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TECHNOLOGY ASSESSMENT HTR

Part 5

Thorium fueled High Temperature gas cooled Reactors

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Preface

The present study is one in a group of studies published in the framework of a technology assessment of the High Temperature Reactor (HTR). This technology assessment was commissioned by the Dutch Ministry of Economic Affairs and executed by the Netherlands Energy Research Foundation (ECN) in cooperation with Utrecht University. The major goal of this research effort was to obtain a better perspective of the social feasibility of employing HTR technology in the long run. Both economic performance and environmental consequences play a central role in determining the social feasibility of this technology. The motive for starting this study was based on the high expectations with respect to the inherently safe characteristics of the technology in comparison with conventional designs. A broad assessment incorporating a wide range of disciplinary skills appeared desirable to address the opportunities and limitations of the HTR technology. The list of studies mentioned on the next page indicates that the scope of topics addressed is indeed very broad. The results of the different studies are presented in a separate executive summary.

The study has been implemented under the direction of C.D. Andriesse, formerly with ECN-Policy Studies, but presently with the Centre for Natural Sciences of Utrecht University. As indicated by the study director in his contribution regarding the sustainability of the HTR it is impossible to come up with one definite final conclusion. The concepts of social feasibility and environmental sustainability are not sufficiently operational and too strongly normative to allow such a result. In the framework of this study no attempt is made to solve on-going debates about the essence of sustainability in general and the social acceptability of nuclear energy in particular. The sustainability of the HTR technology must be viewed primarily in relation with the sustainability of other nuclear technologies. The studies thus provide the background material necessary for a dialogue about choices regarding the direction of future nuclear research, when energy policy makers decide to keep the nuclear option open.

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Reports published in the framework of the HTR-technology assessment

ECN-report	Author(s)	Title
ECN-C--96-050	C.D. Andriesse[1]	Technology assessment HTR Samenvatting en conclusies
ECN-C--96-041	H.M. van Rij[2]	Technology assessment HTR - Deelstudie 1 Thermodynamisch potentieel van de Hoge Temperatuur Reactor
ECN-C--96-042	R. Smit[3] J.G de Beer[3] I.C. Kok[4]	Technology assessment HTR - Deelstudie 2 Inpasbaarheid van Hoge Temperatuur Reactor in industriële processen
ECN-C--96-043	P. Lako[4]	Technology assessment HTR - Deelstudie 3 Economie van nieuwe concepten van de modulaire Hoge Temperatuur Reactor
ECN-C--96-044	A.I. van Heek[2]	Technology assessment HTR - Part 4 Power Upscaling of High Temperature Reactors
ECN-C--96-045	J.H. Bultman[2]	Technology assessment HTR - Part 5 Thorium fueled High Temperature Gas cooled Reactors
ECN-C--96-046	D.H. Dodd[2] J.F.A. van Hienen[2]	Technology assessment HTR - Part 6 The radiological risks associated with the thorium-fuelled High Temperature Reactor: a comparative risk evaluation
ECN-C--96-047	W. de Ruiter[3]	Technology assessment HTR - Deelstudie 7 Maatschappelijk draagvlak voor de introductie van de Hoge Temperatuur Reactor
ECN-C--96-048	W.C. Turkenburg[3]	Technology assessment HTR - Deelstudie 8 Kernenergie en duurzame ontwikkeling
ECN-C--96-049	C.D. Andriesse[1]	Technology assessment HTR - Deelstudie 9 Het duurzame van de Hoge Temperatuur Reactor

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CONTENTS

SUMMARY

Thorium as a fertile fuel component for the High Temperature Gascooled Reactor (HTR) instead of uranium has been reviewed. The use of thorium is generally believed to be more in agreement with philosophies of sustainability, low actinide waste production, and non-proliferation. The HTR is considered because of the inherent safety features, which excludes fuel melting due to decay heat during loss of coolant accidents.

It has been concluded that the use of thorium might be beneficial to reduce the actinide waste production. Preferably, uranium has to be recycled and extra fissile material has to be used in the form of highly-enriched uranium. This is in contradiction with the current doctrines of non-proliferation, although the safeguardability of highly-enriched uranium is generally accepted to be possible.

A self-sustaining fuel cycle might be possible in the HTR, but this could reduce the inherent safety features of the design and it could lead to higher proliferation risks and fuel cycle costs. To obtain breeding in an HTR, a larger core size is necessary, which will reduce the passive cooling capabilities. The higher proliferation risks are due to the occurence of the fissile U-233 in the fuel cycle. This can be prevented by diluting this fissile material with U-238, but this will change the thorium fuel cycle slowly into a fuel cycle containing mainly U-238 instead of Th-232.

1. INTRODUCTION

Recently, thorium fuel has gained renewed interest, because thorium fuel leads to reduced production of plutonium and other transuranic materials. This transuranic material determines the long-term radiotoxicity of the nuclear waste of uranium type of fuel.

At ECN, High Temperature Gas Cooled Reactors are being studied mainly for their passive safety features. Use of thorium fuel in the HTR may be of advantage for material utilization and waste management. The first HTR built in Germany was fueled with highly-enriched uranium with thorium as fertile material. This report presents an assessment of the use of thorium as fertile fuel in HTR. It has been based upon the material gathered during a generic study on thorium performed by ECN in 1994 [1–4] and references quoted therein. Material specific for the HTR has been gathered from reports produced by the Research Center in Jülich KFA [5–8].

This report consists of two parts: a part on general characteristics of thorium-fueled thermal reactors; and a part on the thorium-fueled HTR.

The discussed general characteristics of thorium-fueled thermal reactors are:
- Neutron cross-section data,
- Isotopic composition of the fuel,
- Thermodynamic properties,
- Radiotoxicities of the actinide waste.

Two types of fueling strategies are considered for the thorium-fueled HTR: once-through systems and fuel cycle systems with uranium recycling.

The discussed characteristics of thorium-fueled HTR are:
- Uranium ore requirements,
- Decay heat of the fuel,
- Conversion ratios of HTR systems,
- Influence of enrichment of fissile fuel startup and makeup material (this is added during fuel fabrication to obtain the desired burnup),
- Reactivity coefficients,
- Estimated fuel cycle costs.
- Proliferation issues

This report is produced for the project "Technology Assessment for the High Temperature Gas Cooled Reactor", which is aimed to answer questions about acceptability, economics, and sustainability. The questions to be answered on the bases of this literature review are:
- is the thorium-fueled HTR cleaner than once-through light water reactors with respect to long-term radiotoxicity?
- what are the consequences for inherent safety of the HTR designed for low long-term radiotoxicity of nuclear waste?
- what are the consequences for inherent safety of the HTR designed for self-sustainability?
- what are the proliferation issues related to these ways of operation?

2. CHARACTERISTICS OF THORIUM FUELED THERMAL REACTORS

2.1 Basic Data

2.1.1 Fissile Isotopes

Cross Sections

The most important fissile isotopes for the thorium cycle and the uranium cycle are U-233, U-235, Pu-239, and Pu-241. In table 2.1, the cross sections for capture and fission reactions at the thermal neutron energy of 0.0253 eV are presented. Also, the ratio of capture to fission cross section α, the number of neutrons produced per absorption η_0, the average number of neutrons produced per fission $\overline{\nu}$, and the energy produced per fission w_f are presented. Here α is defined as

$$\alpha = \frac{\sigma_\gamma^0}{\sigma_f^0}, \tag{2.1}$$

and η_0 is defined as

$$\eta_0 = \frac{\overline{\nu}\sigma_f^0}{\sigma_\gamma^0 + \sigma_f^0}, \tag{2.2}$$

where σ_f^0 and σ_γ^0 are the thermal microscopic cross sections for fission and capture, respectively.

The number of neutrons produced per absorption, η_0, is an important quantity to determine whether breeding with this fuel type is possible. Breeding is achieved when for each fissile atom destroyed, more than one fissile atom is produced. In that case, the factor η_0 needs to be higher than two; one neutron is needed to maintain the chain reaction and one is needed to produce the new fissile atom. Furthermore, some neutrons will be absorbed in non-fuel materials or will leak out of the reactor core. Therefore, the quantity $\eta_0 - 2$ is listed to show the perspective for feasibility of a breeder reactor in theory. For U-233, $\eta_0 - 2$ is the highest, and in fact U-233 can be considered as the only candidate for fuel in a thermal breeder. Still, the margins for operating a thermal breeder reactor are very small; parasitic neutron absorptions and neutron leakage should be minimized to be smaller than 0.3 at least. The value of $\eta - 2$ for a fast breeder with uranium/plutonium fuel is almost twice the value of $\eta_0 - 2$ for thermal fission of U-233.

Table 2.1 *Cross section related data for U-233, U-235, Pu-239, and Pu-241*

cross section	U-233	U-235	Pu-239	Pu-241
σ_γ^0 (b)	45.5	98.3	269.3	358.2
σ_f^0 (b)	529.1	582.6	748.1	1011.1
α	0.0861	0.1687	0.3600	0.3543
η_0	2.296	2.0751	2.115	2.1686
$\eta_0 - 2$	0.296	0.0751	0.115	0.1686
$\overline{\nu}$	2.493	2.4251	2.877	2.937
w_f (MeV)	191.3	194	200.1	202.2

Fission Product Yields

The fission product yields are dependent on fissile isotope and neutron spectrum.

In nuclear data libraries, fission product yields for thermal fission of U-233, U-235, and Pu-239 are presented. The yields are presented in figure 2.1 as a function of the number of nucleons in the nucleus as given by the burnup code ORIGEN. There are some distinct differences in the yields; some specific and important ones are presented in table 2.2.

The xenon and samarium isotopes are presented because these have the largest impact on the reactivity of the fuel. Clearly, the highest amount of these absorber fission products are produced by plutonium, and for U-233, the production rate is somewhat lower than for U-235.

Cs-137 and Sr-90 are displayed because these fission products determine the radiotoxicity of the nuclear waste upto two hundred years after discharge. The production rates for U-233 are somewhat higher than for U-235. For Pu-239, the production rate of Sr-90 is significantly lower.

Tc-99 and I-129 are displayed because these isotopes are important considering the possible dose to future populations due to leakage from repositories. The production rate of Tc-99 for U-233 is significantly lower than for U-235, whereas the production rate of I-129 for U-233 is significantly higher than for U-235.

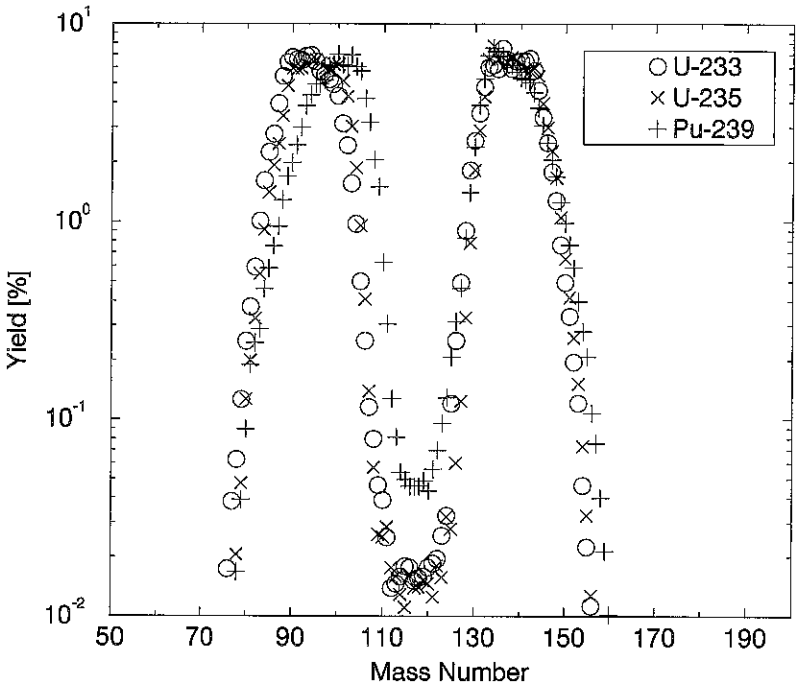

Figure 2.1 *Fission product yields as a function of mass number for U-233, U-235, and Pu-239*

Table 2.2 *Fission product yields in % per fission for some important isotopes for fission of U-233, U-235, or Pu-239*

Isotope	Fission Product Yields [%]		
	U-233	U-235	Pu-239
Xe-135	5.87	6.59	7.24
Sm-149	0.763	1.05	1.25
Sm-151	0.333	0.416	0.762
Sr-90	6.78	5.95	1.99
Cs-137	6.59	6.42	6.63
Tc-99	4.93	6.21	6.20
I-129	1.82	0.782	1.40

Delayed Neutron Fractions
Another parameter which is dependent on the fissile isotope is the delayed neutron fraction β. This fraction for thermal fission is presented in table 2.3 [9]. Clearly, the delayed neutron fractions of U-233 and of Pu-239 are smaller than that of U-235. In general, reactors containing these fissile isotopes instead of U-235 will respond faster to reactivity insertions.

Table 2.3 *Delayed neutron fractions β for thermal fission of U-233, U-235, Pu-239, and Pu-241 [9]*

	U-233	U-235	Pu-239	Pu-241
β	0.00281	0.00700	0.00227	0.00545

2.1.2 Fertile Isotopes

The main difference between the uranium and thorium cycles is of course the fertile material itself. In table 2.4, the capture cross sections σ_γ^0 at a neutron energy of 0.0253 eV and the resonance integrals I_γ for both isotopes are presented. The thorium thermal cross section is larger but the resonance integral is smaller than for U-238.

Table 2.4 *Neutron capture cross-section data for fertile isotopes [10]*

cross section	Th-232	U-238
σ_γ^0 (b)	7.4	2.7
I_γ (b)	85	277
σ_f^0 (b)	.	$4 \cdot 10^{-6}$
I_f (b)	.	$1.54 \cdot 10^{-3}$

It should be noted that the resonance integrals in table 2.4 are given for infinite dilution of the resonance isotope. In reality, the resonance integral in a lattice is important. For realistic lattices, this resonance integral can be given as a function of the surface of the fuel pin S (cm^2) and the mass M (grammes). For uranium oxide it is found that [11]:

$$I_\gamma = 4.15 + 26.6\sqrt{S/M} \qquad (b), \tag{2.3}$$

and for thorium oxide it is found that [11]:

$$I_\gamma = -3.6 + 37.6\sqrt{S/M} \qquad (b). \tag{2.4}$$

This means that for realistic values of S/M, between 0.3 and 0.6 cm^2/g, the resonance integral for thorium oxide and uranium oxide are about the same and between 18 and 25 b.

The different fertile isotopes may have a significant impact on reactivity coefficients, especially the Doppler and void coefficients.

2.2 Thorium Fuel Cycle and Uranium Fuel Cycle

In this section, the major isotope production paths of the thorium fuel cycle and the uranium fuel cycle are discussed. First, the thorium fuel is considered. Neutron capture in Th-232 produces the fissile U-233 according to:

$$\text{Th-232} + n \rightarrow \text{Th-233} \overset{T_{1/2}=22.3m}{\rightarrow} \text{Pa-233} \overset{T_{1/2}=27.0d}{\rightarrow} \text{U-233} \tag{2.5}$$

The Pa-233 causes a major hold up for the formation of U-233 due to the relatively long half life of Pa-233. During this hold up, a neutron might be absorbed and prevent formation of U-233. The rate of neutron absorption in Pa-233 will be higher when the neutron flux is higher.

The major isotope produced in the uranium cycle is Pu-239. This plutonium is formed by neutron capture in U-238 according to:

$$\text{U-238} + n \rightarrow \text{U-239} \overset{T_{1/2}=23.5m}{\rightarrow} \text{Np-239} \overset{T_{1/2}=2.35d}{\rightarrow} \text{Pu-239} \tag{2.6}$$

For the uranium cycle, the parasitic absorption in Np-239 is only of relevance for very high fluxes due to the fast decay of Np-239. In equilibrium, the Np-239 concentration is low. So, the decay of Np-239 during a reactor stop will lead to much smaller reactivity insertions than the decay of Pa-233 in the thorium fuel.

2.3 Fuel temperature coefficient of reactivity

A previous study on the thorium cycle in PWRs and HWRs [1] showed that the Doppler coefficient (which is in most cases equal to the fuel temperature coefficient of reactivity) for the thorium-based fuels does not significantly differ from the uranium-based fuels. As this coefficient is strongly dependent on enrichment, this conclusion could be untrue for fuel with a different enrichment.

2.4 Thermodynamic properties

From a thermodynamic point of view important parameters are the melting point, the thermal conductivity, and the delayed decay heat fraction. The melting point, the thermal conductivity, and the specific heat of thorium fuel are presented in the ECN report from Konings [3]. The melting point of thorium oxide is somewhat higher than for uranium oxide while the thermal conductivity and the specific heat capacity are about the same at operating temperatures. This means that with respect to these properties thorium oxide is the better. These properties might be important in loss of coolant accidents. For these accidents, the decay heat of the fuel is important. This will be discussed specifically for the HTRs.

2.5 Actinide Waste and Thorium Fuel

In this section, we will discuss the results of an ECN study on the actinide waste produced in thorium-fueled Heavy Water Reactors (THWRs) [4]. We expect that these results will be relevant for HTRs as well, because both the THWR and the HTR are systems with a very thermalized spectrum. The normalization of all presented numbers will be different for the HTR due to the much higher burnup (about 80 MWd/kg(HM) for the HTR compared to between 7 and 32 MWd/kg(HM) for the HWR).

In the ECN study performed by Wichers, once-through systems as well as fuel cycle systems with uranium recycling have been considered. For all THWRs, startup and makeup fissile material is needed. In total, 6 THWRs have been considered by Wichers. In this report, we will discuss only the limiting cases for the once-through systems and the fuel cycle systems with uranium recycling with an efficiency of 99.9%.

The results presented in this section are normalized to operation of 1 year of a 1000 MW_e plant. The normalization assumes a thermal efficiency of 33% and a load factor of 100%.

The quantity representing the toxicity of the waste is the ingestion radiotoxicity in ALI/GW_ea . One should bear in mind that this quantity does not represent the risk due to storage of the waste in underground repositories. Complete safety analyses need to be carried out to determine whether waste related to the thorium cycle or the uranium cycle lead to lower risks to future populations.

The limiting case for the once-through systems is the THWR started with highly-enriched uranium. In figure 2.2, the actinide radiotoxicity of the discharged fuel of the THWR is compared to the actinide radiotoxicity of the discharged fuel of the once-through HWR fueled with natural uranium. In figure 2.3, the actinide radiotoxicity is split up for the isotopes U-232, U-233, U-234, and Pu-238.

Upto 4,000 years after discharge, the actinide radiotoxicity for the THWR is a factor of 10 smaller than for the natural uranium fueled once-through HWR. After 40,000 years, the actinide radiotoxicity for the THWR is larger than for the natural uranium fueled once-through HWR. The actinide radiotoxicity for the first 1,000 years is mainly due to Pu-238. The radiotoxicity after 1,000 years is mainly due to U-233 in the spent fuel. Therefore, one may expect a further reduction of the actinide radiotoxicity when uranium is partitioned from the spent fuel and recycled.

The limiting case for the fuel cycle systems with uranium recycling is the THWR with U-233 makeup. In figure 2.4, the actinide radiotoxicity of the discharged fuel of the THWR with uranium recycling is compared to the actinide radiotoxicity of the discharged fuel of the once-through HWR fueled with natural uranium [2]. In figure 2.5, the actinide radiotoxicity is split up for the isotopes Pa-231, U-233, U-234, and Pu-238. Between 300 and 20,000 years after discharge, the actinide radiotoxicity for the THWR is a factor of 100 smaller than for the natural uranium fueled once-through HWR. At 100,000 years after discharge, the actinide radiotoxicity for the THWR and for the natural uranium fueled once-through HWR are almost equal. The remaining radiotoxicity of the THWR with uranium recycling is due to plutonium upto a 1000 years after discharge, and it is due to protactinium (mainly Pa-231) afterthat.

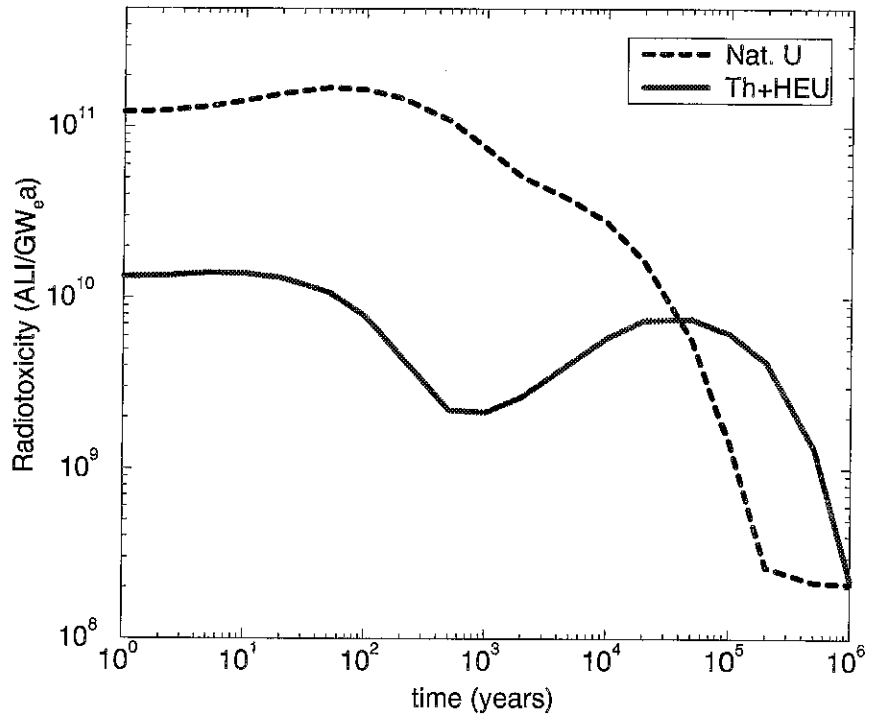

Figure 2.2 *Radiotoxicity of actinide waste in Once-Through HWRs*

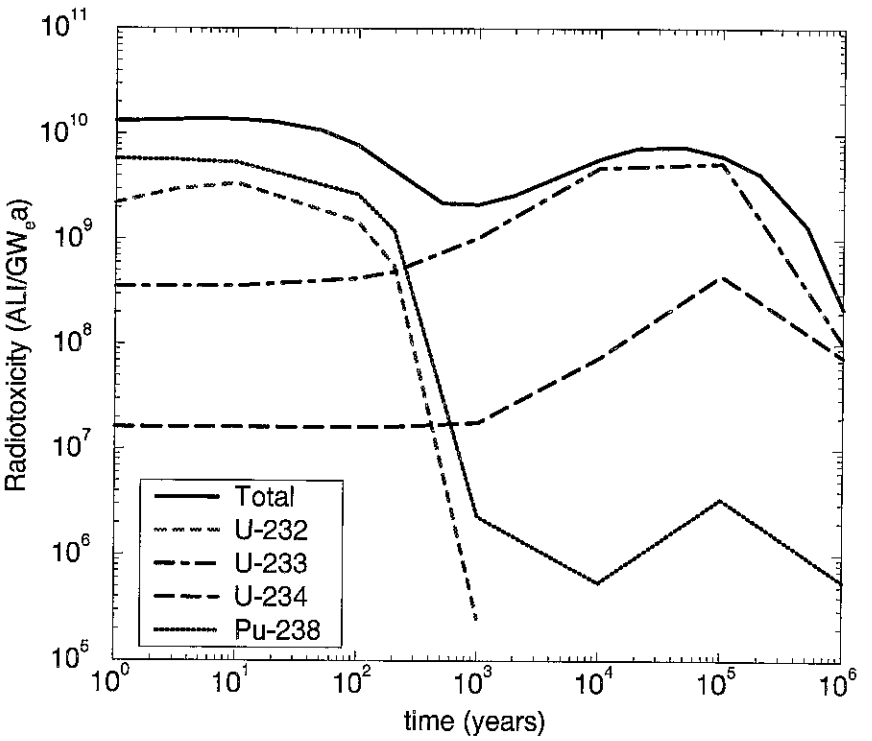

Figure 2.3 *Radiotoxicity of actinide waste in Once-Through THWR with HEU startup material, split up to some nuclides*

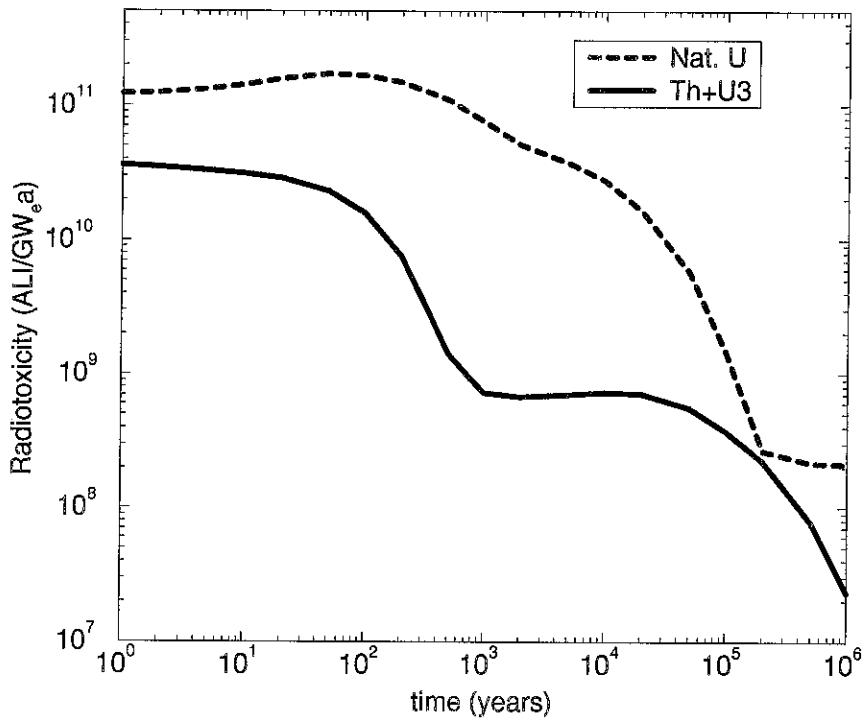

Figure 2.4 *Radiotoxicity of actinide waste in Once-Through HWR and THWR with U-233 makeup and uranium recycling*

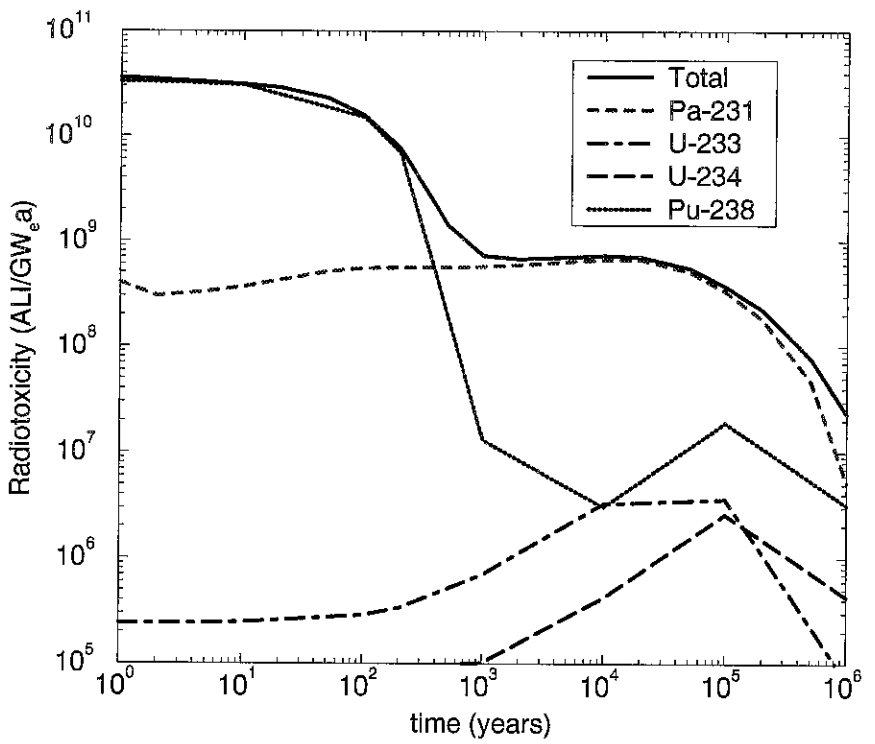

Figure 2.5 *Radiotoxicity of actinide waste in THWR with U-233 makeup and uranium recycling, split up to some nuclides*

A further reduction of the actinide radiotoxicity might be achieved by recycling plutonium and protactinium, but this is not recommended. The plutonium recycling will complicate the reprocessing of the fuel considerably, because one has to operate the THOREX process with additional plutonium extraction. Protactinium recycling will lead to a large buildup of the protactinium isotopes plus a large buildup of U-232. This U-232 is a major complicating factor due to the high energy γ-radiation emitted by its decay products [3].

In table 2.5, the masses per $GW_e a$ are presented for the Once-Through HWR on uranium, the once-through THWR with HEU startup, and the THWR with U-233 makeup and uranium recycling. Some basic decay and radiotoxicity data (half life, emitted radiation, and the Annual Limit on Intake for ingestion (ALI) [12]) of the actinide isotopes are presented as well. In interpreting these data, one should keep in mind that for the radiotoxicity of these isotopes, the decay products are as important, or sometimes even more important than the mother isotope. Also, some sums of isotope masses have been given: the mass per element, the mass of all Artificially Radiotoxic Actinides (ARA), the mass of Transuranics (TRU), and the mass of Minor Actinides (MA). The ARA actinides are all actinides made during irradiation, which were not present before this reactor started operating. So, for the once-through reactors, it refers to all actinides except for U-234, U-235, and U-238. For the THWR with recycling, it refers to all actinides except Th-232 and U-233. The Transuranics are all actinides higher than uranium. So, the transuranics start with neptunium. The Minor Actinides are all transuranics except for plutonium.

The ARA mass at discharge is highest for the once-through THWR with HEU startup material. This reflects the high conversion ratio compared to the once-through HWR with uranium. For the THWR with recycling, the ARA mass at discharge is not interesting but the ARA mass in the waste; that is when 99.9% of the uranium is recycled. The mass in the waste is $5 \cdot 10^4$ $g/GW_e a$, which is more than a factor of 10 smaller than for the once-through cases. The TRU mass shows that the buildup of higher actinides is smaller for the thorium-based fuels than for the uranium-based fuel. However, this quantity is for waste determination not relevant, although it is much refered to when the thorium-based fuels are discussed.

Another interesting quantity presented in this mass table is the mass of the fissile isotopes. This quantity is important when we will discuss the proliferation aspects of these fuel cycles, later in this report. For the once-through uranium case, the fissile plutonium content relative to the total plutonium content is 75%. This is high if one compares to fissile contents for the once-through Light Water Reactors, which are about 55%. This is due to the very low burnup for the HWR; the burnup is 7 MWd/kg(HM) compared to 45 MWd/kg(HM) for LWRs. The fissile plutonium content is not very interesting for the thorium-based fuels because of the small content of plutonium. More interesting is the fissile uranium inventory relative to the uranium content, These are 85% and 57% for the once-through and uranium recycling THWRs, respectively. The high fissile inventory for the once-through case is due to the HEU startup material. The fissile inventory for the case with uranium recycling is due to the recycling itself. During the recycling, non-fissile uranium isotopes, like U-234, build up, whereas fissile isotopes are depleted. For the thorium-based HWR this effect is much smaller than for the uranium-based HWR.

Table 2.5 *Actinide masses for once-through uranium fueled, thorium-HEU fueled HWRs and actinide masses for the self-sustained THWR with uranium cycle. Basic data of actinides is given*

nuclide	Actinide Mass g/GW$_e$a			Basic Actinide Data		
	Nat. U	Th-HEU	U-recyc	half life	decay	ALI [Bq]
Th-230	.	$1.4{\cdot}10^{1}$	$2.3{\cdot}10^{1}$	$7.54{\cdot}10^{3}y$	α	$3{\cdot}10^{5}$
Th-232	.	$1.1{\cdot}10^{8}$	$3.2{\cdot}10^{7}$	$1.41{\cdot}10^{10}$ y	α	$4{\cdot}10^{4}$
Th	.	$1.1{\cdot}10^{8}$	$3.2{\cdot}10^{7}$	.	.	.
Pa-231	.	$1.9{\cdot}10^{3}$	$1.4{\cdot}10^{3}$	$3.28{\cdot}10^{4}$ y	α	$\cdot 10^{4}$
Pa-233	.	$1.1{\cdot}10^{5}$	$3.5{\cdot}10^{4}$	27.0 d	β	$2{\cdot}10^{7}$
Pa	.	$1.1{\cdot}10^{5}$	$3.6{\cdot}10^{4}$	.	.	.
U-232	.	$3.9{\cdot}10^{2}$	$1.5{\cdot}10^{3}$	69.9 y	α	$2{\cdot}10^{5}$
U-233	.	$6.9{\cdot}10^{5}$	$4.7{\cdot}10^{5}$	$1.6{\cdot}10^{5}$ y	α	$7{\cdot}10^{5}$
U-234	$6.8{\cdot}10^{3}$	$4.9{\cdot}10^{4}$	$2.8{\cdot}10^{5}$	$2.46{\cdot}10^{5}$ y	α	$7{\cdot}10^{5}$
U-235	$3.6{\cdot}10^{5}$	$1.0{\cdot}10^{6}$	$5.4{\cdot}10^{4}$	$7.04{\cdot}10^{8}$ y	α	$7{\cdot}10^{5}$
U-236	$1.1{\cdot}10^{5}$	$1.7{\cdot}10^{5}$	$1.1{\cdot}10^{5}$	$2.34{\cdot}10^{7}$ y	α	$7{\cdot}10^{5}$
U-237	$2.2{\cdot}10^{2}$	$1.8{\cdot}10^{2}$	$9.1{\cdot}10^{1}$	6.75 d	β	$2{\cdot}10^{7}$
U-238	$1.6{\cdot}10^{8}$	$1.2{\cdot}10^{5}$	$4.7{\cdot}10^{2}$	$4.47{\cdot}10^{9}$ y	α	$8{\cdot}10^{5}$
U	$1.6{\cdot}10^{8}$	$2.1{\cdot}10^{6}$	$9.6{\cdot}10^{5}$	.	.	.
Fis. U	$3.6{\cdot}10^{5}$	$1.8{\cdot}10^{6}$	$5.6{\cdot}10^{5}$	.	.	.
Np-237	$3.5{\cdot}10^{3}$	$3.7{\cdot}10^{3}$	$6.9{\cdot}10^{3}$	$2.14{\cdot}10^{6}$ y	α	$3{\cdot}10^{4}$
Np-238	$1.6{\cdot}10^{1}$	9.5	$1.7{\cdot}10^{1}$	2.12 d	β	$2{\cdot}10^{7}$
Np-239	$1.4{\cdot}10^{4}$	$1.8{\cdot}10^{-2}$	$1.9{\cdot}10^{-1}$	.	.	.
Np	$3.5{\cdot}10^{3}$	$3.7{\cdot}10^{3}$	$6.9{\cdot}10^{3}$	.	.	.
Pu-238	$4.4{\cdot}10^{2}$	$3.7{\cdot}10^{2}$	$2.1{\cdot}10^{3}$	87.8 y	α	$4{\cdot}10^{4}$
Pu-239	$3.9{\cdot}10^{5}$	$1.9{\cdot}10^{3}$	$5.1{\cdot}10^{2}$	$2.42{\cdot}10^{4}$ y	α	$4{\cdot}10^{4}$
Pu-240	$1.4{\cdot}10^{5}$	$4.4{\cdot}10^{2}$	$1.6{\cdot}10^{2}$	$6.56{\cdot}10^{3}$ y	α	$4{\cdot}10^{4}$
Pu-241	$2.9{\cdot}10^{4}$	$1.5{\cdot}10^{2}$	$7.0{\cdot}10^{1}$	14.4 y	β	$4{\cdot}10^{4}$
Pu-242	$7.3{\cdot}10^{3}$	$2.4{\cdot}10^{1}$	$2.8{\cdot}10^{1}$	$3.73{\cdot}10^{5}$ y	α	$4{\cdot}10^{4}$
Pu	$5.7{\cdot}10^{5}$	$2.9{\cdot}10^{3}$	$2.9{\cdot}10^{3}$	.	.	.
Fis. Pu	$4.3{\cdot}10^{5}$	$2.1{\cdot}10^{3}$	$5.8{\cdot}10^{2}$	.	.	.
Am-241	$2.6{\cdot}10^{2}$	1.9	1.6	$4.32{\cdot}10^{2}$ y	α	$3{\cdot}10^{4}$
Am-242m	1.8	$4.7{\cdot}10^{-3}$	$1.2{\cdot}10^{-2}$	$1.42{\cdot}10^{2}$ y	β	$4{\cdot}10^{4}$
Am-243	$2.7{\cdot}10^{2}$	$9.2{\cdot}10^{-1}$	2.6	$7.36{\cdot}10^{3}$ y	α	$3{\cdot}10^{4}$
Am	$5.3{\cdot}10^{2}$	2.8	4.2	.	.	.
Cm-242	$5.7{\cdot}10^{1}$	$2.8{\cdot}10^{-1}$	$4.5{\cdot}10^{-1}$	162.8 d	α	$9{\cdot}10^{5}$
Cm-244	$1.7{\cdot}10^{1}$	$5.3{\cdot}10^{-2}$	$3.6{\cdot}10^{-1}$	18.1 y	α	$6{\cdot}10^{4}$
Cm	$7.4{\cdot}10^{1}$	$3.2{\cdot}10^{-1}$	$8.1{\cdot}10^{-1}$	.	.	.
ARA	$6.7{\cdot}10^{5}$	$1.0{\cdot}10^{6}$	$5.0{\cdot}10^{5}$	.	.	.
TRU	$5.7{\cdot}10^{5}$	$6.6{\cdot}10^{3}$	$1.6{\cdot}10^{4}$	.	.	.
MA	$4.1{\cdot}10^{3}$	$3.7{\cdot}10^{3}$	$6.9{\cdot}10^{3}$	.	.	.

For the latter fuel cycle, recycling of plutonium is only possible during a few cycles. Then, the plutonium vector is deteriorated too much to operate a safe reactor. After 7 cycles of recycling plutonium, the fissile content of the plutonium vector is reduced to 47% [13].

In the mass table, the phenomenon discussed in paragraph 2.2 can be seen. The Np-239 content at EOC directly after irradiation is much smaller than the Pa-233 content.

2.6 Thorium and non-proliferation

Any nuclear fuel cycle has the risk of mis-use for nuclear weapons fabrication. In this respect, the thorium fuel cycle is no different. To understand the particulars in the discussion of proliferation risks, proliferation and safeguardability are explained first.

Misuse of the nuclear energy technology can occur at three levels:
1. Diversion of nuclear materials from its peaceful use,
2. Utilization of nuclear facilities for the production of weapons material, and
3. Application of the technologies in special facilities for non-peaceful purposes.

The IAEA safeguards system is directed in the first place to nuclear materials. It is centered around the bookkeeping of nuclear materials by the operator in cooperation with the state, with the IAEA in the role of auditor. The measure to achieve good bookkeeping is Nuclear Materials Accountacy and Verification (NMA). Second, the IAEA directs its attention to the mis-use of facilities. An additional measure to safeguard facilities is Containment and Surveillance (C/S).

Proliferation aspects of the thorium cycle are in general not very different from the uranium fuel cycle with two exceptions: First, U-233 is the major fissile isotope in the fuel cycle rather than U-235 and/or Pu-239. Second, highly-enriched uranium should be used as make-up fuel to obtain low long-term radiotoxicity of the waste.

Three characteristics are imported considering the fissile materials:
1. Minimum critical mass. This is important because it determines how fast one can obtain enough material to make a nuclear weapon.
2. Spontaneous fission rate, because these can trigger a bomb to explode earlier than planned and therefore reducing its exploding force.
3. Specific α- and γ-activity. The specific α-activity determines the heat produced in the material. Heat is a difficult factor to deal with in operation of the weapon. The specific γ-activity is important because it determines the dose rate for personnel handling the material.

In the following table, some of these characteristics have been compared.

Table 2.6 *Properties of fissile materials*

Fissile Material	Critical Mass [kg]	Half Life SF [y]	Half Life α-decay [y]
U-233	16	$>3{\cdot}10^{17}$	$1.6{\cdot}10^{5}$
U-235	50	$>10^{19}$	$7.0{\cdot}10^{8}$
Pu-239	16	$>5{\cdot}10^{15}$	24110

U-233 is in respect of critical mass similar to plutonium, but in respect of spontaneous fissioning more like U-235. The specific alpha-activities ($\sim 1/T_{1/2}$) of both U-233 and plutonium are much higher than for U-235. The spontaneous fission rate and the specific activities are mainly determined by short lived polluting actinides. These are absent in the case of U-235 obtained from enrichment facilities. U-233 and Pu-239 obtained from the civil nuclear fuel cycle will always be accompanied by these polluting actinides. The amount of polluting actinides depends strongly on the irradiation history as discussed in section 2.5. The polluting actinides become more abundant when the burnup is increased. The uranium recycled for the thorium cycle is for 55% fissile. This is 47% for the plutonium recycled in the uranium cycle in LWRs [13]. Of this plutonium, a considerable

fraction is Pu-241 (half life is 14.4 years), which decays fast to the fissionable and therefore polluting Am-241.

For uranium, the polluting actinides are U-232, U-234, and U-236. For plutonium, the polluting actinides are Pu-238, Pu-240, Pu-242, and Am-241 produced by decay of Pu-241. The abundance of U-232 and Pu-238 will be approximately the same, because they are produced by (n,2n)- reactions on the fertile material, which cross sections are similar for Th-232 and U-238. The abundance of U-234 and U-236 will be lower for the thorium fuel cycle, due to the much lower chance of neutron capture relative to fission for U-233 compared to Pu-239, as discussed in section 2.1. We will compare the characteristics of the polluting actinides of uranium and plutonium.

1. Spontaneous fission from polluting actinides:
   The uranium polluting actinides have a much lower likelihood of spontaneous fissioning decay than the plutonium polluting actinides.
2. α-activity:
   The α-activity for plutonium is mainly determined by the decay product of Pu-241, Am-241 with half life of 433 years. Uranium does not have a similar polluting actinide.
3. γ-activity:
   The γ-activity is due to the decay products of U-232. Plutonium has a very small fraction of Pu-236 as a polluting actinide decaying to U-232, but this fraction is much smaller than the fraction of U-232 relative to U-233.

These polluting actinides will make nuclear weapon production much more difficult than if pure isotopes were used, but not impossible. In particular, the hard γ's emitted by U-232 will make usage of U-233 more difficult and it will present a major barrier to theft.

Another aspect is the availability of this fissile material in the fuel cycle. For the uranium cycle, plutonium will be separated in reprocessing. At that point, it is usable for weapons fabrication, although it is not a very favorable material to use in nuclear weapons because of the decay heat and the spontaneous fissioning of the polluting plutonium isotopes. For the thorium cycle, highly-enriched uranium make-up material will be available during fuel fabrication unless one opts for the once-through operation on thorium with medium-enriched uranium make-up fuel. The safeguarding of highly-enriched uranium is technically straightforward, but it is considered to be the nuclear material with the highest proliferation risk.

The last aspect in proliferation risk is the type of nuclear weapon which can be produced. A plutonium weapon is of an implosion-type, whereas a uranium weapon can also be of a gun-type. The latter is easier to make.

2.7 Conclusions

In this chapter, we have studied the possible implications of changing the fertile material for a typical thermal reactor from U-238 to Th-232. First, the important physical quantities are compared for the fissile isotopes, the fertile isotopes and the remaining fissionable isotopes. The most important difference is caused by the much lower delayed neutron fraction for U-233. This leads to faster power transients upon reactivity insertions for thorium fuel. Another effect is that the hold up of the production of U-233 is much larger than the hold up of the production of Pu-239. This will lead to larger reactivity insertions during reactor stops for

thorium fuel. Finally, the conversion ratio is higher for fuels based on thorium, which makes thorium the most suitable candidate fuel for thermal breeders.

The actinide radiotoxicities of thorium-based thermal reactors are considerably smaller than for the once-through uranium fueled systems, if uranium is recycled. Between 300 and 40,000 years after discharge, the actinide radiotoxicity of the waste is about a factor of 100 smaller than for once-through uranium systems.

A once-through thorium cycle will reduce radiotoxicity of the waste between 1000 to 40,000 years. For longer decay times, the radiotoxicity becomes higher than for uranium cycles due to U-233.

The results on waste radiotoxicity are for thorium fueled heavy water reactors. We assume that the results for high temperature reactors will be similar. This needs to be verified. The reader should also keep in mind that radiotoxicity by itself is not a good indication of the hazard of the waste in waste repositories.

Thermal reactors based on thorium fuel with uranium recycling can be operated without a large deterioration of the uranium vector. This is in contrast to uranium-based thermal reactors with plutonium recycling, which leads to a strong deterioration of the plutonium vector. This increases the need for extra fissile enrichment to a level which leads to reduced safety parameters. There is reason to believe that thorium-based thermal reactors with multiple uranium recycling will not lead to a high fissile enrichment and that safety parameters will not be reduced. This should be investigated for the THTR and the THWR.

Our conclusion on proliferation issues is that the uranium/plutonium cycle leads to less proliferation risk (according to current doctrines of proliferation issues) than the thorium/uranium cycle when highly-enriched uranium is being used. When this is not the case, the proliferation risks for the two cycles are very similar.

3. THORIUM FUEL IN HTRS

3.1 Fuel Cycles

Six thorium-based fuel cycles have been considered in the HTR [6]:

1. Once-through thorium fuel system with 93% enriched uranium (U-235),
2. Once-through thorium fuel system with 20% enriched uranium (U-235),
3. Thorium fuel system with 93% enriched uranium (U-235) as fuel makeup and uranium recycling,
4. Thorium fuel system with 20% enriched uranium (U-235) as fuel makeup and uranium recycling less than 15% enriched,
5. Thorium-fueled prebreeder system with 93% enriched uranium (U-235) and separate thorium fuel elements to produce U-233 for start-up of the breeder reactor,
6. Thorium-fueled breeder system with U-233 start-up material from prebreeder. This is the self-sustainable thorium cycle, the THTR makes new fissile material for the next cycle in the same reactor; only thorium make-up material needs to be added to the new fuel.

As comparison, the modular 200 MW_e HTR with 20% enriched uranium (MEU; U-235) is considered. Also, the 93% enriched uranium case without recycling is considered. Data as presented by Teuchert are given in table 3.1 [6]. This table will be discussed in the coming paragraphs.

Table 3.1 *Characteristics of THTR and HTR normalized to 1000 MW_e , enrichment of tailings is 0.25%, and reprocessing losses of 1%*

Reactor	MODUL	HTR			THTR		PreB.	B.
number			1	2	3	4	5	6
Fertile Mat.	U-238	U-238	Th-232	Th-232	Th-232	Th-232	Th-232	Th-232
Fis. Makeup	MEU	HEU	HEU	MEU	HEU	MEU	HEU	U-233
Recycling	no	no	no	no	yes	yes	yes	yes
burnup MWd/kg	80	100	100	100	37	53	17	20
CR	0.49	0.58	0.59	0.58	0.84	0.66	0.74	1.0
U-ore Mg/a	169	152	135	143	40	97	196	.
Fiss. Inv. kg/GW_e	1210	1020	920	710	1270	600	2420	3200
U-235 use kg/a	683	613	536	570	155	355	761	.
U-233+U235 loss kg/a	90	99	164	138	6	29	10	13
Pu-239+Pu-241 loss kg/a	66	67	1	15	2	20	8	.
relative fuel cycle costs	1	0.8	0.7	0.8	0.8	0.9	1.1	0.9

3.2 Conversion Ratio and Uranium Ore Requirements

Conversion ratio and uranium ore requirements are strongly related; a higher conversion ratio leads to a more effective use of fissile material and therefore to lower ore requirements. A higher conversion ratio can be obtained in two ways:

- By reducing neutron leakage for instance by increasing the core size.
  The neutron which would otherwize be lost can now be used to make new fissile material from the fertile material.
- By reducing the fuel enrichment.
  In this way, the chance of neutron absorption in fertile material is increased. Of course, the drawback of this method is the lower burnup that can be reached. The fuel is faster depleted and has to be reprocessed at an earlier stage. A way to overcome this is by heterogeneous core design as is done for fast breeder reactors. Then the core consists of driver fuel elements with a high fissile content to obtain a high burnup and breeding fuel elements containing only thorium at startup and obtaining a very low burnup at the end of cycle.

In table 3.1, some of these phenomena are demonstrated. The conversion ratio of the once-through THTR systems is in the order of 0.6, whereas the conversion ratio of the THTR with uranium recycling is considerably higher at a much lower burnup.

The ore requirements are much lower when uranium is recycled, of course due to the use of most fissile material. The uranium requirements when uranium is not recycled are only somewhat less for thorium-fueled HTRs than for uranium fueled HTRs.

3.3 Fissile Materials Balance

The fissile materials balance shows that the HTR is very efficient in its use of the fissile isotopes. The percentage of fissile material discharged relative to the fissile material loaded is 25% for the modular HTR, whereas this is 33% for LWRs.

The amount of fissile plutonium discharged reduces considerable in going from uranium-based fuel to thorium-based fuel. When thorium and HEU makeup fuel is used the discharged fissile plutonium is a factor 60 lower, wheras it is a factor 4 lower for the thorium and MEU makeup fuel.

On the other hand, the amount of fissile uranium discharged is much higher for the once-through thorium case than for the once-through uranium case. The total amount of discharged fissile material is about the same for the once-through systems.

The amount of fissile uranium lost decreases considerable when uranium is recycled. The lowest amount of fissile material lost is when uranium is recycled and HEU topping and makeup is used. Then, the percentage of fissile material lost, relative to the amount of fissile material loaded, is 5%. The amount of fissile material lost is larger for the breeder than for the case with uranium recycling and HEU topping. This is due to the very large inventory and the very low burnup for the breeder reactor.

3.4 Fuel Cycle Costs

Fuel cycle costs are difficult to determine and should be considered with some scepticism. A few statements can be made:

1. Fuel cycle costs increase for modular design compared to large reactors due to economy of size,
2. Fuel cycle costs increase when highly-enriched uranium is used because the enrichment costs increase with enrichment percentage.
3. Fuel cycle costs increase when uranium is recycled due to the costs of reprocessing and fuel refabrication,
4. Fuel cycle costs increase strongly when breeding is pursued, because of the need for reprocessing and the lower burnup.

3.5 Safety and Thorium Fueled HTRs

Already two safety issues have been discussed: the lower delayed neutron fraction for thorium-based fuels compared to uranium-based fuels, and the comparable feedback coefficients for thorium and uranium-based fuels. In literature, some reactivity coefficients on uranium fueled HTRs and thorium-fueled HTRs can be found. The total core temperature coefficient of reactivity of uranium fueled HTRs is in the order of $-5 \cdot 10^{-5} \delta k/K$ [14]. For the breeder THTR, the total core temperature coefficient of reactivity is $-4 \cdot 10^{-5} \delta k/K$ [15]. For the THTRs with or without recycling but at lower conversion ratios, the total core temperature coefficient of reactivity is in the order of $-2 \cdot 10^{-5} \delta k/K$ [16]. Although the latter value is considerably smaller in magnitude, the overall conclusion remains that no safety issues considering the reactivity feedback result in going from uranium-based fuels to thorium-based fuels.

A third issue is the decay heat. Rütten shows that the decay heat curve of thorium reduces more slowly than the decay heat curve of uranium-based fuels as is shown in figure 3.1 [15]. This is due to the larger half life of Pa-233 as compared to Np-239. The decay heat is important for the determination of the fuel temperature in a loss of coolant accident. For time periods after reactor shutdown important for these accidents, the decay heat is more than 30% higher for thorium-based fuels than for uranium-based fuels. This will lead to higher maximum fuel temperatures during a loss of coolant accident assuming the same geometrical reactor layout and assuming that the thermal characteristics of thorium and uranium fuel are the same. Therefore, the power density of thorium-based fuels should be considerably lower to maintain the same safety features. It remains to be determined how much lower the power density should be to maintain the same maximum fuel temperature.

Finally, the self-sustaining thorium cycle is considered. To obtain a conversion ratio equal to one, neutron loss has to be minimized. The HTR has some definite advantage over other thermal reactors because no construction materials are inside the core. Therefore, only one means remains to minimize the neutron loss, and that is minimization of the neutron leakage. This leads to a large core size, which reduces the inherent safety features of this design, because the passive decay heat removal is reduced due to the core enlargement.

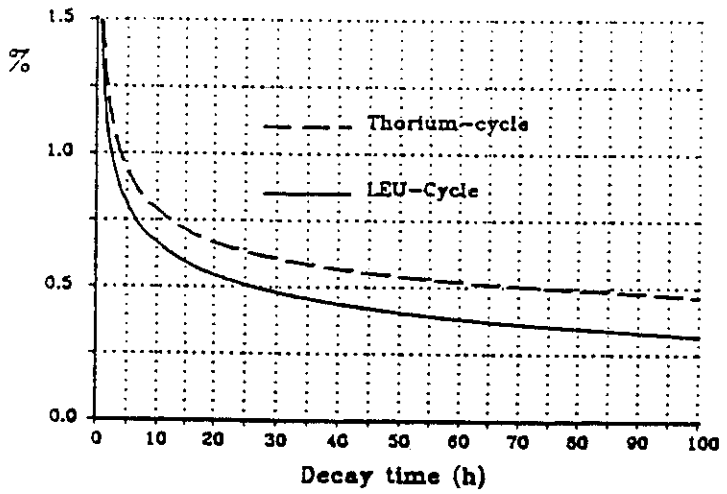

Figure 3.1 *Relative decay heat: Comparison between uranium and thorium-based fuels*

4. DISCUSSION

Several objectives are pursued in the project (Technology Assessment for the HTR) for which this report is prepared:

- Safety in reactor operation ; this is supposed to be the main attraction of the HTR,
- Los resource utilization; this is necessary for the HTR to be an energy-source for the long-term,
- Low long-term nuclear waste production; a new nuclear technology is acceptable when this is achieved,
- Non-proliferation; a new technology is only acceptable when the proliferation risk is not enhanced,
- Economics; costs of a new technology should be reasonable.

From the discussed details it should be clear that these five objectives can not be optimized at the same time. Therefore, we made the following relational table on the basis of the previously discussed results. On the left hand is the quantity to be optimized, on the right hand are the impacts on the other quantities.

Table 4.1 *Relational table for optimizing HTRs*

Optimized quantity	Influence on quantities				
	safety	utilization	waste	non-proliferation	economics
safety	.	-	0	0	-
utilization	-	.	+	-	-
waste	-	+	.	-	-
non-proliferation	0	-	-	.	+
economics	-	-	-	+	.

Optimizing on safety will lead to a small modular reactor. This will lead to a higher resource utilization because more neutrons leak away, which could have been used for the production of new fissile material. The burnup will decrease due to this phenomenon or a higher enrichment may be used. The influence on long-lived waste is negligible when a higher enrichment is used. Modularity is not in conflict with non-proliferation but it leads to higher costs unless the modularisation itself can lead to lower costs per unit by simplification.

Optimization on resource utilization leads to a large core size to obtain a high conversion ratio. This will reduce safety. The amount of waste is minimized due to the re-use of fissile material. Reprocessing and refabrication will increase the proliferation risks and reduce the economics.

The minimization of low long-lived waste leads to the same results as for optimization on resource utilization.

Minimization of proliferation risks, defined according to current doctrines on proliferation, will exclude the usage of highly-enriched uranium for civil energy production. In some countries, also reprocessing is rejected because of assumed proliferation risks. Both doctrines will lead to low resource utilization and high production of long-lived waste.

Minimization of costs leads to a large core without recycling and the use of highly-enriched uranium. Therefore, the waste production is high and the resource utilization is low.

This relational table is important to make a decision in which way thorium could be used as a fertile fuel in HTR. At this point in time, the most important issues for nuclear power production are safety and long-lived waste production. Safety for the HTR asks for modularity. Low long-lived waste production asks for recycling of the major actinides. We have reason to assume that multiple plutonium recycling may lead to reduced safety due to the much higher fissile enrichment requirement. On the other hand, we also have reason to assume that this is not the case for multiple uranium recycled for thorium-based thermal reactors. Therefore, thorium-fueled modular HTRs with recycling of uranium and addition of highly-enriched uranium should be operated to both have the increased safety behaviour of the modular HTR and the low long-lived waste production. However, this could have a negative impact on the proliferation risks as defined by current doctrines.

5. CONCLUSIONS

Thorium has two attractive features: the perspective to achieve a self-sustaining fuel cycle and the low production of higher actinides. However, the lower production of higher actinides in itself is not important considering the radiotoxicity of the waste and the hazard of the waste in repositories. For once-through operation of the thorium, the radiotoxicity of the waste is lower by a factor of 10 between 1000 and 40,000 years. When uranium is recycled, the radiotoxicity of the waste is lower by a factor of 100. Recycling will be necessary to achieve a self-sustaining cycle without externally supplied fissile material. A self-sustaining fuel cycle may only be possible when neutron losses are minimized which leads to a large core size and high fuel cycle costs. A large core is in conflict with inherent safety features of the modular HTR. Modularity is pursued to achieve a lower temperature rise during loss of coolant accidents.

Low long-term radiotoxicity does not depend on low resource utilization. A low long-term radiotoxicity of waste can also be achieved for non-breeders with additional fissile uranium make-up. Use of HEU is in conflict with current doctrines of non-proliferation. Use of MEU in HTRs with uranium recycling will not lead to lower long-term radiotoxicity.

Safety, low long-term nuclear waste production, non-proliferation, resource utilization, and economics can not be optimized at the same time. Safety on the one hand and economics and resource utilization on the other hand are conflicting objectives. Also, low long-term nuclear waste production and non-proliferation are conflicting. Sustainability and economics are conflicting objectives as well.

To obtain increased safety behaviour and reduced long-lived waste production, the thorium-fueled modular HTR with recycling of uranium should be operated because multiple recycling of uranium is possible without increased enrichment. This leads to reduced safety parameters when plutonium is multiple recycled.

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