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THE THORIUM FUEL CYCLE

E. R. Merz

Institute for Chemical Technology
Kernforschungsanlage Jülich GmbH
D-5170 Jülich / Germany

1. INTRODUCTION

Utilization of nuclear energy is based upon two fuel cycles: the uranium-plutonium and the thorium-uranium cycle. Without the breeding possibilities on the two fertile isotopes ^{238}U and ^{232}Th leading to fissile ^{239}Pu and ^{233}U, respectively, the world-wide resources of naturally occuring fissile ^{235}U would soon become exhausted. Breeding provides a simple means for multiplying the amount of fissile nuclear material present in natural uranium resources, thus justifying from a political economy point of view, the immense capital expenditure required for installing a nuclear industry.

Todays nuclear energy exploitation virtually is based exclusively on the uranium-plutonium cycle, applying mostly low enriched uranium in Light Water Reactors (LWR's). In a few countries, Heavy Water-moderated Reactors (HWR's) operated with natural uranium are being in use as well.

Thorium as a fertile material has primarily been recommended for its application in High-Temperature Gas-cooled Reactors (HTR's) since this reactor type is especially well suited for an economic operation with a Th-^{233}U fuel cycle [1, 2]. Unfortunately, the initially very promising outlooks for a fast market introduction of HTR's have failed. The cause however, by no means is due to lacking fuel cycle capabilities, but far more presents a problem of fundamental character:

The question is whether or not there is any demand at all for an additional reactor system besides the presently market dominating water-moderated reactors, e.g. LWR's and possibly HWR's, together with the Fast Breeder Reactors (FBR's), world-wide under development.

This question has been discussed many times at large in the past, sometimes even with great emphasis and emotions. Uncontested are the advantages of achievable maximum helium gas coolant temperature for an optimum transposition into exploitable power, either by increased thermal efficiency applying a closed-cycle gas turbine for electricity generation, or even more, when utilized as process heat [3]. It seems as if only the latter could provide the HTR a real market chance, even though not in the near future. Even in case of a wide-spread application of HTR's as a source of high-temperature process heat, no thorium fuel cycle is required inevitably for their operation. High-temperature gas-cooled reactors may just as well be operated - even though with markedly reduced economy - on a so-called low-enriched uranium cycle [4]. On the other hand, the development of a complete new fuel cycle could be omitted due to the fact that the already existing uranium-plutonium fuel cycle technology is applicable to a considerable extent.

2. ARGUMENTS IN FAVOUR OF THE THORIUM FUEL CYCLE

First of all the question may be raised as to whether or not a thorium fuel cycle is needed at all, and that, independent of a particular reactor type. It seems advisable to answer this question in the light of the following three criteria: prospects of technical realization, economy and long-term fuel supply guaranty.

The most advantageous mode of utilization the thorium fuel cycle is to recycle the generated ^{233}U. This requires reprocessing and refabrication capabilities. Results thus obtained so far from extensive R&D programs performed in several countries provide good confidence that both reprocessing of thorium-containing fuel elements as well as recycling of recovered ^{233}U are technically feasible just as well as it is the case for fuel values from the competitive uranium-plutonium cycle [5, 6].

The open question still to be answered deals with the recycling costs. Because of the fact that todays industrial technology in this particular field is much less advanced than in the uranium-plutonium fuel cycle, the cost estimates exhibit the handicap of a wide margin of error. However, even in connection with comparably pessimistic assessment values for the specific recycling costs, tolerable and sometimes even markedly lower energy production costs for the thorium fuel cycle are calculated. Altogether, the overall economy proves to be favourable in certain cases.

The decisive argument however, in favour of the thorium fuel cycle is the long-term availability and securing of raw materials supply. Operation strategy calculations for different reactor systems lead to the conclusion that a successful application of thorium in several power reactor systems gives rise to remarkable uranium ore savings. A combined application of both the uranium and the thorium fuel cycle thus guarantees long-term fuel supply at tolerable and fairly stable prices [7, 8, 9]. Nevertheless, one should be aware of the fact that there is no need for a thorium fuel cycle in case fast breeder reactors, utilizing ^{239}Pu, become operable on an industrial scale at the beginning of next century. The plutonium 239 recovered from spent LWR fuel elements serves as starting material. With increasing number of FBR power stations in operation, annual requirements of uranium ore will be decreasing to a considerable extent due to the fact that for fertile fuel element fabrication plenty of stockpiled depleted uranium is available. Altogether, the economically exploitable resources of uranium ore estimated will suffice over several centuries applying the combination of LWR's and FBR's.

But what happens if after all fast breeder power reactors are introduced into the market either too late or their introduction fails at all?

This risk may well be overcome by application of a thorium fuel cycle. Basically, there are several reactor types suitable for this purpose.

3. APPLICATION OF THE THORIUM FUEL CYCLE IN VARIOUS REACTOR TYPES

When studying this problem, it seems advisable to distinguish between light water, heavy water and high-temperature reactors operated in the thermal range, on the one hand, and the fast breeder reactors on the other hand. The fast reactors may be either of sodium-cooled or of gas-cooled type. Since, in respect of thorium application, the two concepts exhibit rather similar fuel cycle characteristics, they are being dealt with here as a single system.

A thorough analysis of the possibility of thorium application in various thermal reactor systems reveals reduced uranium ore requirements in all cases. A presupposition is however, the availability of a recycling technology. Under certain circumstances, and particularly in the event of further increasing uranium prices, there should be even slight cost advantages in favour of thorium application. The most promising reactor system seems to be the high-temperature reactor [10] followed by the heavy water-moderated type. Both systems promise equally good overall fuel utilization performance but with substantially higher unit capital costs incurred for the HWR. This is mainly due to additional expenditures resulting from its heavy water inventory (D_2O) [11].

Thorium utilization in LWR's is likewise possible giving rise also to quite measurable uranium ore savings but at distinct increased fuel cycle expenditures [12]. Thus, thorium application in LWR's might at best be of interest if fast breeders would find their way into the market only with great time delay and in case of further increasing uranium prices.

Fast breeder reactors are as well suited for thorium application, and that even under different alternatives, i.e. in the core, in the blanket or in both of them [13].

TABLE I summarizes a semi-quantitative comparison of the different reactor systems considered. Further details may be learned from calculations and studies recently carried out by several authors on which results the statements made are based [12, 14, 15, 16]. The system of LWR + FBR with Pu recycle is competitive with the system of HTR + HWR + possibly LWR with ^{233}U recycle in regard to both consumption of uranium as well as availability of plutonium. If, during the next 30 years, a thorium fuel cycle utilization in thermal reactors for energy production purposes would dominate over the application of a uranium-plutonium cycle in LWR's, the FBR's may become short of the ^{239}Pu required for their start-up phase beginning around the year of 2000. Fast breeder people therefore are afraid of a widespread thorium application, since it could seriously endanger their plutonium supply. The same risk arises from extensive recycling of plutonium in light water reactors.

Both argumentations are neither valid for the years coming. On the one hand, there is no adequate reprocessing capacity available for LWR fuel elements so that plutonium recycling in LWR's is impossible anyhow, and on the other hand, first of all a ^{233}U recycling technology as well as capacity would be required before any competitive situation could materialize at all. Therefore, it is quite impossible during the next ten years to make any wrong decision in this respect. This time period, however, should be utilized for adequate development work in the thorium fuel cycle field so that finally, use can be made of the best option.

4. ADVANTAGES OF THE THORIUM FUEL CYCLE

Owing to the increased conversion factors obtained on ^{233}U in a thermal neutron spectrum as a result of the favourable ratio (α) from neutron capture to fission, the thorium cycle is slightly ahead of the uranium-plutonium cycle. In a fast neutron spectrum, the α-values for ^{233}U and ^{239}Pu are approximately of the same order. Nevertheless, the values for the U-Pu-cycle altogether result in a somewhat more favourable neutron economy (higher ν- and ε'-values) leading to slightly improved conversion rates [17]. Yet the application of a thorium fuel cycle in both reactor types of HTR and HWR might involve a real alternative to fast breeder reactors. In comparison, light water reactors are by far less attractive.

However, the large amount of funding still required for a complete development of this kind of power generating reactor systems can only be justified if the necessity of a back-up solution for the fast breeder is fully accepted.

A further important aspect, namely that of saving uranium, points in the same direction. Supposed, the thorium fuel cycle would be employed soon, valuable amounts of mined uranium could be saved within about the next 30 years prior of a market penetration of the fast breeders. Even subsequent of their market introduction, and having gained the leading position, a mixed fuel service (U-Pu/Th-U) may turn out in the long run to offer the most reliable basis of fuel supply. Next to the HTR, the heavy water-moderated reactor (CANDU-type) represents another interesting possibility for the application of a thorium fuel cycle. In spite of the assumed higher capital costs due to its D_2O-inventory, only slightly increased total energy production costs in mills/kWh are resulting. However, equal uranium ore savings are achieved in both cases.

Another important aspect may well be of interest. HWR's do not require highly enriched uranium, they are operated either with natural uranium or at best with slightly enriched ^{235}U, thus lowering separative work requirements to a large extent. Eventually, it cannot be ruled out that heavy water-moderated reactors could be made marketable within a relatively short period of time, demanding reduced expenditure of development for the system itself as well as for the fuel cycle. The question arises as to whether or not the heavy water reactor after all may be praised the so-called "forgotten reactor"?

The thorium fuel cycle is further characterized by two additional features of less importance: that is to say by recycling of uranium instead of plutonium and by substantially reduced built-up rates for long-lived α-instable transuranic isotopes in the Th-U-chain. Thus the risk of long-term waste storage resulting from reprocessing of spent fuel elements is remarkably reduced by one or two orders of magnitude, assuming comparable separation factors for the transuranic elements for both fuel cycles [18]. Altogether refabrication of ^{233}U has to be carried out remotely under radiation shielding due to ^{232}U-impurities in the recovered ^{233}U, it may turn out in the end that recycling of ^{233}U is easier than that of Pu. This statement is not merely based upon existing psychological reluctance against the element plutonium as such but more because of quantifyable technical and technological facts. Processing of uranium into the most diversified fuel elements is well known to the one, and to the other the sophisticated and problematic plutonium chemistry and metallurgy has lost its relevance to fissile materials recycling.

The high intrinsic radioactivity of ^{233}U which normally requires additional efforts and expenses, may be turned to a positive side. This uranium requires strong shielding for handling in order to avoid hazardous radiation exposure of personnel, thus providing self-protection against unauthorized profileration of fissile material. It has therefore already been proposed by safeguards people to contaminate recovered plutonium values with high artificial radioactivity.

Finally, a last problem should be mentioned. Handling of pure ^{233}U (exactly as in the case of ^{239}Pu) involves safeguards problems whose administrative and technical consequences cannot fully overlooked as yet. It is imaginable that again the thorium fuel cycle turns out to be somewhat more favourable than the plutonium cycle due to the fact that only denatured ^{233}U is permitted for refabrication and recycling purposes, respectively. By limiting the content of ^{233}U to less than 20 % in the ^{238}U, safeguarding problems will be reduced considerably. In this case, fissile and fertile material cannot be separated any more applying purely chemical methods as it may be possible for a plutonium-uranium system. It would require a very costly and difficult achievable isotope separation capability. Therefore, the system presents inherent protection against misuse for military weapons manufacturing.

In conclusion, several profound arguments speak in favour of a thorium fuel cycle. But what is still lacking, is the availability of an adequate reprocessing technology on an industrially proven scale.

5. THORIUM DEPOSITS AND RESERVES

Deposits of thorium in nature are known to be about twice to three times as abundant as those of uranium. Two kinds of thorium minerals are found: the primary magmatic and the sondary sedimentary ones, the former being encountered relatively seldom.

The main ores from which thorium is industrially extracted are monazite, deposits of which are found at many places around the world. As a primary mineral, monazite occurs in small amounts in pegmatites where it is an end product of crystallization of once molten mineral solutions. Monazite is an isomorphous mixture of rare-earth phosphates in which thorium phosphate and silicate are found associated in proportions ranging from 1-20 %. The economically significant deposits of monazite are formed by weathering of pegmatites, followed by gravity concentration of heavy minerals in sand beds through the action of wind and water. Such weathering is more rapid in the tropics than in cooler zones, and it is the tropics that most deposits of monazite sand have been found.

A second mineral is the thorite, a thorium silicate with a thorium oxide equivalent ranging from 5 to 10 %.

The known reserves of thorium are classified at present merely under one price category. It is of the order of < 10 \$/lb ThO_2 according to 1973 price level. Only cheapest deposits are having a chance at all to be exploited. The data available on deposits, extent, etc. are rather fragmentary. Thorium prospecting was never carried out systematically, but far more at random, as demand had always been lower than the assured resources. The world principal thorium reserves known today, estimated as of beginning of the seventieths [19], are shown in TABLE II. Output figures in different countries are largely unknown today. Due to the small demand actually prevailing, they will hardly exceed 10,000 tons per year.

6. FUEL CYCLE TECHNOLOGY

6.1. General aspects

The utilization of the Th-^{233}U fuel cycle in thermal reactors requires the development of a closed fuel cycle. It is characterized by both the supply of the necessary nuclear fuel and the disposal of predominantly radioactive waste products as is illustrated by figure 1. Of course, analogous requirements will also apply to the use of thorium in fast breeder reactors. Desupplying of nuclear power stations, that means reprocessing, refabrication and waste management, are just as decisive steps in the nuclear fuel cycle than are uranium production, uranium enrichment and fuel manufacturing. It was only a few years ago that common awarness of the importance for the feasibility of recycling capabilities compared to the other steps of the fuel cycle has still been very meagre. It is quite understandable that manufacturers as well as utilities are afraid to develop reactor systems on the basis of a thorium fuel cycle and to support their market introduction as long as the technological and economic risks inherent to this system cannot be calculated on the basis of well-proven data sets. The question frequently posed as to whether or not it would be more advisable first to develop a nuclear power plant or the associated fuel cycle, can be answered clearly in this instance.

As matters stand today, first of all the basic R & D problems associated with thorium reprocessing and ^{233}U-refabrication have to be investigated with highest priority followed by the demonstration of their usability on an industrial scale. Fuel element manufacturing, shipping and storage actions, as well as a complete waste management may be tackled only with secondary priority. Suitable processes for the fabrication of thorium-containing fuel elements for the different reactor types under consideration are either already known or if necessary, may rapidly be adapted in line with existing fabrication processes from the uranium cycle to required specifications.

Thorium fuel cycle waste treatment resembles very much to that of a LWR fuel reprocessing plant. Only the graphite combustion in the course of HTR fuel reprocessing requires minor extra efforts. A direct thorium disposal possibly becoming necessary is almost without any problems. All other steps of the thorium fuel cycle are more or less indentical with the uranium cycle and, consequently, do not require any additional expenditure for development.

6.2. Process flow-sheets

To illustrate the most important and characteristic individual steps of reprocessing and refabrication, simplified process flow-sheets are represented in figures 2 and 3.

They give a general idea, at least from a qualitative point of view, of the development work still necessary to close the thorium fuel cycle. It becomes evident that altogether higher expenditures are required for the HTR than is the case with LWR's or HWR's. This is due to the special requirements of the combustion head-end of the graphitic HTR fuel elements. An additional expenditure of considerable extent in comparison with the mixed particle concept is likely to arise in this connection on making use of a separate TRISO-feed and BISO-breed particle system. A TRISO-TRISO feed-breed system presumably would be even more costly.

In the case of refabrication of ^{233}U, the two reactor fuel concepts are counterbalancing each other in respect to necessary development expenditures. Experience gained from cold (fresh) HTR fuel fabrication and from LWR fuel production will facilitate hot recycling development work to a large extent.

Assuming a successful introduction of the thorium fuel cycle in water-moderated reactors, it would be recommendable to take advantage of the good experience obtained in connection with remote HTR fuel kernel production as an alternative refabrication process. The oxide or carbide microsphere preparation process in mind is a sol-gel precipitation starting from a metal oxide sol. Compaction of the sintered kernels in the fuel tubes will then be achieved via vibration, thus replacing the established fuel compact concept.A basic assumption for a successful application of a ^{233}U-refabrication process is its simplicity and reliability. Remote operation capability is the prime necessity.

6.3. Cost and time requirements

Several phases of development are necessary before getting to the final demonstration of an overall process under industrial measures. Firstly, it will start with R & D work on laboratory scale using unirradiated and irradiated fuel material, terminated by the submission of a preliminary process and apparatus flow-sheet. The time needed for this development period is at least 5 years. For most of the individual tasks illustrated in figures 2 and 3, work has already been completed in this start-up phase, particularly as far as the HTR fuel cycle is concerned [5, 6] . Lab-scale work is followed by thorough engineering scale component development. In parallel, it is advisable to start already with the basic design engineering for pilot plants. Time requirement should be in the order of about 4 years.

The next step is a so-called pilot plant operation phase with extensive test operational runs employing unirradiated and as far as possible also irradiated fuel specimens of original size and composition. The minimum necessary time period to perform this program element is about 5 years. In the course of this project phase, conceptual studies for prototype and demonstration units should already be undertaken.

Partially in line or slightly temporal delayed follows the fourth development phase which is devoted to the design, construction and test operation of individual prototypical process units and equipment or complete facilities. Along with it, preliminary design work is performed for the demonstration plant. The time period required for this project phase should be rated at not less than 3 years.

The project will be finally concluded with the fifth phase covering the construction and commissioning of a demonstration plant of representative capacity, which is considered today to be in the order of an equivalent of 5 GWe power plant output. The necessary period of time should hardly be less than 6 years.

A question of fundamental importance concerns the development costs. Any conclusive answer cannot be given here as the relevant charges are different from country to country, dependent on previous work existing, structure of research and economy, etc. A very rough estimate exhibits expenses to the amount of about 250 mill. $ covering all R & D work inclusive of prototype testing; the cost of a demonstration plant for reprocessing and refabrication should not exceed 600 mill. $. In addition, expenses for irradiation experiments of recycle fuel elements in the order of about 30 mill. $ may arise.

This assessment applies to the HTR fuel cycle. Significantly lower overall costs might be estimated for water-moderated reactors.

7. CONCLUSIONS

The statements made in this paper clearly indicate the advantages that can be derived using the thorium fuel cycle. They primarily invclve preservation of uranium resources and long-term low cost energy supply.

Maximum benefits can be achieved applying an HTR system. The heavy-water moderated reactor ranks number two, followed at clear distance by the present-day light-water reactors. A thorium fuel cycle can also be used profitably in future fast-breeder reactors.

Thus, numerous arguments speak in favour of a systematic and consequent development of the thorium fuel cycle. The main emphasis in this connection should be focussed on both reprocessing and partial sectors of refabrication. As a matter of fact, a meaningful R & D program can in part be implemented independent of a specific reactor type, for many problems are the same for all fuel element systems.

The funds to be made available are certainly not low; however, a realistic cost-benefit evaluation presents a very favourable picture in comparison with alternative future sources of energy production.

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TABLE I: Comparison of Different Reactor Systems in Regard to Capital and Fuel Cycle Costs as well as Uranium Ore Savings Applying a Thorium Fuel Cycle

(The estimates are based upon U_3O_8 prices of 60 $/kg and separative work costs of 75 $/kg SWU)

Reactor System	Conversion Rate	Relative Capital Costs for Power Plant *	Fuel Cycle	Fuel Recycling (^{239}Pu resp. ^{233}U)	Relative Specific Fuel Cycle Costs	Uranium Reserves Lasting for Years (Resources $3{,}5 \times 10^6$ t U_3O_8)
LWR	0,6	100	U-Pu	no	100	25
	0,6	100	U-Pu	yes	85	40
	0,8	100	Th-U	yes	135	30
	~ 1,0	110	Th-U	yes	200	50
HWR	0,7	125 φ	U-Pu	no	75	30
	0,8	125 φ	Th-U	yes	75	70
	~ 1,0	125 φ	Th-U	yes	160	> 100
HTR	0,65	105	Th-U	no	90	30
	0,65	105	Th-U	yes	85	55
	0,8	105	Th-U	yes	95	70
	~ 1,0	110	Th-U	yes	140	> 100
FBR	> 1,1	? (≥ 200 ?)	U-Pu +Th-U	yes	105	> 200

* For LWR's an index number of 100 was chosen

φ Includes the costs for D_2O

<u>TABLE II:</u> Estimated Low-Priced World Reserves of Thorium

Country	Reasonably Assured Resources (x 1000 Tons)	Estimated Additional Resources (x 1000 Tons)
Canada	90	90
U.S.A.	60	300
South Africa	20	?
Egypt	20	320
Brazil	5	35
India	350	?

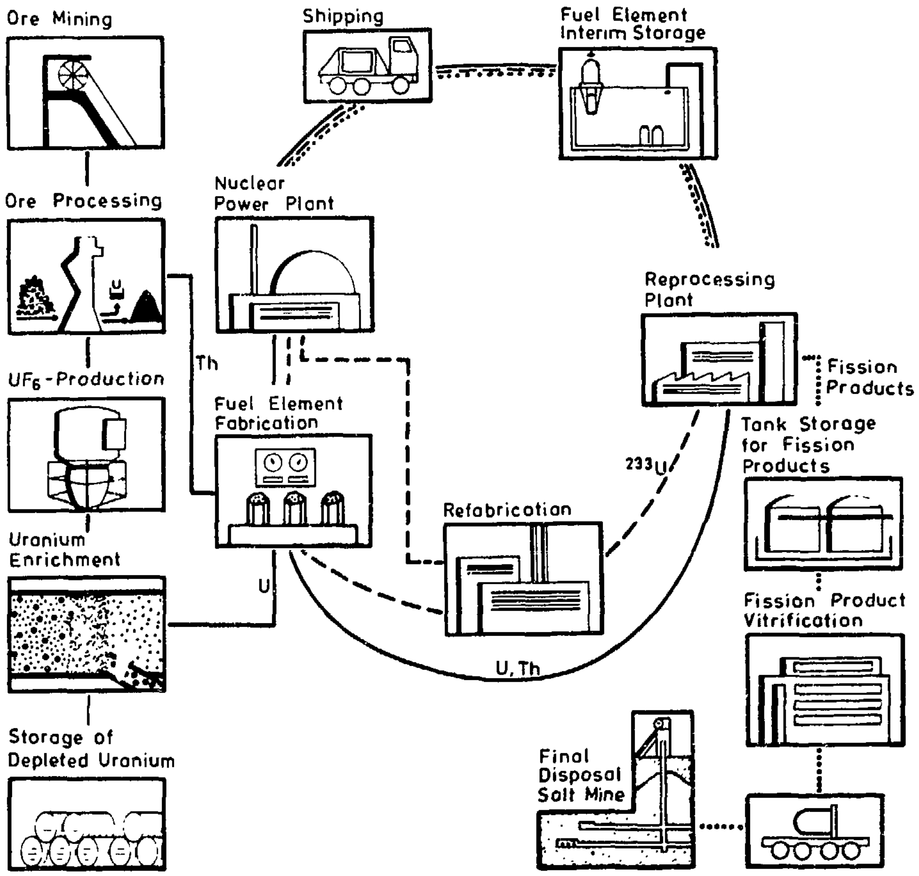

Fig. 1: The Thorium Fuel Cycle

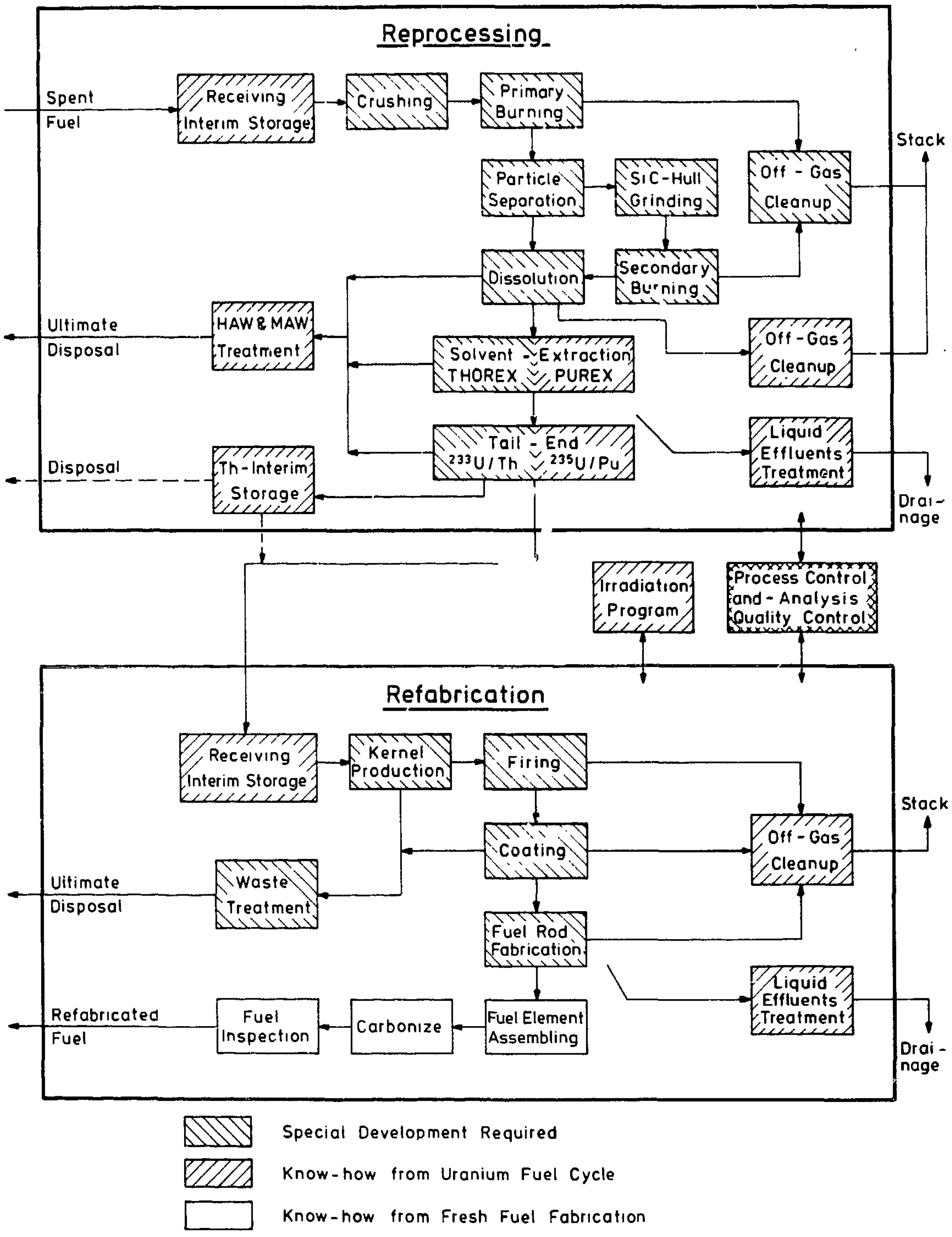

Fig.2: Simplified Flowsheet for Reprocessing, Refabrication and Waste Treatment of Fuel Elements from HTR's

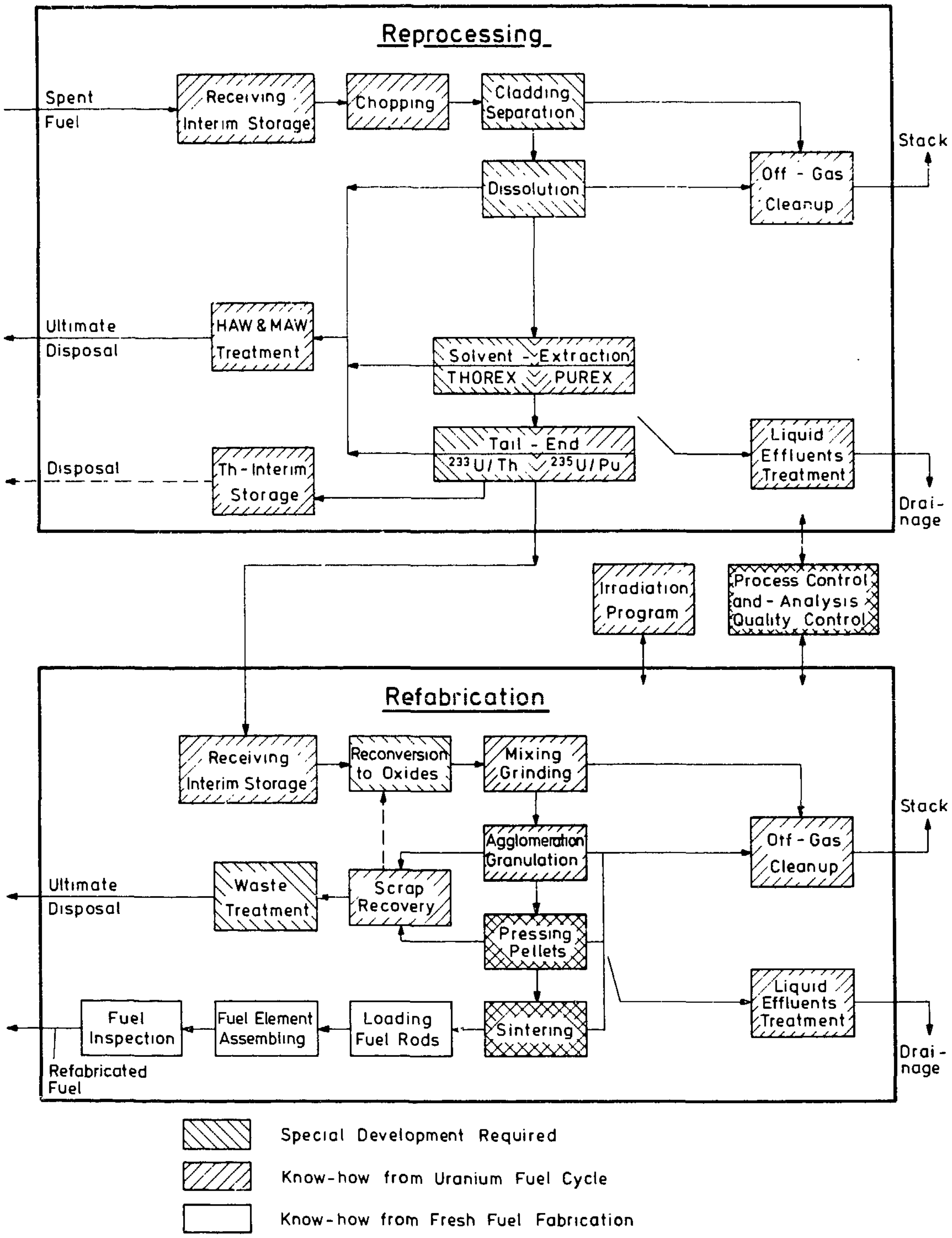

Fig.3: Simplified Flowsheet for Reprocessing, Refabrication and Waste Treatment of Fuel Elements from LWR's, HWR's and FBR's