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MINOR ACTINIDE TRANSMUTATION IN THORIUM AND URANIUM MATRICES IN HEAVY WATER MODERATED REACTORS

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The irradiation of Th-232 breeds fewer of the problematic minor actinides (Np, Am, Cm) than the irradiation of U-238. This characteristic makes thorium an attractive potential matrix for the transmutation of these minor actinides, as these species can be transmuted without the creation of new actinides as is the case with a uranium fuel matrix. Minor actinides are the main contributors to long term decay heat and radiotoxicity of spent fuel, so reducing their concentration can greatly increase the capacity of a long term deep geological repository. Mixing minor actinides with thorium, three times more common in the Earth's crust than natural uranium, has the additional advantage of improving the sustainability of the fuel cycle.

In this work, lattice cell calculations have been performed to determine the results of transmuting minor actinides from light water reactor spent fuel in a thorium matrix. 15-year-cooled group-extracted transuranic elements (Np, Pu, Am, Cm) from light water reactor (LWR) spent fuel were used as the fissile component in a thorium-based fuel in a heavy water moderated reactor (HWR). The minor actinide (MA) transmutation rates, spent fuel activity, decay heat and radiotoxicity, are compared with those obtained when the MA were mixed instead with natural uranium and taken to the same burnup. Each bundle contained a central pin containing a burnable neutron absorber whose initial concentration was adjusted to have the same reactivity response (in units of the delayed neutron fraction β) for coolant voiding as standard NU fuel.

I. INTRODUCTION

Minor actinides (MA) can be disposed of by mixing them with uranium based fuels and re-irradiating them in a heavy water reactor (HWR), but this will cause more problematic minor actinides to be bred from U-238, reducing the efficiency of the process. An alternative approach is to transmute minor actinides in a thorium based reactor fuel, since irradiation of Th-232 produces negligible amounts of the problematic MA. Thorium is fertile, since it breeds the fissile nuclide U-233 during irradiation, but the use of thorium as a reactor fuel requires an initial fissile component to support the nuclear reaction until sufficient quantities of U-233 can be made. If one extracts all the transuranic (TRU) elements (Pu, Np, Am, Cm) from spent light water reactor (LWR) fuel to mix with thorium in an HWR fuel bundle, the fissile isotopes (primarily Pu-239) will serve this purpose.

This study compares burning TRU elements in a uranium matrix versus burning them in a thorium matrix; HWRs are used in both cases. The HWR used in this study is a pressure tube reactor cooled and moderated with heavy water. The fuel mixture is composed of transuranic dioxide ($TRUO_2$), which consists of 15 year cooled group-extracted transuranic elements homogeneously mixed with either a thorium dioxide (ThO_2) matrix or a (natural) uranium dioxide (UO_2) matrix. In both cases, the fuel bundle contained a burnable neutron poison in the central element whose levels were adjusted to reduce the coolant void reactivity (CVR) of the bundle.

II. CALCULATION METHODS

II.A Codes Used

The lattice cell calculations for this study were performed using WIMS-AECL v.3.1.2.1 [1] with an ENDF/B-VII based library[2].

WIMS Utilities version 2.0.3 [3] was used to process WIMS-AECL output and calculate delayed neutron fractions as a function of irradiation.

The LWR TRU composition was estimated by running a pincell simulation of an LWR fuel bundle using the code WOBI [4]. WOBI alternates between calls to: 1) WIMS-AECL to produce a neutron flux solution in the unit cell and absorption, (n, 2n), and fission rates for many major nuclides, 2) LINK16 [4], which merges the WIMS-AECL reaction rates with scale.rev16.xn44, a large AMPX format library containing many nuclides and reactions, 3) COUPLE, which takes the merged library and updates an ORIGEN library with the reaction rates so calculated, and 4) ORIGEN, a widely used depletion code. scale.rev16.xn44, COUPLE and ORIGEN are part of the SCALE 5.1 suite of nuclear analysis codes [5].

II.B. Fuel Bundle Concept

The fuel bundle used for this study is a 54-element concept (Fig. 1). Because coolant voiding generally results in an increase in the neutron flux in the central region of the bundle, the CVR can be reduced by inserting

a neutron poison in the centre, and this is the purpose of the large central pin shown. Other lines in Fig. 1 (e.g. the lines bisecting the fuel pins) divide the assembly into different regions for calculation purposes. Previous sensitivity analyses have shown that the sub-division of the pins in the assembly (Fig. 1) is robust. The results do not vary by more than 2% for any reaction rate compared to finer discretizations for this bundle. Table I lists the specifications for the bundle concept and the HWR.

TABLE I: HWR and Bundle Specifications

Quantity	Value, Units
# of fuel channels	380
# bundles per channel	12
Length of Bundle	49.5 cm
Centre Pin Radius	1.619 cm
Fuel Pin Radius	0.475 cm
Cladding Thickness	0.0325 cm
Natural UO_2 Density	10.60 g/cm^3* [6]
ThO_2 Density	9.67 g/cm^3* [6]
$TRUO_2$ Density	11.112 g/cm^3** [6]
Dysprosia Density	7.81 g/cm^3* [6]
Zirconia Density	5.68 g/cm^3* [6]
Lattice pitch (square)	28.575 cm
Moderator D_2O purity	99.8 wt%D_2O, ~342.15 K
Pressure Tube (PT)	Zr-2.5Nb
Calandria Tube (CT)	Zr-2
Average D_2O Coolant	99 at% D_2O , ~561.15 K
Cladding Material	Zr-4

* Densities are adjusted (smeared) values to account for modelling of a 3D reactor in 2D.

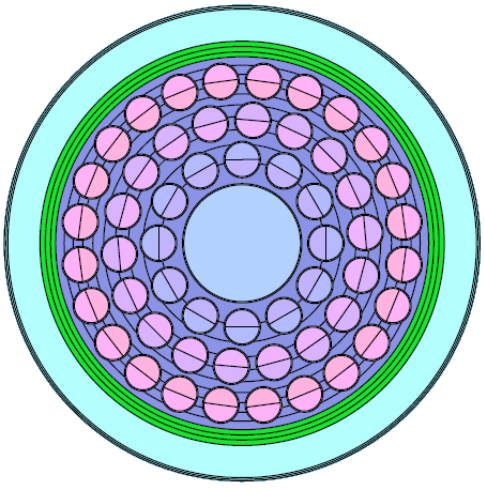

Fig.1. 54-element bundle concept. The lines show the discretization of the fuel pins and moderator.

II.C. Fuel Material Properties

The fuel bundle contains elements of two types of materials: the fuel and a neutron absorbing material. The neutron-absorbing pin in the centre contains a mixture of dysprosia and zirconia. The fuel pins surrounding the centre pin contains a mixture of ThO_2 and $TRUO_2$ or a mixture of UO_2 and $TRUO_2$. The $TRUO_2$ composition, assuming all elements formed a dioxide of the LWR TRU is shown in Table II; the density of $TRUO_2$ was taken to be the smeared density of plutonium (IV) oxide. Note, after 15 years the original amount of Cm-242 is negligible and the reported amount of Cm-242 in Table II is due to the ongoing decay of Am-242m.

II.B.1. Fuel Temperature Calculation for Thorium Matrix

The fuel temperature was calculated using the linear element rating (LER) in an iterative process. Solving Fourier's heat flux transport equation for a cylindrical system with a uniform heat production throughout, and a fixed temperature boundary, yields an average temperature given by:

$$T_{fuel} = \frac{1}{R}\int_0^R \frac{LER}{4\pi k}\left(1-\left(\frac{r}{R}\right)^2\right)dr + T_{coolant} \qquad (1)$$

The value for the thermal conductivity k was estimated, based on the thermal conductivity of a thorium-plutonium mixed oxide (92%Th, 8%Pu)O_2, as this closely resembles the initial composition of the fuel, to be 3.64 W m^{-1} K^{-1} [7] and was held constant through the iteration steps. With a fixed coolant temperature of 561.15 K, and an assumed constant value of k, equation (1) can be written as:

$$T_{fuel} = \frac{2LER}{12\pi k} + T_{coolant} \qquad (2)$$

TABLE II: HWR TRU Input Fuel Composition (15-year-cooled)

Nuclide	Weight %
O-16	11.774
Np-237	4.718
Pu-238	2.116
Pu-239	42.267
Pu-240	18.921
Pu-241	5.988
Pu-242	5.775
Am-241	6.686
Am-242m	8.03 x10^{-3}
Am-243	1.346
Cm-242	3.21x10^{-5}
Cm-243	4.24x10^{-3}
Cm-244	3.47x10^{-1}
Cm-245	4.23x10^{-2}
Cm-246	5.21x10^{-3}
Cm-247	8.82x10^{-5}

Nominal initial temperatures, and an assumed bundle power of 840 kW (= 17,500 kW/m) provided initial LERs for each ring of pins (from WIMS-AECL 3.1.2.1, for fresh fuel) which were used to calculate temperatures using (2). The iteration converged rapidly and the final LERs and temperatures were found to be 9.90 kW/m, 18.69 kW/m, and 51.70 kW/m, 705.4 K, 833.6 K, and 1314.7 K, for the individual pins in the inner, intermediate and outer rings, respectively. The peak LER of between 50 to 60 kW/m is known to be acceptable for uranium oxide fuels [8].

II.B.2. Fuel Temperature Calculation for Uranium Matrix

Similar to the calculation process mentioned for the thorium matrix, the LERs for the uranium matrix were calculated. The fuel temperatures were iterated using equation (2), with an assumed constant thermal conductivity of 4.96 W m^{-1} K^{-1} for uranium dioxide [9].

The LERs were found to be 12.21 kW/m, 22.18 kW/m, and 50.18 kW/m for the inner, intermediate and outer rings respectively. These corresponded to fuel temperatures of 692.0 K, 798.9 K, and 1098.9 K.

II.C. Fuel Burnup

A steady bundle power of 840 kW was assumed for a single bundle. During the simulation, WIMS-AECL automatically calculates the 'time-integrated k_∞' an estimate of the average neutron multiplication of an infinite array of bundles having a distribution from zero to the current burnup. When this value falls to 1.03, the 'exit burnup' is reached and the bundle assumed to be removed from the reactor. This calculation assumes that the reactor as a whole has 3% leakage (from edges, or into flux shape control absorbers) which is a typical value for HWRs. A burnup target of 45 MWd/kg initial heavy elements (IHE) was chosen as it is consistent with the burnups for current LWRs [10] (pressure water reactors specifically). This target was reached by adjusting the fuel composition.

II.D. Delayed Neutron Fraction

The KINPAR module of WIMS Utilities was used to generate the delayed neutron fraction as a function of irradiation. The calculation was performed with 6 delayed neutron precursor groups, but no photoneutron groups.

The delayed neutron fraction for each particular burnup condition are averaged together by a process called 'burnup weighting', which effectively weights the calculated delayed neutron fraction for each burnup step by both the power of the bundle (constant in this case) and time length of the step.

II.E. Target Coolant Void Reactivity

The change in the geometry and design of the bundle, as well as a change in the fuel composition, lead to a change in the reactor kinetics. The fissioning of plutonium produces fewer delayed neutrons. A lower value of the delayed neutron fraction β increases the rate at which the reactor power changes in response to a reactivity increase. To recover the same rate of power rise during a coolant-void incident, the CVR is adjusted in proportion to β.

The burnup-weighted CVR from a base case of a natural uranium fuelled HWR reactor is ~14.4 mk for 100% voiding (1 mk = 1 pcm). From this value, a target CVR for the TRU/thorium matrix simulation was determined to be 9.97 mk, and to be 11.00 mk for the TRU/uranium matrix simulation.

II.F. Decay Calculations

WOBI was used to decay the LWR fuel after the in-reactor simulation to estimate its thermal power and activity after long periods.

In order to compare the effect of re-irradiation of the TRU on the total thermal power of all waste materials, the following procedure was followed. First, the spent fuel composition was obtained from WOBI. Next, the TRU elements were manually removed and the remaining elements were input into ORIGEN-ARP [11]. (ORIGEN-ARP was used for this decay step, rather than WOBI, because a number of fission products produced during irradiation were not present in the SCALE 5.1 standard composition library used by WOBI .)

III. RESULTS

Table III shows a summary of the results of the simulated burnups for the TRU/thorium and TRU/uranium cases, with % Dy_2O_3 in centre pin adjusted to achieve the target CVR (within 0.5%) and irradiation time adjusted to achieve the target burnup (within 0.5%).

TABLE III: Summary of Thorium and Uranium Fuel Burnup and Fuel Characteristics

	Thorium fuel	Uranium fuel
Burnup (MWd/kg)	45.1	44.8
CVR (mk)	9.92	11.00
% $TRUO_2$	7.45	4.20
% Dy_2O_3 (centre)	2.4	1.6
Maximum LER (kW/m)	51.7	50.2
Irradiation time (days)	833	902
TRU throughput (kg/year)	2.69×10^3	1.44×10^3

III.A. Thorium Matrix

The absolute and relative transmutation rates for all transuranics were determined and are summarized in Table IV. In this table, negative rates indicate destruction of the nuclide, while positive rates indicate creation.

For the transuranics as a whole, 44% of the initial mass is fissioned, corresponding to a transmutation rate of 1171 kg/reactor/year. For the minor actinides specifically, 69% (140 kg/year) of the initial Am-241 and 42% (60 kg/year) of the initial Np-237 is transmuted. Against that, 36 kg/year of Am-243 is produced and 57 kg/year of total curium. However, the principal curium nuclides produced are even numbered ones (Cm-242 and Cm-244), both of which have short half lives and can be allowed to decay in place before handling or placement in a repository.

III.A.1.Production of Pa-233 and U-233

Achieving an exit burnup of 45 MWd/kg relies on the breeding of U-233 from Th-232 to sustain a nuclear chain reaction once the initial fissile isotopes are depleted. Through neutron capture, Th-232 becomes Pa-233, which subsequently beta decays to U-233 with a half life of 26.975 days. Fig. 2 shows the production of Pa-233 and U-233, as well as the sum of the production of both nuclides. Pa-233 and U-233 both approach their equilibrium values by the end of the irradiation. The total rate of production of Pa-233 and U-233 is 358.42 kg/reactor/year.

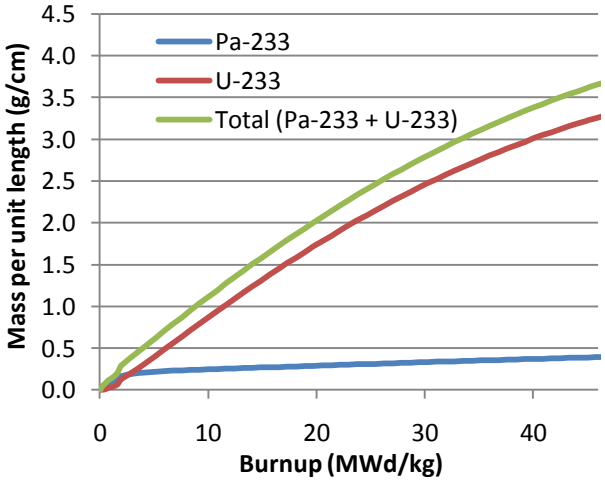

Fig. 2. Amount of Pa-233 and U-233 and their sum as a function of burnup

TABLE IV: Rate of Transmutation for Thorium and Uranium fuel

	Mass Increase (% of initial fuel value)		Rate of Mass Increase (kg/reactor/year)	
	In Thorium Fuel	In Uranium Fuel	In Thorium Fuel	In Uranium Fuel
Np-237	-41.7	-53.8	-60.0	-41.3
Total Np	-41.5	-49.3	-59.8	-37.9
Pu-238	72.3	41.3	46.7	14.2
Pu-239	-81.4	-78.9	-1050.7	-541.6
Pu-240	-15.1	-15.6	-87.1	-48.1
Pu-241	-26.8	-29.5	-48.9	-28.7
Pu-242	42.2	79.5	74.5	74.6
Total Pu	-46.5	-43.4	-1065.5	-529.6
Am-241	-68.6	-88.3	-140.0	-95.9
Am-242m	164.9	-14.1	0.4	0.0
Am-243	88.6	135.2	36.4	29.6
Total Am	-41.8	-50.8	-102.8	-66.3
Cm-242	*very high*	*very high*	25.7	13.1
Cm-243	636.7	637.7	0.8	0.4
Cm-244	268.3	507.1	28.5	28.7
Cm-245	65.3	25.8	0.8	0.2
Cm-246	526.6	816.9	0.8	0.7
Cm-247	*very high*	*very high*	*neg.*	*neg.*
Cm-248	*very high*	*very high.*	*neg.*	*neg.*
Total Cm	465.3	663.60	56.7	43.1
Total MA	-26.3	-28.6	-105.9	-61.1
Total TRU	-43.5	-41.2	-1171.4	-590.6

III.A.2.Supporting Reactors

The support ratio is defined as the power production in LWRs divided by the power production in HWRs for an equilibrium two-reactor system where all the LWR TRU is passed to the HWRs.

The spent nuclear fuel production rate from the Takahama-3 reactor with the fuel assembly SF97-4 [10], which has a thermal power capacity of 2652 MWth and a burnup of 47.02 MWd/kg, was determined to be approximately 20.6 MT/reactor/year. Of the 20.6 metric tons, approximately 1.32% is TRU. Assuming 100% efficiency of the extraction of all TRU from the spent nuclear fuel, the total exit mass of TRU from LWR spent fuel is 271 kg/reactor/year. The annual requirement of TRU for the HWR is 2696 kg/reactor/year.

The Takahama-3 reactor has a net electric output of 830 MWe, and typical HWRs have a net electric output of 725 MWe. Using the specified outputs of the respective reactors the support ratio is 11.4 (GWe LWR per GWe HWR).

III.B. Uranium Matrix

The transmutation rate and percent-of-initial transmuted values determined are summarized in Table IV. Transmutation rates in terms of percent-of-initial mass are largely similar between the two fuel types, but lower in the uranium based fuel (41% total TRU mass transmuted vs. 44% in the thorium matrix). The absolute throughput of TRU is significantly lower in the uranium based fuel (591 kg/year vs. 1171 kg/year) because of the considerably lower initial loading of TRU. This lower initial loading reflects a number of factors, but the lower absorption cross section of U-238 relative to thorium and the presence of fissile U-235 in natural uranium are the most significant.

III.B.1.Supporting Reactors

For the uranium matrix, the initial TRU content in the fuel was 4.2 vol.%; this translates to a consumption of 1435 kg/year of TRU in a HWR with a uranium matrix. Using a similar process outlined in Section III.A.2, the supporting ratio for this case is 6.1 GWe of LWR power per GWe of HWR power.

III.C. Spent Fuel Characteristics

The characteristics of the spent fuel examined were: decay heat (W/kg), radioactivity (Bq/kg) and the radiotoxicity (Sv/kg). The measure of radiotoxicity used was the committed effective dose (CED) per kg integrated over 50 years, denoted $\tilde{E}(50)$, a quantity which includes a biological coefficient for each nuclide which takes into account uptake and residence time in body organs. Other assumptions in this calculation were: exposure by inhalation to 5 μm fuel particles and 'S type' solubility for all nuclides, corresponding to slow release (from the fuel particle matrix).

The fission products and actinides were decayed for 1 million years and compared to a reference case. The reference case assumes the spent nuclear fuel from a LWR is not reprocessed and is sent directly for long-term disposal. The recycled cases assume that the spent nuclear fuel is reprocessed, the TRU is extracted with 100% efficiency, and the remainder of the fuel is prepared for storage. The decay heat, radioactivity and CED/kg of the remaining LWR fuel (uranium and fission products) was added to that from the spent nuclear fuel obtained from the HWR, burning the TRU elements in a uranium or thorium matrix. The overall throughput of fuel in the reactor park scenarios was determined using the support ratios from Sections III.A.2 and III.B.1, normalized to 1 GWe total power for the reactor park, and compared to the reference LWR-only case with the same electrical power output.

The results are shown in Fig. 3 (activity), Fig. 4 (thermal power) and Fig. 5 (radiotoxicity) for both recycling cases vs. the reference case. For fission products, activity and radiotoxicity are suppressed for both the TRU/thorium and TRU/uranium cases (see Fig. 6 and Fig 8) relative to the reference case for a few hundred years, but become very similar to the reference case after that time. For both TRU cases, fission product radiotoxicity increases above the reference case towards $1x10^6$ years due to the enhanced production of such long-lived isotopes as Zr-93/Nb-93m (via U-233 fission in thorium matrix fuels) and Cs-135 (via Pu-239 fission in both TRU fuels). However, fission products are relatively small contributors to the total radiotoxicity over the entire $1x10^1$ to $1x10^6$ range of interest, so this increase has few consequences.

For the actinides there is a larger and longer term difference between the recycling and reference cases. Approximately 50% of the actinide activity, decay heat and toxicity has been suppressed over much of the decay period (see Fig. 6, 7, 8). However, decay heat and toxicity are equal, or slightly higher for times < 100 years, when both of these quantities (for actinides) are dominated by Pu-238 ($t_{½}$ = 87.7 years). Since excess Pu-238 is produced in both the TRU/thorium and TRU/uranium burning scenarios, the otherwise large reduction seen in overall decay heat and toxicity is smaller until it decays.

Between $1x10^4$ and $1x10^6$ years the activity, decay heat and toxicity of the TRU/thorium matrix case rises above the values of the reference case. This is due to the decay of U-233 ($t_{½}$ = 159,200 years) in the spent fuel. A similar feature occurs in the TRU/uranium matrix fuel, but is due to the decay of U-234 ($t_{½}$ = 246,000 years) created by the earlier decay of the Pu-238. U-234 in natural uranium is present in equilibrium with the decay

of U-238 (U-238 → α + Th-234 → β + Pa-234 → β + U-234). During irradiation, it is initially reduced by neutron absorption, but long irradiation produces it by the α decay chain Cm-242 → α + Pu-238 → α + U-234. In the TRU/uranium matrix case, this excess U-234 content does not result in the overall activity, decay heat or toxicity rising above that of the reference case. On both cases, by $1x10^6$ years, activity, decay heat, and toxicity are very similar for the TRU/uranium and TRU/thorium cases and well below the values in the reference scenario.

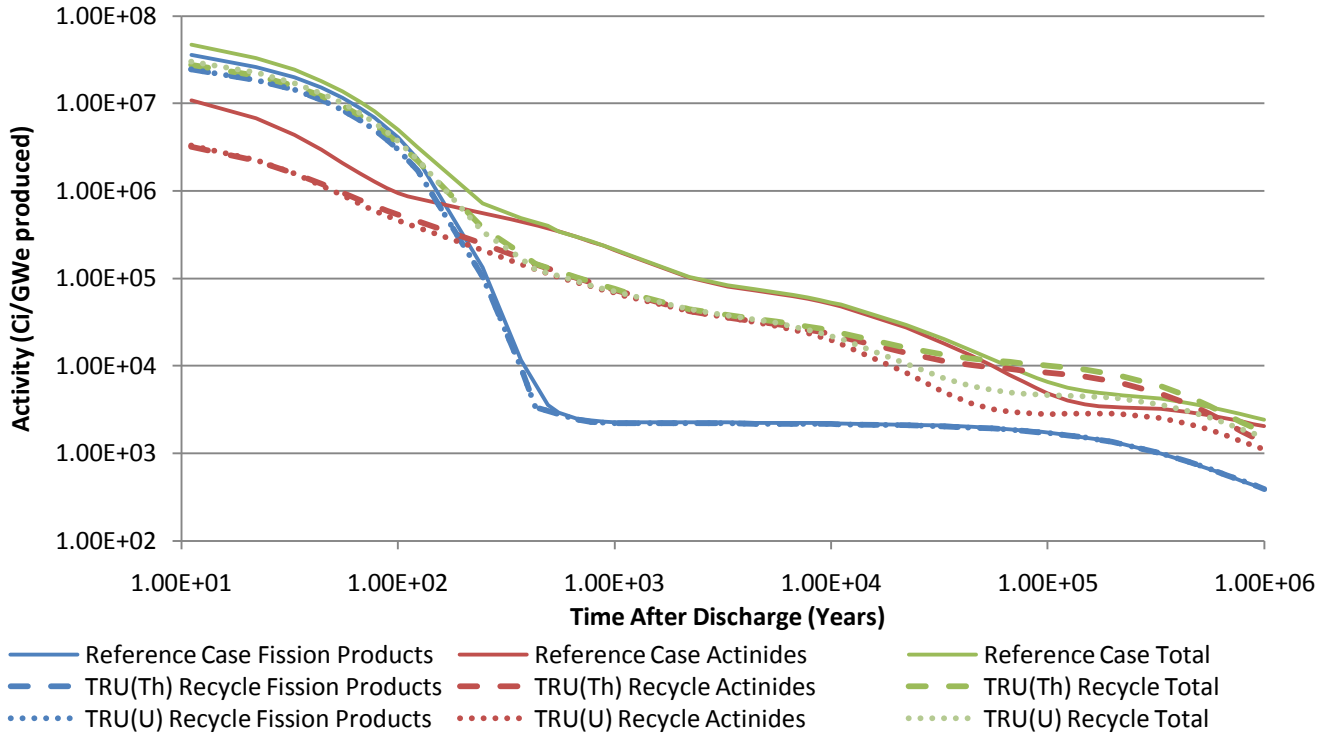

Fig. 3. Radioactivity as a function of time for the reference case and TRU recycled in to thorium and uranium fuel

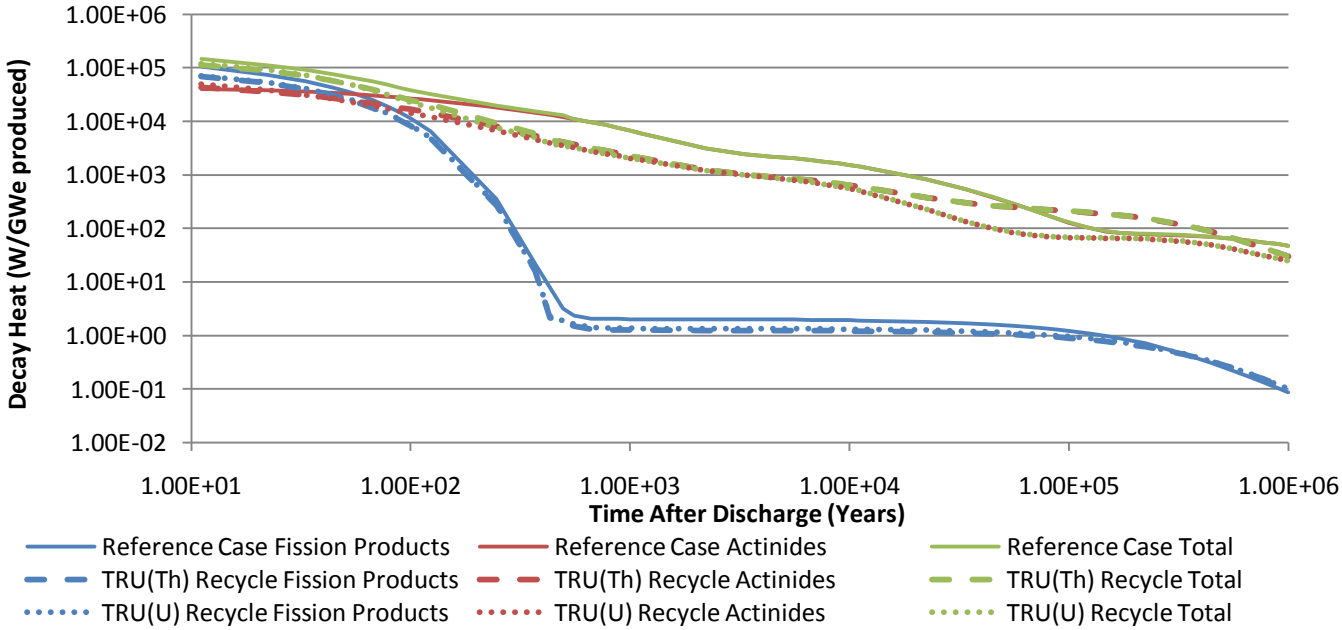

Fig. 4. Decay heat as a function of time for the reference case and TRU recycled in to thorium and uranium fuel

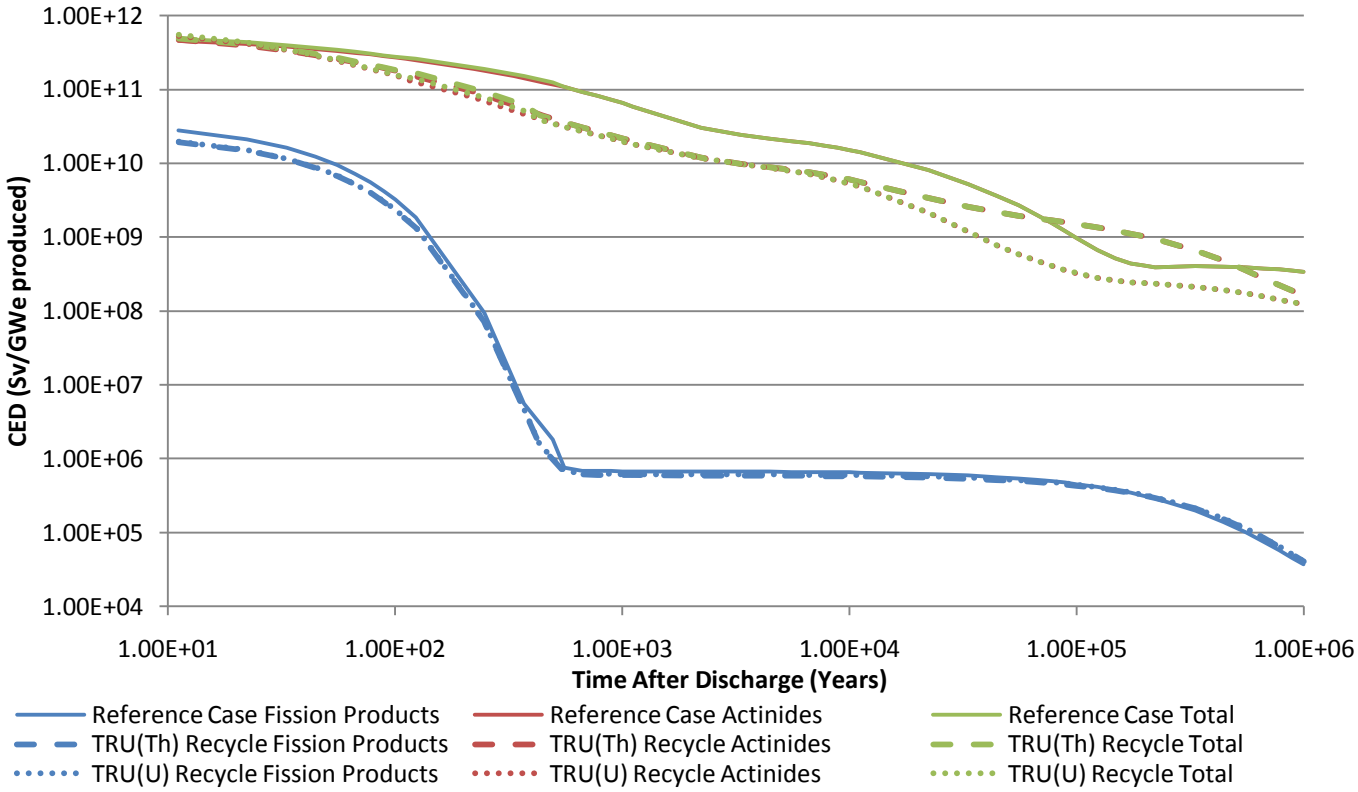

Fig. 5. CED as a function of time for the reference case and TRU recycled in to thorium and uranium fuel

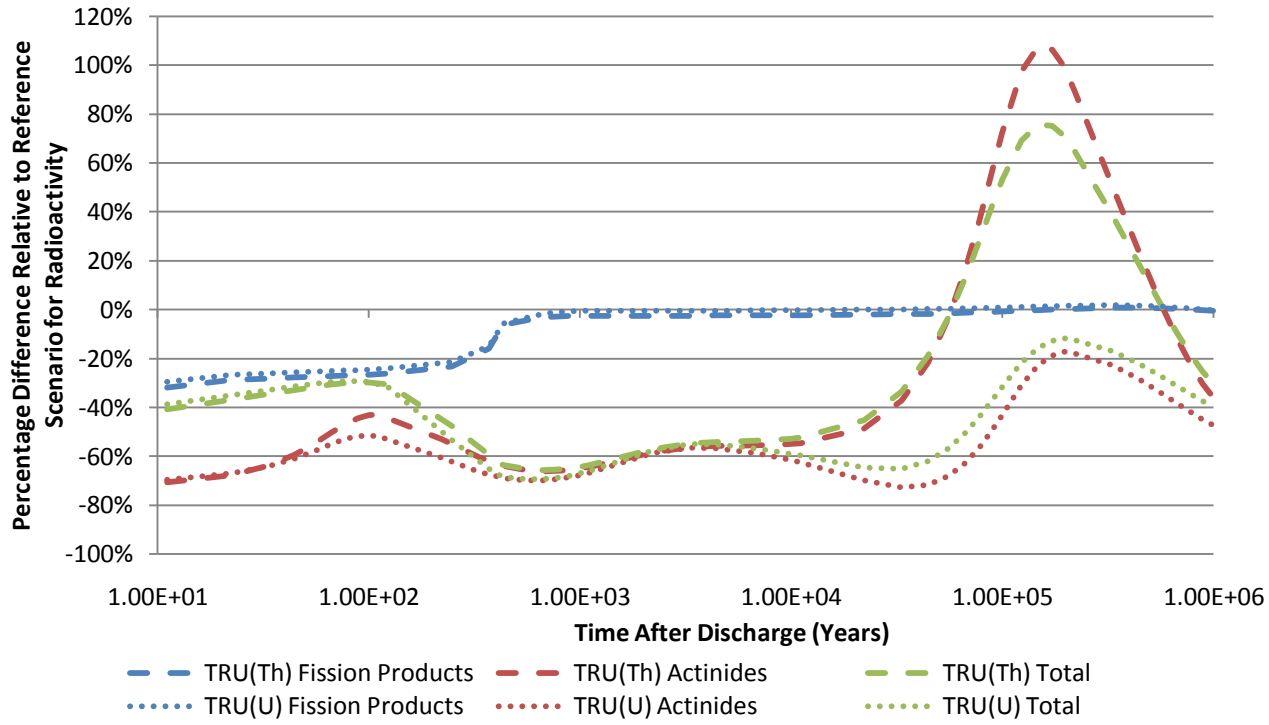

Fig. 6. The percentage difference of radioactivity for the recycled cases relative to the reference case as a function of time

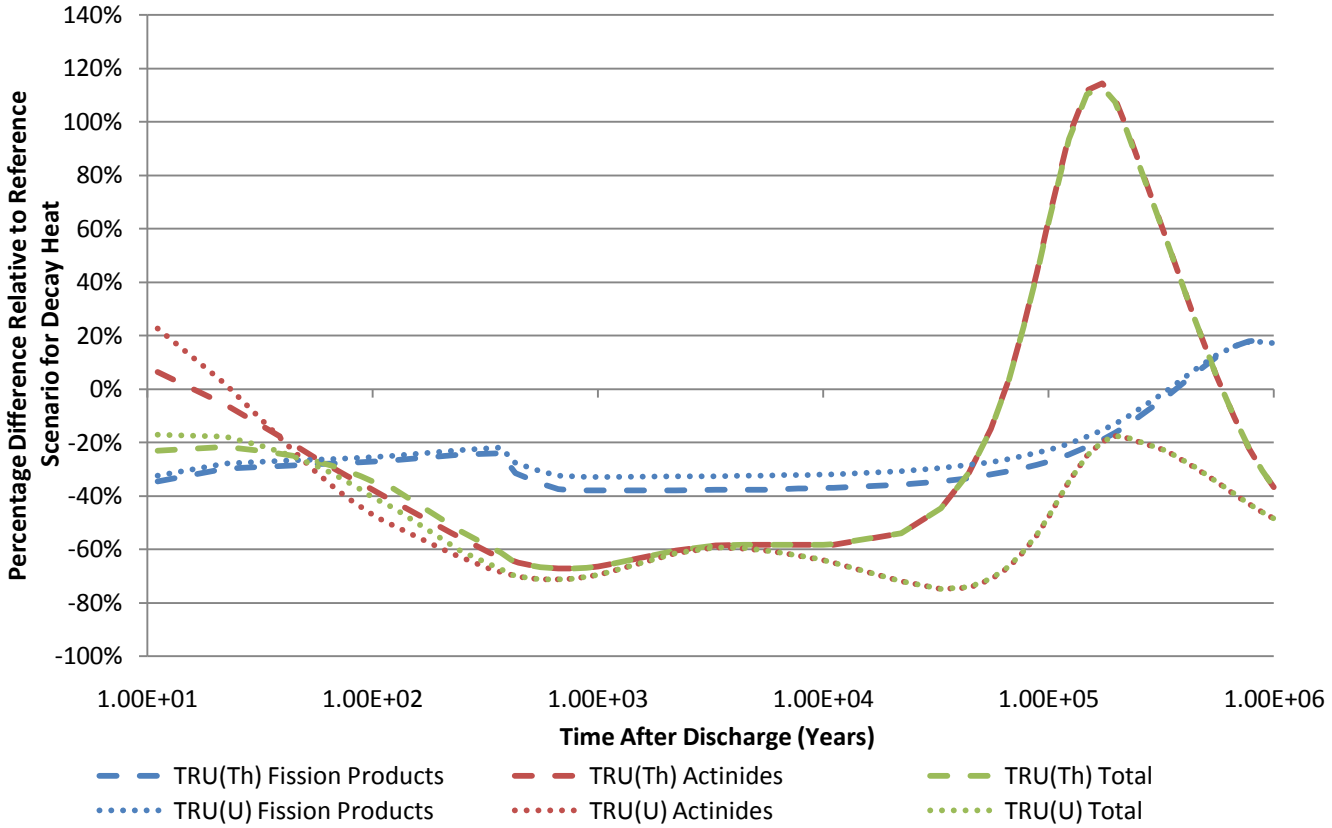

Fig. 7. The percentage difference of the decay heat for the recycled cases relative to the reference case as a function of time

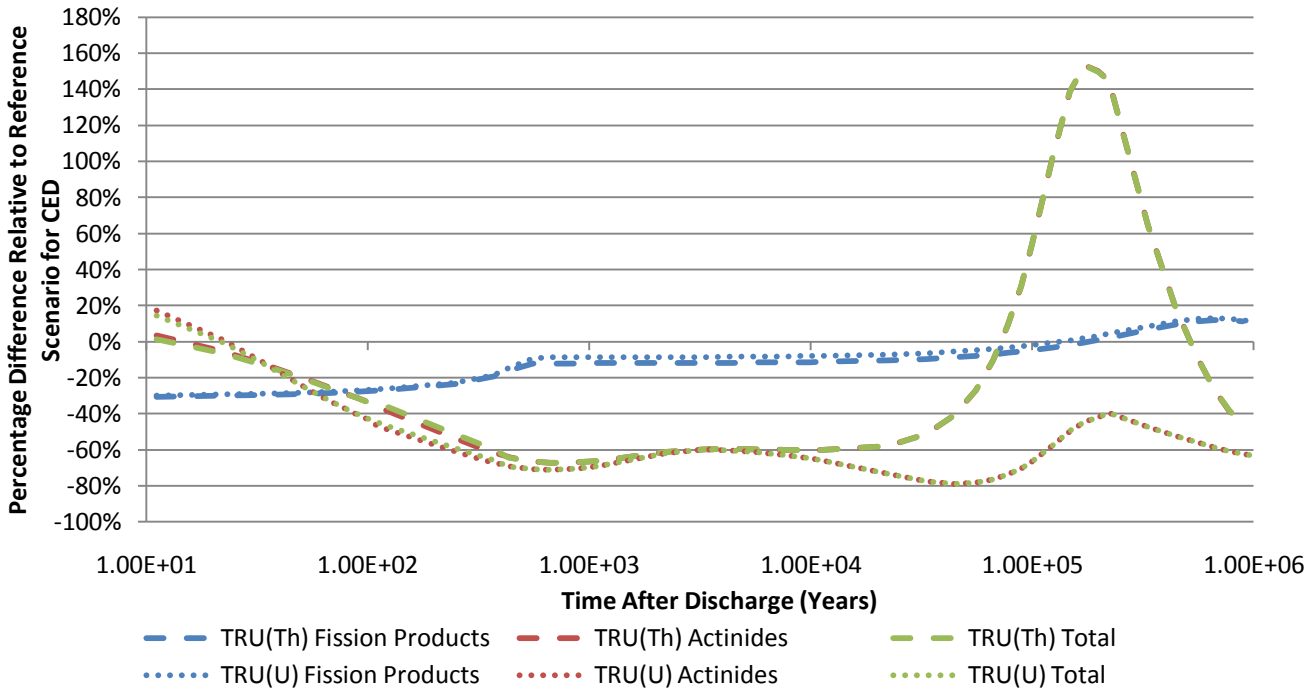

Fig. 8. The percentage difference of the CED for the recycled cases relative to the reference case as a function of time

IV. SUMMARY AND CONCLUSIONS

Transmuting minor actinides from LWR spent fuel in thorium fuel and uranium fuel matrices in HWRs of current design has been examined in this study. The scope of this study was limited to the physics of the lattice cell calculations. A 54-element fuel bundle concept was used with a large centre pin containing a neutron poison to lower the CVR for safety considerations.

Thorium is four times more abundant than uranium, and breeds the valuable fissile nuclide U-233. This is similar in concept to breeding Pu-239 from uranium based fuels to increase the total energy extracted. Because few MA are created by irradiated thorium, it has an advantage as a matrix in which to reduce the total mass of MA from the LWR fuel cycle. In this case, the LWR:HWR support ratio was found to be 11.4:1 and 6.1:1 for TRU/thorium and TRU/natural uranium fuel mixtures, respectively, in a HWR. However, it is observed that in both cases a relatively small number of HWRs is required to deal with the TRU output of LWRs.

Total spent fuel characteristics in the two recycling cases (adding in the LWR fission product and uranium waste not recycled into HWRs) were equal, or better in the two recycling cases for three important spent fuel characteristics: total activity, decay heat, and committed effective dose (radiotoxicity) except in two time periods. The first time period is the first two to three decades of decay, when Pu-238, having high specific decay heat and toxicity, and which is increased relative to the reference case in both recycling scenarios, is most important. The second time period is around 100,000 years, when the U-233 produced in irradiated thorium starts to dominate the fuel activity, leading to an overall increase in decay heat and radiotoxicity in the TRU/thorium case. By $1x10^6$ years the total activity, decay heat and radiotoxicity of the spent fuel for both the TRU cases is less than that of the reference case.

Long-lived fission products such as Zr-93 and Cs-135, created preferentially in the TRU fuels, increase the decay heat and radiotoxicity of these spent fuels towards and beyond $1x10^6$ years of decay, but the actinide components dominate these quantities at this time, and the fission product increase will have little practical effect.

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