



## **CRITICALITY SAFETY ANALYSIS OF SPENT FUEL POOL FOR A PWR USING REPROCESSED NUCLEAR FUELS**

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**Keywords:** Criticality, Spent fuel pool, reprocessed fuel

### **ABSTRACT**

A spent fuel pool of a typical Pressurized Water Reactor (PWR) was evaluated for criticality studies when it uses reprocessed 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<sub>2</sub> 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 at normal operation conditions. 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 with UO<sub>2</sub> fuel in the pool, it would not need to be resized.

### **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 it's 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<sub>2</sub> in terms of crystallographic, physical and neutronic properties. Thus, MOX has been used to replace UO<sub>2</sub> 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-TR(transuranic) has also been studied [7]

Recently, the interest in the thorium cycle has increased and many researches on thorium are carried out [10-13]. The purpose of this paper is to understand the magnitude and trends in the burn-up credit of three reprocessed 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 investigate of the need for modification in pool dimensions to accommodate reprocessed fuels.

## 2. METHODOLOGY

### 1. Criticality Safety Analysis for benchmark validation

In this paper, mixed oxide (MOX), thorium (Th- $\text{UO}_2$ ), transuranic spiked with thorium (TRU-Th) and  $\text{UO}_2$  fuels were considered. MOX and  $\text{UO}_2$  fuels compositions have been derived following Phase IV-B Burn-up Credit Criticality benchmark [8]. The composition of thorium-uranium fuel and reprocessed transuranic fuel spiked with thorium were obtained after successive computational simulations. As to ensure the validation of MOX benchmark, the irradiation using TRITON sequence in the SCALE6.0 code and the cross sections of the ENDF/B-VII library for 1000 generations and 5000 neutrons per generation package was performed. For this irradiation history, three operating cycles are requested; two cycles consisting of 420 days full power with end of cycle (EOC), burn-up equal to 16 GWd/teHM followed by 30 days downtime, and one cycle consisting of 420 days full power with EOC burn-up equal to 16 GWd/teHM [8].

For this study, MOX multiplication factor was found using KENO-VI module with v7-238-energy-group library as well as MOX benchmark composition and assembly geometry, related to a typical 17x17 PWR. The fuel assemblies modeled are rectangular (21.59 cm x 21.59 cm) with 3.657 m of height and 1.26 cm of pitch distance. Reflective boundary conditions were 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 tubes and assembly geometry is presented in Tab. 1 [8]:

Tab. 1. Fuel model parameters considering guide and instrumented tubes dimensions

| 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 benchmark. The non-fissile material compositions are specified in Tab. 2[8].



Tab 2. Non-fissile material compositions

| Nuclide  | Atoms/barn.cm |
|--|---------------|
| <b>Zircaloy-2 (5.8736 g/cm<sup>3</sup> - reduced density)</b>    |               |
| Zr   | 3.8657E-02    |
| Fe   | 1.3345E-04    |
| Cr   | 6.8254E-05    |
| <b>Coolant/moderator (600 ppm boro, 0.7245 g/cm<sup>3</sup>)</b> |               |
| H  | 4.8414E-02    |
| O  | 2.4213E-02    |
| <sup>10</sup> B  | 4.7896E-06    |
| <sup>11</sup> B  | 1.9424E-05    |

The benchmark data used for MOX and UO<sub>2</sub> assemblies' pre-irradiation composition are shown in Tab. 3 and Tab. 4, respectively. MOX fuel assembly considers three enrichment zones as shown in Tab. 5 and the geometry is shown in Fig. 1 [8].

For UO<sub>2</sub> fuel, following the benchmark, the assembly has an initial enrichment of 4.3 w/o <sup>235</sup>U/U. The same assembly geometry was adopted for all fuels in this investigation.

Tab. 3. Initial MOX fuel compositions (grams)

| Nuclide | High       | Medium     | Low        | Average (for pin cell calculation) |
|---------|------------|------------|------------|------------------------------------|
| U-234   | 1.2318E-06 | 1.2636E-06 | 1.2889E-06 | 1.2425E-06                         |
| U-235   | 2.5798E-04 | 2.6400E-04 | 2.6740E-04 | 2.5996E-04                         |
| U-238   | 1.0194E-01 | 1.0486E-01 | 1.0577E-01 | 1.0286E-01                         |
| Pu-238  | 2.4777E-04 | 1.7228E-04 | 1.3673E-04 | 2.2328E-04                         |
| Pu-239  | 5.3759E-03 | 3.7617E-03 | 2.9638E-03 | 4.8505E-03                         |
| Pu-240  | 2.5500E-03 | 1.7903E-03 | 1.4178E-03 | 2.3028E-03                         |
| Pu-241  | 9.2692E-04 | 6.4757E-04 | 5.1085E-04 | 8.3605E-04                         |
| Pu-242  | 6.9936E-04 | 4.8833E-04 | 3.8544E-04 | 6.3073E-04                         |
| O-16    | 2.2202E-01 | 2.2132E-01 | 2.2107E-01 | 2.2195E-01                         |

Tab. 4. Initial composition for 3.4w/o <sup>235</sup>U/U UO<sub>2</sub> fuel

| Nuclide | 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    |

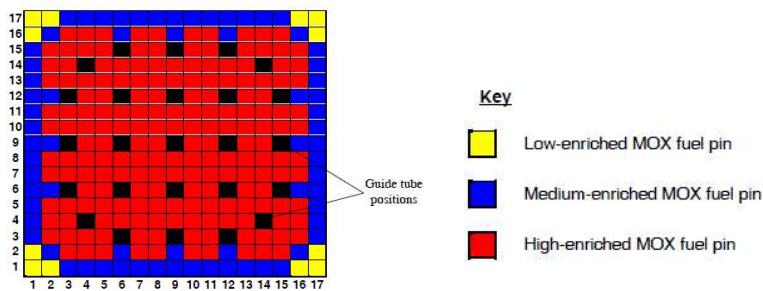


Fig. 1. MOX fuel assembly

Tab. 5. Initial fuel enrichments

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

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  $ThO_2-UO_2$  and TRU-Th fuels were obtained.

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 were adopted. The Transuranic fuel was spiked with thorium. After perform successive computational simulations, was verified that a 10 w/o Th spiked in this fuel would give the closer value of multiplication factor obtained by the benchmark.

## II. Burn-up calculation

After setting all assemblies composition, the burnup depletions for all assemblies investigated in this paper were made using the same conditions as MOX benchmark irradiation, previously specified.

## III. Spent fuel pool criticality

After irradiation, all fuels assemblies shall be conveyed to a spent fuel pool, maintaining the system under the upper criticality limit of 0.95 [9]. The criticality was calculated using KENO-V sequences making use of CSAS5 module and v7-238-energy-group library, including bias and uncertainty.

The pool model used in this study was based on the cooling pool described in the Angra 2, Final Safety Analysis Report - FSAR (2013) [9]. The pool dimension is 15.636 x 5.569 m and 367.7 of depth. The criticality safety analysis considers the minimum boron concentration of 2300 ppm specified in FSAR and required for spent fuel pool [9]. Fig. 2 shows the two cases of spent fuel pool.

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. Aiming to make a close analysis of the nuclear power plants and to ensure a safety project, three mixed-pools were designed and filled with a quarter of reprocessed SNF and three-quarter of standard UO<sub>2</sub>. The criticality was analyzed considering entirely pool capability and pool water at room temperature.

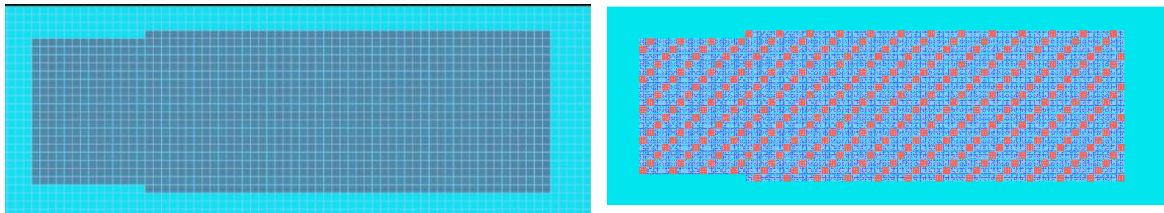


Fig. 2. Spent fuel pool using Angra-II FSAR (2013) as model and mixed spent fuel pool designed for reprocessed SNF.

### 3. RESULTS AND DISCUSSION

#### 1. Criticality Safety Analysis

In MOX benchmark, a  $k_{inf}=1.1540 \pm 0.0037$  was verified while for this study, an initial  $k_{inf} = 1.1517 \pm 0.0033$  was obtained, ensuring benchmark validation. In the same benchmark an initial  $k_{inf}=1.3312 \pm 0.0044$  was found and the composition that gives this  $k_{inf}$  was used in this paper.

In order to attain a sensible comparison of results, Tab. 6 summarizes the three operating cycles (EOC 1, EOC 2 and EOC 3) taking into account the average  $k_{inf}$  for the eight groups that contributed for MOX benchmark as well as the value for the beginning of cycle burn-up obtained in this study [8].

Tab.6.  $K_{inf}$  for each EOC reported in benchmark and  $k_{inf}$  values for MOX burn up.

|                    | <b><math>k_{inf}</math> for participants*</b> | <b>MOX <math>k_{inf}</math> obtained</b> |
|--------------------|---|--|
| Beginning of cycle | $1.1540 \pm 0.0037$                           | $1.1517 \pm 0.0033$                      |
| EOC 1              | $1.0578 \pm 0.0040$                           | $1.0609 \pm 0.0047$                      |
| EOC 2              | $1.0010 \pm 0.0054$                           | $1.0077 \pm 0.0031$                      |
| EOC 3              | $0.9544 \pm 0.0066$                           | $0.9598 \pm 0.0041$                      |

\*NUPEC, CEA, GRS, PSI, BNFL, JAERI, DTLR, ORNL.

Based on MOX fuel initial  $k_{inf}$  reported above, it was found the pre-irradiation composition for the ThO<sub>2</sub>-UO<sub>2</sub> fuel assemblies which provided an initial  $k_{inf}=1.1587 \pm 0.0036$ , using the same assembly geometry adopted for MOX fuel. The pre-irradiation fuel composition obtained is shown in Tab. 7.



Tab. 7. Initial composition for 16 w/o  $^{235}\text{U}/\text{U}$  Th-UO<sub>2</sub> fuel

| Isotope | Composition (grams) |
|---------|---------------------|
| Th-232  | 2.5160E-01          |
| U-234   | 1.2868E-04          |
| U-235   | 1.3304E-02          |
| U-238   | 6.8838E-02          |
| O-16    | 6.6613E-01          |

For TRU-Th fuel, the 10w/o on fissile material composition showed an initial  $k_{\text{inf}}=1,1531 \pm 0.0041$ . The pre-irradiation fuel composition for the TRU-Th fuel assemblies is shown in Tab. 8.

Tab. 8. Initial TRU fuel compositions grams

| Isotope | Composition | Isotope | Composition | Isotope | Composition |
|---------|-------------|---------|-------------|---------|-------------|
| Th-232  | 7,3847E-01  | Pu-240  | 3,0020E-02  | Cm-241  | 8,1851E-27  |
| Np-235  | 2,2836E-12  | Pu-241  | 1,6284E-02  | Cm-242  | 1,0782E-07  |
| Np-236  | 5,3150E-09  | Pu-242  | 8,5531E-03  | Cm-243  | 5,7228E-06  |
| Np-237  | 4,6380E-03  | Pu-243  | 1,4444E-18  | Cm-244  | 4,3061E-04  |
| Np-238  | 1,8087E-12  | Pu-244  | 5,7068E-07  | Cm-245  | 2,2711E-05  |
| Np-239  | 1,1620E-09  | Pu-246  | 5,4094E-24  | Cm-246  | 2,9232E-06  |
| Pu-236  | 6,2732E-09  | Am-241  | 4,9653E-03  | Cm-247  | 3,2156E-08  |
| Pu-237  | 2,8746E-21  | Am-242m | 1.332E-05   | Cm-248  | 1,8776E-09  |
| Pu-238  | 2,4498E-03  | Am-242  | 1,5921E-10  | Cm-250  | 1,0220E-17  |
| Pu-239  | 7,1653E-02  | Am-243  | 1,8689E-03  | O-16    | 1,2061E-01  |

Once that for the three reprocessed fuels, the initial  $k_{\text{eff}}$  values acquired are close to each other, and equal to 1.15, the irradiation at same condition was performed. Tab. 9 summarizes the pre-irradiation  $k_{\text{inf}}$  values.

Tab. 9. Initial  $k_{\text{eff}}$  obtained for the four cases fuel.

| Fuel             | Pre irradiation $k_{\text{eff}}$ |
|------------------|----------------------------------|
| MOX              | 1.1517 $\pm$ 0.0033              |
| ThO <sub>2</sub> | 1.1587 $\pm$ 0.0036              |
| TRU              | 1.1531 $\pm$ 0.0041              |
| UO <sub>2</sub>  | 1.3312 $\pm$ 0.0044              |

### 1. Burn-up calculation

In order to preserve maximum accuracy during a depletion calculation, for Th-UO<sub>2</sub> and TRU-Th fuels, the legacy addnux value of 3 has been included once that these fuels have a high concentration of Th-232 and U-233 nuclides and this option include most additional set of nuclides in transport updates and cross-section. Fig. 3 shows the criticality curves plotted considering the 3 cycles for all the investigated fuels.

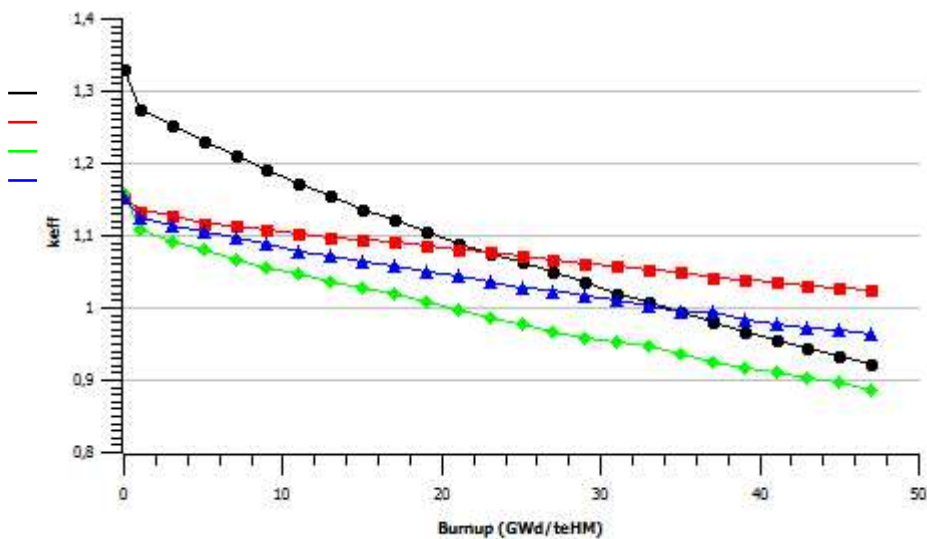


Fig. 3.  $K_{inf}$  for 3 cycle for a single assembly irradiated in PWR reactor.

UO<sub>2</sub> 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<sub>2</sub> fuel curve. Although Th-UO<sub>2</sub> 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<sub>2</sub> 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-Th 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 burn-up potential after the 3 cycles

The composition, after burn-up, containing the mainly nuclides for all fuels assemblies studied are shown in Tab. 10.

Tab. 10. Mainly nuclides and composition after burnup (grams).

| Composition after irradiation in PWR reactor |            |                         |             |                      |
|--|------------|-------------------------|-------------|----------------------|
| Nuclide                                      | MOX Fuel   | Th-UO <sub>2</sub> Fuel | TRU-Th Fuel | UO <sub>2</sub> Fuel |
| U-232  | 6.9299E-10 | 5.4321E-05              | 4.4483E-05  | 8.4300E-10           |
| U-233  | 9.4437E-10 | 1.0679E-02              | 1.2292E-02  | 3.6848E-09           |
| U-234  | 3.8957E-05 | 1.8614E-03              | 9.3184E-04  | 1.5694E-04           |
| U-235  | 1.0174E-03 | 6.7618E-03              | 1.4356E-04  | 7.9860E-03           |
| U-236  | 2.3948E-04 | 4.8959E-03              | 1.6267E-05  | 5.4392E-03           |
| U-237  | 1.5658E-06 | 6.6124E-06              | 2.1557E-08  | 8.5699E-06           |
| U-238  | 7.9961E-01 | 1.7777E-01              | 5.9083E-08  | 8.3208E-01           |
| Pu-238                                       | 1.4892E-03 | 1.9306E-04              | 5.3649E-03  | 2.6605E-04           |
| Pu-239                                       | 2.0186E-02 | 1.9387E-03              | 3.4156E-02  | 5.8191E-03           |
| Pu-240                                       | 1.6344E-02 | 7.2889E-04              | 2.8460E-02  | 2.5711E-03           |
| Pu-241                                       | 9.1731E-03 | 6.5728E-04              | 1.5005E-02  | 1.6010E-03           |
| Pu-242                                       | 6.0207E-03 | 3.4178E-04              | 9.7330E-03  | 6.9120E-04           |
| Th-232                                       | 1.3881E-11 | 6.4739E-01              | 7.3773E-01  | 3.5963E-10           |
| Th-230                                       | 1.9528E-10 | 7.6074E-07              | 1.5402E-06  | 1.4819E-09           |
| Am-242m                                      | 2.1801E-05 | 5.0290E-07              | 1.8294E-04  | 1.1967E-06           |
| Xe-135                                       | 8.7474E-09 | 6.9030E-09              | 9.1923E-09  | 7.6692E-09           |

### I. Spent fuel pool criticality

After irradiation, the insertion of 1252 assemblies for each fuel individually was simulated and safety parameters behavior for the different fuels was analyzed. It was obtained the required safe criticality by considering the minimum pitch distance between the assemblies. In Fig. 4 are represented the assemblies for the different fuels.

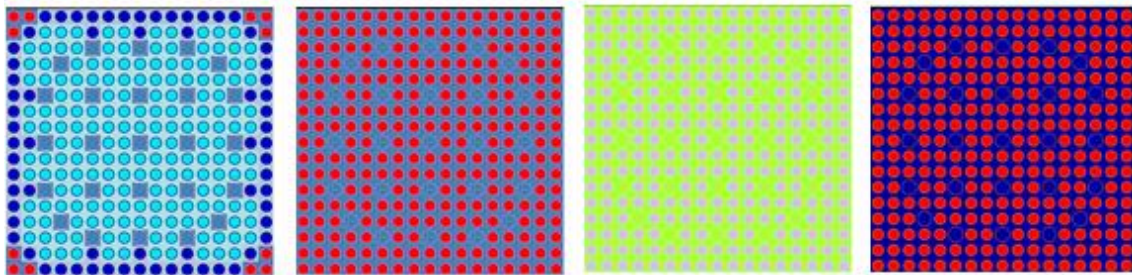


Figure 4: Fuel elements geometry adopted for spent fuel pool criticality analysis.  
 a) MOX, b) ThO<sub>2</sub>-UO<sub>2</sub>, c) TRU-Th and d) UO<sub>2</sub>

Tab. 11 summarizes the  $k_{inf}$  values for the fuels in the pool containing only one type of fuel and  $k_{inf}$  for mixed spent fuel pools.

Tab. 11.  $k_{inf}$  when the fuel assemblies were inserted into the pool.

| Only one fuel in pool | $k_{inf}$             | Mixed-pool ( $\frac{3}{4}$ UO <sub>2</sub> ) | $k_{inf}$             |
|-----------------------|-----------------------|--|-----------------------|
| MOX                   | $0.84409 \pm 0.00032$ | $\frac{1}{4}$ MOX                            | $0.81084 \pm 0.00027$ |
| Th-UO <sub>2</sub>    | $0.44730 \pm 0.00025$ | $\frac{1}{4}$ Th-UO <sub>2</sub>             | $0.75507 \pm 0.00023$ |
| TRU-Th                | $0.93698 \pm 0.00028$ | $\frac{1}{4}$ TRU-Th                         | $0.85139 \pm 0.00026$ |
| UO <sub>2</sub>       | $0.78256 \pm 0.00029$ | --   | --                    |





The values for multiplication factor showed in Tab. 11 are in accordance with curves plotted in Fig. 2 once that follow the same  $k_{\text{eff}}$  descending order.

Fill the  $\text{UO}_2$  fuel in a mixed spent fuel pool, together with MOX or TRU fuels, made the criticality decrease 3.939% and 9.135% respectively. Even though criticality has increased on 68.81% when  $\text{UO}_2$  fuel assemblies were inserted with Th- $\text{UO}_2$  fuel assemblies in the pool, for all three cases of mixed spent fuel pool a  $k_{\text{safe}} \leq 0.95$  remains guaranteed as established by Angra 2, Final Safety Analysis Report [9].

#### 4. CONCLUSIONS

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 reprocessed fuel based on transuranic elements and thorium spiked 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.

#### ACKNOWLEDGMENT

The authors are also grateful to Brazilian research funding agencies, CNEN – Comissão Nacional de Energia Nuclear (Brazil), CNPq – Conselho Nacional de Desenvolvimento Científico e Tecnológico (Brazil), CAPES – Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (Brazil) and FAPEMIG – Fundação de Amparo à Pesquisa do Estado de Minas Gerais (MG/Brazil) for the support. Furthermore, we are also grateful to sponsors and donor volunteers for their support of this event.

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