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VHTR, ADS, and PWR Spent Nuclear Fuel Analysis

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Abstract

The aim of this study is to analyze and compare the discharged-spent fuel of three types of nuclear systems: a Very High-Temperature Gas Reactor (VHTR), a lead-cooled Accelerator-Driven System (ADS) and a standard Pressurized Water Reactor (PWR). The two first systems, VHTR, and ADS were designed to use reprocessed fuels. UREX+ and GANEX techniques were used for the reprocessing processes respectively. The fuel burnup simulated for the systems in other works have been used to obtain the final composition of the spent fuel discharged. After discharge, the radioactivity, the radiotoxicity, and the decay heat were evaluated through the ORIGEN 2.1 code until 10^7 years and compared to the literature.

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Keywords: Reactors VHTR, ADS, and PWR; reprocessing fuel; nuclear fuel cycle; SCALE 6.0; MONTEBURNS and ORIGEN 2.1

1. Introduction

One of the major nuclear problems is the spent nuclear waste (SNF) produced by the neutron irradiation of uranium. Because the neutron radiative capture of uranium produces minor actinides and plutonium isotopes, which

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pose long-term radioactivity and high radiotoxicity. Therefore, to reduce the amount SNF going to the final disposal, reprocessing techniques have been proposed to recover the uranium and plutonium that can be reused in the nuclear power plant. Some techniques such as GANEX and UREX+ [1,2]also recover the minor actinides and the plutonium into the matrix due to non-proliferation issues. It is known that the minor actinides have a high fission cross section for high-energy neutrons, such as, the ones produced in fast reactors or ADS which can transmute them reducing the long-term final disposal of the minor actinides (MA). The main goal is to evaluate and compare through ORIGEN 2.1 code [3]the differences in the SNF of each fuel (reprocessed fuels and UO_2) after irradiation in different reactors VHTR[4], ADS[5] and PWR[6] analyzing the radioactivity, radiotoxicity, and the decay heat after irradiation in these reactors. The burnup conditions and the fuels used for each reactor were different, which make each of them unique. Therefore, this work would help to understand the canister design needs for a final repository.

2. Methodology

The SNF matrix was obtained from three different reactors with different burnup features: PWR,VHTR, and ADS. After discharge, parameters such as the composition evolution, the radioactivity, the radiotoxicity, and the decay heat have been evaluated through the ORIGEN 2.1 code until 10^7 years. Some characteristic of the fuel and conditions of burnup are described below. More details can be obtained in the references.

The reprocessed fuels used in ADS and VHTR were GANEX and UREX+ methods respectively. Both of them were obtained from the spent fuel of a standard PWR fuel, which had been submitted to a burnup of 33 GWd/tHM during three years and then left for five years in the pool[4,5]. After that, the spent fuel had reprocessed by GANEX or UREX+ method depending on the reactor used.

2.1. Pressurized Water Reactor (PWR)

The SNF data for the PWR was obtained from the ORNL/TM-6051 report[6]. The PWR conditions were the fuel was UO_2 enriched to 3.2% submitted to a burnup of 33000 MWd/tHM during three years and thermal power of 3800 MW_t.

2.2. Very High-Temperature Gas Reactor (VHTR)

The SNF data for the VHTR comes from a burnup of 97.80 GWd/tHM with a thermal power of 600 MW_t during three years, simulated through the TRITON6[7] module (SCALE 6.0). It used reprocessed fuel by UREX+ process spiked with thorium, containing 15% of fissile material[4].

2.3. Accelerator Driven Subcritical Reactor System (ADS)

The SNF data for the ADS comes from a burnup of 237.6 GWd/tHM with a thermal power of 515 MW_t during 20 years. It was simulated in MONTEBURNS (MCNP/ORIGEN2.1) code[8]. This system used reprocessed fuel spiked with thorium with 12% of fissile material[5].

Table 1 shows a summary of the main characteristics related to the studied systems and Table 2 presents the initial and final compositions after the burnup.

3. Results

3.1. Radioactivity

Figure 1 shows the SNF radioactivity for each reactor PWR, VHTR, and ADS during 10^7 years. The highest radioactivity for the reactors with reprocessed fuel is due to the higher concentrations of plutonium isotopes on the reprocessing fuel, which are much lower amounts on the PWR. The SNF for the ADS and VHTR have higher radioactivity than the one for PWR SNF. The VHTR radioactivity's begins lower than the ADS one. Nevertheless, after 1000 years due to the plutonium contribution the radioactivity for the VHTR overcomes the one for the ADS

during 5000 years. Then, it drops off ADS again until 10^7 years. After ten million years, all spent fuels show almost similar radioactivity values. Figures 2 show the contribution of the actinides and fission products, in which can be seen that the fission products domain the radioactivity from the three SNF the first 100 years.

Table 1. The main characteristics of the reactors.

Reactor Type	PWR	VHTR	ADS
Initial enrichment or fissile material (%)	3.2	15	12
Power (MW_t)	3800	600	515
Burnup (GWd/tHM)	33	97.80	237.6

Table 2. Concentrations of the principal actinides in the irradiated fuel.

Reactor	PWR		VHTR		ADS	
	Concentrations (g)					
Nuclide	Load	Disch.	Load	Disch.	Load	Disch.
^{232}Th	-	$2.13x10^{-4}$	$7.60x10^{5}$	$7.49x10^{5}$	$8.20x10^{5}$	$6.88x10^{5}$
^{233}U	-	$1.43x10^{-3}$	-	$3.16x10^{4}$	$3.78x10^{-6}$	$7.66x10^{4}$
^{234}U	$2.90x10^{2}$	$1.85x10^{2}$	-	$6.76x10^{3}$	$2.82x10^{-1}$	$1.26x10^{4}$
^{235}U	$3.2x10^{4}$	$8.19x10^{3}$	-	$1.45x10^{3}$	$1.47x10^{1}$	$1.81x10^{3}$
^{236}U	-	$4.07x10^{3}$	-	$1.21x10^{2}$	$7.51x10^{0}$	$3.47x10^{2}$
^{238}U	$9.7x10^{5}$	$9.70x10^{5}$	-	$1.28x10^{-1}$	$1.79x10^{3}$	$1.44x10^{3}$
^{237}Np	-	$4.43x10^{2}$	$7.91x10^{3}$	$3.90x10^{3}$	$8.64x10^{3}$	$1.88x10^{3}$
^{238}Pu	-	$1.35x10^{2}$	$4.19x10^{3}$	$9.70x10^{3}$	$3.38x10^{3}$	$5.33x10^{3}$
^{239}Pu	-	$5.07x10^{3}$	$1.22x10^{5}$	$4.84x10^{4}$	$8.85x10^{4}$	$1.41x10^{4}$
^{240}Pu	-	$2.37x10^{3}$	$5.13x10^{4}$	$4.90x10^{4}$	$3.02x10^{4}$	$2.79x10^{4}$
^{241}Pu	-	$1.25x10^{3}$	$2.77x10^{4}$	$2.75x10^{4}$	$2.84x10^{4}$	$4.75x10^{3}$
^{241}Am	-	$4.63x10^{2}$	$8.48x10^{3}$	$5.49x10^{3}$	$1.51x10^{3}$	$4.09x10^{3}$
^{242}Cm	-	$9.88x10^{2}$	$1.95x10^{-1}$	$1.01x10^{3}$	$4.71x10^{2}$	$1.54x10^{2}$
^{244}Cm	-	$5.13x10^{3}$	$7.78x10^{2}$	$5.28x10^{3}$	$5.40x10^{2}$	$1.89x10^{3}$
^{99}Tc	-	$7.87x10^{2}$	$2.32x10^{3}$	$2.32x10^{3}$	-	$5.48x10^{3}$
^{129}I	-	$1.82x10^{2}$	-	-	-	$2.42x10^{3}$
^{137}Cs	-	$1.22x10^{3}$	$4.18x10^{3}$	$4.18x10^{3}$	-	$9.09x10^{3}$
^{147}Pm	-	$1.39x10^{2}$	-	-	-	$6.51x10^{2}$
^{151}Sm	-	$1.37x10^{1}$	-	-	-	$6.28x\ 10^{2}$

Figure 3 present the radioactivity contribution by actinide, in which can be seen that the plutonium domains most of the radioactivity until 10^5 except for the interval in which the americium radioactivity is higher than the plutonium. On one hand, the intervals in which this minor actinide (MA) has the higher contribution for the PWR and ADS is approximately between 100-1000 and 250 -750 years, respectively. On the other hand, the curium concentration on the VHTR is higher than in the others, which contributes at the same level of radioactivity as the plutonium in the first 100 years. Then, it decreases until being negligible. After 10^5 years, the uranium, actinium, and thorium contributions become strong and start to dominate the radioactivity. The following graphics (Fig. 3 (a, b and c)) show how the differences for nuclide amount influence in the radioactivity behavior during 10^7 years according to each reactor and reprocessed technique used.

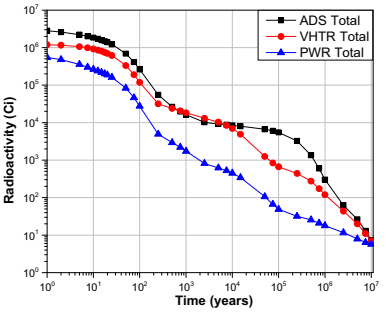

Fig. 1. Total radioactivity for each reactor.

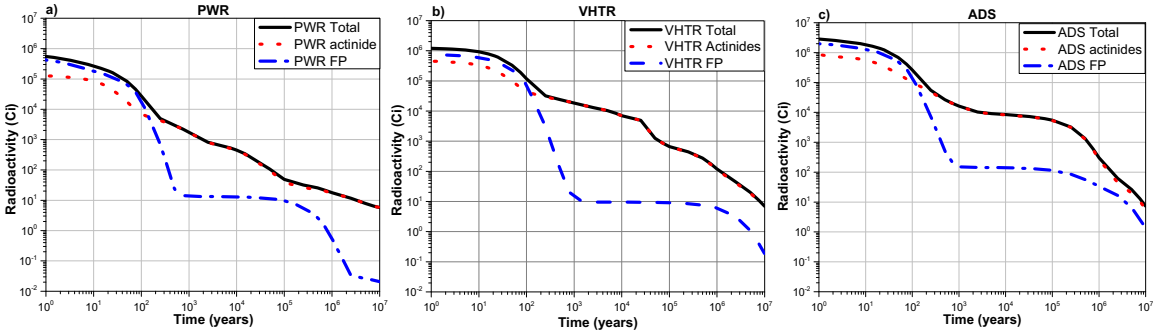

Fig. 2. Contribution to the radioactivity of Actinides and FP for each reactor: a) PWR, b) VHTR and c) ADS.

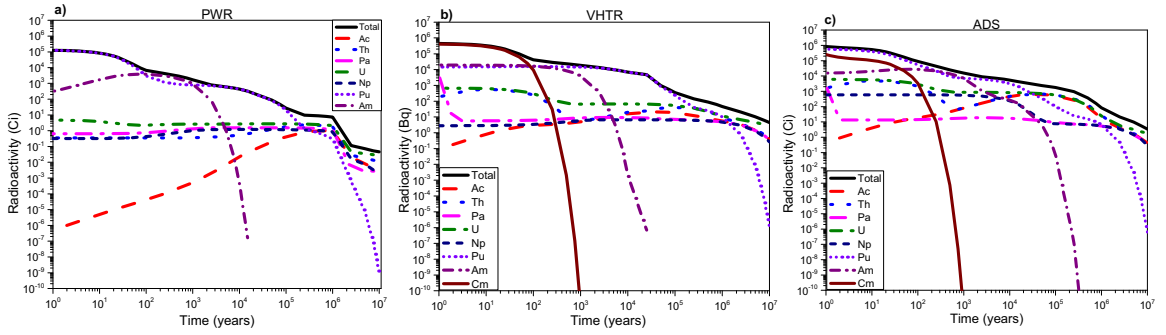

Fig. 3. Actinides contribution to the total radioactivity for each reactor: a) PWR, b) VHTR and c) ADS.

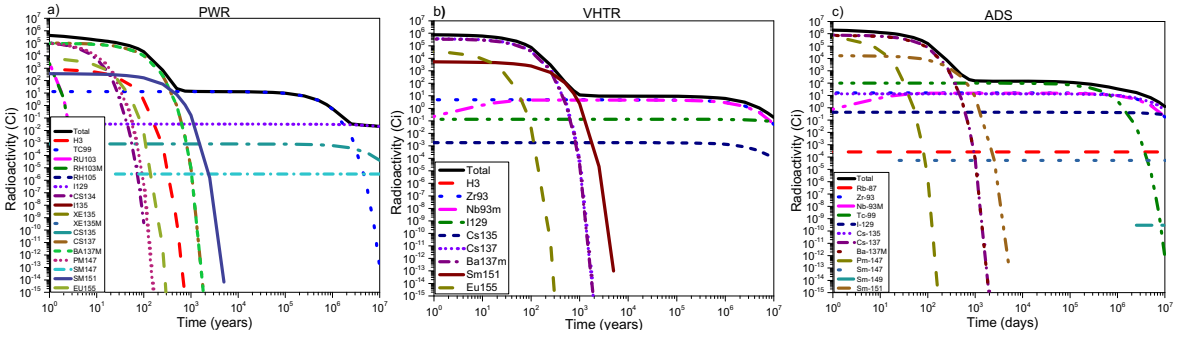

Fig. 4. Fission products contribution to the total radioactivity for each reactor: a) PWR, b) VHTR and c) ADS.

However, Figure 4 shows the fission products contribution by nuclide for each reactor. It can see that the most of the FPs decay after 2000 years, the barium-137m and caesium-137 are the highest contributors to the radioactivity during the first 500 years. After that, in general, the technecium-99, and iodine-129 are the major contributors to the radioactivity.

3.3. Radiotoxicity for Ingestion

The potential hazard of nuclear waste is characterized by its toxicity, taking into account the sensitivity of biological systems to inhalation or ingestion of radioisotopes[9]. Figure 5 shows the total ingestion hazard in the spent fuel materials studied. The ADS spent fuels have the highest radiotoxicity during 7.5 million years then the PWR becomes the one with higher toxicity. On the other hand, the VHTR SNF is the second most radiotoxic until the 2.5 million years. After this point, the PWR has higher radiotoxicity. As shown in Figure 6, the responsible for the high radiotoxicity are the actinides, which not happen on the PWR. In this case, the fission products are responsible for the highest radiotoxicity for the first 40 years. Then the actinides dominate the radiotoxicity levels. The major difference between the VHTR and ADS is that in the VHTR the curium contribution is larger provoking differences between the FP and actinides ingestion toxicity, as can be seen in Figure 7. Thorium is one of the principal contributors to radiotoxicity around $2x10^5$ to 10^7, 10^5 to 10^7 and 10^4 to 10^7, for the PWR, VHTR, and ADS, respectively. As shown in Figure 8, for the FP case, the highest contributor to the first 2000 years is the cesium. After, this is the iodine which continues until the 10^7 years.

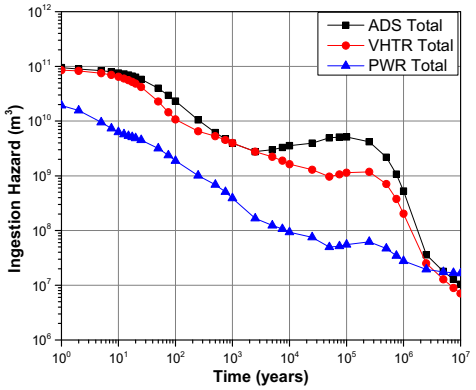

Figure 5. Total ingestion hazard for spent fuel materials.

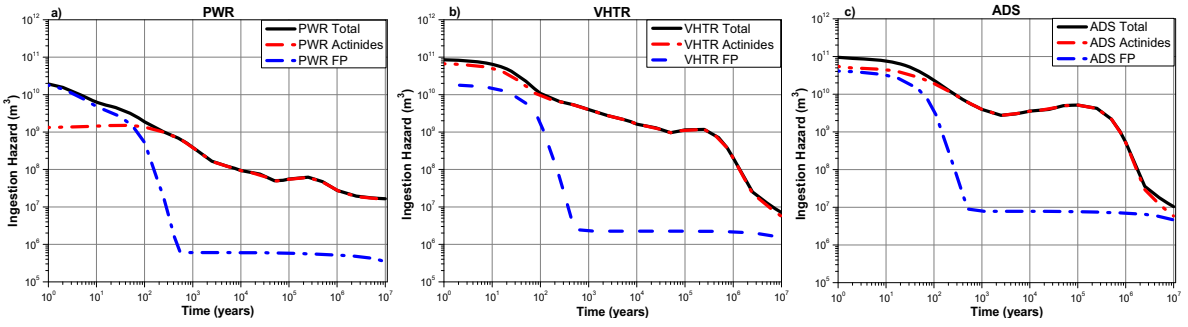

Figure 6. Ingestion hazard for each reactor: a) PWR, b) VHTR and c) ADS.

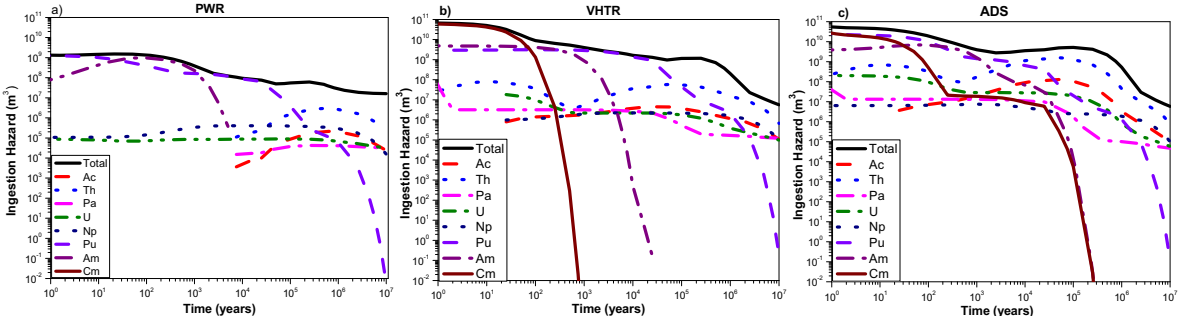

Figure7. Ingestion hazard by actinide for each reactor: a) PWR, b) VHTR and c) ADS.

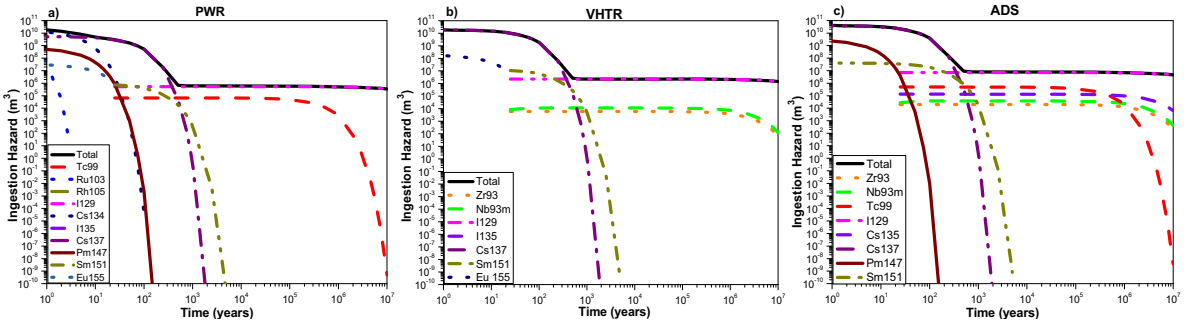

Figure 8. Ingestion hazard by fission product for each reactor: a) PWR, b) VHTR and c) ADS.

3.4. Decay Heat

The decay heat of nuclear fuels is extremely important because it involves the proper manner to manage the spent fuel cooling system, the transport of spent fuel and the repositories design[10]. Fig. 9 presents a comparison of the spent fuel decay heat, in which the VHTR and the ADS spent fuels have higher decay heat than PWR spent fuel. Nevertheless, after 10 million years the decay heat values reach almost the same values between the three spent fuels. As shown in Fig. 10 and 11, the first years after the discharge, the VHTR spent fuel releases more decay heat. It is due to the actinide contribution, especially curium, and plutonium. Fig. 10 and 12 present Cs-134 as the principal contributor to the decay heat for the PWR. It has upper contribution than the actinides for the first 25 years after discharge to the decay heat. For the VHTR and the ADS, Ba and Cs have the highest contributions for the first 1000 years. Then, the zirconium and niobium overtake the highest contribution from the FP to the decay heat.

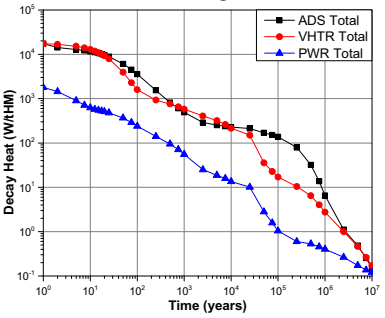

Fig. 9. Total decay heat for spent fuel materials.

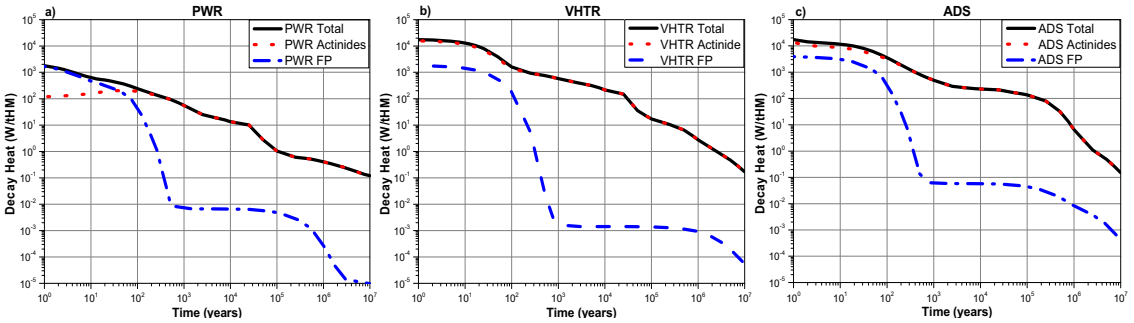

Fig. 10. Decay heat for each reactor: a) PWR, b) VHTR and c) ADS.

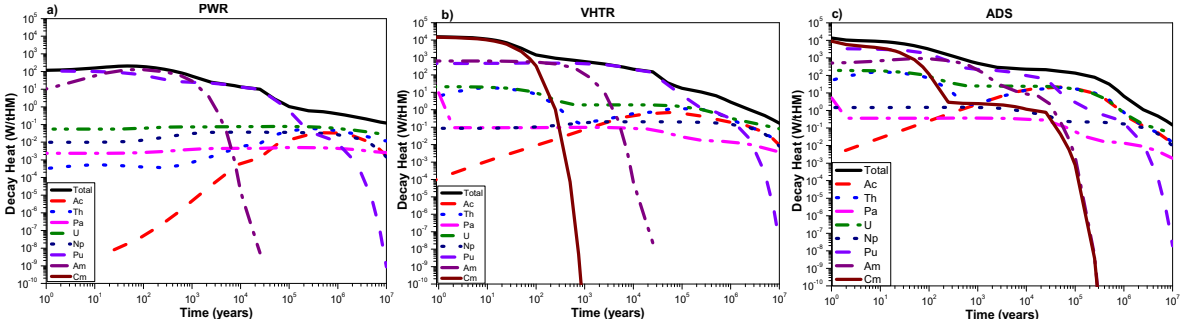

Fig. 11. Decay heat by actinide for each reactor: a) PWR, b) VHTR and c) ADS.

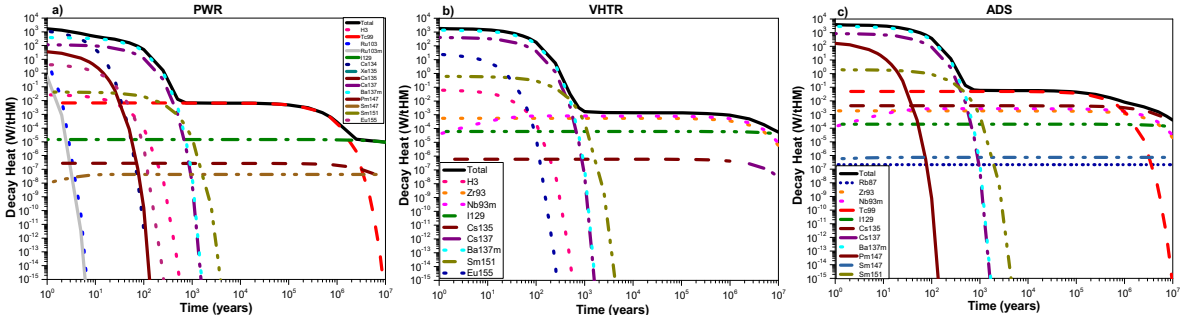

Fig. 12. Decay heat by fission product for each reactor: a) PWR, b) VHTR and c) ADS.

4. Conclusions

This paper is a preliminary fuel cycle analysis of the SNF behavior after irradiation for different reprocessing techniques and systems. The SNFs coming from reprocessing techniques and burned up in advanced reactors show that the radiotoxicity decreases below a conventional SNF from a typical PWR for the time studied. The VHTR and ADS have higher values of radioactivity, radiotoxicity, and decay heat, because of the greater concentrations of plutonium and curium in these reactors than in the PWR. Fission products have the greatest contribution the first 25 years over the parameters studied for a PWR. The most harmful fission products are Ba-137m, Tc-99, I-129 and Nb-93m and for the actinides is the plutonium and curium. The presence of actinium and thorium increases over time

and turning significant in all parameters studied. The decay heat data will be useful for the design of spent fuel pools, canister, SNF cask for transportation, and final repository.

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