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Criticality safety analysis of spent fuel pool for a PWR using UO_2, MOX, $(Th\text{-}U)O_2$ and $(TRU\text{-}Th)O_2$ fuels

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ABSTRACT

A spent fuel pool of a typical Pressurized Water Reactor (PWR) was evaluated for criticality studies when it uses spent nuclear fuels. PWR nuclear fuel assemblies with four types of fuels were considered: standard PWR fuel, MOX fuel, thorium-uranium fuel and reprocessed transuranic fuel spiked with thorium. The MOX and UO_2 benchmark model was evaluated using SCALE 6.0 code with KENO-V transport code and then, adopted as a reference for other fuels compositions. The four fuel assemblies were submitted to irradiation using three operating cycles with burnup equal to 16 GWd/teHM. The burnup calculations were obtained using the TRITON sequence in the SCALE 6.0 code package. The fuel assemblies modeled use a benchmark 17x17 PWR fuel assembly dimensions. After irradiation, the fuels were inserted in the pool. The criticality safety limits were performed using the KENO-V transport code in the CSAS5 sequence. It was shown that mixing a quarter of reprocessed fuel withUO_2 fuel in the pool, it would not need to be resized

Keywords: reprocessed fuel, Spent Fuel Pool, criticality, PWR

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1. INTRODUCTION

There has been a continued interest in reprocessing nuclear fuels to recycle useful nuclear materials such as uranium, thorium, and plutonium [1]. Today, nuclear reactors operate mainly with uranium-plutonium cycle but since the beginning of nuclear power development, thorium was considered as an alternative fuel option for reactors [2].

Mixed Oxide (MOX), as well as thorium and transuranic spiked with thorium are alternatives to the Low-Enriched Uranium (LEU) fuel used in Light Water Reactors (LWRs). There has been a revival of interest in the use of thorium in light water reactors because its use in the nuclear fuel could provide longer life cycles and high burnup in the reactors while increasing in-repository durability [3]. Moreover, thorium is three times more abundant in nature compared to uranium and has an attractive potential for breeding to a fissile isotope [4]. On the other hand, in-reactor, MOX fuel behavior is similar to that of UO_2 in terms of crystallographic, physical and neutronic properties. Thus, MOX has been used to replace UO_2 in thermal reactors [5].

The reactivity of nuclear fuel decreases with irradiation (or burnup) due to the transformation of heavy nuclides and the formation of fission products. Burn-up credit studies aim at accounting for fuel irradiation in criticality studies of the nuclear fuel cycle (transport, storage, etc.). Several benchmark exercises were conducted in order to compare computation tools used in this context [6]. In addition to MOX recycle, other non-proliferating reprocessing fuels such as Th-Pu, Th-U e Th-TRU(transuranic) has also been studied [7].

Recently, the interest in the thorium cycle has increased and many researches on thorium are carried out [11-14]. The purpose of this paper is to understand the magnitude and trends in the burn-up credit of three spent nuclear fuel (SNF); being mixed oxide, natural thorium and natural thorium with plutonium, as well as typically low-enriched uranium fuel, at same conditions. This approach is then used to calculate the criticality under spent fuel pool based on Angra 2 pool. This work aims to use the criticality safety parameter to investigate whether or not the Angra 2 pool size needs to be remodeling in case of using spent nuclear fuel as disposal.

2. MATERIALS AND METHODS

2.1. MOX and UO_2 Benchmark description

In this paper, standard PWR fuel, mixed oxide (MOX), thorium $(Th\text{-}U)O_2$ and transuranic fuel spiked with thorium $(TRU\text{-}Th)O_2$ from a typical Pressurized Water Reactor (PWR) were evaluated. MOX and UO_2 fuels compositions have been derived following Phase IV-B Burn-up Credit Criticality benchmark [8].

The MOX fuel adopted from the Phase IV-B benchmark derived from the reprocessing of thermal reactor UO_2 fuels. This exercise is based upon fuel compositions provided by the MOX benchmark organizers and considered the impact of plutonium isotopic compositions in the MOX fuel, associated with first-generation MOX recycle. The MOX assembly pre-irradiation fuel composition is shown in Table 1.

Table 1: Initial MOX fuel compositions for given fuel pin (Atoms/barn.cm) [8].

Nuclide	High	Medium	Low	Average (for pin cell calculation)
U-234	2.5718E-07	2.6436E07	2.6789E-07	2.5952E-07
U-235	5.3798E-05	5.5300E-05	5.6040E-05	5.4287E-05
U-238	2.1194E-02	2.1786E-02	2.2077E-02	2.1387E-02
Pu-238	4.1677E-05	3.6128E-05	2.8473E-05	4.6610E-05
Pu-239	1.1259E-03	7.8717E-04	6.2038E-04	1.0156E-03
Pu-240	5.3500E-04	3.7403E-04	2.9478E-04	4.8255E-04
Pu-241	1.9392E-04	1.3557E-04	1.0685E-04	1.7491E-04
Pu-242	1.4636E-04	1.0233E-04	8.0644E-05	1.3201E-04
O-16	4.6602E-02	4.6553E-02	4.6529E-02	4.6586E-02

The MOX fuel assembly geometry adopted is a 17 x 17 PWR fuel assembly with three enrichment zones as shown in Figure 1. The initial MOX enrichments for these zones are shown in Table 2 [8].

Figure 1: *MOX fuel assembly [8].*

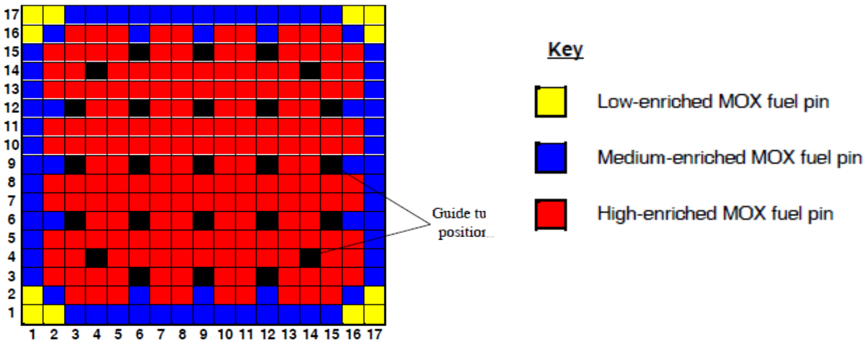


Table 2: Initial MOX fuel enrichments from Phase IV-B benchmark [8].

MOX Fuel Case A	MOX Fuel Enrichment, w/o $Pu_{fissile}$/[U+Pu]
High	5.692
Medium	3.984
Low	3.142
Average	5.136

The UO_2 fuel assemblies have an initial enrichment of 4.3 w/o ^{235}U/U taken from Phase IV-B exercise as well as a typical 17 x 17 PWR fuel assembly geometry. The composition of the UO_2 fuel is presented in Table 3 [8].

Table 3: Initial composition for 3.4w/o ^{235}U/U UO_2 fuel, from Phase IV-B exercise [8].

Nuclide	Composition Atoms/barn.cm
U-234	8.1248E-06
U-235	1.0113E-03
U-236	8.0558E-06
U-238	2.2206E-02
O	4.6467E-02

In Phase IV-B exercise, the same geometry related to a typical PWR assembly with 3.6568 m of height and 1.26 cm of pitch distance was adopted for MOX and UO_2 fuels. The assembly geometry with reflective boundary conditions was considered and no air gap between fuel and cladding is assumed. The 24 guide tubes and one instrumented tube were modelled as water-filled zircaloy-2 tubes. The assembly dimensions are presented in Table 4 [8].

Table 4: Fuel model parameters considering guide and instrumented tubes dimensions [8].

Parameter	Dimension
Fuel pin pitch	1.26 cm
Fuel pin radius	0.475 cm
Fuel pellet radius	0.410 cm
Cladding thickness	0.065 cm
Guide tube outer radius	0.613 cm
Guide tube inner radius	0.571 cm
Wall thickness	0.042 cm

A reduced-density zircaloy has been specified for the fuel pin cladding to take into account the air gap between the fuel and cladding. The guide tubes were also modelled using this reduced-density zircaloy composition as reported in the Phase IV-B benchmark. The non-fissile material compositions are specified in Table 5 [8].

Table 5: Non-fissile material compositions [8].

Nuclide	Atoms/barn.cm
Zircaloy-2 (5.8736 g/cm³ - reduced density)	
Zr	3.8657E-02
Fe	1.3345E-04
Cr	6.8254E-05
Coolant/moderator (600 ppm boro, 0.7245 g/cm³)	
H	4.8414E-02
O	2.4213E-02
^{10}B	4.7896E-06
^{11}B	1.9424E-05

The benchmark validation for MOX and UO_2 fuels was performed using KENO-VI sequence making use of CSAS6 module in the SCALE6.0 code and ENDF/B-VII collapsed 238-energy-group library. The irradiation history was performed following Phase IV-B benchmark. The material temperatures are specified in Table 6 [8].

Table 6: Material temperatures.

Material	Temperature (K)
Fuel temperature	900
Cladding temperature	620
Coolant/moderator temperature	575

Attempting to find a multiplication factor for thorium and thorium-transuranic fuels as close as possible to the MOX fuel multiplication factor, supplied by the benchmark, successive simulations using SCALE6.0 code with KENO-VI module and ENDF/B-VII collapsed 238-energy-group library were performed and the composition for $(Th\text{-}U)O_2$ and $(TRU\text{-}Th)O_2$ fuels were obtained.

After setting all assemblies composition, the fuels assembly were irradiated over three operating cycles, following Phase IV-B Burn-up Credit Criticality benchmark, two cycles consisting of 420 days full power with end of cycle (EOC) burnup equal to 16 GWd/teHM followed by 30 days downtime, and one cycle consisting of 420 days full power with EOC burnup equal to 16 GWd/teHM. For the irradiation, SCALE 6.0 code with KENO-VI sequence and ENDF/B-VII collapsed 238-energy-group library was adopted.

After irradiation, all fuels assemblies shall be conveyed to a spent fuel pool (SFP), maintaining the system under the upper criticality limit of 0.95 [10]. The criticality was calculated using KENO-V sequences making use of CSAS5 module and v7-238-energy-group library, including bias and uncertainty.

3. RESULTS AND DISCUSSION

3.1. MOX and UO_2 Benchmark Validation

The MOX and UO_2 benchmark results were compared with the results obtained in this study (DEN). Table 7 summarizes the three operating cycles (EOC 1, EOC 2 and EOC 3) taking into account the values obtained for k_{inf} as well as Average (Ave), Standard Deviation (SD) and relative Standard Deviation (RSD) calculations for the eight groups that contributed for MOX and UO_2 benchmark and DEN value.

Table 7: k_{inf} and reactivity change up for all benchmark participants and DEN.

Participant		k_{inf}			Relative difference in k_{inf} (%)		
		EOC 1	EOC 2	EOC 3	EOC 1	EOC 2	EOC 3
NUPEC		1.05978	1.00753	0.96100	0.19	0.45	0.63
CEA		1.05624	0.99968	0.94869	-0.14	-0.32	-0.66
GRS		1.05910	0.99909	0.94752	0.13	-0.38	-0.79
PSI		1.06088	1.00618	0.95837	0.30	0.31	0.35
BNFL		1.04976	0.99654	0.94974	-0.75	-0.64	-0.55
JAERI		1.05541	0.99749	0.95292	-0.22	-0.55	-0.21
DTLR		1.05900	1.00460	0.95100	0.12	0.16	-0.42
ORNL		1.06269	1.01166	0.96610	0.47	0.87	1.16
DEN		1.0567	1.0041	0.9598	0.10	0.11	0.50
Average	Before	1.05786	1.00108	0.95442			
	After	1.05773	1.00298	0.95502			
Stand.dev.	Before	0.00402	0.00543	0.00667			
	After	0.003430	0.00455	0.00667			
Stand.dev (%)	Before	0.38	0.54	0.70			
	After	0.32	0.45	0.69			

According to Table 7, the value obtained for k_{inf} using the different library with KENO-VI sequence is within the range of value obtained by other institutions, validating thus, the procedure adopted. In the Phase IV-B benchmark, a k_{inf} =1.1540 ± 0.0037 was verified for MOX fuel while in this study, an initial k_{inf} =1.1517 ± 0.0033 was obtained, ensuring benchmark validation. In the same benchmark an initial k_{inf}=1.3312 ± 0.0044 was found for the standad UO_2 fuel and the composition that gives this k_{inf} was used in this paper.

3.2. Th-UO_2 and TRU-ThO_2 Fuels

For thorium fuel, a mixture at 94% of theoretical density consisting of 75w/o Th and 25w/o U on a heavy metal basis latter enriched to 16 w/o U-235 giving an overall enrichment of 3.985 w/o U-235 in total heavy metal was adopted. It was found the pre-irradiation composition for the Th-UO_2 fuel assemblies which provided an initial k_{inf} =1.1587 ± 0.0036, using the same assembly geometry adopted for MOX fuel. The pre-irradiation fuel composition obtained is shown in Table 8.

Table 8: Initial composition for 16 w/o $^{235}U/U$ (Th-U)O_2 fuel

Isotope	Composition (Atoms/barn.cm)
Th-232	1.61215E-02
U-234	8.24518E-06
U-235	8.52488E-04
U-238	4.41091E-03
O-16	4.26835E-02

For the reprocessed transuranic fuel spiked with thorium, denominated (TRU-Th)O_2, characterization, the composition of the UO_2 fuel of a typical PWR, with initial enrichment of 3.1% with 33 GWd/tonHM of burnup, after 5 years in cooling pool was considered. Then, it was theoretically reprocessed by UREX+ reprocessing technique, a non proliferation reprocessing technique, that proposes the uranium and actinides co-extraction. The first stage involves the recuperation of uranium, plutonium and neptunium by extraction in 16 steps with 19% TBP in kerosene diluent and organic-to-aqueous ratio (O/A) equal to 3. With this stage, 99.99% U, 99.9% Pu, and 95% Np, beyond 0.1% of Am and Cm are recovered. There is also some contamination with these removals: 1.2% Ru, 1.6% Zr, 0.2% Ce and 0.1% Nd [9]. The reprocessed fuel was then spiked with Th-232 and the amount of fissile material contained therein being varied; starting at 1% and increasing to 30% in order to obtain the infinite neutron multiplication factor as close as possible of the MOX fuel benchmark. Table 9 represents the initial thorium-transuranic fuel composition. TRU-ThO_2 fuel also uses Phase IV-B exercise assembly dimensions.

Table 9: Initial (TRU-Th)O_2 fuel compositions atoms/barn.cm

Isotope	Composition	Isotope	Composition
Th-232	2.185E-02	Pu-240	8.586E-04
Np-235	2.286E-14	Pu-241	4.638E-02
Np-236	5.315E-11	Pu-242	2.426E-04
Np-237	1.343E-04	Pu-244	5.706E-09
Np-238	5.217E-14	Am-241	1.414E-04
Np-239	3.337E-11	Am-242m	3.770E-07
Pu-236	6.273E-09	Am-243	5.279E-05
Pu-237	2.874E-21	Cm-242	1.078E-07
Pu-238	7.066E-05	Cm-243	5.728E-06
Pu-239	2.058E-03	Cm-244	4.306E-04

The (TRU-Th)O_2 fuel composition was obtained after successive simulations and verified that a 43 w/o spiked with thorium , would give an overall fissile material of 10 w/o in this fuel and an initial k_{inf}=1,1531 ± 0.0041, the closest value of the initial MOX benchmark k_{inf}.

3.3. Fuels Assemblies

Once that MOX, (Th-U)O_2 and (TRU-Th)O_2 fuels have the initial k_{inf} values close to each other, and equal to 1.15, the four assemblies were then irradiated in a PWR core unit using the same three operating cycles previously described from Phase IV-B Burn-up Credit Criticality benchmark. Figure 2 represents the four assemblies simulated and Table 10 summarizes the pre-irradiation k_{inf} values.

Figure 2: *Fuel elements geometry adopted for Spent Fuel Pool criticality analysis. a) MOX, b) (Th-U)O_2, c) (TRU-Th)O_2 and d) UO_2.*

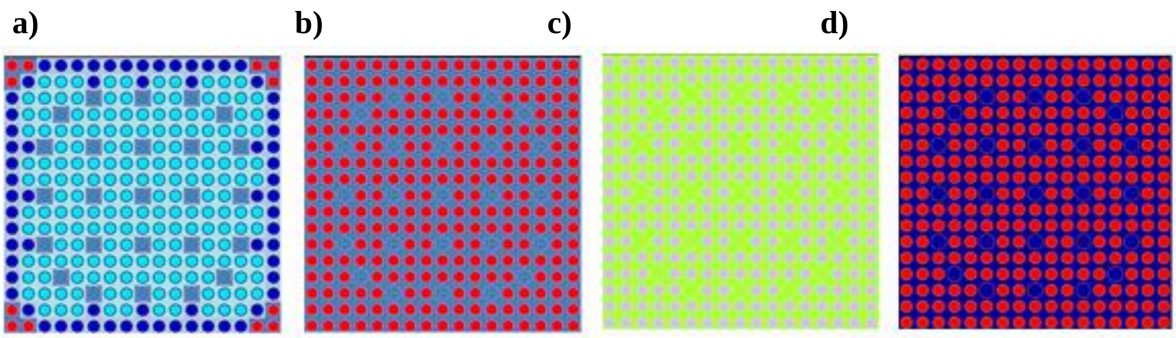


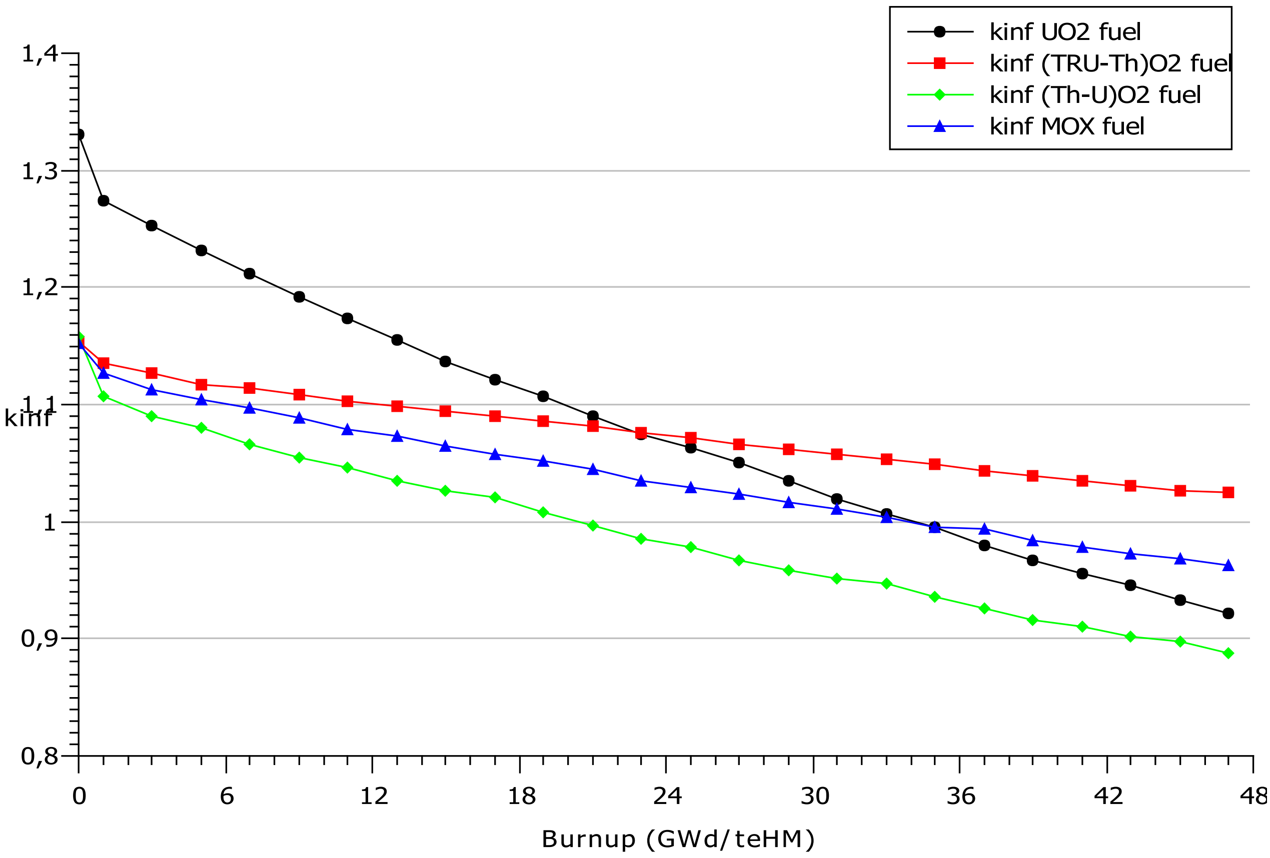
Table 10: Initial k_{inf} obtained for the four cases fuel.

Fuel	Pre irradiation k_{inf}
MOX	1.1517 ± 0.0033
(Th-U)O_2	1.1587 ± 0.0036
(TRU-Th)O_2	1.1531 ± 0.0041
UO_2	1.3312 ± 0.0044

3.4. Burnup Calculation

In order to preserve maximum accuracy during the depletion calculation, for (Th-U)O_2 and (TRU-Th)O_2 fuels, the legacy addnux value of 3 has been included. TRITON allows the user to determine the set of nuclides added to the combustible material by means of control parameter parm = (addnux = N), where N is an integer $0 \leq N \leq 3$. In N = 3, 166 nuclides are added, adding a total of 232 allowing a more detailed configuration. At these high burnup levels, these nuclides have a small effect on the neutron spectrum of the system but generally contribute to the overall reactivity of the system. Figure 3 shows the criticality curves plotted considering the 3 cycles for all the investigated fuels.

Figure 3: *k_{inf} considering the 3 cycles for a single assembly irradiated in PWR reactor.*



UO_2 fuel has a significant concentration of U-238 and U-235. In U-238 and U-235 chain decay there are no other fissile materials which explains the rapid decrease of UO_2 fuel curve. Although $(Th\text{-}U)O_2$ fuel has a high concentration of Th-232, achieving 75%, and the consequent produce of U-233, a fissile material, it has also a considerable concentration of U-238 and U-235 (20.61 and 3.985% respectively) which do not give rise to new fissile materials, ensuring a steep incline, but not as much as the curve of UO_2 fuel. MOX fuel, in reverse, it is a low-enriched uranium (LEU) fuel with only 0.233% of U-235 and 5.344% on fissile material. The presence of fissile material such as Pu-239 makes possible the chain reaction maintenance, ensuring a smooth curve for this fuel. $(TRU\text{-}Th)O_2$ fuel is spiked with thorium, so it has 82.72% of Th-232, this isotope can absorb neutrons and become U-233. This fact, added to any presence of uranium in this fuel, contribute for the smoother inclination of the curve.

The above mentioned results suggest that the thorium-transuranic fuel still have burnup potential after the 3 cycles.

The composition, after burnup, containing the mainly nuclides for all fuels assemblies studied are shown in Table 11.

Table 11: Fuel's Composition after burnup (atom/barn.cm).

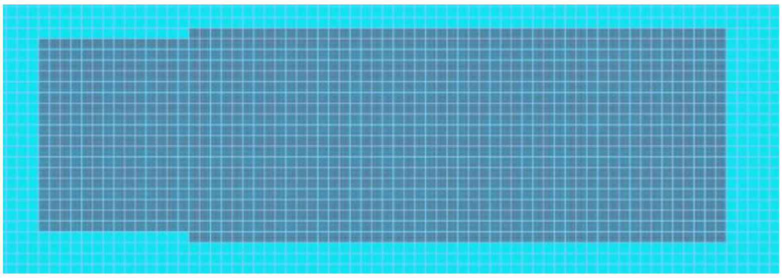
Composition after irradiation in PWR reactor				
Nuclide	MOX Fuel	(Th-U)O_2 Fuel	(TRU-Th)O_2 Fuel	UO_2 Fuel
U-232	1.837E-11	1.209E-06	1.269E-06	2.226E-11
U-233	2.494E-11	2.509E-04	3.528E-04	9.687E-11
U-234	1.024E-06	3.911E-05	2.659E-05	4.108E-06
U-235	2.663E-05	2.442E-04	4.083E-06	2.082E-04
U-236	6.241E-06	1.358E-04	4.611E-07	1.412E-04
U-237	2.063E-06	1.498E-07	5.932E-10	2.215E-06
U-238	2.067E-02	3.989E-03	1.662E-09	2.141E-02
Pu-238	3.848E-05	4.423E-06	1.509E-04	6.848E-06
Pu-239	5.195E-04	4.684E-05	5.956E-04	1.491E-04
Pu-240	4.188E-04	1.620E-05	7.951E-04	6.563E-05
Pu-241	2.341E-04	1.504E-05	4.168E-04	4.070E-05
Pu-242	1.530E-0-4	6.509E-06	2.961E-04	1.750E-05
Th-232	3.680E-13	1.555E-02	2.130E-02	9.495E-12
Th-230	1.565E-12	1.667E-08	4.085E-08	3.947E-11
Am-242m	5.541E-07	5.501E-07	5.068E-06	3.029E-08
Xe-135	3.986E-10	6.631E-10	4.550E-10	3.480E-10

3.5. Spent fuel pool criticality

The pool model used in this study was based on the cooling pool described in the Angra 2, Final Safety Analysis Report - FSAR (2013) [10]. The pool's dimensions are 15.914 x 5.668 m and 11.6568 m depth. The criticality safety analysis considers the minimum boron concentration of 2300 ppm specified in FSAR and required for spent fuel pool [10]. It was important to evaluate the criticality under spent fuel pool conditions for these four fuels individually, considering the fuel assemblies at 298 K once that at this temperature would be expected the higher multiplication factor possible due to the Doppler effect.

The minimum pitch distance in the SFP was found using KENO-V sequences making use of CSAS5-S module in the SCALE 6.0 code. It was verified that a 0.695cm pitch distance would maintain the criticality under the upper criticality limit of 0.95 and assure the maximum elements' capacity in the SFP maintain the sub criticality. Figure 4 illustrates the SFP filled in with its entirely capacity.

Figure 4: *Spent fuel pool using Angra 2 FSAR (2013) as a model.*



Aiming to make a close analysis of the nuclear power plants and to ensure a safety project, two different load patterns were designed for assemblies while into the pool. In a first moment, an uniform configuration was adopted with just a single type of fuel. In a second instance, the reprocessed fuels or (Th-U)O_2 fuel were placed together with the standard fuel into the SFP taking into consideration a ratio of 1:3, correlatively. Figure 5 shows mixed distribution for the assemblies in the SFP.

Figure 5: *1:3 ratio configuration for SFP using MOX ,(Th-U)O_2 or (TRU-Th)O_2 with standard UO_2 fuel.*

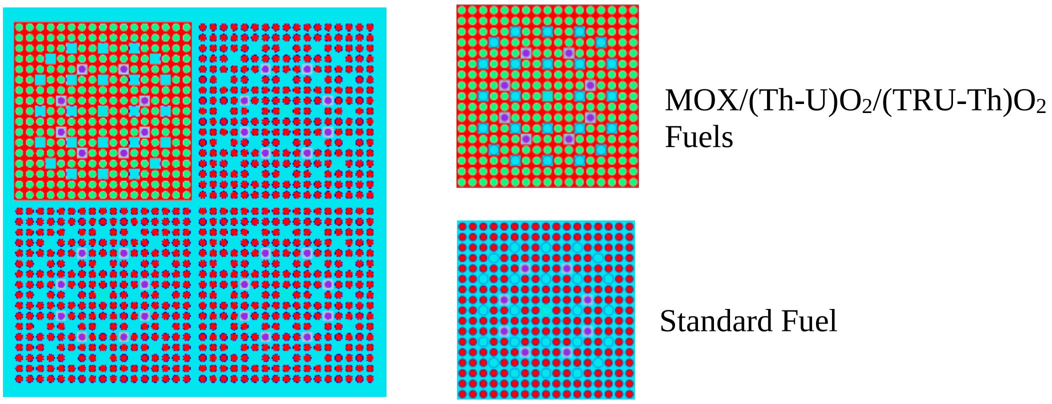


Table 12 summarizes the k_{inf} values for the fuels in the pool containing only one type of fuel and k_{inf} for the mixed spent fuel pool using the reprocessed fuels (MOX, $(TRU\text{-}Th)O_2$) or $(Th\text{-}U)O_2$ fuel with the standard PWR UO_2 fuel .

Table 12: k_{inf} when the fuel assemblies were inserted into the pool.

Only one fuel in pool	k_{inf}	Mixed-pool ($^3/_4 UO_2$)	k_{inf}
MOX	0.84409 ± 0.00032	¼ MOX	0.81084 ± 0.00027
$(Th\text{-}U)O_2$	0.64962 ± 0.00022	¼ $(Th\text{-}U)O_2$	0.75507 ± 0.00023
$(TRU\text{-}Th)O_2$	0.93698 ± 0.00028	¼ $(TRU\text{-}Th)O_2$	0.85139 ± 0.00026
UO_2	0.78256 ± 0.00029	--	--

The values for multiplication factor showed in Table 11 are in accordance with curves plotted in Figure 3 once that follow the same k_{inf} descending order.

Fill the UO_2 fuel in a mixed spent fuel pool, together with MOX or $(TRU\text{-}Th)O_2$ fuels, made the criticality decrease 3.939% and 9.135% respectively. Even though criticality has increased on 13.97% when UO_2 fuel assemblies were inserted with $(Th\text{-}U)O_2$ fuel assemblies in the pool, for all three cases of mixed spent fuel pool a $k_{safe} \leq 0.95$ remains guaranteed as established by Angra 2, Final Safety Analysis Report [10].

4. CONCLUSION

The results presented in the MOX benchmark were firstly validate and then compared with the results obtained from the simulations performed in this work. This study demonstrated the possibility of insertion of spent fuels based on transuranic elements and spiked with thorium in a PWR core as it extends the burning, decreases radioactive waste and decreases the risk of proliferation. The insertion of the fuels in the pool showed that the system remains subcritical. It was shown that by using a quarter of reprocessed fuel in the mixed spent fuel pool, the dimensions of the pool would not need to be modified.

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