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EVALUATION OF THORIUM BASED NUCLEAR FUEL

Actinide Waste

V.A. WICHERS

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Abstract

Use of thorium based fuel has recently been proposed as a possible way to reduce the amount of actinide waste from nuclear power. To examine this possibility, burnup calculations were done of five once-through Thorium Heavy Water Reactor (THWR) systems, and three THWR systems with uranium recycle. The natural uranium once-through system was adopted as reference. The studied THWR fuel systems differed in the choice of fissile makeup fuel and exit burnup. The HWR was chosen because of its good neutron economy. Actinide waste production (in mass per GW_ea) and radiotoxicity (in ALI per GW_ea) for storage times up to 10^6 a were calculated for each system. The study shows that the THWR system with uranium recycle and High Enriched Uranium (U-235) makeup fuel performed best, producing both the lowest amount of plutonium and actinide waste with the lowest radiotoxicity. Relative to the natural uranium in HWR once-through system, radiotoxicity is reduced by a factor varying between 2 and 50 for the full range of storage times up to 10^6 a.

Keyword 1: Thorium fuel cycles

Keyword 2: Actinide waste

Keyword 3: Heavy Water Reactor

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1. INTRODUCTION

Reduction of long-lived actinide waste from nuclear power has become a major topic of study. At first sight, the thorium fuel cycle appears to offer an easy solution, since the production of plutonium and higher actinides is low when U-238 is absent. This report aims to clarify the potential of the thorium fuel cycle for reduced actinide waste production. To that end fuel depletion calculations for various thorium fuel cycles are done.

The definition of actinide waste is controversial. In this report, those actinides for which in a given fuel cycle no use is foreseen are considered as waste. This view is consistent with the definition of radioactive waste given in [1]: "Any material that contains or is contaminated with radionuclides at concentrations or radioactivity levels greater than the 'exempt quantities' established by the regulatory body and *for which no use is foreseen*". What is considered as waste therefore depends on the system under study.

This study is focused on thorium-thermal reactor systems; thorium-FBR systems are not considered. As makeup fuel, fissile plutonium and U-235 are studied. The use of plutonium as fissile makeup fuel can be motivated because it is available, but also because plutonium consumption can be a goal in itself. Motivations for this goal are that plutonium is an anthropogenic, long-lived radiotoxic substance and that continued stockpiling of for example LWR plutonium, a material which can be used directly as nuclear material in nuclear weapons, may be viewed as presenting a proliferation risk. However, from the point of view of reduced actinide production in thorium systems, the choice of plutonium as makeup fuel may be doubtful. The goal of thorium cycles in general is to reduce actinide production; for this aspect, the HWR once-through natural uranium system is adopted as reference. The goal of Pu-topped thorium cycles in particular is to reduce the overall "world" actinide inventory; for this aspect, the PWR uranium-plutonium MOX cycle is adopted as reference.

The evaluation is based on burnup calculations of various thorium-based fuels in Heavy Water Reactors (HWRs). Calculations have been done for homogeneously distributed fuels. HWRs were chosen because of their good neutron economy. This property allows high conversion ratios, and thus leads to a better utilization of the thorium itself, and to a lower amount of possible additional fissile fuels. The model used in the calculations is based on the Canadian Deuterium Uranium (CANDU) HWR. The burnup calculations are done with the SAS6 code sequence [2]. Once-through fuel modes, as well as systems with uranium recycle, are examined.

Although thorium-FBR systems are outside the scope of this study, they may have some promises. Thorium fuel may have a lower fast sodium void effect in fast reactors than uranium-plutonium fuel due to two causes. First, η (the number of neutrons produced per neutron absorbed in the fuel) of U-233 increases at a higher energy (≈ 5 MeV) than η of plutonium and U-235 (≈ 1 MeV). Second, the fission cross section of Th-232 is lower and has a somewhat higher threshold energy than the fission cross section of U-238 [3].

The burnup computations provide the masses of nuclides in discharged fuel. However, masses do not give an impression of risk. Therefore, also the corresponding radiotoxicity of actinides and their daughters in the waste is presented. The radiotoxicity is expressed in units of Annual Limits on Intake (ALI) as a function of time up to 10^6 a after discharge. All calculated results are normalized to one GW_e a of generated electricity.

2. SELECTED FUEL CYCLES

Seven Thorium in HWR (THWR) fuel cycles were studied: four once-through fuel systems, and three systems with uranium recycle (see Table 1). The choice is motivated in the following sections. The HWR once-through natural uranium system is included as reference. The adopted fuel terminology is explained in Fig. 1; a topping isotope is a fissile isotope in the makeup fuel.

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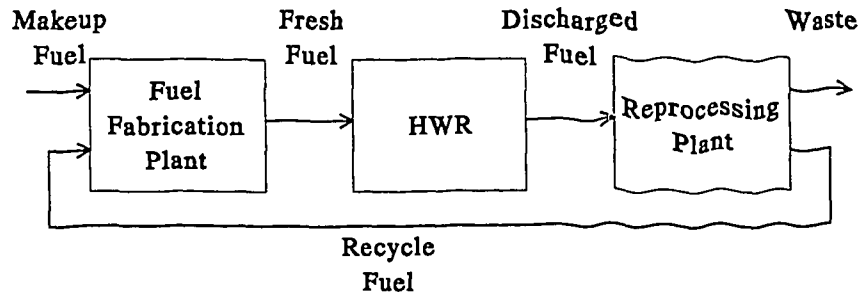


Fig. 1. Terminology of fuels occurring in thorium fuel cycles.

Only recycling of uranium is studied. This was thought sufficient to reveal the principal behaviour of closed thorium fuel cycles with respect to long-lived actinide waste. Recycling of other actinides, if desired, could be analyzed in a follow-up study.

Table 1. Overview of studied HWR Fuel Cycles.

	Fuel Cycle	Fuel	Topping Isotope
1	HWR once-through natural uranium system	UO ₂	—
2	Pu-THWR once-through system (1)	Th,PuO ₂ (Pu-fissile 1.66 wt% HM)	Pu-fissile
3	Pu-THWR once-through system (2)	Th,PuO ₂ (Pu-fissile 2.19 wt% HM)	Pu-fissile
4	HEU-THWR once-through system	Th,UO ₂ (U-235 1.86 wt% HM)	U-235 as HEU
5	MEU-THWR once-through system	Th,UO ₂ (U-235 2.00 wt% HM)	U-235 as MEU
6	HEU-THWR system with uranium recycling	Th,UO ₂ (U-fissile 2.12 wt% HM)	U-235 as HEU
7	MEU-THWR system with uranium recycling	Th,UO ₂ (U-fissile 2.16 wt% HM)	U-235 as HEU
8	Pu-THWR system with uranium recycling	Th,U,PuO ₂ (Fissile fraction 2.26 wt% HM)	Pu-fissile

2.1 Fissile Makeup Material

All examined thorium fuel cycles require externally supplied fissile fuel. This is obvious for once-through fuel modes, since natural thorium does not contain fissile isotopes. For the closed systems studied here, it appears that the conversion ratio is less than unity, even at equilibrium. However, no optimization was attempted.

Concerning the topping isotope, the possible choices are U-235, U-233 or fissile plutonium. Uranium mixtures of two enrichments in U-235 are considered: HEU-235 (U-235 fraction > 90%) and MEU-235.

MEU is defined by the following limit on the enrichment of *fresh* fuel [5]:

$$\frac{\text{Weight of U-233} + 0.6 \times \text{Weight of U-235}}{\text{Weight of total uranium}} \leq 0.12. \quad (1)$$

This implies for fuel without U-233 that the U-235 fraction is less than 20%. For once-through thorium systems, all uranium in fresh fuel is makeup fuel, and eq. (1) therefore also holds for the makeup fuel. For closed fuel cycles, the fresh fuel also contains recycled uranium, and eq. (1) cannot hold for the makeup fuel in an equilibrium thorium fuel cycle (see Section 2.3.)

The fissile isotope U-235 is present in natural uranium. The fissile isotope U-233 is not present in nature and can only be obtained through reprocessing of irradiated thorium fuel. This material is not available in commercial quantities. Therefore, U-233 is not a viable source of makeup fuel. Of the uranium makeup fuels, only HEU-235 and MEU-235 were considered.

Separated plutonium is available from stocks of reprocessed LWR fuel, and possibly also from military stocks of plutonium. The isotopic composition of the plutonium makeup fuel used in the present study is listed in Table 2. This plutonium corresponds to PWR fuel with a discharge burnup of 45 MWd/kg. Two Pu-THWR once-through systems, with different enrichments and consequently different exit burnups, were analysed in order to study the effect of burnup on produced actinide waste. A property of interest is the plutonium consumption potential of the Pu-THWR fuel cycles. This potential is compared with that of MOX fuelled PWRs [12]. Table 2 also lists the isotopic composition of the plutonium contained in this MOX fuel.

Table 2. *Isotopic composition in wt% of plutonium in HWR thorium-based fuel (this work) and plutonium in PWR uranium-plutonium MOX fuel [12].*

Isotope	Fraction (wt% Pu) of makeup fuel	Fraction (wt% Pu) [12]
Pu-238	2.63	1.8
Pu-239	53.27	59.0
Pu-240	24.40	23.0
Pu-241	13.09	12.2
Pu-242	6.61	4.0

2.2 Once-Through Fuel Systems

Considerable attention is paid to once-through fuel systems. Once-through thorium fuel systems are claimed to produce lower quantities of actinides than current LWR cycles. Once-through fuel systems are definitely technologically simpler, and in most cases more economical at present, than closed fuel cycles. Thorium once-through systems derive much of their attractiveness from this promising combination of qualities. Detailed characteristics of the once-through systems will be provided in Section 3.4, Table 5 and in Section 4.1, Table 7.

2.3 Closed Thorium HWR Fuel Cycles

Three THWR fuel cycles with U recycling have been examined with different choices of the makeup fuel, respectively HEU-235, MEU-235 and plutonium. The closed fuel cycles are analysed for the equilibrium situation. Under this condition, all reloads are identical. Uranium is reprocessed after sufficiently long cooling to allow decay of Pa-233, and is recycled minus an out-of-core loss of 0.1%.

In the MEU-Th cycle with uranium recycle, the makeup fuel is still HEU-235 (except for the first cycle). The reason for this is that during residence in the reactor, U-238 is far less depleted than U-235. Thus the makeup fuel must be highly enriched uranium. Otherwise, the U-238 fraction of recycle uranium would rapidly increase with recycle generation number, so that, with time, the cycle would shift (in about 10 a) from a thorium cycle to a uranium-plutonium cycle.

Detailed characteristics of the closed fuel cycles will be provided in Section 3.4, Table 5 and in Section 4.1, Table 8. The possibility of self sustaining thorium fuel cycles [4] is not studied here.

3. CALCULATION METHODS

3.1 Geometrical Model

The two-dimensional cell model used for the neutron spectrum calculations is shown in Fig. 2. It comprises one half of a CANDU fuel bundle of 37 pins and moderator.

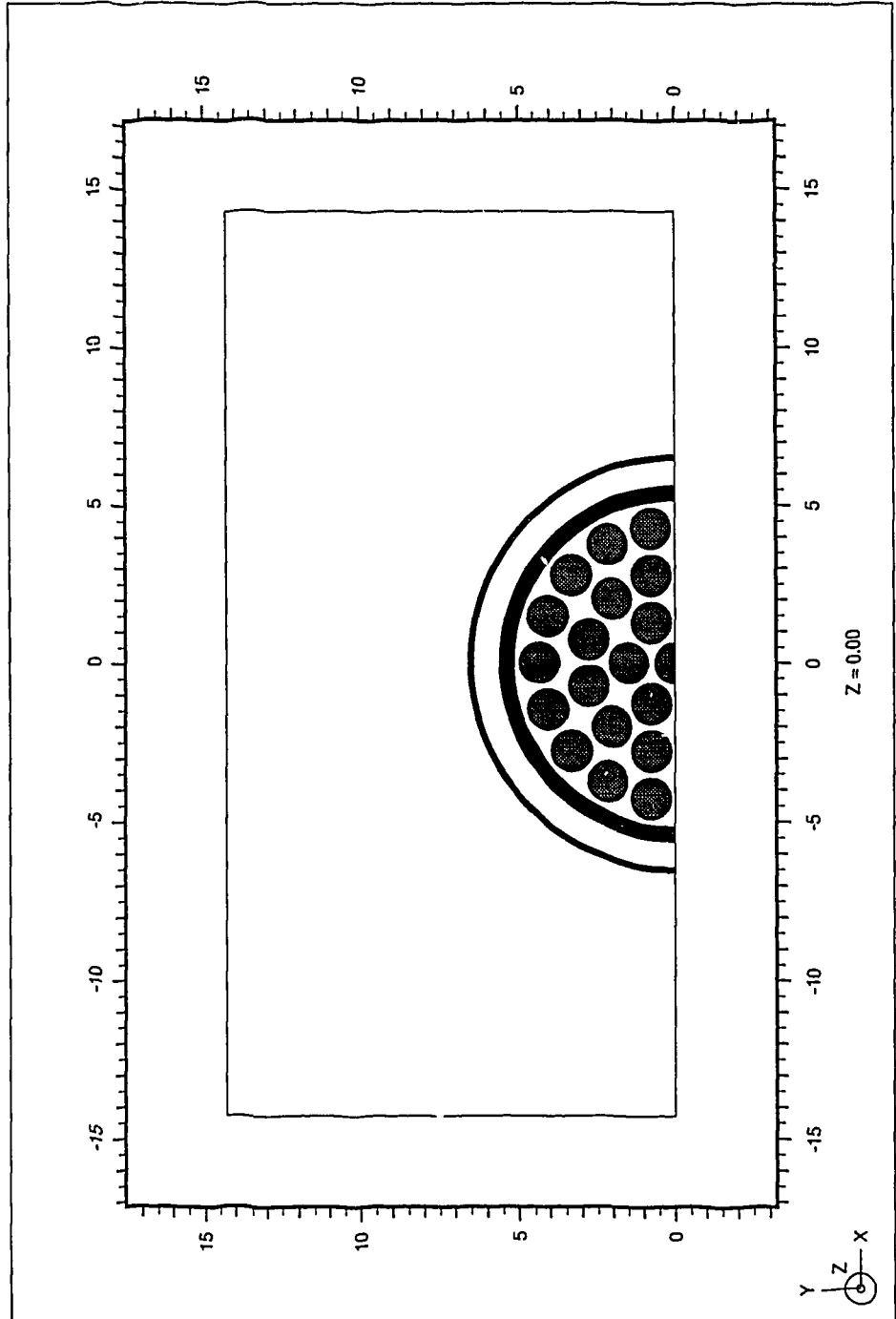


Fig. 2. Geometry of the standard HWR fuel bundle. Only one of two identical halves is shown. From centre to outside: central pin, second, third and fourth annuli, with high pressure coolant; pressure tube; He filled annulus; calandria tube; low pressure moderator.

The parameters selected for the model are largely based on a CANDU HWR of the type located at Darlington, Canada. However, the specific power is 26.3 W/g HM, in accordance with the value adopted in INFCE [5], but differing from the value of 22.8 W/g HM adopted in [6]. This is done to facilitate comparison of our fuel cycle data with those of the INFCE studies [5]. The fuel bundle parameters are summarized in Table 3.

Table 3. *Parameters of the standard fuel bundle.*

Parameter	Value
Number of fuel pins per bundle	37
Number of fuel bundles per channel	12
Diameter fuel pellet (cm)	1.21
Outer diameter cladding (cm)	1.31
Inner diameter pressure tube (cm)	10.338
Outer diameter pressure tube (cm)	11.242
Inner diameter calandria tube (cm)	12.8956
Outer diameter calandria tube (cm)	13.2016
Lattice pitch (cm)	28.6
Channel length (cm)	600
Effective fuel temperature (K)	960
Effective clad temperature (K)	563
Effective coolant temperature (K)	563
Effective moderator temperature (K)	344
Specific Power (W g ⁻¹ HM; Core average)	26.3

3.2 Burnup Sequence Method

The fuel depletion calculations were done with the SAS6 burnup driver code [2], which uses the spectral code WIMS-D (version 4) and the point-depletion code ORIGEN-S. Cross section updating and inclusion in the spectrum calculations differed for the nuclides listed in Table 4 and the remaining nuclides. The former were identified in preliminary runs as the major neutron absorbers. For these nuclides, each burnup step started with updating the cross section working library with self-shielded cross sections corresponding to the current nuclide densities as computed by ORIGEN-S. Self-shielded cross sections were calculated by the SCALE codes BONAMI and NITAWL, respectively for resonance self-shielding in the unresolved region based on the Bondarenko method, and in the resolved region based on the Nordheim method. The remaining nuclides are approximately accounted for in the neutron spectrum calculations through equivalent pseudo nuclides.

Table 4. Nuclides explicitly included in the calculations.

Actinides		Fission Products		
Th-232	Pu-238	Kr-83	Xe-135	Sm-147
Pa-231	Pu-239	Zr-93	Cs-133	Sm-149
Pa-233	Pu-240	Mo-95	La-139	Sm-150
U-232	Pu-241	Tc-99	Pr-141	Sm-151
U-233	Pu-242	Ru-101	Nd-143	Sm-152
U-234	Am-241	Rh-103	Nd-144	Eu-153
U-235	Am-242m	Rh-105	Nd-145	Eu-154
U-236	Cm-242	Pd-105	Nd-146	Eu-155
U-238	Cm-243	Pd-108	Nd-147	
Np-237	Cm-244	Ag-109	Pm-148	
Np-239		Xe-131	Pm-148m	

3.3 Nuclear Data

The cross section data applied in the neutron spectrum calculations are primarily based on the JEF2.2 evaluated file. The cross section data used in the burnup calculations are based on the JEF2.2 file if available, and on the EAF-3 file for the other nuclides and for all isomer branching ratios. Cross sections from EAF-3 are applied for nuclides at infinite dilution. The approach of using new JEF2.2 based, ORIGEN-S working libraries in order to apply a consistent set of cross section data for all nuclides was described in [7].

3.4 Data Analysis

The fuel cycle data are summarized in Table 5. The theoretical density of mixed thorium-uranium-plutonium $\text{Th}_{1-x-y}\text{U}_x\text{Pu}_y\text{O}_2$ oxide fuel is given by [13]

$$\rho_{ox} = 10.001 + 0.914x + 1.390y \quad [\text{g/cm}^3]. \quad (2)$$

In all cases an actual fuel density of $\approx 90\%$ theoretical density is adopted.

Figs. 3 and 4 show the calculated k_{∞} as a function of burn-up; calculated values for a CANDU operating on natural uranium fuel from [9] are also shown for comparison. The discharge burn-up is determined following the criterion given by *Janssen* [8] that, to operate the reactor with sufficient margin, the cycle-averaged k_{∞} should not be smaller than 1.05. In order to reach a reasonable exit burnup, the initial enrichment should be chosen such that k_{∞} initially is well above 1.15, since k_{∞} drops by ≈ 0.04 after start up. The remarkably small slope of k_{∞} versus burnup for the Thorium-HEU and Thorium-MEU systems, which is due to a high conversion ratio, enables reaching higher exit burnups for a slight increase in enrichment.

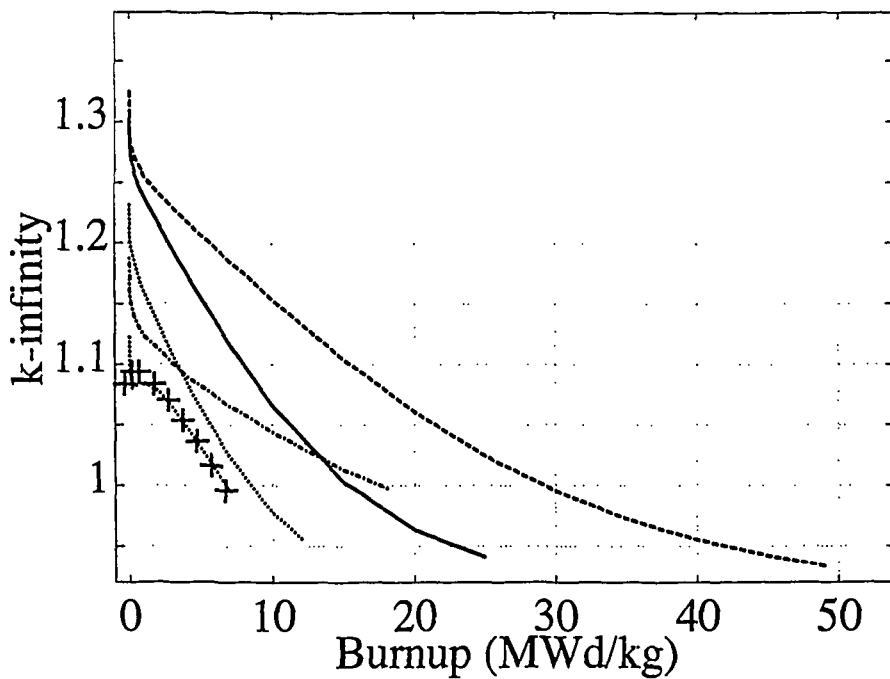


Fig. 3. k_{∞} of HWR once-through fuel systems:

- ++ : CANDU with natural uranium [9]. : Natural uranium (this work)
- : Thorium-Pu (1.6 wt% enrichment). - - - - : Thorium-HEU.
- : Thorium-Pu (2.2 wt% enrichment). - - - - : Thorium-MEU.

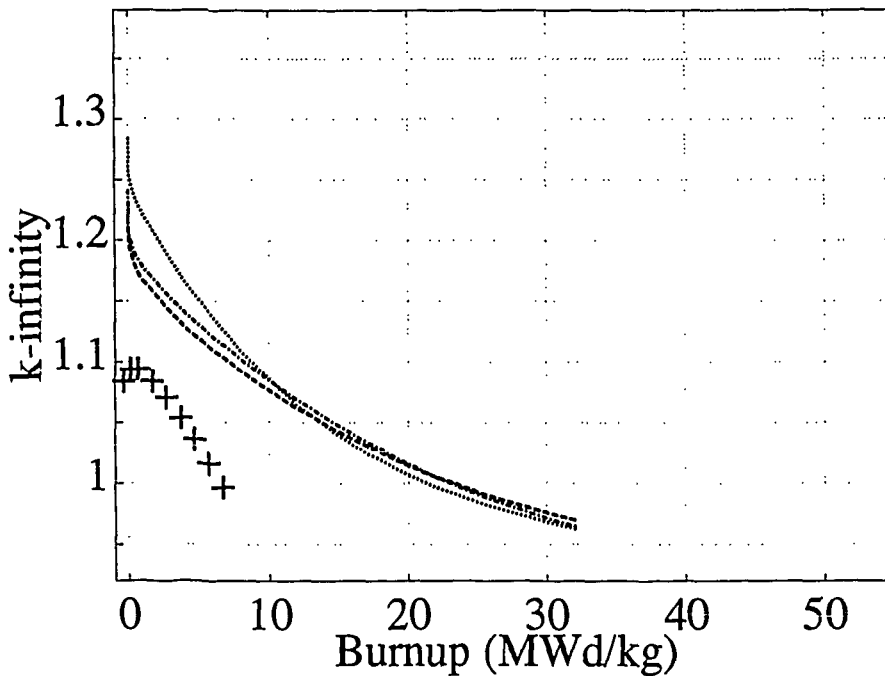


Fig. 4. k_{∞} of thorium in HWR systems with uranium recycle:

- ++ : CANDU with natural uranium [9].
- - - - : Thorium-HEU. - - - - : Thorium-MEU. : Thorium-Pu.

Table 5. Fuel cycle data (OT: Once-through mode; UR: with uranium recycle). Notes: the quoted density ρ_{HM} refers to heavy metal, i.e. without oxygen; the exit burnup is the assembly average.

Fuel Cycle	Fuel	x (wt%)	y (wt%)	ρ_{HM} (g/cm ³)	Exit Burnup (MWd/kg)	C
HWR NatU	U	—	—	8.533	7	1.85+07
Pu-THWR OT (1)	Th _{1-y} Pu _y	—	2.5	7.938	12	1.16+07
Pu-THWR OT (2)	Th _{1-y} Pu _y	—	3.3	7.947	25	5.57+06
HEU-THWR OT	Th _{1-x} U _x	2.0	—	7.932	49	2.85+06
MEU-THWR OT	Th _{1-x} U _x	10.0	—	7.985	18	7.70+06
HEU-THWR UR	Th _{1-x} U _x	4.1	—	7.984	32	4.33+06
MEU-THWR UR	Th _{1-x} U _x	7.6	—	8.019	32	4.31+06
Pu-THWR UR	Th _{1-x-y} Pu _y U _x	2.9	1.1	7.977	32	4.33+06

Normalisation of the results to values per 1 GW_ea requires definite assumptions for the *net efficiency* and the *load factor*.

The *net efficiency* is assumed to be 0.33. The *load factor* is assumed to be 100%, since, because of their cost structure, the nuclear power plants will normally be part of the base load capacity of the power program of a utility.

The values adopted for the above factors are common for all fuel cycles analysed in this study. Therefore they do not affect the relative results for the amounts of radionuclides or the radiotoxicities. However, these values are important when our results are compared with those of other authors. As an example, we obtain 158 tU/GW_ea for the uranium requirement of a HWR operating on natural uranium. Our value for an efficiency value of 0.33 is consistent with the value of 171 tU/GW_ea stated in [10] for an efficiency value of 0.30.

The nuclide quantities (whether in number of nuclides or gram per cm³) resulting from SAS6 are averaged over the four fuel annuli (see Fig. 2), by weighting with the fraction of pins in each annulus, and are normalized to 1 GW_ea by multiplication with

$$C = \frac{365.25 \cdot 10^6}{\eta \rho_{HM} BU} \quad [cm^3/GW_e a]. \quad (3)$$

Here, η is the net thermal efficiency, ρ_{HM} is the mass density of heavy metal (in g/cm³), and BU is the exit burnup (in MWd/kg).

4. RESULTS

4.1 Fuel Cycle Information

The fuel cycle information of the studied fuel cycles is presented in Tables 7 and 8. The actinide masses in discharged fuel are for 180 d cooling time. Note the high natural uranium and separative work (SW) requirements of the once-through MEU-THWR system. The resource utilization of the HEU-THWR once through system is better, but is obtained for an exit burnup of 49 MWd/kg. Typical values per GW_ea for the once-through uranium-plutonium LWR system are 217 t natural uranium and 160 tSW and for the system with plutonium-uranium recycle 120 t natural uranium and 104 tSW. Regarding resource utilization, HEU- or MEU-thorium once-through systems therefore compare unfavourably with uranium systems.

For the systems with uranium recycle, an approximate equilibrium cycle was adopted. Searching for equilibrium is very time consuming. Equilibrium was defined in Chapter 2 as the situation when all reloads are identical, accounting for an out-of-core loss of 0.1%. Two factors greatly complicate the search for equilibrium. First, all 37 pins in the fuel bundle receive fresh fuel of identical composition at BOL. However, the fuel compositions of the four annuli at EOL are obviously different. Second, the coupled differential equation approach for determining equilibrium concentrations cannot be applied when an a priori unknown amount of makeup fuel must be added in order to attain a required fissile contents of fresh fuel.

Equilibrium was searched for iteratively. Equal exit burnups in the iterations was reached by appropriate choice of the makeup fuel. The number of iterations was typically four. The results in this Chapter are therefore approximations for the values at equilibrium. Table 6 lists the relative differences in the masses of uranium isotopes in the discharge fuel of consecutive cycles (after sufficient cooling) for the adopted equilibrium cycles. The deviations are sufficiently small to determine the potential of THWR cycles for reduced actinide waste production.

Table 6. *Relative differences (in %) between amounts of uranium isotopes in the discharged fuel of consecutive cycles, based on weighted averages over the four fuel annuli. U-238 is ignored for the HWR with Th-Pu fuel, because of its low weight fraction of $2 \cdot 10^{-2}$ wt% U.*

Isotope	HWR with Th-HEU fuel and U Recycle	HWR with Th-MEU fuel and U Recycle	HWR with Th-Pu fuel and U recycle
U-232	-1.4	-13.0	-8.0
U-233	-0.1	-3.3	-5.5
U-234	-2.8	-2.2	-2.8
U-235	+0.1	+2.7	-3.8
U-236	+10.8	+8.6	-0.7
U-238	-0.0	-2.2	—

Table 7. Fuel Cycle Information - THWR Systems with Once-through Fuel Systems (0.2 wt% tails enrichment).

Fuel Cycle	NatU	Th-Pu (1)	Th-Pu (2)	Th-HEU	Th-MEU
Topping isotope	—	Pu-fiss	Pu-fiss	U-235	U-235
Fuel Residence Time (EFPD)	266	456	951	1863	684
HM Replacement ($t/GW_e a$)	158	92.1	44.3	22.6	61.5
Reload Enrichment (wt% HM)	0.71	1.66	2.19	2.80	2.00
Average Discharge Burnup (GWd/t)	7	12	25	49	18
<u>Reload Fuel Requirements</u>					
U-235 Enrichment Makeup (wt%)	0.71	—	—	93	20
Pu-fiss/Pu-tot (wt%)	—	66	66	—	—
Natural U ($t/GW_e a$)	158	0	0	123	236
SW ($tSW/GW_e a$)	0	0	0	160	279
U ($t/GW_e a$)	158	0	0	0.68	6.1
Pu ($t/GW_e a$)	0	2.3	1.5	0	0
Th ($t/GW_e a$)	0	89.8	42.8	21.9	55.3
<u>Discharged Fuel Properties</u>					
U-fiss/U (wt %)	0.23	95	92	65	16
Pu-fiss/Pu (wt %)	74	39	44	34	72
Th ($t/GW_e a$)	0	89	42	21	54
Pa ($kg/GW_e a$)	0	2.8	2.3	1.1	1.6
U ($t/GW_e a$)	156	0.7	0.5	0.52	5.9
Np ($kg/GW_e a$)	3.5	0.01	0.03	5.7	4.3
Pu ($t/GW_e a$)	0.56	1.4	0.6	0.004	0.04
Am ($kg/GW_e a$)	0.54	30	30	0.060	0.19
Cm ($kg/GW_e a$)	0.045	4.3	7.6	0.017	0.022

Table 8. Fuel Cycle Information - THWR Systems with U Recycle (0.2 % tails enrichment. Eqm = equilibrium.)

Fuel Cycle	Th-HEU	Th-MEU	Th-Pu
Topping isotope	U-235	U-235	Pu-fiss
Fuel Residence Time (EFPD)	1217	1217	1217
HM Replacement (t/GW _e a)	34.57	34.56	34.59
Eqm Reload Enrichment (wt% HM)	2.12	2.16	2.26
Average Discharge Burnup (GWd/t)	32	32	32
<u>Reload Fuel Requirements</u>			
U-235 Enrichment Makeup (wt%)	93.0	68.0	—
Pu-fiss/Pu-tot (wt%)	—	—	66
Eqm Natural U (t/GW _e a)	38	49	0
Eqm SW (tSW/GW _e a)	49	63	0
Eqm U Makeup (t/GW _e a)	0.21	0.37	0
Eqm Pu Makeup (t/GW _e a)	0	0	0.37
Eqm Th (t/GW _e a)	33.2	30.0	33.2
<u>Discharged Fuel Properties</u>			
Eqm U-fiss/U (wt %)	45	12	52
Eqm Pu-fiss/Pu (wt %)	42	56	50
Eqm Th (t/GW _e a)	32	29	32
Eqm Pa (kg/GW _e a)	1.4	1.3	1.6
Eqm U (t/GW _e a)	1.2	4.25	0.97
Eqm Np (kg/GW _e a)	13	16	11
Eqm Pu (kg/GW _e a)	9.6	39	117
Eqm Am (kg/GW _e a)	0.079	0.47	12
Eqm Cm (kg/GW _e a)	0.014	0.088	3.7

4.2 Comparison with INFCE Fuel Cycle Information

Thorium fuel cycles were extensively studied by Working Group 8 of the International Nuclear Fuel Cycle Evaluation [5]. The emphasis of the INFCE studies was not on actinide waste, but on uranium utilization. Although the conclusions of these studies are of little use for the present study, the reported fuel cycle information of the thorium-HEU and thorium-Pu HWR systems with uranium recycle may give some valuable information. Table 9 presents a comparison of the INFCE information of these fuel cycles with corresponding data of Table 8.

The correspondence is fair in general, with a few exceptions. The annual thorium requirements found by INFCE are about 20% higher than our values, but the equilibrium enrichments (fissile fractions) are equal. Also the INFCE values for fissile fractions of uranium and plutonium in spent Th-HEU fuel are much higher than the present values. Hence, the conversion ratio of thorium fuel cycles was overestimated by INFCE, which explains the lower natural uranium and SW requirements.

Table 9. Comparison with Fuel Cycle Information from INFCE for THWR Systems with U Recycle. Eqm = equilibrium; n.s. = not specified.

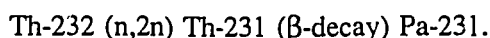
Fuel Cycle	Th-HEU		Th-Pu	
	Present	INFCE	Present	INFCE
Topping isotope	U-235		Pu-fiss	
Fuel Residence Time (EFPD)	1217	1029	1217	1053
HM Replacement ($t/GW_e a$)	34.57	42.7	34.59	41.4
Eqm Reload Enrichment (wt% HM)	2.12	2.11	2.26	2.17
Average Discharge Burnup (GWd/t)	32	29.3	32	30
<u>Reload Fuel Requirements</u>				
U-235 Enrichment Makeup (wt%)	93.0	93.0	—	—
Pu-fiss/Pu-tot (wt%)	—	—	66	n.d.
Eqm Natural U ($t/GW_e a$)	38	33.3	0	0
Eqm SW ($tSW/GW_e a$)	49	43.4	0	0
Eqm U Makeup ($t/GW_e a$)	0.21	0.18	0	0
Eqm Pu Makeup ($t/GW_e a$)	0	0	0.37	0.35
Eqm Th ($t/GW_e a$)	33.2	41	33.2	40
<u>Discharged Fuel Properties</u>				
Eqm U-fiss/U (wt %)	45	85	52	n.s.
Eqm Pu-fiss/Pu (wt %)	42	75	50	n.s.
Eqm U ($t/GW_e a$)	1.2	0.84	0.97	0.79
Eqm Pu ($kg/GW_e a$)	10	3.4	4.25	n.s.

4.3 Fresh Fuel and Discharged Fuel Composition

The discharged actinide masses per GW_ea are listed in Tables 10 and 11. The various types of fresh fuel components cause different "transmutation" products in the discharged fuel. This transmutation behaviour is responsible for the basic differences between the actinide contents of discharged fuel of the various fuel cycles.

4.3.1 Thorium

Irradiation of thorium mainly produces isotopes of protactinium and uranium isotopes. For instance, Pa-231 is produced via the reaction sequence



Pa-231 appears to be a major contributor to the radiotoxicity of discharged thorium fuel. This will become evident from the radiotoxicities presented in the next section and has been confirmed elsewhere (see e.g. [11]).

Also of importance are U-232 and U-234. These uranium isotopes are mainly produced via the (n,2n)-reaction in U-233, and the (n, γ)-reactions in U-233 and Pa-233, respectively.

4.3.2 Recycle Fuel

Recycle uranium contains large fractions of even uranium isotopes, of which U-234, U-236, U-238 are the most abundant. U-234 and U-236 are both neutron absorbers, neutron absorption in U-234 producing fissile U-235¹, whereas neutron absorption in U-236 leads to the production of harmful Np-237. Consequently, the Np-237 production rate (in mass/ GW_ea) in fuel cycles with U recycle is larger than in all once-through fuel systems.

One of the negative aspects of uranium recycling is the build-up of U-238. It appears that the fractions of U-238 in discharged fuel are 0.5 wt% of HM and 0.1 wt% of HM, respectively for the uranium recycling and the once-through HEU-THWR systems.

Conclusion: The large fractions of even uranium isotopes in recycle uranium lead to significant Np-237 and plutonium production rates and thus to high radiotoxicity of the actinide waste.

¹Note however that although U-234 in fresh fuel is of value as a fertile fuel, the U-235 production mechanism via U-234 gives a negative neutron balance, since from Th-232 up to fission of U-235, 3 neutrons are consumed and ≈ 2.5 neutrons are produced.

Table 10. *Masses of actinides in discharged fuel in g per GW_e a for HWR NatU and Thorium Once-through fuel systems (at discharge).*

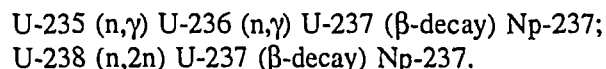
Nuclide	Natural U	Th-Pu (1)	Th-Pu (2)	Th-HEU	Th-MEU
Th-230	1.4E-02	2.9E+01	2.6E+01	1.5E+01	1.4E+01
Th-232	1.4E-03	8.9E+07	4.2E+07	2.1E+07	5.4E+07
Pa-231	.0	2.8E+03	2.3E+03	1.0E+03	1.6E+03
Pa-233	.0	8.8E+04	4.2E+04	2.4E+05	5.9E+04
U-232	.0	5.6E+02	8.9E+02	9.0E+02	5.5E+02
U-233	1.6E-03	5.7E+05	4.2E+05	2.7E+05	5.1E+05
U-234	6.8E+03	3.1E+04	4.0E+04	5.9E+04	4.6E+04
U-235	3.6E+05	2.1E+03	4.2E+03	3.9E+04	3.5E+05
U-236	1.1E+05	1.6E+02	4.6E+02	9.0E+04	1.3E+05
U-237	2.2E+02	2.1E-01	5.6E-01	7.0E+01	1.2E+02
U-238	1.6E+08	4.6E-01	8.0E-01	3.4E+04	4.8E+06
Np-237	3.5E+03	1.3E+01	3.0E+01	5.7E+03	4.3E+03
Np-238	1.6E+01	3.0E-02	6.9E-02	1.5E+01	1.1E+01
Np-239	1.4E+04	1.8E-02	2.0E-02	9.8E+00	4.5E+02
Pu-238	4.4E+02	3.7E+04	1.8E+04	2.0E+03	7.3E+02
Pu-239	3.9E+05	3.4E+05	8.0E+04	1.1E+03	2.6E+04
Pu-240	1.4E+05	5.6E+05	2.8E+05	4.9E+02	9.7E+03
Pu-241	2.9E+04	1.9E+05	8.7E+04	2.7E+02	4.0E+03
Pu-242	7.3E+03	2.2E+05	1.7E+05	2.6E+02	1.3E+03
Pu-243	1.3E+00	1.9E+01	1.2E+01	3.5E-02	1.8E-01
Am-241	2.6E+02	8.9E+03	6.7E+03	9.8E+00	8.7E+01
Am-242	1.2E+00	1.7E+01	1.2E+01	2.5E-02	2.2E-01
Am-242m	1.8E+00	7.2E+01	5.8E+01	8.4E-02	6.8E-01
Am-243	2.7E+02	2.1E+04	2.3E+04	5.0E-01	9.9E+01
Cm-242	5.7E+01	2.2E+03	2.3E+03	4.2E+00	2.2E+01
Cm-244	1.7E+01	3.2E+03	6.5E+03	1.5E+01	1.1E+01
Total	1.6E+08	9.1E+07	4.3E+07	2.3E+07	6.0E+07

Table 11. *Masses of actinides in discharged fuel in g per GW_e a for HWR Closed Thorium fuel cycles (at discharge).*

Nuclide	Th-HEU	Th-MEU	Th-Pu
Th-230	2.3E+01	2.1E+01	2.6E+01
Th-232	3.2E+07	2.9E+07	3.2E+07
Pa-231	1.4E+03	1.3E+03	1.6E+03
Pa-233	3.4E+04	3.1E+04	3.3E+04
U-232	1.8E+03	1.7E+03	1.9E+03
U-233	4.5E+05	4.1E+05	4.6E+05
U-234	2.6E+05	2.3E+05	2.6E+05
U-235	7.3E+04	7.9E+04	5.0E+04
U-236	2.5E+05	3.6E+05	2.0E+05
U-237	1.6E+02	2.1E+02	1.4E+02
U-238	1.6E+05	3.1E+06	1.9E+02
Np-237	1.3E+04	1.6E+04	1.1E+04
Np-238	3.1E+01	3.8E+01	2.6E+01
Np-239	3.7E+01	2.8E+02	1.3E-01
Pu-238	3.7E+03	4.6E+03	6.7E+03
Pu-239	3.3E+03	1.8E+04	6.3E+03
Pu-240	1.4E+03	9.3E+03	4.1E+04
Pu-241	7.1E+02	4.0E+03	1.7E+04
Pu-242	4.3E+02	2.6E+03	4.6E+04
Pu-243	5.7E-02	3.3E-01	4.2E+00
Am-241	2.3E+01	1.4E+02	1.3E+03
Am-242	5.5E-02	3.2E-01	2.7E+00
Am-242m	2.0E-01	1.1E+00	1.2E+01
Am-243	5.5E+01	3.3E+02	9.8E+03
Cm-242	7.8E+00	4.7E+01	5.8E+02
Cm-244	1.0E+01	6.3E+01	3.4E+03
Total	3.3E+07	3.3E+07	3.3E+07

4.3.3 Enriched Uranium Makeup Fuel

A major minor actinide produced from *enriched uranium* makeup fuel is Np-237. The main production paths are



The amounts of thus produced Np-237 in the Th-Pu once-through fuel systems are about two orders of magnitude less than in the other once-through systems. For long storage times ($> 10^4$ a), the Pu-241 and Am-241 decay path to Np-237 becomes important. The contribution from this path is significant for the Pu-topped THWR fuel cycles.

Table 12. Amounts of Np-237 after long storage times ($> 10^4$ a).

Fuel cycle	Production rate (kg/GW _e a)
Natural uranium once-through system	33
Th-Pu once-through system (1)	200
Th-Pu once-through system (2)	94
Th-HEU once-through system	4
Th-MEU once-through system	8
Th-HEU system with uranium recycle	14
Th-MEU system with uranium recycle	20
Th-Pu system with uranium recycle	29

Plutonium production by irradiation of U-238 proceeds via Np-239. The amount of Np-239 in fuel during irradiation is a measure of the magnitude of plutonium production. To emphasize that the good properties of thorium fuel cycles in this respect are without doubt, Table 13 recapitulates the Np-239 production rate data from Tables 10 and 11.

Table 13. Np-239 production rates.

Fuel cycle	Production rate (g/GW _e a)
Natural uranium once-through system	140000
Th-Pu once-through system (1)	0.02
Th-Pu once-through system (2)	0.02
Th-HEU once-through system	36
Th-MEU once-through system	450
Th-HEU system with uranium recycle	37
Th-MEU system with uranium recycle	280
Th-Pu system with uranium recycle	0.13

For the Pu-THWR, HEU-THWR and MEU-THWR once-through systems, the plutonium production rates are in the ratio of $10^{-6} : 2.5 \cdot 10^{-3} : 0.03$, with the natural uranium once-through system as reference. The three closed fuel cycles show a similar picture as their once-through counter parts, although less pronounced. This picture confirms the intuitive notion that the plutonium production rate in Th-based fuels is much lower than in U-based fuels.

4.3.4 Plutonium Makeup Fuel

Irradiation of *plutonium* makeup fuel leads to a shift in the plutonium isotopic vector towards higher isotopes and to the production of higher actinides. The net americium and curium production rates are predominantly determined by the initial plutonium contents of the fuel, and to a lesser extent by the U-238 contents. For the Pu-THWR, HEU-THWR and MEU-THWR once-through systems, the americium+curium production rates are in the ratio of $60 : 0.003 : 0.33$, with the natural uranium once-through system as reference. For the Pu-THWR, HEU-THWR and MEU-THWR systems with uranium recycling, the americium+curium production rates are in the ratio of $25 : 0.14 : 1$, with the natural uranium once-through system as reference.

4.3.5 Comparison with Borssele PWR

Table 14 lists the production rates of some actinides in the HEU-THWR cycle with uranium recycle and in the Borssele PWR. Note that the rates for Np-237 and Pu-238 are almost equal.

Table 14. *Net production rates of actinides in the Borssele NPP [16] and in the HEU-THWR cycle with uranium recycle (this work).*

Nuclide	Net production rate (kg/GW _e a)	
	Borssele NPP	HEU-THWR with U recycle
Np-237	14	13
Pu-238	4.4	3.7
Pu-239	221	3.3
Pu-240	78	1.4
Pu-241	55	0.7
Pu-242	15	0.4
Am-241	1.9	0.002
Am-243	2.5	0.000
Cm-242	0.6	0.008
Cm-244	0.6	0.010

4.4 Total Radiotoxicity of Actinide Waste

The evolution with time of the radiotoxicity of actinide waste from the THWR systems is shown in Figs. 5 and 6. The radiotoxicity is expressed in units of ALI per GW_ea. The time scale is set by the decay times of the involved radionuclides and the geologic time scale, and runs up to 10⁶ a.

In the once-through fuel systems, all actinides in the discharged fuel are waste. Fig. 5 therefore presents the radiotoxicities corresponding to the masses listed in Table 10. With the HEU-THWR and MEU-THWR systems, a reduction by a factor of 10 is achieved at 10³ a with respect to the reference natural uranium fuel cycle. Unfortunately, the reduction vanishes at 10⁵ a and for larger storage times even an enhancement is seen. With Pu-THWR systems, the radiotoxicity is always significantly higher than for the reference system. The effect becomes less at higher burnups. However, Fig. 5 does not reflect that feeding plutonium into the THWR system leads to an overall reduction of the radiotoxicity of the world anthropogenic actinide inventory.

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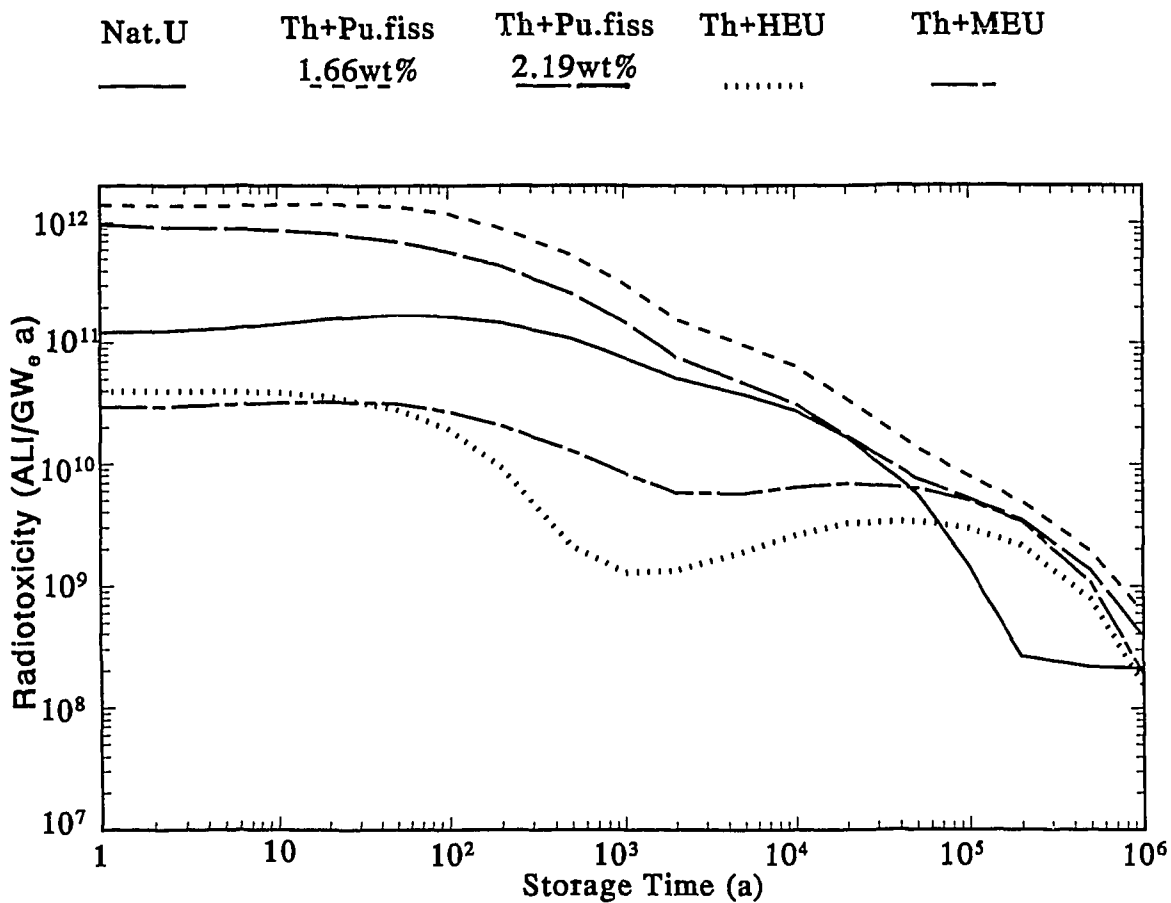


Fig. 5. Radiotoxicity of actinide waste in THWR once-through systems compared to the natural uranium in HWR system.

In the closed fuel cycles, 0.1% of the uranium and 100% of all other actinides in discharged fuel are waste. The recycled fraction of uranium (99.9%) is part of the core inventory. This part is not included in Fig. 6. Note that for long prolongation times of the fuel cycle, the contribution of the core inventory to the radiotoxicity per GW_ea of generated electricity vanishes.

The results of Fig. 6 show a disappointingly small reduction of the radiotoxicity of the actinide waste. This perhaps unexpected outcome can only be understood by inspecting the radiotoxicity of each actinide element separately, which is done in the next section.

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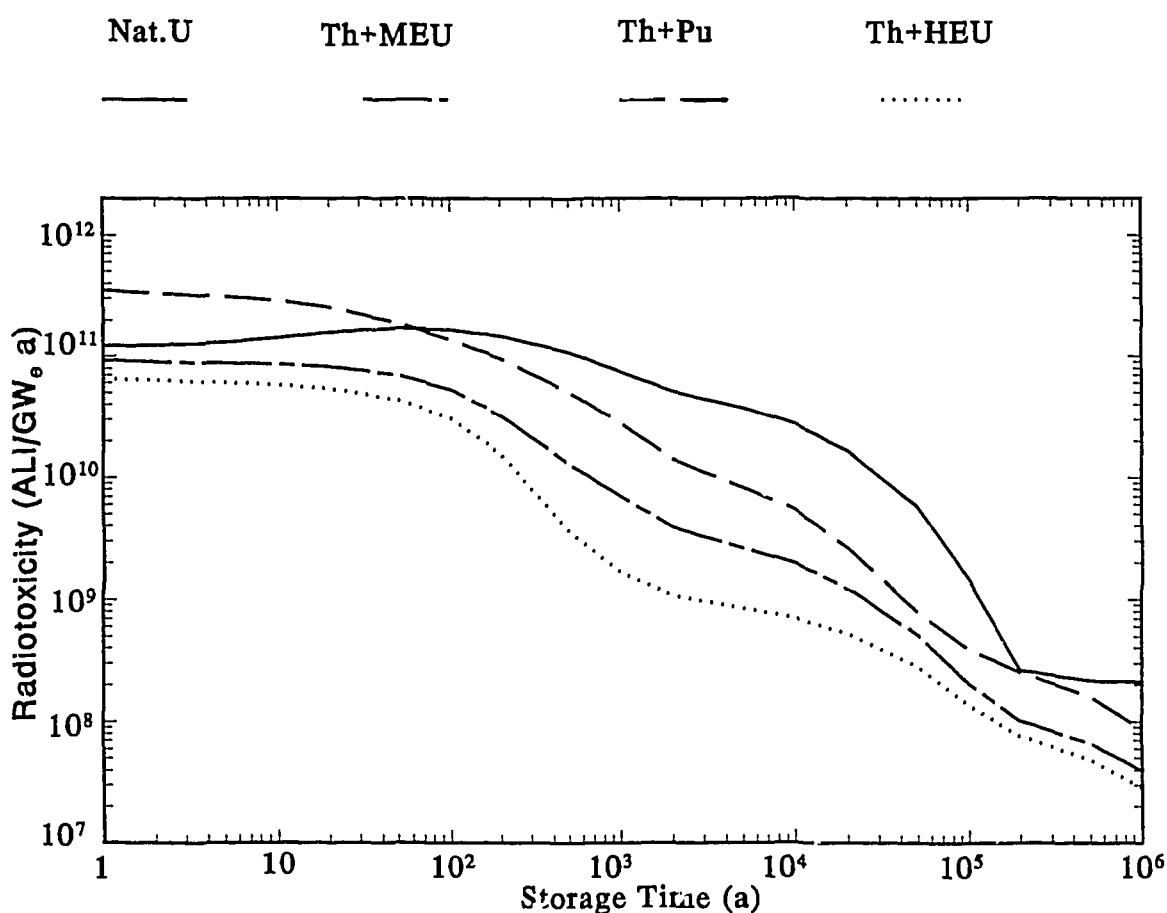


Fig. 6. Radiotoxicity of actinide waste in THWR systems with uranium recycle compared to the natural uranium in HWR once-through system.

4.5 Radiotoxicity per Element of Actinide Waste

4.5.1 Once-Through Systems

The radiotoxicities per element for once-through THWR systems are shown in Figs. 7-10. The results for each fuel cycle will be discussed briefly.

Fuel cycle 1. In the natural uranium HWR once-through system (Fig. 7), plutonium clearly is the dominant contributor to the radiotoxicity upto storage times of 10^5 a, whereas uranium dominates for longer times.

Fuel cycles 2 and 3. The Pu-THWR once-through systems (Fig. 8; only the 2.19 wt% Pu system is shown) roughly give the same picture as the natural uranium once-through system. Differences are that initially there is much more curium, that uranium becomes dominant earlier (after 10^4 a) and that the radiotoxicity of this uranium is also larger by a factor of ≈ 30 . Note that the discharged uranium is not irradiated natural uranium, but is produced from thorium and by further transmutation. The isotope U-233 contributes 80% of the radiotoxicity of uranium at long storage times, the remaining 20% being due to U-234.

Fuel cycle 4. In the HEU-THWR once-through system (Fig. 9), the radiotoxicities of plutonium and uranium in the waste are almost the same for the first 1000 a, after which uranium becomes the dominant contributor. Also this uranium is predominantly produced from thorium and further transmutation.

Fuel cycle 5. In the MEU-THWR once-through system (Fig. 10), plutonium is the dominant contributor upto 10^4 a, and uranium for longer storage times.

One could conclude from the dominance of uranium that the uranium should be recycled in order to reduce the radiotoxicity of the actinide waste. However, recycling has a negative side-effect: deterioration of the isotopic vector. In case of uranium recycling, the weight fractions of U-234, U-236 and U-238 will increase with increasing generation number. This effect provides the explanation for the disappointing picture emanating from Fig. 6. The influence of this effect on the long-term radiotoxicity behaviour should become clear when inspecting the radiotoxicities per element for closed cycles.

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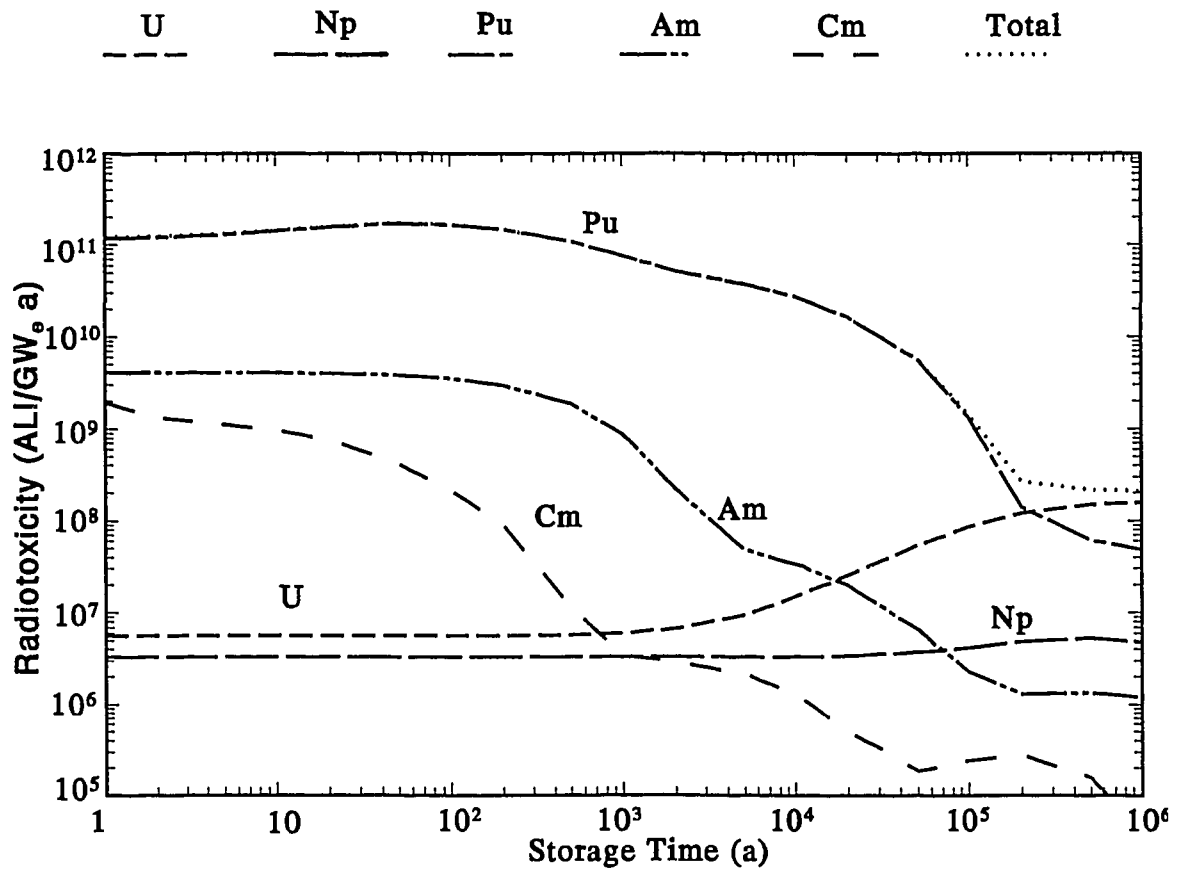


Fig. 7. Radiotoxicity of actinides in the HWR natural uranium once-through system (Fuel Cycle 1).

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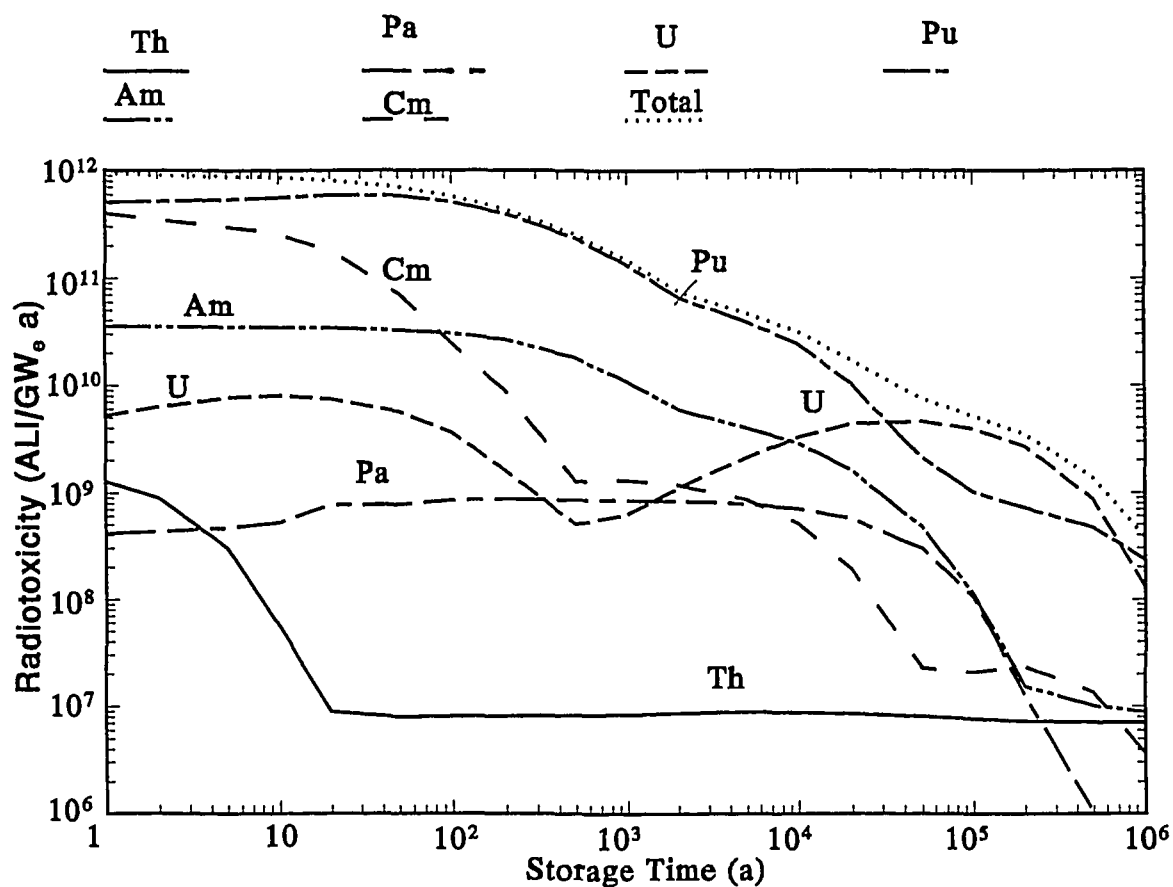


Fig. 8. Radiotoxicity of actinides in the Pu-THWR (Pu fresh fuel fraction: 2.19 wt% HM) once-through system (Fuel Cycle 3).

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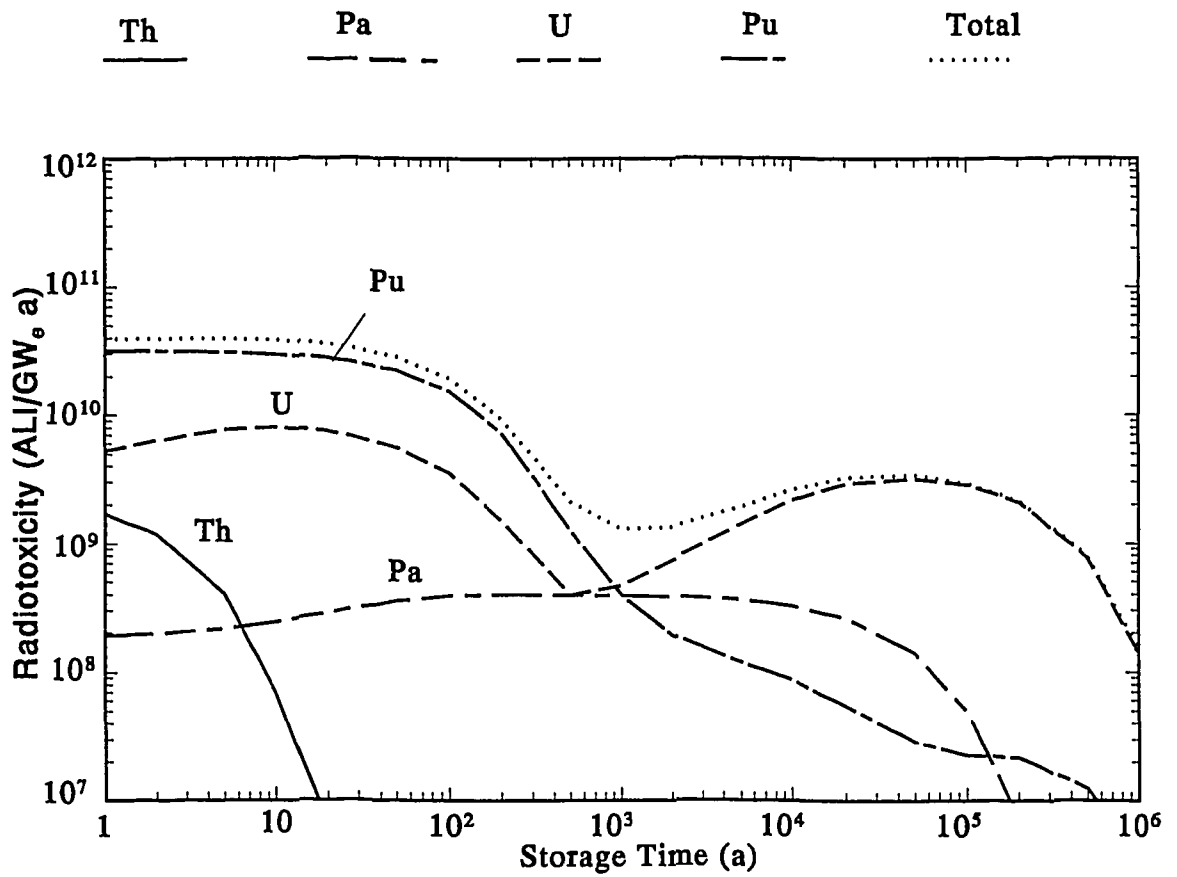


Fig. 9. Radiotoxicity of actinides in the HEU-THWR once-through system (Fuel Cycle 4).

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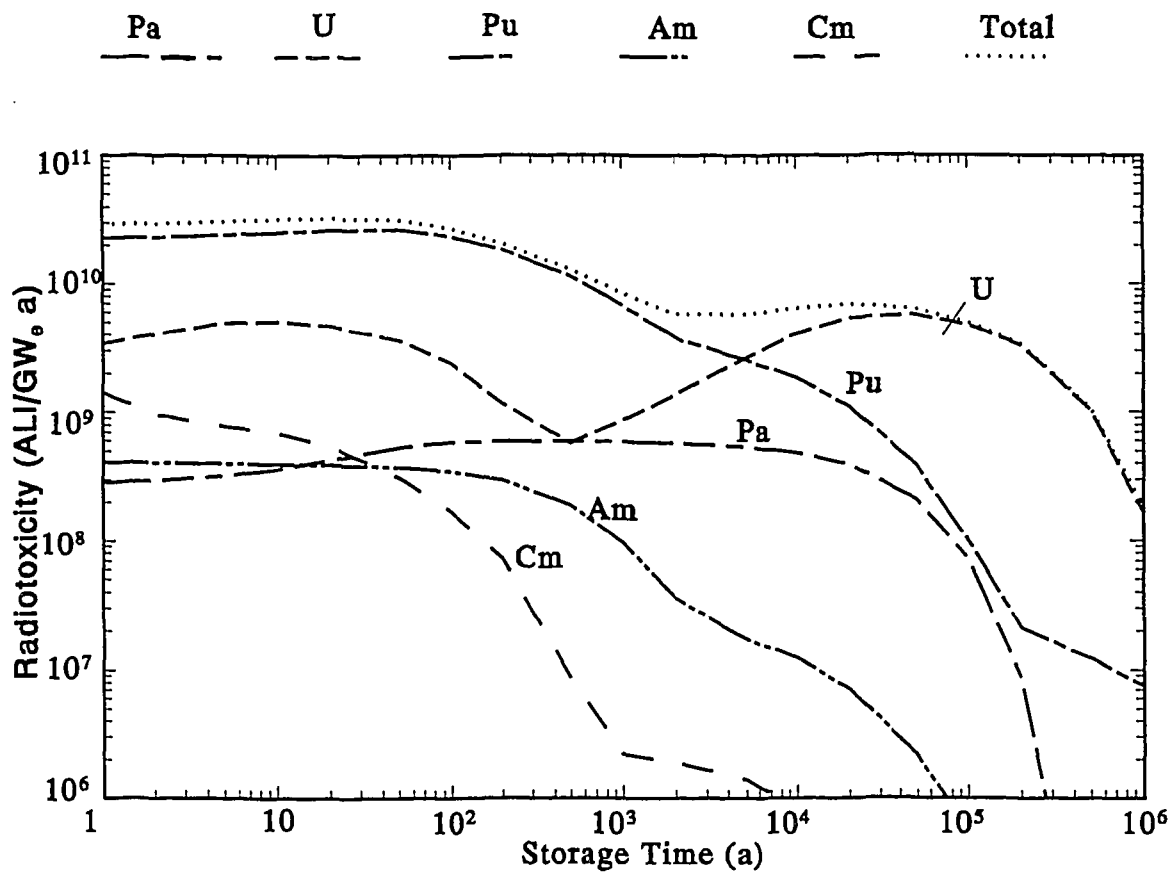


Fig. 10. Radiotoxicity of actinides in the MEU-THWR once-through system (Fuel Cycle 5).

4.5.2 Closed Systems

The radiotoxicities per element for THWR systems with uranium recycle are shown in Figs. 11, 12 and 13.

Fuel cycle 6. In the HEU-THWR system with uranium recycle (Fig. 11), plutonium dominates the actinide radiotoxicity for storage times up to 100 a. Between 10^3 and 10^5 a, Pa-231 and plutonium contribute equally. Hence, further actinide waste reduction requires recycling of both Pu and Pa-231. However, recycling of Pa-231 will lead to the build-up of U-232. The U-232 descendants Bi-212 and Tl-208 are hard γ -ray emitters and reach maximum activities after 10 years of storage. Recycling of Pa-231 would thus lead to high fuel fabrication costs and is not recommended.

Fuel cycle 7. The MEU-THWR system with uranium recycle (Fig. 12) shows the same trends, but the plutonium contribution is a factor of ≈ 2 larger and americium becomes a significant contributor.

Fuel cycle 8. Also the Pu-THWR system with uranium recycle (Fig. 13) offers the same picture, however curium is also important for storage times up to 100 a.

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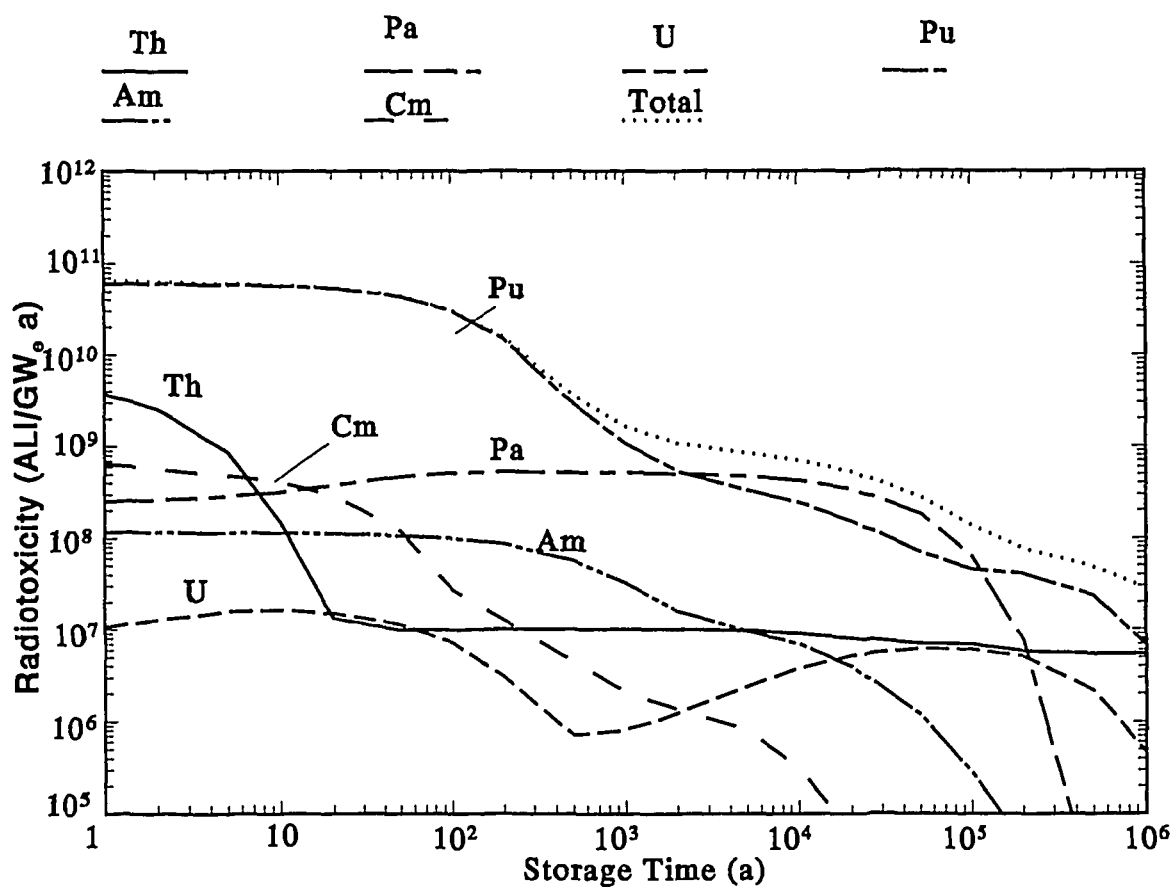


Fig. 11. Radiotoxicity of actinides in the HEU-THWR system with uranium recycle (Fuel Cycle 6).

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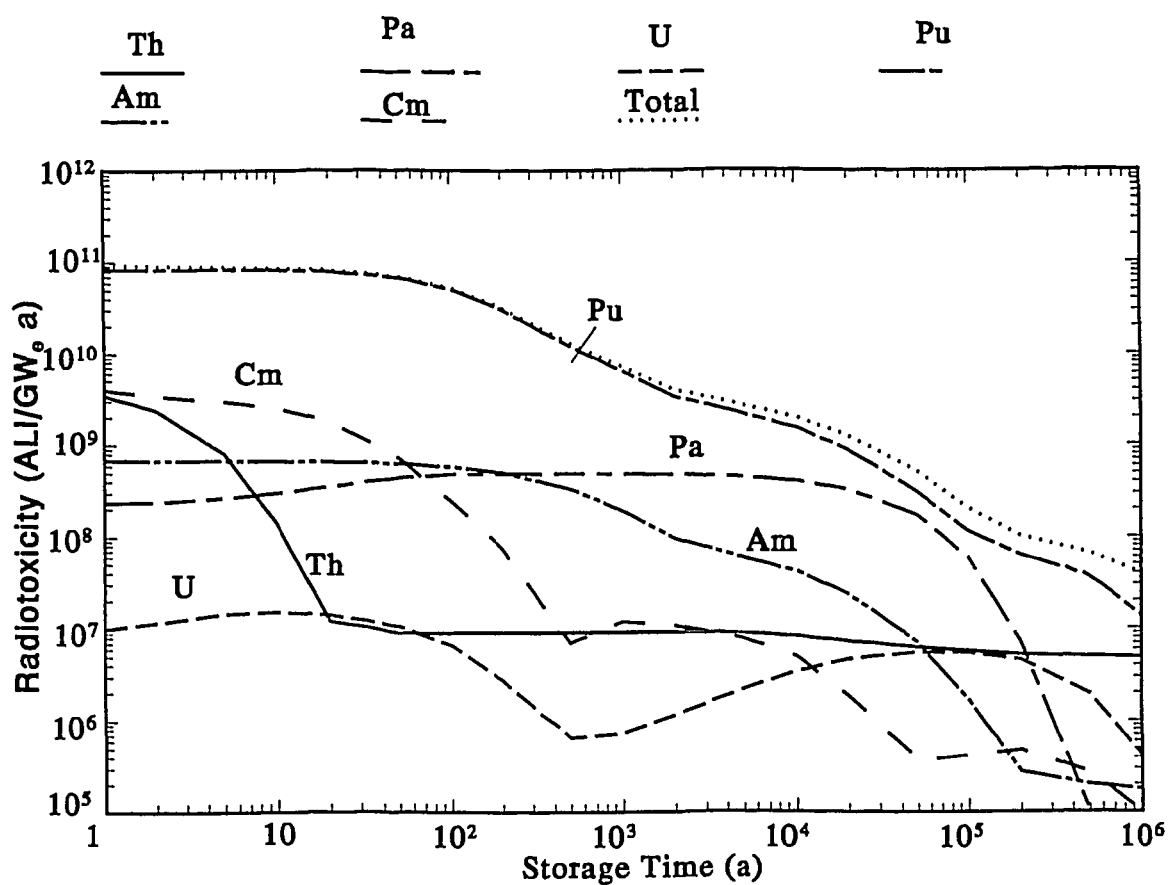


Fig. 12. Radiotoxicity of actinides in the MEU-THWR system with uranium recycle (Fuel Cycle 7).

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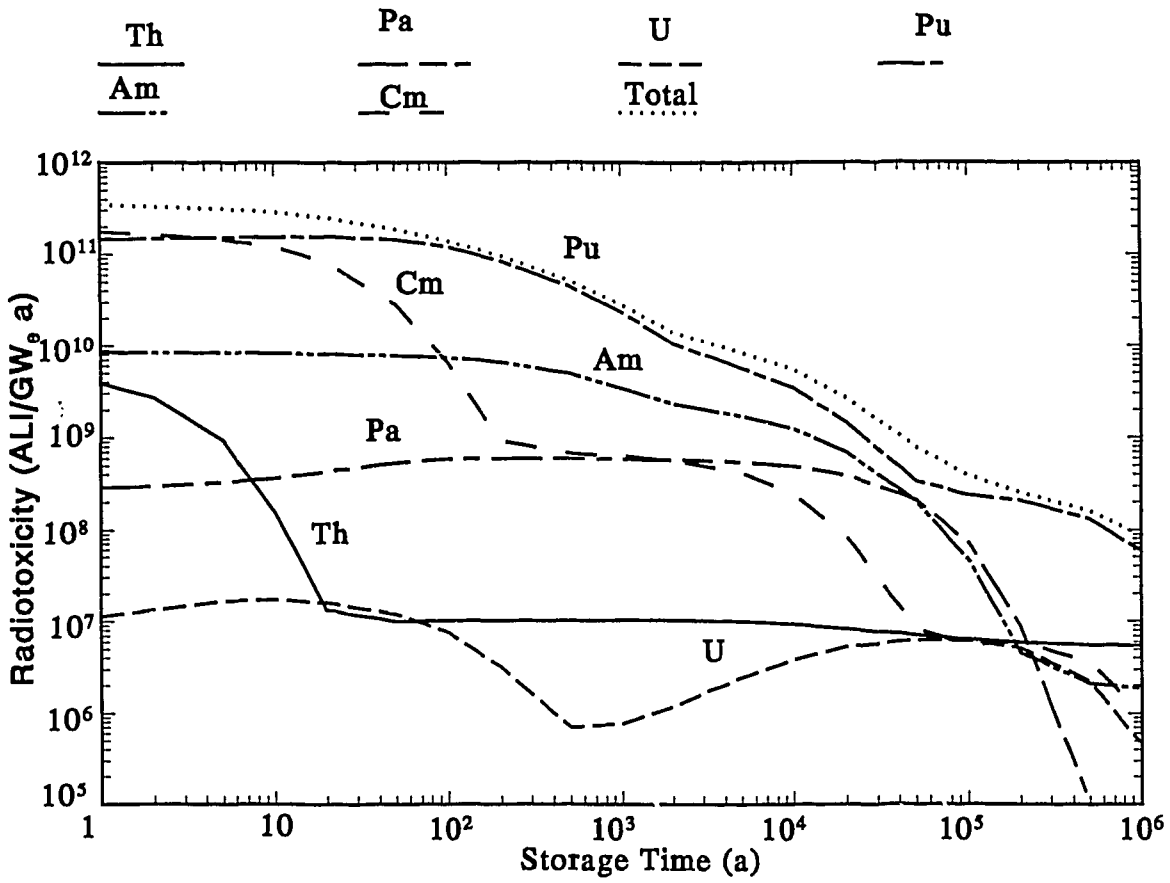


Fig. 13. Radiotoxicity of actinides in the Pu-THWR system with uranium recycle (Fuel Cycle 8).

4.6 Pu burning in Thorium

Table 15 lists the plutonium consumption rates (consumed mass per GW_ea) of plutonium-topped thorium fuel cycles. Note that these rates are net rates, i.e. consumption minus production. Table 15 shows that Pu-THWR once-through systems are about a factor of 2.5 more effective for plutonium burning than a 100% or full MOX-fuelled PWR. This is predominantly due to different plutonium production rates (see section 4.3.3). The closed Pu-THWR fuel cycle is less effective for Pu-burning than the MOX-PWR cycle. This is due to the high conversion ratio of the THWR.

Table 15. *Plutonium consumption rate in $\text{kg}/\text{GW}_e\text{a}$ of Pu-THWR fuel cycles.*

Fuel cycle 2: Pu-THWR once-through system with Pu-fissile of 1.66 wt% HM and exit burnup of 12 MWd/kg.

Fuel cycle 3: Pu-THWR once-through system with Pu-fissile of 2.19 wt% HM and exit burnup of 25 MWd/kg.

Fuel cycle 8: Pu-THWR system with uranium recycle and total fissile content of 2.26 wt% HM and exit burnup of 32 MWd/kg.

PWR-MOX: U-Pu full MOX in PWR fuel cycle, first recycle, with a burnup of 50 MWd/kg and with plutonium isotope fractions (in wt%) Pu-238: 1.8; Pu-239: 59.0; Pu-240: 23.0; Pu-241: 12.2; Pu-242: 4.0 [12].

Isotope	Fuel Cycle			
	2	3	8	PWR-MOX
Pu-238	20	20	0	0
Pu-239	890	700	190	420
Pu-240	0	80	50	30
Pu-241	110	100	30	-60
Pu-242	-70	-70	-20	-50
Pu-total	950	830	250	340

4.7 Actinide Waste from the Fuel Cycle Front-End

The front end of the nuclear fuel cycle also gives rise to actinide waste. The main sources in this respect are mining and milling and uranium enrichment. These stages will be analysed here, but only briefly since the focus of this report is on actinide waste from spent fuel. Radiotoxicity values will be given for a storage time of 10^6 a.

Regarding mining, for both uranium and thorium an extraction efficiency of 95% is assumed [14], a value which is still representative for current industry practice [15]. In mining and milling, the geologic immobilization of the radioactive elements is destroyed. It is assumed here that, despite the long storage times considered, this immobilization is not restored. Enrichment produces a depleted tailings stream, besides the desired enriched product stream. The tailings are not used in the fuel cycles considered here, and thus are waste, in accordance with the adopted definition of waste [1]. The masses of the front end actinide waste streams are derived from Tables 7 and 8.

Table 16 lists the corresponding radiotoxicities. The adopted specific radiotoxicities for ingestion of natural uranium, uranium enrichment tailings and natural thorium are 1.095, 0.952 and 0.168 ALI/g, respectively. Note that concerning actinide radiotoxicity, the front- and back-end contributions of the HEU-THWR and MEU-THWR fuel cycles (both once-through and closed) are of the same order of magnitude at 10^6 a. For the Pu-THWR fuel cycles, the front-end contribution is negligible, but the plutonium topping fuel is obtained from external systems. These systems are not included in the analysis.

4.8 Infinite Recycling

Thermal reactor thorium fuel cycles in general may offer an advantage not mentioned yet. In current thermal uranium-plutonium systems, multiple recycling of plutonium is limited. This is due to degradation of the isotopic vector of recycle plutonium with increasing generation number, which necessitates increasing plutonium fractions. However, the allowed value of the plutonium fraction is limited due to safety considerations, and thus also the number of recycles is limited. The reprocessed uranium can be recycled to thermal reactors in several ways: via re-enrichment to produce low enriched uranium (LEU) fuel; via mixing with reprocessed plutonium to produce MOX fuel; or even via mixing with HEU to produce LEU fuel. The amount of reprocessed plutonium is insufficient for all reprocessed uranium to be mixed into MOX fuel. For topping with U-235 as HEU, significant supplies of natural uranium and separative work are required. Hence in current practice, the re-enrichment route is followed with LWR reactors. Thus the bulk of reprocessed uranium is left for final disposal as enrichment tails.

By contrast, infinite recycling of bred uranium in a thermal reactor thorium fuel cycle is expected to pose no problem from safety or other reactor-physical considerations [17]. Also all thorium can be recycled, using HEU as the preferred topping fuel. The amount of topping is comparatively small due to the intrinsic properties of U-233. High converting reactors, such as the HWR, are needed to further reduce the required amount of topping fuel, leading to less natural uranium requirements, less mining and milling waste, less uranium enrichment tails and lower separative work.

Table 16. Actinide waste from the front-end of the nuclear fuel cycle. Radiotoxicities are specified for a storage time of 10^6 a.

	Mass t/GW_{ca}			Radiotoxicity 10^6 ALI/ GW_{ca}			
	U in mining and milling tailings	U in enrichment tailings	Th in mining and milling tailings	U in mining and milling tailings	U in enrichment tailings	Th in mining and milling tailings	Total
Natural uranium once through	8.32	0	0	9.11	0	0	9.11
Pu-THWR once through	0	0	4.73	0	0	0.80	0.80
Pu-THWR once through	0	0	2.25	0	0	0.38	0.38
HEU-THWR once through	6.47	122.8	1.15	7.08	117	0.19	124
MEU-THWR once through	12.4	230.3	2.91	13.6	219	0.49	233
HEU-THWR uranium recycle	2.00	37.9	1.75	2.19	36.1	0.29	38.6
MEU-THWR uranium recycle	2.63	49.6	1.58	2.88	47.2	0.26	50.3
Pu-THWR uranium recycle	0	0	1.75	0	0	0.29	0.29

5. CONCLUSIONS

Actinide waste production rates were established in terms of actinide mass per unit of electricity produced ($1 \text{ GW}_e\text{a}$) and in terms of actinide radiotoxicity per GW_ea . Of the seven THWR systems studied, the closed HEU-THWR fuel cycle performed best, having both the smallest plutonium production rate and the lowest radiotoxicity. Relative to the natural uranium HWR once-through system, actinide waste radiotoxicity is reduced by a factor varying between somewhat less than 2 to 50 for the full range of storage times up to 10^6 a.

A further reduction of the radiotoxicity of actinide waste in the closed HEU-THWR cycle would require removal of not only plutonium, but also of Pa-231. Recycling of the latter is undesirable since it would lead to enhanced levels of U-232, necessitating costly radiation protection for fuel fabrication.

Pu-THWR once-through systems are about a factor of 2.5 more effective for plutonium burning than a 100% or full MOX-fuelled PWR. This is predominantly due to different plutonium production rates. However, closed Pu-THWR fuel cycles are less effective for Pu-burning than the MOX-PWR cycle, due to the high conversion ratio of the THWR.

Infinite recycling of the two major actinides (thorium and uranium) seems possible in thorium fuel systems. This feature would enable a considerable reduction of the waste mass with respect to current uranium-LWR systems. Further study is needed to assess the feasibility of infinite recycling in thorium systems. This could include various types of reactors and hybrid reactor-accelerator-based systems.

6. LIST OF ABBREVIATIONS

ALI	Annual Limit on Intake
BOL	Begin of Life
CANDU	Canadian Deuterium Uranium reactor
EFPD	Equivalent Full Power Days
EOL	End of Life
HWR	Heavy Water (moderated) Reactor
INFCE	International Fuel Cycle Evaluation
HEU	High Enriched Uranium. The IAEA Safeguards definition for HEU is: <i>uranium enriched to 20 wt % U-235 or more</i> . At the time this definition was established, U-233 was not present in the fuel cycle, which explains why this nuclide is omitted. HEU is considered as special fissionable material and as direct-use material.
HM	Heavy Metal
LEU	Low Enriched Uranium
LWR	Light Water Reactor
MEU	Medium Enriched Uranium. MEU was defined in INFCE (see text, p. 4), but is not adopted by IAEA Safeguards.
MOX	Mixed oxide; term used in nuclear chemistry for a mixture of uranium oxide and plutonium oxide
NatU	Natural Uranium
PWR	Pressurized Water Reactor
SSET	Self-sustaining Equilibrium Thorium fuel cycle, i.e. the conversion ratio of the fuel cycle is slightly larger than unity to overcome losses of fissile fuel due to processing.
SW	Separative Work
THWR	Thorium fuelled Heavy Water moderated Reactor

7. REFERENCES

- [1] International Atomic Energy Agency, *Radioactive Waste Management Glossary*, IAEA-TECDOC-264 (1982).
- [2] J.M. Li, P.F.A. de Leege, and J.L. Kloosterman, *SAS6: Two-Dimensional Depletion and Criticality Analysis Code*, Petten, Netherlands Energy Research Foundation (ECN), ECN-I--94-041 (to be published).
- [3] A.J. Janssen, private communication.
- [4] A.R. Dastur, A.S. Gray, N. Gagnon, D.B. Buss, and R.A. Verral, *The Role of CANDU in Reducing the Radiotoxicity of Spent Fuel*, Global '93 Conference on Future Nuclear Systems, Emerging Fuel Cycles and Waste Disposal Options, Seattle, Washington, USA, 12-17 September 1993, pp. 1229-1236.
E. Critoph, S. Banerjee, F.W. Barclay, D. Hamel, M.S. Milgram, J.I. Veeder, *Prospects for Self-Sufficient Equilibrium Thorium Cycles in CANDU Reactors*, ANS Winter Meeting, San Francisco, 16-21 November 1975, AECL-5501.
- [5] International Nuclear Fuel Cycle Evaluation, *Advanced Fuel Cycle and Reactor Concepts*. Report of INFCE Working Group 8; IAEA, Vienna, 1980. STI/PUB/534. ISBN 92-0-159880-7.
- [6] J.L. Kloosterman and J.M. Li, *Transmutation of Tc-99 and I-129 in Fission Reactors*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--95-002 (1995).
- [7] J.L. Kloosterman, *New Working Libraries for Transmutations Studies*, Proc. Global '93 Conference on Future Nuclear Systems, Emerging Fuel Cycles and Waste Disposal Options, Seattle, Washington, USA, 12-17 September 1993, pp. 1229-1236.
- [8] A.J. Janssen, *Transmutation of Fission Products in Reactors and Accelerator-Driven Systems. Some Critical Remarks*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--94-001 (1994).
- [9] A.R. Dastur, Private Communication of 24 June 1993.
- [10] Data Base for a CANDU-PHW Operating on a Once-Through Natural Uranium Cycle, draft submission to INFCE Working Group 8: Advanced Fuel Cycle and Reactor Concepts. INFCE/WG.8/CAN/DOC 2, 1978 October, p. 175.
- [11] M. Srinivasan, K. Subba Rao, M.V. Dingankar, *The "Actinide Waste" Problem in Perspective*, waar????
- [12] V.A. Wichers, J.M. Li, T.T.J.M. Peeters, *Computational Benchmark on Plutonium Recycling in PWRs. ECN solution of OECD WPPR Benchmark*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--94-021 (1994).

- [13] R.J.M. Konings, P.J.A.M Blankenvoorde, E.H.P. Cordfunke, K. Bakker, *Evaluation of Thorium Based Nuclear Fuel: Chemical Aspects*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--95-007, 1995.
- [14] International Nuclear Fuel Cycle Evaluation, *Waste Management and Disposal*, Report of INFCE Working Group 7, IAEA, Vienna, 1980. STI/PUB/534. ISBN 92-0-159780-0.
- [15] International Atomic Energy Agency, *Uranium Extraction Technology*, IAEA, Vienna, 1993, STI/DOC/10/359, ISBN 92-0-103593-4.
- [16] J.L. Kloosterman, *Incentives for Transmutation of Americium in Thermal Reactors*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--94-022 (1994).
- [17] J.H. Bultman, W.J.M. de Kruijf, *Evaluation of Thorium Based Nuclear Fuel: Reactorphysics*, Petten, Netherlands Energy Research Foundation (ECN), ECN-R--95-006 (1995).