AN EVALUATION OF THE PWR CYCLE WITH INSERTION OF MAs

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ABSTRACT

The insertion of different percentage values of MAs (minor actinides) in UO$_2$ and MOX fuels was analyzed to a fuel assembly of PWR reactor such as that of ANGRA-I. The main aim is to evaluate the neutronic parameters and the isotopic composition to an extended burnup. The variations in the final fuel composition and in the neutronic parameters for the different percentage of MAs inserted are small. The studies show that it is possible to have an extended burnup to MOX fuel. However, to UO$_2$ type, the fuel temperature coefficient becomes positive in the end of the burnup. Therefore, to UO$_2$ fuel, it is necessary to modify the fuel composition or the geometry to obtain satisfactory results.

1. INTRODUCTION

The strategies for the management of spent fuel in each country vary from reprocessing to direct disposal. However, if no decisions on management strategies are taken, the large amount of spent fuel in storage will continue to increase [1]. The disposition of spent fuel can be either disposal in geological repositories or recycled, through reprocessing techniques. The minor actinides (MAs) from spent fuels are very important in nuclear waste management mainly because of the long-term radiotoxicity due their extremely long half-lives. Theoretical investigations have demonstrated that it is possible to insert MAs in UO$_2$ and MOX fuels and using these fuels in the Angra-I PWR reactor, in Brazil, burning a considerable quantity of actinides during the safe reactor operation [2, 3, 4, 5].

In this work, a 16 x 16 fuel assembly with the geometric characteristics of Angra I reactor [6], has been considered. The neutronic behavior of the fuel pins of such fuel assembly was evaluated considering different percentages of MAs insertion. Eight fuels were analyzed being four with UO$_2$ fuel and four with MOX fuel. The aim is to study the isotopic composition and the neutronic parameters during the burnup evaluating the possibility to reach an extended burnup of 55000 MWd/tHM. The following neutronic parameters were analyzed: infinite multiplication factor, hardening of the spectrum and reactivity temperature coefficients.
2. METODOLOGY

2.1. The code

The simulations were performed using the WIMS (Winfrith Improved Multi-group Scheme) code. This code is applied for nuclear system cells calculations. It allows to consider fuel geometries as cylinders or slabs in homogeneous arrays or in clusters (heterogeneous) and to obtain different approximation for neutronic calculations defined for the user. It is possible also to evaluate the problem with multicells. In such case, the user defines two or more cells and assigns a probability that a neutron leaving one cell enters another cell. The code gives the infinite multiplication factor ($k_{\text{inf}}$) and the effective multiplication factor ($k_{\text{eff}}$). The last one is calculated if buckling data are given. The code generates few group constant taking into account the buckling to be used as input on diffusion codes with multigroup. There is an option to perform investigations of buckling for criticality analyses and reactions rate for many isotopes [7].

2.2. Geometry Model

The geometry of the model takes into account previous study of fuel rod dimensions [6]. In such study, the fuel assembly has 16 x 16 pins being 235 fuel rods, 20 control rod thimbles and 1 instrumentation thimble. The pitch distance is 1.232 cm, the external radius of the fuel rod and of the fuel pin are 0.4572 cm and 0.3923 cm, respectively. In the simulations, the model considers the quantity of the fuel rods in a fuel assembly and in the dimensions of these fuel rods. The problem has been considered 235 equal cells and it representative cell is show in the Fig. 1 bellow.

![Figure 1. The geometry for one fuel pin cell in the WIMS code.](image)

2.3. Fuel composition

The LWR spent fuel was reprocessed by UREX technique [8] where the MAs and uranium recovered were inserted in two types of fuels: UO$_2$ and MOX. Different percentage values of recovered material were evaluated in this study. To values between 1% and 3% the variations in the neutronic parameters behavior were small. Therefore, the percentages of the recovered material inserted in the fresh fuel presented in this work are 1%, 5%, 10% and 15%.

The percentage of fissile material was based in the typical fuel enrichment of the Angra I and in the studies to obtain an extended cycle [6]. The analyzed cases and the compositions for each case are presented in the Table 1 and Table 2, respectively.
Table 1. Percentage of recovered material for both fuel types.

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>1.0%</th>
<th>5.0%</th>
<th>10.0%</th>
<th>15.0%</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO₂+MA</td>
<td>Case 1</td>
<td>Case 2</td>
<td>Case 3</td>
<td>Case 4</td>
</tr>
<tr>
<td>MOX+MA</td>
<td>Case 5</td>
<td>Case 6</td>
<td>Case 7</td>
<td>Case 8</td>
</tr>
</tbody>
</table>

Table 2. Initial composition for the studied cases in weight percentage.

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
<th>Case 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>²³⁵U</td>
<td>1.003 10⁻⁵</td>
<td>5.014 10⁻⁵</td>
<td>1.003 10⁻⁴</td>
<td>1.504 10⁻⁴</td>
</tr>
<tr>
<td>²³⁴U</td>
<td>4.993 10⁻⁶</td>
<td>4.964 10⁻⁶</td>
<td>4.993 10⁻⁶</td>
<td>4.891 10⁻⁶</td>
</tr>
<tr>
<td>²₃⁹U</td>
<td>4.159 10⁻⁶</td>
<td>2.080 10⁻⁶</td>
<td>4.159 10⁻⁵</td>
<td>6.239 10⁻⁵</td>
</tr>
<tr>
<td>²³⁷Np</td>
<td>3.847 10⁻⁴</td>
<td>1.923 10⁻⁴</td>
<td>3.847 10⁻³</td>
<td>5.770 10⁻³</td>
</tr>
<tr>
<td>²³⁹Pu</td>
<td>2.057 10⁻⁴</td>
<td>1.019 10⁻³</td>
<td>2.037 10⁻³</td>
<td>3.056 10⁻³</td>
</tr>
<tr>
<td>²₃⁹Pu</td>
<td>5.945 10⁻³</td>
<td>2.973 10⁻₂</td>
<td>5.945 10⁻²</td>
<td>8.918 10⁻²</td>
</tr>
<tr>
<td>²₃⁹Pu</td>
<td>2.493 10⁻²</td>
<td>1.247 10⁻²</td>
<td>2.493 10⁻²</td>
<td>3.740 10⁻²</td>
</tr>
<tr>
<td>²⁴¹Pu</td>
<td>1.348 10⁻¹</td>
<td>6.738 10⁻¹</td>
<td>1.348 10⁻²</td>
<td>2.022 10⁻²</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>7.096 10⁻⁵</td>
<td>3.548 10⁻⁵</td>
<td>7.096 10⁻⁴</td>
<td>1.064 10⁻⁴</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>4.123 10⁻⁴</td>
<td>2.062 10⁻³</td>
<td>4.123 10⁻³</td>
<td>6.185 10⁻³</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>1.103 10⁻⁶</td>
<td>5.514 10⁻⁶</td>
<td>1.103 10⁻⁵</td>
<td>1.654 10⁻⁵</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>1.548 10⁻⁴</td>
<td>7.742 10⁻⁴</td>
<td>1.548 10⁻³</td>
<td>2.323 10⁻³</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>8.942 10⁻⁹</td>
<td>4.471 10⁻⁸</td>
<td>8.942 10⁻⁸</td>
<td>1.341 10⁻⁸</td>
</tr>
<tr>
<td>²⁴³Cm</td>
<td>4.75 10⁻⁷</td>
<td>2.375 10⁻⁶</td>
<td>4.75 10⁻⁶</td>
<td>7.125 10⁻⁶</td>
</tr>
<tr>
<td>²⁴⁴Cm</td>
<td>3.571 10⁻⁵</td>
<td>1.786 10⁻⁴</td>
<td>3.571 10⁻⁴</td>
<td>5.357 10⁻⁴</td>
</tr>
</tbody>
</table>

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3. RESULTS

3.1. Isotopic composition

The Figures from 2 up to 7 show the weight variation (in percentage) to the isotopes U, Pu, Np, Cm and Am in the beginning (BOC) and in the end of cycle (EOC). The cases from 1 up to 4 presents the UO$_2$ fuel and the cases from 5 up to 8 the MOX fuel both with insertion of reprocessed material. Before this insertion, the UO$_2$ contains only uranium and the MOX presents uranium and plutonium in its composition. Therefore, after the insertion of reprocessed material, the MOX will have more Pu isotopes than the UO$_2$ fuel. This behavior can be verified in the Figures from 2 up to 4 where the MOX fuel presents more Pu isotopes than the UO$_2$ fuel in BOC. The figures show also the decrease of $^{238}$Pu, $^{239}$Pu and $^{240}$Pu to the MOX fuel while there is an increase of the Pu isotopes to the UO$_2$ fuel type. The $^{239}$Pu and $^{241}$Pu are products of the transmutation-decay chain beginning with the fertile isotope $^{238}$U [9]. Therefore, the increase of the Pu isotopes in the UO$_2$ fuel is probably due to the transmutation of $^{238}$U (see Fig. 6). On the MOX fuel, the considerable presence of Pu isotopes in the BOC provides its depletion during the burnup. The $^{238}$Pu and $^{240}$Pu have large thermal cross section to thermal energy and their decrease is caused by the transmutation in $^{239}$Pu and $^{241}$Pu, respectively. The Fig. 4 shows an increase of $^{241}$Pu and the Fig. 2 presents a decrease of $^{239}$Pu to MOX fuel. As the $^{238}$Pu is a fissile material, its isotopic composition decreases during the depletion along the burnup.

The $^{235}$U decreases and the $^{236}$U and $^{237}$U increase for all the cases studied (see Fig. 2 and 5). As the $^{235}$U is a fissile material, its reduction is due to its depletion during the burnup. The radioactive capture of $^{235}$U contributes to increase of $^{236}$U. Moreover, the neutron absorption by the $^{236}$U isotope provides the increase of $^{237}$U (see Fig. 6).

The Fig. 3 shows an increase of $^{237}$Np and $^{239}$Np. After the transmutation-decay chain, the $^{236}$U and $^{237}$U transmute in $^{237}$Np and $^{239}$Np, respectively (see Fig. 6). Therefore, the increase of Np isotopes may be explained by such chain reaction.

The Fig. 5 shows an increase of $^{243}$Am and $^{244}$Cm, caused probably by the increase of $^{242}$Pu (see Fig. 4). The $^{242}$Pu transmutes to $^{243}$Pu by radioactive capture and, after then, a beta decay transmutes $^{243}$Pu in $^{243}$Am. If this last isotope has a new radioactive capture, it transmutes to $^{244}$Am and then, after a beta decay, the $^{244}$Am transmutes in $^{244}$Cm (see Fig. 6). Therefore, the increase of $^{243}$Am and $^{244}$Cm may be also explained by this chain reaction.

There are small variations in the final isotopic composition if considering the different percentages of the reprocessed material inserted. To a same type of the fuel (UO$_2$ or MOX), there is small variation in the final isotopic composition. The cases from 1 up to 4 present practically the same values of weight in percentage at the EOC. The same behavior is observed to the cases from 5 up to 8. Despite this small variation in the final composition, there are cases that present more accumulation or depletion of the studied isotopes. The cases 1 and 5, with 1% of reprocessed material insertion, present higher depletion of $^{238}$Pu, $^{239}$Pu and $^{240}$Pu and higher accumulation of $^{241}$Pu and $^{242}$Pu. The cases 1 and 5 too present the lower accumulation of $^{237}$Np, $^{239}$Np and $^{242}$Cm. In addition, the cases 1 and 8 presents lower accumulation of $^{243}$Am and $^{244}$Cm.
Figure 2. Isotopic composition of $^{235}$U, $^{236}$U and $^{239}$Pu at the BOC and EOC.

Figure 3. Isotopic composition of $^{237}$Np, $^{239}$Np and $^{238}$Pu at the BOC and EOC.
Figure 4. Isotopic composition of $^{240}$Pu, $^{241}$Pu and $^{242}$Pu at the BOC and EOC.

Figure 5. Isotopic composition to $^{237}$U, $^{243}$Am and $^{244}$Cm at the BOC and EOC.
3.2. Neutronic parameters

The Figures from 7 up to 10 present the behavior of the neutronic parameters evaluated. The differences in the UO$_2$ and the MOX compositions cause considerable differences in the neutronic parameters for both types of fuel. However, for a same type of the fuel, the differences in the neutronic parameters behavior are small. In this case, the difference is in the BOC isotopic composition.

The Fig. 7 and 8 show, respectively, the infinity multiplication factor (k$_{inf}$) and the fast to total flux rate ($\phi_F/\phi_T$) during the burnup. These figures present differences in the values of the two neutronic parameters for UO$_2$ (cases from 1 up to 4) and MOX (cases from 5 up to 8) fuels due different isotopic plutonium compositions for both fuels. The MOX has more isotopes of plutonium than the UO$_2$ fuel. The absorption cross section for plutonium isotopes is higher than that for uranium isotopes in thermal spectrum. Consequently, at the BOC, the MOX fuel presents k$_{inf}$ values lower than the UO$_2$ one. However, the decrease of the k$_{inf}$ values during the burnup is lower to the MOX fuel. This behavior is due to $^{239}$Pu to have low $\eta$ value in comparison with the $^{235}$U and due the transmutation of $^{240}$Pu in the fissionable $^{239}$Pu [9]. In addition, the neutrons fission spectrum is more energetic for the $^{239}$Pu than for the $^{235}$U. Consequently, the MOX fuel presents values of $\phi_F/\phi_T$ rate higher than the UO$_2$ one, that is, the MOX spectrum is more hardened than that for the UO$_2$ type, as it is clearly demonstrated in the Fig. 8.

To UO$_2$ fuel, the increase of reprocessed material percentage in the composition causes an increase of Pu isotopes. In the other hand, to MOX fuel, the increase of reprocessed material percentage generates a reduction of such isotopes in the composition (see Fig. 3 and 4). As mentioned before, the plutonium isotopes provide less variation in the k$_{inf}$ values and cause hardening of the spectrum. Therefore, to UO$_2$ fuel, the increase of the reprocessed material produces less variation in the k$_{inf}$ values and generates a hardening of the neutron spectrum.

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In the other hand, to MOX fuel, the increase of the reprocessed material causes considerable variation in the $k_{\text{eff}}$ values and generates a softening of the neutron spectrum. These behaviors are shown in the Figs. 7 and 8.

The Fig. 9 and 10 show the behavior of the reactivity temperature coefficient to fuel ($\alpha_{TF}$) and moderator ($\alpha_{TM}$). To analyze the $\alpha_{TF}$ and $\alpha_{TM}$ parameter the corresponding temperatures were increased in 10K. The $\alpha_{TM}$ parameter remained negative during the complete burnup for all the analyzed cases. For the UO$_2$ fuel, the $\alpha_{TF}$ parameter increased during the burnup becoming positive after 49500 MWd/thM limiting the burnup. Because of the large epithermal absorption resonances for the plutonium isotopes, the moderator and fuel temperature coefficients tend to be more negative for MOX in comparison with the UO$_2$ fuel [8]. This behavior can be verified in the Figures 9 and 10. To a same type of the fuel (UO$_2$ or MOX), the different percentages of reprocessed material insertion cause small variations in the $\alpha_{TF}$ and $\alpha_{TM}$ parameters. To UO$_2$ fuel, increasing the percentage of reprocessed material insertion causes values a few more negatives to $\alpha_{TF}$ and $\alpha_{TM}$ along the burnup. To MOX fuel, increasing the percentage of reprocessed material insertion causes values more negatives to $\alpha_{TM}$ and more positives to $\alpha_{TF}$.

![Figure 7. Infinity multiplication factor versus burnup.](image-url)
Figure 8. Fast to total flux ratio versus burnup.

Figure 9. Fuel moderator coefficient versus burnup.
In this work, it was verified that it is possible to reach an extended burnup for the MOX fuel with MAs insertion. The UO$_2$ fuel presents positive $\alpha_{TF}$ values after 49500 MWd/tHM, limiting the burnup as it was verified in the analyses. Therefore, for the UO$_2$ fuel, it is necessary to change the composition or the geometry of the fuel assembly to obtain an extension of burnup.

The different percentages of the reprocessed material inserted in the same type of fuel (MOX or UO$_2$) generate small variations in final isotopic compositions and in the neutronic parameters behavior. In spite of this, increasing the insertion of reprocessed material in the UO$_2$ fuel causes decrease in the $k_{\text{inf}}$ values, hardening of the spectrum and values more negatives to $\alpha_{TF}$ and $\alpha_{TM}$ parameters. To MOX fuel, the increase of insertion of reprocessed material in the composition generates small increase in the $k_{\text{inf}}$ values, a softening of the spectrum and values more positives for $\alpha_{TF}$ and more negatives to $\alpha_{TM}$. In this way, the MOX presents better characteristics to an extended burnup in comparison with the UO$_2$ fuel. The fuels with less amount of reprocessed material insertion (cases 1 and 5) presented higher depletion of $^{238}\text{Pu}$, $^{239}\text{Pu}$ and $^{240}\text{Pu}$, lower accumulation of $^{237}\text{Np}$, $^{239}\text{Np}$ and $^{242}\text{Cm}$, but had higher accumulation of $^{241}\text{Pu}$ and $^{242}\text{Pu}$.

The present work was performed using the WIMS-D5 code for multi-cell of the fuels. In future studies will be investigated the behavior of fuel assembly with mixed fuels. In addition, other nuclear code will be used for to evaluate the reactor core behavior filled with $\frac{1}{3}$ or more of fuel with MAs insertion.
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REFERENCES