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# EVALUATION OF THORIUM BASED NUCLEAR FUEL

## Extended Summary

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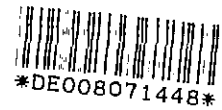



# EVALUATION OF THORIUM BASED NUCLEAR FUEL

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## Abstract

Application of thorium based nuclear fuels has been evaluated with emphasis on possible reduction of the actinide waste. As a result three ECN-reports are published, discussing in detail:
- the reactor physics aspects, by comparing the operation characteristics of the cores of Pressurized Water Reactors and Heavy Water Reactors with different fuel types, including equilibrium thorium/uranium fuel, once-through uranium fuel and equilibrium uranium/plutonium fuel [1],
- the chemical aspects of thorium based fuel cycles with emphasis on fuel (re)fabrication and fuel reprocessing [2],
- the possible reduction in actinide waste as analysed for Heavy Water Reactors with various types of thorium based fuels in once-through operation and with reprocessing [3].

These results are summarized in this report together with a short discussion on non-proliferation and uranium resource utilization.

It has been concluded that a substantial reduction of actinide radiotoxicity of the disposed waste may be achieved by using thorium based fuels, if very efficient partitioning and multiple recycling of uranium and thorium can be realized. This will, however, require large efforts to develop the technology to the necessary industrial scale of operation.



## Keywords

Thorium
Fuel Cycle
Reactor Physics
Nuclear Waste

# CONTENTS

# 1. Introduction

Uranium, either natural or enriched in the fissile isotope $^{235}U$, is being applied as fuel for almost all nuclear power reactors currently in operation. The energy value of the initial fissile material $^{235}U$ is increased considerably because new fissile material $^{239}Pu$ is being produced during reactor operation by neutron absorption in the fertile $^{238}U$ and successive beta-decays. Most of this plutonium is fissioned in-situ before the fuel is unloaded. This formation of fissile plutonium leads to a substantially increased burnup of the uranium fuel and to a more economical use of the primary uranium resources.

Natural thorium $^{232}Th$ may be applied as fertile material instead of $^{238}U$. Similar to the $^{239}Pu$ production in uranium fuel, $^{232}Th$ can be transmuted to the fissile uranium isotope, $^{233}U$. This process is the basis of a series of alternative fuel cycles: the thorium fuel cycles. As, however, no fissile thorium isotopes occur in nature, thorium cycles have to be ignited with external fissile material, such as $^{235}U$ extracted from natural uranium or plutonium from reprocessed LWR fuel or weapons inventories.

In the past, the thorium cycles were studied worldwide as an option to improve the uranium utilization at the, then envisaged, large expansion of nuclear energy production; thorium resources are supposed to exceed uranium resources by at least a factor of four [4-9]. Conceptual designs have been made for the use of thorium in a variety of reactor types [9-12]. However, due to the world-wide stagnation in nuclear power generation, sufficient fissile material (uranium and plutonium) is available at low price, as fuel for the present-day nuclear reactor capacity even for a substantial period of time. So, resource optimization alone would not justify, at present, the expensive development and implementation of thorium based fuel cycles.

Recently the use of thorium has regained attention because of its prospects to reduce the radiotoxicity of actinide waste. The usual spent uranium fuel contains substantial amounts of transuranium elements, i.e. various plutonium, americium, neptunium and curium isotopes and these long-lived radioactive materials are dominantly responsible for the long-term radiotoxicity of the spent fuel. Much effort is spent, nowadays, to develop waste management strategies for reducing this long-term radiotoxicity by advanced partitioning and transmutation processes. Reduction by one or two orders of magnitude may be feasible in this way, if reprocessing techniques will be improved to partition with high efficiency the various actinides and if special burner reactors will be operated for large-scale transmutation [13,14].

The thorium cycles may offer a complementary approach to handle this part of the waste problem. Replacing $^{238}U$ by $^{232}Th$ will reduce considerably the production of transuranium nuclei per unit of fission energy [19,20]. On the other hand the non-natural uranium isotopes $^{232}U$, $^{233}U$ and $^{234}U$, typical for spent thorium fuel, are produced and may have an adverse impact on the long-term radiotoxicity of the waste.

At ECN the perspectives and drawbacks of thorium cycle options have been analysed with special attention to the nuclear waste reduction, both reduced actinide production and enhanced burning capability. This study included reviews on basic reactor physics characteristics and on fuel cycle technology as well as a generic computational analysis on long-term radiotoxicity of the waste of thorium fuel in comparison to uranium fuel.

These items are discussed in detail in the following reports, resulting from this ECN study:

* *Evaluation of Thorium Based Nuclear Fuel; Reactor Physics*, By J.H. Bultman and W.J.M. de Kruijf [1].

* *Evaluation of Thorium Based Nuclear Fuel; Chemical Aspects*, By R.J.M. Konings, P.J.A.M. Blankenvoorde and E.H.P. Cordfunke [2].

* *Evaluation of Thorium Based Nuclear Fuel; Actinide Waste*, by V.A. Wichers [3].

The present report is an extended summary of the above mentioned ECN-reports and may be regarded as a general review of the thorium cycle, especially with respect to the nuclear waste aspects. Two subjects are additionally addressed in this report: non-proliferation and uranium utilization connected to fuel cycles with thorium.

# 2. Thorium Fuel Cycles

In thorium based fuels the isotope $^{232}Th$ is applied as fertile material. As no fissile thorium isotopes exist in nature, thorium-fuelled reactors have to be started up with some external fissile material (*startup fuel*). When reactor operation is progressing, new fissile material, $^{233}U$, is bred from thorium. The amount of $^{233}U$ approaches equilibrium at a concentration of about 1.5%, and this material is then (partially) feeding the fission process in the reactor. In practice, fissile material has to be added to the fresh fuel (*makeup fuel*) to ensure reactor criticality for the designed burnup period at appropriate (economical) reactor conditions.

Several fissile materials are candidates for startup or makeup fuel:

Uranium enriched in $^{235}U$, either as High Enriched (about 92%) Uranium, HEU, or as Medium Enriched (less than 20%) Uranium, MEU. HEU is the obvious option when limitation of the $^{238}U$ inventory in the fuel is pursued, but it will require fabrication, transport and handling of this weapon-grade fuel material.

Plutonium, either from reprocessed spent uranium fuel or from weapon inventories. Unlike weapon plutonium, the reprocessed LWR plutonium contains substantial amounts of fissionable (non-fissile in a thermal spectrum) Pu-isotopes.

Uranium-233, from previously irradiated and reprocessed thorium fuel.

The various thorium fuel cycles can be characterized by the type of fissile material selected as startup or makeup fuel and by the way the back-end of the cycle is arranged, either final storage of spent fuel (once-through) or recycling of useful fuel components.

*Once-through thorium cycles* with high, medium or low enriched uranium additives have been considered from the very beginning [9]. Presently also thorium/plutonium based fuels are studied as a possible way to burn the plutonium from reprocessed LWR fuel or from weapons inventories [17]. "Once-through" variants have been studied at which low enriched uranium fuel (LEU) and natural thorium are supplied to the reactor core in separate elements (the so called mixed uranium/thorium cycle) [17]; such a configuration permits optimal, but different burnup for the LEU fuel and for the thorium elements, in which as much of the produced $^{233}U$ as possible has to be burned in-situ. This type of operation requires reactor systems with very flexible fuel management systems, such as CANDU or HTR.

*Closed fuel cycles* are considered to optimize the energy potential from $^{233}U$; these cycles are of long-term strategic interest [5]. Thorium and uranium are reprocessed and re-used for the next fueling. If sufficient breeding could be achieved in the reactor then no additional makeup fuel but only some fresh thorium is required for fuel refabrication, otherwise also some "fresh" makeup fuel has to be added.

# 3. Reactor Physics Characteristics

The evaluation of reactor physics characteristics includes a discussion of the basic nuclear data of the fissile and the fertile components and an analysis of steady state behaviour and dynamic characteristics of reactors operating with thorium.

## 3.1 Basic Data of the Fissile and Fertile Materials

### 3.1.1 The Fissile Isotopes

Only four nuclides, $^{233}U$, $^{235}U$, $^{239}Pu$ and $^{241}Pu$, are candidate materials for the fissile component in both uranium and thorium based fuels for large scale nuclear reactor applications. Although a thorough analysis on fission characteristics and behaviour, specific for each of these materials, can only be performed in combination with realistic reactor designs, some general remarks can be made on the basis of basic nuclear data. In table 3.1 some data with respect to thermal capture and thermal fission are presented.

Table 3.1 *Basic nuclear data for the fissile isotopes [1]*

| | $^{233}U$ | $^{235}U$ | $^{239}Pu$ | $^{241}Pu$ |
|---|---|---|---|---|
| $\sigma_\gamma$ [b] | 45.5 | 98.3 | 269.3 | 358.2 |
| $\sigma_f$ [b] | 529.1 | 582.6 | 748.1 | 1011.1 |
| $\alpha$ | 0.086 | 0.169 | 0.360 | 0.354 |
| $\nu$ | 2.493 | 2.425 | 2.877 | 2.937 |
| $\eta$ | 2.296 | 2.075 | 2.115 | 2.169 |
| $\beta$ | 0.0028 | 0.0070 | 0.0023 | |

$\sigma_\gamma$ : thermal cross section for neutron capture (in barn)
$\sigma_f$ : thermal cross section for fission (in barn)
$\alpha$ : ratio $\sigma_\gamma / \sigma_f$
$\nu$ : average number of neutrons produced per fission
$\eta$ : average number of neutrons produced per absorption
$\beta$ : delayed neutron fractions

The thermal capture cross section of $^{233}U$ is relatively small, whereas its fission cross section is almost equal to the value for $^{235}U$. The ratio of thermal capture to thermal fission ($\alpha$) is the smallest for $^{233}U$, indicating that this nuclide is relatively most efficient for thermal fission.
This is also illustrated by the parameter $\eta$: the number of neutrons created per neutron, absorbed by the fissile nuclide. The value of $\eta$ is of prime interest for considerations of neutron economy. Of the newly created $\eta$ neutrons, obviously one neutron is requested to maintain the nuclear chain reaction. Furthermore, a fraction of a neutron is required as compensation for the loss of neutrons by absorption in construction materials, coolant, control materials and fission products and for the neutrons that escape by leakage from the core. Finally, the remaining balance of neutrons is available for

converting fertile into fissile material. This conversion process is characterized by the quantity CR, the conversion ratio, which is defined as the average number of fissile nuclides produced in a reactor per consumed (fission or capture) fuel nuclide. Higher values of $\eta$ enable higher values of CR and so a more economic use of the initial fissile inventory. The specific value of $\eta$ is spectrum (reactor type) dependent as shown in table 3.2.

Table 3.2 *Values of $\eta$ for some typical spectra*

| | Thermal (2200 m/s) spectrum [1] | Typical HTR spectrum [4] | Typical FBR spectrum [1] |
|---|---|---|---|
| $^{233}U$ | 2.28 | 2.24 | 2.31 |
| $^{235}U$ | 2.07 | 1.95 | 1.93 |
| $^{239}Pu$ | 2.11 | 1.78 | 2.49 |
| $^{241}Pu$ | 2.15 | | 2.72 |

For thermal spectra (CANDU or HTR) the nuclide $^{233}U$ is the most efficient fissile isotope, whereas the plutonium isotopes may be most appropriate for fast breeder reactors.
It has always been a challenge for reactor designers to achieve a thorium based, thermal reactor system with breeding properties, i.e. for each fissile nuclide destroyed, at least one new fissile atom has to be produced by conversion of fertile thorium. In that case, at least two neutrons are required to maintain the nuclear reaction and to keep the fissile inventory at constant level. The remainder ($\eta$-2) neutrons are available to counterbalance parasitic absorptions and neutron leakage and to achieve reactor control in practice. The margin $\eta$-2 is, at thermal spectra, rather limited for all fissile nuclides but for $^{233}U$ this margin is the highest. It may, in principle, be possible to realize a thermal breeder [9,15,16], but it will be hard to achieve economical operation. It seems worthwhile to study the capability of fast reactors for $^{233}U$ breeding, but this is beyond the scope of the present work.

### 3.1.2 The Fertile Isotopes

The fertile materials considered are $^{232}Th$ and $^{238}U$. These materials are of importance for the reactor physics properties of the fuel because they are the major components of the fuel. In table 3.3 some basic data are given.

Table 3.3 *Basic data of the fertile materials [1]*

| | $^{232}Th$ | $^{238}U$ |
|---|---|---|
| $\sigma_\gamma$ (thermal neutrons) (b) | 7.4 | 2.7 |
| $L_\gamma$ (b) | 85 | 227 |
| Fission Threshold (MeV) | 1.5 | 0.8 |

$\sigma_\gamma$ : cross section for thermal neutron capture (in barn)
$L_\gamma$ : resonance integral for neutron absorption (in barn)

The thermal capture cross section is for thorium higher than for the uranium isotope and for this reason higher enrichment in fissile material is required for thorium-based fuel than for uranium fuel, for equivalent burnup in a reactor of the same power output.

## 3.2 Reactor operation and control

### 3.2.1 Protactinium Holdup

The basic breeding reaction sequence for thorium fuel is:

$$^{232}\mathrm{Th}\ (n,\gamma)\ ^{233}\mathrm{Th} \xrightarrow[22m]{\beta} {}^{233}\mathrm{Pa} \xrightarrow[27d]{\beta} {}^{233}\mathrm{U}$$

which is rather similar to the reaction sequence for the uranium cycle:

$$^{238}\mathrm{U}\ (n,\gamma)\ ^{239}\mathrm{U} \xrightarrow[24m]{\beta} {}^{239}\mathrm{Np} \xrightarrow[2.3d]{\beta} {}^{239}\mathrm{Pu}$$

Main differences between the two breeding reactions are the cross section for thermal neutron capture (the capture cross section for $^{232}$Th is about three times larger than for $^{238}$U) and the half-life of the intermediate nuclide in the beta-decay process (half-life of $^{233}$Pa is 27 days, much larger then the 2.3 day half-life of $^{239}$Np). After startup of a reactor new fissile material ($^{239}$Pu or $^{233}$U) will buildup, evolving to a certain equilibrium concentration. For the thorium cycle the equilibrium concentration is about 1.5 % $^{233}$U per amount of thorium and this equilibrium is reached on a time scale, dictated by the decay to $^{233}$U (27 days). The equilibrium concentration of $^{239}$Pu in $^{238}$U is only 0.3 % and is achieved on a much shorter time scale. The relatively long half-life of $^{233}$Pa causes a protactinium holdup in the reactor core. The equilibrium protactinium concentration is strongly flux dependent, increasing at higher fluxes [1,10].

There is a number of effects related to the protactinium holdup:

- a neutron may be absorbed by $^{233}$Pa and the created $^{234}$Pa will decay rapidly to $^{234}$U. This has a negative effect on the neutron economy, non-fissile $^{234}$U is formed and creation of fissile $^{233}$U is prevented.
- a relatively high concentration of protactinium may still be present in the reactor core at shut down, decaying slowly to fissile $^{233}$U. The reactor control system should account for this reactivity increase after shut down.
- the protactinium concentration in the core is dependent on local flux history; emanating power peaking problems must be avoided.

In some reactor concepts the fuel elements with high $^{233}$Pa concentration are unloaded or placed in positions with low flux, to favour decay to $^{233}$U in stead of parasitic absorption in $^{233}$Pa.

### 3.2.2 Reactivity of Stationary Systems

In the reactor physics part of the ECN study [1] the reactivity of stationary (equilibrium) systems has been analysed. The neutron balance of typical PWR and typical HWR (CANDU) geometries has been determined for three different fuel types:

- pure thorium/$^{233}$U fuel assuming recycling,
- uranium/plutonium fuel (MOX) assuming recycling
- 3.3% enriched uranium (fresh fuel) as reference case for PWR and natural uranium (fresh fuel) as reference case for CANDU.

Conversion ratios higher than one can be obtained for the thorium cycle, but the excess reactivity is very small. So, the reactivity loss by buildup of absorbing fission products can only be compensated for a short period, allowing for only very limited burnup. In this respect CANDU is more promising than PWR.

The burnup may be increased substantially by adding neutrons to the reactor from an external source. Then, the reactor can be operated even when the effective multiplication factor is smaller than one. Recently Rubbia et al [18] made a proposal for a system at which a subcritical, thorium fuelled reactor was driven by an external neutron source, consisting of a proton accelerator with spallation target.

### 3.2.3 Dynamic Behaviour

Bultman and De Kruijf [1] extended the analyses to the impact on the dynamic reactor behaviour, for the same reference cases, mentioned above.

Two aspects are important when comparing thorium and uranium fuels:

- the fraction of delayed neutrons as a measure for the margin to prompt criticality
- the inherent feedback mechanisms (such as Doppler effect and coolant temperature effect) as measures for the ability to limit power excursions.

As shown in table 3.1 the delayed neutron fraction for $^{233}$U is considerably smaller than for $^{235}$U and this shall be a point of concern at the design of the reactor control system.

Computational results [1] with respect to the fuel temperature coefficient did not show a relevant difference between thorium and uranium for both PWR and CANDU type of reactors. The coolant void coefficient is smaller for thorium fuel, negative for the PWR and positive for CANDU.

# 4. Chemical Aspects of the Thorium Fuel Cycle

## 4.1 Occurrence and Ore Processing of Thorium

*Occurrence of thorium*
Thorium ores are found in many countries but systematic exploration has not been performed. Large "reasonably assured resources" (RAR) are present in Brazil, India, Norway, Turkey and USA. The RAR thorium resources in the world (excluding Eastern Europe, the former USSR and China) are at least about 2 $10^3$ Mg and about the same quantity of thorium is reported as "estimated additional resources" (EAR) [21].

*Thorium ore processing*
The ore processing of monazite, the principal thorium ore, is well established and is quite similar to the ore processing of uranium: concentration of the ore by electromagnetic techniques followed by dissolution in acid, selective precipitation and solvent extraction. Presently, the thorium production is limited. It is primarily a by-product of the exploitation of heavy metal ores (titanium, rare earths, zirconium).

## 4.2 Fuel Fabrication

Many types of thorium fuels can be distinguished, dependent on the amount of fissile makeup (ranging from 1 wt% to 10 wt%), on the type of fissile material ($^{233}U$, $^{235}U$, $^{239}Pu$) and on the chemical composition of the fuel (metal, oxide, nitrate or carbide). So, a variety of different fuel materials is possible, all with their specific needs at the fabrication process.
Some experience exists on the fabrication of thoria and mixed thoria-urania pellets for water-cooled reactors and on the fabrication of coated particles with thoria for high-temperature gas-cooled reactors.

### 4.2.1 Thoria and Mixed Thoria/Urania for LWRs

Much of the technology for fabricating thoria and thoria/urania fuel pellets comes from the light-water breeder reactor (LWBR) programme in the USA [9]. In-pile irradiation tests indicated that high density and high integrity of the fuel pellets were required to obtain, among others, a good thermal conductivity, to minimize gas-release and fuel dimensional changes and to reduce fuel-cladding interaction. Based on these results a process was developed for the fabrication of such high-density, high-integrity thoria-based fuel pellets from oxide powders [22]. This process is also suited for remote control fabrication, which is required because of the radiotoxicity of natural thorium. However, the pellets produced in this way did not always meet the required quality due to variabilities in the thoria starting material.

A sol-gel process was developed for the production of thoria-urania micro-spheres, the so-called SOLEX process [23]. The major advantages of this

process are its simplicity, which makes it excellently suited for remote operation, and the minimal production of dust, which is the major concern in radiation protection.

### 4.2.2 Thoria Coated Particles for HTRs

The microspheres for HTR fuel of German design [24] are produced by a sophisticated technique: the solution flows through oscillating nozzles, forming droplets with subsequent solidification. The so formed microspheres are then coated with several protective layers. This process is well established and excellently suited for remote operation.

## 4.3 Irradiation Performance

The available information on the irradiation behaviour of $ThO_2$-based fuels, reviewed in ref. [25], indicates that $(Th,U)O_2$ is comparable to, and in many respects superior to $UO_2$. However, the majority of the experiments were performed with low $UO_2$ content (<10%). Data on fuels with $UO_2$ content greater than 20% are scarce. The capability of this fuel type needs to be demonstrated, particularly under off-normal conditions.

## 4.4 Fuel Reprocessing, the THOREX Process

For reprocessing of thorium-based spent fuel, the THOREX process has been designed, especially for oxide type of fuel. This process is, like the PUREX process for uranium spent fuel, a hydrochemical method based on solvent extraction techniques, but with special adaptations for the dissolution step of thorium fuel with a fluoride catalyst and for the partitioning of thorium and uranium. Due to limited experience, the THOREX process is technically less advanced than the PUREX process and there is little experience on an industrial scale.

### 4.4.1 Chemistry

The first step in the THOREX process is to prepare the spent fuel for solubilization of the thorium and the uranium dependent of the fuel type:

- $ThO_2$-$UO_2$ spent fuel in stainless steel or zircaloy cladding which is removed mechanically.
- HTR spent fuel, consisting of coated particles of $ThO_2$ (or $ThC_2$) and $UO_2$ (or $UC_2$), embedded in a graphite matrix. The particles are crushed and the carbon is burned off.
- Th or $ThO_2$ in aluminium cladding, which can be dissolved with or without cladding.

Next, the fuel has to be prepared for the solvent extraction process. During dissolution of the fuel in a (13 M $HNO_3$ + 0.05 M HF + 0.1 M $Al(NO_3)_3$) solution, thorium and uranium form aqueous ions. In case of oxide fuel, thorium will react to give $Th^{4+}$(aq) and uranium will be oxidized from U(IV) to U(VI) to give $UO_2^{2+}$(aq). During the latter reaction, gaseous nitrous oxides

are formed such as NO and $NO_2$ which will escape from the solution together with the fission gases (Kr, Xe, $I_2$) and volatile fission products (e.g. Ru).

Uranium and thorium are extracted from the aqueous phase with a solution of tri-n-butyl phosphate (TBP) in kerosene. The ions are specifically concentrated in the organic phase or in the aqueous phase, depending upon their complex formation with the TBP. At high TBP concentration (≈ 30 %) both uranium and thorium are extracted into the organic stream whereas the fission products are concentrated in the aqueous stream.

A thorium/uranium partitioning by changing the valency of thorium is not possible, in contrast to the uranium/plutonium partitioning in the PUREX process. Thorium is separated from uranium by contacting the loaded organic phase with an aqueous phase containing 0.5 M $HNO_3$. At this condition thorium is selectively re-extracted to the aqueous phase. By using multiple separation steps more than 99.99% of thorium is separated from the uranium stream. In order to recover the uranium, the organic phase is subsequently multiple contacted with a 0.01 M $HNO_3$ solution. In this way more than 99.99% of uranium is recovered.

In case that also an appreciable amount of plutonium is present in the U/Th mixture, all the plutonium is co-extracted as Pu(IV) with U and Th in the extraction step. By the addition of a reducing agent, $Pu^{4+}$ is reduced to $Pu^{3+}$ which is re-extracted to the aqueous phase. Thorium can be stripped in the next step, and after that the uranium. Finally, the uranium and thorium and/or plutonium streams can be purified by ion exchange.

If a small amount of minor actinides (Am, Np, Cm) is present in the thoria fuel, the Am and Cm concentrate in the aqueous waste stream, whereas Np accompanies the U product. In order to recover the Am and Cm, the aqueous waste can be treated in the TRUEX or DIAMEX process.

### 4.4.2 Development and Status

The THOREX process was initially developed at Oak Ridge National Laboratory in the USA. A pilot plant has been operated in which 1.3 Mg irradiated $ThO_2$ was reprocessed. In addition, military programmes have been executed in the USA to produce large amounts of $^{233}U$ [26]. In Germany attempts were made to develop variants of the THOREX process for the reprocessing of (high burnup) fuel from HTRs. A single-cycle process was proposed, in which the requested decontamination factors could be realized. However, the recommended flow sheet has not been tested on pilot plant scale, using real spent fuel.
In the eighties, German and Brazilian researchers investigated thorium based fuels for PWRs. They proposed a single-cycle acid THOREX process, at which reprocessing of $(ThU)O_2$ fuel in zircaloy cladding will be feasible.
In India, flow sheets have been developed for selective recovery of uranium from irradiated thoria fuel. Because the uranium product still contains significant amounts of thorium as impurity, additional purification steps are needed. In pilot plant operation the thorium contamination could be reduced to 50-150 ppm. [27]

## 4.5 Fuel Refabrication

The $^{233}U$ obtained from the reprocessing of thorium fuel always contains some $^{232}U$. This isotope produces very intense $\gamma$-emitting daughter nuclides among which $^{208}Tl$ is notable ($E_\gamma$=2.6 MeV). Similarly, the reprocessed thorium contains significant amounts of $^{228}Th$, a decay product of $^{232}U$, leading again to the intense gamma-radiation of $^{208}Tl$. As a result, fuel refabrication using reprocessed uranium or thorium can only be done in gamma-shielded hot-cell laboratories.

## 4.6 Waste Management

Final treatment of the waste from thorium based fuel may be performed according to similar processes as the waste from uranium fuel. More attention must, however, be paid to the vitrification process, where the presence of fluoride ions enhances corrosion, and to the handling procedures, due to the very intensive gamma irradiation from $^{232}U$-daughters. Moreover, the glass volume of wastes from the THOREX process is estimated to be considerably greater ( about 70%) compared to wastes from the PUREX process [28].

# 5. Actinide Waste

The production of actinide waste has been studied for various thorium cycles in comparison to the once-through uranium cycle. In this context, actinide waste is defined as those actinides for which in a given fuel cycle no re-use is foreseen and which is therefore disposed. The evaluation is based on burnup calculations, performed at ECN [3], for the case of a Heavy Water Reactor (HWR). The HWR is selected as reference reactor because of its good neutron economy, thus enabling better conversion ratios, better utilization of the thorium itself, and reduced requirement of additional fissile makeup fuel.

## 5.1 Overview of the Studied Thorium Fuel Cycles

Nine HWR fuel cycles with oxide type fuel are considered:

- a *reference fuel cycle*, standard for present CANDU operation.
  * Natural uranium fuel; exit burnup 7 MWd/kg (cycle 1)

- four *once-through fuel modes* with fresh fuel consisting of thorium and additive makeup fuels of, respectively, high (HEU) or medium (MEU) enriched uranium or of plutonium with isotopic composition corresponding to PWR spent fuel with discharge burnup of 45 MWd/kg.
  * Th,$PuO_2$; Pu-fissile-fraction: 1.66 wt% HM;
    Exit burnup 12 MWd/kg (cycle 2)
  * Th,$PuO_2$; Pu-fissile-fraction: 2.19 wt% HM;
    Exit burnup 25 MWd/kg (cycle 3)
  * Th,HEU ; $^{235}$U-fraction : 1.86 wt% HM;
    Exit burnup 10 MWd/kg (cycle 4)
  * Th,MEU ; $^{235}$U-fraction : 2.00 wt% HM;
    Exit burnup 18 MWd/kg (cycle 5)

- four *closed fuel cycles* with fresh fuel, which consists of three components: thorium, uranium recycled from previously irradiated spent thorium fuel and additive makeup fuel, either HEU or Pu or "pure" $^{233}$U. Only recycle of the uranium is considered, with reprocessing after sufficiently long cooling to allow decay of $^{233}$Pa. An out-of-core loss of 0.1% is assumed. The analysis is made for equilibrium fuel cycles, when all successive reloads are supposed to be identical in composition.
  * Th with U-recycle and HEU topping;
    U-fissile-fraction: 2.12 wt% HM
    Exit burnup 32 MWd/kg (cycle 6)
  * Th with denatured U-recycle and HEU topping;
    U-fissile-fraction: 2.16 wt% HM
    Exit burnup 32 MWd/kg (cycle 7)
  * Th with U-recycle and Pu topping;
    Fissile-fraction : 2.26 wt% HM
    Exit burnup 32 MWd/kg (cycle 8)
  * Th with U-recycle and $^{233}$U topping;
    U-fissile-fraction: 2.01 wt% HM
    Exit burnup 32 MWd/kg (cycle 9)

## 5.2 The Effect of Fresh Fuel Components

*Thorium*
Irradiation of thorium generates various isotopes of protactinium and uranium. The basic reaction for thorium is the conversion reaction to $^{233}U$ (see section 3.2). Furthermore, the parasitic (n,2n)-reaction of thorium leads to $^{231}Th$ and, after beta-decay, to $^{231}Pa$, and this isotope gives a major contribution to the actinide-radiotoxicity of the spent thorium fuel. Also the uranium isotopes $^{232}U$, produced from $^{233}U$ by a (n,2n) reaction and $^{234}U$, produced by (n,$\gamma$) reactions from $^{233}U$ or $^{233}Pa$, are relevant in this respect.

*Recycle Fuel*
Recycle uranium contains, besides the fissile $^{233}U$ and $^{235}U$, also large fractions of $^{234}U$ and $^{236}U$ and a small fraction of $^{232}U$. Neutron absorption in $^{236}U$ leads to $^{237}Np$. The isotope $^{232}U$, decaying with a half-life of 71.7 year, produces very intense $\gamma$-emitting daughter nuclides, which are the main source for the high gamma activity of the recycle fuel.

*Makeup Fuels*
Makeup fuels on the basis of enriched uranium contain, in various compositions, the uranium isotopes $^{235}U$ and $^{238}U$. Higher actinides, such as $^{237}Np$, Pu-isotopes, $^{241}Am$, $^{243}Am$ and small amounts of curium are produced during irradiation, like the actinide production in uranium based fuel. Irradiation of plutonium makeup leads to the production of higher plutonium isotopes and to americium and curium isotopes.

## 5.3 Results of the Calculations

Burnup calculations have been performed on a two-dimensional cell model for a standard CANDU fuel bundle and moderator geometry. Details on geometry, material composition, computational technique and basic data are given in ref. [3]. Values for the exit burnup have been fixed on the basis of the criterion that the reactor is operated with sufficient reactivity margin to ensure an assembly-averaged burnup $k_\infty$ of at least 1.05.

### 5.3.1 Radiotoxicity of Actinide Waste; once-through fuel modes

For the once-through modes, all actinides in the discharged fuel are considered to be waste. The total actinide radiotoxicity of this waste is presented in fig. 5.1 for the various fuel types, for storage times up to $10^6$ years. In comparison with natural uranium fuel, the thorium/HEU system shows a significant reduction of the radiotoxicity, at least for a period of $10^4$ years (up to one order of magnitude reduction). After that period the radiotoxicity will even exceed the value for the natural uranium case, mainly due to decay of $^{233}U$ and, to a minor extent, of $^{231}Pa$.

Plutonium makeup is not a reasonable choice when reduction of the radiotoxicity of the spent fuel is pursued. For the plutonium cycles the radiotoxicity of the waste exceeds the value of the natural uranium case during the full storage period. One has, however, to keep in mind that the plutonium, fed into the fresh thorium fuel, represents a certain amount of radiotoxicity and this plutonium will be mostly destroyed during the reactor

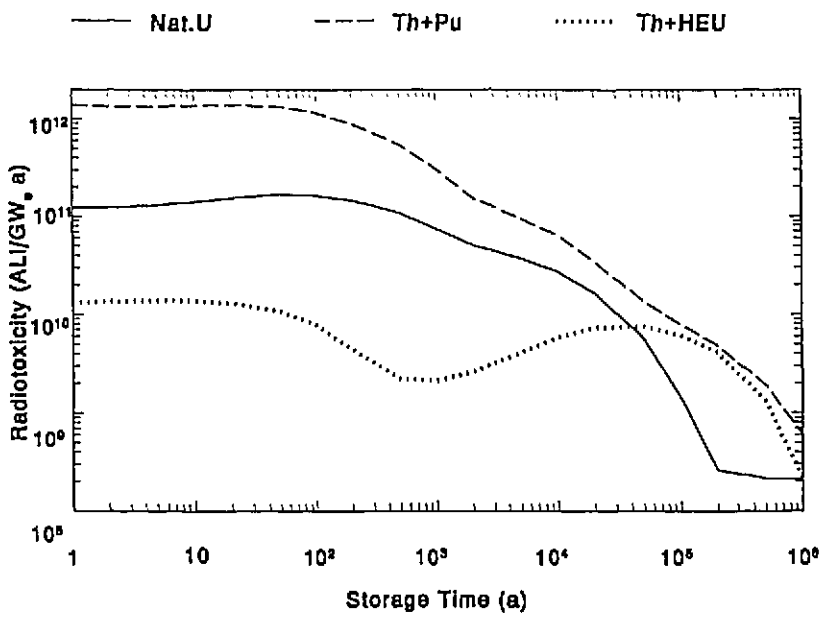


Figure 5.1 *Radiotoxicity of actinide waste in HWR once-through systems*

operation. This effect is not taken into account in fig. 5.1, because it would involve the analysis of waste per unit of electricity generated in a park consisting of LWRs and HWRs. The thorium/Pu cycle is most efficient for net plutonium burning; a net Pu-consumption of about 900 kg/GWe.a can be realized, much higher than achievable in present LWRs with MOX fuel loaded in 1/3 of the core.

### 5.3.2 Radiotoxicity of Actinide Waste; closed cycles

The out-of-core uranium losses (0.1%) and all other actinides in the spent fuel are considered to be the actinide waste. The computational results are related to fuel cycles approximating equilibrium, when all fuel reloads are identical.

In MEU/Thorium fuel the fissile isotopes ($^{233}U$ and $^{235}U$) are, during residence in the reactor, far more depleted than $^{238}U$. If the recycled uranium would be made up with fresh MEU, then the $^{238}U$ fraction of the recycled fuel would rapidly increase with recycling generation number and, within about ten years, the cycle would shift from a thorium cycle to a pure uranium-plutonium cycle. So, the option of a closed thorium cycle with MEU makeup will not be relevant to consider.

By using HEU (about 91% to 93% enrichment) as makeup, the total uranium fraction in the equilibrium fuel could be stabilized on a level of about 0.04 (52% enriched in fissile isotopes). Due to the rather substantial fractions of higher uranium isotopes there will be a buildup of mainly plutonium, explaining the relatively high level of the radiotoxicity of the waste during the first centuries of storage, see figure 5.2.

The fuel cycle of thorium with uranium recycle and $^{233}U$ makeup (derived from a hypothetical, external $^{233}U$-source) has been considered, because this cycle can be regarded as the cycle with the lowest possible transuranics production.

As shown in figure 5.2, the closed cycles with both HEU and $^{233}U$ makeup show the lowest long-term radiotoxicity. The results for these two cycles are rather similar:

- a major radiotoxicity contribution from $^{238}Pu$ during the first $10^3$ years
- a long-term contribution from $^{231}Pa$, originating from the (n,2n) reaction with thorium, to be considered as the threshold value for any thorium cycle. During the storage period from $10^3$ to $10^5$ years a maximum reduction by a factor of 100 can be noticed in comparison to the radiotoxicity of once-through natural uranium fuel.

Thorium itself gives a negligible contribution to the radiotoxicity of the waste. Recycling of plutonium could be considered, but it would reduce the actinide radiotoxicity only for the first $10^3$ years. Further reduction of the long-term radiotoxicity would require the recycling of $^{231}Pa$, which is considered highly undesirable, because it will lead to increased concentrations of protactinium and of $^{232}U$ in the spent fuel. Reprocessing and fuel refabrication will, then, become extremely difficult due to the very high gamma activity of the decay products of $^{232}U$.

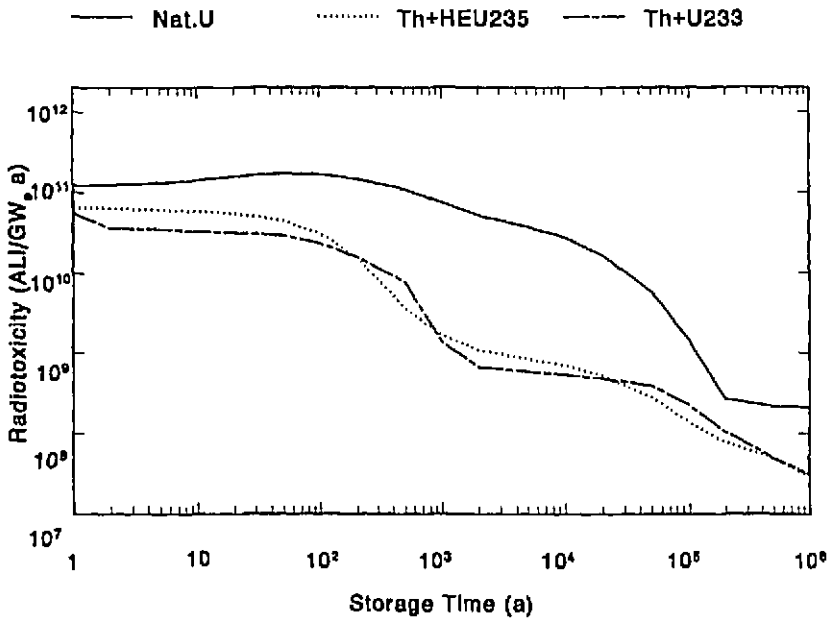


Figure 5.2 *Radiotoxicity of actinide waste of HWR cycles with U- recycling*

# 6. Non-proliferation

The properties of the thorium cycle concerning the issue of proliferation were already systematically investigated during INFCE [29]. The recent interest in the thorium cycle, motivated by promises with respect to long-lived waste, has re-evocated the discussion on proliferation resistance of this cycle.

Non-proliferation analysis should address the safeguardability of the thorium fuel cycle. In general, advanced fuel cycles may differ in the expenditure (initial and recurrent) required for meeting the technical IAEA Safeguards Objective "*to timely detect the diversion of a significant quantity of nuclear material*". These costs may be high compared with the current LWR fuel cycles, and in extreme cases the technical principles of fuel cycle facilities may make it intrinsically impossible to meet this IAEA Safeguards Objective.

The front end stages of the thorium cycle, comprising mining and milling, refining and fuel fabrication are very similar to the uranium fuel cycle.

In order to startup the reactor or to extend the fuel burnup period, external $^{235}U$ may be used as additional fissile material. To minimize the amount of transuranics produced, the $^{238}U$ content of this uranium is preferably as low as possible, so use of highly enriched uranium HEU is recommended in this respect. Therefore fresh HEU has to be produced in the enrichment plant and will be present in the fuel fabrication plant. HEU is a direct-use weapon material. Although verification of HEU is technically straightforward, severe requirements on the detection of misuse or diversion will be demanded. Similar reasoning holds when plutonium is used as startup or makeup fuel (as is the case for MOX fuel production).

Safeguarding will also be required for the protactinium, present in unloaded spent thorium fuel. Fast partitioning of the protactinium, followed by decay to $^{233}U$, may lead to production of high-quality weapons grade $^{233}U$ material.

When final disposal is selected as back end option, then no major distinctions are to be expected between safeguards issues of the disposal of spent uranium fuels and of spent thorium fuels, both qualitatively and quantitatively.

Concerning closed cycles, the uranium, partitioned from thorium fuel, and the plutonium, partitioned from uranium fuel, both direct-use materials, will not only be of the same safeguards category of nuclear materials, but also their quantities (annual throughput, inventories) in commercial-scale reprocessing plants will be similar. So, the same requirements on timeliness and accuracy of material accountancy measures must be adopted to uranium in a thorium reprocessing plant as to plutonium in a uranium reprocessing plant.
On the other hand the uranium, reprocessed from thorium fuel, will emit very intensive $\gamma$-radiation ($E_\gamma$=2.6 MeV), which can be regarded as an additional barrier against theft.

# 7. Utilization of Uranium Resources

Improvement of the energy potential of the uranium resources has, in the past, been the main incentive to study thorium cycles. In the present study the nuclear waste problem is the main point of view. Possible introduction of the thorium cycle cannot be realized on short term and will imply an important change in the nuclear fuel system. For this reason the long-term availability of resources is, still, relevant. From literature [7,8,29,30,31] a set of data has been deduced for the uranium ore requirements per unit of generated energy, for various fuel cycles and reactor systems. These data, shown in figure 7.1, are good approximations for the amounts of uranium ore required for the regular fuel reloads of the reactor, but do not account for the initial reactor inventory. Variations in initial inventories may have their impact on the lifetime uranium requirements of the reactors but are neglected here.

The following cases were considered:
- a typical PWR with 3.2% enriched uranium fuel, operating in once-through mode or with uranium and plutonium recycle
- CANDU with natural uranium fuel or with 1.2 % enriched uranium fuel, operating in once-through mode
- HTR-MODULE with 8% enriched uranium fuel, operating in once-through mode
- HTR-MODULE with thorium and HEU make-up fuel, with uranium recycle
- CANDU with thorium and HEU make-up fuel with uranium recycle
- large HTR with thorium and MEU (20%) make-up fuel, operating in closed cycle.

LWR operation without recycling (once-through) turns out to be most consuming with respect to uranium resources. Increased burnup of the fuel will lead to some improvement. Uranium and plutonium recycle in LWRs may further reduce the uranium ore requirement to about the same amount as needed for once-through HTR and CANDU.
For all reactor types the uranium requirement at once-through operation tends to decrease at increasing burnup of the fuel. However, the opposite trend is noticed for the cases with recycling, due to deteriorating fissile properties of the recycle uranium and plutonium at higher burnup.

Application of thorium will lead to a large reduction in primary uranium ore requirement if the spent fuel is reprocessed and the uranium is recycled.This is true for the small sized HTR-MODULE, even more for the large HTR and certainly for CANDU reactors, in which operation at thermal breeding may be approached.

For accelerator-based systems, such as recently proposed by CERN [18], no more uranium will be required after start-up of the system.

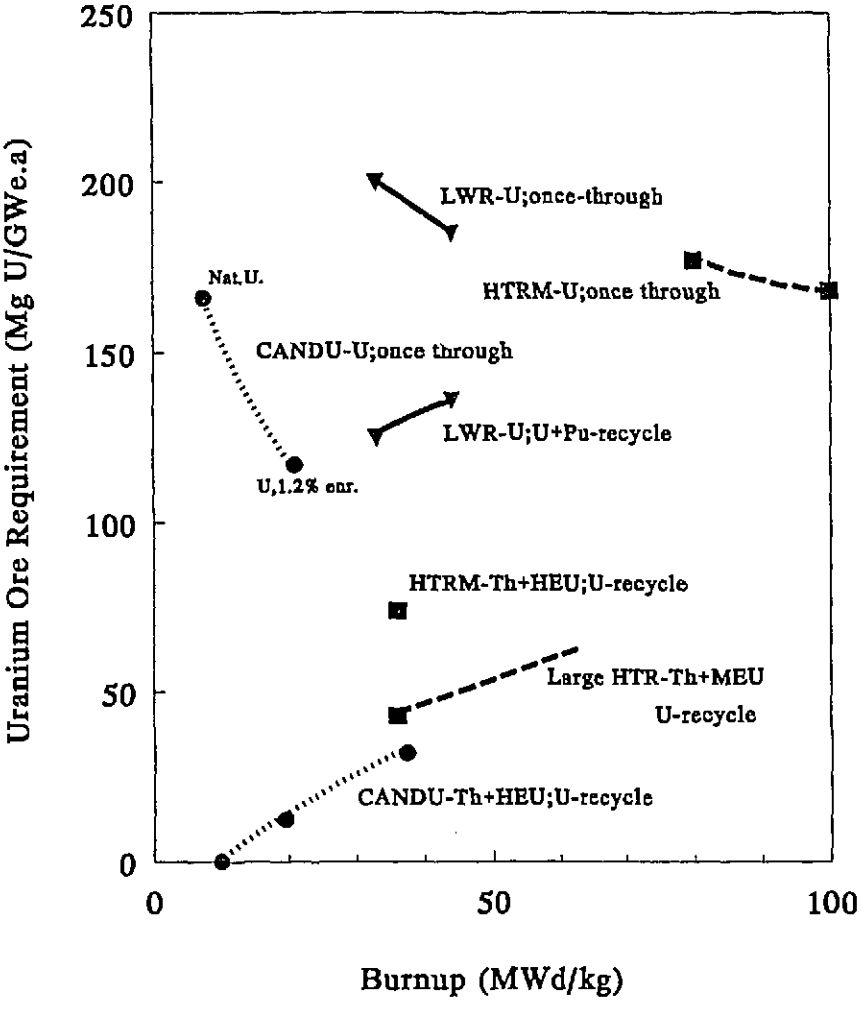


Figure 7.1: *Uranium ore requirements for annual reload*

# 8. Conclusions

*With respect to reactor physics:*
Application of thorium instead of uranium fuel will not have a serious impact on the reactor physics safety characteristics of present day reactors, although minor adaptations may be required to handle typical aspects such as protactinium holdup and decreased fraction of delayed neutrons.

*With respect to fuel cycle chemistry:*
All chemical aspects of the thorium cycle, from mining to reprocessing, have been studied in the past and technical solutions for all cycle steps have been indicated and sometimes even tested on pilot scale. The technologies have not been developed to large-scale, commercial applications. Due to the very hard gamma radiation, the handling of reprocessed uranium and thorium will require severe shielding and remote control operation for all process steps. The glass volume of wastes from the THOREX process is estimated to be considerably larger than of the wastes from a comparable PUREX process.

*With respect to actinide waste of HWR systems:*
For the once-through systems a reduction in radiotoxicity by a factor of ten (compared to the natural uranium case) may be achieved for thorium with HEU makeup, at least for the first $10^4$ years of storage time. After that period the radiotoxicities for all once-through thorium systems will be larger due to decaying $^{233}U$, abundantly present in the spent fuel.
In the case of uranium recycling a reduction of the radiotoxicity can be achieved by a factor of 100 (compared to once-through natural uranium), at least for the storage period from $10^3$ to $10^5$ years. $^{231}Pa$, originating from a (n,2n) reaction with thorium, dictates the threshold value for the long-term radiotoxicity: a common limit for all closed thorium cycles (figure 5.2). Fuel cycles, made up with "pure" $^{233}U$ or with HEU(235) show almost equal long-term radiotoxicities. Due to the high conversion ratio of the HWR considered, only small amounts of makeup fuels have to be added and further limitation of the makeup will not lead to decreased long-term radiotoxicity.

The radiotoxicity of the directly stored thorium/plutonium fuel is larger than the value for natural uranium, for the full storage period. Plutonium makeup is a prospective option to burn the plutonium.

*With respect to non-proliferation:*
The safeguardibility of most process steps in thorium-based fuel cycles is very similar to the uranium cycles.
There may be an additional concern when HEU is used as makeup material. In case of reprocessing of the thorium fuel, then uranium, highly enriched in $^{233}U$, has to be handled, comparable with plutonium in the uranium cycle. However, unlike plutonium this reprocessed uranium is strongly gamma-active. This has a bearing on physical protection and detection, but it will not affect proliferation resistance.

*The major advantages of thorium fuel cycles seem to be:a reduction of the volume of the actinide waste; a reduction of actinide radiotoxicity by a factor of about 50 to 100; and an improved utilization of uranium resources. The condition is that efficient partitioning and multiple recycling of uranium and thorium from the spent fuel would be feasible. This has still to be proven for large scale reactor operation. It will require huge efforts to develop the various fuel cycle steps, such as reprocessing, irradiation in power reactors and fuel refabrication, to the requested scale of operation.*

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