td>CaO 0.47</td>
<td>La + Ce + Pr</td>
<td></td>
</tr>
<tr>
<td>PbO 0.10</td>
<td>(Murata's index) = 70.7</td>
<td></td>
</tr>
<tr>
<td>Pb₂O₄ 27.27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SiO₂ 2.62</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total 100.13</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The thorium present in monazite is explained by several authors to be the result of the replacement of trivalent cerium lanthanides by quadrivalent thorium. It is believed by the writer that the chemical composition of monazite in beach placers derived from the Nile will be more or less constant owing to the excellent mixing and sorting during sedimentation, although varieties of the mineral have been reported. The chemical and spectrographic analysis of monazite reported lately by El Hinnawi [18] is given in Table III; it shows a Murata index of 70.7 for the sum of the more basic rare earths, thus suggesting a granitic derivation for monazite.
**Reserves of economic minerals, monazite, thorium and uranium in beach placers**

An estimation of the reserves of economic minerals as a whole and monazite containing ThO₂ and U₃O₈ in particular in the beach placers of Nile derivation is given in Table V. The estimation is based on the aerial radiometric surveys covering the areas of concentration on the Mediterranean beach between Port Said and Abu Qir; the flight lines which are 250 m apart are perpendicular to the beach, as well as the sampling and mineralogical analysis of the beach sands from Damietta eastwards towards Port Said, and to the east and west of Rosetta.

In the Damietta area economic minerals are particularly concentrated to the east of the mouth of the Nile Branch; the economic mineral content reaches its maximum (more than 60%) at a distance 11 km to the east of the mouth and monazite at a distance of about 3 km (Figs. 2 and 3). In the first 12 km near the mouth concentration of economic minerals constitutes 27.23% and monazite 1.05% on the average, while in the 40 km farther from the mouth concentration of economic minerals constitutes 11.98% on the average, and the monazite 0.11%. Beach sands containing more than 10% economic minerals are found in an area of the coast (31 km X 0.5 km) where the average values for the samples collected from this part are: economic minerals 22.33% and monazite 0.44%, which are used as a basis for calculating the estimated reserves of the Damietta area (Tables IV and V).

An estimation of the reserves of black sands in the top metre of an area 4.9 km² on the beach to the east of the Rosetta branch of the Nile has been given by Higazy and Naguib [20]. The coastal strip to the east of Rosetta has been sampled and mineralogically examined by Meshref [17]. Moreover, EBASCO [21] estimated the reserves in an area 6 km X 2 km and to a depth of 20 m which is located about 20 km eastwards from the Nile mouth at Rosetta. The proven reserves given for this part are 37 200 000 t of economic minerals (comparatively lower grade with an average grade of 7.75% of economic minerals excluding magnetite). EBASCO [21] also examined an area 0.5 km X 4.0 km at a depth of 20 m and a distance of 10 km westwards from the Rosetta mouth. The sand is also on the lower grade side with an average value of economic minerals of 6.75% excluding magnetite, and the proven reserves of economic minerals are 5 400 000 t.

Rosetta beach sands are characterized by the presence of lenticular stretches of particularly high grade ore; the averages of a lens of this type located 2.5 km east of Rosetta are: economic minerals 73.93%, monazite 1.16%, and a ratio of monazite to economic minerals of 1:64 [17]. This grade of highly concentrated black sands has been used for industrial mineral separation; mineralogical analysis for the normal feed used is given by Rittmann and Nakhla [22] as follows: economic minerals 77.05% monazite 1.06% and the ratio of monazite to economic minerals of 1:73. The latter values are near enough to the field results of Meshref. Calculations of the reserves of Rosetta east and west are based, however, on the averages of the normative grade beach sands which cover an area of 34 km X 1 km in the east and 12 km X 0.5 km in the west with economic minerals constituting 28.95% and monazite 0.22% on the average.
FIG. 2. Distribution of the total economic minerals content in black sands along the Mediterranean beach from Damietta towards Port Said.

FIG. 3. Distribution of monazite along the beach.
<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Distance from the mouth of Damietta branch (km)</th>
<th>Economic minerals (%)</th>
<th>Monazite (%)</th>
<th>Monazite/economic minerals</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>8.92</td>
<td>0.28</td>
<td>1:32</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>10.72</td>
<td>0.22</td>
<td>1:48</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>12.41</td>
<td>0.40</td>
<td>1:31</td>
</tr>
<tr>
<td>4</td>
<td>4</td>
<td>23.88</td>
<td>0.60</td>
<td>1:40</td>
</tr>
<tr>
<td>5</td>
<td>5</td>
<td>35.59</td>
<td>1.65</td>
<td>1:22</td>
</tr>
<tr>
<td>6</td>
<td>6</td>
<td>34.19</td>
<td>1.74</td>
<td>1:20</td>
</tr>
<tr>
<td>7</td>
<td>7</td>
<td>13.12</td>
<td>0.31</td>
<td>1:43</td>
</tr>
<tr>
<td>8</td>
<td>8</td>
<td>27.82</td>
<td>0.71</td>
<td>1:39</td>
</tr>
<tr>
<td>9</td>
<td>9</td>
<td>44.29</td>
<td>1.65</td>
<td>1:27</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
<td>59.73</td>
<td>1.80</td>
<td>1:33</td>
</tr>
<tr>
<td>11</td>
<td>11</td>
<td>60.47</td>
<td>2.96</td>
<td>1:20</td>
</tr>
<tr>
<td>12</td>
<td>12</td>
<td>18.30</td>
<td>1.33</td>
<td>1:14</td>
</tr>
<tr>
<td>13</td>
<td>13</td>
<td>4.80</td>
<td>0.02</td>
<td>1:236</td>
</tr>
<tr>
<td>14</td>
<td>18</td>
<td>14.02</td>
<td>0.05</td>
<td>1:260</td>
</tr>
<tr>
<td>15</td>
<td>23</td>
<td>9.28</td>
<td>0.005</td>
<td>1:1974</td>
</tr>
<tr>
<td>16</td>
<td>28</td>
<td>27.84</td>
<td>0.64</td>
<td>1:43</td>
</tr>
<tr>
<td>17</td>
<td>33</td>
<td>11.66</td>
<td>0.02</td>
<td>1:490</td>
</tr>
<tr>
<td>18</td>
<td>38</td>
<td>18.85</td>
<td>0.11</td>
<td>1:156</td>
</tr>
<tr>
<td>19</td>
<td>43</td>
<td>9.52</td>
<td>0.004</td>
<td>1:2213</td>
</tr>
<tr>
<td>20</td>
<td>48</td>
<td>3.77</td>
<td>0.01</td>
<td>1:419</td>
</tr>
<tr>
<td>21</td>
<td>53</td>
<td>2.92</td>
<td>0.01</td>
<td>1:198</td>
</tr>
</tbody>
</table>

The estimated reserves for the top metre and the top twenty metres of the main occurrences of beach sands derived from the Nile are given in Table V taking the value of two to represent the normal specific gravity of the sands. According to this table the top twenty metres contain the following quantities in metric tonnes: economic minerals, 616,046,000; monazite, 6,268,000; ThO₂, 371,060; and U₃O₈, 28,200. The potential resources of the placer sands in the UAR are actually considerably higher, as they are de-
TABLE V

ESTIMATED RESERVES OF ECONOMIC MINERALS, MONAZITE, ThO$_2$ AND U$_3$O$_8$ IN BEACH PLACERS DERIVED FROM THE NILE

<table>
<thead>
<tr>
<th></th>
<th>Damietta east</th>
<th>Rosetta east and Rosetta west</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Area (km)</strong></td>
<td>$31 \times 0.5$</td>
<td>$34 \times 1$ and $12 \times 0.5$</td>
<td></td>
</tr>
<tr>
<td><strong>Economic minerals, average (%)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Monazite, average (%)</td>
<td>22.33</td>
<td>29.85</td>
<td></td>
</tr>
<tr>
<td><strong>Monazite/economic minerals</strong></td>
<td>$1/52$</td>
<td>$1/136$</td>
<td></td>
</tr>
<tr>
<td><strong>Estimated reserves of economic minerals in top metre (t)</strong></td>
<td>$922 , 300$</td>
<td>$398 , 000$</td>
<td>$30 , 020 , 300$</td>
</tr>
<tr>
<td><strong>Estimated reserves of economic minerals in top 20 m (t)</strong></td>
<td>$138 , 446 , 000$</td>
<td>$477 , 600 , 000$</td>
<td>$616 , 046 , 000$</td>
</tr>
<tr>
<td><strong>Estimated monazite reserves in top metre</strong></td>
<td>$137 , 400$</td>
<td>$176 , 000$</td>
<td>$313 , 400$</td>
</tr>
<tr>
<td><strong>Estimated monazite reserves in top 20 m (t)</strong></td>
<td>$274 , 800$</td>
<td>$352 , 000$</td>
<td>$626 , 800$</td>
</tr>
<tr>
<td><strong>Estimated ThO$_2$ reserves in top metre (t)</strong></td>
<td>$8 , 134$</td>
<td>$10 , 419$</td>
<td>$18 , 553$</td>
</tr>
<tr>
<td><strong>Estimated ThO$_2$ reserves in top 20 m (t)</strong></td>
<td>$162 , 680$</td>
<td>$208 , 380$</td>
<td>$371 , 060$</td>
</tr>
<tr>
<td><strong>Estimated U$_3$O$_8$ reserves in top metre (t)</strong></td>
<td>$618$</td>
<td>$792$</td>
<td>$1 , 410$</td>
</tr>
<tr>
<td><strong>Estimated U$_3$O$_8$ reserves in top 20 m (t)</strong></td>
<td>$12 , 380$</td>
<td>$15 , 840$</td>
<td>$28 , 220$</td>
</tr>
</tbody>
</table>

Posited in one of the favourable areas for accumulation of heavy minerals in the world.

**Physical concentration of monazite**

A pilot plant for the physical concentration of local monazite from the beach sands has been constructed in the UAR Atomic Energy Establishment. The purpose of this plant is to produce high grade monazite on a small scale and to test the various parameters and the optimum conditions for the separation of monazite and other valuable minerals from the beach sands.
A monazite concentrate of 79% purity is passed on three wet tables of Wilfley type by 9-in hydraulic classifiers used for feeding and sometimes for desliming the feed. The capacity of the wet shaking tables is 50 kg/h each. The monazite concentrate yields, after passing on the tables, an upper concentrate containing monazite and zircon, and a lower concentrate containing zircon, ilmenite and a little monazite. The upper concentrate, after drying in a furnace, is fed into a Ding's cross belt magnetic separator, under a field of moderate magnetic intensity; a magnetic fraction containing ilmenite is separated from a relatively non-magnetic fraction with zircon and monazite. The last fraction is passed again in the magnetic separator, this time under a field of high magnetic intensity, thus yielding a fraction mainly constituted of zircon and another fraction composed mainly of monazite with some zircon belonging to a magnetic variety and ilmenite. The last fraction is passed into a Carpco electrostatic separator (capacity 20 kg/h), first under an electrostatic field to separate zircon in one fraction, and monazite and ilmenite in a second fraction, and lastly under a mixed field to separate monazite 97 to 98% purity from ilmenite [22]. The mineral analysis of the monazite concentrate produced is as follows [23]: monazite 97.8%, ilmenite 0.4%, magnetite 0%, zircon 1.1%, rutile 0.3%, garnet 0%, green silicates 0.4%, quartz 0%; total 100%.

In the field of industrial mineral separation of black sand products a project is contemplated by the Egyptian Black Sands Company using relatively low grade sands located some 20 km to the east of the Rosetta mouth. The general outline of the project of mineral production is as follows: beach sands amounting to 3,228,000 t and containing 7.75% economic minerals (excluding magnetite) are to be mined per year, and will be treated to produce 137,500 t of ilmenite, 43,750 t of magnetite, 18,750 t of zircon, 12,500 t of garnet, 3,750 t of rutile and 1,250 t of monazite annually.

**Chemical processing of monazite**

A pilot plant for the production of thorium-uranium cake with a capacity of 25 kg per batch has been constructed in the UAR Atomic Energy Establishment. This pilot plant is based on the alkaline process and is considered as a preliminary step towards the large-scale chemical processing of local monazite, and as a medium for testing the various conditions for the treatment of this mineral.

The monazite of 97 to 98% purity is first ground to -325 mesh in a ball mill of 30 kg/h capacity. Ground monazite, caustic soda and water in the proportions of 1:1.5:2.5 are mixed together and heated for three hours to a temperature of 145°C. Water is added and the product is digested in hot water at 110°C to dissolve the trisodium phosphate present. Filtration of a cake of rare earths, thorium and uranium hydroxides is carried out in a filter press of nickel plated stainless steel. Complete dissolution of the cake is affected in 37% hydrochloric acid for 30 min and later the solution is diluted with water. The pH is adjusted between 5.8 and 6 by the addition of caustic soda, thus thorium-uranium hydroxides are precipitated leaving the rare earths in solution. The thorium-uranium cake thus produced is filtered on a hard rubber filter press and later dried; it contains 12% ThO₂.
1% $\text{U}_2\text{O}_8$ with impurities of rare earths, oxides and iron oxides. The rare 
earth remaining in solution are precipitated as hydroxides by adding 40% 
caustic soda [24].

Further improvements in the above procedure are taking place, as well 
as the construction of small pilot plants to separate uranium from thorium 
using an anion exchange resin and covering the range of the processes of 
the chemical treatment of monazite.

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ECONOMIC INCENTIVE FOR
THORIUM REACTOR DEVELOPMENT*

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OAK RIDGE, TENN., UNITED STATES OF AMERICA

THE ENERGY SUPPLY PROBLEM

Throughout the nuclear power industry, people have in recent years become increasingly aware of the fact that mining uranium for its fissile content can at best be only a temporary solution to the energy supply problem. There is just not enough low cost uranium ore to sustain mankind's power needs for long, especially in view of the rapid growth of these needs. Although the vast quantity of high-cost fissile uranium existing in the earth's crust can conceivably be considered a source of energy, to utilize this source as such would entail expenditures far greater than could be tolerated by our expanding civilization. The solution to the long-term energy supply problem therefore, as everyone well recognizes, is to produce all future fissile needs by breeding rather than by mining $^{235}$U. Since an abundant supply of bred fissile material means an abundant supply of neutrons, which in turn can be used to produce more fissile material from $^{238}$U or thorium, breeding is effectively a "renewable" source of energy whose costs are no longer sensitive to the costs of mined source material. Latent energy sources such as fossil fuels and mined (and separated) $^{235}$U, on the other hand, even though they may be "relatively inexhaustible" will always be confronted by the problem of rising production costs. When application by application, these costs become prohibitive, the use of such latent sources must necessarily dwindle and the renewable sources take over.

Of the many possible renewable energy sources that exist in the world, such as wind, tides, solar energy, farm wastes, etc., only the breeding of fissile material appears to be sufficiently economic, adequate and adaptable to serve as an overall long-term energy source. Thus the question to be answered for each individual need is not "Must we develop nuclear power breeders?", but rather, "When must we develop nuclear power breeders?". In this context, the word develop should be taken to mean "develop to the point where they are economically competitive with alternative power sources" because regardless of their state of technical development no breeders will be exploited commercially unless they are economically competitive. It is obvious that the answer to the above question is "As soon as possible" merely because renewable energy sources are economically stable over a long period whereas latent energy sources are not. The main problem then is to find the optimum, shortest path to follow to develop such breeders. It is the purpose of this paper to show how thorium fits into this overall picture.

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation.
The development of any power system whether it be nuclear or conventional is complicated because of the many opposing forces which influence the path taken. In general, economic factors such as investment and operating costs, costs of money and anticipated profits dictate the direction to follow; however, for any new unit being considered, these are tempered by an assessment of the reliability and performance of the unit and the overall risk involved. Because most power systems are developing very rapidly, gross changes in technology and particularly in the fuel supply picture are likely to occur during the life span of any individual plant. In spite of this, most decisions are based mainly on prevailing conditions rather than on future possibilities, even though these latter might indicate a different direction to take.

The nuclear industry, in particular, is characterized by such an approach. Present lines of development are based almost entirely on historical origins rather than on future outlooks. These historical lines of development unfortunately involve reactor designs and systems, such as the light-water-moderated reactors, which have been optimized for minimum power costs in an era of cheap and abundant uranium. For this reason, most present-day reactors are wasteful of neutrons and thereby have very poor fissile production characteristics. Because of this, they consume rather than produce fissile material and in order to produce cheap power are more or less completely dependent on a continuing supply of low-cost uranium from the ground. Fortunately, present estimates indicate that resources of cheap uranium are relatively plentiful compared to present and near-future needs. Thus, current-technology light-water reactors appear to be an acceptable short-term solution to the energy supply problem even though they may be inadequate as a long-term energy source. The question now becomes, "How can the gap between present-day reactors and breeders best be bridged?"

One obvious approach, of course, is to start with present-day reactors and gradually improve their neutron utilization characteristics (measured in terms of fissile material produced) with the hope that they might evolve into breeders. Although this approach may result in reactors with very good fuel utilization characteristics, the disparity between neutron utilization in current reactor types and what is needed for the reactors to be efficient breeders seems too great to overcome. It is not enough for reactors to eliminate the consumption of fissile material; they must go beyond this point and produce excess fissile material at a rate adequate to meet all needs.

A second approach to the establishment of a breeder industry, which is the one generally being pursued, is to support an independent line of fast reactor development leading hopefully to commercially exploited fast breeders. The chances of success in this direction are very good; however, the technology of sodium-cooled fast breeders on which most hopes are based, is radically different from that of current technology water reactors. Unknown problems and uncertain economics confront the fast breeder line of development, as a result of which, the time required for complete success is quite uncertain. Thus, a considerable time might elapse
during which a large amount of uranium must be mined to support the $^{235}\text{U}$ consuming industry. If too long a time is allowed to elapse before bringing fast breeders on the line, the amount of money spent on mining uranium and separating the $^{235}\text{U}$ in diffusion plants will reach astronomical proportions.

A third approach which represents the middle ground between the two approaches previously described may be the answer. This approach involves the development of reactor systems whose technology is not appreciably different from that of present-day reactors but which offer the possibility of developing into efficient breeders. Such reactors cannot only reduce the consumption of uranium in the interim period but without drastic changes in technology can also solve the long range problem of providing a new source of fissile material by breeding. It is to this category of reactors that thorium-fuelled reactors belong.

FUTURE ROLE OF THORIUM CONVERTERS AND BREEDERS

The role of thorium reactors may be twofold; first, as recycle converters which make better use of mined $^{235}\text{U}$ than $^{238}\text{U}$-based converters; and second, as high-performance thermal breeders playing a primary or secondary role to fast breeders. Implementation of the first role, namely the development of advanced thorium converters, may achieve the following three objectives:

(a) Reduction in nuclear fuel cycle costs
(b) Reduction in the requirement for mined uranium and associated $^{235}\text{U}$ separation in diffusion plants
(c) Development of technology which might result in high-performance thermal breeders.

The justification for pursuing these objectives, however, depends on what it costs to attain them compared to future savings that might result. In the last section of this paper, a quantitative assessment of this aspect is given, which indicates that the present worth of future savings achievable through the successful development of advanced thorium converters followed by thermal breeders may far exceed presently planned expenditures for the development of such reactors.

The primary incentive for pursuing the longer-range goal of establishing a thermal breeder complex of significant size is that such breeders have lower specific inventories ($\text{kg fissile/MW(e)}$) than fast breeders and comparably low doubling time (~10 yr to double fissile inventory). Thus thermal breeders would require a smaller total investment in fissile material to reach the point at which the system can be sustained on bred material only. The economics of this aspect of thermal breeders is also analysed in the last section of this paper.

ECONOMICS OF FUEL UTILIZATION

The two nuclear characteristics of a reactor which are important from the standpoint of the economics of fuel utilization are (a) fuel inventory and (b) net fuel burn-up. The first of these, the fuel inventory, is the amount
of fuel in the reactor and associated fuel processing plants required to supply a given electrical capacity. This is usually expressed as kilograms of fissile material per megawatt of electrical capacity (kg/MW(e)) since in most reactor designs, costs associated with the inventory of fertile material are relatively unimportant. The second fuel utilization characteristic, net fuel burn-up, represents the net loss (or gain) of fissile material associated with the production of a given amount of electrical energy. Because this is such a small unit in terms of kilograms per megawatt-hour of electricity, it is convenient to express net burn-up in terms of kilograms fissile per megawatt-year of electricity (1 MW-yr = 8760 MWh = 8.76 × 10^8 kWh). The actual annual consumption or production of fissile material, of course, depends on the load factor of the reactor and the net burn-up figure must be corrected accordingly.

The two fuel utilization characteristics just described are important when comparing the economics of various future nuclear power systems because first of all, only these two factors will be influenced by changes in the cost or value of fissile material. Secondly, as nuclear power systems grow and the individual plants become very large, construction costs of various types of plants tend to approach one another. This means that capital costs will not greatly influence the choice of what kind of system to build. Finally, in large-scale fabrication and processing plants, i.e. plants handling 5-10 ton/d of nuclear fuels, unit fuel costs become very low and relatively insensitive to the type of fuel being handled. Thus, as a first approximation, an analysis of the economics of fuel utilization should provide a good indication of the probable long-term competitiveness of various nuclear power systems.

Before considering the fuel utilization of various systems, however, a few words of caution should be introduced. Although the fuel utilization characteristics of a reactor type may be a good measure of its long-term merit, reactor designers can vary these characteristics over a wide range to optimize their system for prevailing or anticipated economic conditions. Thus, any single measure of the inventory and net burn-up characteristics of a reactor type is not necessarily representative of that type optimized under other economic ground rules. The following analysis, therefore, is subject to this limitation.

FUEL UTILIZATION CHARACTERISTICS OF VARIOUS REACTOR TYPES

The fuel utilization characteristics of current-technology light-water reactors are compared with those of more advanced systems in Table I. These data may be converted into short tons of U\textsubscript{3}O\textsubscript{8} on the basis of 3.77 kg \textsuperscript{235}U extractable/short ton U\textsubscript{3}O\textsubscript{8}. Corresponding separative work requirements of course depend on the degree of enrichment of the fuel loading. In the case of the light-water reactors which use slightly enriched \textsuperscript{235}U, 110 kg separative work are required per kg fissile to supply the initial inventory and 180 kg separative work per kg fissile are required for \textsuperscript{235}U makeup. In the case of the remaining reactors which are fuelled with highly enriched \textsuperscript{235}U or recycled fissile material, 240 kg separative work per kg fissile are required.
Table I

FUEL UTILIZATION CHARACTERISTICS OF VARIOUS REACTOR TYPES

<table>
<thead>
<tr>
<th>Reactor type</th>
<th>Inventory (kg fissile/MW(e))</th>
<th>Net burn-up (kg fissile/MW-yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Current-technology light-water</td>
<td>3.8</td>
<td>0.38</td>
</tr>
<tr>
<td>High-temperature gas-cooled</td>
<td>3.2</td>
<td>0.11</td>
</tr>
<tr>
<td>Thorium-fuelled heavy-water</td>
<td>2.1</td>
<td>0.25</td>
</tr>
<tr>
<td>Seed blanket</td>
<td>4.8</td>
<td>0.073</td>
</tr>
<tr>
<td>Molten-salt converter</td>
<td>2.0</td>
<td>0.073</td>
</tr>
<tr>
<td>Westinghouse-type fast breeder</td>
<td>5.0</td>
<td>-0.31</td>
</tr>
<tr>
<td>Molten-salt thermal breeder</td>
<td>1.5</td>
<td>-0.10</td>
</tr>
</tbody>
</table>

FUEL REQUIREMENTS OF LIGHT-WATER REACTORS

Table II gives the amount of uranium required to support a nuclear industry based solely on light-water reactors with the fuel utilization characteristics given in Table I. The results are also based on the recent growth of the nuclear industry projected by the USAEC in December 1964 but rounded off so that the incremental additions to the system during any given five-year period follow a smooth curve. Also shown in Table II are the possible costs of the uranium based on (1) use of USA resources only or (2) USA plus Canadian resources, respectively, as shown in Table III. It is seen that with light-water reactors only, the price of natural uranium would rise to about $30/lb by about the year 2010. What increased uranium costs would do to the price of fully enriched $^{235}$U and to the corresponding cost of electrical power is shown in Table IV.

Applying the data in Tables II and IV to the cumulative nuclear power generation anticipated during the next 50 yr results in added power costs of many billions of dollars as shown in Table V.

The increased power costs shown in Table V are based on the assumption that separative work costs (i.e. the cost of $^{235}$U enrichment) will remain unchanged. The separative work capacity of the existing diffusion plant complex, however, is only about 20 million kg/yr according to unclassified estimates [Nucleonics, Sept. 1964]. On this basis, three new diffusion plants of about the same capacity as existing plants will have to be built by the year 2000 and at least one new plant every five years after that time. If this were done with private capital, increased costs of money might well cause the cost of separative work to increase which would make the incremental cost of power even higher than that shown in Table V.
<table>
<thead>
<tr>
<th>Year</th>
<th>Installed nuclear electricity capacity (Thousands of MW(e))</th>
<th>Thousands of short tons of U₃O₈ Requited</th>
<th>Required separative work capacity (Million kg/yr)</th>
<th>Cost of U₃O₈ ($/lb)</th>
<th>USA only</th>
<th>USA + Canada</th>
</tr>
</thead>
<tbody>
<tr>
<td>1970</td>
<td>7</td>
<td>7</td>
<td>2</td>
<td>8.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>1975</td>
<td>28</td>
<td>30</td>
<td>6</td>
<td>8.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>1980</td>
<td>75</td>
<td>90</td>
<td>12</td>
<td>8.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>1985</td>
<td>175</td>
<td>230</td>
<td>20</td>
<td>8.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>1990</td>
<td>320</td>
<td>460</td>
<td>34</td>
<td>10.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>1995</td>
<td>510</td>
<td>810</td>
<td>53</td>
<td>10.00</td>
<td>5.00</td>
<td></td>
</tr>
<tr>
<td>2000</td>
<td>745</td>
<td>1300</td>
<td>75</td>
<td>10.00</td>
<td>10.00</td>
<td></td>
</tr>
<tr>
<td>2005</td>
<td>1030</td>
<td>1960</td>
<td>100</td>
<td>30.00</td>
<td>20.00</td>
<td></td>
</tr>
<tr>
<td>2010</td>
<td>1360</td>
<td>2790</td>
<td>125</td>
<td>30.00</td>
<td>30.00</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td>1740</td>
<td>3830</td>
<td>144</td>
<td>30.00</td>
<td>30.00</td>
<td></td>
</tr>
<tr>
<td>2020</td>
<td>2165</td>
<td>5070</td>
<td>183</td>
<td>30.00</td>
<td>30.00</td>
<td></td>
</tr>
<tr>
<td>2025</td>
<td>2645</td>
<td>6550</td>
<td>200</td>
<td>30.00</td>
<td>30.00</td>
<td></td>
</tr>
</tbody>
</table>
TABLE III
RESOURCES OF URANIUM IN THE USA AND CANADA

<table>
<thead>
<tr>
<th>Price range per pound U\textsubscript{3}O\textsubscript{8} ($)</th>
<th>USA resources (thousands of short tons U\textsubscript{3}O\textsubscript{8})</th>
<th>Canadian resources (thousands of short tons U\textsubscript{3}O\textsubscript{8})</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 - 10</td>
<td>300 - 350</td>
<td>650</td>
</tr>
<tr>
<td>10 - 15</td>
<td>460</td>
<td></td>
</tr>
<tr>
<td>15 - 20</td>
<td>430</td>
<td></td>
</tr>
<tr>
<td>20 - 30</td>
<td>700</td>
<td></td>
</tr>
<tr>
<td>30 - 50</td>
<td>8000</td>
<td></td>
</tr>
</tbody>
</table>


TABLE IV
INFLUENCE OF INCREASED URANIUM COSTS ON THE PRICE OF FULLY ENRICHED \textsuperscript{235}U AND ELECTRIC POWER COSTS

<table>
<thead>
<tr>
<th>Cost of U\textsubscript{3}O\textsubscript{8} ($/lb)</th>
<th>\textsuperscript{235}U cost\textsuperscript{a} ($/g)</th>
<th>Incremental added power cost (milli/kWh)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>10</td>
<td>0.3</td>
</tr>
<tr>
<td>30</td>
<td>20</td>
<td>1.6</td>
</tr>
<tr>
<td>50</td>
<td>30</td>
<td>3.0</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Based on separative work costs of $22.50/kg.

REDUCTION IN URANIUM AND SEPARATIVE WORK REQUIREMENTS THROUGH THE INTRODUCTION OF FAST BREEDERS

It is obvious from the foregoing that because of the added cost of power, which would be experienced by light-water-type nuclear plants if low-cost sources of uranium become exhausted, there is a tremendous economic incentive to develop breeders to the point where they can provide needed fissile material. To really meet the fissile requirement problem, moreover, such
TABLE V
INCREMENTAL USA NUCLEAR POWER COST WITH LIGHT-WATER REACTORS ONLY

<table>
<thead>
<tr>
<th>Year</th>
<th>Cumulative power generation - 10^3 MW·yr</th>
<th>Added power cost (Billion $)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1970</td>
<td>16</td>
<td>-</td>
</tr>
<tr>
<td>1980</td>
<td>290</td>
<td>-</td>
</tr>
<tr>
<td>1990</td>
<td>1610</td>
<td>-</td>
</tr>
<tr>
<td>2000</td>
<td>5250</td>
<td>10</td>
</tr>
<tr>
<td>2010</td>
<td>12800</td>
<td>100</td>
</tr>
<tr>
<td>2020</td>
<td>25600</td>
<td>300</td>
</tr>
</tbody>
</table>

a Assuming use of USA plus Canadian resources.

breeders must produce excess fuel at a rate sufficient to supply fissile inventories for new plants being added to the system and also to make up for the fissile material consumed in non-breeders. Indications are that to do this job, breeders should produce new fuel at an annual rate of about 5 to 10% of their total fissile inventory (i.e., 20- to 10-yr inventory doubling times). Achievement of the lower doubling time, moreover, has the added attribute that if annual interest charges on fissile inventories are 10%, sale of bred fuel at its purchased price would just balance these charges. In other words, the value or price of fissile material would remain constant. It is apparent, therefore, that if breeders are inventoried with low-cost fissile material, they can sell bred fuel cheaply, whereas, if they are inventoried with high-cost fissile material they must get high prices for their bred fuel. Thus the long-term price of fissile material hinges on what it will cost to supply inventories for the breeders to the point where they become self-sustaining on bred fuel.

Using the projected nuclear power growth figures given in Table II and the performance characteristics of a Westinghouse-type fast breeder as a basis, the natural uranium required to support the industry was determined as a function of the time the breeders are introduced into the system. The results shown in Fig. 1 indicate that introduction of breeders of the given type by the year 2000 (assume one constructs only breeders after that time) would limit the total amount of uranium mined for its 235U content to about 3.6 million tons. But under such conditions, the price of fissile material would rise to about $20/g which is undesirable. Switching over to this type of breeder by 1980 would not alleviate the situation as the total amount of mined uranium would still be more than 2 million tons which exceeds the estimated low cost ($5-10/lb U₃O₈) resources by a factor of two. Introducing the fast breeders by 1980, however, does limit the uranium requirement to approximately the amount that might be available at low cost.
The corresponding separative work capacity to enrich the $^{235}$U that is mined is shown by Fig. 2. It is seen that only by introducing the given type of fast breeder as early as 1980 can one limit the needed capacity to that which might be satisfied by existing diffusion plants.

**NATURAL URANIUM-D$_2$O-FAST BREEDER COMPLEX**

An alternative to the early introduction of fast breeders has been suggested; namely, to build natural-uranium heavy-water reactors before breeders. This would not only save on the consumption of $^{235}$U but also provide the plutonium needed to inventory the fast breeders. Figure 3 shows the influence on ore requirements of replacing light-water reactors with natural-uranium heavy-water reactors during the period 1975 to 1990. Two
types of fast breeders are considered: (1) a 16-yr doubling time, 5 kg fissile per MW(e) design and (2) a 10-yr doubling time, 4 kg fissile per MW(e) breeder. It is seen in Fig. 3, that the introduction of heavy-water reactors saves about 400,000 short tons per short ton of mined U$_3$O$_8$; however, a more important saving (i.e. 700,000 short tons of U$_3$O$_8$) is achieved by improving the performance of the fast breeder. The combination of heavy-water reactors followed in 1990 by 10-yr-doubling-time fast breeders does, however, appear to limit the mined re-requirement to that which might be available at low cost.

INFLUENCE OF HIGH PERFORMANCE THERMAL BREEDERS ON ORE AND SEPARATIVE WORK REQUIREMENTS

A similar situation exists for developing a self-sustaining breeder industry based on high performance thermal breeders rather than fast breeders. In contrast to the high inventory-high breeding gain characteristics of fast breeders, however, thermal breeders have lower inventory-lower breeding gain characteristics but approximately the same doubling times. A switch from water reactors to 15-yr-doubling-time thermal breeders in 1990, however, would alleviate, but not solve, the fissile shortage problem. As shown in Fig. 4, the required amount of mined uranium would still be about 1.4 million tons. It is doubtful that such thermal breeders could be developed before 1990; therefore, earlier cases have not been considered.

ROLE OF ADVANCED THORIUM CONVERTERS

There is some indication that advanced thorium converters now being developed as part of the United States Civilian Nuclear Power Program may be a possible answer to the fissile shortage problem. Such reactors make considerably better use of mined $^{235}U$ than the light-water reactors and if introduced into the industry at an early enough date then can save on fissile
consumption to the point where the high cost $^{235}$U need never be used. Figure 4 illustrates this point for four advanced thorium converters; namely, thorium-fuelled heavy-water, large seed-blanket, high-temperature gas-cooled and molten-salt converter. It is seen that replacement of the water reactors with any one of these reactor types during the period 1975 to 1990 would limit the requirement for mined $^{235}$U to that available in low-cost ore. Thus, the role of advanced converters may be that of conserving low-cost $^{235}$U such that enough remains to inventory a self-sustaining breeder industry.

To achieve this role, however, the advanced thorium converters must be able to produce energy more cheaply than light-water reactors. This may seem a formidable barrier to their introduction into the industry. However, because of the more favorable fuel utilization characteristics of advanced thorium converters, they should be able to generate electricity around half a mill per kilowatt-hour cheaper than the present types of light-water reactors. If no serious technical problems are encountered in their development, advanced thorium converters offer the potential of being an economical solution to the $^{235}$U conservation problem.

ECONOMIC INCENTIVE FOR THE EARLY DEVELOPMENT AND INTRODUCTION OF BREEDERS AND ADVANCED CONVERTERS

By applying ore costs and separative work costs to the previously derived requirements it is now possible to determine the dollar requirement for mining and for $^{235}$U separation for the various approaches to a self-sustaining breeder industry. These costs can then be put on a present-worth basis to establish the present economic incentive for the early introduction of breeders and the earlier introduction of advanced converters.

Table VI gives the present worth of future savings in mining uranium and separating $^{235}$U for a system based solely on advanced converters followed by thermal breeders. Costs of uranium were taken as $20/kg and separative work as $25/kg. It is seen in this table that the present worth (calculated using a 5% discount factor) of introducing thermal breeders in 1990 compared to 2000 is $6.2 billion. Replacing water reactors with ad-
TABLE VI

ECONOMIC INCENTIVE FOR EARLY INTRODUCTION OF ADVANCED CONVERTERS AND THERMAL BREEDERS

<table>
<thead>
<tr>
<th>Year advanced converter introduced</th>
<th>Present worth of future savings (Billion $) for thermal breeders introduced by the year</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2000</td>
</tr>
<tr>
<td>base case</td>
<td>3.1</td>
</tr>
<tr>
<td>1990</td>
<td>3.3</td>
</tr>
<tr>
<td>1985</td>
<td>4.9</td>
</tr>
<tr>
<td>1980</td>
<td>6.3</td>
</tr>
</tbody>
</table>

Advanced converters in the period 1975 to 1990 would increase the present worth of future savings to $9.2 billion. It is apparent, on this basis, that a very sizable development programme is justified as a means of achieving these two goals.

IMPORANCE OF THORIUM RESOURCES

In considering the development of thorium-fuelled converters and breeders, a question that is often raised is "Are thorium resources adequate to support a large-scale industry based on the use of thorium?"

In answering this question it is important to point out that thorium does not in itself constitute a nuclear fuel but merely an indirect source of such fuel. Thus, thorium is not mined for its fissile content as is natural uranium and future requirements will be considerably lower. Existing designs of thorium converters and thorium breeders require between 100 and 200 kg of thorium per MW(e) for initial inventory of such reactors and an additional 50 kg/MW(e) for thorium consumed during a 30-yr plant life. If these data and the previously mentioned projected growth of the USA nuclear industry are used as a basis, and if it is assumed that all reactors in the industry are thorium-fuelled, the future thorium requirements might be as much as shown in Table VII.

These projected requirements may be compared with estimated USA resources of thorium shown in Table VIII.

It is seen that the low-cost domestic reserves of thorium would not be exhausted before the year 2010 even though the USA nuclear power system consisted solely of thorium-based reactors. The use of higher-cost thorium, moreover, would not greatly influence the economics of thorium reactors.

Since world reserves of low-cost thorium ore are many times greater than US resources, it is unlikely that any pressing demand for higher-cost material will be encountered in other parts of the world during the next half century.
### TABLE VII

**PROJECTED GROWTH OF NUCLEAR INDUSTRY AND CORRESPONDING THORIUM REQUIREMENTS IN THE USA**

<table>
<thead>
<tr>
<th>Year ending</th>
<th>MW(e) x 10^6</th>
<th>ThO₂ required (thousands of short tons)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1980</td>
<td>75</td>
<td>20</td>
</tr>
<tr>
<td>1990</td>
<td>335</td>
<td>88</td>
</tr>
<tr>
<td>2000</td>
<td>730</td>
<td>194</td>
</tr>
<tr>
<td>2010</td>
<td>1380</td>
<td>375</td>
</tr>
<tr>
<td>2020</td>
<td>2200</td>
<td>660</td>
</tr>
</tbody>
</table>

### TABLE VIII

**USA THORIUM RESOURCES - 1981**

(Thousand short tons ThO₂)

<table>
<thead>
<tr>
<th>Price range ($) per pound ThO₂</th>
<th>Reasonably assured resources</th>
<th>Possible additional resources</th>
<th>Total resources</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 - 10</td>
<td>100</td>
<td>300\textsuperscript{a}</td>
<td>400</td>
</tr>
<tr>
<td>10 - 30</td>
<td>100</td>
<td>100\textsuperscript{a}</td>
<td>200</td>
</tr>
<tr>
<td>30 - 50</td>
<td>3 000</td>
<td>7 000\textsuperscript{b}</td>
<td>10 000</td>
</tr>
<tr>
<td>50 - 100</td>
<td>8 000</td>
<td>17 000\textsuperscript{b}</td>
<td>25 000</td>
</tr>
<tr>
<td>100 - 500</td>
<td>1 000 000</td>
<td>2 000 000\textsuperscript{b}</td>
<td>3 000 000</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Depends on new discoveries which may yield smaller or larger reserves than indicated.

\textsuperscript{b} Extensions of known uraniferous (thoraniferous) formations.
INTRODUCTION

Various reactor systems that would use the thorium-uranium-233 cycle are under development because of their potential for achieving low power costs and high fuel utilization. The achievement of low power costs depends to a great extent upon the economics, and consequently the technology, involved in the recycle of fuel from such reactors. Technical development is required for shipping, chemical processing, fuel preparation, and refabrication of bred fuel. Since the cost of fabricating fuel is a major contributor to the cost of any fuel cycle, it is essential that we consider the technology and economics of refabricating fuel elements of various reactor systems.

There is little experience that can be used in the solution of the technical problems which exist in the refabrication of fuel. An assessment, at the present time, of the refabrication technology for the Th-U cycle must be considered very preliminary for several reasons. Current fuel element designs are based principally on processes optimized for initial and, therefore, contact fabrication. This could lead to erroneous conclusions since heavy penalties might be applied to fuel element costs because certain contact steps used conventionally become extremely awkward and expensive when they are automated and performed under conditions imposed by recycle fuel. On the other hand, if we assume a design and a fabrication technique to be suitable for refabrication, we may err in estimating the extent to which a process can be extrapolated from meagre data to a large-scale refabrication plant. There is little experience with recycle fuel and none with certain types of fuel elements. There is no relevant experience with large plant processing or the economies to be realized in large plants fabricating fuel elements for power reactors.

Because of the uncertainties that currently shadow the technology and economics of refabricating fuel, it is necessary that development programmes be carried out to gain an understanding of the technology and to seek methods for eventual economical utilization of recycle fuel in power reactor systems. This necessity has led various investigators [1-8] and ourselves to programmes for detailed evaluation of the technical and economic feasibility of refabrication. In this paper, we present a number of factors which must be considered if one is to refabricate bred fuel and suggest methods of attacking problems encountered in refabrication technology. We discuss our plans for pilot-scale demonstration of the refabrication of two types of fuel elements. The paper concludes with an analysis of the effect of production rate and isotopic content of fuel on refabrication cost.

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation.
PROBLEMS IN FUEL REFABRICATION

Refabrication economics are influenced by a number of major factors which are readily recognized. Among those factors are the fuel element design, the process used for fabrication and the isotopic content of the fuel; the refabrication plant and equipment design greatly affect fabrication costs.

Effect of fuel isotopic content and recycle scheme on fabrication plant design

For any proposed fabrication plant, one has the problem of selecting for the given isotope the type or mode of fabrication that is to be employed in the fabrication of either first cycle or recycle fuel. The possibilities are contact, hooded, glove box, semi-remote, or remote operation. We have chosen to define these terms as follows: (1) Contact operations are those in which the operator has direct contact with the material. (2) Hooded operations are those which are contained in ventilated enclosures that are not hermetically sealed. (3) Glove box operations are those requiring hermetic sealing of the equipment. (4) Semi-remote operations are those requiring light shielding. (5) Remote operations are those requiring heavy shielding and totally remote operation. The type and quantity of the isotope in the fuel and the quantity of the fuel being processed dictate the type of operation to be selected.

Several recycle systems can often be used. For example, in the High-Temperature Gas-Cooled Reactor (HTGR) one may choose not to recycle the thorium, but to recycle only the $^{233}\text{U}$. With this particular fuel element design, one then has the choice of using either homogeneous (U-Th)C$_2$ particles or discrete particles of UC$_2$ and ThC$_2$. With (U-Th)C$_2$ particles, all of the material must be handled in the refabrication plant, but when the discrete particles are used the particles containing virgin thorium can be prepared in a hooded operation. The other alternative is to recycle the thorium and the $^{233}\text{U}$. Similar possibilities arise in other fuel element designs where it is feasible to mechanically separate $^{233}\text{U}$ and thorium during some of the fabrication steps.

In the fuel cycle of interest, Th-$^{233}\text{U}$, the mode of fabrication depends upon the $^{232}\text{U}$ concentration of the fuel, its age, and whether virgin or recycle thorium, which contains $^{228}\text{Th}$, is being used. The amount of fuel processed, its concentration, and its proximity to the operator is also important. We have analysed the effect of $^{232}\text{U}$ concentration on the type of facility which should be employed in the fabrication of HTGR fuel and Spectral Shift Control Reactor (SSCR) fuel.

In the analysis for the SSCR fuel, we used the flow sheet shown in Fig. 1. The SSCR fuel element has approximately 200 fuel rods containing (U-Th)O$_2$ in an 8-in. square array. Each fuel rod is approximately 0.4-in., in diameter and clad with 0.025-in., wall Zircaloy-2 tubing. The fuel element is designed for mechanical assembly. The basic structure is an open cage assembly into which the fuel rods are simply inserted; no welding or brazing of the structure after loading of fuel rods is required. To perform the shielding calculations and determine the mode of fabrication, the fuel element fabrication facility was divided into three zones.
Zone 1. Bulk oxide is received from the fuel reconstitution facility and prepared for loading in the fuel rods.
Zone 2. Fuel rods are loaded, end-capped and inspected.
Zone 3. Fuel rods are autoclaved, placed into the pre-assembled fuel element framework and inspected.

Typical material flow rates for fabrication of the SSCR fuel element are shown in Table I. It is notable that material hold-up in Zone 3 is high because of the necessity of storing fuel rods containing different enrichments for loading into a single fuel element. We used the fuel element design shown
### TABLE I

SSCR MATERIAL LOCATION, QUANTITY, AND AGE IN FABRICATION PLANT

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>Zone 2</th>
<th>Zone 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>20</td>
<td>64</td>
</tr>
<tr>
<td>(2) Time since material received from sol-gel plant (h)</td>
<td>3</td>
<td>8</td>
</tr>
<tr>
<td>(3) Material held up in equipment (kg)</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>5</td>
<td>5</td>
</tr>
</tbody>
</table>

Plant capacity (kg heavy metal/d) of 60, 230, 930, 3700
in Fig. 2 in the analysis for the HTGR fuel element. The element has a diameter of 4\frac{1}{2} in. and has ten equally spaced 3/4-in.-diam. holes in the cross-section between the outside diameter and the 3/4-in. inside diameter. These holes are filled with fuel bodies consisting of an aggregate of coated fuel particles held together by a graphite matrix. The fuel element is 20 ft in overall length and has two identical sub-assemblies, each having an active fuel length of 7\frac{1}{2} ft. The fuel assemblies can be fabricated separately, can be attached to either a reflector assembly, or a fission-product trap assembly, and can be joined by the central coupling to form a complete fuel element. The flowsheet used is shown in Fig. 3. We considered both discrete particles of UC₂ and ThC₂ and particles of (U-Th)C₂. For fabrication of the HTGR fuel element, the facility was divided into three zones.

Zone 1. The oxide microspheres are received from the fuel reconstitution facility and converted to carbide and inspected.

Zone 2. The fuel particles are coated with pyrolytic carbon and inspected.

Zone 3. Compacts are prepared, inspected, and loaded into the graphite sleeves. Finally, two fuel segments are assembled together with end pieces, inspected, and shipped to the reactor site.

Typical material flow rates for (U-Th)C₂ particles are shown in Table II; those for UC₂ particles in Table III.

The following assumptions were made in calculating the shielding requirements for all of the plants.

1. The time between solvent extraction and receipt of material at the fuel element fabrication plant is five days.

2. A major clean-up of the equipment and enclosures is performed after five working days.

3. No substantial quantity of the material is located closer than 1 ft to the enclosure wall.

4. The amount of material retained in the equipment (on the walls, in crevices, etc.) during processing is 3 kg, but the material is released and continued in the process at five-day clean-up intervals.

5. The plant processes 110\% of the quantity shipped to allow for internal rejection of product not meeting specifications.

Shielding was calculated for plants having daily processing capacities ranging from 10 to 3700 kg of heavy metal per day. Typical results of these calculations are summarized in Table IV. Unless otherwise indicated, the results are for combinations of virgin thorium and ²³³U. As can be observed in the table, we have used approximately 3.5 in. of steel as a practical limit for semi-remote fabrication because of the difficulty of working through a greater distance with glove hands or tongs. Also, radiation from sources requiring greater than 3.5 in. of steel would prohibit, or at least greatly inhibit, contact maintenance of equipment. In all of the results, the shielding is calculated to limit body exposure to 1 mR/h. The data in Table IV show the limit of semi-remote fabrication for SSCR fuel to be approximately 5 ppm ²³²U in heavy metal, that for HTGR fuel containing (U-Th)C₂ particles 2 to 5 ppm depending upon the plant capacity, and that for discrete particles of UC₂ + ThC₂ only 1 ppm. It is notable that the discrete particles of UC₂ required greater shielding than (U-Th)C₂ particles because they are not diluted with thorium during the conversion and coating operations.
In fuel cycles where all the fuel is recycled, it is probable that the concentration of $^{232}\text{U}$ will exceed 5 ppm in heavy metal. We therefore conclude from our analyses that a remote plant is required for refabrication of such fuel.

This conclusion raises two serious questions: (1) How does one approach a satisfactory technical and economical solution to the problem of remotely fabricating fuel elements? (2) What are the cost penalties that one could expect for fabrication under such rigorous conditions? To answer these questions, it is appropriate that we consider the requirements and features of a remote fuel refabrication plant operating at a significant production rate.

Requirements and features of the remote fuel element production plant

The plant should incorporate automatic processing equipment insofar as practicable for several reasons. (1) The rate of unit processing has reached a value at which it is economical to invest in capital plant and minimize operating manpower expense. (2) The requirement for remote processing makes manual control of processing equipment extremely difficult. (3) Reproducibility and quality control are better assured by automatic systems.

The plant must provide substantial shielding from gamma irradiation depending on the isotopes being fabricated. The actual amount of shielding is not an important point since, if it is assumed that the process will proceed automatically, manipulation will not be used for normal operations.

The plant must offer suitable containment for the isotopes being fabricated. Generally, fuel refabrication operations can be divided into two categories according to the degree of contamination of the environment which can be expected from them: (1) contaminated fabrication that includes operations in which the fuel material is not completely contained in cladding; and (2) clean fabrication that includes operations in which the material is contained. These two categories of operation require two different types of shielded work areas. The work area for contaminated fabrication must be hermetically sealed and ventilated for control of particulate contamination. The probability of spreading particulate contamination during clean fabrication operations is minimal; therefore, adequate ventilation for cooling of the equipment and for a contingent contamination incident must be provided, but no other precautions are necessary.

These categories of refabrication allow two general methods of equipment maintenance. The equipment used in contaminated fabrication would require completely remote maintenance, unless the work area could be decontaminated to an extent that protected personnel could enter the area. The most likely possibility for repair of equipment components is that the components will be decontaminated and removed to a gloved maintenance area. Personnel can enter the cell and directly maintain equipment in a clean fabrication cell after removal of gamma-radiation sources.

Need for simplicity in processing equipment

It is desirable that all processes for fabrication of nuclear fuels be simple. In the case of remote fabrication of fuel elements, simple processes of equipment are not only desired, but mandatory, for several reasons.
FIG. 2. Reference design for an HTGR fuel element for remote fabrication
FIG. 3. Flowsheet for fabrication of HTGR fuel elements
TABLE II
MATERIAL LOCATION, QUANTITY, AND AGE IN FABRICATION PLANT PROCESSING \((U-\text{Th})\)C\(_2\)

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>Plant capacity (kg heavy metal/d) of 60  230  930  3700</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>(1) In-process material at given time (kg)</td>
<td>22  88  352  1408</td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (h)</td>
<td>16  16  16  16</td>
</tr>
<tr>
<td>(3) Material held up in equipment (kg)</td>
<td>3  3  3  3</td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant, (d)</td>
<td>5.7  5.7  5.7  5.7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zone 2</th>
<th>Plant capacity (kg heavy metal/d) of 60  230  930  3700</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>20  80  320  1280</td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (h)</td>
<td>26  26  26  26</td>
</tr>
<tr>
<td>(3) Material held up in equipment (kg)</td>
<td>3  3  3  3</td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>6  6  6  6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zone 3</th>
<th>Plant capacity (kg heavy metal/d) of 60  230  930  3700</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>15  30  60  60</td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (d)</td>
<td>1.7  1.3  1.3  1.2</td>
</tr>
<tr>
<td>(3) Hold-up material in any element (kg)</td>
<td>3  3  3  3</td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>6.7  6.4  6.3  6.3</td>
</tr>
</tbody>
</table>
# TABLE III

<table>
<thead>
<tr>
<th>Zone 1 (UC\textsubscript{2} only)</th>
<th>Plant capacity (kg heavy metal/d) of</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>3</td>
<td>4</td>
<td>16</td>
<td>64</td>
<td></td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (h)</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td></td>
</tr>
<tr>
<td>(3) Material held up in equipment (kg)</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>5.7</td>
<td>5.7</td>
<td>5.7</td>
<td>5.7</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zone 2 (UC\textsubscript{2} only)</th>
<th>Plant capacity (kg heavy metal/d) of</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>3</td>
<td>6</td>
<td>24</td>
<td>96</td>
<td></td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (h)</td>
<td>26</td>
<td>26</td>
<td>26</td>
<td>26</td>
<td></td>
</tr>
<tr>
<td>(3) Material held up in equipment (kg)</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zone 3 (UC\textsubscript{2} + ThC\textsubscript{2})</th>
<th>Plant capacity (kg heavy metal/d) of</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) In-process material at a given time (kg)</td>
<td>15</td>
<td>30</td>
<td>60</td>
<td>60</td>
<td></td>
</tr>
<tr>
<td>(2) Time since in-process material received from sol-gel plant (d)</td>
<td>1.7</td>
<td>1.3</td>
<td>1.3</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>(3) Hold-up material in any element (kg)</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>(4) Time since hold-up material received from sol-gel plant (d)</td>
<td>6.7</td>
<td>6.4</td>
<td>6.3</td>
<td>6.2</td>
<td></td>
</tr>
</tbody>
</table>
### TABLE IV

<table>
<thead>
<tr>
<th>$^{233}$U $^a$</th>
<th>SSCR Fuel</th>
<th>HTGR Fuel - (U-Th)$_2$ particles $^b$</th>
<th>HTGR Fuel - UC$_2$ + ThC$_2$ particles $^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(ppm)</td>
<td>230</td>
<td>930</td>
<td>3700</td>
</tr>
</tbody>
</table>

#### Semi-remote fabrication (in. steel)

| 1  | 0.8 | 1.2 | 1.4 | 1.2 | 1.9 | 2.0 | 2.8 | 2.8 | 3.2 |
| 2  | 1.7 | 2.1 | 2.3 | 2.0 | 2.7 | 2.9 |     |     |     |
| 5  | 2.8 | 3.3 | 3.5 | 3.1 |     |     |     |     |     |

#### Remote fabrication (in. concrete)

| 2  | 12.2 | 13.8 | 14.8 | 13.2 | 15.6 | 17.0 | 15.9 | 15.9 | 17.2 |
| 5  | 15.0 | 16.7 | 17.8 | 16.0 | 18.5 | 20.0 | 18.7 | 18.7 | 20.0 |
| 10 | 18.9 | 20.6 | 21.8 | 19.6 | 22.2 | 24.1 | 21.4 | 21.4 | 22.8 |
| 20 | 21.7 | 23.5 | 24.8 | 22.4 | 25.0 | 27.2 | 25.1 | 25.1 | 26.5 |
| 50 | 28.4 | 30.3 | 31.8 | 28.8 | 31.6 | 34.3 | 34.3 | 34.3 | 35.8 |
| 100| 31.2 | 33.9 | 34.8 | 31.6 | 34.4 | 37.4 | 37.0 | 37.0 | 38.6 |
| U-Th $^d$ | 36.0 | 38.5 | 40.0 | 36.0 | 39.0 | 42.5 | 41.5 | 41.5 | 43.0 |

---

$a$ Parts per million in heavy metal (U + Th).

$b$ Each particle containing a mixture of $^{233}$U and virgin thorium.

$c$ $^{233}$UC$_2$ process alone in parts of the process.

$d$ $^{233}$U-recycle thorium-assumptions.
(1) The complexity of any operation tends to be magnified when that operation is done remotely. (2) The performance of fabrication operations, many of which are performed by hand in conventional nuclear facilities, are inherently difficult in remote facilities. (3) Repair of equipment in remote facilities is extremely difficult.

Requirements for simplicity in fuel element design

The design of fuel elements should be such that (1) the number of remote operations required is an absolute minimum and (2) the operations required are reasonably straightforward and easily accomplished. Also, fuel element designs should be optimized for minimum fuel fabrication costs. For example, the reduction of the number of pieces of hardware which must be handled per kilogram of fuel usually effects economies in fuel fabrication operations.

SELECTION OF SIMPLIFIED PROCESSES AND OPTIMUM FUEL ELEMENT DESIGNS

Now that certain requirements have been set forth for refabrication plants, we can discuss some approaches to the objectives of simplification and minimal cost.

Importance of fuel element design

The fuel element design will, of course, determine the process and the degree of inspection which is required for the fuel elements. The designer must carefully consider the need for certain specifications as well as the geometry of the fuel element. Two fuel elements [9] as examples will illustrate this point; the SSCR fuel element designed by the Babcock and Wilcox Company and the advanced HTGR element by General Atomic Division of the General Dynamics Corporation.

The SSCR fuel element was designed for vibratory compaction and for mechanical assembly of the fuel rods into a fuel bundle. The fuel element assembly scheme consists of pushing loaded fuel rods into a pre-positioned and assembled lattice which has been constructed outside the fuel fabrication plant. This is a significant improvement over the operations involved in the manufacture of a brazed fuel element assembly.

In any rod-bundle design, there are a number of parameters which greatly affect cost; such as, type of fuel material, fuel diameter, fuel length, fuel density, type cladding, clad thickness, and number of fuel rods in an assembly [10]. Figure 4 illustrates the results of independent variation of some of these parameters over an extensive range. These curves relate to an advanced PWR-type fuel element of rod-bundle design fuelled with low-enrichment UO₂ pellets. All of these effects are due to the number of pieces which must be handled per kilogram of fuel during fabrication. Obviously, the fuel element designer can, by judicious selection of geometry and dimensions consistent with core thermal performance requirements, minimize costs.
One concept [11] for an advanced HTGR fuel element is that of loose particles contained in a graphite body. This, of course, considerably shortens the process in that the fabrication of graphite bodies containing dispersed fuel particles is eliminated. The fuel particle spheres in the HTGR element are coated with pyrolytic carbon. As shown in Fig. 5, costs are affected substantially by the particle diameter and the coating thickness which is selected for a particular fuel element [12]. Over the range shown in the figure, the cost of fabrication increases with increasing particle size and initial particle diameter. In the process, a fluidized bed is used conventionally for coating the fuel particles. In such beds, for larger particles
a much increased gas velocity is required to fluidize the particles; there is poor gas-solid contact; the particle motion characteristics change; and there might be increased maintenance from sooting. This analysis could be negated by the development of more efficient devices for coating larger particles. The explanation for the effect of coating thickness is straightforward; one simply has to apply more carbon, but at a specified rate of deposition.

Another economy would be the use of coated-oxide microspheres instead of carbide. Other factors, such as the amount of fuel per fuel element, the number of holes per fuel element, and the length of the fuel element, would also have a bearing on fabrication costs.

Simplified processes

Process steps selected should be short, easy to operate and control, and they should have a high degree of utilization of the material put into the process. That is, there should be little recycle of material within the process. We shall use two examples to illustrate several points.

Process for metal-clad oxide fuel elements

First, in fabricating metal-clad oxide fuel bundles there are several basic methods of consolidating fuel and introducing it to fuel tubes; these are pelletizing, extrusion, swaging, and vibratory compaction. From the standpoint of remote operation, pelletizing operations have two drawbacks. (1) Although pelletizing has been used extensively, centreless grinding of pellets is still required to meet the usual dimensional tolerances. (2) High-temperature sintering operations are required for densification of pellets. The material from the grinding operation creates a substantial waste and recycle problem. The pelletizing step does, however, have the advantage of mature technology and of adaptability to mass production. Extrusion techniques, although not fully developed, have similar drawbacks, that is, a necessity for high-temperature sintering and for grinding. The process is, however, adaptable to higher production rates. We should also note that both pelletizing and extrusion require that the material be handled in several operations before the formation of green fuel bodies. Swaging has been extensively investigated as a method for consolidating powders but has not yet seen large-scale use because of difficulties in determining whether cladding quality has been affected by the working that occurs during swaging. Even without this effect, swaging has not proved to be substantially cheaper than pelletizing operations.

Vibratory compaction is a method which has been under extensive investigation during the past few years. At ORNL, we have produced 1100 fuel rods in a pilot demonstration of the fabrication of $^{233}\text{U}-\text{Th}$ oxide-bearing fuel rods [13-18]. Fuel rods were fabricated at a rate of 10 to 15 per day using the bulk oxide-vibratory compaction route. Approximately 900 of the rods, 1/2-in. in diameter and 46-in. in length, and 200 shorter, 18-in. long, but otherwise identical, rods were fabricated in a semi-remote, lightly shielded (4.5-in. steel) facility, known as the Kilorod Facility. During the production,
an average density of 90% of theoretical was obtained using Th-3 wt.% $^{233}$U oxide. The vibratory compaction process used has the advantage of utilizing almost 100% of the material charged to the process.

It was specified that (1) each rod have a fuel density of ± 2% of the average density for the entire shipment and (2) that the density within a fuel rod at any position be within ±2% of the average for that fuel rod. To meet this specification, a mixture of coarse and fine powders consisting of 55% -6 +16 mesh and 45% unclassified fines was produced. The density variation along the length of the fuel rods was determined by a gamma-scanning operation using a 1/8-in. X 3/8-in. gamma beam, which was done immediately after vibratory compaction. During the early operations in the facility, the recycle rate between the compaction and gamma-scanning steps was quite high; nearly 20%. In the later campaigns, the recycle between the two steps because of insufficient homogeneity reached a low of less than 1%.

There were, however, mechanical difficulties with the pneumatic vibratory compactor. It required more maintenance than any other item of equipment in the process line, largely from the breakage of components of the compaction machine. The vibratory compactor was the rate controlling machine in the process; the performance of the complete step for a single rod required approximately 17 min [18]. Unless techniques are developed for simultaneous vibration of a number of fuel rods, one faces the proposition of having a large number of vibratory compactors in a fuel fabrication plant of large capacity. Because the sol-gel process produces dense oxide suitable for vibratory compaction very cheaply and because the difficulties with the vibratory compactor can probably be overcome, we believe it to be the most economical method of consolidating material in oxide metal-clad fuel bundles under remote conditions.

It should be noted that we had little difficulty in the Kilorod Facility with the remainder of the process steps after initial operation.

**Fuelled-graphite process**

Two steps in the process require extensive consideration if the costs of fabricating fuelled-graphite elements are to be reduced. One is the technology for making spherical particles of either oxide or carbide. The second is the process for pyrolytic-carbon coating of these microspheres. Heretofore, the technology for making spherical particles has consisted of various mechanical methods of consolidating combinations of the solid materials $\text{UO}_2$, $\text{ThO}_2$, and carbon and then treating these at high temperature to complete the necessary reactions and to densify the material. These methods invariably involve tedious techniques which become more complicated when special operating conditions are imposed by recycle fuel. Therefore, an objective of ORNL has been to simplify the manufacture of spherical particles.

We believe the sol-gel process [19] meets this objective. Clinton [20] at ORNL has demonstrated on a pilot scale the preparation of thorium-uranium microspheres which can be used as oxide or converted to carbide. Following the flow-sheet shown in Fig. 6, the microspheres are prepared by dispersing the sol at room temperature in an immiscible organic liquid
which has some solubility for water. The sol droplets must be suspended in the organic phase until enough water is extracted from the aqueous sol to cause gelation. After drying, the gel microspheres are calcined at 1150°C to complete the preparation of dense (99.4% of theoretical is typical) oxide microspheres. Microspheres produced by this method can be directly converted to carbide \((U\text{-Th})C_2\) at 2150°C [21].

The coating operation accounts for 25% of the cost of fuel fabrication in a plant capacity of 60 kg/d and 38% in plants at 3700 kg/d [12]. Therefore, to reduce costs, one should strive to increase the efficiency of the coating step. At ORNL, we are investigating methods of improving the efficiency of coating apparatus. These studies involve theoretical calculations of process conditions required in fluidized beds and rotary-drum coaters, as well as model studies on such devices [22, 23].

We are now preparing a pilot-scale facility for demonstration of improved methods of spherical particle preparation, conversion of oxide particles to carbide, and of coating with pyrolytic carbon. Most of this work is directed toward the scale-up of these processes. In the pilot facility, we shall accumulate data that we hope will lead to efficient remote fabrication processes and equipment. The facility is to process non-recycle material.

**OAK RIDGE NATIONAL LABORATORY PILOT-SCALE DEMONSTRATION**

The absence of accurate data on costs and information on the technology for remote fabrication of fuel has led us to the construction of a facility in which we shall attempt the development of practical and economic solutions to refabrication problems. The facility is known as the Thorium-Uranium Recycle Facility (TURF) [24, 25].

**Thorium-Uranium Recycle Facility**

The TURF will furnish the necessary space and shielding to perform all of the operations required for the processing of a spent-fuel element through the various phases of the fuel cycle. The facility has been sized to accom-
modate integrated recycle processes with equipment scaled down from production plant sizes so that realistic and reliable bases for technical and economical analyses will exist. The design of the facility is such that two fabrication processes can be accommodated in the facility at one time.

The facility will be an irregularly shaped three-story building approximately 162 ft long, 124 ft wide, with a partial basement. The first floor, shown in Fig. 7, provides space for offices, change rooms, operating areas around the cell enclosures, a fuel storage basin, and a receiving area. The second floor provides space for chemical make-up, sampling of in-cell processes, cask decontamination, a shop for contaminated equipment, a development laboratory, and the mechanical and electrical equipment for the building. The third floor is a high bay area which includes the cell roof area and provides facilities for entry of cell services and cell access. The bay is provided with a 50-ton crane to handle casks large enough to accommodate fuel elements up to 12 ft in length. The basement will provide space for access to the equipment storage cell and for the vulnerable equipment associated with the chemical cell. The building acts as a second line of containment for the cell complex.

The primary zone of containment for the facility consists of six shielded cells and associated glove maintenance room and air lock, all of which are depicted in Fig. 8. Four of the cells provide the operating space for the process equipment while two provide supporting functions. The mechanical processing and chemical processing cells will be used for operations incidental to irradiated fuel recovery and reconstitution of fissile and fertile materials into forms suitable for use in fuel element manufacture. The contaminated fabrication cell provides space for fabrication operations through the point where all fissile and fertile material is contained and sealed in fuel cladding. The clean fabrication cell will be used for final assembly and inspection of fuel elements. The two large cells, mechanical processing and contaminated fabrication, are to be maintained remotely; the clean fabrication cell will be maintained by a contact means. The chemical cell has the flexibility of allowing either method of maintenance.

All of the remote maintenance cells and the decontamination cell are lined with stainless steel. The service penetrations for all cells are provided with seals to confine radioactivity. Provisions have been made for future conversion to an inert atmosphere in the remote maintenance cells, the decontamination cell, equipment storage cell, and the glove maintenance room to permit processing of pyrophoric materials on a large scale.

The cells are capable of processing and refabricating fuel assemblies as long as 12 ft and containing as much as 35 kg of Th-U fuel irradiated to 25 000 MWD/t and decayed for 90 d. All of the operating cells are provided with the equivalent of 5½ ft of normal concrete up to the electromechanical manipulator bridge level and 4½ ft of concrete above this level.

**Process and equipment design for TURF**

We are now designing and constructing equipment for demonstration of remote fabrication processes in the TURF. The two processes involved are those for fabrication of metal-clad oxide fuel elements and fuelled-graphite
elements. Before discussing these two processes in detail and the equipment which we have designed, it would be appropriate to consider some of the criteria on which the process and equipment design is predicated.

Criteria for process and equipment design

All of the general considerations which have been previously discussed were taken into account in the selection of processes and equipment designs. Therefore, we have tried to select a simple fuel element design, to select a process that is short as possible, and to reduce to a minimum the number of operations that must be done remotely. In addition to these general considerations, there are a number of details that should be mentioned.

The layout of the fabrication equipment was divided into contaminated and clean fabrication zones according to the degree of contamination that is expected from the operations. These assignments greatly influence the type of equipment which should be designed for operation in the particular area. For example, each item of equipment located in the contaminated fabrication cell must be capable of being remotely installed, maintained, and removed by the cell remote handling system. Therefore, motor switches and complex mechanical components which are susceptible to wear, damage, or failure must be replaceable with a minimum of effort and without removal of the complete equipment item. Contact maintenance can be employed in the clean fabrication cell; therefore, provisions for maintenance by manipulators in the cell need not be extensive.

In the design of the equipment, we have used mild steel or aluminium extensively. Stainless steel has been used only for parts in direct contact with the fuel or where corrosive liquids or atmospheres dictate its use. Elastomers and organic materials have been used only where absolutely necessary; and, when used, provisions have been made for their easy replacement.

One of the principal problems involved in remote fabrication is that of transfer of materials between operational steps in the process scheme. At the onset of design, it was recognized that special handling systems would be required to precisely locate and to quickly transfer the article being fabricated from one operation to the next. Our analysis showed that if the general manipulation provided in the cell complex were used for transfer, the percent utilization of the process equipment would be extremely low; that is, the principal bottleneck in the process would be the general purpose manipulators. Therefore, we concluded that transfer of bulk solid material from one step to the next should be done by gravity flow through pipes or by enclosed powder conveyors and that transfer of fuel rods or other large components between operational steps should be accomplished with special transfer machines.

We can best illustrate our approaches to these problems by discussing the processes that we have designed and the concepts of the equipment for these processes [26, 27].
FIG. 7. Oak Ridge National Laboratory thorium-uranium recycle facility, first floor
FIG. 8. Oak Ridge National Laboratory thorium-uranium recycle facility, hot cells

Fabrication of oxide-bearing metal-clad fuel bundles

One objective of the TURF is to demonstrate the feasibility of remote fabrication and assembly of metal-clad oxide fuel elements. The equipment is to be capable of accommodating fuel rods from 2 to 10 ft in length and 1/4 to 3/4 in. in diameter. The flow sheet for the oxide fabrication process is presented in Fig. 9; the layout of the equipment in Fig. 10. As can be seen in Fig. 10, fabrication and inspection equipment will be constructed in the form of a multi-station system. Transfer of rods in the multi-station system will be accomplished with two special fuel rod transfer machines. Versatility to accommodate the wide dimensional variation of the fuel rods will be achieved when possible by minor adjustment of components. However, in most cases, the cost of constructing versatile units has dictated that partial retooling or complete interchange of certain equipment items be done.

The design of equipment for powder conditioning before vibratory compaction has been relatively straightforward. In the Kilorod Facility, there was little mechanical difficulty with this equipment. Therefore, we have simply extended those designs to remote conditions by adding appropriate control systems and conveyors.
MATERIAL FROM REJECTED FUEL RODS

FIG. 9. Flowsheet for fabrication of metal-clad oxide fuel bundles in thorium-uranium recycle facility.
Figure 10. Schematic layout of equipment for fabrication of oxide fuel bundles in thorium-uranium recycle facility.
Particular attention was given to the vibratory compaction step. In the Kilorod Facility, the principal problem was the frequent maintenance required by the equipment. Also, as has been previously noted, the equipment had a low rate of unit processing which could give scale-up problems. Therefore, we attempted to design components in which stress risers were eliminated; and we attempted to design a unit which would vibrate two fuel rods simultaneously.

Another piece of equipment which has given particular difficulty in design is the gamma scanner. It is required to detect a 2% density variation over a 3/8-in. interval of rod length. The scanner consists of a collimated gamma beam, a NaI detection crystal, and a means of moving the beam along the length of the rod. If either a cobalt-60 source or a caesium-137 source is used, the scanning speed is very slow; therefore, it was necessary to evolve a concept for multiple rod scanning.

The design of the equipment for all of the other operations has been relatively straightforward, and we envision no problem in scaling these to higher production rates. Most of the problems have been the ones which usually accompany the design of such specialized equipment. Little attention has been given to the problem of fuel element assembly because of the lack of a firm fuel element design for fabrication. The assembly equipment would, of course, be greatly dependent upon the particular geometry and configuration of the fuel element to be assembled.

Fuelled-graphite equipment

We are now conceptually designing equipment to be used for demonstration of refabrication technology for the fuelled-graphite element depicted in Fig. 2. The flow sheet that is being used for the conceptual design is shown in Fig. 11. The equipment is to be a production type so that information gained will be applicable both technically and economically to a full-scale production facility. The equipment is to be sized for the production of approximately 35 kg of heavy metal per day. The principal barrier to this goal as far as capacity is concerned is the coating process. We do not yet have a solution to the relatively low processing rate which is inherent to the step.

A preliminary layout of the fabrication equipment for the fuelled-graphite element in the TURF is shown in Fig. 12. This equipment will occupy approximately one-half of the space in the contaminated fabrication and clean fabrication cells. It is to be located on the opposite side of the cells from the oxide fabrication equipment. In the process, dried sol-gel oxide microspheres are fed from the mechanical processing cell by a transfer conveyor; and then the material flows through various transfer conveyors from one step to another according to the sequence depicted on the flow sheet and in the equipment layout.

Because of the handling limitations imposed by the building clearances, a fuel element longer than 12 ft cannot be handled in the facility. Therefore, it will be necessary to accomplish the assembly operations for the fuelled-graphite element in another facility or at the reactor site. This could be done by means of a central fitting of the threaded or bayonet type, and the joint could be sealed by an inductively heated braze joint.
All of the hardware which is used in the fabrication of the fuel elements will be loaded into magazines and brought into the cell by means of the cell manipulation and crane system.

Present status of the facility and equipment

The construction of the TURF was started in April 1965 and is expected to be completed in early 1967. All of the oxide fabrication equipment has been conceptually designed, and a substantial portion of the detailed design of this equipment has been accomplished. Two items of oxide fabrication equipment, the vibratory compaction machine and the fuel transfer machine, are now nearing completion of construction.

We have just commenced the conceptual design of the equipment for fabrication of fuelled-graphite elements. The accomplishment of detailed design of a great number of the items in this process line depends upon the performance of equipment now being readied for pilot operation and upon data obtained in other experimental programmes which we have under way.

REFABRICATION ECONOMICS

From the experience gained in the operation of the pilot demonstration in the Kilorod Facility, the design of the TURF and its equipment and other studies we have conducted, we are in a position to estimate the cost of refabricating various fuel elements. Certainly, our estimates represent an extrapolation of existing technology; and they must be confirmed by additional and more accurate experience. But, they do suffice to give an indication of the cost penalties which might be incurred in the refabrication of fuel under various processing schemes. Therefore, let us consider two types of fuel elements and assume that reasonable refinement and improvement of fabrication processes have been accomplished.

Method of analysis

In our economic analyses, one objective has been to do the complete analysis of the refabrication costs for any reactor fuel element. This objective requires a tremendous number of calculations. Therefore, during the past two years we have developed computer programmes for the calculation of the cost of fabricating various types of fuel elements. The computer programmes were written from data evolved in the following manner. The basic equipment required in the plant was selected and the uninstalled cost of this equipment was estimated. These costs were multiplied by various factors to determine the final capital cost of the fabrication plant. The multiplication factors were obtained from studies conducted jointly with construction engineers in which detailed plant layouts were examined for various sizes and types of fabrication plants. These studies yielded detailed breakdowns of such cost elements as building construction, equipment installation, instrumentation, engineering and pre-operation charges. In the computer programmes, equations are included for each step in the process so that,
FIG. 11. Flowsheet for fabrication of fuelled-graphite elements in thorium-uranium recycle facility
FIG. 12. Schematic layout of equipment for fabrication of fuelled-graphite elements in thorium-uranium recycle facility
upon description of the size and type of plant in which the fabrication is to occur, proper capital costs of that step can be obtained.

Operating costs were determined by similar methods. Material costs in the manufacture of the product were obtained through consultation with industrial manufacturers.

Cost of preparing fuel materials by the sol-gel method

As has been previously noted, we favour the use of sol-gel material as input for vibratory compaction and for the fuelled-graphite process. The computer programmes do not calculate the cost of preparing materials produced by the sol-gel technique, but the cost of producing such material has been estimated by Harrington and Chandler [28]. Their data are presented in Figs. 13 and 14 for bulk oxide material and spherical oxide particles, respectively. The cost of sol-gel material preparation is not included, unless specifically noted, in the fabrication costs presented in this paper.

Basis for calculation of fabrication costs

In calculating the fabrication costs which are presented in this paper, we made the following assumptions:

1. The fabrication plants would be single-purpose; that is, only one particular type of fuel element would be fabricated.
2. The fabrication plants would operate 260 days per year, three shifts per day.
3. The plants would be amortized at an annual rate of 22% of the capital cost.

The data do not include charges for profit and for inventory or cost of source or fissionable material. Non-nuclear hardware costs are included.
FIG. 14. Cost of preparation of oxide microspheres by the sol-gel process

FIG. 15. Effect of production rate and mode of fabrication on the cost of fabricating SSCR fuel elements

Cost of a typical metal-clad oxide fuel element

The reference metal-clad oxide fuel element was the SSCR element described previously, and the flow sheet was that presented in Fig. 1. It was assumed that the fuel elements would be autoclaved. Figure 15 shows the effect of mode of fabrication and production rate on the cost of fabricating the SSCR-type fuel element. As can be seen, the production rate has a substantial effect on the cost of fabrication. This cost advantage cannot be realized unless sustained production of identical fuel elements is allowed. If dimensions or configurations are changed, then penalties would be incurred for retooling of the facility.
One comparison of interest in recycling fuel is that of the cost penalties incurred because of $^{232}\text{U}$ concentration in the fuel or because of $^{228}\text{Th}$ in recycle thorium. Figure 16 shows the effect of $^{232}\text{U}$ concentration on the cost of fabrication of the SSCR fuel element in four plants with different capacities, 60, 230, 930, and 3700 kg/d. It should be noted that we do not know exactly the transition point (the first break in the curves) between semi-remote and remote operation; but, as can be seen from the curves, it is not mandatory that these points be determined precisely since the cost penalty incurred for remote fabrication over semi-remote fabrication is not substantial. The recycle thorium case is depicted as being equivalent to 4000 ppm $^{232}\text{U}$ in heavy metal.

![Figure 16. Effect of $^{232}\text{U}$ concentration on the cost of fabricating SSCR fuel elements](image)

**Cost of fabrication of fuelled-graphite elements**

The reference fuel element for calculation of fuel fabrication costs for the HTGR is shown in Fig. 2; the process used, in Fig. 3. Figure 17 presents the effect of mode of fabrication and plant capacity on fabrication costs of fuel elements containing (U-Th)$_2$C$_2$ particles. Figure 18 presents the effect of $^{232}\text{U}$ concentration on fabrication costs at several different plant capacities. We should note that the cost penalty for recycling thorium as discrete particles is quite high at low production rates because of the necessity of maintaining two remote production lines, one for $^{233}\text{U}$ and one for thorium. These curves show that the cost relationships for the HTGR fuel element behave in a manner similar to those of the SSCR.

**Cost penalties for remote fabrication**

Detailed analysis of the data used in the curves previously presented reveals the incremental cost factors which can be applied to similar fabrication processes to obtain the refabrication cost if the cost of the fabricating non-recycle fuel is known. Tables V and VI present, respectively, the ratios
of remote fabrication cost to hooded fabrication cost for SSCR and for the fuelled-graphite element.

It is clear that the capital and operating factors decrease as plant capacity increases. The decrease in these factors with plant capacity is understandable because there are certain costs which are only first costs in remote operations. For example, health physics facilities are required in a 60-kg/d plant as well as a 3700-kg/d plant; and probably the facilities would be of the same type and very nearly the same size in both cases. Thus, we are observing the effect of more efficient utilization of certain service personnel and facilities in the larger plants.
COST RATIOS FOR COMPARISON OF REMOTE AND HOODED PLANTS FABRICATING SSCR FUEL ELEMENTS

<table>
<thead>
<tr>
<th>Ratio of remote-to-hooded cost for plant capacity</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital</td>
<td>1.23</td>
<td>1.21</td>
<td>1.18</td>
<td>1.14</td>
</tr>
<tr>
<td>Operating</td>
<td>1.52</td>
<td>1.38</td>
<td>1.41</td>
<td>1.34</td>
</tr>
<tr>
<td>Total (including hardware)</td>
<td>1.29</td>
<td>1.21</td>
<td>1.15</td>
<td>1.10</td>
</tr>
</tbody>
</table>

\( ^{232} \text{U} \) in heavy metal, virgin thorium.

COST RATIOS FOR COMPARISON OF REMOTE AND HOODED PLANTS FABRICATING (U-Th) PARTICLES

<table>
<thead>
<tr>
<th>Ratio of remote-to-hooded cost for plant capacity</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital</td>
<td>1.87</td>
<td>1.79</td>
<td>1.73</td>
<td>1.75</td>
</tr>
<tr>
<td>Operating</td>
<td>1.47</td>
<td>1.43</td>
<td>1.37</td>
<td>1.37</td>
</tr>
<tr>
<td>Total (including hardware)</td>
<td>1.53</td>
<td>1.42</td>
<td>1.34</td>
<td>1.35</td>
</tr>
</tbody>
</table>

\( ^{232} \text{U} \) in heavy metal, virgin thorium.

The cost ratios for the SSCR fuel elements and the HTGR fuel elements are different. This is because a greater number of the operations involved in the fabrication of SSCR fuel elements are done externally to the remotely operated facilities and do not incur the penalties for remote fabrication.

This point can be further illustrated by Table VII, which presents the cost ratios for comparison of remote and hooded plants fabricating fuelled-graphite elements containing discrete particles of UC\(_2\) and ThC\(_2\). In this particular case, a great portion of the fabrication (the conversion and coating of virgin thorium oxide) is done in facilities which are hooded. Therefore, the cost ratios are considerably lower than those presented in Fig. 6 for plants fabricating fuelled-graphite elements containing (U-Th)C\(_2\) particles.
REFABRICATION TECHNOLOGY

TABLE VII

COST RATIOS FOR COMPARISON OF REMOTE AND HOODED PLANTS FABRICATING UC₂ AND ThC₂ PARTICLES

<table>
<thead>
<tr>
<th>Ratio of remote-to-hooded cost for plant capacity (kg heavy metal/d) of</th>
<th>60</th>
<th>230</th>
<th>930</th>
<th>3700</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capital</td>
<td>1.29</td>
<td>1.20</td>
<td>1.18</td>
<td>1.09</td>
</tr>
<tr>
<td>Operating</td>
<td>1.26</td>
<td>1.22</td>
<td>1.18</td>
<td>1.16</td>
</tr>
<tr>
<td>Total (including hardware)</td>
<td>1.24</td>
<td>1.16</td>
<td>1.12</td>
<td>1.08</td>
</tr>
</tbody>
</table>

*50 ppm \(^{232}\)U in heavy metal, virgin thorium.

SUMMARY

The investigation of the technology for refabrication of bred fuel is just beginning. Much more evaluation needs to be done and much thought must go into systems required for recycle of such fuel. However, it is encouraging that the cost penalties associated with the refabrication of such fuel do not appear prohibitive.

Recycle fuel in the thorium-uranium-233 fuel cycle will very probably require remote fabrication. This probability dictates that the fuel element design be simple and be optimized for minimal cost and that the processes be simple, easy to operate, short, and capable of being done remotely. Furthermore, we must take into account the fact that some operations which typify the fabrication of non-recycle fuel are not economically feasible in remote situations.

In the cost analyses, we have been reasonably optimistic in that we have assumed the satisfactory solution of certain of the problems which have been previously mentioned. This does not mean, however, that present-day practices can be directly applied to remote fabrication technology. It is, therefore, clear that we must press development in a number of areas if we are to attain economical Th-\(^{233}\)U recycle technology for any particular type of reactor.

We are conducting a comprehensive programme at the Oak Ridge National Laboratory aimed at meeting the objectives of economical recycle of fuel. With further development of the pilot-scale demonstration of methods for vibratory compaction and for preparation of particles for fuelled-graphite elements, we should have a much better basis for the design of equipment for remote fabrication of fuel in the TURF. Once we have designed and operated equipment in the TURF, we shall have a much better basis than we do now for evaluation of the technical feasibility and cost of refabricating fuel elements of any particular type.
ACKNOWLEDGEMENTS

Obviously, the work represented in this paper is the result of the efforts of a number of persons. We would particularly like to acknowledge the help of those who have made direct contributions: R.B. Pratt and J.D. Sease for their work in the engineering of the remote fabrication systems; T.N. Washburn for his assistance in the analysis of fuel refabrication economics; and R.L. Pilloton for valuable contributions to the economic and technical analyses for refabrication of fuelled-graphite elements. Finally, we should note that all of these persons have made valuable contributions to the formulation of our programmes and concepts for refabrication.

REFERENCES


INTRODUCTION

Thorium metal has an isotropic body-centred cubic crystal structure, higher strength, and a 600°C higher melting point than uranium which has an anisotropic orthorhombic crystal structure. Thorium dioxide crystallizes in the same stable cubic fluorite lattice as uranium dioxide but has a melting point some 500°C higher and forms a complete series of solid solutions with uranium dioxide with the same lattice configuration. The carbides of thorium and uranium are similar and also form a series of solid solutions. All of these physical facts indicate the promise of improved performance for thorium-base fuels even if one ignores its breeding potential.

Historically, thorium and thorium alloys or compounds have been studied much less intensively than uranium and its alloys. Only recently have a number of investigators begun to seriously study the thorium-base compounds, both oxides and carbides for potential application to power reactors.

The programme at Oak Ridge National Laboratory has emphasized the sol-gel [1] process for the preparation of thorium-base fuel compounds as potentially more economical with a denser, more uniform product. Calcined sol-gel thoria-urania fuel has been semi-remotely processed and fabricated into rods by vibratory compaction techniques [2]. The sol-gel material can be processed into truly round, smooth microspheres of theoretical density for coated particle applications [1].

The data from the irradiation tests reported have shown that thorium-base fuels, both metallic and ceramic, have the basic characteristics required for power reactors. Interest has shifted to determining the maximum performance characteristics to take advantage of the indicated potentials for higher temperature operation and increased power ratings with improved stability. This paper reviews the data obtained from different irradiation-test programmes on sol-gel and conventionally processed thorium-base fuels, as metal-clad bulk oxides and as-coated particles. Finally, there is a section which reviews the available data on the irradiation characteristics of thorium-base metallic fuels.

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation.
IRRADIATION BEHAVIOUR OF Th-U ALLOYS

BULK OXIDE FUELS OF ThO₂–UO₂

In support of the Thorium Utilization Fuel Cycle Program a series of irradiation tests on metal-clad bulk oxide fuels containing intimately mixed ThO₂ and UO₂ was started at the Oak Ridge National Laboratory in 1961. This programme has concentrated on ceramic fuels produced by the sol-gel [1] process and fabricated into rods by the use of vibratory compaction. The initial objectives were to compare the basic nuclear fuel performance characteristics of this chemically produced ceramic with those of arc-fused material and the performance of vibratory compacted fuel with pressed and sintered pellets. The early tests indicated a favourable comparison and the objectives have been expanded to include:

1. Effects of sol-gel processing variables
2. Effects of fuel-rod geometry (diameters and lengths)
3. Determination of the maximum performance characteristics of vibratory compacted sol-gel fuels
4. Potential of mixed progeny sol-gel fuels (e.g. ThO₂–PuO₂)
5. Effects of semi-remote fabrication of vibratory compacted ThO₂–{²³³}UO₂ sol-gel fuels [2]

Table 1 lists the groups of fuel rods, their principal objectives, and their current status.

Most of the tests have been made with non-instrumented rods in the Materials Test Reactor (MTR), Engineering Test Reactor (ETR), and the Chalk River National Research Experimental Reactor (NRX). Three tests were conducted in the Oak Ridge Research Reactor (ORR) [3], one a trefoil cluster in a pressurized-water loop operating at 250°C and 1750 lb/in² and the other two were instrumented rods in the Poolside Facility with 538 and 704°C cladding temperatures.

Post-irradiation examinations include dimensional analysis for swelling or bowing, fission-gas release measurements, gamma scanning, burn-up determinations, X-ray diffraction analysis, and metallographic examinations.

As with any irradiation programme, and particularly with one as broad in scope as this, there are time overlaps in obtaining data to evaluate the various objectives. The initial scoping studies are complete and all other portions are being pursued. Many of the earlier tests have been reported in detail [4–8] and have shown that sol-gel ThO₂–UO₂ has the basic performance characteristics for a power reactor fuel.

Effects of high burn-up

The effects of higher burn-up on three types of ThO₂–UO₂ fuels have recently become available. One of the advantages of the ThO₂ base fuels is the relatively high breeding ratio. It can be calculated that in an appropriate thermal flux a thoria fuel containing approximately 1 wt.% ²³⁵U will produce essentially constant power to a burn-up of approximately 50,000 MWd/t (Th+U). However, in most of our tests a 4 to 5 wt.% ²³⁵U has been used to shorten the time required for significant burn-up. Total depletion of this initial uranium will produce approximately 30,000 MWd/t but it is necessary to move the test rods periodically to higher flux posi-
### Table 1. Summary of Thorium Fuel Cycle Program Irradiations of Powder-Packed Capsules (Rods) of Type 304 Stainless Steel*

<table>
<thead>
<tr>
<th>Designation</th>
<th>Type of Oxide</th>
<th>Density, % theor.</th>
<th>Fuel Rod Dimensions, in.</th>
<th>Linear Heat Rating, Btu/hr-ft</th>
<th>Peak Burnup, Mwd/tonne metal</th>
<th>Status</th>
<th>Objective</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTR-I</td>
<td>Arc-fused</td>
<td>86 to 87</td>
<td>11</td>
<td>0 312 0 025</td>
<td>40,000</td>
<td>2 examined, 4 being examined, 1 in pile</td>
<td>Provide base-line data to use in comparing sol-gel and arc-fused oxide</td>
</tr>
<tr>
<td>MTR-II</td>
<td>Sol-Gel S</td>
<td>88 to 89</td>
<td>22</td>
<td>0 312 0 025</td>
<td>62,000</td>
<td>In Pile</td>
<td>Obtain higher heat rating by increasing enrichment</td>
</tr>
<tr>
<td>MTR-III</td>
<td>Sol-Gel 35</td>
<td>86 to 89</td>
<td>12</td>
<td>0 438 0 025</td>
<td>85,000</td>
<td>In Pile</td>
<td>Compare oxide calcining atmospheres and higher heat ratings obtained by increasing diameter</td>
</tr>
<tr>
<td>ETR-I</td>
<td>Sol-Gel 35</td>
<td>86 to 89</td>
<td>12</td>
<td>0 438 0 025</td>
<td>&gt; 100,000</td>
<td>Being Examined</td>
<td>Same as for MTR-III</td>
</tr>
<tr>
<td>NRX-I</td>
<td>Sol-Gel A and B</td>
<td>86 to 87</td>
<td>11</td>
<td>0 312 0 025</td>
<td>17,000</td>
<td>Examined</td>
<td>Provide base-line data</td>
</tr>
<tr>
<td>NRX-II</td>
<td>Sol-Gel C</td>
<td>83 to 86</td>
<td>22</td>
<td>0 312 0 025</td>
<td>22,000</td>
<td>Examined</td>
<td>Study effect of increased length</td>
</tr>
<tr>
<td>NRX-III</td>
<td>Sol-Gel S</td>
<td>88 to 89</td>
<td>39</td>
<td>0 312 0 025</td>
<td>28,000</td>
<td>Being Examined</td>
<td>Study effect of increased length</td>
</tr>
<tr>
<td>ORR Loop</td>
<td>Sol-Gel 26</td>
<td>84 to 85</td>
<td>22</td>
<td>0 460 0 015</td>
<td>52,000</td>
<td>Examined</td>
<td>Study in pressurized water at 260°C and 1750 psi</td>
</tr>
<tr>
<td>ORR Poolside</td>
<td>Sol-Gel D</td>
<td>85</td>
<td>7</td>
<td>0 625 0 020</td>
<td>35,000</td>
<td>Examined</td>
<td>Measure effective thermal conductivity using central thermocouple in Na-K at 315 psig, 540 and 705°C</td>
</tr>
<tr>
<td>ETR-II</td>
<td>BNL-Sol-Gel</td>
<td>90</td>
<td>19</td>
<td>0 499 0 035</td>
<td>~65,000</td>
<td>In Pile</td>
<td>Study effects of remote fabrication and oxide recalcining</td>
</tr>
<tr>
<td>ETR-III</td>
<td>Sol-Gel ThO₂</td>
<td>88</td>
<td>19</td>
<td>0 499 0 035</td>
<td>~80,000</td>
<td>Being Prepared</td>
<td>Study ThO₂ blanket material with gradually increasing heat rating and provide high Pa low-fission-gas-product material for chemical processing</td>
</tr>
</tbody>
</table>

*Except ETR-II, III and two of the three ORR loop specimens were Zircaloy clad

**Temp packed
tions to maintain essentially constant heat ratings. Test rods from the MTR Group I containing either sol-gel or arc-fused vibratory compacted ThO₂-4.5% UO₂ have been examined after burn-ups in excess of 70,000 MWd/t (Th+U). A drawing of these fuel rods is shown in Fig. 1. In addition, four rods, containing pressed and sintered pellets of ThO₂-4.5% UO₂ at burn-ups between 37,000 and 119,000 MWd/t (Th+U), have been examined.

The fabrication details, irradiation histories, and the significant post-irradiation findings and calculations are presented in Tables 2a, b, and c. The time-average values for the various heat ratings are for the peak burn-up region in each rod calculated from the maximum burn-ups at the same location, while the maximum instantaneous values are based on fuel depletion calculations and flux steps during irradiation and do not include any changes in flux profile during the various test-reactor cycles. All of the values for

$$
\frac{T_s}{k d \theta}
$$

assume that the surface temperature of the fuel ($T_s$) is the same as the temperature calculated for the inside surface of the cladding and that the thermal conductivity data of Kingery [9] apply for

$$
\frac{T_s}{k d \theta}
$$

at temperatures below this surface temperature. Although the reliability of these assumptions is questionable, as will be seen later in this paper, they do provide a consistent method of comparison.
Table 2a. Effects of High Burnup on ThO\textsubscript{2}-UO\textsubscript{2} Fuels: Description of Irradiation Test Rods\textsuperscript{a}

<table>
<thead>
<tr>
<th>Group</th>
<th>Number</th>
<th>Fuel Rod Identification</th>
<th>Type of Oxide</th>
<th>Total Uranium\textsuperscript{b} (wt %)</th>
<th>Oxide Particle Size</th>
<th>Vibrated Density\textsuperscript{c} (% TD)</th>
<th>Fuel Rod Dimension</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Length (cm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Outside Diameter (cm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Wall (cm)</td>
</tr>
<tr>
<td>MTR-I</td>
<td>43-59</td>
<td>U-1</td>
<td>Arc fused</td>
<td>3.96</td>
<td>d</td>
<td>85.7</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-60</td>
<td>U-2</td>
<td>Arc fused</td>
<td>3.96</td>
<td>d</td>
<td>85.6</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-61</td>
<td>U-3</td>
<td>Arc fused</td>
<td>3.96</td>
<td>d</td>
<td>85.5</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-62</td>
<td>U-5</td>
<td>Sol-gel E</td>
<td>3.96</td>
<td>d</td>
<td>87.0</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-63</td>
<td>Z-5</td>
<td>Sol-gel E</td>
<td>3.96</td>
<td>d</td>
<td>86.2</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-64</td>
<td>Z-7</td>
<td>Sol-gel E</td>
<td>3.96</td>
<td>d</td>
<td>86.2</td>
<td>28.70</td>
</tr>
<tr>
<td></td>
<td>43-65</td>
<td>Z-8</td>
<td>Sol-gel E</td>
<td>3.96</td>
<td>d</td>
<td>85.5</td>
<td>28.70</td>
</tr>
<tr>
<td>MTR</td>
<td>43-39</td>
<td>712</td>
<td>Arc fused</td>
<td>3.92</td>
<td>Pellets</td>
<td>93</td>
<td>11.43</td>
</tr>
<tr>
<td></td>
<td>43-40</td>
<td>729</td>
<td>Arc fused</td>
<td>3.92</td>
<td>Pellets</td>
<td>93</td>
<td>11.43</td>
</tr>
<tr>
<td></td>
<td>43-41</td>
<td>730</td>
<td>Arc fused</td>
<td>3.92</td>
<td>Pellets</td>
<td>93</td>
<td>11.43</td>
</tr>
<tr>
<td>ETR</td>
<td>43-44</td>
<td>645</td>
<td>Arc fused</td>
<td>3.92</td>
<td>Pellets</td>
<td>93</td>
<td>11.43</td>
</tr>
</tbody>
</table>

\textsuperscript{a}Clad with type 304 stainless steel with operating cladding temperature at 100°C.

\textsuperscript{b}Enriched in $^{235}$U to 93%.

\textsuperscript{c}Based upon a calculated density depending on composition (approx 10.04 g/cm\textsuperscript{3}).

\textsuperscript{d}Distribution A: 60 wt % -10 +16 mesh; 15 wt % -70 +140 mesh; 25 wt % -200 mesh.
Table 2b. Effects of High Burnup on ThO₂-UO₂ Fuels: Irradiation History

<table>
<thead>
<tr>
<th>Experiment Identification</th>
<th>Average External Pressure (psia)</th>
<th>Average Reactor Full Power Time (Days)</th>
<th>Peak Linear Heat Rating Average (W/cm)</th>
<th>Peak Linear Heat Rating Instantaneous (W/cm)</th>
<th>Peak Cladding Heat Flux Average (W/cm²)</th>
<th>Peak Cladding Heat Flux Instantaneous (W/cm²)</th>
<th>Maximum Burnup (Nw/d/tonne Th+U)</th>
<th>Maximum Burnup (fissions/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTR-I</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>U-1</td>
<td>40</td>
<td>110.2</td>
<td>299</td>
<td>362</td>
<td>120</td>
<td>145</td>
<td>12,300</td>
<td>2.50</td>
</tr>
<tr>
<td>U-2</td>
<td>40</td>
<td>376.2</td>
<td>297</td>
<td>388</td>
<td>119</td>
<td>155</td>
<td>42,100</td>
<td>8.56</td>
</tr>
<tr>
<td>U-3</td>
<td>40</td>
<td>707.5</td>
<td>267</td>
<td>419</td>
<td>107</td>
<td>168</td>
<td>71,000</td>
<td>14.43</td>
</tr>
<tr>
<td>Z-5</td>
<td>40</td>
<td>110.2</td>
<td>341</td>
<td>426</td>
<td>137</td>
<td>172</td>
<td>14,000</td>
<td>2.87</td>
</tr>
<tr>
<td>Z-7</td>
<td>40</td>
<td>375.2</td>
<td>286</td>
<td>347</td>
<td>114</td>
<td>138</td>
<td>40,200</td>
<td>8.23</td>
</tr>
<tr>
<td>Z-8</td>
<td>40</td>
<td>691.1</td>
<td>311</td>
<td>507</td>
<td>124</td>
<td>202</td>
<td>81,000</td>
<td>16.46</td>
</tr>
<tr>
<td>MTR</td>
<td>712</td>
<td>40</td>
<td>905</td>
<td>652</td>
<td>168</td>
<td>261</td>
<td>119,400</td>
<td>26.41</td>
</tr>
<tr>
<td>729</td>
<td>40</td>
<td>660</td>
<td>461</td>
<td>507</td>
<td>185</td>
<td>285</td>
<td>96,000</td>
<td>24.6</td>
</tr>
<tr>
<td>730</td>
<td>40</td>
<td>497</td>
<td>270</td>
<td>394</td>
<td>108</td>
<td>182</td>
<td>37,000</td>
<td>10.8</td>
</tr>
<tr>
<td>ETR</td>
<td>645</td>
<td>180</td>
<td>406</td>
<td>394</td>
<td>158</td>
<td>250</td>
<td>50,000</td>
<td>12.8</td>
</tr>
</tbody>
</table>

*Based on 200 MeV/fission.
Table 2c. Effects of High Burnup on ThO₂-UO₂ Fuels: Postirradiation Observations and Calculations

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel Rod Identification</th>
<th>Time Average (W/cm)</th>
<th>Maximum Instantaneous (W/cm)</th>
<th>$^{85}\text{Kr}$ Release (%)</th>
<th>Dimensional Changes</th>
<th>Radial Extent of Microstructural Changes</th>
<th>Equiaxed Grains (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTR-I</td>
<td>U-1</td>
<td>33.8</td>
<td>39.8</td>
<td>2.4</td>
<td>+0.025</td>
<td>+0.025</td>
<td>a</td>
</tr>
<tr>
<td></td>
<td>U-2</td>
<td>33.7</td>
<td>41.9</td>
<td>7.2</td>
<td>0.000</td>
<td>-0.051</td>
<td>a</td>
</tr>
<tr>
<td></td>
<td>U-3</td>
<td>31.6</td>
<td>44.7</td>
<td>6.4</td>
<td>+0.005</td>
<td>+0.013</td>
<td>a</td>
</tr>
<tr>
<td></td>
<td>Z-5</td>
<td>37.8</td>
<td>44.4</td>
<td>0.5</td>
<td>+0.025</td>
<td>+0.075</td>
<td>a</td>
</tr>
<tr>
<td></td>
<td>Z-7</td>
<td>32.8</td>
<td>38.4</td>
<td>13.2</td>
<td>-0.025</td>
<td>-0.037</td>
<td>a</td>
</tr>
<tr>
<td></td>
<td>Z-8</td>
<td>34.6</td>
<td>52.7</td>
<td>17.0</td>
<td>+0.015</td>
<td>+0.058</td>
<td>0.1b</td>
</tr>
<tr>
<td>MTR</td>
<td>712</td>
<td>44.8</td>
<td>65.4</td>
<td>22.8</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>729</td>
<td>48.2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>730</td>
<td>31.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>c</td>
</tr>
<tr>
<td>ETR</td>
<td>645</td>
<td>42.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

---

*a* Sintering boundary not delineated.

*b* Calculated heat flux for equiaxed grain growth $\int \frac{dT}{kdT} / T_o = 31.5$ W/cm.

*c* Gas samples diluted with air in sampling and partially lost.
The dimensional changes on the MTR-I group indicate no significant swelling in these highly restraining clads. Pre-irradiation measurements were not made on the pellet rods, since they were originally intended for chemical processing studies only; however, the post-irradiation diameters were within the tolerance specifications for the tubing used as cladding. The fission-gas data indicate a slightly higher release for the sol-gel material, but there is no evidence of a breakaway phenomenon. The principal factor in gas release at these heat ratings seems to be time at temperature, which is consistent with a diffusion mechanism for gas release.

The macroscopic appearance of the various sections shown in Figs. 2, 3, and 4 shows a remarkable similarity in the appearance of the fuels at these high burn-up levels. Sections of the rods with less burn-up are similar, although the radial position of the more or less circumferential cracks appear to move outward with increased exposure. The radial location of these cracks appears to be associated with the extent of in-pile sintering or equiaxed grain growth. Since this phenomenon is dependent on time as well as temperature, the radial extent of such changes would be expected to increase with higher burn-up (i.e. time) at a constant heat flux. The microstructures on these rods clearly show the time dependence of such diffusional processes as sintering and grain growth. Figures 5 and 6 are composite radial micrographs of the arc-fused vibratory compacted rods at various levels of exposure. Figures 7 and 8 are similar composites of the sol-gel vibratory compacted rods, while Figs. 9, 10, 11, and 12 show...
FIG. 3. MTR group I, capsule Z-8: macroscopic views of selected section locations, as-cut

FIG. 4. Pellet rod No. 712: macroscopic views of selected section locations, as-cut
FIG. 5. Composite photomicrographs showing typical transverse sections of unirradiated and irradiated vibratory compacted arc-fused ThO$_2$-4.5% UO$_2$. Radial area from the outside diameter to the centre of the fuel. As-polished.
FIG. 6. Composite photomicrographs showing typical transverse sections of irradiated vibratory compacted arc-fused ThO$_2$-4.5% UO$_2$. Radial area from the outside diameter to the centre of the fuel. As-polished
FIG. 7. Composite photomicrographs showing typical transverse sections of unirradiated and irradiated vibratory compacted sol-gel E ThO$_2$-4.9% UO$_2$. Radial area from the outside diameter to the centre of the fuel. As-polished.
FIG. 8. Composite photomicrographs showing typical transverse sections of irradiated vibratory compacted sol-gel $\text{ThO}_2$-$4.5\% \text{UO}_2$. Radial area from the outside diameter to the centre of the fuel. As-polished
(a) As-polished. ×500. (b) Etched. ×250
FIG. 10. Representative regions of fuel from fuel rod 43-41. \(10.8 \times 10^{23} \text{ fissions/cm}^3\). (a) Peripheral region. (b) Mid-radial region. (c) Central region. As-polished. \(\times 250\). Reduced 16.3%
IRradiation behaviour of Th-U alloys

FIG. 11. Representative regions of fuel from fuel rod 43-44. \( (12.8 \times 10^{18} \text{ fissions/cm}^3) \). (a) Cladding-fuel interface. (b) Mid-radial region. (c) Central region. As-polished. x250. Reduced 25%
FIG. 12. Representative areas from fuel rod 43-40. \( (24.6 \times 10^{20} \text{ fissions/cm}^2) \). (a) Cladding-fuel interface. (b) Central region. As-polished. \( \times 250 \). Reduced 34%
selected micrographs at various radial positions for the pressed and sintered pellets.

In the case of the arc-fused material, the cracks in the original fuel fragments were first accentuated in the mid-radius region while the porosity in the centre agglomerated. With continued irradiation, the cracks apparently healed and a grain structure developed within the fuel fragments while the pore sizes in the centre increased. At 70 000 MWd/t, $14 \times 10^{20}$ fissions/cm$^3$, the central region appears to have developed a dense laminar-like structure with some inter-particle sintering and the outer two-thirds shows a uniform dispersion of fine porosity with a band of larger pores at the dividing line between the two structures.

The sol-gel fuel fragments in the unirradiated condition are crack-free but do contain uniformly distributed sub-microscopic pores. On irradiation, this porosity first appears to agglomerate and accentuate the fine-particle size. Continued irradiation develops a distinct grain structure in the centre of the fuel with equiaxed grains 6 to 8 μm in size separated from the balance of the fuel by a circumferentially oriented crack. At 81 000 MWd/t, $16 \times 10^{20}$ fissions/cm$^3$, the development of equiaxed grains is very distinct with a maximum size of 60 μm. The circumferential crack still separates this area from the remainder of the fuel, but there appears to be some inter-fragment sintering in the outer two-thirds and, as with the arc-fused material, a band of moderately large porosity is found in this area.

With both fuels, after irradiation there is a second white metallic-appearing phase developed in the central region. Initially, this appears to be closely associated with porosity but at exposures of $8 \times 10^{20}$ fissions/cm$^3$, or 375 reactor full-power days, this phase appears to be randomly distributed throughout the matrix. A third light-grey phase was also found located in discrete limited areas of some specimens. Although this phase has not been definitely identified, the appearance and the association with fine fuel fragments indicate that it is probably aluminium silicate contamination from the ball-milling operations.

The evolution of structural changes in the pressed and sintered pellets is not so clearly defined since an equiaxed grain structure exists before irradiation. As with the other fuels, there is an accumulation of porosity. There is a gradual degradation of the distinct grain structure starting at the centre and spreading out towards the cladding. The one exception to this is the one rod exposed in the ETR where the maximum instantaneous heat flux may have been considerably higher. In this case the central degradation is apparent but distinct grain growth is noted between the mid-radius and the cladding. All structures show a second white metallic phase particularly in the central regions. The grey impurity phase seen in the unirradiated, as well as the irradiated, structures has been identified by X-ray diffraction as Al$_2$O$_3$ and 6 Al$_2$O$_3$·CaO. The effect of these high levels of impurities on the microstructures is not known.

One rod of vibratory compacted sol-gel fuel is still under irradiation and will be exposed to a burn-up in excess of 100 000 MWd/t (Th+U). However, from the findings to date it is apparent that there is no significant difference between the performance of vibratory compacted sol-gel and arc-fused ThO$_2$-UO$_2$ fuels. Both compare favourably with pressed and sintered pellet fuels at linear heat ratings between 300 and 400 W/cm ($\beta$kd6 34-45
<table>
<thead>
<tr>
<th>Group</th>
<th>Rod</th>
<th>Fuel Material(^a)</th>
<th>Burnup (fissions/cm(^3))</th>
<th>Unit Cell(^b)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Preirradiation</td>
<td>Postirradiation</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(\times 10^{20})</td>
<td></td>
</tr>
<tr>
<td>MTR-I</td>
<td>U-2</td>
<td>Arc-fused</td>
<td>8.6</td>
<td>5.594 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>U-3</td>
<td>Arc-fused</td>
<td>14.4</td>
<td>5.594 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>Z-7</td>
<td>Sol-gel</td>
<td>8.2</td>
<td>5.593 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>Z-8</td>
<td>Sol-gel</td>
<td>16.5</td>
<td>5.593 ± 0.001</td>
</tr>
<tr>
<td>Pellet</td>
<td>645</td>
<td>Pressed and sintered</td>
<td>12.8</td>
<td>5.590 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>729</td>
<td>Pressed and sintered</td>
<td>24.6</td>
<td>5.590 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>712</td>
<td>Pressed and sintered</td>
<td>26.4</td>
<td>5.590 ± 0.001</td>
</tr>
</tbody>
</table>

\(^a\) ThO\(_2\)-4.5\% UO\(_2\), 235U enriched 93%.

\(^b\) Nelson-Riley Function.
IRRADIATION BEHAVIOUR OF Th-U ALLOYS

In addition, the ThO$_2$-5% UO$_2$ fuels show no evidence of breakaway swelling or fission-gas release at burn-ups as high as 2.6 x 10$^{21}$ fissions/cm$^3$, 120 000 MWd/t, where three-fourths of the fission energy has been derived from the $^{233}$U bred into the fuel during irradiation. According to swelling estimates [10] on the pellet rods, the ThO$_2$-4.5% UO$_2$ change in volume was limited to 0.46% $\Delta V/10^{20}$ fissions cm$^3$. This is roughly one-half the rate proposed by Anderson [11] and others for UO$_2$ ($0.8 \Delta V/10^{20}$ fissions cm$^3$). X-ray diffraction data on the various irradiated ThO$_2$-UO$_2$ fuels have also tended to confirm the stability of the cubic fluorite crystal structure lattice and indicate a considerable capacity for fission products, as can be seen from the negligible changes in lattice parameters in Table 3.

Effects of high heat ratings

Although the high burn-up test rods have shown the usefulness of the thorium-base fuels at moderate heat ratings comparable to current pressurized water-power reactors, the full utilization of the sol-gel ThO$_2$-UO$_2$ fuels requires an investigation of the maximum performance characteristics and an understanding of the effect of processing variables on these characteristics. The ORR loop and ORR Poolside Experiments were the first steps in this phase of the investigation. More recently, experiment groups MTR-II, MTR-III, and ETR-I were prepared and inserted in the reactor for this purpose. Tables 4a, b, and c present the fabrication characteristics, irradiation histories, and principal post-irradiation findings on the rods examined to date.

The findings on the ORR loop and Poolside experiments have been reported [5-8], and the ETR-I rods are currently being evaluated. This latter group of three rods was subjected to the highest flux with the expectation of producing central melting. A common sol-gel ThO$_2$-5% UO$_2$ preparation was used as fuel in these rods, however, it was calcined in three different atmospheres: air, Ar-4% H$_2$ (the standard atmosphere), and nitrogen to produce sol-gel batches 35-1, 35-2, and 35-3, respectively. The post-irradiation dimensional analyses have shown no indications of swelling or bowing. Fission-gas samples and gamma scans were obtained. Based on the gamma scans, which showed no anomalies, and the structures found, these rods were sectioned and are currently being evaluated metallographically. Figures 13, 14, and 15 show the resulting as-cut structures at the various section locations.

From these sections, it is apparent that the linear heat ratings were sufficient to develop a central void in all three of the rods. The levels of the gamma scans and the results of the burn-up analysis indicate that there was a real difference in fission rate and consequently in linear heat rating as shown in Tables 4b and c. The extent of the central voids appears to match the indicated heat ratings, but the lack of any anomalies in the gamma scans and the appearance of the sections indicate that the anticipated condition of central melting was not achieved although columnar grain growth is significant. Columnar grain growth has been shown to occur without melting in UO$_2$ [12] and in ThO$_2$-UO$_2$ in the ORR loop experiment L-1-B [7], but the probability of the effective thermal conductivity of the as-packed
Table 4a. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO\textsubscript{2}-UO\textsubscript{2} Fuel Rods: Description of Irradiation-Test Fuel Rods

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel Rod Identification</th>
<th>Type of Oxide</th>
<th>Total Uranium (wt.%)</th>
<th>Cladding</th>
<th>Oxide Particle Size</th>
<th>Vibrated Density (% TD)</th>
<th>Fuel Rod Dimensions (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ORR Loop</td>
<td>L-1-A</td>
<td>Sol-gel 26</td>
<td>5.35</td>
<td>304 Stainless Steel</td>
<td>c</td>
<td>85.2</td>
<td>54.61</td>
</tr>
<tr>
<td></td>
<td>L-1-B</td>
<td>Sol-gel 26</td>
<td>5.35</td>
<td>Zircaloy 2</td>
<td>c</td>
<td>84.1</td>
<td>54.61</td>
</tr>
<tr>
<td></td>
<td>L-1-C</td>
<td>Sol-gel 26</td>
<td>5.35</td>
<td>Zircaloy 2</td>
<td>c</td>
<td>84.1</td>
<td>54.61</td>
</tr>
<tr>
<td>ORR Poolside</td>
<td>03-5</td>
<td>Sol-gel D</td>
<td>2.50</td>
<td>304 Stainless Steel</td>
<td>d</td>
<td>84.8</td>
<td>17.80</td>
</tr>
<tr>
<td></td>
<td>03-6</td>
<td>Sol-gel D</td>
<td>2.50</td>
<td>304 Stainless Steel</td>
<td>d</td>
<td>85.5</td>
<td>17.78</td>
</tr>
<tr>
<td>ETR-I 43-80</td>
<td>18</td>
<td>Sol-gel 35-1</td>
<td>5.02</td>
<td>304 Stainless Steel</td>
<td>e</td>
<td>88.1</td>
<td>30.48</td>
</tr>
<tr>
<td></td>
<td>43-80</td>
<td>Sol-gel 35-2</td>
<td>5.02</td>
<td>304 Stainless Steel</td>
<td>e</td>
<td>89.4</td>
<td>30.48</td>
</tr>
<tr>
<td></td>
<td>43-81</td>
<td>Sol-gel 35-3</td>
<td>5.02</td>
<td>304 Stainless Steel</td>
<td>e</td>
<td>85.1</td>
<td>30.48</td>
</tr>
</tbody>
</table>

\textsuperscript{a}Enriched in \textsuperscript{235}U to 93%.
\textsuperscript{b}Based upon a calculated density depending on composition (approx 10.04 g/cm\textsuperscript{3}).
\textsuperscript{c}Distribution H: 60 wt \% -6 +16 mesh; 25 wt \% -50 +140 mesh; 15 wt \% -200 mesh.
\textsuperscript{d}Distribution A: 60 wt \% -10 +16 mesh; 15 wt \% -70 +140 mesh; 25 wt \% -200 mesh.
\textsuperscript{e}Distribution C: 60 wt \% -8 +16 mesh; 15 wt \% -70 +100 mesh; 25 wt \% -200 mesh.
Table 4b. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO2-UO2 Fuel Rods: Irradiation History

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel Rod Identification</th>
<th>Cladding Temperature (°C)</th>
<th>Average External Pressure (psia)</th>
<th>Exposure Time Reactor Full-Power Days</th>
<th>Peak Linear Heat Rating Time Average (W/cm)</th>
<th>Instantaneous (W/cm)</th>
<th>Peak Cladding Heat Flux Time Average (W/cm²)</th>
<th>Instantaneous (W/cm²)</th>
<th>Maximum Burnup a (Mwd/tonne (fissions/cm³))</th>
</tr>
</thead>
<tbody>
<tr>
<td>ORR Loop</td>
<td>L-1-A</td>
<td>260</td>
<td>1750</td>
<td>29.6</td>
<td>381</td>
<td>389</td>
<td>104</td>
<td>106</td>
<td>1,600 x 10^20</td>
</tr>
<tr>
<td></td>
<td>L-1-B</td>
<td>260</td>
<td>1750</td>
<td>29.6</td>
<td>499</td>
<td>509</td>
<td>136</td>
<td>139</td>
<td>2,100 0.45</td>
</tr>
<tr>
<td></td>
<td>L-1-C</td>
<td>260</td>
<td>1750</td>
<td>29.6</td>
<td>410</td>
<td>418</td>
<td>112</td>
<td>114</td>
<td>1,730 0.38</td>
</tr>
<tr>
<td>ORR Poolside</td>
<td>03-5</td>
<td>705</td>
<td>315</td>
<td>215.2</td>
<td>304</td>
<td>337</td>
<td>61</td>
<td>68</td>
<td>5,220 1.13</td>
</tr>
<tr>
<td></td>
<td>03-6</td>
<td>540</td>
<td>315</td>
<td>215.2</td>
<td>275</td>
<td>297</td>
<td>55</td>
<td>60</td>
<td>4,640 0.88</td>
</tr>
<tr>
<td>ETR-I</td>
<td>18</td>
<td>100</td>
<td>180</td>
<td>140.5</td>
<td>870</td>
<td>1050</td>
<td>304</td>
<td>367</td>
<td>20,400 4.28</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>100</td>
<td>180</td>
<td>140.5</td>
<td>865</td>
<td>1044</td>
<td>302</td>
<td>365</td>
<td>20,000 4.26</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>100</td>
<td>180</td>
<td>140.5</td>
<td>914</td>
<td>1103</td>
<td>319</td>
<td>385</td>
<td>22,000 4.46</td>
</tr>
</tbody>
</table>

aBased on 200 MeV/fission.
Table 4c. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO2-UO2 Fuel Rods: Postirradiation Observations and Calculations

<table>
<thead>
<tr>
<th>Fuel Rod</th>
<th>Time Average (W/cm)</th>
<th>Maximum Instantaneous (W/cm)</th>
<th>Release (%)</th>
<th>Dimensional Changes</th>
<th>Radial Extent of Microstructural Changes</th>
<th>Various Changes (W/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Tc</td>
<td>kdθ</td>
<td>T0</td>
<td>Tg</td>
<td>Teg</td>
<td>Tν</td>
</tr>
<tr>
<td>Experiment</td>
<td>Identification</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ORR Loop</td>
<td>L-1-A</td>
<td>46.5</td>
<td>47.4</td>
<td>2.3</td>
<td>+0.025</td>
<td>+0.102</td>
</tr>
<tr>
<td></td>
<td>L-1-B</td>
<td>57.0</td>
<td>58.1</td>
<td>Failure</td>
<td>0.000</td>
<td>+0.229</td>
</tr>
<tr>
<td></td>
<td>L-1-C</td>
<td>49.6</td>
<td>50.6</td>
<td>3.9</td>
<td>0.000</td>
<td>+0.102</td>
</tr>
<tr>
<td>ORR Poolside</td>
<td>03-5</td>
<td>54.5</td>
<td>60.5</td>
<td>18.3</td>
<td>-0.025</td>
<td>-0.229</td>
</tr>
<tr>
<td></td>
<td>06-5</td>
<td>46.7</td>
<td>50.4</td>
<td>Gas lost</td>
<td>0.000</td>
<td>+0.051</td>
</tr>
<tr>
<td>ETR-I</td>
<td>18</td>
<td>82.3</td>
<td>99.4</td>
<td>38.6</td>
<td>+0.025</td>
<td>+0.051</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>82.2</td>
<td>99.2</td>
<td>27.8</td>
<td>+0.037</td>
<td>+0.056</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>86.2</td>
<td>104.0</td>
<td>21.4</td>
<td>+0.027</td>
<td>+0.041</td>
</tr>
</tbody>
</table>

a: Time average value.
b: Sintering boundary not delineated.
FIG. 13. ETR group I, capsule 18: macroscopic views of selected specimen locations, as-cut

FIG. 14. ETR group I, capsule 10: macroscopic views of selected section locations, as-cut
oxides being high enough to permit this in these experiments at full power was unlikely. Examination of the reactor start-up on the first cycle of exposure of these rods shows that for the first 9½ h of exposure the reactor was never over 50% of full power. Since this would have produced an $\frac{1}{k_0}$ very close to the 54 W/cm defined by experiment L-1-B for columnar grain growth it is entirely possible that during this time sufficient sintering and grain growth occurred to raise the effective thermal conductivity, develop a central void, and prevent melting. Whether or not this is the case, it is apparent that at a linear heat rating in excess of 1000 W/cm the maximum performance capabilities of these vibratorily compacted sol-gel fuels have not been reached.

The calcining atmosphere appears to influence the fission-gas release rates. This conclusion will need verification from the lower rated rods in the MTR-III group which contain the same fuels but are still under irradiation. Other experiments at Chalk River [13] have shown an effect on grain growth in hydrogen-sintered and air-sintered ThO$_2$–UO$_2$ pellets when subjected to short-time rabbit-type irradiation tests, with the air-fired material showing more grain growth.

The microstructures on the ETR-I rods, shown in Figs. 16, 17, and 18, are similar to those developed in UO$_2$ fuel rods at high heat ratings.

Although the maximum performance characteristics have not been defined, it would appear that sufficient data are available for a preliminary comparison of ThO$_2$–UO$_2$ and UO$_2$ fuels. The most common method of comparison for bulk fuels of this type is the use of the $\frac{1}{k_0}$ values for specific
FIG. 16. Composite photomicrograph of vibratorily compacted sol-gel ThO$_2$-UO$_2$ calcined in air and irradiated to $4.28 \times 10^{20}$ fissions/cm$^3$ at a linear heat rating of approximately 1000 W/cm

FIG. 17. Composite photomicrograph of vibratorily compacted sol-gel ThO$_2$-UO$_2$ calcined in Ar-4% H$_2$ and irradiated to $4.26 \times 10^{20}$ fissions/cm$^3$ at a linear heat rating of approximately 1000 W/cm

FIG. 18. Composite photomicrograph of vibratorily compacted sol-gel ThO$_2$-UO$_2$ calcined in H$_2$ and irradiated to $4.46 \times 10^{20}$ fissions/cm$^3$ at a linear heat rating of approximately 1000 W/cm

microstructural changes [14]. A review of the very extensive literature on UO$_2$ experiments will show that there are considerable differences of opinion among the various investigators on the precise values of $k/d_0$ for various structural changes. These differences develop primarily from the treatment of the fuel-to-cladding conduction evaluation, the interpretation of the extent of melting, and the thermal conductivity value chosen for temperatures below about 500°C. The temperatures for equiaxed grain growth (1500°C) and columnar grain growth (1700°C) in UO$_2$ are, however, fairly well established but they have not been investigated for ThO$_2$-UO$_2$ fuels as yet.

Because of the differences in UO$_2$ experimental interpretations and the sparsity of data on ThO$_2$-UO$_2$ fuels an absolute comparison is impossible.
Table 5. Comparison of UO₂ and ThO₂-UO₂ Vibratorily Compacted Fuel Rods

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel Material (a)</th>
<th>Density (% TD)</th>
<th>Heavy Element</th>
<th>Inside Burn Up</th>
<th>Surface Temperature</th>
<th>Thermal Rating (W/cm)</th>
<th>Linear Heat Rating (W/cm)</th>
<th>Linear Heat Rating (W/cm)</th>
<th>Linear Heat Rating (W/cm)</th>
<th>Linear Heat Rating (W/cm)</th>
<th>85Kr Release (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ORR (c) 7N1</td>
<td>UO₂</td>
<td>87.1</td>
<td>-</td>
<td>4,780</td>
<td>353</td>
<td>434</td>
<td>32.5</td>
<td>20.7</td>
<td>12.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Loop 701</td>
<td>UO₂</td>
<td>87.1</td>
<td>-</td>
<td>5,140</td>
<td>361</td>
<td>465</td>
<td>34.8</td>
<td>21.1</td>
<td>47.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7F1</td>
<td>UO₂</td>
<td>86.9</td>
<td>-</td>
<td>6,290</td>
<td>383</td>
<td>566</td>
<td>41.2</td>
<td>20.0</td>
<td>72.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8N1</td>
<td>UO₂</td>
<td>85.6</td>
<td>-</td>
<td>4,810</td>
<td>347</td>
<td>404</td>
<td>30.9</td>
<td>22.8</td>
<td>17.5</td>
<td>77.0</td>
<td></td>
</tr>
<tr>
<td>8O1</td>
<td>UO₂</td>
<td>85.8</td>
<td>-</td>
<td>5,040</td>
<td>351</td>
<td>423</td>
<td>31.3</td>
<td>21.2</td>
<td>16.1</td>
<td>25.0</td>
<td></td>
</tr>
<tr>
<td>8P1</td>
<td>UO₂</td>
<td>85.5</td>
<td>-</td>
<td>6,880</td>
<td>387</td>
<td>578</td>
<td>40.8</td>
<td>23.9</td>
<td>20.3</td>
<td>25.0</td>
<td></td>
</tr>
<tr>
<td>ORR L1A</td>
<td>ThO₂-5% UO₂</td>
<td>85.2</td>
<td>-</td>
<td>1,600</td>
<td>341</td>
<td>381</td>
<td>30.3</td>
<td>No Void</td>
<td>20.5</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td>Loop L1B</td>
<td>ThO₂-5% UO₂</td>
<td>84.1</td>
<td>-</td>
<td>2,100</td>
<td>368</td>
<td>499</td>
<td>39.7</td>
<td>No Void</td>
<td>36.6</td>
<td>17.7</td>
<td></td>
</tr>
<tr>
<td>1LC</td>
<td>ThO₂-5% UO₂</td>
<td>84.1</td>
<td>-</td>
<td>1,730</td>
<td>348</td>
<td>410</td>
<td>32.6</td>
<td>No Void</td>
<td>20.7</td>
<td>3.9</td>
<td></td>
</tr>
<tr>
<td>ETR-I 18</td>
<td>ThO₂-5% UO₂</td>
<td>88.1</td>
<td>-</td>
<td>20,400</td>
<td>211</td>
<td>870</td>
<td>63.0</td>
<td>35.8</td>
<td>34.7</td>
<td>38.0</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>ThO₂-5% UO₂</td>
<td>89.4</td>
<td>-</td>
<td>20,000</td>
<td>209</td>
<td>865</td>
<td>66.4</td>
<td>24.6</td>
<td>19.8</td>
<td>28.0</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>ThO₂-5% UO₂</td>
<td>85.1</td>
<td>-</td>
<td>22,000</td>
<td>218</td>
<td>914</td>
<td>61.3</td>
<td>28.2</td>
<td>25.3</td>
<td>21.0</td>
<td></td>
</tr>
</tbody>
</table>

(a) UO₂ fuel was arc-fused crushed and vibratorily compacted; oxygen-to-uranium ratio 2.002:2.003; UO₂ enriched 5 to 6%. ThO₂-UO₂ fuel was sol-gel material crushed and vibratorily compacted; UO₂ enriched 93%.

(b) $\int_c^v$ is center to surface of fuel; $\int_s^v$ is void to surface of fuel; $\int_s^{cg}$ is the limit of columnar grain growth to surface of fuel; and $\int_s^{eg}$ is the limit of discernible equiaxed grain growth to surface of fuel.

However at ORNL vibratorily compacted fuel rods of both UO$_2$ and ThO$_2$-6% UO$_2$ have been tested in the pressurized loop at essentially identical conditions. The UO$_2$ work has been reported [15-16] and the data listed in Table 5 were derived from these reports. All $j/kd\theta$ values are quoted to the fuel surfaces which should have been at essentially the same temperature.

The most obvious difference is the lack of void formation in the ThO$_2$-UO$_2$ loop rods at $j/kd\theta$ values where significant voids were formed in the UO$_2$ rods. To illustrate void formation in ThO$_2$-UO$_2$ the ETR-I rods which operated at lower surface temperatures but at higher power levels and to much higher burn-ups have been included in this table. The extent of columnar grain growth is also fairly well defined in post-irradiation microstructures and again the $j/kd\theta$ values show the ThO$_2$-UO$_2$ material to be better in the comparable loop exposures. The extent of equiaxed grain growth or sintering is difficult to determine in these powder compacts but it would appear that similar temperatures and $j/kd\theta$ values apply to ThO$_2$-UO$_2$ and UO$_2$ fuels.

Any attempt to place a quantitative value on the superiority of ThO$_2$-UO$_2$ compacts from these data would be highly speculative. However, Rao [13] at Chalk River has compared 3-min rabbit tests of UO$_2$ and ThO$_2$-UO$_2$ using pressed and sintered pellets. He has concluded that similar microstructural changes in ThO$_2$-1% UO$_2$ compared with UO$_2$ will require a 10% higher power output. Such short-time tests have proven to be good guides in the past and from the data reported here it appears that his estimate may prove to be conservative. Irradiation tests at higher linear heat ratings are planned and additional out-of-pile data are being obtained including thermal simulation and thermal conductivity measurements. These out-of-pile tests are needed to remove some of the uncertainties associated with evaluation of the microstructures from in-pile tests.

**Mixed progeny and remotely fabricated sol-gel ThO$_2$-233UO$_2$ fuel tests**

We have done very little testing with mixed progeny fuels but three fuel rods containing sol-gel ThO$_2$ mixed with PuO$_2$ before calcining and tamp-packed into fuel rods have been examined after exposure at linear heat ratings up to 245 W/cm to burn-ups of 29 000 MWd/t (Th+Pu). The fission-gas release rates were less than 5% and the microstructures were similar to ThO$_2$-UO$_2$ exposed under the same conditions.

The semi-remotely fabricated, vibratorily compacted rods containing sol-gel ThO$_2$-233UO$_2$ (ETR Group III) have been in the ETR for four months and appear to be performing satisfactorily.

**Summary**

A rather extensive series of irradiation tests over the past five years has shown that sol-gel vibratorily compacted ThO$_2$-5% UO$_2$ fuel rods perform as well as similar arc-fused ThO$_2$-UO$_2$ fuel rods and rods containing pressed and sintered pellets of ThO$_2$-UO$_2$ at burn-ups up to 80 000 MWd/t. There has been no evidence of breakaway swelling or sudden increases in fission-gas release in these rods at linear heat ratings between 300 and 350 W/cm ($j/kd\theta$ 32-38 W/cm).
Although the maximum performance characteristics have not been determined, sol-gel ThO$_2$-5% UO$_2$ vibratory compacted fuels have been operated at linear heat ratings of 1000 W/cm (kWd of 100 W/cm) to burn-ups over 20,000 MWD/t with no evidence of swelling, some void formation, but no central melting and with fission-gas release rates less than 30%

It appears that ThO$_2$-base fuels with low UO$_2$ contents will permit significantly higher power densities than similar metal-clad UO$_2$ fuels and that vibratory compaction is a satisfactory rod fabrication technique.

**THORIUM-CONTAINING PYROLYTIC-CARBON-COATED PARTICLE FUELS**

Several irradiation tests of thorium-bearing fuels in the form of carbides and oxides have been conducted in support of high-temperature gas-cooled reactor design requirements. The fuel has all been in the form of small particles coated with pyrolytic carbon for fission-product retention; both loose beds of particles and graphite matrix elements have been irradiated. Most ORNL irradiations of HTGR fuels to date have utilized high concentrations of uranium to obtain accelerated burn-up and experience with thorium is limited. Much of the testing of thorium-bearing fuels has been related to pebble-bed reactor application, first for the PBRE concept [17] and later for the German AVR [18]. Some recent tests have been in support of the TARGET [19] reactor fuel concept.

**Coated-particle capsule irradiation tests**

In the development of these fuels it has proven desirable first to evaluate the particles separately and then the fuel element assembly. Significant results of irradiation tests on unsupported coated particles at ORNL have been described in detail [20, 21], except for the results of the more recent tests involving coated oxide particles. The test facilities have included static capsules inserted in the Low-Intensity Test Reactor and the Materials Test Reactor, sweep capsules in the Oak Ridge Research Reactor, and, for coated oxide particles only, the ORR loop No 1 facility [3].

The results of static tests have generally corroborated those of sweep tests. Results from sweep facilities are rather emphasized in this discussion because in these tests a continuous account of the behaviour of the test material is provided by analysis of the helium gas that is passed through the capsule. This gas is monitored continuously for total activity and sampled periodically for fission-gas analysis. Descriptions of the coated particles, the techniques used, and their characterization in coating application have been reported in detail [22-25]. Information on performance tests involving multilayer coatings on uranium and uranium-thorium is listed in Table 6 and the characterization of coatings in Table 7. The results are summarized below.

Coated carbide particles

The following conclusions were drawn from extensive testing of coated, high-density UC$_2$ and (Th, U)C$_2$ fuel particles with nominal particle diam-
etters of 200 μm, (1) multilayer coatings are superior to the monolithic coatings tested; (2) the performance of monolithic coatings with average thicknesses of 50 to 75 μm is substantially inferior to that for 100-μm-thick coatings; (3) spheroidal particles generally perform better than irregularly shaped particles with sharp edges and protrusions; (4) in experiments in which no coatings have ruptured during irradiation, the fission-gas release, expressed as the ratio of the release rate to the birth rate (R/B) for \(^{88}\text{Kr}\), has been of the order of \(10^{-5}\) to \(10^{-7}\) and can be accounted for by the uranium contamination in the coatings as determined before irradiation by alpha-counting; and (5) the performance to substantial burn-up of coated (Th, U)\(\text{C}_2\) particles with thorium to uranium ratios up to 2:1:1 has been comparable to that of similarly coated \(\text{UC}_2\) particles.

Coated oxide particles

The highest burn-up demonstrated, at this writing, for loose coated oxide particles is 12 at.% heavy metal at 1400°C in a sweep capsule (experiment A9-2). This test has operated for about 9 weeks and, by comparison with results from other tests in this type of facility, the fission-gas release has been constant and very low. Thermocouple failure occasioned termination of a previous sweep capsule test (experiment B9-20) on coated \(\text{UO}_2\) particles after a burn-up of 4 2 at.% heavy metal at 1370°C. None of the coatings were ruptured in this test as the result of irradiation, and metallographic examination revealed only minor damage by wedge-shaped fractures, none of which extended beyond one-half the thickness of the inner coating. Sol-gel (Th, и)\(\text{O}_2\) particles containing 8% \(\text{UO}_2\) and coated with a two-layer coating have been irradiated at 1200°C to a short burn-up in experiment B9-19 and for an extended time in the ORR loop No 1.

Typical photomicrographs of unirradiated and irradiated particles of both uranium and uranium-thorium fuels are shown in Figs. 19 and 20.

**Fuelled graphite irradiation performance**

Sweep capsule tests

Evaluation of a reactor fuel element must take into account irradiation test performance. Although it is desirable to test in actual power-reactor service, no suitable facility yet exists for in-service testing of H1'GR fuels. The irradiation capsule and loop tests now employed in evaluating spherical fuel elements offer advantages for instrumentation and control and for measurement of fission-gas release as a function of irradiation experience. Most ORNL capsule irradiations have been conducted in the ORR Poolside Facility, which provides the reference conditions, including fission power density, temperature, and a sweep atmosphere of helium. Other capsules in core positions have been used to obtain high burn-up. The design of the poolside irradiation capsule is shown in the schematic diagram of Fig. 21 and the irradiation test facilities have been described in detail [3].

Specially equipped hot cells are used for examination of the spheres following irradiation. The examinations include a gross gamma scan, visual and photographic observations, dimensional measurements, crushing and
Table 6. Irradiation Test Conditions and Fission-Gas Release Data for Unsupported Coated Fuel Particles with Multilayer Pyrolytic-Carbon Coatings

<table>
<thead>
<tr>
<th>Experiment Designation</th>
<th>Sample Type of Coating</th>
<th>Fuel Structure</th>
<th>Temperature (°C)</th>
<th>Burnup (at. % heavy metal)</th>
<th>Ratio of R/B for ( ^{88}\text{Kr} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1-11</td>
<td>GA-310</td>
<td>(Th,U)C2</td>
<td>1200</td>
<td>14.7</td>
<td>( 3.6 \times 10^{-6} )</td>
</tr>
<tr>
<td>B9-10</td>
<td>NCC-AD</td>
<td>UC2</td>
<td>1150</td>
<td>2.6</td>
<td>( 2.4 \times 10^{-6} )</td>
</tr>
<tr>
<td>B9-11</td>
<td>NCC-208-2</td>
<td>(Th,U)C2(d)</td>
<td>1150</td>
<td>2.6</td>
<td>( 1.3 \times 10^{-6} )</td>
</tr>
<tr>
<td>B9-14</td>
<td>GA-309</td>
<td>UC2</td>
<td>1150</td>
<td>2.6</td>
<td>( 1.6 \times 10^{-3} )</td>
</tr>
<tr>
<td>C1-15</td>
<td>GA-314</td>
<td>(Th,U)C2(e)</td>
<td>1370</td>
<td>18.7</td>
<td>( 4.0 \times 10^{-3} )</td>
</tr>
<tr>
<td>B9-15</td>
<td>GA-309</td>
<td>UC2</td>
<td>1370</td>
<td>18.7</td>
<td>( 1.0 \times 10^{-3} )</td>
</tr>
<tr>
<td>B9-16</td>
<td>NCC-216</td>
<td>UC2</td>
<td>1370</td>
<td>18.8</td>
<td>( 1.0 \times 10^{-3} )</td>
</tr>
<tr>
<td>B9-17</td>
<td>GR-138</td>
<td>UC2</td>
<td>1370</td>
<td>14.9</td>
<td>( 1.0 \times 10^{-3} )</td>
</tr>
<tr>
<td>B9-19</td>
<td>GR-206</td>
<td>(Th,U)O2(d)</td>
<td>1200</td>
<td>14.9</td>
<td>( 2.3 \times 10^{-6} )</td>
</tr>
<tr>
<td>B9-20</td>
<td>GR-201</td>
<td>UO2</td>
<td>1370</td>
<td>4.2</td>
<td>( 1.5 \times 10^{-3} )</td>
</tr>
<tr>
<td>A9-2</td>
<td>OR-298</td>
<td>UO2</td>
<td>1370</td>
<td>12.0(1)</td>
<td>( 2.0 \times 10^{-7} )</td>
</tr>
<tr>
<td>Loop 1-14(j)</td>
<td>OR-206</td>
<td>(Th,U)O2(e)</td>
<td>1370</td>
<td>2.7(1)</td>
<td>( 2.5 \times 10^{-5} )</td>
</tr>
</tbody>
</table>


\(^b\)Periodic thermal cycles were applied between 925 and 1315°C.


\(^d\)Thorium-to-uranium ratio 0.6:1.0.

\(^e\)Thorium-to-uranium ratio 2.1:1.0.


\(^g\)Particles contain 8 wt % UO2 and were prepared by the ORNL sol-gel process.

\(^h\)Test terminated due to thermocouple failure.

\(^i\)Test still in progress.

\(^j\)This test is described in a later section of this report. About 9 months of irradiation was accumulated at conditions indicated.
Table 7. Characterization of OR-2D Coatings Deposited from Methane and Listed in Table 6.

<table>
<thead>
<tr>
<th>Sample Designation</th>
<th>OR-138</th>
<th>OR-182</th>
<th>OR-201</th>
<th>OR-205</th>
<th>OR-206</th>
<th>OR-298</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type of Fuel Particle</td>
<td>UC₂</td>
<td>(Th,U)O₂</td>
<td>UO₂</td>
<td>ThO₂</td>
<td>(Th,U)O₂</td>
<td>UO₂</td>
</tr>
<tr>
<td>Average Fuel-Particle Diameter, μ</td>
<td>186</td>
<td>217</td>
<td>229</td>
<td>243</td>
<td>206</td>
<td>302</td>
</tr>
<tr>
<td>Inner Layer</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coating temperature, °C</td>
<td>1400</td>
<td>1400</td>
<td>1400</td>
<td>1400</td>
<td>1400</td>
<td>1400</td>
</tr>
<tr>
<td>CH₄ flow rate, cm³ min⁻¹ cm⁻²</td>
<td>0.17</td>
<td>0.15</td>
<td>2.8</td>
<td>0.83</td>
<td>0.83</td>
<td>0.16</td>
</tr>
<tr>
<td>Coating thickness, μ</td>
<td>40</td>
<td>55</td>
<td>40</td>
<td>50</td>
<td>35</td>
<td>70</td>
</tr>
<tr>
<td>Outer Layer</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coating temperature, °C</td>
<td>1700</td>
<td>1800</td>
<td>1800</td>
<td>1800</td>
<td>1800</td>
<td>1900</td>
</tr>
<tr>
<td>CH₄ flow rate, cm³ min⁻¹ cm⁻²</td>
<td>0.17</td>
<td>0.33</td>
<td>0.27</td>
<td>0.17</td>
<td>0.17</td>
<td>0.10</td>
</tr>
<tr>
<td>Coating thickness, μ</td>
<td>72</td>
<td>45</td>
<td>80</td>
<td>80</td>
<td>63</td>
<td>65</td>
</tr>
</tbody>
</table>
FIG. 19. Pyrolytic-carbon-coated $\text{UO}_2$ particles from batch OR-201. (a) Unirradiated. $\times$100. (b) Irradiated to 4.2 at.\% U burn-up at 1370°C in capsule B9-20. As-polished. $\times$200
FIG. 20. Pyrolytic-carbon-coated (Th,U)O$_2$ particles from batch OR-182. (a) Unirradiated. x250. (b) Irradiated to 0.25 at. % heavy metal burn-up at 1200°C in capsule 0.1-8. x200. Note the bright metallic-appearing fragments at the interface and within the inner coating after irradiation. As-polished
FIG. 21. ORR Poolside capsule for testing 8-cm-diam. fuelled-graphite spheres
impact strength measurements, burn-up determinations, metallography, and radiochemical analysis of parts from the graphite structures. The apparatus and techniques used for the examinations were described in a recent publication [26].

Irradiation tests of spherical fuel elements such as that shown in Fig. 22 have been conducted principally to investigate the effect of fabrication variables. These findings have been summarized recently [27], and have been reported in greater detail in ORNL progress reports [28-31].

Operating conditions and fission-gas release during irradiation for the spheres used in several tests and representative of both uranium and thorium-uranium elements are given in Table 8. It is notable that only one sphere showed a ratio of fission-gas release rate to birth rate, R/B, significantly in excess of 10⁻³. This sphere, GA-VS11-2, fabricated as shown in Table 8 showed evidence of excessively high temperatures in the matrix and probably operated at a much higher temperature than was estimated because of lack of bonding and the development of a gap, which was observed in post-irradiation examination. Relative fractional release of noble fission gases is illustrated in Fig 23, which shows little difference between the moulded and machined shells. A somewhat wider variation is apparent for different elements having machined shells, however, this probably indicates variation in the effectiveness of bonding between the matrix and shell. The shell bonding difficulty may not be inherent in the design, since these were all experimental elements, but this does represent an important problem area. Sphere 3M-VS16 showed, at the start, very low fission-gas release but the release increased steadily with burn-up. This behaviour is attributed to progressive failure of monolithic-laminar coatings with increasing burn-up (see above).

No significant difference is apparent for the fission-gas release behaviour of uranium and thorium fuels. These release rates are considered quite satisfactory for pebble-bed reactor service. Observations concerning release of solid fission products have been more limited, but again no difference has been observed between uranium and thorium-bearing coated particle fuels.

In-pile loop test of thorium-uranium oxide pyrolytic-carbon-coated particle fuel element

Perhaps the most significant test by ORNL of thorium-bearing fuel for HTGR reactors has been conducted in a recirculating loop, ORR No. 1 [3]. The irradiation assembly designated as experiment 14 contains two cylindrical fuelled-graphite elements with loose coated particles. This type of fuel element is of interest for advanced reactor designs [19].

The design of the fuel elements for experiment 14 is shown in Fig 24. The two cylindrical fuel elements are aligned one above the other in the loop. They differ in that the coated particles in the upper element are contained within an annulus between a tube and a rod of type ATJ graphite. The lower element consists of a solid graphite rod with five longitudinal holes containing loose coated particles located symmetrically around the axis of the rod. The lower element is contained within a sealed stainless steel capsule and the upper element is vented to the helium coolant in the loop via a porous
Table 8. Operating Conditions and Fission-Gas Release Data for 6-cm-diam OBR-Irradiated Fuel Spheres Containing Coated Carbide Fuel Particles, Poolside Capsules

<table>
<thead>
<tr>
<th>Type of Shell</th>
<th>Shell Thickness (cm)</th>
<th>Insert Fabrication</th>
<th>Unfueled Shell</th>
<th>Particle Type</th>
<th>Coating metal</th>
<th>Burnup (at. %)</th>
<th>Power Time (days)</th>
<th>Average Power Density (W/cm³)</th>
<th>Operating Temperature, °C</th>
<th>Average Temperature, °C</th>
<th>R/B</th>
<th>86Kr x 10⁻⁴</th>
</tr>
</thead>
<tbody>
<tr>
<td>Machined GA-V311</td>
<td>1.1</td>
<td>Hot pressed at 750°C, baked at 1800°C</td>
<td>Type ATJ</td>
<td>(Th,U)C₂</td>
<td>Triplex</td>
<td>3.2</td>
<td>27</td>
<td>30.0</td>
<td>930</td>
<td>1370</td>
<td>1 to 43b</td>
<td>1 to 43b</td>
</tr>
<tr>
<td>GA-VS15</td>
<td>0.8</td>
<td>Hot pressed at 750°C, baked at 1800°C</td>
<td>Type ATJ</td>
<td>(Th,U)C₂</td>
<td>Triplex</td>
<td>2.0</td>
<td>92</td>
<td>18.6</td>
<td>910</td>
<td>Not determined</td>
<td>2.5</td>
<td>6.0</td>
</tr>
<tr>
<td>GA-VS16a</td>
<td>0.8</td>
<td>Warm pressed at 100°C, baked at 1800°C</td>
<td>Type ATJ</td>
<td>(Th,U)C₂</td>
<td>Triplex</td>
<td>2.3</td>
<td>96</td>
<td>18.6</td>
<td>920</td>
<td>1370</td>
<td>2.0</td>
<td>4.0</td>
</tr>
<tr>
<td>NCL-VS1</td>
<td>1.1</td>
<td>Mold injection baked at 1400°C</td>
<td>Type ATJ</td>
<td>(Th,U)C₂</td>
<td>Duplex</td>
<td>1.5</td>
<td>46c</td>
<td>18.6</td>
<td>920</td>
<td>1320</td>
<td>2.0</td>
<td>4.0</td>
</tr>
<tr>
<td>ORNL-VS3</td>
<td>0.8</td>
<td>Warm pressed at 150°C, baked at 1000°C</td>
<td>Type ATJ</td>
<td>UC₂</td>
<td>Duplex</td>
<td>1.2</td>
<td>62</td>
<td>16.0</td>
<td>850</td>
<td>1180</td>
<td>0.2</td>
<td>0.3</td>
</tr>
<tr>
<td>Molded GA-VS12d</td>
<td>0.63</td>
<td>Warm pressed at 150°C</td>
<td>Not known</td>
<td>(Th,U)C₂</td>
<td>Triplex</td>
<td>4.3</td>
<td>146</td>
<td>30.0</td>
<td>920</td>
<td>Not determined</td>
<td>3.0</td>
<td>2.5</td>
</tr>
<tr>
<td>GA-VS12d</td>
<td>0.63</td>
<td>Warm pressed at 750°C, baked at 1800°C</td>
<td>Not known</td>
<td>(Th,U)C₂</td>
<td>Triplex</td>
<td>2.7</td>
<td>87</td>
<td>30.0</td>
<td>940</td>
<td>Not determined</td>
<td>2.0</td>
<td>2.5</td>
</tr>
<tr>
<td>3M-VS16b</td>
<td>0.63</td>
<td>Not known</td>
<td>Not known</td>
<td>(Th,U)C₂</td>
<td>Laminar</td>
<td>3.0</td>
<td>140</td>
<td>18.6</td>
<td>910</td>
<td>1270</td>
<td>0.01 to 0.01</td>
<td>1a</td>
</tr>
</tbody>
</table>

a R/B increased steadily throughout irradiation.
b Similar spheres.
c Test terminated by leak in capsule.
d Similar spheres.

OLSEN et al.
IRRADIATION BEHAVIOUR OF Th-U ALLOYS

SHELL PLUG WITH CARBON CEMENTED JOINT

THORIUM-URANIUM CARBIDE (200 MICRON DIAM)

PYROLYTIC CARBON COATING (100 MICRON THICKNESS)

COATED FUEL PARTICLES IN GRAPHITE MATRIX

MACHINED AT GRAPHITE SHELL

FIG. 22. Schematic of a typical coated-particle fuel element

$R/B$ Values of $^{88}$Kr and $^{133}$Xe for 6-cm-diam Fuel Spheres
Irradiated in ORR Poolside Facility

FIG. 23. $R/B$ values of $^{88}$Kr and $^{133}$Xe for 6-cm-diam fuel spheres irradiated in ORR Poolside Facility
FIG. 24. Fuel assembly for experiment 14 in ORR loop No. 1
nickel plug. The upper capsule is instrumented with nine thermocouples, one of which is located at the axial centre of the fuel. The lower capsule is uninstrumented, but the heat generation rate is so low that its temperature is not much higher than the outlet gas temperature so no instrumentation is necessary. The stainless steel container is required to protect the fuel element from damage during insertion into and removal from the loop.

The fuel used in experiment 14 is comprised of a mixture of two batches of pyrolytic-carbon-coated particles fabricated at ORNL and designated as OR-205 and -206. The particles for both batches were prepared by the sol-gel technique. The particles in batch 205 consist of pure ThO₂, whereas those in batch 206 contain approximately 8 wt.% UO₂ (93% ²³⁵U). A detailed description of these coated particles is given in Table 7. The mixture was prepared by blending 53.415 g from batch OR-206 with 10.551 g from batch OR-205; it was then poured into the individual compartments.

The fuel burn-up in the (Th, U)O₂ particles in the upper vented region is estimated to be 2.7 at.% heavy metal after approximately 9 months of operation. Since initially only 7.2% of the heavy metal in these particles was ²³⁵U, the ²³⁵U consumption, including fission and conversion to ²³⁶U, is 44 at.%. Burn-up of fuel in the lower, sealed element which contains the same fuel but was in a lower neutron flux is estimated to be 2.2 at.% heavy metal.

The temperature in the upper region has been maintained at 1370°C in the centre of the fuel and 1040°C in the surrounding graphite fuel support tube. The fuel temperature of the uninstrumented lower region is estimated to be 700°C.

Equilibrium concentrations of noble gas fission products in the loop coolant have been nearly constant and indicate R/B values for the upper region of approximately:

\[
\begin{align*}
{^{88m}}Kr & : 1.9 \times 10^{-5} \\
{^{87}}Kr & : 1.0 \times 10^{-5} \\
{^{88}}Kr & : 2.5 \times 10^{-5} \\
{^{135}}Xe & : 1.5 \times 10^{-5} \\
{^{133}}Xe & : 1.4 \times 10^{-5}
\end{align*}
\]

The contaminant gases CO, CO₂, and H₂ have tended to build up slowly but continuously in the loop coolant, although the rate has decreased with time. The contaminants have been controlled by replacement of the coolant about once every 3 to 4 weeks when the concentration of CO and CO₂ reached approximately 125 and 75 ppm by volume, respectively. Approximately 30% Ne was added to the helium to lower the thermal conductivity of the coolant. This has made it possible to maintain the central fuel temperature in the vented region at 1370°C despite the high consumption of ²³⁵U.

The element has been removed from the loop for post-irradiation examination, but has not yet been opened. It is significant that the residual radioactivity of the loop did not increase measurably over the period of this test, as determined by external gamma scanning. Analysis of samples from exposed surfaces near the compressor and on the fuel element support also indicate very low activity from metallic fission products.
Table 9. Description of Fueled Spheres and Average Operating Conditions for Eight-Ball Irradiation Capsule FI-8B-5

<table>
<thead>
<tr>
<th>Position</th>
<th>Sphere Designation</th>
<th>Fuel Loading</th>
<th>Unfueled Shell Thickness</th>
<th>Average Temperature Center</th>
<th>Estimated Burnup</th>
<th>Fabricators</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Total U, U 235, Th</td>
<td>(g) (g) (g)</td>
<td>(°C)</td>
<td>(at. %) (Heavy Metal) (Fissions/cm³)</td>
<td></td>
</tr>
<tr>
<td>1 (top)</td>
<td>3M-S13-10</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.25 molded</td>
<td>973 c</td>
</tr>
<tr>
<td>2</td>
<td>GA-S14-2A</td>
<td>1.46</td>
<td>0.28</td>
<td>6.27</td>
<td>0.20 ATJ</td>
<td>869</td>
</tr>
<tr>
<td>3</td>
<td>NCF-S24-L3</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.00</td>
<td>902</td>
</tr>
<tr>
<td>4</td>
<td>3M-S13-7</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.20 ATJ</td>
<td>886</td>
</tr>
<tr>
<td>5</td>
<td>NCF-S25-V10</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.00</td>
<td>854</td>
</tr>
<tr>
<td>6</td>
<td>3M-S13-14e</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.25 molded</td>
<td>841</td>
</tr>
<tr>
<td>7</td>
<td>GA-S14-3A</td>
<td>1.46</td>
<td>0.28</td>
<td>6.27</td>
<td>0.20 ATJ</td>
<td>808</td>
</tr>
<tr>
<td>8 (bottom)</td>
<td>3M-S13-9</td>
<td>1.58</td>
<td>0.29</td>
<td>1.00</td>
<td>0.20 ATJ</td>
<td>773</td>
</tr>
</tbody>
</table>

Additional Notes:

a) Fabricators: GA, General Atomic Division of General Dynamics; 3M, the 3M Company; NCF, Carbon Products Division of Union Carbide.

b) Calculated on basis of fueled-core volume.

c) Only the top sphere had a central thermocouple.

d) The shell of this sphere was cracked prior to irradiation by dropping 24 times from a height of 4 meters on to a bed of 1.5-in.-diam graphite spheres.

e) This sphere was dropped 25 times without cracking under the same conditions as (d).
Eight-ball static capsule test

The eight-ball capsule contains eight 1.5-in.-diam spheres placed inside individual graphite containers and assembled inside a graphite tube with end insulators. The graphite tube assembly is inserted in stainless steel primary and secondary containment tubes; this tube assembly makes up the capsule which is tested in the F-1 position of the ORR core. A fifth and final test of this type, F1-8B-5, has been irradiated to higher fuel burn-up than any other ORNL test of thorium-uranium carbide HTGR fuel elements. The fuelled spheres used in this experiment are described in Table 9. Heavy metal fuel burn-up in this capsule was approximately 25 at.% with approximately 45% for $^{235}$U during 9 months exposure in the ORR. None of the spheres showed visible damage; overall linear shrinkage for spheres having machined shells was approximately 1% with approximately 2% for moulded shells and elements. Observation of the coated particles indicates performance about as expected from previous high burn-up tests of uranium carbide particles.

Summary

Limited test data for thorium-uranium HTGR fuels at low thorium concentration or low burn-up show no observable difference from uranium fuel performance for comparable irradiations. Overall performance for these coated particle fuels has been favourable when compared with reactor service requirements where release to birth ratio (R/B) of $10^{-4}$ for noble gases has been considered acceptable. Both thorium-uranium carbide and oxide fuels have been tested; results for oxides, although limited, have been excellent.

THORIUM-BASE METAL FUELS

The potential advantage of high fuel density available with metallic fuels has long been recognized, but has not been exploited for power reactors because fuel swelling and growth phenomena can cause fuel element failures at relatively low burn-up. As part of the studies on the use of nuclear reactors for desalination, a review of the available irradiation data on uranium, thorium, and their alloys has been made. Recent developments in metallic fuel technology, the lower fuel temperatures of interest in such reactors; the low fuel cycle costs for metallic fuel, and probable fabrication economies with large reactor complexes suggest that metallic fuels should again be considered.

There has been very little work reported on thorium and thorium alloy irradiations. However, several experiments on thorium metal have indicated that it has considerably greater dimensional stability during irradiation than uranium. Thorium, which has an isotropic body-centred cubic crystal structure, is not subject to growth effects and the attendant internal stresses that develop in the anisotropic orthorhombic crystals of uranium.

Early data on thorium irradiation experiments have been collected by Bauer et al. [32]. An Argonne National Laboratory investigation of thorium-uranium alloys has been summarized by Kittel et al. [33], who irradiated
small specimens (0.14-in. diam × 0.875 in. long) of a number of thorium-uranium alloys at temperatures up to 1000°C and burn-ups to 10 at.%. Swelling for all alloys increased from approximately 1% per at.% burn-up at lower temperatures to about 2.5% per at.% burn-up at 650°C. At higher temperatures the swelling rate increased somewhat, reaching a value of 6% per at.% burn-up at 800°C. Volume increases were linear with burn-up and independent of uranium content. The data are plotted in Fig 25. Specimens containing in excess of 25% U became warped and distorted, but those containing 20% U, or less, did not show significant distortion or surface roughness. In a few cases, excessive swelling was observed as a result of locally high temperatures at the top end of the specimen. The authors attribute the excellent behaviour to the fact that chill-cast specimens were used. Uranium particles thus were very thin so that fission products were largely trapped in the thorium matrix.

Data on the swelling of specimens of cast and swaged Th-11 wt.% U irradiated to 1.5 at.% burn-up at temperatures up to 650°C were reported by Battelle [34] and by Atomics International [35]. Specimens irradiated below 590°C decreased in density at the rate of 3% per at.% burn-up. Specimens irradiated at higher temperatures showed greater density decreases even at burn-ups as low as 0.2%, but evidence of considerable overheating was also observed. Slugs of Th-7.5 wt.% U, 3/4 in. in diameter and clad in stainless steel with a 0.025-in.-thick annulus of sodium, were used as fuel in Core II of the Sodium Reactor Experiment [36]. The maximum coolant outlet temperature was 490°C and maximum fuel central temperatures were near 650°C. Thorium fuel elements were operated to exposures of 5260 MWd/t, and satisfactory operation was achieved [37], but no data on examination of the fuel are yet available.
Hanford is irradiating three 8-in.-long test elements of Zircaloy-2 clad annular tubes (1.75 in. OD, 1.05 in. ID) of Th-232 wt.% U-238 wt.% Zr alloy in the ETR [38] high-temperature pressurized-water loop. Preliminary data at the maximum measured exposure of 9300 MWD/t show an 0.9% volume increase. Maximum fuel operating temperatures have ranged between 460 and 585°C, and specific power between 45 and 69 W/g [39]. The irradiations are being continued.

From these data it is apparent that thorium metal fuels are not subject to the excessive "cavitation" swelling reported for uranium metal in the 400 to 550°C temperature range [40-42]. However, the effects of fabrication variables and alloying or dispersion hardening are not known. In addition, the fuel temperature at which the $\% \Delta V$/at $\%$ burn-up increases rapidly needs to be defined. The compatibility of the metallic fuels with possible lower cost cladding materials must also be investigated. In general the successful utilization of metallic fuels, particularly thorium-base fuels, appears feasible, but their performance will depend on the adequate control of a number of variables. Further research and development is required to define the requirements necessary to achieve optimum performance.

ACKNOWLEDGEMENTS

The efforts of many people are represented in the work reviewed in this paper, and it is impossible to give full credit to each. In the bulk oxide fuel irradiations programme special note is due to J.W. Ullmann and S.D. Clinton of the Chemical Technology Division for pre-irradiation planning and calculations and from the Metals and Ceramics Division to W.S. Ernst, J.W. Tackett and R.W. McClung for fuel rod fabrication, welding, and non-destructive testing; E.J. Manthos for metallography; Y. Hirose, a foreign visitor from the Hitachi, Ltd., Japan, for post-irradiation calculations, and to W.C. Thurber and S.A. Rabin who supervised most of the early experiments. In the coated particle fuel irradiation programme three Laboratory divisions are involved. H.C. McCurdy, in the Reactor Division, G.M. Watson, in the Reactor Chemistry Division, and P. Patriarca, in the Metals and Ceramics Division, supervised most of the recent effort. Special note is due to R.L. Beatty for coating studies, F.L. Carlsen for evaluation of spheres, C.O. Smith for impact studies, O. Sisman and F.R. McQuilkin for irradiation tests, J.L. Scott for thermal conductivity studies, and E.L. Long for metallography.

REFERENCES


INTRODUCTION

Reactor fuels based on thorium or its compounds are important for fuel economy in advanced converters and breeders. The development of such fuels requires considerable physical, mechanical, and other data on potential materials. Our purpose has been to survey the literature on these materials, report selected data, and indicate the areas where further data are needed.

THORIUM AND ITS ALLOYS

Although thorium metal has potential application as a nuclear fuel base, up to now it has been used only as the Core II loading for the Sodium Reactor Experiment. It has been proposed for some power reactors, and considerable development on thorium metallurgy and properties has been carried out. The results of this research have been summarized in several reviews [1-4], the most comprehensive have covered available data up to the period of about 1958 to 1960. We have examined some of the more recent data on thorium and its alloys as an aid in planning additional research directed toward establishing the feasibility of thorium-fuelled reactors.

Physical properties of thorium

In many respects the physical and mechanical properties of thorium compare favourably with those of uranium. The physical properties are compared in Table I. The isotropic face-centred cubic crystal structure of thorium is a significant advantage, since the orthorhombic crystals of uranium expand unequally in different directions. This anisotropy is responsible for growth effects and internal stresses that lead to accelerated swelling when uranium is irradiated in the temperature range of 400 to 550°C. The phase transformation in uranium effectively prohibits its operation as a fuel above about 660°C, since fission gases are released from the lattice during atomic rearrangements that occur during transformation. The phase transformation of thorium is at about 1400°C, and it is decreased only slightly by many alloying elements, as shown by Bannister [5].

The thermal conductivity of thorium is about 30% greater than that of uranium at 100°C and about 8% greater at 650°C. The density of thorium is significantly less than that of uranium metal.

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation.
### PHYSICAL PROPERTIES OF THORIUM AND URANIUM

<table>
<thead>
<tr>
<th></th>
<th>Pure thorium</th>
<th>Uranium (β-transformed)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal structure</td>
<td>face-centered cubic</td>
<td>orthorhombic</td>
</tr>
<tr>
<td>Lowest transformation temperature (°C)</td>
<td>1400</td>
<td>661</td>
</tr>
<tr>
<td>Melting point (°C)</td>
<td>1750</td>
<td>1130</td>
</tr>
<tr>
<td>Thermal conductivity coefficient (W/cm deg C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>at 100°C</td>
<td>0.38</td>
<td>0.28</td>
</tr>
<tr>
<td>400°C</td>
<td>0.42</td>
<td>0.35</td>
</tr>
<tr>
<td>650°C</td>
<td>0.46</td>
<td>0.42</td>
</tr>
<tr>
<td>Thermal expansion coefficient (10⁻⁶/deg C (average))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25-200°C</td>
<td>11.0</td>
<td>15.5a</td>
</tr>
<tr>
<td>25-650°C</td>
<td>12.5</td>
<td>19</td>
</tr>
<tr>
<td>25-1000°C</td>
<td>11.7</td>
<td>19.0</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>11.0</td>
<td>19.0</td>
</tr>
<tr>
<td>Elastic constants (25°C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Modulus of elasticity (lb/in²)</td>
<td>10.4 x 10⁶</td>
<td>29.3 x 10⁶</td>
</tr>
<tr>
<td>Shear modulus (lb/in²)</td>
<td>4.1 x 10⁶</td>
<td>12.0 x 10⁶</td>
</tr>
<tr>
<td>Poisson's ratio</td>
<td>0.27</td>
<td>0.22</td>
</tr>
</tbody>
</table>

* a Average values for beta-transformed uranium values for the three directions are [100], 27.4 x 10⁻⁶, [010], 0, [001], 23.2 for the range 0 to 300°C.

A selection from Ames Laboratory's [6] values of the heat capacity of thorium iodide follows:

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>25</th>
<th>100</th>
<th>200</th>
<th>400</th>
<th>600</th>
<th>800</th>
<th>1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat capacity (cal/mole degC)</td>
<td>6.56</td>
<td>6.75</td>
<td>6.99</td>
<td>7.49</td>
<td>7.99</td>
<td>8.62</td>
<td>9.54</td>
</tr>
</tbody>
</table>

Darnell and McCollum [7] have studied the vapour pressure of thorium in equilibrium with ThO₂ and found by extrapolation the following relation for the vapour pressure of pure metal

\[
\log P(\text{atm}) = -27.960/T + 5.575.
\]

The predominate vapour species above liquid solutions of ThO₂ in thorium was ThO, and its vapour pressure was described as

\[
\log P(\text{atm}) = -(22.200 \pm 700)/T + (4.70 \pm 0.31).
\]

The solid solubility of ThO₂ in thorium was 3.5% at 2090°K, and 8.2% at 2450°K.
Thorium has elastic and mechanical properties at room temperature somewhat inferior to those of uranium. The mechanical properties of thorium metal are dependent on impurity and alloy content and on the textures and cold work developed during fabrication treatments. The purest thorium, made by decomposition of thorium iodide, has, when annealed, tensile strengths of 16,000 to 20,000 lb/in$^2$, yield strengths from 5000 to 10,000 lb/in$^2$, and considerable ductility. Commercial thorium, produced by bomb-reduction of thorium fluoride, contains carbon and other interstitial impurities which strengthen thorium, as shown in Fig. 1. In the cast or wrought and annealed condition this metal has good ductility and tensile and yield strengths varying upward from about 25,000 and 18,000 lb/in$^2$, respectively. Thorium work hardens rapidly, and tensile strengths are increased 50% and yield strengths doubled by cold reductions of 50% or less. Ductility is reduced by cold work, but not excessively so. Representative data [1] are shown in Table II.

Thorium metal is readily fabricated by casting, powder metallurgy, extrusion, rolling, and other methods. Cold worked material starts to recrystallize and anneal at slightly above 500°C, although temperatures up to 700°C may be required to ensure complete annealing for slightly worked material.

The modulus of elasticity [1] of thorium is about $10.4 \times 10^6$ lb/in$^2$ at 25°C and it decreases linearly with increasing temperature to about $7 \times 10^5$ lb/in$^2$ at 500°C. Slightly higher values of $11.6 \times 10^5$ lb/in$^2$ at 25°C decreasing by 7400 lb/in$^2$ degC to 300°C were more recently reported by Livesey [8] as the result of dynamic measurements. For uranium, values of $27 \times 10^5$ lb/in$^2$ at 25°C and $23.4 \times 10^5$ lb/in$^2$ at 300°C have been reported [2].

**Thorium alloys**

Considerable research has been done on alloying thorium to improve the mechanical properties for use in reactor fuel elements. Many potential hardeners were found, some of the most effective being C, Zr, Al, In, Mo, and U. Detailed information on effects of specific elements and metallurgical factors involved can be found in earlier reviews [1-3]. More recent studies of thorium have been directed toward improvement of high-temperature properties, and we shall briefly summarize the data from several recent studies on potential fuel alloy systems.

Solid solution hardening, precipitation hardening, and dispersion hardening have all been considered for improving high-temperature properties of thorium. For exploratory studies, hot hardness has been a frequent method of evaluation, but hot tensile tests and creep tests have also been used. The results of separate studies are not directly comparable, probably because of differing impurity constituents in the base thorium and slightly differing experimental techniques.

**Thorium-uranium alloys**

Uranium is an essential constituent in thorium fuel alloys because it is added to provide a fissionable isotope and because it is a product of neutron irradiation. The solubility of uranium was reported by Rough and Bauer [9]
FIG. 1. Effect of some interstitial solutes normally found in bomb-reduced thorium on the room-temperature tensile properties and hardness of iodide thorium (Base metal compositions and metallurgical treatments: BMI-0.012% C, 0.080% O; forged and hot rolled at 700°C, cold reduced 50%, annealed in argon 2 h at 850°C; ORNL-0.015% C, 0.080% O; arc-melted, cold reduced 80%, vacuum annealed 0.5 h at 650°C.)
TABLE II
TYPICAL PROPERTIES OF WROUGHT-ANNEALED AND COLD-WORKED THORIUM AT ROOM TEMPERATURE

<table>
<thead>
<tr>
<th>Condition when tested</th>
<th>Tensile strength (lb/in²)</th>
<th>Yield strength 0.2% offset (lb/in²)</th>
<th>Elongation (%)</th>
<th>Gauge length (in)</th>
<th>Reduction in area (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Iodide</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wrought-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>sheet(^a)</td>
<td>17 300</td>
<td>6 900</td>
<td>36</td>
<td>2</td>
<td>62</td>
</tr>
<tr>
<td>Wrought-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>sheet(^b)</td>
<td>19 700</td>
<td>11 200</td>
<td>44</td>
<td>1</td>
<td>60</td>
</tr>
<tr>
<td><strong>Bomb-reduced</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Extruded-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>rod(^c)</td>
<td>30 000</td>
<td>21 700</td>
<td>51</td>
<td>2</td>
<td>74</td>
</tr>
<tr>
<td>Extruded-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>rod(^d)</td>
<td>34 400</td>
<td>27 600</td>
<td>51</td>
<td>2</td>
<td>73</td>
</tr>
<tr>
<td>Wrought-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>rod(^e)</td>
<td>33 700</td>
<td>26 300</td>
<td>55</td>
<td>1.4</td>
<td>69</td>
</tr>
<tr>
<td>Extruded-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>rod(^f)</td>
<td>38 500</td>
<td>31 900</td>
<td>48</td>
<td>2</td>
<td>69</td>
</tr>
<tr>
<td>Wrought-annealed</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>sheet(^g)</td>
<td>39 600</td>
<td>30 300</td>
<td>--</td>
<td>--</td>
<td>52</td>
</tr>
<tr>
<td>Cold rolled 37.5%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>rod(^h)</td>
<td>49 000</td>
<td>45 400</td>
<td>20</td>
<td>1.4</td>
<td>61</td>
</tr>
<tr>
<td>Cold rolled 25%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>sheet(^i)</td>
<td>58 700</td>
<td>54 800</td>
<td>11</td>
<td>1</td>
<td>39</td>
</tr>
<tr>
<td>Cold rolled 50%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>sheet(^i)</td>
<td>65 400</td>
<td>61 800</td>
<td>5</td>
<td>1</td>
<td>16</td>
</tr>
</tbody>
</table>

\(^a\) Average of 4 lots, cold reduced 85%, annealed 0.5 h at 650°C.
\(^b\) Forged and hot rolled at 700°C, cold rolled 50%, annealed 2 h at 850°C.
\(^c\) Average of 4 lots, 0.05% C, annealed 0.5 h at 750°C after extrusion.
\(^d\) Average of 4 lots, 0.07% C, annealed 0.5 h at 750°C after extrusion.
\(^e\) Average of 4 lots, 0.06 to 0.085% C, reduced 95% by extrusion, cold reduced 37.5%, annealed 0.5 h at 750°C.
\(^f\) Average of 4 lots, 0.09% C, annealed 0.5 h at 750°C after extrusion.
\(^g\) Forged and hot rolled at 700°C, annealed 1 h at 750°C, 0.11% C.
\(^h\) Average of 3 lots, 0.06 to 0.085% C, reduced 72% by extrusion, cold rolled 37.5%.
\(^i\) 0.11% C, forged and hot rolled at 700°C before cold rolling.

as about 1.8% at 1100°C, 1.1% at 600°C, and 0.7% at 25°C. Essentially similar results were reported by Murray [10] and Bentle [11] but a maximum solubility limit of 2.7% at 1343°C has been suggested by Bannister [5]. Bentle's results predicted somewhat higher solubility at 1250°C. Uranium in excess of the solubility limit exists as free uranium, which at levels in excess of about 15% is present as a grain boundary network regardless of
fabricating techniques used [12]. Below about 15% U, the composition of the alloy and the metallurgical treatments will govern the size and distribution of the uranium phase, which in turn affect the physical properties and irradiation performance. High-temperature heat treatment tends to cause agglomerations of uranium and decrease strength [13]. Battelle [12] has studied the effect of casting and fabricating techniques on hot hardness and creep strength of alloys containing up to 20% U. Limited data indicated that Th-5% U had greater creep strength at 600 and 700°C than alloys containing greater amounts of uranium. Kittel [14] found superior irradiation performance on alloys containing 15% U or less.

Creep data by Atomics International [15] shown in Table III for annealed, swaged, and swaged and thermally cycled Th-9% U, show that the strengthening effects of mechanical work are retained at 600°C.

**TABLE III**

**CREEP DATA FROM Th-9% U TESTED AT 600°C**

<table>
<thead>
<tr>
<th>Alloy condition</th>
<th>Stress (lb/in²)</th>
<th>Strain rate (%/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annealed</td>
<td>4000</td>
<td>0.017</td>
</tr>
<tr>
<td>Annealed</td>
<td>4500</td>
<td>0.033</td>
</tr>
<tr>
<td>Swaged</td>
<td>6000</td>
<td>0.010</td>
</tr>
<tr>
<td>Swaged</td>
<td>7000</td>
<td>0.026</td>
</tr>
<tr>
<td>Swaged</td>
<td>8000</td>
<td>0.060</td>
</tr>
<tr>
<td>Swaged and thermally cycled</td>
<td>5000</td>
<td>0.0035</td>
</tr>
<tr>
<td>Swaged and thermally cycled</td>
<td>5700</td>
<td>0.0060</td>
</tr>
<tr>
<td>Swaged and thermally cycled</td>
<td>7000</td>
<td>0.017</td>
</tr>
</tbody>
</table>

Ternary alloys containing uranium

Battelle Memorial Institute [12] also investigated high-temperature mechanical properties of fabricated ternary alloys containing 10% U. Relative behaviour of the alloys was the same at 600 and 700°C. Of the alloys tested in creep, those containing 0.5% Be, 1.5% Mo, or 2% Nb had creep strengths appreciably above that of the binary 10% U alloy, some appeared to be very slightly stronger than the binary Th-5% U alloy. Alloys containing both 10% U and 10% Zr were somewhat inferior to the binary 10% U alloy in creep strength but had significantly higher tensile and yield strengths at both 600 and 700°C. Hot hardness test results were also reported for cast specimens containing the various ternary additions to Th-10% U. Alloys containing 10% Zr, 0.1% Be, 0.2% C, and 1.5% Mo were significantly harder than the 10% U base alloy at 600°C and below.
Cole and Wilkinson [13] of Advanced Technology Laboratories (ATL) also studied ternary additions to thorium containing 5 or 10% U. Tensile and stress-rupture strengths of the base alloys at 600 and 800°C could essentially be doubled. Most noticeable improvement was attained with 2 and 5% Zr, although additions of Nb, Mo, C, Al, and Be also significantly improved strength. Solid solution hardening and dispersion hardening were obtained. The mechanical properties were sensitive to the size and distribution of the uranium-phase particles, and careful control of melting and heat-treatment procedures was utilized to achieve optimum structures and properties. Carbon at levels of 0.2 and 0.25% only slightly improved the properties of the binary Th-U alloys at 600°C and above but was very effective at 400°C and below. However, carbon was detrimental in alloys that also contained zirconium.

Of the alloys tested by ATL, superior strengths were obtained for quenched and aged alloys. Table IV shows typical results including the effects of extended aging treatments. The data indicate that over-aging has no detrimental effects. The extended aging treatments did not significantly alter the microstructure.

High-temperature strengthening

Burka and Hammond [16] at ORNL studied the effects of many binary and some ternary additions to arc-melted thorium and evaluated results by hot hardness traverses on hot rolled plate. Carbon was found to be principally effective at below 600°C, although 0.2% C in combination with Cr, Mo, U, or Be appeared to enhance hot hardness at 600°C. Best hardening alloy additives at 600 and 750°C were 2 or 4% In and 5% Zr. The hardness of the various alloys at 600 and 750°C is shown in Table V. Indium was less effective as a hardener in alloys prepared by powder metallurgy [17]. Murray [18] has studied the solubility of indium in thorium and found a eutectic at 8.9% In and 1160°C. The solubility decreased to 2.5% at 1000°C and 0.85% at 800°C. Indium, of course, is not a desirable alloying element for thermal reactor fuel.

Burka and Hammond [17, 19] also investigated the hardening of thorium by fine particle dispersions of ThO₂. The compacts, fabricated by extrusion of ball-milled TH₄ and ThO₂ powders, showed high strength and hardness and considerable ductility at 800°C. The experiments also indicated that control of the oxide and carbon pick-up during the milling was quite difficult.

Compatibility with cladding materials

We have not yet surveyed information on the compatibility of thorium with potential clad materials, but little appears available, particularly for the temperature range of power reactors. Hanford [20] has successfully clad tubular Th-2.5% U-1.0% Zr by co-extrusion with Zircaloy-2 and is now testing the material under irradiation in pressurized water. Zircaloy-2 and the fuel alloy inter-diffuse slightly at 700°C, but the materials are compatible in the operating temperature range of water-cooled power reactors.
### EFFECTS OF AGING TREATMENTS ON THE STRENGTH OF HEAT-TREATED Th-U-Zr ALLOYS

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Aging conditions</th>
<th>Ultimate tensile strength (lb/in²)</th>
<th>Yield strength (lb/in²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Time (h)</td>
<td>Temp. (°C)</td>
<td></td>
</tr>
<tr>
<td>Th-5% U-2% Zr</td>
<td>2</td>
<td>850</td>
<td>12900</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>800</td>
<td>7700</td>
</tr>
<tr>
<td></td>
<td>170</td>
<td>850</td>
<td>8200</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>850</td>
<td>8350</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>800</td>
<td></td>
</tr>
<tr>
<td>Th-5% U-5% Zr</td>
<td>2</td>
<td>850</td>
<td>13230</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>800</td>
<td>11800</td>
</tr>
<tr>
<td></td>
<td>170</td>
<td>850</td>
<td>14115</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>850</td>
<td>12050</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>800</td>
<td></td>
</tr>
<tr>
<td>Th-10% U-2% Zr</td>
<td>2</td>
<td>850</td>
<td>9015</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>800</td>
<td>10500</td>
</tr>
<tr>
<td></td>
<td>170</td>
<td>850</td>
<td>8450</td>
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<tr>
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<td>2</td>
<td>850</td>
<td>10550</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>800</td>
<td></td>
</tr>
<tr>
<td>Th-10% U-5% Zr</td>
<td>2</td>
<td>850</td>
<td>7900</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>800</td>
<td>9250</td>
</tr>
<tr>
<td></td>
<td>170</td>
<td>850</td>
<td>11150</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>850</td>
<td>8000</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>800</td>
<td></td>
</tr>
</tbody>
</table>

*a* Tested at 800°C.

### Summary

The data on physical and mechanical properties of thorium metal suggest that it may have considerable potential as a reactor fuel. A preliminary analysis of creep data, high-temperature strength, and hot hardness data suggests that at temperatures above about 500°C thorium is significantly stronger than uranium. Even at temperatures of 400 to 550°C, anisotropy causes internal stresses under thermal cycling or neutron irradiation, which decrease the creep strength of uranium significantly below that obtained at isothermal conditions in the absence of irradiation. Since thorium, with its
Table V

Thorium-base alloys in order of decreasing hardness at 600 and 750°C

<table>
<thead>
<tr>
<th>Alloy (wt.%)</th>
<th>600°C DPH</th>
<th>750°C DPH</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 In</td>
<td>91</td>
<td>55</td>
</tr>
<tr>
<td>2 In</td>
<td>63</td>
<td>55</td>
</tr>
<tr>
<td>10 Zr</td>
<td>59</td>
<td>54</td>
</tr>
<tr>
<td>5 Zr</td>
<td>59</td>
<td>33</td>
</tr>
<tr>
<td>5 Zr-0.2C</td>
<td>54</td>
<td>33</td>
</tr>
<tr>
<td>20 Zr</td>
<td>54</td>
<td>33</td>
</tr>
<tr>
<td>5 V-0.2C</td>
<td>50</td>
<td>31</td>
</tr>
<tr>
<td>4 In-5 Zr</td>
<td>49</td>
<td>25</td>
</tr>
<tr>
<td>2 In-0.2C</td>
<td>49</td>
<td>25</td>
</tr>
<tr>
<td>2 Cr-0.2C</td>
<td>47</td>
<td>25</td>
</tr>
<tr>
<td>2 Mo-0.2C</td>
<td>44</td>
<td>24</td>
</tr>
<tr>
<td>5 U-0.2C</td>
<td>44</td>
<td>23</td>
</tr>
<tr>
<td>0.2 Al-0.2C</td>
<td>43</td>
<td>20</td>
</tr>
<tr>
<td>8 Nb</td>
<td>43</td>
<td>20</td>
</tr>
<tr>
<td>0.2 Be-0.2C</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>5 Ti-0.2C</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>0.2 C</td>
<td>38</td>
<td>20</td>
</tr>
<tr>
<td>5 Ti</td>
<td>37</td>
<td>19</td>
</tr>
<tr>
<td>20 U</td>
<td>36</td>
<td>19</td>
</tr>
<tr>
<td>0.4 Al</td>
<td>35</td>
<td>18</td>
</tr>
<tr>
<td>8 Cr</td>
<td>35</td>
<td>18</td>
</tr>
<tr>
<td>0.2 Al</td>
<td>32</td>
<td>18</td>
</tr>
<tr>
<td>5 V</td>
<td>32</td>
<td>17</td>
</tr>
<tr>
<td>5 U</td>
<td>30</td>
<td>17</td>
</tr>
<tr>
<td>8 Mo</td>
<td>30</td>
<td>17</td>
</tr>
<tr>
<td>0.15 C</td>
<td>29</td>
<td>15</td>
</tr>
<tr>
<td>2 Nb-0.2C</td>
<td>29</td>
<td>15</td>
</tr>
<tr>
<td>10 U</td>
<td>28</td>
<td>14</td>
</tr>
<tr>
<td>4 Mo</td>
<td>27</td>
<td>14</td>
</tr>
<tr>
<td>4 Cr</td>
<td>27</td>
<td>13</td>
</tr>
<tr>
<td>4 Nb</td>
<td>26</td>
<td>13</td>
</tr>
<tr>
<td>2 Nb</td>
<td>26</td>
<td>13</td>
</tr>
<tr>
<td>2 Mo</td>
<td>24</td>
<td>11</td>
</tr>
<tr>
<td>0.10 C</td>
<td>23</td>
<td>11</td>
</tr>
<tr>
<td>Thorium</td>
<td>22</td>
<td>11</td>
</tr>
<tr>
<td>2 Cr</td>
<td>21</td>
<td>11</td>
</tr>
<tr>
<td>0.2 Be</td>
<td>21</td>
<td>9</td>
</tr>
</tbody>
</table>

isotropic structure, may not be subject to such internal stresses, its properties may not be degraded to the same extent. However, the effect on creep strength of fission events during irradiation of thorium has yet to be established.

Recent developments with uranium alloy fuels have emphasized the importance of microstructural control for improving irradiation behaviour.
of the metal, and data indicate that fine particle dispersions are most effective in increasing dimensional stability of fuel alloys at high temperature. Similar methods may be effective with thorium and should be investigated.

Thorium-fuelled reactors will likely have greatest economy if initial uranium concentrations are low and if generated $^{233}$U contributes substantially to the burn-up. Thus, long-time operation of the fuel will be required. Experiments are needed to establish the radiation performance of thorium in which burn-up of generated $^{233}$U is substantial.

Mechanical property data suggest that thorium fuels may be able to operate at considerably higher temperatures than uranium. If such abilities are to be utilized, consideration must be given to compatibility of the fuel with potential clad materials.

CERAMIC COMPOUNDS OF THORIUM

Many thorium compounds exist with the high-temperature stability required for useful ceramic bodies. These include ThBe$_3$, ThB$_4$, ThB$_6$, ThC, ThC$_2$, ThN and higher nitrides, ThO$_2$, and several phosphides, silicides, and sulphides. Time does not permit reviewing here the state of knowledge of the properties of most of these except to say that for the most part it is incomplete and unverified. The literature on ceramic compounds of thorium is rampant with guesses, misquotations, and incomplete qualifications, so the original sources must be traced for all data. This we have not done for this brief review, but we are preparing an extensive collection of data on thorium ceramic compounds [21]. Our attention here will emphasize the compounds of most immediate nuclear application.

Thorium dioxide

Thorium dioxide is without doubt the best characterized ceramic compound of thorium. Although this partly stems from its study for nuclear purposes, a great deal of information exists because of the non-nuclear usefulness of the material. Since thoria is the highest melting and the most stable to reduction of all the refractory oxides, it is a superior crucible material for the melting of reactive metals. Thoria is generally prepared in powder form by the thermal decomposition of a purified salt, usually the oxalate. This powder can be consolidated by usual ceramic fabrication techniques, such as slip casting, pressing and sintering, or hot pressing. The fabricability and ceramic properties can often be related to conditions of preparation of the starting salt and firing. Fabrication, properties, and uses of thoria ceramics have recently been reviewed by Hepworth and Rutherford [22] and by Ryshkewitch [23].

Thorium dioxide exists up to its melting point as a single cubic phase with the fluorite crystal structure, isomorphous and completely miscible with UO$_2$. Unlike UO$_2$, thoria does not dissolve oxygen to a measurable extent. Therefore it is stable to high temperature in oxidizing environments. On prolonged heating to 1800 to 1900°C in vacuum it blackens with loss of oxygen, although the loss is insufficient to be reflected in chemical analysis.
or lattice-parameter measurement. On reheating in air to 1200 or 1300°C
the white colour is restored. When uranium dioxide is incorporated in thoria,
the lattice can take up extra oxygen in proportion to the uranium content.

Table VI summarizes the more important physical and mechanical prop-
erties of thoria, along with analogous properties of uranium dioxide taken
from the compilation by Belle [24]. In some cases the data are a small
selection of what are available. The heat capacity is known with precision
from near absolute zero up to over 900°C, and other thermodynamic functions
have been derived from it [25, 26]. Less precise data exists [27, 28] up
to 2400°C. The heat and free energy of formation are from a compilation
[29] that extends up to 2000°C, although it is not based on the best and latest
data. The lack of a value for the heat of fusion illustrates the state of con-
fusion on properties of thoria. A value of 21.4 kcal/mole has been attri-
buted to Lambertson, Mueller and Gunzel [30]. These authors needed a
value for a theoretical interpretation of their UO₂-ThO₂ melting point data
and guessed it by taking 3R as the entropy of fusion. Mechanical properties
up to 900°C and higher are given in the compilation by Ryshkewitch [23] and
elsewhere, but these should be taken as examples of particular specimens
and not as values to be expected. The actual values depend on density, poro-
sity, grain size, and other qualities dependent on fabrication, and they vary
considerably. Preliminary measurements on pure dense specimens pre-
pared at Oak Ridge National Laboratory [31] indicate higher strengths than
those tabulated. Recent data [31] on the compressive creep of thoria are
given in Fig. 2. Thoria is a high-temperature semiconductor, so its thermal
and, even more so, electrical conductivity depend on fabrication history
and impurity content. The latter property shows quite wide variations.

Although a thorium-base fuel will necessarily contain uranium, little
information exists on the properties of thoria-urania solid solutions. Physi-
cal and mechanical properties are probably in most cases intermediate
between those of the pure components fabricated similarly. However, this
is not true of transport properties such as thermal conductivity. Kungery [42]
founds lower conductivity for mixed oxides than for either pure oxide and still
lower thermal conductivity after the specimens had been heated under
oxidizing conditions.

Excellent compatibility of thoria with metals reflects its high thermo-
dynamic stability. It is the most stable solid oxide at high temperatures,
and thus it resists attack by many reactive metals. Table VII summarizes
results from some tests of compatibility of thoria with refractory and other
metals. Data on compatibility with the usual reactor structural metals at
modest temperatures is lacking, probably because no incompatibility has
been found and because anything compatible with UO₂ would be less reactive
toward thoria. A further observation [37] was that thoria contained less
than 1% W from the crucible after prolonged studies of vaporization to 2600°C.
One limitation to thoria is that it is subject to thermal shock.

**Thorium carbides**

Next to thoria, the most prominent thorium ceramic compounds for
nuclear application are the carbides. The monocarbide ThC and the di-
carbide ThC₂ can both be prepared by a variety of techniques. Arc-melting
### Table VI

**IMPORTANT PROPERTIES OF THORIUM AND URANIUM DIOXIDES**

<table>
<thead>
<tr>
<th>Property</th>
<th>Value for ThO₂</th>
<th>Reference</th>
<th>Value for UO₂ [24]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Crystal structure</strong></td>
<td>Face-centred cubic (CaF₂ type)</td>
<td></td>
<td>Face-centred cubic (CaF₂ type)</td>
</tr>
<tr>
<td>Space group</td>
<td>O₈ Fm3m</td>
<td></td>
<td>O₈ Fm3m</td>
</tr>
<tr>
<td>Lattice parameter (Å)</td>
<td>5.5974 at 26°C</td>
<td></td>
<td>5.4704 at 20°C</td>
</tr>
<tr>
<td></td>
<td>5.6448 at 942°C</td>
<td>[32]</td>
<td>5.5246 at 946°C</td>
</tr>
<tr>
<td><strong>Theoretical density (g/cm³)</strong></td>
<td>10.00</td>
<td></td>
<td>10.96</td>
</tr>
<tr>
<td><strong>Interatomic distances (Å)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>M-M</td>
<td>3.958</td>
<td></td>
<td>3.868</td>
</tr>
<tr>
<td>O-O</td>
<td>2.799</td>
<td></td>
<td>2.735</td>
</tr>
<tr>
<td>M-O</td>
<td>2.424</td>
<td></td>
<td>2.368</td>
</tr>
<tr>
<td><strong>Thermal properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Melting point (°C)</td>
<td>3300 ± 100</td>
<td>[30]</td>
<td>2760 ± 30</td>
</tr>
<tr>
<td>Spectral emissivity (λ = 0.65 μm)</td>
<td>0.53 at 300°C to 0.21 at 800°C</td>
<td>[33]</td>
<td>0.416 ± 0.026 (near m.p.)</td>
</tr>
<tr>
<td></td>
<td>0.2 to 0.65 at 1300°C depending on sample history</td>
<td>[34]</td>
<td>0.850 at 727°C</td>
</tr>
<tr>
<td>Thermal conductivity (W/cm deg C)</td>
<td>0.103 at 100°C</td>
<td></td>
<td>0.370 at 1947°C</td>
</tr>
<tr>
<td></td>
<td>0.086 at 200°C</td>
<td></td>
<td>0.105 at 100°C</td>
</tr>
<tr>
<td></td>
<td>0.060 at 400°C</td>
<td></td>
<td>0.0815 at 200°C</td>
</tr>
<tr>
<td></td>
<td>0.044 at 600°C</td>
<td></td>
<td>0.0590 at 400°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0452 at 600°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0376 at 800°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0361 at 1000°C</td>
</tr>
<tr>
<td>Heat capacity (cal/mole deg C)</td>
<td>17.060 + 18.06(10⁻⁴)T - 2.6166(10⁵)/T²</td>
<td>[26]</td>
<td>18.46 + 2.431(10⁻³)T - 2.272(10⁵)/T²</td>
</tr>
<tr>
<td>(298 to 1200°C)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Debye temperature (°K)</td>
<td>200</td>
<td></td>
<td>870°K</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>&lt; 600°K</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(390-600°K)</td>
</tr>
<tr>
<td>Property</td>
<td>Value for ThO₂</td>
<td>Reference</td>
<td>Value for UO₂ [24]</td>
</tr>
<tr>
<td>-----------------------------------------------</td>
<td>-----------------</td>
<td>-----------</td>
<td>----------------------------------------</td>
</tr>
<tr>
<td>Coefficient of linear expansion (°C⁻¹)</td>
<td>6216(10⁻⁶) + 3.541(10⁻³)T - 0.1125/T²</td>
<td>[36]</td>
<td>10.8 x 10⁻⁶ (20-926°C) 9.9 x 10⁻⁶ (25-800°C) 10.0 x 10⁻⁶ (400-900°C) 10.52 x 10⁻⁶ (26-1000°C)</td>
</tr>
<tr>
<td>Vapour pressure (atm)</td>
<td>7.64 - 3.440(10⁴)/T (2200-2900°K) 9.02 - 3.78 (10⁴)/T (2170-2400°K)</td>
<td>[37]</td>
<td>log P = - 33115/T - 4.026 log T + 23.111</td>
</tr>
<tr>
<td>Thermodynamic properties</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat of formation, Δ₂₉₈ (kcal/mole)</td>
<td>233.2 ± 0.4</td>
<td>[29]</td>
<td>259.2 ± 0.6</td>
</tr>
<tr>
<td>Free energy of formation, -ΔF₂₉₈ (kcal/mole)</td>
<td>279.2 ± 0.65</td>
<td>[29]</td>
<td>246.6 ± 0.6</td>
</tr>
<tr>
<td>Entropy, 298°K, (cal/mole deg C)</td>
<td>15.59 ± 0.02</td>
<td>[25]</td>
<td>18.6 ± 0.1</td>
</tr>
<tr>
<td>Heat of sublimation (kcal/mole)</td>
<td>158.7 ± 2.5, in range 2000-3000°K</td>
<td>[37]</td>
<td>137.1 ± 1.7 at 1800°K</td>
</tr>
<tr>
<td>Entropy of sublimation (cal/mole deg C)</td>
<td>35.3 ± 1.0, in range 2000-3000°K</td>
<td>[37]</td>
<td>36.4 at 1800°K</td>
</tr>
<tr>
<td>Mechanical properties</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Elastic properties</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Young's modulus (kilobars, lb/in²)</td>
<td>1370, 19.8(10⁶)</td>
<td>[23]</td>
<td>1930, 28.0(10⁹) at room temperature 1837, 26.5(10⁹) at room temperature 1655, 24.0(10⁹) at 800°C</td>
</tr>
<tr>
<td>Shear modulus (kilobars, lb/in²)</td>
<td>990, 14.3(10⁶) at 30°C 390, 5.6(10⁵) at 1300°C</td>
<td>[39]</td>
<td>745, 10.8(10⁹) at room temperature</td>
</tr>
<tr>
<td>Poisson's ratio</td>
<td>0.17</td>
<td></td>
<td>0.302 at room temperature</td>
</tr>
<tr>
<td>Modulus of rupture (kilobars, lb/in²)</td>
<td>0.83, 12000</td>
<td>[40]</td>
<td>0.965-1.10, 14,000 - 16,000</td>
</tr>
<tr>
<td>Compressive strength (kilobars, lb/in²)</td>
<td>16, 214000</td>
<td>[23]</td>
<td>4.14-9.65, 60,000 - 140,000</td>
</tr>
<tr>
<td>Fracture strength (kilobars, lb/in²)</td>
<td>1, 14000</td>
<td>[23]</td>
<td>0.16-0.37, 2300-5400</td>
</tr>
<tr>
<td>Hardness, Knoop</td>
<td>640 (500-g load)</td>
<td>[41]</td>
<td>666 ± 14</td>
</tr>
<tr>
<td>Electrical, magnetic, and optical properties</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electrical resistivity (ohm cm)</td>
<td>1 to &gt; 10⁴ at 1600°C, depending on sample and treatment</td>
<td>[23]</td>
<td>3 x 10⁻⁴ - 10⁻⁶ at room temperature</td>
</tr>
<tr>
<td>Index of refraction</td>
<td>2.09</td>
<td></td>
<td>about 10⁻¹ at 500°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>about 10⁻¹ at 1000°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.95</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.2-2.7</td>
</tr>
</tbody>
</table>
FIG. 2. Compressive creep behaviour of thorium. Specimens were isostatically pressed at 35,000 lb/in² and fired in air for 2 h at 1800°C. Density 97.5% of theoretical, average grain diameter about 10 µm.

of the elements, pressing and sintering of the powdered elements, and carbon reduction of the oxide are among the frequently encountered methods. The monocarbide phase apparently is capable of appreciably less but no greater than the stoichiometric carbon content. The same is true of the dicarbide, and preparations of it frequently are hypostoichiometric.

Information on properties of thorium carbides is very scant compared with that on the oxide. Properties of the two carbides are summarized in Table VIII. Comparison with Table VI points up the many gaps in our knowledge. Many of these gaps are probably already being filled, because of the growing interest in these materials. For example, several new publications [46–51] relate to the thermodynamic properties.
### Table VII

**REACTIVITY OF THORIA TOWARD METALS**

<table>
<thead>
<tr>
<th>Behaviour</th>
<th>Temperature (°C) of observed behaviour with</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>No reaction on rapid heating to temperature</td>
<td></td>
<td>[43]</td>
</tr>
<tr>
<td>Reactors on rapid heating</td>
<td></td>
<td>[43]</td>
</tr>
<tr>
<td>Heated 20°C/min and held 15 min at temperature</td>
<td></td>
<td>[44]</td>
</tr>
<tr>
<td>No reaction</td>
<td>1400 1800 1800 1800 1400 1600 1600</td>
<td></td>
</tr>
<tr>
<td>Slight attack</td>
<td>1600 1800 1800 1800</td>
<td></td>
</tr>
<tr>
<td>Severe attack</td>
<td>1600</td>
<td></td>
</tr>
<tr>
<td>No surface damage</td>
<td>1800 1800 1800 1800 1800</td>
<td></td>
</tr>
<tr>
<td>Visible reaction in 4 min (although little reaction to 2300°C)</td>
<td>1900</td>
<td>[45]</td>
</tr>
<tr>
<td></td>
<td>2200</td>
<td></td>
</tr>
</tbody>
</table>

Information on the compatibility of thorium carbides with other materials is scarce. This survey has revealed nothing on compatibility of pure carbides with metals, although it is likely that the thorium carbides resemble the uranium carbides in this. The carbides are reactive toward air and water. Powders have to be handled in inert atmospheres. Various hydrocarbons result from the reaction of the carbides with water [52, 57].

In contrast with the oxide, the carbides have been studied principally mixed with uranium. Complete isomorphous solid solutions exist between uranium and thorium monocarbides. The binary dicarbide system is more complex; an equilibrium diagram has been proposed by Hill and Cavin [53]. The thorium compound is monoclinic but very nearly orthorhombic. On sufficient heating or substitution with uranium it gradually transforms to body-centred tetragonal. This phase is not completely miscible with the body-centred tetragonal uranium compound, but the two-phase region is narrow.

Several thorium compounds, including both carbides, were prepared by arc-melting with 10 and 20% substitution of uranium for the thorium [58]. Table IX lists several properties measured and Table X gives results of compatibility tests with various materials. The same authors also reported graphically weight gains in air for the carbides with 10% substitution. Cook et al. [54] measured the thermal expansion over the range 40 to 1000°C for ThC₂ and several mixed dicarbides containing up to 75% U substituted. Increasing the uranium content or the temperature increased the thermal expansion, the increase with uranium content was monotonic except at the two-phase region.
<table>
<thead>
<tr>
<th>Property</th>
<th>ThC&lt;sub&gt;2&lt;/sub&gt;</th>
<th>Reference</th>
<th>ThC</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal structure</td>
<td>Pseudo-orthorhombic</td>
<td>[52]</td>
<td>Face-centred cubic (NaCl)</td>
<td>[52]</td>
</tr>
<tr>
<td>Lattice parameter (Å)</td>
<td>10.555</td>
<td></td>
<td>5.346</td>
<td></td>
</tr>
<tr>
<td></td>
<td>8.233</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.201</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Theoretical density (g/cm&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>9.6</td>
<td></td>
<td>10.61</td>
<td></td>
</tr>
<tr>
<td>Interatomic distance (Å)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Th-Th</td>
<td>3.780</td>
<td></td>
<td>2.673</td>
<td></td>
</tr>
<tr>
<td>C-C</td>
<td>3.780</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Th-C</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal Properties</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Melting point (°C)</td>
<td>2655</td>
<td>[52]</td>
<td>2625</td>
<td>[52]</td>
</tr>
<tr>
<td>Specific heat (cal/mole deg C)</td>
<td>13.55</td>
<td>[53]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coefficient of linear expansion (degC&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>7.29(10&lt;sup&gt;-6&lt;/sup&gt;) for 40-400°C</td>
<td>[54]</td>
<td>8.80(10&lt;sup&gt;-6&lt;/sup&gt;) for 40-1000°C</td>
<td></td>
</tr>
<tr>
<td>Thermodynamic Properties</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat of formation, -ΔH&lt;sub&gt;298&lt;/sub&gt; (kcal/mole)</td>
<td>44.8</td>
<td>[56]</td>
<td>29.6±4.8</td>
<td>[46]</td>
</tr>
<tr>
<td>Free energy of formation, -ΔF (kcal/mole, 298-2300°K)</td>
<td>45-2.6(10&lt;sup&gt;-3&lt;/sup&gt;)T</td>
<td>[56]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Entropy, 298°K (cal/mole deg C)</td>
<td>19.3</td>
<td>[56]</td>
<td>16.38</td>
<td>[46]</td>
</tr>
<tr>
<td>Mechanical Properties</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hardness, diamond pyramid, 200-g load (kg/mm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>600</td>
<td>[52]</td>
<td>850</td>
<td>[52]</td>
</tr>
<tr>
<td>Electrical Properties</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electrical resistivity (ohm cm, 25°C)</td>
<td>30(10&lt;sup&gt;-6&lt;/sup&gt;)</td>
<td>[52]</td>
<td>25(10&lt;sup&gt;-6&lt;/sup&gt;)</td>
<td>[52]</td>
</tr>
</tbody>
</table>
TABLE IX

PROPERTIES OF ARC-MELTED THORIUM-URANIUM CARBIDES

<table>
<thead>
<tr>
<th>Property</th>
<th>Temperature ('С)</th>
<th>Value for</th>
<th>Value for</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Th₀.₉ U₀.₁ C</td>
<td>Th₀.₉ U₀.₁ C₂</td>
</tr>
<tr>
<td>Thermal conductivity,</td>
<td>200</td>
<td>0.13</td>
<td>0.20</td>
</tr>
<tr>
<td>(W/cm deg C)</td>
<td>800</td>
<td>0.17</td>
<td>0.25</td>
</tr>
<tr>
<td>Thermal expansion (%)</td>
<td>0-400</td>
<td>0.24</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>0-900</td>
<td>0.64</td>
<td>0.69</td>
</tr>
<tr>
<td>Hardness (kg/mm²) (1-kg load)</td>
<td>25</td>
<td>260</td>
<td>1100</td>
</tr>
<tr>
<td>Compressive fracture stress (lb/in²)</td>
<td>25</td>
<td>1.3 x 10⁵</td>
<td>1.5 x 10⁵</td>
</tr>
<tr>
<td></td>
<td>975</td>
<td>6.8 x 10⁴</td>
<td>2.3 x 10⁴</td>
</tr>
<tr>
<td>Thermal expansion coefficient a (degC⁻¹)</td>
<td>40-400</td>
<td>7.84 x 10⁻⁶</td>
<td></td>
</tr>
<tr>
<td></td>
<td>40-1000</td>
<td>9.31 x 10⁻⁶</td>
<td></td>
</tr>
</tbody>
</table>

a Values for 12.3% UC₂ taken from Ref. [54].

TABLE X

COMPATIBILITY PROPERTIES OF THORIUM-URANIUM CARBIDES

<table>
<thead>
<tr>
<th>Contact materials</th>
<th>Behaviour of (a) Th₀.₉ U₀.₁ C, (b) Th₀.₉ U₀.₂ C, (c) Th₀.₉ U₀.₁ C₂, (d) Th₀.₄ U₀.₂ C₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al, Zr</td>
<td>all four, no reaction in 1000 h at 540°С</td>
</tr>
<tr>
<td>Inconel, type 321 stainless steel</td>
<td>all four, no reaction in 1000 h at 815°С</td>
</tr>
<tr>
<td>Ta, W</td>
<td>all four, no reaction in 1000 h at 1095°С</td>
</tr>
<tr>
<td>Nb, Mo</td>
<td>all four, no reaction in 1000 h at 815°С</td>
</tr>
<tr>
<td></td>
<td>all four, no reaction in 100 h at 1095°С</td>
</tr>
<tr>
<td></td>
<td>all four, &lt; 0.002 in. penetration in 1000 h at 1095°С</td>
</tr>
<tr>
<td>H₂O at 90°С</td>
<td>(a), (c), ThC, ThC₂ all very poor</td>
</tr>
<tr>
<td>Santowax R at 350°С</td>
<td>Weight loss (mg/cm² d) (a), 2.0, (c), 0.3; ThC, 0.35; ThC₂, 0.5</td>
</tr>
<tr>
<td>NaK at 650°С</td>
<td>Weight loss (mg/cm² d) (a), 0.045; (c), 0.015</td>
</tr>
</tbody>
</table>

Thorium nitrides

The thorium nitrides reported are ThN, Th₂N₃, and Th₃N₄. The latter two may not both exist, and their use at high temperatures, if possible at all, would require high nitrogen pressures. The mononitride is stable to its melting point of 2730°C in the presence of sufficient nitrogen; less than
PROPERTIES OF THORIUM

1 atm is needed [59]. It has a face-centred cubic crystal structure (NaCl type) with a lattice parameter of 5.1584 Å and a narrow composition range. The decomposition pressure in atmospheres is given by

$$\log P = 8.086 - 33.244/T + 0.958 (10^{-17})T^6.$$ 

It can be prepared by induction melting of thorium under 2 atm N\textsubscript{2} or by hot pressing powder from decomposition of Th\textsubscript{2}N\textsubscript{3}. With ThC it forms a complete series of solid solutions [60]. Thorium mononitride is more reactive toward water than UN [61]. Otherwise, thorium nitride information is lacking except for some guessed thermodynamic properties.

**Miscellaneous thorium compounds**

Thorium forms two borides, ThB\textsubscript{4} and ThB\textsubscript{6}. Some available physical and thermal data [62, 63] indicate that the tetraboride is a promising ceramic material, and a few properties have been reported [58] for material with incorporated uranium. However, the nuclear usefulness of this material requires separated \textsuperscript{11}B. With silicon and beryllium, thorium forms the potentially useful Th\textsubscript{3}Si\textsubscript{2}, ThSi, and ThBe\textsubscript{13}. The data on these compounds is quite scanty, but a few properties have been studied for uranium substitution products [58]. Thorium forms several high-melting sulphides and an oxysulphide ThOS. Several of these show promising ceramic properties, but we have not yet completed a survey of them. The compounds ThS and Th\textsubscript{2}S\textsubscript{3} have received the most attention. There is no information to indicate that phosphides of thorium would have nuclear application.

**Summary**

The data on physical properties of thorium oxide are quite extensive. The extensive data on mechanical properties is misleading, however, since these properties depend greatly on the fabrication of the specimen. Data on diffusion in thoria is completely lacking. Compatibility with many materials is excellent, either known from experiment or safely predictable. However, information on thoria containing uranium is very meagre. Despite the regular isomorphous replacement, the disorder introduced precludes prediction of properties by interpolation between those of the components; the scant data on thermal conductivity prove this.

In contrast is the situation on the carbides, which are of interest only for application in nuclear energy. Information on these compounds is quite scanty, but the rate at which new knowledge is being reported is quite high. Reflecting the nuclear interest, the information on mixed carbides with uranium is keeping pace with or even exceeding that on pure thorium carbides.

The compounds ThBe\textsubscript{13}, ThB\textsubscript{4}, ThN, ThSi, Th\textsubscript{3}Si\textsubscript{2}, ThS, and Th\textsubscript{2}S\textsubscript{3} have been partially characterized for ceramic nuclear use.
REFERENCES

PROPERTIES OF THORIUM

[31] YUST, C.S., Oak Ridge National Laboratory, private communication.
[33] PIRANI, M., "Radiation properties of different substances within the temperature range 250°C to 800°C", J. Sci. Instrum. 16 (1939) 373-78.


1. INTRODUCTION AND SUMMARY

This paper reviews developments in the United States of America in the field of chemical reprocessing of thorium-bearing spent fuels from nuclear reactors. To limit the length of the paper, consideration is restricted to fuels of the most promising reactor types in the USA advanced converter reactor programme. Emphasis is placed on the differences in reprocessing methods and costs between these thorium fuels and standard low-enrichment uranium metal or oxide fuels clad in aluminum, zirconium, or stainless-steel alloys. The relationship between reprocessing and the rest of the fuel cycle for the most important proposed thorium-uranium recycle schemes is discussed. Head-end processes, starting with irradiated fuel elements and ending with a nitrate solution suitable for feed to solvent extraction, are described for the various fuel types. Solvent extraction flowsheets for separating the uranium and thorium from each other and from the fission products are evaluated. The interim and ultimate waste disposal problems associated with the various recycle schemes, head-end processes and solvent extraction flowsheets are presented. Finally, comments are made on the costs of reprocessing thorium fuels, by comparison with standard uranium fuels and with respect to the economic problems associated with sizing and starting a reprocessing plant when the fuel load is small initially but promises to increase substantially with time.

Primary consideration is given to (1) pyrolytic carbon-coated thorium-uranium oxide or carbide microspheres contained in massive graphite fuel elements, i.e. fuels for "Target" type, high-temperature, gas-cooled reactors (HTGR); and to (2) thorium-uranium metal, oxide or carbide clad with zirconium or aluminium alloys, which includes fuels for heavy-water-cooled and moderated reactors (HWR), heavy-water-moderated, organic-cooled reactors (HWOCR), and also spectral shift control reactors (SSCR) and light-water-cooled and moderated seed-blanket reactors (SBR or LPR). For purposes of comparison, standard fuel reprocessing methods and costs are assumed to be those typical of Hanford, Savannah River, and the Nuclear Fuel Services plant. The thorium fuels are all non-standard in at least some of the following aspects:

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation
(1) Graphite, carbon and carbide-type fuels may require pre-treatment steps, such as burning or pyrohydrolysis or grinding, not presently provided in industrial-scale reprocessing plants.

(2) Thorium fuels in general require more powerful dissolvents, e.g. more concentrated nitric acid and the presence of fluoride as a catalyst, than uranium fuels to get acceptable dissolution rates, and even so may dissolve at significantly lower rates than similar uranium fuels. The use of these dissolvents may complicate the succeeding feed adjustment, solvent extraction and waste disposal steps as a result of enhanced corrosion and dissolution of part or all of the cladding material along with the thorium.

(3) Thorium fuel dissolver solutions usually require a feed adjustment step to remove excess acid, perhaps all the way to an acid-deficient condition, (and perhaps also to decompose organic compounds in the case of carbide fuels after low-temperature hydrolysis or leaching) before the solvent extraction step.

(4) In solvent extraction, the phase equilibria and the contacting problems associated with third-phase formation are such that thorium fuels typically must be processed through a given size of equipment at a lower rate than uranium fuels.

(5) Thorium fuels as a class tend to have higher economic-optimum fissile enrichments and burn-ups than uranium fuels, and this may lead to throughput-rate limitations or other processing restrictions, because of criticality control or permissible fission-product off-gas release, etc.

(6) The fact that $^{233}\text{U}$ production involves the relatively slow decay of 27-d $^{233}\text{Pa}$ and the growth of the gamma-active daughters of $^{232}\text{U}$ and $^{228}\text{Th}$ into the recovered products complicates the pre-and post-processing handling of thorium fuels by comparison with uranium fuels.

(7) If thorium is recovered, the $^{228}\text{Th}$ and $^{234}\text{Th}$ in it will cause it to have a lower market value than virgin thorium, because of the extra fabrication costs involved, and it may even have a negative value in the sense that the cheapest thorium recycle scheme might include 10- to 15-yr storage of the recovered thorium to provide for decay of the $^{228}\text{Th}$.

(8) If thorium is not recovered and/or if aluminum nitrate is used as a salting agent in solvent extraction, the high-level fission-product waste will have to be stored in larger volume in the acid condition in stainless steel tanks, which is more expensive than the highly concentrated, neutralized storage in mild-steel tanks of wastes from standard uranium fuels.
Some of these differences can involve an increased cost of a factor of two or more on a weight basis, or may even prevent the processing of certain fuels, in a plant designed for standard uranium fuels. Because of their higher burn-up and/or thermal efficiency, some thorium fuels can stand the increased cost-per-unit-weight without incurring much of a cost-per-kilowatt-hour penalty; but for other fuels, especially those which cannot be processed in existing plants, the disadvantage is quite serious.

On the other hand, design and cost studies reviewed in this paper show that reprocessing plants built specifically to handle thorium-bearing fuels, using processes described in this paper, could process these fuels at only slightly higher cost-per-unit-weight than standard uranium fuels, hence permitting the thorium-fuelled advanced converter reactors to take almost full advantage of their higher burn-up and/or thermal efficiency in achieving lower power costs. Thus, the fuel processing problems of thorium-fuelled reactors are viewed as temporary "start-up" problems, perhaps involving some initial economic disadvantages for the early thorium-fuelled advanced converters, but which will fade away as the industry reaches a reasonable size.

2. HEAD-END PROCESSES

2.1. Burn-leach processes for HTGR fuels

This discussion will be limited to processing methods for HTGR fuels that contain pyrolytic carbon-coated thorium and/or uranium carbide or oxide fuel particles in massive graphite elements. A promising processing method consists of burning the fuel in a fluidized bed of inert alumina and then leaching with fluoride-catalysed nitric acid (Thorex reagent) to recover the uranium and thorium [1]. Laboratory- and engineering-scale studies of this process have been made with unirradiated prototype fuel specimens, and a few hot-cell experiments have been carried out with irradiated material.

The burn-leach process for graphite-base fuels is shown schematically in Fig. 1. Initially, the fuel is chopped or crushed to a convenient size for handling and fed to a fluidized-bed burner where it is burned at 700 to 750°C in a fluidized bed of granular alumina. Burning is started by injecting preheated oxygen into the fluidized bed and simultaneously heating the bed by external heaters. When the fuel starts to burn, the heaters are turned off, and the heat of reaction is removed by air-cooling the bed. For efficiency, continuous operation, with feeding of fuel, fresh alumina, and oxygen to the burner, and withdrawal of ash, all at the proper rates, is preferred. Under normal operation, nearly quantitative consumption of the oxygen is achieved, resulting in an off-gas composed mainly of CO₂ with less than 5% of carbon monoxide in the off-gas. Particles in the off-gas are removed mostly by filters, and a gas-cleanup system prevents the release of all radioactivity except the noble gases. After burning, the product bed is transferred to a leacher where the uranium and thorium are dissolved. The alumina may be recycled or discharged to waste. Uranium and thorium recoveries should be greater than 99.5%.
Design of the burner and the leaching system may be dependent on the type of fuel being burned and whether it is desirable to prevent isotopic mixing of the high $^{236}\text{U}$ content material remaining in the fuelled particle and the freshly bred $^{233}\text{U}$ in the thorium particle. Burning of fuels containing carbon-coated Th-U dicarbide particles converts the carbides to finely powdered oxides, dispersed homogeneously throughout the bed. Consequently, to recover the uranium and thorium, the entire bed must be leached. However, oxide fuel particles of high ThO$_2$ content may not be affected during combustion in a fluidized bed and probably could be separated from the alumina if desired before the leaching operation. This leads to the possibility of preventing isotopic mixing by a physical separation or by a selective chemical dissolution to separate the ThO$_2$ particles containing the bred $^{233}\text{U}$ from the U$_3$O$_8$ derived from either uranium carbide or uranium oxide fuel particles.

2.1.1. Fluidized-bed combustion of graphite-base fuel

2.1.1.1. Fuels containing carbon-coated carbide particles. Tests of the combustion of fuel containing carbon-coated carbide fuel particles were conducted in 2-in.-diam. and 4-in.-diam. fluidized beds [2, 3]. The 4-in.-diam. fluidized bed (Fig. 2) used at Oak Ridge National Laboratory for pilot plant studies had the essential features of a plant-size burner. The unit was made of nickel 201, although type 310 stainless steel would probably be preferred for a large-scale burner because of the better high-temperature mechanical properties of this material. The fluidized bed of alumina was an efficient heat-transfer medium and it diluted and suspended the fuel oxides formed during combustion so that the product was a free-flowing powder. The com-
bustion was easily controlled and good operation, with loading of the alumina up to 30 wt.% U-Th oxides, was achieved when the starting bed was composed of equal parts of 60- and 90-mesh fused alumina. Attrition of the alumina was negligible in a one-week test [3].

In a typical experiment, chopped or crushed fuel was added to approximately 20 kg of alumina, and the bed was heated to the ignition temperature of 650°C. Bed centerline temperatures and wall temperatures were held at about 750 and 700°C, respectively, by air-cooling the finned exterior of the fluidized bed. The CO₂ and CO contents of the off-gas were continuously monitored and were relatively constant when there was an excess of carbon in the burner. A decrease in the CO₂ and CO contents showed that the carbon inventory in the bed was being depleted and more fuel was added as needed to maintain the desired oxidation rate. Alumina was added periodically when the product was continuously withdrawn. Any small particles of carbon entrained in the alumina below the grid were rapidly burned in the hot oxygen, and it was possible to continuously withdraw a product stream containing less than 0.1% carbon and 30 wt.% U-Th oxides from the bottom of the bed. Toward the end of a combustion run, when the carbon concentration in the bed was low, it was necessary to supply heat to the burner to ensure combustion of the last traces of carbon. The superficial gas velocity in the bed was about 0.76 ft/sec at the bed mid-point pressure of 17.6 lb/in² abs. and average temperature of 725°C.

Continuous oxidation rates varying from 1.1 to 1.4 kg of carbon per hour were obtained in pilot plant tests with a 4-in.-diam. fluidized bed by varying the oxygen flow rate over the range of 1.3 to 1.6 scfm. Oxygen utilization decreased from 97 to 90% as the flow rate was increased. The heat-transfer coefficient from bed to wall was estimated at 85 Btu/h ft² degF and temperatures could be easily controlled by air cooling the finned outer wall. Plugging of the filters was not a problem, and the filter blowback system was not used during routine operations. Micropore filtration [3, 4] of the off-gas showed that practically no particles escaped through the primary sintered-metal filters. The typical off-gas consisted of about 90% CO₂, 5% CO, and 5% O₂. Corrosion of the burner was negligible [2].

2.1.1.2. Fuels containing carbon-coated oxide particles. To date, no fuel containing carbon-coated ThO₂ and UO₂ or mixed ThO₂-UO₂ microspheres has been burned in a fluidized bed. A potential problem with this fuel arises from the uncertainties concerning the integrity of the ThO₂ or mixed ThO₂-UO₂ microspheres after irradiation to projected burn-ups of 50 000 to 80 000 MWD/t. If the ThO₂ or mixed ThO₂-UO₂ microspheres retain their shape, it might be possible to separate them from most of the alumina after burning the carbon. This might result in a simpler leaching system in that only the fuel particles would be fed to the leacher. For example, preliminary tests have shown that ThO₂-UO₂ particles containing up to 75% ThO₂ are unaffected by oxygen at 800°C while pure UO₂ is readily oxidized to U₃O₈. However, if the particles are broken during irradiation or combustion, they will be dispersed throughout the alumina with the U₃O₈ derived from oxidation of the UO₂ seed particles; thus all the alumina must be leached.
2.1.1.3. Fuel and reprocessing systems to permit segregation of $^{236}$U.

In the TARGET concept [5] for the HTGR, the uranium fuel particles after irradiation contain a relatively large amount of $^{236}$U, the recycle of which is undesirable if high conversion ratios are to be achieved. Some possibilities for the design of the fuel and the reprocessing system to permit the $^{236}$U to be withdrawn from the fuel cycle are given in the following, which assumes fluidized-bed oxidation of the fuel as the first step:

<table>
<thead>
<tr>
<th>Fuel system</th>
<th>Segregation of the $^{236}$U might be achieved by:</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO$_2$</td>
<td>(1) Leaching the ash with dilute nitric acid to remove the $^{235,6}$U$_3$O$_8$ before dissolving the ThO$_2$-$^{233}$UO$_2$ particle in Thorex reagent.</td>
</tr>
<tr>
<td>UC$_2$</td>
<td>or (2) Physical separation of the ThO$_2$-$^{233}$UO$_2$ from the alumina containing the $^{235,6}$U$_3$O$_8$.</td>
</tr>
</tbody>
</table>

This oversimplifies the problem, since there are many possibilities for including refractory materials (such as ZrO$_2$, ZrC, SiC, BeO, etc.) in the fuel or fertile particle or as a coating for the same. Obviously, this would complicate the reprocessing of these fuels.

2.1.1.4. Fission product behaviour during combustion. The behaviour of the fission products was not studied during actual fluidized-bed combustion but was examined cursorily in laboratory-scale tube-furnace experiments. In one series of experiments [6], in which a large excess of oxygen was used to burn prototype Peach Bottom fuel irradiated to about 10 000 MWd/t, up to 35% of the caesium and 96% of the ruthenium were volatilized from the high-temperature zone during 6-h combustions at 800°C. Experiments [7] in the same equipment with a slightly irradiated fuel at 700°C showed that up to 1.1% of the caesium and 65% of the ruthenium were volatilized in 6 h, thus illustrating the desirability of low combustion temperatures. In each case, practically all the fission products were trapped in the cool end of the reaction tube and nearly all remaining activity was removed by filtering the off-gas through a sintered metal filter. The overall decontamination factor was greater than $10^4$ in all experiments. In other studies [8] only a small amount of caesium and ruthenium were volatilized from the hot zone when the fuel was burned in a deficiency of oxygen at 800°C.

Supplemental treatment of the off-gas may be required. An attractive method might be to mix steam with the off-gas, condense the vapour, and then filter through absolute filters. Waste-calcination work [9, 10] indicated that a system combining sintered-metal filters, condensation of vapour, and finally absolute filtration can yield decontamination factors $\geq 10^8$ for the off-gas. If rare gas retention ever becomes necessary for the reprocessing plant, the large amount of CO$_2$ in the off-gas may be a serious problem for the rare gas retention system.
2.1.2. Leaching of fluidized-bed products

2.1.2.1. Products from fuels that contain carbon-coated carbide particles. An efficient bench-scale batch leacher was devised [3] in which the leaching acid was recirculated upflow at a low rate through the bed, fluidizing the bed for better contact of the solids and the acid. After leaching, the product solution was drained from the bed, and the bed was washed with water. The bed material for these studies was produced by burning unirradiated Peach Bottom fuel compacts (carbon-coated Th-U dicarbide particles dispersed in a graphite matrix) in a fluidized bed of Norton RR alumina at 700 to 750°C. In laboratory-scale 5-h leaches, more than 99.5% of the uranium and thorium were recovered when the HNO₃ concentration was 4 M or higher, and when the HF concentration was 0.02 to 0.05 M. Uranium and thorium recoveries were inadequate with 13 M HNO₃ and with 2 M HNO₃ - 0.05 M HF. Less than 2% of the alumina was dissolved.

2.1.2.2. Products from fuels containing carbon-coated oxide particles. We have not yet done experiments with fuels containing carbon-coated mixed ThO₂-UO₂ particles. Leaching of uranium and thorium may simply involve dissolution of ThO₂-UO₂ microspheres in the presence of a small amount of alumina if the microspheres are physically separated from the alumina after burning. Laboratory tests showed that unirradiated ThO₂-UO₂ microspheres probably cannot be dissolved readily in a dilute Thorex solution; however, dense 300- to 600-μm-diam. ThO₂ microspheres were dissolved in 3 to 6 h in boiling 13 M HNO₃-0.05 M HF, even in the presence of a large excess of alumina. In other studies [11, 13], irradiated ThO₂-UO₂ pellets appeared to dissolve faster than unirradiated oxide, and a 6-h dissolution period is estimated to be adequate.

2.1.3. Conceptual design for a large HTGR fluidized-bed burner and leacher system

A conceptual design was prepared [1] for a head-end reprocessing facility to permit reprocessing HTGR fuel at a multipurpose reprocessing plant (e.g., the Nuclear Fuel Services, Inc. plant). The conceptual design of a large burner is shown in Fig. 3; it is a scale-up of the pilot plant burner and would be operated similarly. Provisions for removing heat from central portions of large fluidized beds must be made to avoid excessive centreline temperatures. It was assumed that the fuel contained only mixed ThO₂-UO₂ microspheres, that the microspheres would not be broken, and that a classifying operation after burning would permit leaching the microspheres in the presence of very little alumina. The conceptual design of the leaching and feed adjustment system is shown in Fig. 4. For criticality control, use of two geometrically safe slab leachers in series is envisaged for dissolving practically all the fuel particles before the solution and alumina slurry flow into large-diameter feed adjustment vessels. The leachers would be equipped with thermosiphon heating and solids pumping loops and would operate continuously in series. Leachant would be pumped into the first slab-shaped tank and maintained at its boiling point throughout the leaching system. Solids and solution from the first leacher would overflow continuously into
FIG. 2. Four-inch-diameter fluidized-bed burner for pilot plant studies
FIG. 3. Conceptual design for a large, fluidized-bed burner for graphite-base fuels
FIG. 4. Conceptual leaching and feed-adjustment system for graphite-base fuels that contain ThO$_2$-UO$_2$ microspheres

the second one. Alumina would be transported through the system without being attacked appreciably by the dissolvent. Solution from the second leacher would be transferred to a feed adjustment system where any small fuel particles still remaining in the leacher overflow would be dissolved rapidly.

A conceptual design of a complete HTGR head-end facility is shown in Fig. 5. It provides for fuel element receipt and storage, crushing, burning, leaching, and feed adjustment for up to 40 elements per day and up to 225 days per year in two parallel processing lines. The elements were assumed to be 4.5-in.-diam., 20-ft long graphite logs, each containing 107 kg of carbon plus 10.9 kg of thorium plus uranium plus fission products. A total capital investment of about $9 million was estimated for construction and start-up of this facility:

<table>
<thead>
<tr>
<th>Item</th>
<th>Cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Building, cells and services</td>
<td>$ 2,787,000</td>
</tr>
<tr>
<td>Process equipment</td>
<td>$ 1,098,000</td>
</tr>
<tr>
<td>Process piping</td>
<td>$ 906,000</td>
</tr>
<tr>
<td>Process and radiation instrumentation</td>
<td>$ 350,000</td>
</tr>
<tr>
<td>Site improvements and utilities</td>
<td>$ 481,000</td>
</tr>
<tr>
<td><strong>Sub-total</strong></td>
<td><strong>$ 5,622,000</strong></td>
</tr>
<tr>
<td>Design and contingency</td>
<td>$ 281,000</td>
</tr>
<tr>
<td>Interest, working capital</td>
<td>$ 607,000</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>$ 9,040,000</strong></td>
</tr>
</tbody>
</table>

2.2. Chemical decladding and dissolution processes

In this section we will consider primarily thorium-uranium oxide, metal and carbide fuels clad in zirconium or aluminum alloys, with only limited discussion of other fuel types such as those clad in stainless steel, since they are of less interest in advanced converter reactors.
2.2.1. Zirconium- or Zircaloy-clad fuels

Zirconium claddings can be separated chemically from core materials by either aqueous or non-aqueous methods. The aqueous method (Zirflex process) involves dissolution of the cladding in ammonium fluoride-ammonium nitrate solutions [14-17]. The non-aqueous techniques are oxidative disintegration in HF-O2 mixtures [18-20], the Thermox process [21] and hydrochlorination (Zircex process) [22]. After decladding, the core materials are dissolved in appropriate reagents in preparation for solvent extraction recovery of the uranium and thorium. Each of these methods is discussed briefly below.

2.2.1.1. Zirflex decladding [14-17]. Zirconium or Zircaloy claddings are readily dissolved in boiling 4 to 6 M NH4F - 0.5 to 1 M NH4NO3. The overall reaction is approximated by the equation:

\[ \text{Zr} + 6 \text{NH}_4\text{F} + 0.5 \text{NH}_4\text{NO}_3 \rightarrow (\text{NH}_4)_2\text{ZrF}_6 + 5 \text{NH}_3 + 15 \text{H}_2\text{O} \]

Actually, about 0.1 mole of hydrogen is also evolved per mole of zirconium dissolved. In the absence of ammonium nitrate a gas having the composition 33% H2 - 67% NH3 is liberated. The average dissolution rate of Zircaloy-2 that has been exposed to high-temperature air or water is about 5 mg/min cm²; thus, a 0.03-in-thick cladding is penetrated in 2 to 3 h. Since the solubility of (NH4)2ZrF6 at 25°C decreases markedly with increasing excess NH4F concentration, the most concentrated decladding waste solutions are obtained by consuming as much of the ammonium fluoride as possible. To optimize both dissolution rate and volume of waste solution, the decladding is conducted with an overall F-Zr atom ratio of about 7, making the maximum attainable zirconium concentration in the waste solution about 0.6 M. Pilot plant studies with unirradiated fuel [17] showed that the ammonia must be removed from the dissolver continuously to prevent an increase in pH of the solution and subsequent precipitation of zirconium oxide, which causes a marked reduction in the rate of reaction. Ammonia removal was effected by using a high boil-up rate and a high temperature in the downdraft condenser or by steam sparging at a rate that maintained a constant liquid level in the dissolver. Suitable materials of construction for the dissolver were stainless steel or Monel. The decladding solution probably will require centrifugation to recover core material fines before discharge to the waste system.

Preliminary experiments [23] with unirradiated ThO2-UO2 7 fuel pellets (4.2% UO2 7, 83% of theoretical density) indicated that zirconium claddings could be dissolved in 6 M NH4F - 1 M NH4NO3 with attendant uranium and thorium losses of less than 0.4%. Even lower losses are expected with higher density ThO2-UO2 fuel. No studies of the decladding of irradiated zirconium-clad ThO2-UO2 have yet been made; however, soluble uranium and plutonium losses in the decladding of Zircaloy-clad UO2 irradiated up to 17 000 MWd/t averaged only about 0.05% [24]. Thus, Zirflex decladding of thorium oxide core fuels appears feasible.

No data appear to exist on the behaviour of Th-U alloy in ammonium fluoride solutions. Uranium metal is attacked [15] at a rate of about
0.05 g/h cm² in boiling 6 M NH₄F with the rate increasing about a factor of 2 when the NH₄NO₃ concentration of the solution is 0.5 to 1 M. If Th-U alloy were attacked at a comparable rate, a significant fraction of the alloy would be converted to insoluble ThF₄ and UF₄. Consequently, the practicability of Zirflex decladding for Th-U alloy fuel is questionable at this time.

Reaction of ThC-UC with ammonium fluoride solutions has not been investigated. If the irradiated carbide were not passive (irradiated UC is passive in boiling water and 6 M NaOH [25] hydrolysis with NH₄F would be expected to yield solid ThF₄ - UF₄ and a gas composed mainly of methane and hydrogen [26, 27]. This formation of an insoluble fluoride would be highly undesirable.

2.2.1.2. Non-aqueous decladding [18-20]. Treatment of zirconium-clad fuels with HF-O₂, with steam, or with HCl in fluidized beds of inert alumina, such as Norton RR and Alcoa T-61 grades, results in a product from which uranium and thorium can be recovered by acid leaching. Reaction of zirconium or Zircaloy with a gaseous mixture of HF and oxygen results in conversion of zirconium to ZrO₂ and simultaneous disintegration of the cladding. Optimum conditions appear to be about 625°C and a gas mixture containing 20 to 40% HF [18]. The penetration rate is at a maximum of about 0.04 in/h with 40% HF-60% O₂ at 625°C. The product bed can contain a total of up to about 40 wt.% ZrO₂ plus core oxides such as U₃O₈ and ThO₂, and always contains 3 to 8% fluorine. This amount of fluorine is very high relative to the amount of uranium (F-U atom ratio in the bed usually is 1 to 3). Thus, removal of the fluorine by pyrohydrolysis before leaching is recommended. In laboratory-scale experiments, greater than 90% of the fluorine was removed from typical product beds in 4-h reactions with steam (1 atm pressure) at 600°C. During HF-O₂ decladding, UO₂ cores are converted primarily to
U\textsubscript{3}O\textsubscript{8} powder which is dispersed throughout the bed. The behaviour of ThO\textsubscript{2}-UO\textsubscript{2}, Th-U, and ThC-UC in this system has not yet been tested. It is expected, however, that both the alloy and carbide would react, although perhaps slowly. Mixed oxide of high ThO\textsubscript{2} content would be expected to be relatively inert.

A similar technique for converting zirconium claddings to ZrO\textsubscript{2} powder involves reaction of the cladding with oxygen-water vapour mixtures at about 825°C using nitrogen as a catalyst [21]. This Thermox decladding of zirconium-clad UO\textsubscript{2}, followed by oxidation of the UO\textsubscript{2} to U\textsubscript{3}O\textsubscript{8}, and leaching of the product with nitric acid gave uranium recoveries of greater than 99%. The Thermox method has not been tested with Th-U alloy or ThO\textsubscript{2}-UO\textsubscript{2} fuels. Of the core materials being considered, only the carbide would be expected to react rapidly with the decladding reagent.

An alternative non-aqueous decladding route is removal of the zirconium cladding as volatile ZrCl\textsubscript{4} by reaction with HCl at about 500°C [22]. The ZrCl\textsubscript{4} would be converted to ZrO\textsubscript{2} in a separate fluidized bed by reaction with steam allowing disposal of the cladding as a solid waste. The application of this Zircex process to thorium-bearing fuels has not yet been studied. Again, only the carbide core materials would be expected to react with the HCl during decladding. The oxide fuels are practically inert.

2.2.1.3. Core dissolution. After decladding, the core materials would be dissolved to produce solutions suitable as feeds for a solvent extraction recovery system. Thorium metal, thorium oxide, and ThO\textsubscript{2}-UO\textsubscript{2} mixtures dissolve in nitric acid containing small amounts of hydrofluoric acid as catalyst (fluoride-catalysed nitric acid) [25, 28]. Dissolution of unirradiated ThO\textsubscript{2}-UO\textsubscript{2} in the optimum reagent, 13 M HNO\textsubscript{3}-0.05 M HF, is slow, with up to 40 h being required for a batch dissolution yielding a 1 M Th(NO\textsubscript{3})\textsubscript{4}-9 M HNO\textsubscript{3} solution. Irradiated ThO\textsubscript{2}-UO\textsubscript{2} dissolves much more rapidly, regardless of the method of preparation. Pelletized, arc-fused, and sol-gel-derived oxides (about 5% UO\textsubscript{2}) were greater than 99.9% dissolved in about 7 h after irradiation to 3000 to 98 000 MWD/t (U+Th) [11-13, 29]. Irradiated thorium metal fuel has been processed on a pilot-plant scale [30, 31]. Metal slugs were dissolved in boiling 13 M HNO\textsubscript{3}-0.04 M HF using an 8-h dissolution period and leaving a 100% heel. The dissolution product was 6.5 M in HNO\textsubscript{3} and 1 M in Th. Thus, a feed adjustment step is probably required after dissolution of both ThO\textsubscript{2}-UO\textsubscript{2} and Th-U alloy to provide suitable feed solutions for solvent extraction.

After non-aqueous conversion of the cladding to ZrO\textsubscript{2}, or its removal as ZrCl\textsubscript{4}, the product bed would be transferred to another vessel and leached with fluoride-catalysed nitric acid. Tests with simulated zirconium-clad sol-gel ThO\textsubscript{2}-UO\textsubscript{2} fuel that had been treated with HF-O\textsubscript{2} showed that when the fluorine present in the product bed is removed by pyrohydrolysis, greater than 99% of the uranium and thorium, but only up to 15% of the ZrO\textsubscript{2}, are leached in 5 h with boiling 13 M HNO\textsubscript{3}-0.05 M HF. The product solution was about 0.2 M in Th, 0.02 M in U, 0.02 M in Zr, 0.1 M in F, and 0.03 M in Al. If the fluorine had not been removed from the bed before leaching, up to 70% of the ZrO\textsubscript{2} and 90% of the fluorine would have been leached and the product solution would have been about 0.2 M in Th, 0.1 M in Zr, and 0.4 M in F. In all tests with fluidized-bed products, less than 2% of the alumina
was dissolved. The leached alumina can, therefore, either be recycled or discharged to waste.

Dissolution of ThC has received only slight attention. Preliminary laboratory studies [32] indicate that arc-melted ThC dissolves readily in boiling fluoride-catalysed nitric acid but only slowly in nitric acid itself. Dissolution in fluoride-catalysed nitric acid results in a large fraction of the carbide carbon being converted to soluble organic species such as oxalic acid and mellitic acid. In this respect, the behaviour of ThC is identical to that of the uranium carbides in nitric acid [33]. Preparation of a suitable solvent extraction feed would, therefore, probably require dissolution of the ThC-UC in 13 M HNO₃-0.05 M HF followed by digestion of the resultant solution in acid permanganate to oxidize most of the soluble organic species. Such a process was tested on a laboratory scale with irradiated UC specimens [34]. An alternative to direct dissolution in nitric acid solutions is pyrohydrolysis of ThC followed by dissolution of the resulting ThO₂ in fluoride-catalysed nitric acid. Although this method has not yet been studied, it is expected that ThC will behave like UC. Uranium monocarbide reacts rapidly with steam at 700 to 750°C, to give UO₂, CO₂, CO, and hydrogen[34]. Pyrohydrolysis of irradiated UC resulted in practically no volatilization of fission products. Combustion, in oxygen, is an alternative to pyrohydrolysis for carbide fuels, although the rate of oxidation of arc-melted carbides at 700 to 750°C is lower than the rate of reaction with steam. This fact, and the possibility of volatilizing ruthenium and caesium during combustion, makes the pyrohydrolysis method an attractive non-aqueous approach.

Other core materials such as UO₂-ZrO₂ and Th-Zr alloy have been considered for various reactors. No satisfactory aqueous dissolution process is evident for UO₂-ZrO₂, but it might be possible to dissolve the alloy in fluoride-catalysed nitric acid.

2.2.2. Aluminium-base clad fuels

Dispersions of Al₂O₃ in aluminium (designated as SAP or AMP) are being considered as fuel claddings, especially for organic-cooled reactors. Very little work has been done on the processing of SAP-clad fuels. Preliminary laboratory-scale experiments show that SAP can be dissolved either in NaOH-NaNO₃ solutions or in mercury-catalysed nitric acid solutions. In experiments using boiling 2 M NaOH-1.78 M NaNO₃ (Na-Al atom ratios of 2 and 4), the initial dissolution rate of a SAP sample containing about 6% Al₂O₃ was about 20 mg/min cm². This rate is high enough to allow penetration of a 0.03-in-thick clad in 2 to 3 h, and is about the same as that obtained for pure aluminium under the same conditions. Reaction with the NaOH-NaNO₃ solution left an Al₂O₃ residue corresponding to about 80% of the alumina in the original sample. On the other hand, dissolution of the SAP in boiling 4 M HNO₃-0.005 M Hg(NO₃)₂ (HNO₃-Al mole ratios of 4 to 8) resulted in practically complete solubilization of the sample, although the rate of reaction was much lower than that obtained in NaOH-NaNO₃ solution. Complete dissolution required about 20 h; the initial rate of dissolution was only about 0.5 mg/min cm². In contrast, type 2S aluminium and extruded 15% U-85% Al alloy dissolved [35] under the same conditions at rates of about 140 mg/min cm².
Soluble losses of uranium and thorium in caustic decladding should be negligible with each of the core materials being considered. Although un-irradiated ThC reacts readily with NaOH solutions [36], it is highly probable that after irradiation the carbide will be inert, as is the case with uranium carbide [25]. If this is true, caustic decladding of carbide (and the other types of) fuels probably would be a practicable approach.

2.2.3. Stainless steel-clad fuels

Stainless steel claddings can be chemically separated from core materials either by aqueous or non-aqueous methods. The aqueous method (Sulfex process [14, 37-40]) involves dissolution of the cladding in boiling 4 to 6 M H$_2$SO$_4$. The non-aqueous method is oxidative disintegration in HF-O$_2$ mixtures [18-20].

2.2.3.1. Sulfex decladding. With a 200% stoichiometric excess of boiling acid, the initial dissolution rate increases from about 2 to 30 mg /min cm$^2$ as the sulphuric acid concentration increases from 2 to 8 M. In cold pilot plant studies, penetration rates of 0.003-0.004 in/h were obtained in a recirculating dissolver [40]. Stainless steel that has been in contact with high-temperature water may be passive to sulphuric acid; in this event, dissolution is initiated by contacting the fuel with a piece of soft iron. The solubility of stainless steel sulphates at 25°C decreases from about 80 to to 20 g of stainless steel per litre as the sulphuric acid concentration increases from 2 to 8 M [38]. Nickel appears to be suitable as a material of construction for the dissolver.

Irradiated UO$_2$ and ThO$_2$-UO$_2$ are practically inert to boiling 4 to 6 M H$_2$SO$_4$. Decladding of stainless steel-clad UO$_2$ fuel specimens that had been irradiated up to 28 200 MWd/t resulted in soluble uranium and plutonium losses of only about 0.05% [24]. Similar experiments with stainless steel-clad ThO$_2$-UO$_2$ fuel [11-13] irradiated up to about 25 000 MWd/t showed that uranium and thorium losses were 0.5% or less. No tests have been made of the reactivity of irradiated Th-U alloy in sulphuric acid; however, experiments with unirradiated uranium metal [22] showed it to be relatively inert to boiling 2 to 8 M H$_2$SO$_4$. The reaction of ThC with sulphuric acid has not been studied; however, both unirradiated UC and UC irradiated up to 6000 MWd/t reacted with 6 M H$_2$SO$_4$ at 80°C yielding a gas composed mainly of methane and hydrogen and a solid which was probably U(SO$_4$)$_2$·4H$_2$O [25]. Conversion of the uranium to an insoluble sulphate is highly undesirable; thus, the use of Sulfex decladding for carbide fuels will probably not be practical.

After Sulfex decladding, the core materials would be washed and dissolved by the techniques described in section 2.2.1.3.

2.2.3.2. HF-O$_2$ disintegration. Stainless steel-clad fuels react with gaseous HF-O$_2$ mixtures in a fluidized bed in a manner similar to that of zirconium [18-20]. The stainless steel is converted to its respective oxides at the optimum temperature of about 650°C at a rate of about 0.06 in/h. The optimum gas composition appears to be about 40% HF-60% O$_2$. Bed products containing up to 30% U$_3$O$_8$ and 15% stainless steel oxides have been produced.
from stainless steel-clad UO₂ fuels. After removal of the fluorine by pyro-
hydrolysis, leaching with 1 to 15 M HNO₃ resulted in the recovery of greater
than 99.9% of the uranium. The product solution was about 0.2 M in U. The
amount of iron oxide dissolved increased from 16 to 85% as the nitric acid
concentration increased from 1 to 15 M. As discussed in section 2.2.1.2,
the effect of HF-O₂ on carbide, metal, or ThO₂-UO₂ cores has not yet been
investigated.

2.3. Mechanical head-end processing

The mechanical approach to head-end processing of spent reactor fuels
has been investigated during the past eight years in the USA. The investi-
gation has included disassembly, mechanical decladding, grind-leach, and
shear-leach methods. Some of these pre-chemical mechanical treatment
methods may offer attractive alternatives to the heretofore discussed chemi-
cal processes for thorium-bearing advanced converter fuels, e.g. crushing,
grinding and leaching of graphite fuels, and the shearing and leaching of
metal-clad fuels.

2.3.1. Crushing, grinding and leaching of graphite-type fuels

In the burn-leach process described in section 2.1, crushing of the
graphite fuel is required as the first step unless the coated particles can
be separated from the massive graphite log. Following crushing, an alterna-
tive to burning the graphite in a fluidized bed would be to further size-reduce
the rough crushed fuel by additional crushing or grinding. All of the coated
particles must be broken to permit recovery of fissile and fertile values by
leaching. At ORNL, investigation of this alternate route of grind-leach has
begun with HTGR fuel. Some very preliminary early work [41] has indi-
cated wear problems in hammer-mill crushing equipment and difficulties in
attaining complete leaching and washing of the graphite fines. Crushing,
either with or without burning to destroy the bulk graphite, followed by
grinding and leaching may be the only way to recover fuels with refractory
coatings such as SiC on the fuel particles.

2.3.2. Shear-leach processing

The shear-leach process has been intensively investigated [42-44] and
is to be used commercially by Nuclear Fuel Services, Inc., to process
power reactor fuels clad with stainless-steel or zirconium alloys and
containing cores of UO₂, ThO₂-UO₂, U-Mo metal, or Th-U metal. The
process, developed at Oak Ridge National Laboratory, involves the shearing
of the fuel bundle into short lengths to expose the fuel oxide or alloy, after
which the exposed fuel is leached from the cladding.

The method is capable of processing any aluminum-, stainless steel-, or
Zircaloy-2-clad oxide or metal fuel, using the same equipment. Currently,
there is no apparent advantage in processing graphite fuels by shear-leach,
unless a shearing or breaking operation is used to subdivide graphite fuel
logs. There are distinct advantages for the metal-clad fuels with core
materials which can be leached without dissolving the clad. One consider-
able advantage is the lower cost of storing the cladding material as metal waste rather than as Zirflex or Sulfex type liquid waste. It has been estimated that the waste storage cost for the leached metal clad is about 1/20th the cost of storing the corresponding chemical decladding waste [43-45]. Existing processing plants could be adapted by the addition of a mechanical head end to process a greater variety of fuel in conventional stainless steel equipment.

2.3.2.1. Shear-leach flowsheet. A typical shear-leach process flowsheet for stainless-steel-clad or Zircaloy-2-clad uranium oxide and/or thorium oxide fuel is presented in Fig. 6. Fuel assemblies that have been manufactured by high temperature brazing are disassembled by sawing off the inert end fittings, sheath, and tube sheets. The resulting fuel tube bundles are then sheared. The sheared pieces are collected in a perforated basket containing a consumable carbon steel liner, and leached in a batch leacher. The liner is required to contain the fuel fines during transport of the basket to the leacher. The leached cladding is sent to underground waste storage along with the other metallic scrap from the fuel element.

The newer type of fuel element that has been assembled by retaining the fuel rod with wire grids or spring clips can be sheared intact; however, the grids and clips tend to remain as large pieces, complicating the shear operation and basket loading somewhat. An alternative method is the withdrawal of tubes from the parent assembly. In demonstration tests with Consolidated Edison Core B fuel, mechanical equipment, consisting of a hydraulic cylinder, bumper, support rack, elevating jacks, and ejector, withdrew 14 fuel tubes simultaneously using a force of only 300 lb. Fuel tubes removed from the fuel assembly in this manner can be satisfactorily sheared as a loose bundle.

The 250-ton-prototype shear has been used successfully to shear unirradiated stainless-steel-clad uranium oxide and thorium-uranium-oxide fuel assemblies up to about 6 in. square, and containing up to 144 fuel tubes, into lengths of from 1/2 to 2 in. Zircaloy-2-clad oxide-type fuel has also been sheared satisfactorily, with some sparking but without encountering any significant safety problems caused by the presence of Zircaloy-2 metal fines. Zircaloy-2-clad uranium metal fuel assemblies of the NPR type were successfully sheared into 1/2- to 2-in. lengths. In this case also, some sparking occurred during shearing, but there appeared to be no real fire hazard involved.

2.3.2.2. Shear-leach design concept. A conceptual mechanical head-end and leaching equipment layout for shear-leach processing of about one metric ton of fuel per day is illustrated in Fig. 7. This layout is based largely on the results of the developments carried out over the past several years. The basic unassembled equipment cost, exclusive of the shielded cell, manipulators and supporting facilities, is very roughly estimated to be $600,000, broken down as follows:

<table>
<thead>
<tr>
<th>Shear</th>
<th>$225,000</th>
<th>Multiple tube puller</th>
<th>$10,000</th>
</tr>
</thead>
<tbody>
<tr>
<td>Handling table (carriage)</td>
<td>20,000</td>
<td>Fuel leach baskets (36)</td>
<td>36,000</td>
</tr>
</tbody>
</table>
FIG. 6. Shear-leach process for spent reactor fuels
With the exception of remote maintenance, all operations have been investigated sufficiently to show that each is feasible in a hot cell.

2.3.2.3. Leaching. Some typical bench-scale data are presented in Table I for shearing and leaching of unirradiated UO₂-ThO₂ and UO₂. The leacher used was similar to that of Fig. 8, which was used for engineering-scale studies.

In the case of Zircaloy-2-clad urania-thoria fuels, some of the Zircaloy cladding and fines dissolved in the fluoride-catalysed nitric acid required for core dissolution [47]. In tests conducted with pelletized, sol-gel and arc-fused thoria containing 4 to 5% urania in boiling 13 M HNO₃-0.04 M HF-0.04 M Al(NO₃)₃ dissolvent in the presence of Zircaloy-2 cladding and fines, from 1-5% of the massive cladding dissolved along with 60-80% of the minus-10 mesh fines. Recent shearing tests with Zircaloy-clad UO₂ indicate less than 1% of minus-10 mesh fines are formed. The rate of dissolution of ThO₂-UO₂ in fluoride-catalysed nitric acid was diminished in the presence of zirconium but not enough to render shear-leach unfeasible for this type fuel.

Hot cell tests on the batch dissolution and leaching of irradiated sol-gel-derived, pelletized, or arc-fused ThO₂-UO₂ in boiling 13 M HNO₃-0.04 M NaF-0.04 M Al(NO₃)₃ indicated that irradiation increased the dis-
<table>
<thead>
<tr>
<th>Fuel</th>
<th>Cladding</th>
<th>Shearing force (ton)</th>
<th>Recommended sheared length (in.)</th>
<th>Core dislodged (%)</th>
<th>Clad dislodged (%)</th>
<th>Packing density (g/cm³)</th>
<th>Void fraction (%)</th>
<th>Time to batch leach 99.9% (h)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO₂</td>
<td>Stainless steel</td>
<td>50-90</td>
<td>1</td>
<td>36</td>
<td>2</td>
<td>4.8</td>
<td>50</td>
<td>1‡</td>
<td></td>
</tr>
<tr>
<td>(pellets)</td>
<td>or Zircaloy-2</td>
<td></td>
<td>1½</td>
<td>28</td>
<td>2½</td>
<td></td>
<td>55</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>UO₂-ThO₂</td>
<td>Stainless steel</td>
<td>50-75</td>
<td>½</td>
<td>85</td>
<td>8</td>
<td>4.4</td>
<td>48</td>
<td>8</td>
<td>Sparged</td>
</tr>
<tr>
<td>(pellets)</td>
<td></td>
<td></td>
<td>1</td>
<td>36</td>
<td>2</td>
<td></td>
<td>50</td>
<td>12</td>
<td>Not sparged</td>
</tr>
<tr>
<td>UO₂-ThO₂</td>
<td>Stainless steel</td>
<td>50-75</td>
<td>½</td>
<td>85</td>
<td>8</td>
<td>4.4</td>
<td>48</td>
<td>20</td>
<td>25% heel</td>
</tr>
<tr>
<td>(sol-gel)</td>
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<td></td>
<td>1</td>
<td>36</td>
<td>2</td>
<td></td>
<td>50</td>
<td>65</td>
<td>No heel</td>
</tr>
</tbody>
</table>
solution rate over unirradiated oxides, with up to 95% in solution at 8 h and 99.8% in 24 h [11]. Greater than 99.8% of the thorium and uranium was recovered in leaching tests with sheared fuel pieces. Uranium and thorium losses were less than 0.05%. In other tests, sheared stainless-clad UO₂ irradiated to about 8000 MWd/t was easily leached in 4 M HNO₃ [48]. Only about 0.6% of sheared stainless-steel cladding dissolves in fluoride-catalysed nitric acid in 20 h.

The batch leaching of stainless steel clad unirradiated UO₂-ThO₂ pellets sheared into 1/2- or 1-in. lengths has been investigated in an engineering-scale Pyrex glass and stainless steel leacher (Fig. 8). Dissolvent is circulated by convection. Variables affecting dissolution rates were studied with boiling (120°C) 12.7 M HNO₃-0.1 M Al(NO₃)₃-0.04 M NaF as the dissolvent.
Typical UO$_2$-ThO$_2$ leaching data are presented in Fig. 9. The consumable carbon steel liner dissolves almost immediately. During the dissolution of the liner (about 2 min) 14 to 38% of the core is discharged from the basket and settles to the bottom of the leacher. A dissolvable 0.001-in-thick carbon steel liner adds about 11 g(Fe)/kg of uranium or thorium to the solvent extraction feed.

The data of Fig. 9 show the effect on dissolution time of terminal thorium loading, boil-up rate as represented by steam pressure, and a comparison of 1/2 and 1 in. sheared lengths. The basis of the comparison is the time to dissolve 99.9% of the fuel. It required 8.5 h to produce a solution 0.5 M in thorium at 60 lb/in$^2$ gauge steam as compared to 26 h to produce a 1 M thorium solution. Fifteen hours were required to attain 99.9% dissolution when producing a solution 0.5 M thorium at 20 lb/in$^2$ gauge steam as compared to the 8.5 h at 60 lb/in$^2$ gauge steam. About 18 to 19 h were required to leach both 1/2 and 1 in. sections when operating at the same conditions of terminal thorium loading and steam pressure. While producing a 1 M thorium solution at 20 lb/in$^2$ gauge steam, only 95.5% was leached in 24 h as compared to 99.9% leached in 26 h at 60 lb/in$^2$ gauge steam. It was concluded that a rapid boil-up rate enhances dissolution and 1 in. sections are leached as efficiently as 1/2 in. sections. The amount of core remaining as unleached UO$_2$-ThO$_2$ was negligible in all runs. The empty hulls or leached shells were washed free of product solution by four separate water washes. The volume of wash water used each time was about one-fifth of the volume of empty hulls.

In sharp contrast to thoria fuels, unirradiated stainless clad UO$_2$ sheared into 1-in. lengths are easily completely leached in the same leacher with boiling 7 M HNO$_3$ in about 2 h.

Although shear-leach studies have not yet been performed using the advanced converter fuels under discussion, ample information has been
REPROCESSING METHODS AND COSTS

developed with stainless-clad and Zircaloy-2-clad urania-thoria fuels to indicate that the projected fuels can be processed by shear-leach techniques. There are, however, two principal areas of doubt: (1) SAP cladding may tend to dissolve in the fluoride-catalysed nitric acid dissolvent, and (2) carbide fuels may cause an unusually high wear rate of shear blades. There are also the chemical problems associated with the hydrolysis of carbides.

2.3.2.4. Recommended practice. In applying the shear-leach process to a given fuel, the following practices are recommended:
(a) Use sheared lengths of 1 in.
(b) Use perforated basket(s) in single or multi-legged recirculating type leacher. The free or open area of basket can range from 5% to 25% with the sheared fuel retained by a dissolvable liner of carbon steel or aluminum.
(c) Leach UO₂-ThO₂ to a 25% heel in a period of 20-25 h.
(d) Use hull wash water (or acid) to make up acid for next leaching step.

3. SOLVENT EXTRACTION PROCESSES

3.1. General flowsheet considerations

The emphasis of this discussion is on solvent extraction flowsheets suitable for reprocessing plants designed specifically to support a thorium-fuelled power reactor industry, with secondary consideration given to the problems of processing thorium fuels in plants designed to handle standard uranium fuels. Recovery and purification of both the uranium and the thorium is emphasized, since discarding the thorium with the high-level fission-product waste is not a desirable long-term solution for a large thorium-fuelled power reactor industry from either the fuel-utilization or the waste-disposal points of view. The implications of the thorium recycle scheme, whether immediate recycle or delayed recycle after decay of \(^{228}\)Th, on the choice of solvent extraction processes are considered.

3.1.1. Standard uranium fuels

The standard uranium fuel reprocessing method is the Purex Process [49, 50] based on extraction of both the low-enrichment uranium and the plutonium with 30% tri-n-butyl-phosphate (TBP) in a suitable diluent. The uranium and plutonium may be partitioned in the first cycle of extraction, or they may be co-stripped in the first cycle and partitioned in a second cycle. After partitioning, additional decontamination of the separated uranium and plutonium is obtained by one or more additional cycles of solvent extraction or, in the case of plutonium, by anion exchange. For high-enrichment \(^{235}\)U fuels, the 25-TBP Process [51] recovers the uranium by extraction with 1.5-to-6% TBP if aluminum nitrate is used as a salting agent or with 10-to-30% TBP if only nitric acid salting is desired. The corresponding processes for thorium fuels are described below in some detail. The differences in the flowsheets for uranium fuels and thorium fuels are: (1)
uranium is the major constituent in one case and the minor constituent in the other; (2) thorium is extracted less strongly than uranium; and (3) the build-up of the gamma-active daughters of $^{232}\text{U}$, $^{228}\text{Th}$ and $^{234}\text{Th}$ in the recovered products makes high-degree decontamination from fission products a relatively less important consideration than in uranium-plutonium recovery. Standard uranium fuels usually can be dissolved to give acceptably high uranium concentration and acceptably low excess acid concentration for feeding directly to solvent extraction. On the other hand, thorium fuels usually require a feed adjustment step after dissolution to increase the thorium concentration and remove excess acid.

3.1.2. Protactinium

Protactinium recovery is not assumed to be of interest in power reactor fuel processing for the purposes of this discussion. Since the 27-d half-life $^{233}\text{Pa}$ is not normally extracted with the uranium [52], its primary effect on processing is the pre-processing time delay required for its decay to negligible levels. For thorium power reactor fuels this probably means a minimum of 120 d between reactor discharge and fuel processing and a typical delay of 180 to 210 d. This compares with a minimum delay of 90 d and a typical delay of 120 to 150 d for uranium power reactor fuels, for which the controlling factor is the decay of 8-d $^{131}\text{I}$.

3.1.3. Extraction of uranium only

When only the uranium is to be recovered from the thorium fuel, dilute TBP or di-sec-butyl-phenylphosphonate (DBPP) may be used for the extraction. The Acid Interim-23 Process [53] uses 2.5-to-10% TBP (Fig. 10). The higher concentration permits higher processing rates but lower decontamination from fission products and a smaller separation factor from thorium would result. The lower concentration might be useful for criticality control, at the price of reduced throughput. The Kilorod Interim-23 Process [54, 55, 56] used 2.5% DBPP, which has a higher uranium-thorium separation factor than TBP by about a factor of 4 and also provides excellent decontamination from fission products. The thorium which remains in the aqueous phase may be recovered by a second extraction, either immediately or at a later date, or may be permanently discarded with the fission products. As discussed below, however, if the thorium is to be recovered a simultaneous co-extraction with the uranium probably is preferable.

These Interim-23 flowsheets, using thorium nitrate as the primary salting agent, are, of course very similar to the 25-TBP flowsheets using aluminum nitrate as salting agent.

3.1.4. Co-extraction of uranium and thorium

Several systems have been developed for the co-extraction of uranium and thorium with TBP. In the original Thorex flowsheet [57] aluminium nitrate resulting from the dissolution of the aluminium cladding of the thorium metal slugs acted as the salting agent for the extraction of thorium and uranium from an acid-deficient solution into 42.5% TBP. A modification
of this process involves a similar co-extraction from a solution containing both aluminium nitrate and nitric acid [58]. This modification gives good, though not equal, decontamination and has the advantage of not requiring a feed adjustment step; however, the acid in the feed decreases the processing capacity of a given size plant by decreasing the solubility of the thorium-TBP complex in the diluent, requiring operation with lower thorium concentration in the solvent phase to avoid formation of a third phase (i.e. a second organic phase)[59]. Another variation of the Thorex flowsheet is the Acid Thorex Process [60] shown in Fig. 11, for fuel solutions which do not contain aluminium. It involves the extraction of thorium and uranium from an acid deficient solution into 30% TBP, with nitric acid added at a lower stage in the contactor to provide salting when the thorium nitrate concentration has been reduced. This process results in maximum processing capacity and excellent decontamination, and reduces the volume of waste to be stored by eliminating the non-volatile aluminium from the aqueous waste. It does require a feed adjustment to produce the acid-deficient condition.

3.1.5. Effects of fuel type and cladding

Any of the afore-mentioned flowsheets can be adapted for the processing of either oxide or metal fuels. Carbide fuels may also be processed, but if the thorium is dissolved by a low-temperature hydrolysis in nitric acid a feed adjustment step would be required to destroy the organic materials in the solution before feeding it to solvent extraction [34].

The type of cladding must also be considered in the choice of a fuel recovery process. If the clad is dissolved with the fuel the added metal salts will act as salting agents for the solvent extraction, but they also will restrict the flowsheet to be chosen and affect the waste disposal operations.
Large quantities of stainless steel or zirconium salts in the solution would eliminate consideration of acid-deficient flowsheets since large amounts of precipitates would be formed in these cases. Even small amounts of solids in the feed, resulting from partial dissolution of the cladding or even from high concentrations of fission products in high burn-up fuels, may cause trouble. Batch contactors or pulse columns can handle solutions containing up to several per cent solids but mixer-settlers are usually designed for solids-free solutions.

3.1.6. Non-nitrate systems

All of the fuel recovery systems being considered at present are based on nitrate solutions. Small amounts of other anions, such as fluoride, may be tolerated though adjustments may have to be made to compensate for their presence. If other solvents, such as hydrofluoric, hydrochloric or sulphuric acids, were required to dissolve the fuel, new solvent extraction flowsheets would have to be developed for these systems, perhaps using other organo-phosphorus compounds or amines.

3.1.7. Equipment consideration

A few equipment items should receive special consideration in a thorium processing facility. A feed adjustment tank, in which the dissolved fuel can
be heated to \(\sim 160^\circ C\), adds to flexibility by allowing a choice between acid and acid-deficient feed solutions for the solvent extraction system, and would also provide a means of destroying the organic materials which may be formed during the dissolution of carbide fuels. Equipment designers also should consider the possibility of third-phase formation in the thorium extraction systems. In a mixer-settler, the second organic phase accumulates between the organic and aqueous exit parts of the settler so that it cannot move out of the equipment. Since there is low turbulence in the settler unit, redissolution of this TBP heavy phase in the organic system is quite difficult. As a result, the operating conditions must be maintained conservatively away from the region of third-phase formation. On the other hand no major difficulties occur if there is a small amount of third-phase formation in a pulse column and recovery is practically instantaneous when operating conditions are corrected. Also, as mentioned already, pulse columns have a much greater tolerance for solids in the aqueous feed to the solvent extraction system.

### 3.2. Power reactor fuel processing

Figure 12 shows a general purpose solvent extraction flowsheet for thorium-bearing power reactor fuels. Insofar as practicable, cladding materials and any other unnecessary cationic or anionic constituents should be kept out of the dissolver solution, to permit a choice of the best possible feed adjustment and solvent extraction conditions and to avoid complicating waste treatment and disposal problems. Feed adjustment to maximum thorium concentration, preferably 1.5 M, and to minimum acidity, pre-
ferably slightly acid-deficient, is desirable to permit maximum processing rate and maximum decontamination from fission products. Co-extraction of the thorium and uranium with 30% TBP, using the Acid Thorex flowsheet, decontamination by scrubbing with nitric acid, and selective stripping of first thorium and then uranium, will provide good recovery, separation and decontamination in a single cycle of solvent extraction. With an acid-deficient feed the decontamination factors from rare earths and ruthenium will be about $10^4$ and $10^3$, respectively, for both uranium and thorium. If it is necessary to use an acid feed the decontamination factors will be somewhat lower though not by more than a factor of 10. In either case, this degree of decontamination is more than is required from the reactor physics point of view, so that any additional decontamination requirement must be justified by fuel refabrication and other handling requirements before it is put back into the reactor. No additional fission product decontamination of the thorium is justified since the gamma activity of the daughters of $^{228}$Th and $^{234}$Th will require either remote fuel refabrication anyway or else storage for a sufficiently long time that the activity from extractable fission products will reach direct handling levels before the $^{228}$Th does. Additional fission-product decontamination of the uranium may be justified if it is to be recycled with virgin or long-decayed thorium, and an optional second uranium cycle is indicated to take care of this case.

3.2.1. Extraction equipment capacity

In general, the maximum thorium processing rate through solvent extraction equipment of a given size will be less than the maximum uranium processing rate for a similar flowsheet. For example, the total volumetric capacity (combined aqueous and organic flow rates) of pulsed extraction-scrub columns is about 900 gal/h ft$^2$ for either the Purex or the Acid Thorex processes [61]; but since the maximum capacity of the solvent for thorium is only about half that for uranium, the effective capacity of the equipment is only about half as much for thorium as for uranium. A similar argument applies to the partitioning and strip columns. In a two- or three-column system designed for Purex the overall effect would be greater than a factor of two, since the capacity of a Purex plant for processing thorium is limited by the partitioning columns. In the Purex system the minor constituent, plutonium, is being selectively stripped from the major, uranium; whereas in the Thorex system, the major constituent, thorium, is being selectively stripped from the minor. For processing thorium fuels in an existing Purex plant, one might choose to use an Interim-23 first cycle, either discarding the thorium or recovering it from the waste in a second extraction, or one might choose to co-extract and co-strip the thorium in the first cycle and then separate them in an Interim-23 second cycle.

3.2.2. Waste volumes

For low burn-up fuels, the volume of concentrated high-level fission-product waste from either the Purex or the Acid Thorex flowsheets is about 50 gal/t of uranium or thorium processed. The waste concentrate is pri-
Reprocessing Methods and Costs

3.2.3. Close-coupled processing and fabrication

If economic evaluation of thorium fuels cycles indicates that immediate recycle of the thorium is competitive despite the cost penalties of remote fuel refabrication, further attention should be given to the possible economic advantages of low-decontamination-factor processes closely coupled to the fabrication operations. A single cycle of Acid Thorex followed immediately by Sol-Gel oxide preparation and vibratory compaction into fuel tubes is an example of a promising close-coupled process. Even the solvent extraction step might be further simplified since decontamination factors as low as ten might be acceptable in the overall fuel recycle scheme if this permitted sufficient cost savings to outweigh the disadvantages.

For recycle of uranium with virgin or long-decayed thorium, a study has been made of the Sol-Gel vibratory compaction route as regards the relationship between radiation dose to personnel, type of handling, amount of $^{232}\text{U}$ in the uranium and scale of the uranium-thorium production operation [62]. For fabricating fuels containing 3% $^{233}\text{UO}_2$ in Th$_2$O$_3$, if the $^{233}\text{U}$ contains no more than 20 ppm $^{232}\text{U}$ a production rate of 100 kg/d can be achieved in an unshielded facility without overexposure of personnel. For the same conditions except for 250 ppm $^{232}\text{U}$, shielding of 2 in. of lead would be required. For higher $^{232}\text{U}$ contents or larger production rates or for use of recycled thorium without $^{228,234}\text{Th}$ decay, remote fuel fabrication techniques will be required.

The direct maintenance of the equipment used in the Sol-Gel flowsheet is simplified by ease of decontamination. The Kilorod Facility [56], in which the fission-product contamination of the feed was negligible, was easily decontaminated with a vacuum cleaner to the degree that maintenance could be performed in "air suits" without excess worker exposure. The presence of fission products in the recycled uranium or thorium would complicate fabrication operations not only by increasing the shielding requirements but also by increasing the difficulty of equipment maintenance (and possibly even by affecting the Sol-Gel chemistry, though there is not enough data available at present to show what concentration of fission products can be tolerated). When the gel is fired to 1050°C, ruthenium, and perhaps other fission products also, would be volatilized and deposited in process equipment,
especially the furnace. Previous experience has shown that decontamination would be difficult in such a case, and remote maintenance may be required.

The economic dependence of the overall fuel cycle cost upon the degree of decontamination, the fabrication and maintenance techniques, the amount of $^{232}$U and $^{238,234}$Th in the fuel being processed, and the scale of the production operation is sufficiently complex that further studies, both experimental and theoretical, are needed to indicate the best long-term route and timing for thorium fuel cycle development to follow, either in general or for a particular reactor and fuel type. In the short term, the equipment available and the particular reactor and fuel type considered will have a marked effect on the optimization of the fuel cycle.

4. POWER REACTOR FUEL REPROCESSING COSTS

4.1. USAEC reference fuel-processing plant

From 1957 to 1963 the standard basis for evaluating the spent-fuel processing contribution to nuclear power costs was the calculated cost for processing the fuel in question in the "AEC Reference Fuel-Processing Plant"[63], a hypothetical plant capable of recovering purified uranium and plutonium from irradiated fuel at the rate of 1000 kg of uranium per day, up to 3% enrichment. At higher enrichments the capacity of this conceptual plant decreased, as a result of criticality considerations, to, for example, 930 kg/d at 4% enrichment, 537 kg/d at 10%, and 44 kg/d at 93%. The reference plant could process thorium fuels at the rate of 1000 kg/d if only the enriched uranium were to be recovered or 600 kg/d if thorium also had to be recovered, again subject to criticality limitations on the enriched-uranium processing rate. The USAEC announced in the Federal Register of 12 March 1957 that it would provide spent-fuel processing services at calculated charges based on the conceptual plant "on an interim basis... until the time when processing is available commercially". Initially, the standard USAEC daily charge was $15,300, both for the calculated number of processing days required for a batch of fuel and for the calculated number of "turn-around" days (the time required between processing batches for shut-down, clean-out, and start-up), but there was a provision for escalation that increased this figure to more than $17,000 in 1961 [64] and to an estimated $19,800 by 1965 [65]. The charges on a per-metric-ton basis were typically 25 to 100% or more higher than on the per-day basis, depending on batch size and daily processing rate.

4.2. NFS commercial processing plant

In 1963 the USAEC accepted an offer by Nuclear Fuel Services, Inc., to provide fuel processing services on a commercial basis, beginning in 1965 [65]. Nuclear Fuel Services (NFS) is now completing a plant with a nominal capacity of 1000 kg/d for uranium of up to 3% enrichment irradiated to burn-ups of up to 20,000 MWh/t and lower capacities for higher enrichments and burn-ups. The nominal capacity for thorium fuels is 500 kg/d for recovering only the enriched uranium. An extra charge will be assessed for the dis-
posal of the thorium-bearing waste. The initial base charge of $23,500 per "revenue day" (processing plus turn-around time) is subject to future escalation. Since the minimum turn-around time under the NFS formula is one third the processing time, the minimum per-metric-ton price for processing is $31,300. For 3% enriched fuel irradiated to a burn-up of 20,000 MWD/t at a thermal efficiency of 31%, this corresponds to a processing cost contribution to nuclear power of about 0.21 mill/kWh(e). Typical NFS processing costs for first-generation power reactor cores will be considerably higher than this, for example [65]:

<table>
<thead>
<tr>
<th>mill/kWh(e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Southern California Edison</td>
</tr>
<tr>
<td>Indian Point UO₂</td>
</tr>
<tr>
<td>Indian Point ThO₂</td>
</tr>
</tbody>
</table>

These higher charges are the result of lower burn-up, lower thermal efficiency, smaller processing batch size (which leads to a higher ratio of turn-around time to processing time) or higher enrichment than in the example calculation above. The large difference in the Indian Point UO₂ and ThO₂ figures is caused primarily by the 2-to-1 processing rate ratio between uranium and thorium, and secondarily by the extra charge for disposal of the high level waste containing the thorium. This extra charge results from larger volumes and the necessity of storing the wastes in the acid condition in relatively small stainless steel tanks, instead of neutralized storage in large mild-steel tanks.

4.2.1. Through-put rate

Since the NFS price schedule is based on daily charges, the amount of fuel that can be processed per day determines the unit cost of processing, which is normally reported as $/kg (of uranium or thorium) for fuel cycle cost purposes. The NFS nominal throughput rate is 1000 kg(U)/d up to 3% enrichment, and falls to 880 kg/d at 4%, 465 kg/d at 10%, and 40 kg/d at 93%. The nominal processing rate for thorium is 500 kg/d up to 8.5% highly enriched uranium content, and is inversely proportional to the uranium content above this level. At present these enrichment penalties are based on pre-irradiation enrichment. There is some possibility that these criticality penalties may be relaxed in the future, by use of post-irradiation enrichment and/or by use of nuclear poisons in the processing solutions and materials of construction. The NFS-AEC contract [65] has an "isotopic limits per processing lot" clause which can result in the processing rate being inversely proportional to burn-up above approximately 20,000 MWD/t. This limitation may be waived insofar as actual operating experience permits, and the actual limitation may be nearer 30,000 MWD/t [66]. The throughput rate penalties for enrichment, burn-up, thorium, etc., are calculated separately and only the most restrictive applied, rather than all of the penalties being applied consecutively. For example, there would be no burn-up penalty for thorium fuels up to 40,000 MWD/t (or possibly 60,000).
4.2.2. Fuel type

The "standard" fuel for NFS is UO$_2$ or ThO$_2$-UO$_2$ sheathed in stainless steel, zirconium, or zircaloy, in an assembly up to 16 ft long and up to 6 in. diam., weight up to one ton, with assembly casing and end fittings easily removable in the NFS mechanical cell, with individual fuel element diameters up to 0.75 in. and cladding thickness up to 0.05-in., and with metallic hardware inside the assembly up to 1/8 in. thickness. Other fuel types may suffer processing rate penalties imposed by the physical or chemical limitations of the NFS plant; for example, for U-Zr or U-Al alloy fuel clad in Zr or Al the processing rate is 400 kg/d gross weight of alloy plus cladding. An important consideration in advanced power reactor planning is that NFS is not now equipped to process certain fuel types, e.g. graphite- or carbon-type fuels such as HTGR. In connection with proposals to build an HTGR, there has been some consideration of the possibility of adding a special head-end facility to crush and burn the fuel to permit more-or-less standard aqueous processing. A substantial extra charge would have to be assessed to an HTGR fuel to pay for the extra capital and operating charges involved.

4.2.3. Turn-around time

The standard NFS turn-around time requirement is 8 d or one third of processing time, whichever is greater. To minimize turn-around charges relative to processing charges, the processing batch size should thus be equivalent to 24 or more processing days. For small fuel batches, requiring less than 8 processing days, the turn-around time can be reduced to equal to processing time (down to a minimum of 2 d turn-around) provided that these small batches can be combined with other similar small batches and also provided that processing can be delayed by NFS to permit convenient scheduling of combined small batches.

4.2.4. Waste disposal charges

For "standard" uranium fuels, as defined above, the $23,500/d base charge includes interim radioactive waste storage in mild-steel tanks by NFS and eventual perpetual maintenance by New York State. Fuel types which generate more high-level liquid processing waste than standard uranium fuels are subject to extra charges for interim and ultimate waste disposal. At present, thorium is not recovered and must be stored with its fission products in stainless steel tanks at an extra charge on the order of $9-16/kg. Alloy fuels such as U-Mo, U-Zr, and U-Al are also subject to substantial extra waste charges.

4.2.5. Escalation and other costs

The NFS price schedule is subject to escalation to cover increased labour pay rates and material prices. This is estimated to increase the base daily charge from $23,500 to $25,000 by mid-1970.
The processing charge does not include inventory or use charges on fuel before, during or after processing. For a typical standard fuel this might involve 120 d pre-shipping hold-up, 60 d shipping plus pre-processing hold-up at the plant, 30 d processing hold-up, plus another 30 d post-processing and shipping hold-up. Losses of nuclear material during processing, up to 1-1.5% at NFS, are not included in the processing charge. Shipping costs and costs of converting recovered material to forms other than concentrated nitrate solution are likewise not included.

4.3. Future processing costs

4.3.1. NFS

The base NFS price schedule discussed above is based on a nominal 300 revenue days per year for 15 yr, 1965-1980. The actual amount of power reactor fuel to be processed during the first years of this period will be less than the nominal NFS capacity but will exceed it in the early 1970's and grow rapidly thereafter, according to estimates of increase in nuclear power generation. NFS probably will be able to handle up to 350 revenue days per year with minor increased costs, and the NFS-USAEC contract provides for a corresponding reduction in the daily charge, up to 10% at a load of 350 or more revenue days per year. Thus the Oyster Creek [67] cost analysis assumes $21,150/d after 31 Dec. 1974. Assuming that burn-up and enrichment penalties can be relaxed to cover standard uranium fuel irradiated to 30,000 MWD/t at 31% thermal efficiency, the lower daily charge would correspond to only 0.13 mill/kWh(e). On the other hand, escalation may override the base price reduction and optimum burn-up for large PWR reactors may be only 20,000-25,000 MWD/t, so that 0.2 mill/kWh(e) may still be a more normal cost of processing. At the expense of modest additional capital investment and operating costs, NFS may be able to significantly increase their processing rate capability. This should permit a substantial reduction in unit processing costs, but when and whether this actually occurs probably depends on future competitive conditions (see below). Modifications to NFS to permit thorium recovery (in addition to uranium) would eliminate the extra thorium waste disposal charge, but may substitute an interim thorium storage charge for the 7-15 yr period required for $^{228}\text{Th}$ decay.

4.3.2. Other near-term commercial plants

For an industry predicted to grow as fast as is estimated for nuclear fuel processing after about 1973, it would be normal for other private companies to enter the field, in competition with NFS. The General Electric Co. has announced its interest in building a processing plant in the Western USA. Westinghouse has indicated its interest in offering a complete fuel cycle service, including processing. Other companies also have expressed an interest in processing; for example, a plant designed specifically for thorium fuels has been suggested. If this proliferation of processing plants occurs during the next decade, these new plants probably will be approxi-
mately the same size as NFS, and the economies of large-scale processing will be postponed.

4.3.3. Large processing plants

A design study [68] has shown that a 10 ton/d processing plant should cost less than twice as much to build and operate as a 1 ton/d plant, indicating a reduction in unit processing costs by a factor of approximately five. Depending on the burn-up and thermal efficiency, a 10 ton/d plant could service a very large nuclear power reactor economy, a size which may be many years away. On the other hand, a large nuclear desalination industry using natural uranium fuel might need such a large processing plant sooner than a power-only reactor industry [69].

4.3.4. Ultimate waste disposal

The NFS base daily charge includes approximately $700 for perpetual maintenance of the liquid-waste tank farm, plus a similar amount for interim (15-yr) waste disposal costs. For a typical power reactor this amounts to approximately 0.01-to-0.02 mill/kWh(e). ORNL studies of the waste problem [70, 71] indicate that this amount may be inadequate to cover perpetual tank storage costs, that perpetual tank storage of liquid wastes may not be adequately safe in any event, and that a safer ultimate disposal scheme (calcination to dryness and storage in a salt mine) may cost 0.02-0.03 mill/kWh(e) on a large scale. These studies did not include the cost of disposal of the cladding waste. For disposal of these as leached metallic solids the cost is relatively small, but recent ORNL studies [72] of chemical decladding waste solution disposal indicate that this can easily cost as much as the high-level waste, because of the large volumes and their chemical composition, i.e. another 0.02 mill/kWh(e). The large volume of Interim-23 waste, containing the thorium plus the aluminum nitrate salting agent, also would cost about 0.02 mill/kWh(e) more than the Acid Thorex type waste [72].

4.3.5. Advanced converter fuels

In support of the Advanced Converter Evaluation programme [73], fuel processing cost estimates were made for six types of advanced converter reactors: (1) uranium-fuelled, pressurized-water (PWR), assumed to include also boiling water; (2) thorium-fuelled, spectral-shift-controlled (SSCR); (3) uranium-fuelled, pressure-tube, heavy-water-cooled and moderated (HWR-U); (4) thorium-fuelled, heavy-water (HWR-Th); (5) thorium-fuelled, high-temperature, gas-cooled (HTGR), of the TARGET type; and (6) uranium-fuelled, sodium graphite (SGR).

The cost estimates, summarized in Table II, are based on 15% annual fixed charge rate (FCR) on total capital investment, which rate is approximately equivalent to that applicable to the first private commercial reactor fuel processing plant, Nuclear Fuel Services (NFS) [65]. At a 22% FCR, which would be more typical of most private chemical companies, the unit costs would be about 25% higher.
These cost estimates for the evaluation were made for hypothetical future nuclear power economies of 15 000 MW(e) of a given reactor type, with all of the fuel from that reactor type being processed in a single-purpose plant designed to exactly match its load. Thus, differences in annual throughput rate and nominal daily capacity in the various plants were caused by differences in burn-up, thermal efficiency and discharge batch size for the various reactor types. Table II shows the estimated processing contribution to nuclear power cost as a function of industry size, from 5000 to 20 000 MW(e). In this table the burn-up values are held constant at approximately the economic optimum indicated by consideration of overall fuel cycle costs at 15 000 MW(e), though the optimum burn-up would be higher for smaller industry size, and vice versa. All other things being equal, the power cost of processing decreases with increasing burn-up, thermal efficiency and batch size, is lower for uranium fuels than for thorium, and is lower for metal-clad oxides than for carbon-carbide-graphite or sodium-bonded fuels. The combined interplay of all these considerations in this study was to minimize differences in cost per kWh(e) though the costs in dollars per kilogram of fuel varied widely.

For all of the fuels except HTGR all of the uranium and plutonium, or thorium and uranium, in one processing batch (assumed to be the same as one reactor discharge batch) were dissolved together and then partitioned and decontaminated by solvent extraction. For HTGR three alternate schemes proposed under the TARGET [5] concept were considered: (1) mixed thorium-uranium fuel, as with the other reactors, (2) thorium and uranium in separate particles before irradiation, with the thorium-plus-bred-uranium particles processed separately from the high burn-up uranium particles; and (3) separate particles as in (2) but with the high burn-up uranium particles discarded directly to waste disposal instead of being processed, on the basis that their high $^{236}$U content makes recycle of this uranium to HTGR reactors undesirable from the overall economics and physics points of view. Scheme (2) costs more than scheme (3) and would have to be justified on the basis of a market value (for reactors other than HTGR) in excess of the additional cost of $1 to $2 per fissile gram recovered.

These cost estimates were based on modifications of previous estimates of processing plant costs, by duPont [68], and of ultimate waste disposal costs, by ORNL [70]. They are conservatively high in that they would predict higher costs than the actual NFS pricing formula for a plant of comparable fuel processing capability, and also in penalizing thorium fuels for their known processing disadvantages vis-a-vis uranium while not granting any cost credit for potential advantages such as the possibility that only one solvent extraction cycle will be sufficient since the thorium-uranium recycle scheme may require remote fabrication anyway. On the other hand, they may not be quite so conservative in assigning only a moderate head-end cost penalty to SGR and HTGR fuels on the assumption that present development programmes will be successful. These estimates for the Advanced Converter Evaluation agree with those made earlier for large desalination reactors [69], except that the earlier estimates made less allowance for turn-around time between batches and for ultimate waste disposal and used 7.7% fixed charge rate (for municipal or similar financing).
### TABLE II

**FUEL PROCESSING COSTS AS A FUNCTION OF NUMBER AND TYPE OF REACTORS**

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>PWR-U</th>
<th>HWR-U</th>
<th>SGR-U</th>
<th>SSCR-Th</th>
<th>HWR-Th</th>
<th>HTGR-Th</th>
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</thead>
<tbody>
<tr>
<td>Burn-up, MWd/t</td>
<td>21,000</td>
<td>11,100</td>
<td>22,000</td>
<td>28,800</td>
<td>27,800</td>
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<tr>
<td>Thermal efficiency, %</td>
<td>31.1</td>
<td>26.8</td>
<td>43.6</td>
<td>31.2</td>
<td>26.1</td>
<td>44.4</td>
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<tr>
<td>Batch size, t</td>
<td>33.9</td>
<td>19.5</td>
<td>12.2</td>
<td>66.3</td>
<td>13.5</td>
<td>7.89</td>
</tr>
<tr>
<td>Number of 1000 MW(e) Reactors</td>
<td>Processing Cost in mill/kWh(e), 15% F.C.R.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.332</td>
<td>0.396</td>
<td>0.321</td>
<td>0.357</td>
<td>0.381</td>
<td>0.306&lt;sup&gt;1&lt;/sup&gt;</td>
</tr>
<tr>
<td>10</td>
<td>0.201</td>
<td>0.237</td>
<td>0.195</td>
<td>0.212</td>
<td>0.232</td>
<td>0.184&lt;sup&gt;1&lt;/sup&gt;</td>
</tr>
<tr>
<td>15</td>
<td>0.154</td>
<td>0.177</td>
<td>0.144</td>
<td>0.159</td>
<td>0.173</td>
<td>0.149&lt;sup&gt;2&lt;/sup&gt;</td>
</tr>
<tr>
<td>20</td>
<td>0.125</td>
<td>0.136</td>
<td>0.104</td>
<td>0.116</td>
<td>0.125</td>
<td>0.099&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

Footnote: The superscripts 1, 2, 3 refer to three alternate HTGR processing schemes (see text).
4.3.6. Costs in an expanding economy

The cost studies described above assumed an "equilibrium" economy with the fuel from 5000 or 10,000 or 15,000 or 20,000 MW(e) of a given reactor type being processed in a plant designed to match its load and operating at constant full load. A dynamic economy starting small and growing large over a period of years cannot automatically expect to experience processing costs as low as those equivalent to a static economy of the same size at any given time. A processing plant must be built at a particular time and with a particular design capacity; it may be able to increase its actual capacity somewhat over a period of time as a result of technological improvements and inherent over-capacity in its basic design; but it cannot be built initially to match a small reactor economy and then expanded incrementally each year at marginal additional cost to keep up with the load as the reactor economy grows. Thus, in general a processing plant will be over-sized initially, enough so that it eventually can achieve unit costs low enough to give it an economic life long enough to permit it to recover its capital investment plus an acceptable rate of return on investment and still meet actual or potential competition from other plants. Since such a plant must start up on less than a full load, its average load over its life will be less than its equilibrium capacity and hence its average unit costs over its life will be higher than the calculated equilibrium costs. This "start-up penalty" can be appreciable for plants, such as spent fuel processing plants, which have a high ratio of capital cost to operating cost. In the case of the NFS plant, the USAEC is providing a "base load" during the first five years of operation, permitting a pricing policy based on a full load for an assumed 15-yr plant life. The USAEC has indicated a willingness to provide a base load also for a second private processing plant, but this type of support cannot be assumed for all future plants in a private competitive economy.

A study of optimum processing plant size, timing and location in a growth economy has been started. A computer code will be developed to calculate the minimum cost strategy as a function of input assumptions regarding growth curve, cost scaling factors, financing conditions, regulatory and competitive conditions, etc. As a first step in this study, an economic evaluation of HTGR head-end processing costs was made for the design and cost estimate presented in section 2.1.3.1. This head-end facility could handle the fuel from up to 10,000 MW(e) of HTGR reactors, but a reasonable estimate is that it might be 10 yr after the first commercial-size HTGR begins to discharge fuel before the HTGR industry reaches 10,000 MW(e). A present-worth economic analysis, similar to that of Vondy [73], indicated head-end capital and operating unit charges varying from less than $30/kg for a plant with a full load for 15 yr to more than $130/kg for the same same plant with a growing load for only seven years. It was indicated that the plant size was not optimum for the growth curve assumed, but the optimum size has not yet been calculated.

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Programmes for the development of thorium fuel cycles and reactors which use thorium are most often justified by the argument that better fuel utilization is obtained with thorium than with uranium. Numerous analyses of expanding nuclear power economies have shown that less uranium ore must be mined using thorium converter reactors which are started-up with $^{235}\text{U}$ and the bred uranium recycled than using uranium converters with recycle of plutonium. The argument for the use of thorium is extended further by the contention that thorium converters can develop into "break-even" breeders, or even breeders that produce enough excess fuel to inventory other reactors.

There is no doubt as to the importance of conserving fuel resources, but other factors must also be considered to determine whether thorium systems should be developed and which thorium reactor types are most likely to be built. These relate to the economics of thorium systems and to the technical difficulties that lie in the way of their development.

Two points should be made about economics. First, unless restraints are applied, power system operators are going to select the reactor type that they believe will yield the lowest power cost. In addition, they are going to operate at conditions producing the lowest cost, although the fuel utilization of a reactor can always be improved at the expense of higher cost. Conservation of fuel resources may be an important national or world objective, but it will be decisive only if it affects the economics of a reactor or if it is imposed by governmental constraint. Of course, in evaluating economics, future conditions must be properly weighted and attitudes of governmental agencies, as reflected in the regulation of financing and rates, or in tax provisions, taken into account.

The second point is the converse of the first. Some thorium reactors may have lower power costs than competing uranium reactors, and those that do may be developed and built even if their fuel utilization is not particularly favourable.

Technical factors which determine whether a reactor system can actually perform as assumed in the analysis of fuel utilization and economics are of obvious importance. But the question is not just whether a system can be developed to perform as assumed, but also when the postulated performance can be achieved in full-sized plants. In addition, it must be recognized that before the calculated economics can be achieved, the fuel processing and

* Research sponsored by the United States Atomic Energy Commission under contract with the Union Carbide Corporation.
fabrication industry postulated in the economics analysis must exist. One who is considering developing a new system that looks attractive, of course, must also consider the cost of development.

The questions regarding development are particularly applicable to thorium systems. Most of the reactors which appear attractive with thorium are less well developed than the competing uranium systems. In addition, the presence of $^{232}$U in recycle fuel requires that special fuel fabrication facilities be built; and in some cases, new head-end processing methods must be developed and the processing facilities built. Thus, in the United States at least, the thorium converters appear to be caught in a squeeze. On the one hand, the status of development of the thorium reactors is such that they will not be brought into operation in any quantity before the early 1970's, and by then the pressurized and boiling light-water reactors will have a considerable head start. On the other hand, there are large programmes directed towards the development of fast plutonium-uranium breeders, and the postulate of the United States programme is that these reactors will reach the point of economic attractiveness before 1990. It would appear, therefore, if thorium converters are to play an important role, they must become competitive with other converters and be built in quantity in the 1970's. Thermal breeders with low fissile inventories, however, may be able to enter the competition later. If their costs are favourable relative to fast reactors, the period in which they are attractive could extend far into the future.

In view of the foregoing, a study was undertaken at ORNL to estimate the performance and power costs that will be achieved with various proposed thorium reactors if they are built in the 1970's. The design information was taken from conceptual designs of 1000 MW(e) reactors provided by developers of the concepts. A set of economic factors that represent the average conditions over the life of reactors built during that period was defined and physics performance, capital, operating, and fuel cycle costs for all of the reactors were estimated on a consistent basis. For comparison with the thorium reactors, similar estimates were made for a 1000 MW(e) light-water-moderated, pressurized-water reactor. Although we attempted to make our cost estimates correct on an absolute basis, the emphasis in all of the estimates was on accuracy of the costs of the various systems relative to each other.

To determine that the reactors could attain the performance postulated in the design and to identify any development that is required, the design and predicted engineering performance of the reactor plants were reviewed and evaluated. Taking into consideration differences in design requirements revealed by this review, normalized estimates were made of the capital costs for all the plants.

In the sections which follow, the reactors considered are described briefly and their engineering performance requirements discussed, the economic context for the evaluation is outlined, some general comments are made on the physics of thorium reactors, the fuel utilization of specific reactors is presented, and the fuel cycle, capital and operating costs of the reactor are reported.
DESCRIPTION OF REACTORS

Six thorium-fuelled reactors have been evaluated, along with a large pressurized-water reactor which serves as a basis for comparison. We have not attempted to design reactors ourselves but have taken designs proposed by groups who are the developers or proponents of the systems. The technical characteristics of the reactors are summarized in Table I.

The Pressurized-Water Reactor (PWR) design is based on a Westinghouse study [1, 2] of a 1000-MW(e) reactor and differs from the Westinghouse concept mainly in the use of Zircaloy fuel cladding rather than collapsed stainless steel. A 15.8 ft inside diameter by 11.2-in. thick reactor vessel contains the core, and the peak-to-mean power density is 3.3.

The Spectral Shift Control Reactor (SSCR), which utilizes a mixture of light and heavy water as the moderator and coolant, is based on a design [1] by Babcock and Wilcox. In this concept, the entire core is refuelled at one time; fuel depletion and fission product build-up are compensated by increasing the H2O/D2O ratio to reduce the probability of neutron capture in thorium resonances. Twelve zones having different uranium-to-thorium ratios are used to flatten the power distribution, thus reducing the peak-to-average to 1.9 from the higher value of the PWR. This permits a high power density, and the SSCR reactor vessel size is reduced from that for the PWR.

The Heavy-Water Reactor (HWR) is a pressure-tube type based on a design study [1] by E.I. du Pont de Nemours and Company (Savannah River Laboratory). Pressurized heavy-water coolant passes through 688 Zr-2.5% Nb pressure tubes which have an inside diameter of 4.43 in. and are 0.16 in. thick. Concentric tubular fuel elements are used, and the reactor is refuelled while at power.

The Seed-Blanket Reactor (SBR) is based on the movable-fuel concept proposed by Bettis Atomic Power Laboratory [3, 4]. In this concept, fully-enriched uranium is located in annular seed regions distributed throughout a thorium blanket. The SBR is controlled by axial movement of portions of the seed, which changes the effective thickness of the seed annulus. This results in a change in the leakage of neutrons from the highly reactive seed regions into the sub-critical blanket regions, thereby providing criticality control. Control poisons are thus eliminated, and the neutron economy improved.

In the converter recycle concept considered here, the seeds are initially fuelled with 235U, and 233U is recovered from the blanket. After about three converter cycles using 235U seeds, the accumulated 233U is used to fuel the core. According to Bettis, self-sustaining recycle is achieved afterwards with no additional fuel make-up required.

The design evaluated here has 61 seed modules, and the reactor vessel is a 28-ft inside diameter by 9-in. thick sphere.

The High-Temperature Gas-Cooled Reactor (HTGR) is a helium-cooled, graphite and BeO-moderated reactor based on the TARGET concept [1, 5] of General Atomics. Graphite fuel elements contain loose pyrolytic-carbon-coated fuel particles in holes located in a "phone-dial" arrangement. Two sizes of particles are used, one containing only uranium and the other only
### Table I
**Reactor Design Characteristics**

<table>
<thead>
<tr>
<th>Item</th>
<th>Pressurized-water</th>
<th>Spectral shift controlled</th>
<th>Heavy-water</th>
<th>High temperature gas-cooled</th>
<th>Seed blanket</th>
<th>Molten-salt converter</th>
<th>Molten-salt breeder</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net electrical capacity (MW(e))</td>
<td>1,002</td>
<td>1,000</td>
<td>1,010</td>
<td>1,008</td>
<td>1,000</td>
<td>1,060</td>
<td>1,000</td>
</tr>
<tr>
<td>Reactor power (MW(th))</td>
<td>3,220</td>
<td>3,270</td>
<td>3,870</td>
<td>2,270</td>
<td>3,200</td>
<td>2,250</td>
<td>2,250</td>
</tr>
<tr>
<td>Steam temperature/pressure (°C/atm)</td>
<td>257/43</td>
<td>254/40</td>
<td>241/33</td>
<td>565/238</td>
<td>247/37</td>
<td>538/238</td>
<td>538/238</td>
</tr>
<tr>
<td>Net station efficiency (%)</td>
<td>31</td>
<td>31</td>
<td>26</td>
<td>44</td>
<td>31</td>
<td>44</td>
<td>44</td>
</tr>
<tr>
<td>Moderator</td>
<td>H₂O</td>
<td>H₂O</td>
<td>H₂O</td>
<td>Graphite</td>
<td>H₂O</td>
<td>Graphite</td>
<td>Felt salt</td>
</tr>
<tr>
<td>Coolant</td>
<td>H₂O</td>
<td>D₂O H₂O</td>
<td>D₂O</td>
<td>Helium</td>
<td>H₂O</td>
<td>Felt salt</td>
<td>Felt salt</td>
</tr>
<tr>
<td>Coolant temperature, inlet-outlet (°C)</td>
<td>285-314</td>
<td>279-312</td>
<td>264-304</td>
<td>382-800</td>
<td>271-306</td>
<td>566-732</td>
<td>566-732</td>
</tr>
<tr>
<td>Coolant pressure (atm)</td>
<td>140</td>
<td>130</td>
<td>130</td>
<td>140</td>
<td>140</td>
<td>140</td>
<td>7</td>
</tr>
<tr>
<td>Control method</td>
<td>Soluble poison</td>
<td>Moderator mix</td>
<td>Continuous fuelling</td>
<td>Poison rod</td>
<td>Seed position</td>
<td>Continuous fueling</td>
<td>Continuous fueling</td>
</tr>
<tr>
<td>Core diameter x height (m)</td>
<td>3.9 x 3.4</td>
<td>3.1 x 3.3</td>
<td>1.9 x 4.6</td>
<td>9.5 x 4.8</td>
<td>5.3 x 2.2</td>
<td>5.1 x 6.2</td>
<td>2.6 x 2.6</td>
</tr>
<tr>
<td>Specific power (kW/kg fertile)</td>
<td>32</td>
<td>48</td>
<td>48</td>
<td>27</td>
<td>27</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>Core power density (kW/t)</td>
<td>81</td>
<td>150</td>
<td>130</td>
<td>16</td>
<td>7</td>
<td>17</td>
<td>160</td>
</tr>
<tr>
<td>Fuel elements</td>
<td>Rod bundle</td>
<td>Rod bundle</td>
<td>Concentric tubes</td>
<td>Graphite cyl</td>
<td>Rod bundle</td>
<td>Molten salt</td>
<td>Molten salt</td>
</tr>
<tr>
<td>Fuel rod or tube diam (in.)</td>
<td>0.25-0.38</td>
<td>0.42</td>
<td>1.342 0.88, 0.83</td>
<td>4.5</td>
<td>0.25 0.33, 0.74</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel</td>
<td>UO₂</td>
<td>UO₂-ThO₂</td>
<td>UO₂-ThO₂</td>
<td>UC₂-ThO₂</td>
<td>UO₂-ThO₂</td>
<td>UC₂-ThO₂</td>
<td>UF₄-ThF₄-LiF-BeF₃</td>
</tr>
<tr>
<td>Fuel management</td>
<td>1/3 scatter</td>
<td>Batch</td>
<td>On line refuelling</td>
<td>1/12 scatter refuelling</td>
<td>Batch</td>
<td>Continuous</td>
<td>Continuous</td>
</tr>
</tbody>
</table>

*Calculated through external heat exchangers*
Thorium. The two types of particles are separated at the end of the cycle and processed separately. Material from the uranium particle is sold or discarded to reduce the build-up of $^{236}\text{U}$ in the system. Bred uranium recovered from the thorium particle is combined with make-up $^{235}\text{U}$ and refabricated into the uranium-only particles for a subsequent fuel cycle. Thus, the make-up $^{235}\text{U}$ is never recycled and bred uranium is recycled for only one pass. A BeO spine in the centre of the graphite body has a volume that gives a carbon-to-beryllium atomic ratio of 2.4.

Use of a 56-ft inside diameter pre-stressed concrete reactor vessel that contains the core, the steam generators, and the blowers is a major design feature of the plant.

The Molten-Salt Converter Reactor (MSCR) is essentially a scaled-up design [6] of the MSRE (Molten-Salt Reactor Experiment) now in operation at Oak Ridge. It consists of a Hastelloy-N vessel filled with graphite moderator. A fuel salt, consisting of fluorides of uranium, thorium, lithium and beryllium, and melting at about 475°C, is circulated through passages in the core graphite and then through an external heat exchanger. A side stream of the fuel salt is processed continuously for fission product removal in a fluoride volatility and vacuum distillation processing plant integrated with the reactor.

The Molten-Salt Breeder Reactor (MSBR) is similar to the MSCR, except that the fissile and fertile materials are maintained in separate streams. The fertile stream not only circulates through the core graphite, but also surrounds the core forming a large blanket. The fuel stream circulates through the core within graphite tubes. The two-stream design makes possible a smaller core, lower neutron leakage, and decreased neutron losses to protactinium.

ENGINEERING PERFORMANCE AND STATUS OF DEVELOPMENT

Evaluation of the thermal designs of the solid-fuel reactors indicated that all appear to be adequate, with the reservation that the power peaking factors were not checked by physics calculations and the enthalpy rise factor of 1.8 claimed for the pressurized-water reactor has not yet been demonstrated. With regard to fuel element requirements, the PWR and SSCR appear to be designed on the basis of current technology. The clad on the blanket rods of the SBR appears not to be "free-standing", but by proper selection of dimensions it can very likely be made to operate satisfactorily. 1

The remaining reactors are designed on the basis of extrapolations of fuel element technology well beyond existing data. Tests of the nested fuel elements for heavy-water reactors have not progressed to the burn-ups required at the specified heat ratings, and neither the largest size tubes nor assemblies of tubes have yet been tested. The HTGR concept requires further testing of coated particles, of large sizes and to high burn-up, and demonstration of the operation of graphite fuel elements for a 6-yr life. 1

1 It was announced recently that Zircaloy-clad ZrO$_2$-diluted UO$_2$ rods typical of seed elements for long-life SBR cores have failed in irradiation experiments, but we do not yet know the details of the failures and their applicability to the shorter-life breeder cores.
The PWR, SSCR and SBR all require large heavy-walled pressure vessels that are beyond present experience, but the problems associated with their construction appear to be solvable. The HWR uses Zr-2.5% Nb alloy pressure tubes on which corrosion and creep information are favourable but which require excessive extrapolation to predict a 30-yr behaviour. An on-line refuelling machine specified for the heavy-water reactor will require high reliability under severe conditions. Adequately low losses of heavy water from the SSCR and HWR can probably be achieved, but care with closures, seals, and leakage recovery systems will impose some burdens during operation and maintenance.

Feasibility of the particular HTGR design proposed depends largely on successful demonstration of the prestressed-concrete reactor vessel that houses the entire primary system, including the core, helium-circulating blowers, and steam generators. While French and British experience with concrete reactor vessels is applicable, and both the French and British have "all-inside" plants under construction, successful performance of vessels of the high-temperature, gas-cooled reactor type remains to be demonstrated.

A preliminary analysis of plant availability indicates that a high degree of reliability will be required of all reactor equipment. Because of the scheduled down-time required for the reactors which are shut down for re-fuelling, only a few per cent of the operating time will be available for unscheduled equipment outages if those plants are to have an availability of 90%.

The state of development of the systems and components required for the operation of the reactors is far from equal. The light-water-cooled reactors have the most extensive operating experience to call on. Heavy-water reactors have the advantage of NPD, CVTR, PRTR, and HWCTR experience, with the operation of the 200-MW(e) CANDU reactor anticipated soon. Until operation of the Peach Bottom, Dragon, EGCR, and Oldbury stations is under way, the technology of the HTGR must be considered the least firmly established among the solid-fuelled reactors.

The molten-salt reactors are in a separate line of development, represented at present by two small reactor experiments, but no full-scale plants. The feasibility of the molten-salt concept was demonstrated in 1954 with the Aircraft Reactor Experiment, a 2.5 MW(th) reactor with a 900°C maximum temperature. The Molten-Salt Reactor Experiment, which went critical on 1 June 1965, is a 10-MW(th) graphite-moderated reactor operating at 680°C, and is designed to demonstrate the compatibility of the construction materials, Hastelloy-N and graphite, with the molten fluoride fuel, as well as the operation and maintenance of the pumps and other components.

The MSCR is essentially a scale-up of the Molten-Salt Reactor Experiment but is designed for a somewhat higher temperature, 730°C, and for a higher power density. The assumed on-line processing has not been demonstrated, but the extension of the well-developed fluoride volatility process [6] should be straightforward. A process for removing rare-earth fission products from the fuel by vacuum distillation has been tested on a laboratory scale. Rapid removal of xenon will be tested in the Molten-Salt Reactor Experiment and uncertainties regarding the behaviour of some other fission products clarified. The plant design is based on use of an intermediate coolant between the fuel salt and the steam system, but all of the coolants considered present some design or development problems.
The MSBR involves several features that are still under development. The use of graphite tubes to separate the fuel stream from the fertile stream has not been demonstrated, although a method has been developed for making brazed graphite-to-metal joints, remotely if necessary. Replacement of the tubes will be possible, but a design which allows adequate tube life to be obtained in spite of graphite shrinkage will be required. Another engineering problem is the close-coupling of the primary heat exchanger to the reactor that is required to minimize the fuel inventory.

ECONOMIC CONTEXT AND COST FACTORS FOR THE COMPARISON OF REACTORS

As noted earlier, the period of interest for the introduction of thorium reactors appears to be the 1970's. We have, therefore, defined a set of economic assumptions that would represent the average conditions over the 30-yr life of a reactor concept if a number of reactors of that design were built in the United States between 1970 and 1980. These are as follows:

1. The technology used in the reactor and power plant designs was, in general, restricted to that which would be available for smaller, prototype reactors to be built for start-up in 1970.

2. Private ownership of the reactors, the fuel, and of the fabrication and processing plants was assumed as the reference case, and annual charges were taken as 10% on fuel, heavy water, and \(^7\)Li, 12% on the reactor plant, and 22% on the fuel fabrication and processing plants. Alternate plant fixed charges of 7% and inventory costs of 5% were used to represent public ownership.

3. Fuel fabrication and processing costs are based on single-purpose plants, each capable of serving 15 000 MW(e) capacity of the concept under consideration. Because the plant size is specified in terms of electrical capability, the capacity in tonnes per day is different for each fuel burn-up.

4. Levelized fuel cycle costs were computed by determining the "present worth" of all expenditures and receipts during a reactor life-time of 30 yr using a discount factor of 6%. The reactor plant availability was required to be 0.9 in to achieve an average plant factor of 0.8 in over the plant life. The unit costs of fuel fabrication and processing were held constant throughout the reactor life.

5. Assumed costs of nuclear materials are $17.60/kg \(\text{U}_3\text{O}_8\) and $30/kg separative work ($12/g fully enriched \(^{235}\text{U}\), $11/kg \(\text{ThO}_2\), $14/g \(^{233}\text{U}\), $10/g fissile Pu, $37.40/kg \(\text{D}_2\text{O}\), and $0.12/g \(\text{Li}_2\text{O}\).

6. The thorium reactors are fuelled initially with fully enriched \(^{235}\text{U}\) and the bred uranium subsequently recycled with fully enriched \(^{235}\text{U}\) make-up as required. Plutonium from the PWR was assumed to be sold at $10/g, which studies have indicated gives about the same fuel cycle cost as if the plutonium were recycled.

7. The times for fuel fabrication, shipping, post-irradiation cooling, etc. were estimated for each case. Ownership of fuel during fabrication and processing was assigned to the reactor plant, and the inventory charge includes the full out-of-pile inventory. Shipping costs were estimated for each
fuel element design and irradiation condition assuming that the fabrication and processing plants are located at a site 1000 miles from the reactor.

The ground rules and procedure followed are similar to those stated in more detail in [1], with, however, some changes in the values of materials.

PHYSICS OF THORIUM CYCLE REACTORS

The outstanding characteristic of thorium cycle thermal reactors is the favourable neutron economy which is obtained using $^{233}\text{U}$ as the fissionable material. This results largely from the value of $\eta$, the number of neutrons produced per neutron absorbed, which at 2200 m/s is 2.29 for $^{233}\text{U}$ but only 2.07 and 2.10 for $^{235}\text{U}$ and $^{239}\text{Pu}$, respectively. In the thermal spectrum of an actual reactor the $\eta$'s of all three nuclides will be less than the 2200 m/s values because of the necessity of elevated moderator temperatures and because of hardening of the neutron energy spectrum. A significant fraction of neutron absorptions occur at epithermal energies where the values of $\eta$ for all of the nuclides decrease. The effect of epithermal absorptions is generally least harmful with $^{233}\text{U}$, but there has been a troublesome uncertainty in the $^{233}\text{U}$ epithermal cross-sections. A cross-section set derived from various differential cross-section measurements, mainly fast chopper measurements, yields an average capture-to-fission ratio (alpha) of about 0.23. Integral epithermal measurements by Halperin [7] at ORNL and by Feiner at KAPL [8] have indicated, however, that the average value of alpha in a $1/E$ spectrum above 0.5 eV is about 0.17. The weight of evidence appears to favour the lower value of alpha, and it has been used in this study.

Several other differences, less important than the value of $\eta$, occur in the comparison of the thorium cycle with the uranium cycle. One of these is that the fission product yield curve is shifted slightly toward lower mass numbers in the case of $^{233}\text{U}$ fissions. This appears to give a favourable effect on the average absorption cross-section, particularly in the vicinity of mass 149 where there are a number of high cross-section nuclides, and the yield from $^{233}\text{U}$ is only 0.0076 compared to the $^{235}\text{U}$ yield of 0.0113.

An unfavourable and well-known aspect of the thorium cycle is the relatively long half-life (27 d) and high cross-section (43 b thermal and 925 b resonance integral) of the $^{233}\text{Pa}$ which is intermediate in the conversion of thorium to $^{233}\text{U}$. When a neutron is absorbed in $^{233}\text{Pa}$ the loss in conversion ratio is equivalent to two neutrons. In order that there be a favourable result in the competition between decay of $^{233}\text{Pa}$ into $^{233}\text{U}$ and absorptions in $^{233}\text{Pa}$, the flux in the regions containing thorium should not be too high. In most designs the specific power is kept lower than it would otherwise be for this reason. Small cores with breeding in external blankets or continuous movement of the thorium in and out of the high flux regions could also increase the fraction of $^{233}\text{Pa}$ which decays into $^{233}\text{U}$.

The initial buildup of $^{233}\text{Pa}$ and consequent delay in generation of $^{233}\text{U}$ in a fuel element causes a rapid decrease in reactivity of thorium-cycle reactors during the first several half-lives of the $^{233}\text{Pa}$. This reactivity is recovered when the $^{233}\text{Pa}$ decays during a shut-down, and losses to the control poisons that are required to offset these "proactinium transients" can
be important in a reactor which is shut down frequently. A typical plot of reactivity versus burn-up in Fig. 1 shows the contrast of a $^{233}$U-thorium reactor with a partially enriched uranium reactor, which has an initial reactivity increase as the $^{239}$Pu, with its high fission cross-section, builds up.

Although the ultimate goals of thorium reactor designs may include breeding, it is difficult to reconcile the other, often conflicting, demands of low fuel-cycle cost and low capital cost with the requirement of high enough neutron economy for breeding. Neutron leakage and parasitic captures in moderator, coolant, and structure can take an appreciable fraction of the available neutrons. The losses to fission products become important if the fuel fabrication and reprocessing costs require high fuel burn-up. A further reduction in neutron economy comes from the fact that non-breeding systems must use $^{235}$U (or possibly plutonium) as make-up to supplement the bred $^{233}$U. The lower $\eta$ of these isotopes plus the increased build-up of $^{236}$U and $^{237}$Np further reduce the breeding ratio. A rough rule-of-thumb is that the decrease in breeding ratio from any increased parasitic capture, such as in fission products, will be doubled when one takes into account the greater proportion of $^{235}$U in the recycled fuel. The use of thorium in non-breeding reactors serves principally to extend burn-up and increase the utilization of fissile uranium.

We have calculated the performance of the PWR, SSCR, HWR, and HTGR described earlier as to breeding ratios, burn-ups, and mass balances over the thirty-year history. The isotopic depletion calculations were typically done with 15 energy groups using cross-sections that were condensed from a 98 group set. The fuel management scheme recommended by the sponsor—
was used in the calculations with, in one case, as little as 1/12 of the core refuelled at a time. An appropriate delay time was considered between removal of spent fuel from the core and recycle of the reprocessed fuel. Several cases were calculated for each reactor type with variation of fuel enrichment to determine the optimum burn-up.

In the case of the seed blanket reactor, we performed no physics calculation ourselves for the specific design considered but used values provided by Bettis. However, we previously analysed an SBR core which was designed to have a breeding ratio greater than 1.0 and found that starting with pure $^{233}$U, it was capable of operating for over two years with no net consumption of fuel. Our physics calculations for the MSR's were made using an automated computer programme that determined the economic optimum design and computed the physics performance and fuel cycle cost at equilibrium conditions. Thirty-year-average values were obtained by correcting the equilibrium results for the time variation in nuclide concentration assuming that the reactors are started with $^{235}$U. Results are given in Table II for near-optimum cases of each reactor type.

**TABLE II**

<table>
<thead>
<tr>
<th>Concept</th>
<th>Feed enrichment $^a$</th>
<th>Exposure (MWD/t of U + Th)</th>
<th>Breeding ratio $^b$</th>
<th>Index of fuel consumption $^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR</td>
<td>2.2</td>
<td>21 000</td>
<td>0.60</td>
<td>1.20</td>
</tr>
<tr>
<td>SSCR</td>
<td>4.0</td>
<td>25 400</td>
<td>0.75</td>
<td>0.80</td>
</tr>
<tr>
<td>HWR</td>
<td>2.1</td>
<td>28 800</td>
<td>0.84</td>
<td>0.60</td>
</tr>
<tr>
<td>HTGR</td>
<td>2.1</td>
<td>52 300</td>
<td>0.90</td>
<td>0.22</td>
</tr>
<tr>
<td>SBR</td>
<td>1.4</td>
<td>12 000</td>
<td>1.00</td>
<td>0.00</td>
</tr>
<tr>
<td>MSCR</td>
<td>2.6</td>
<td>2 300</td>
<td>0.96</td>
<td>0.09</td>
</tr>
<tr>
<td>MSBR</td>
<td>(d)</td>
<td>(d)</td>
<td>1.06</td>
<td>-0.13</td>
</tr>
</tbody>
</table>

$^a$ Per cent by weight, based on total heavy metal.
$^b$ Gross estimated for last cycle of 30-yr reactor life.
$^c$ ($1.0$-breeding ratio)/thermal efficiency, a measure of relative fuel consumption.
$^d$ Not applicable because fertile and fissile streams have different cycle times.

The SSCR has the lowest breeding ratio of the thorium reactors. This is attributable to the low $\eta$ in the under-moderated condition of the first part of the cycle and the parasitic captures in hydrogen in the last part of the cycle. The HWR gives the next higher breeding ratio in this comparison. The use of heavy water for both coolant and moderator can result in very good neutron economy since there are virtually no absorptions in the heavy water. The fact that the breeding ratio for the HWR was only 0.84 is caused by the parasitic absorptions in the pressure tube and by the economic necessity of reducing heavy water inventories. HWR's could give breeding
ratios above unity if the economics favoured such a design. The neutron economy of the HTGR is hurt slightly by the absorptions in the graphite and BeO moderator, but it is helped by the \((n, 2n)\) reaction in the beryllium and by a device of keeping the bred fuel separate from the make-up \(^{235}\text{U}\) and recycling only the bred fuel. After being fuelled with \(^{233}\text{U}\), the SBR has the highest breeding ratio of the solid fuel element reactors. It achieves a breeding ratio of 1.0 in light water by having a high moderator content in the seed for thermalization where most fissions occur and having a low moderator content in the blanket to reduce parasitic losses there.

The MSCR has good neutron economy by virtue of using graphite moderator and continuously stripping out the volatile fission products so that there is little poisoning from xenon. The MSBR also used graphite moderator and continuous stripping of volatile fission products. In addition it has separate fertile and fissile fuel streams and is able to minimize neutron losses to \(^{233}\text{Pa}\) by allowing most of the Pa to decay in low flux regions.

The cumulative ore requirements for inventory and make-up of the reactors have been computed and are shown in Fig. 2. In each case the nuclear power industry of the USA was assumed to consist entirely of the reactor concept considered, and the power growth rate was taken from the estimate of the November 1962 "AEC Report to the President". The ore commitment at a specified date was taken to include the inventory in the operating reactors and associated processing plants as well as the make-up which would be required for existing reactors over the remainder of a 30-yr life. In the case of the MSBR, however, the ore requirement at a particular time was not reduced by bred fuel to be produced in the future.

By the measure used in Fig. 2, the ore requirement for the SBR over the next 50 yr could be as high as that of a converter which has a lower conversion ratio but also a smaller fuel inventory. Hence comparison on this basis is somewhat unfair to the SBR, since at the same ore commitment it would actually consume less fuel than a converter. On the other hand, the
converters were not optimized for high fuel utilization, and higher conversion ratios could be achieved in them at the expense of higher power cost.

An example of the breeding ratios that can be obtained when a thorium cycle reactor is designed for maximum breeding is shown in Fig. 3. The calculations are for the HTGR described earlier, operating at an average core power density of 7 W/cm$^3$. If one assumes that all fission products are to be retained in the fuel until it is reprocessed (the current design aim) the maximum breeding ratio is 1.07 after deducting processing losses. If, instead, a fuel were designed to release instantaneously all of the volatile fission products (the noble gases and halogens) the maximum breeding ratio would be 1.11. The release of volatile fission products plus the lithium formed in the (n, a) reaction in the beryllium would give a maximum breeding ratio of 1.12. The corresponding doubling times, assuming an out-of-reactor hold-up time for reprocessing and refabrication of 60 d, are 55 yr, 30 yr, and 24 yr. Since complete instantaneous fission product release from solid fuel elements is probably not achievable, the maximum attainable breeding ratio for this concept is probably somewhere between 1.07 and 1.12.

It should be kept in mind that the low fuel burn-ups for the cases shown in Fig. 3 would give excessive fuel-cycle costs unless extremely cheap processing and fuel fabrication procedures which have low fuel hold-up were developed. For this reason, fluid fuel reactors appear to be particularly apt candidates for thermal breeders. The MSBR referred to in Table II has, for example, a breeding ratio of 1.06 with a low fissile inventory (0.7 kg fissile U/MW(e)) and is thereby able to achieve a system doubling time of 10 yr.

FUEL CYCLE COSTS

Fuel fabrication, processing (including ultimate waste disposal), and shipping costs were estimated for each reactor as functions of plant throughput using the methods described in [1]. For the PWR, SSCR, HWR, and
TECHNICAL AND ECONOMIC CHARACTERISTICS

HTGR, these unit costs were combined with the thirty-year mass balances and, using present worth accounting, levelized fuel cycle cost was obtained as a function of fuel exposure. This procedure yielded the minimum costs for the core designs considered, but it should be noted that dimensions, fuel-to-moderator ratio, plant design conditions, etc., were those specified by the reactor designers, and such factors were not varied.

In the case of the seed blanket reactor, we used only the single set of mass balances provided by Bettis and no optimization was done. As mentioned earlier, calculations for the MSR's were made using a computer programme that selected the optimum design and computed the equilibrium fuel cycle cost. Thirty-year average costs were obtained by correcting the equilibrium results for the time variation in nuclide concentration assuming that the reactors are started with $^{235}$U.

A near-optimum example of the operating conditions and fuel cycle cost breakdowns for each concept is presented in Table III. The inventory costs reported in the table include the fuel held up during processing and fabrication.

The costs for the solid-fuel reactors in Table III reflect the large capacity fabrication and processing plants assumed for this study. Costs associated with fabrication and processing range only from 26% of the total fuel cycle cost for the SSCR to 45% for the SBR. An effect of lowering the importance of the costs for processing and fabrication is to increase the importance of the costs associated with owning and consuming uranium, particularly for a 10% inventory charge. Consequently, the fuel cycles generally tend to optimize at lower burn-ups than those cited by the concept sponsors.

The HTGR achieves the lowest fuel cycle costs of the solid-fuel reactors by virtue of its good neutron economy and long fuel life, and its high thermal efficiency gives it a special advantage over the water-cooled reactors with which it is compared. Still lower costs, down to perhaps 1.2 mill/kWh, could be achieved in the HTGR if the BeO were eliminated, but the breeding ratio would be lower and the fuel utilization poorer.

Good neutron economy and low fuel fabrication costs assist the HWR in maintaining low fuel cycle cost (if heavy-water cost is not included). While the PWR has somewhat higher burn-up costs than the SSCR, the difference is more than offset by the lower inventory cost of the PWR that results from use of partially rather than fully enriched uranium.

The high fuel cycle cost of the SBR results from a combination of large fissile inventory and short core life. The SBR was not optimized for minimum power cost as were the other reactors in the table but was designed specifically to obtain a breeding ratio above 1.0 after accumulation of a core loading of $^{233}$U. Probably lower fuel cycle costs could be obtained if it were optimized for the ground rules used.

The MSCR achieves low fuel cycle costs through the good neutron economy it gets from xenon removal and continuous fuel processing, from essentially eliminating fuel fabrication cost, and from high thermal efficiency. Although the fuel processing plant for the molten-salt reactors is at the reactor site and integrated with the reactor, the capital charges (at 12% per year) and operating costs for the processing plant have been isolated and reported in the fuel cycle breakdown as processing costs. By use of separate fissile and fertile streams, with a thorium blanket surrounding the core, the MSBR achieves improved neutron economy over the MSCR and a lower
<table>
<thead>
<tr>
<th>Reactor</th>
<th>PWR&lt;sup&gt;a&lt;/sup&gt;</th>
<th>SSCR</th>
<th>HWR</th>
<th>HTGR</th>
<th>SBR</th>
<th>MSCR</th>
<th>MSBR</th>
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<tr>
<td>Equilibrium or last-cycle data</td>
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<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Exposure, MWD/t of fuel</td>
<td>21,000</td>
<td>25,400</td>
<td>29,400</td>
<td>52,300</td>
<td>12,000</td>
<td>2300</td>
<td>---</td>
</tr>
<tr>
<td>Fuel lifetime, full-power years</td>
<td>1.83</td>
<td>1.46</td>
<td>1.64</td>
<td>4.95</td>
<td>2.18</td>
<td>0.21</td>
<td>0.08</td>
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<tr>
<td>Feed enrichment, wt.% fissile material</td>
<td>2.2</td>
<td>4.8</td>
<td>2.1</td>
<td>3.1</td>
<td>1.4</td>
<td>2.6</td>
<td>---</td>
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<tr>
<td>Fabrication and processing plant size for 15,000 MW(e), tonnes of fuel per year</td>
<td>670</td>
<td>555</td>
<td>590</td>
<td>189</td>
<td>1,300&lt;sup&gt;c&lt;/sup&gt;</td>
<td>(f)</td>
<td>(f)</td>
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<tr>
<td>Fabrication cost, $/kg of fuel</td>
<td>52.50</td>
<td>47.20</td>
<td>37.50</td>
<td>115.00</td>
<td>51.50&lt;sup&gt;d&lt;/sup&gt;</td>
<td>0</td>
<td>0</td>
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<td>Processing cost, $/kg of fuel</td>
<td>31.20</td>
<td>39.00</td>
<td>39.70</td>
<td>105.00</td>
<td>23.00&lt;sup&gt;e&lt;/sup&gt;</td>
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<tr>
<td>Fuel shipping costs (fabricated + irradiated), $/kg of fuel</td>
<td>4.00</td>
<td>8.00</td>
<td>5.90</td>
<td>25.60</td>
<td>5.00</td>
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<td>0</td>
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<tr>
<td>Out-of-pile hold-up time, d</td>
<td>310</td>
<td>360</td>
<td>330</td>
<td>390</td>
<td>290</td>
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<tr>
<td>Initial fissile inventory, kg</td>
<td>2,060</td>
<td>2,910</td>
<td>1,450</td>
<td>2,910</td>
<td>3,500</td>
<td>1750</td>
<td>620</td>
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<td>Value of first core loading, $10^6</td>
<td>15.05</td>
<td>35.82</td>
<td>18.33</td>
<td>36.00</td>
<td>43.82</td>
<td>21.00</td>
<td>7.44</td>
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<tr>
<td>Fabrication cost of first core, $10^6</td>
<td>5.44</td>
<td>3.20</td>
<td>3.00</td>
<td>16.40&lt;sup&gt;1&lt;/sup&gt;</td>
<td>8.60</td>
<td>1.30&lt;sup&gt;g&lt;/sup&gt;</td>
<td>4.39&lt;sup&gt;g&lt;/sup&gt;</td>
</tr>
<tr>
<td>Reactor</td>
<td>PWR(^a)</td>
<td>SSCR</td>
<td>HWR</td>
<td>HTGR</td>
<td>SBR</td>
<td>MSCR</td>
<td>MSBR</td>
</tr>
<tr>
<td>------------------</td>
<td>------------</td>
<td>------</td>
<td>-----</td>
<td>------</td>
<td>-----</td>
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</tr>
<tr>
<td>Fuel-cycle cost, mill/kWh(e)</td>
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<tr>
<td>Fabrication</td>
<td>0.34</td>
<td>0.25</td>
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<td>0.26</td>
<td>0.61</td>
<td>0.04</td>
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<tr>
<td>Net fuel burn-up and losses (^b)</td>
<td>0.71</td>
<td>0.60</td>
<td>0.40</td>
<td>0.19</td>
<td>0.12</td>
<td>0.07</td>
<td>-0.08</td>
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<tr>
<td>Processing</td>
<td>0.20</td>
<td>0.20</td>
<td>0.24</td>
<td>0.19</td>
<td>0.27</td>
<td>0.08</td>
<td>0.12</td>
</tr>
<tr>
<td>Shipping</td>
<td>0.03</td>
<td>0.04</td>
<td>0.04</td>
<td>0.05</td>
<td>0.05</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>Fissile and fertile inventory</td>
<td>0.25</td>
<td>0.72</td>
<td>0.43</td>
<td>0.51</td>
<td>0.85</td>
<td>0.02</td>
<td>0.06</td>
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<tr>
<td>Interest on fabrication</td>
<td>0.08</td>
<td>0.03</td>
<td>0.05</td>
<td>0.17</td>
<td>0.08</td>
<td>0.37</td>
<td>0.17</td>
</tr>
<tr>
<td>Interest on processing</td>
<td>-0.01</td>
<td>-0.01</td>
<td>-0.01</td>
<td>-0.03</td>
<td>-0.03</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>Net fuel-cycle cost</td>
<td>1.60</td>
<td>1.83</td>
<td>1.37</td>
<td>1.34</td>
<td>2.05</td>
<td>0.58</td>
<td>0.35</td>
</tr>
</tbody>
</table>

\(^a\) Fuelled with partially enriched uranium.
\(^b\) Losses = 1% in processing, +0.2% in fabrication.
\(^c\) For breeder, pre-breeder through-put is 860 t/yr.
\(^d\) For breeder, pre-breeder costs, $64/kg.
\(^e\) For breeder, pre-breeder costs, $30 50/kg.
\(^f\) Integrated processing.
\(^g\) Costs associated with carrier salt, including \(^7\)Li.
\(^h\) Losses = 0.1% per pass through processing.
\(^i\) Includes cost of initial purchase of BeO.
fissile inventory. Low processing cost is obtained even with quite short cycle times by the use of fluoride volatility and distillation processes; the estimated cost of an integrated processing plant for the MSBR is less than $5,000,000.

Because the inventory and interest charges used in this study may not apply in many cases, and because the price of uranium ore may differ from the reference value used, the fuel cycle costs reported in Table III have been recalculated for different economic assumptions. The cycles have not been re-optimized for the changed conditions, however, and somewhat lower costs than those given could be achieved by re-optimization.

As seen in Table IV, doubling the reference cost of U₃O₈ (making it $35.2/kg) would increase the fuel cycle costs of the MSBR by 0.03 mill/kWh, the MSCR by 0.15, the HTGR by 0.24, the HWR by 0.29, the SBR by 0.37, and the PWR and SSCR by about 0.46 mill/kWh. Reducing the inventory and interest charge from 10 to 5% would benefit the SBR the most because of its large fuel inventory and benefit the MSR's and PWR the least because of their low inventory costs.

POWER COSTS

Capital and operating costs of the reactors were estimated with the help of Sargent and Lundy Engineers from information provided by the sponsors of the concepts. Since design assumptions varied from one concept to the next, common features or conditions of plants were made comparable before the costs were normalized. A plant design was not available for the SBR and its capital cost was taken as that of the PWR. The costs for the molten-salt reactors were obtained by perturbation of the costs estimated for the sodium graphite reactor in [1]. A change from [1] for all reactors is the use of total indirect costs equal to 32% of the direct costs rather than 41%; the lower value seems more consistent with costs reported for large plants.

Table V presents the estimated capital and operating costs and combines them with the fuel cycle costs given in Table III to obtain power costs. Values are given for both the reference economic condition and for inventory rates and fixed charges that are more appropriate for government ownership or for special financing and taxation arrangements. The cost of heavy water includes both inventory charges and an assumed 2% per year loss rate.

The low capital costs of the HTGR are attained largely through use of the prestressed-concrete vessel that contains the entire primary system. The SSCR cost is lower than that of the PWR because of savings resulting from the high core power density. Elimination of a mechanical fuel handling system, reduced control requirements, low system pressure, and small heat transfer areas in the heat exchangers kept the costs of the molten-salt reactor from being high in spite of the high melting coolant and the intermediate heat transfer loop.

The capital cost values shown of course have considerable uncertainty associated with them. More confidence, however, can be placed in the costs of one concept relative to the others. An examination of the details of the estimates indicates that the capital cost of any of the solid fuel plants relative to the others could be as much as 0.1 mill/kWh lower than the values shown for the reference conditions. Only the HWR, the HTGR, and the molten-salt...
<table>
<thead>
<tr>
<th>Fuel cost for reference conditions (mill/kWh(e))</th>
<th>PWR</th>
<th>SSCR</th>
<th>HWR</th>
<th>HTGR</th>
<th>SBR</th>
<th>MSCR</th>
<th>MSBR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effect of change in fuel inventory and fabrication interest charges to 5% per year (mill/kWh(e))</td>
<td>1.44</td>
<td>1.46</td>
<td>1.13</td>
<td>1.00</td>
<td>1.53</td>
<td>0.38</td>
<td>0.23</td>
</tr>
<tr>
<td>Effect of change in uranium ore cost, with bred fuel values changed in proportion to fully-enriched uranium (mill/kWh(e))</td>
<td>1.30</td>
<td>1.55</td>
<td>1.20</td>
<td>1.19</td>
<td>1.83</td>
<td>0.47</td>
<td>0.33</td>
</tr>
<tr>
<td>At $8.8/kg U₃O₈ ($0.54/g ²³⁵U)</td>
<td>2.07</td>
<td>2.29</td>
<td>1.66</td>
<td>1.58</td>
<td>2.42</td>
<td>0.73</td>
<td>0.38</td>
</tr>
</tbody>
</table>
| At $35.2/kg U₃O₈ ($16.23/g ²³⁵U) | *Values given are perturbations of the base cases without reoptimization.*
### TABLE V

**CONTRIBUTIONS TO POWER COST**

<table>
<thead>
<tr>
<th>Reactor concept</th>
<th>PWR</th>
<th>SSCR</th>
<th>HWR</th>
<th>HTGR</th>
<th>SBR</th>
<th>MSCR</th>
<th>MSBR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plant capital cost ($/kW(e))</td>
<td>124</td>
<td>116</td>
<td>126</td>
<td>110</td>
<td>124</td>
<td>123&lt;sup&gt;a&lt;/sup&gt;</td>
<td>128&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Power cost with 12% fixed charges on reactor plant and 10% fuel inventory and fabrication interest charges (mill/kWh(e))</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Capital</td>
<td>2.1</td>
<td>2.0</td>
<td>2.2</td>
<td>1.9</td>
<td>2.1</td>
<td>2.1</td>
<td>2.2</td>
</tr>
<tr>
<td>Operating</td>
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<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>Fuel cycle</td>
<td>1.6</td>
<td>1.8</td>
<td>1.4</td>
<td>1.3</td>
<td>2.0</td>
<td>0.6</td>
<td>0.4</td>
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<td>Heavy water</td>
<td>—</td>
<td>0.1</td>
<td>0.5</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Total</td>
<td>4.0</td>
<td>4.2</td>
<td>4.4</td>
<td>3.5</td>
<td>4.4</td>
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<td>Power cost with 7% fixed charges on reactor plant and 5% fuel inventory and fabrication interest charges (mill/kWh(e))</td>
<td></td>
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<tr>
<td>Capital</td>
<td>1.2</td>
<td>1.2</td>
<td>1.3</td>
<td>1.1</td>
<td>1.2</td>
<td>1.2</td>
<td>1.3</td>
</tr>
<tr>
<td>Operating</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
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<tr>
<td>Fuel cycle</td>
<td>1.4</td>
<td>1.5</td>
<td>1.1</td>
<td>1.0</td>
<td>1.5</td>
<td>0.4</td>
<td>0.2</td>
</tr>
<tr>
<td>Heavy water</td>
<td>—</td>
<td>0.1</td>
<td>0.3</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Total</td>
<td>2.9</td>
<td>3.1</td>
<td>3.0</td>
<td>2.4</td>
<td>3.0</td>
<td>1.9</td>
<td>1.8</td>
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<sup>a</sup> Excluding on-site processing facilities, for which charges are included under "Fuel Cycle Cost".
reactors could be much higher than the estimates: as much as 0.1 mill/kWh in the case of the HWR, and up to 0.2 mill/kWh for the HTGR (largely because of uncertainties about the concrete reactor vessel). The cost estimates of the molten-salt reactors are more uncertain than the others because less is known about the designs. Since much of the equipment in power plants is similar for all concepts, and since we have tried to be conservative in making the estimates, it would not appear that the capital costs of the MSR's could be underestimated by more than 0.2 mill/kWh.

Although the same operating cost is used for all plants, there will clearly be some variations, but insufficient information is available at present to make a significant distinction.

CONCLUSIONS

The results of our evaluation show that there are thorium cycle reactors which can have both lower power cost and better fuel utilization than a PWR. As often occurs in comparative evaluations, however, the systems which are the least developed or about which the least is known, the HTGR and the MSR's, appear to have the lowest costs. Since our estimates are intended to represent what the costs would be if the reactors were successfully developed, they do not reflect uncertainties in the technology.

The molten-salt reactors have the lowest power costs of all the reactors studied as a result of their low fuel cycle cost, but there are a number of design and development problems which need to be solved. The MSBR, in fact, appears to violate the ground rule that the technology used in the designs be limited to that which could be used in a prototype to go on line in 1970. However, the use of on-site processing for the MSR's does eliminate one economic uncertainty in that achievement of low fuel cycle costs would not await the growth of a large support industry. In spite of the development problems which need to be solved, the MSBR offers a particular virtue in that it can breed and at the same time could have low enough power costs to make it attractive to power system operators.

Of the solid fuel reactors, the HTGR appears particularly promising, but both the graphite fuel elements and the "all-inside" concrete pressure vessel require demonstration in a power reactor. Thus while the HTGR concept is flexible and alternate design approaches are available, its success will depend on the favourable outcome of forthcoming tests of its important features and on creation on an adequate scale of the somewhat unique fabrication and processing facilities it requires.

The heavy-water reactor evaluated in this study has good fuel utilization but its high power cost makes it unattractive. However, studies by others of using organic or boiling light-water coolants for HWR's have shown that appreciable reductions in capital costs, including the D₂O inventory, can be obtained. By a substitution of coolant the power cost of the HWR thus possibly could be lowered significantly from the value we obtained. Since the SSCR offers neither lower power costs nor significantly better fuel utilization than the PWR and has the disadvantage of having heavy water in the coolant, its further development seems unlikely.
The SBR has good fuel utilization but high power cost. It is difficult to compare it with the other reactors, however, since it was designed for a particular breeding ratio and was not optimized for the conditions of our study.

ACKNOWLEDGEMENTS


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