Id.: EN49

PRELIMINARY EVALUATION OF RADIOACTIVE WASTE FROM REPROCESSED FUELS SPIKED WITH THORIUM OR DEPLETED URANIUM FROM A PWR: A COMPARATIVE ANALYSIS

[1,2,3]Lucas Chaves, [1,2]João P. Cunha Melo, [1,2,3]Samuel Fernandes, [2,3]Patricia A. L. Reis, [4]Rafael Miró, [2,3]Claubia Pereira

[1]Departamento de Física – Instituto de Ciências Exatas (ICEX) –
Universidade Federal de Minas Gerais (UFMG)
[2]Departamento de Engenharia Nuclear – Escola de Engenharia –
Universidade Federal de Minas Gerais (UFMG)
[3]INCT de Reatores Modulares e Inovadores
Av. Antônio Carlos, 6627 campus Pampulha 31.270-901, Belo Horizonte, MG, Brasil
[4]Instituto Universitario de Seguridad Industrial, Radiofísica y Medioambiental. Universitat Politècnica de València, Camí de Vera(s/n), 46022, València, España
lucasvgc@ufmg.br, lucasvgc97@gmail.com

Keywords: Angra 2, PWR, burnup, Assembly evaluation, reprocessed fuels

ABSTRACT

This work proposes transuranic fuels reprocessed by GANEX, spiked with depleted uranium or thorium, and analyzes the decay timing of the reprocessed fuels after burnup. The goal is to compare the behavior of reprocessed spent fuels to UOx spent fuels (once-through) in a long-term time spam. The parameters analyzed were specific activity, decay heat, and inhalation and ingestion radiotoxicity. The assemblies have been modeled and burned separately during a full life cycle, 868 days at 33GWd/MTHM, and left to decay for 1000 years. The assemblies are based on Angra 2, a PWR power reactor, and the studies were developed with the SERPENT Monte Carlo Code. The results show no major parameter changes in fission products, comparing UOx and transuranic fuels. The actinides parameters for TRU fuels are higher than the UOx fuels. However, it is important to emphasize that the TRU fuel spiked with depleted U is more closely related to the original fuel, in the parameters observed.

1. INTRODUCTION

The final disposal of spent fuel, the depleted uranium from the enrichment process, and the extraction of uranium ore are the main contributors to the nuclear energy environmental impact. Alongside the scientific community's concerns, there is great pressure from the international community against nuclear energy due to its radioactive waste disposal. Therefore, it is pertinent for the nuclear engineering to find solutions for an eco-friendlier disposal of these spent fuels/depleted uranium. A possible solution would be the spent fuels reprocessing, and the reuse of the depleted uranium, derived from the enrichment process, in these fuels.

Over 40 reactors worldwide use mixed oxide fuels (MOX) to take advantage of plutonium and other fissile materials in the spent fuel's matrix. In a PWR burnup at 47.5GWd/TonHM, the spent fuel matrix has approximately 0.76% of Pu(239+241) and 0.41% of Pu(236+240+242), and 0.75% of U-235, showing that spent nuclear fuel still contains a considerable amount of fissile material for energy output [1].

This work aims to analyze the impact of spent reprocessed fuels in a long-term decay (1000 years), compared to regular UOx fuels. This work also analyzes the difference between spent reprocessed fuels spiked with thorium and depleted uranium in the decay process since using depleted U not only avoids thorium mining but also finds a better use for depleted U. The work uses a model based on the Angra 2 reactor and all reactor characteristics are based on the

reactor FSAR [2]. Angra 2 is a commercial reactor in operation, located in Angra dos Reis – RJ. It is a PWR reactor, SIEMENS/KWU model, and it has been operating since 2001. According to FSAR [2], Angra 2 uses uranium dioxide (UO_2) enriched with 1.9% ~ 3.2% of U-235. The Angra 2 reactor was heavily studied on DEN[3-5], and a study based on reprocessed transuranic fuels using this reactor was also carried out [6].

The assemblies chosen for fuel replacement (UOx to TRUOx) were assemblies with gadolinium, aiming to match the absorption characteristics from transuranic actinides to the standard gadolinium absorber. The fissile concentration for each reprocessed fuel assembly was designed to replace one type of standard fuel assembly for a reprocessed fuel assembly in the modeled core, adjusting the reprocessed fissile material until the global k-eff between the standard UOx core and the core with a reprocessed replacement matches. This process proposed 6 reprocessed fuel assembly types, 3 types spiked with depleted U, and 3 spiked with Th, using the GANEX recuperation method [7]. All assemblies were left to decay for 1000 years after a full life burnup (868 days at 33GWd/MTHM); specific activity, decay heat, and inhalation and ingestion radiotoxicity were analyzed during that time.

2. METHODOLOGY

The work methodology can be divided into three parts:

1. Core and assemblies modeling
2. Reprocessed fuels proposition using the Angra 2 core model,
3. Angra 2 assembly models burnup.

All the models were modeled using SERPENT 2.2.1 - a Continuous-energy Monte Carlo Reactor Physics Burnup Calculation Code [8]. The ENDF/B-VII cross-section data was used in all simulations [9].

2.1 Core and assemblies modeling

The reactor core was modeled in its active part according to FSAR, at working temperatures (WT), with control rods completely withdrawn. In addition to the core active length, an additional geometry was added to the model, a reflector at the model top and bottom, seeking to moderate and reflect neutrons on the upper and lower surfaces. The model also has been separated into 34 nodes axially; 32 nodes are the actual core, and 2 nodes are the top/bottom reflectors. No boron is added to the model.

The reactor core has 193 fuel assemblies, each with 236 fuel rods and 20 guide tubes, half-pitch = 1.43 cm, and a height of 391.6 cm in WT. The guide tubes use Zircaloy 4 as composition material, and the same for the claddings. The Angra 2 core has 6 types of fuel assemblies. Names were addressed for each one of them to facilitate the evaluation:

- Type 19: UOx enriched with 1.9% U-235
- Type 25: UOx enriched with 2.5% U-235
- Type 25V12: UOx enriched with 2.5% U-235, 12 rods of $UO_2Gd_2O_3$
- Type 32: UOx enriched with 3.2% U-235
- Type 32V8: UOx enriched with 3.2% U-235, 8 rods of $UO_2Gd_2O_3$
- Type 32V12: UOx enriched with 3.2% U-235, 12 rods of $UO_2Gd_2O_3$

On the original design, the $UO_2Gd_2O_3$ rods are divided into 3 parts: the top and bottom portions with a fresh UOx fuel with the same enrichment compared to the associated assembly type, and the middle section are the $UO_2Gd_2O_3$ pellets, with the UO_2 in its natural composition.

For this work, all fuel rods with enriched UO_2 + $UO_2Gd_2O_3$ were homogenized for simplification.

Tab. 1. Fuel pins geometry and temperature.

Material:	**Temp:**	**Outter Radius:**
Fuel	873K	4.583
Gap (He)	873K	4.659
Cladding (Zircaloy 4)	618K	5.385
moderator (H20)	573K	7.150 (pitch)

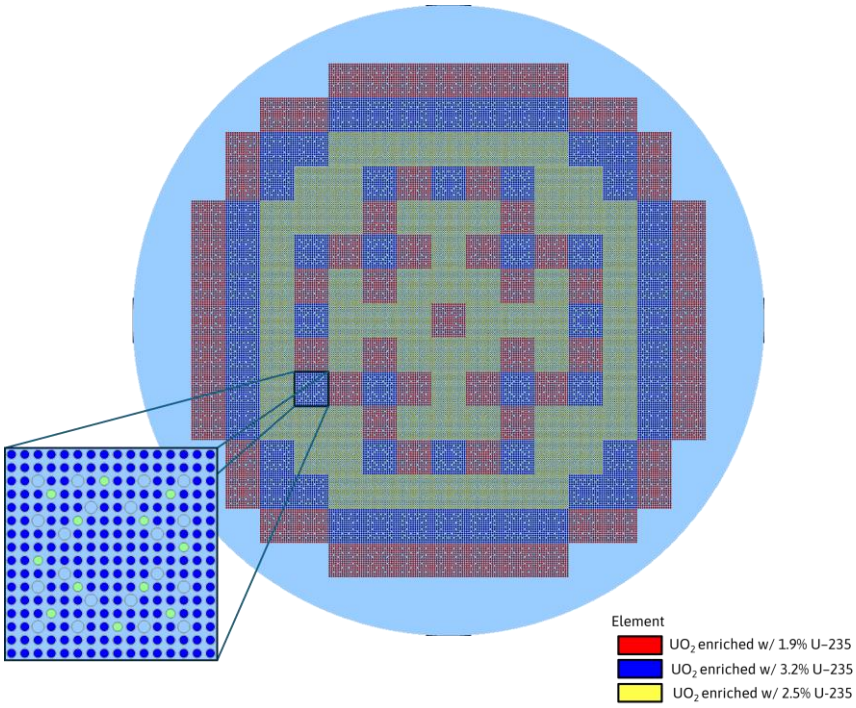

Fig. 1. Angra 2 XY core cross-section. SERPENT auto-generated image.

2.2. Reprocessed fuels proposition

To propose the exchange of fresh UOx + gadolinium fuels for reprocessed fuels, the following method was used: design a reprocessed fuel assembly; replace a single fresh fuel assembly by the correspondent reprocessed assembly; and adjust the fissile material from reprocessed assembly comparing the global k-eff between standard core model only with UOx + gadolinium and the core model replacing one type of UOx + gadolinium assembly with a reprocessed fuel assembly.

The fissile material from the reprocessed fuel assemblies was properly adjusted when the k-eff difference between the standard model and the model with the reprocessed fuel candidate was less than 100 pcm. This method was used until it got 6 reprocessed fuel assembly types, replacing only the UOx + gadolinium assemblies, 3 rep. assemblies using $(TRU,U)O_2$, and 3 rep. assemblies using $(TRU,Th)O_2$. The recovery factor used was the GANEX (Group Actinide Extraction), and the reprocessing process was carried out using a spent fuel matrix from the Angra I PWR reactor, with 3.1% enrichment, burned at 33 GWd/t [10][11]. A steady state running in the standard core was simulated, using 20000 neutrons for 200 cycles,

discarding the first 10%, with the boundary conditions set as "black" (neutrons can escape from the model). The standard UOx model k_{eff} = 1.16496 ± 0.00053. Names were addressed for each one of the reprocessed assemblies as well. The TRU-U's are spiked with depleted uranium, and the TRU-Th's, with thorium.

Tab. 2. Reprocessed fuel assemblies obtained for this work.

Standard Assembly Types	Reprocessed Alternatives	% of Fissile Material	Core k-eff w/ the assembly type replacement
25V12	TRU-U25V12	0.97	1.16489 ± 0.00060
	TRU-Th25V12	2.37	1.16474 ± 0.00064
32V8	TRU-U32V8	4.58	1.16491 ± 0.00066
	TRU-Th32V8	7.47	1.16480 ± 0.00067
32V12	TRU-U32V12	2.03	1.16457 ± 0.00061
	TRU-Th32V12	4.40	1.16464 ± 0.00063

2.3. Angra 2 assembly models burnup.

After the definition of reprocessed fuel assembly candidates, the UOx fuel assemblies and the TRU fuel assemblies were burned, with burnup and the specific power set as 33GWd/MTHM and 38MW/MTHM, respectively, according to FSAR [2]. These specifications led to an operation period of 868 days (~2.37 years), the total core lifetime. This evaluation used 20000 neutrons and 1000 cycles, discarding the first 30%. The boundary conditions for the assembly's burnup were set as reflective. After the assembly's burnup, the depleted assemblies have been analyzed during 1000 years of decay.

3. RESULTS

From the decay data, were analyzed the decay heat, specific activity, inhalation, and ingestion radiotoxicity for all types of assembly, for the actinides and fission products, and compared the UOx assemblies with their respective reprocessed fuel assembly candidates.

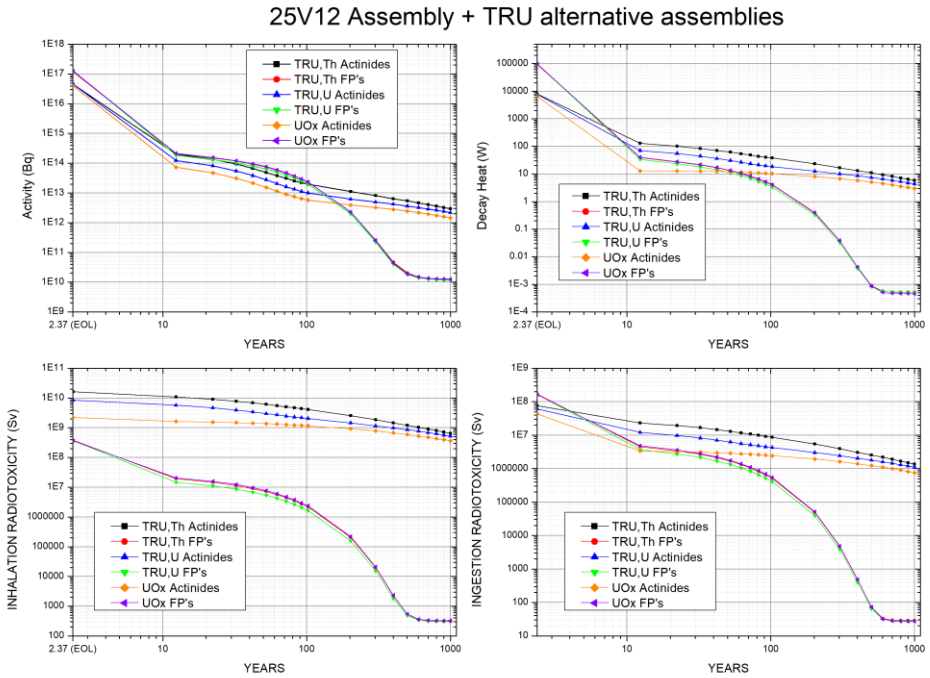

Fig. 2. Top to bottom, left to right: Specific activity (Bq/TonHM), Decay heat, Inhalation Radiotoxicity and Ingestion Radiotoxicity for actinides and FP's, analyzing the 25V12 assembly type and its alternative reprocessed fuel assemblies: TRU-U25V12 and TRU-Th25V12.

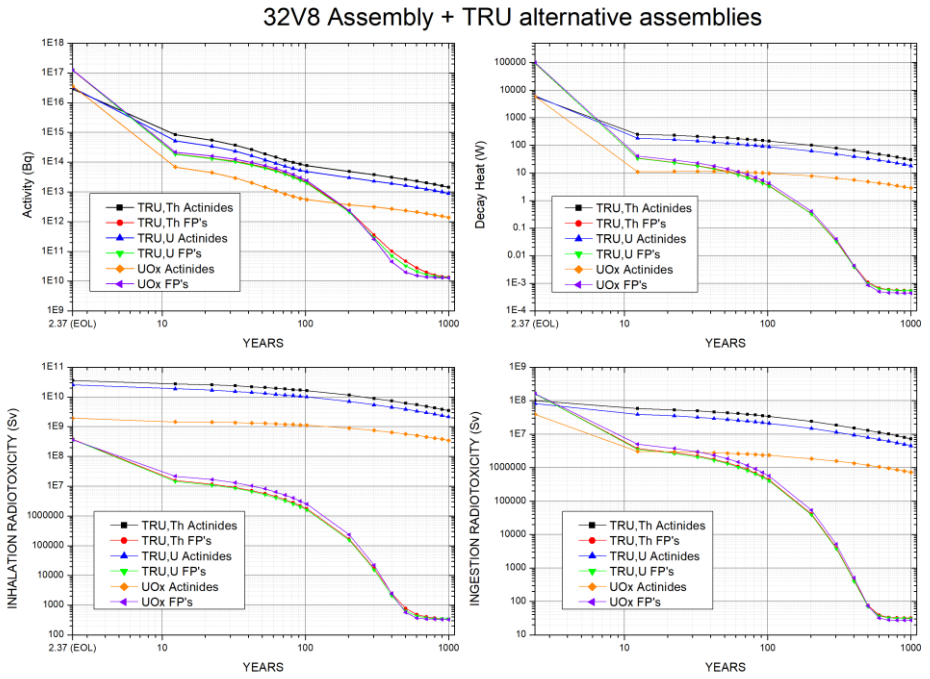

Fig. 3. Top to bottom, left to right: Specific activity (Bq/TonHM), Decay heat, Inhalation Radiotoxicity and Ingestion Radiotoxicity for actinides and FP's, analyzing the 32V8 assembly type and its alternative reprocessed fuel assemblies: TRU-U32V8 and TRU-Th32V8.

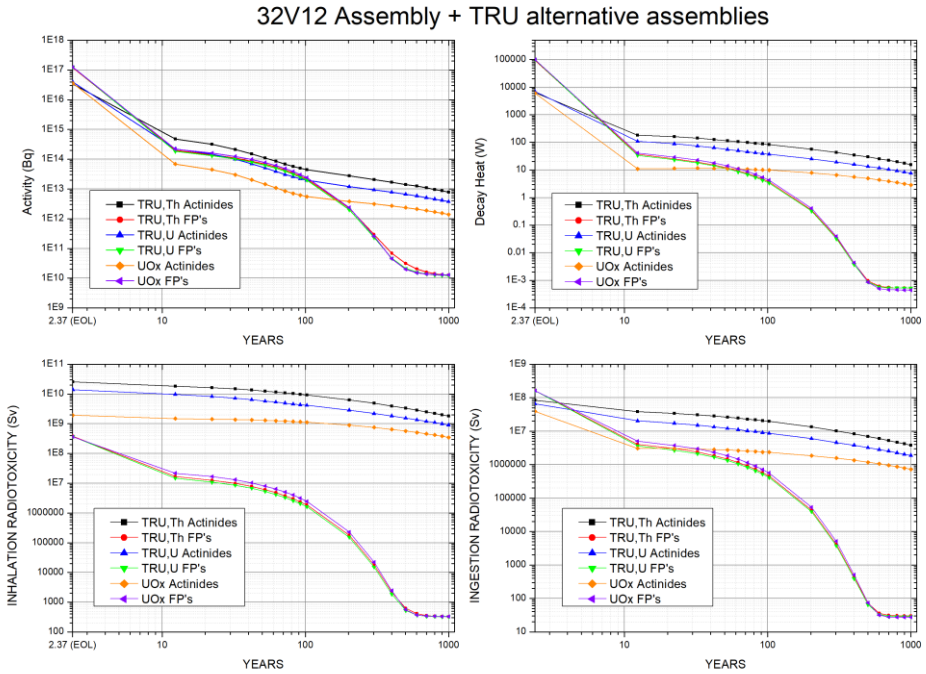

Fig. 4. Top to bottom, left to right: Specific activity (Bq/TonHM), Decay heat, Inhalation Radiotoxicity and Ingestion Radiotoxicity for actinides and FP's, analyzing the 32V12 assembly type and its alternative reprocessed fuel assemblies: TRU-U32V12 and TRU-Th32V12.

In global characteristics, the same tendencies can be seen on graphs 2 – 4 when comparing reprocessed fuels and UOx fuels. FPs, for example, don´t have a significant difference between the UOx and TRU fuels for all parameters, except for a small difference in the specific activity for TRU fuels on the 32V8 candidates, the candidates with more fissile material. The TRU fuels have higher values in all parameters, that's expected due to the actinide's behavior. The Pu isotopes have about twice the integral absorption cross-section value compared to U isotopes on the absorption resonances in the epithermal (0.3 to 1.5 eV) range [12]. Therefore, a higher gamma/beta emission from these isotopes is expected due to de-excitation/transmutation, increasing the decay heat and composition activity. The Am isotopes are great absorbers as well.

The TRUs spiked with Th-232 have the highest values for actinides, and the TRUs spiked with depleted uranium behave more similarly to UOx fuel from actinide's point of view. As shown in figure 2, the 25v12 type and its reprocessed assembly candidates behave more similarly than the other assembly models from different enrichments and fissile material percentages. It's worth mentioning that the TRU models corresponding to the 25V12 have the lowest amounts of fissile material. The decay heat and the specific activity for the 3 sets of assemblies start at 2.37 yrs virtually with the same value and sparse in the first 10 years, maintaining a certain constant value distance after that. The radiotoxicity is higher at the EOL, but the value difference between the assembly types tends to be less evident as time evolves.

4. CONCLUSION

This work aims to analyze the impact of spent reprocessed fuels in a long-term decay (1000 years) compared to regular UOx fuels. Reprocessed fuel was made using the GANEX method, aiming to replace standard UOx + Gd fuels. The fissile concentration for each reprocessed fuel

assembly was chosen, replacing one type of standard fuel assembly for a reprocessed fuel assembly and adjusting the fissile material until the global k-eff between the standard UOx core and the core with a replacement matches. This process was made for all the 6 fuel assembly types. After the definition of reprocessed fuel assembly candidates, the UOx and TRU fuel assemblies were burned at 33GWd/MTHM, for 868 days (~2.37 years), the total core lifetime. After the assembly's burnup, the depleted assemblies have been analyzed during 1000 years of decay. The specific activity, decay heat, and inhalation and ingestion radiotoxicity were analyzed.

All results have a convergence in their behavior for all parameters analyzed. All these radiation values are bound to each other in a way that it's expected that a certain type of fuel with a specific composition has higher values for all parameters compared than other compositions. With more activity, there is more chance of radiotoxicity hazard and a higher decay heat, although the decay heat also depends on other specific transmutation factors. The FPs, in this case, doesn´t have a significative difference between the UOx and TRU fuels for all parameters, except for a small difference in the specific activity for TRU fuels on the 32V8 candidates, the candidates with more fissile material. The radiotoxicity is higher at the EOL, but the value difference between the assembly types reduces as time passes, making evidence that, even if the original UOx fuel is less radiotoxic soon after it leaves the reactor, more in-depth studies need to be carried out to analyze whether this feature compensates for the carbon footprint generated by uranium mining or by not using spent/depleted materials. Further studies will be made using these reprocessed fuels, to check the neutronic/thermohydraulic viability using reprocessed fuels in this model.

ACKNOWLEDGEMENTS

The authors are thankful for the financial support provided by CAPES, CNEN, CNPQ, and FAPEMIG.

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