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Neutronic analysis of a U–Mo–Al fuel and europium as burnable poison

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ABSTRACT

This work presents the neutronic analysis of the U–Mo–Al dispersion fuel concerning uranium density increase and shows comparisons relatively to the U_3Si_2–Al fuel. The U_3Si_2–Al uranium density varied from 3.0 to 5.5 g U/cm^3 while that of U–Mo–Al fuel varied from 4.0 to 7.5 g U/cm^3. The molybdenum mass content in the former case varies from 7% to 10% in mass. Here, it is also proposed the utilization of burnable poison nuclides in the U–Mo–Al fuel meat. Since the fuel is metallic, gadolinium and europium were chosen as candidates to cope with this task. A recently developed cell code at IPEN (HRC) composed of the coupling of the codes HAMMER-TECHNION for the cell analysis, ROLAIDS for the actinide self-shielding calculations and CINDER-2 for the actinide and fission transmutation was employed for the neutronic analyses of U–Mo–Al. The simulated reactor core was similar to the one of RMB (Brazilian Multipurpose Reactor) composed of an array of 5×5 positions with 23 fuel elements and 2 aluminum blocks. A second analysis of the europium case employed the SERPENT code in an explicit core modeling. The burnup calculations were performed considering a power of 30 MW during three cycles of RMB and 30 days. The analyses revealed that the molybdenum content has a great impact in the core reactivity due to its high absorption cross section. A value of 7% was found adequate for the molybdenum mass content. The analyses also reveal that europium is a better burnable poison than gadolinium for the core cycle length and power level under consideration. However, for the U–Mo–Al case, k_∞ increases up to a maximum value and decreases afterwards. This is a striking result since the reactivity for the U–Mo–Al fuel does not increase steadily as verified for the U_3Si_2–Al case. Beyond a certain uranium density, the reactivity will decrease making useless the addition of more uranium.

1. Introduction

Due to the restrictions imposed to the commerce and utilization of highly enriched nuclear fuels (enrichment higher than 20% in mass of ^{235}U), the goal for fuels that allow higher uranium densities for utilization in research reactor has been of major concern. The United States launched the RERTR (Reduced Enrichment for Research and Test Reactors) in 1978 having the main objective the development of the necessary technologies for the conversion of HEU (high enriched fuel) employed in research reactors to LEU (Low Enriched Fuel).

Besides of new materials, new fabrication techniques had to be developed, giving rise to the dispersion fuels, where an uranium compound is homogeneously dispersed in aluminum. Powder technology was employed to cope with this task since it was impossible to combine these new materials starting from their alloy form. In addition to the homogeneous distribution of the fissile phase, it was possible to reach concentrations that did compromise the subsequent fabrication steps employing this fuel manufacture technique. However, the uranium concentration in the phase volume had to be limited to 45% in volume (Tissier, 1991) in order to guarantee the mechanical integrity of the plates.

The International Meeting on Reduced Enrichment for Research and Test Reactor held in Buenos Aires, Argentina, in 1987 presented several related contributions that gave as qualified the dispersions UAl_x–Al, U_3O_8–Al and U_3Si_2–Al respectively with the following densities, 2.3 g U/cm^3, 3.2 g U/cm^3, and 4.8 g U/cm^3, all with 42.5% in volume.

Even with these qualified fuels, high flux research reactors like ATR with a power of 250 MW, as an example, need high enriched

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fuel for its operation. The main fuel currently in study to convert these reactors to low enrichment fuels is the uranium–Molybdenum (U–Mo) alloy.

2. Objectives of this work

The main objective of this work is to analyze the neutronic aspects of the dispersion fuel U–Mo–Al for future utilization in the Brazilian Multipurpose Reactor (RMB) (Perrotta and Obadia, 2011). The power of this reactor is 30 MW and its typical neutron thermal flux in the core is 1.80×10^{14} neutron/cm^2-s. The analyses employed the HAMMER-TECHNION (Barhein et al., 1978), ROLAIDS (Greene et al., 1976) and CINDER-2 (England et al., 1976) – HRC, CITATION (Fowler et al., 1971) and SERPENT (Leppänen, 2013) computer codes and the emphasis will be placed on the uranium density increases in this type of fuel. The molybdenum content will range from 7% to 10%. The final results will be compared to those of U_3Si_2 fuel. Since the U–Mo fuel is metallic, this work also proposes the utilization of a metallic element added directly in the fuel meat that has neutronic properties of burnable poison. Due to their high (n,γ) cross sections, gadolinium and europium were the chosen elements to cope with this task.

3. U–Mo properties

The uranium alloys that show very promising results against irradiation are those that can be maintained in their cubic crystalline structure (gamma phase). Alloys that satisfy this requirement and have tendency to form gamma phase are: U–Cr, U–Mo, U–Nb, U–Re, U–Ru, U–Ti, U–Zr, etc. Among those, the alloy that shows a large extension of the γ phase is the U–Mo alloy. Below 560 °C, a stable structure of the U–Mo is a mixture of alpha phase (α – U orthorhombic) and γ' phase (metastable). However, applying a rapid cooling in the gamma phase, the U–Mo alloy preserves the metastable phase. The U–Mo alloy maintains the metastable phase during the fuel fabrication and under irradiation and shows good compatibility to the aluminum. The U–Mo alloy is currently studied to be the fuel for the research reactors in the future.

The U–Mo alloy is obtained by the fusion of uranium with molybdenum, obtaining a U–Mo ingot. The molybdenum content generally studied in the U–Mo alloys are 7% (U-7 wt% Mo) or 10% (U-10 wt% Mo) in mass.

There are several techniques to obtain the U–Mo powder. Brasil has the hydriding–dehydriding technique originally developed in CNEA, Argentine (Balart et al., 2000). In the hydriding technique, the U–Mo ingot is heated in a atmosphere with hydrogen gas. In an adequate temperature, the ingot absorbs the hydrogen and gets weakened. After that, the ingot is dissolved. After the hydriding, the hydrogen is removed by heating the powder in a vacuum environment (dehydriding), getting as final product just the powder.

Among several others, the major advantages of this fuel alloy are: it allows a higher uranium density relatively to the current fuels, it allows a better efficiency in the fuel reprocessing and it can be fabricated either as a dispersion fuel (employing a U–Mo powder) or as monolithic U–Mo plate (Lopes et al., 2012).

The neutronic behavior of the U–Mo fuel is strongly dependent to the molybdenum content since the Mo absorption cross section is considerably higher mainly relative to that of silicon. Fig. 1 shows the comparison of the molybdenum and silicon absorption cross sections (Cross Section Plotter, Accessed in: 2015).

The U–Mo alloy research was launched by the RERTR program in the mid-80s. This type of alloy allows an uranium density of about 8 g U/cm^3 in its disperse phase. However, the irradiation tests detected a layer of interaction between uranium and aluminum, produced by the diffusion of aluminum particles into the U–Mo particles. As a consequence, the fuel swallowed and lost the thermal conductivity beyond a burnup of 60%.

Current studies aim to solve the aluminum diffusion problems in the fuel meat. The results found so far indicated as one of possible solutions the addition of silicon in the matrix, stabilizing and reducing the interaction problems. However, there is no clear limit of silicon content in the matrix. The researchers in general are being performed with values between 3% and 7% of Si (Ryu et al., 2010). Another difficult in the determination of silicon content is the increase of interaction layer during irradiation which induces a silicon dilution and a consequent loss of efficiency.

Besides of silicon addition, another important parameter to reduce the layer interaction is the size of U–Mo particles employed in the dispersion (Ryu et al., 2011).

The U–Mo fuel also allows the monolithic form (laminar plate of U–Mo). The thickness of this plate ranges between 0.254 and 0.381 mm making it possible to get densities close to 16 g U/cm^3. Of vital importance of this type of study is the conversion of high performance research reactors as the "Advanced Test Reactor" – ATR of Idaho National Laboratory in Idaho. This type of reactor can operate in a power of 250 MW (Glagolenko et al., 2010).

4. Research reactors

Material Testing Reactors are of vital importance for the radioisotope production and supply and research in general. The world faces currently great difficulties of radioisotope production and supply, mainly concerning molybdenum ^{99}Mo which is the source of ^{99}Tc. Several projects of multipurpose reactors are underway around the world. In this context, two examples of such reactors, one in operation and the other under construction, can be mentioned:

The reactor Open Pool Australian Lightwater (OPAL) started its operation in August 2006. This reactor has a power of 20 MW and utilizes low enriched (LEU) U_3Si_2–Al, it is moderated by light water (H_2O), its reflector is heavy water(D_2O) and its operational cycle ranges between 30 and 35 days. The OPAL reactor has two objectives: radioisotope production and scientific and industrial researches (Storr, 2009).

The Jules Horowitz Reactor (JHR) is being built in Cadarache, France under an international collaboration. The JHR thermal power is 100 MW and it will be moderated and cooled by light water. The future fuel of the JHR will be U–Mo with a density of 8 g/cm^3. Since the U–Mo (either dispersion or monolithic) is still in a developing phase and it will not be in the startup of the JHR reactor. The solution was to employ high enriched U_3Si_2–Al (27% de ^{235}U) (The Jules Horowitz Reactor (JHR), Accessed in: 12 January 2015).

The Brazilian Multipurpose Reactor (RMB) is under design in Brazil and pursues the same purposes as the reactors mentioned previously. The RMB will utilize light water (H_2O) as moderator and its core will be reflected by heavy water (D_2O). Its operational cycle is 30 days in a power of 30 MW. The fuel of RMB will be U_3Si_2–Al. As in OPAL, the RMB will employ cadmium wires inside of the fuel elements as burnable poisons.

5. Burnable poisons

Burnable poisons are nuclides that have high absorption cross section and produce as a results of these reactions nuclides with lower absorption cross sections. The utilization of burnable poisons in a reactor is of vital importance since it lowers the initial reactivity excess and burns as the reactor operates; allowing a desirable operational cycle. The decrease of the reactivity at the beginning

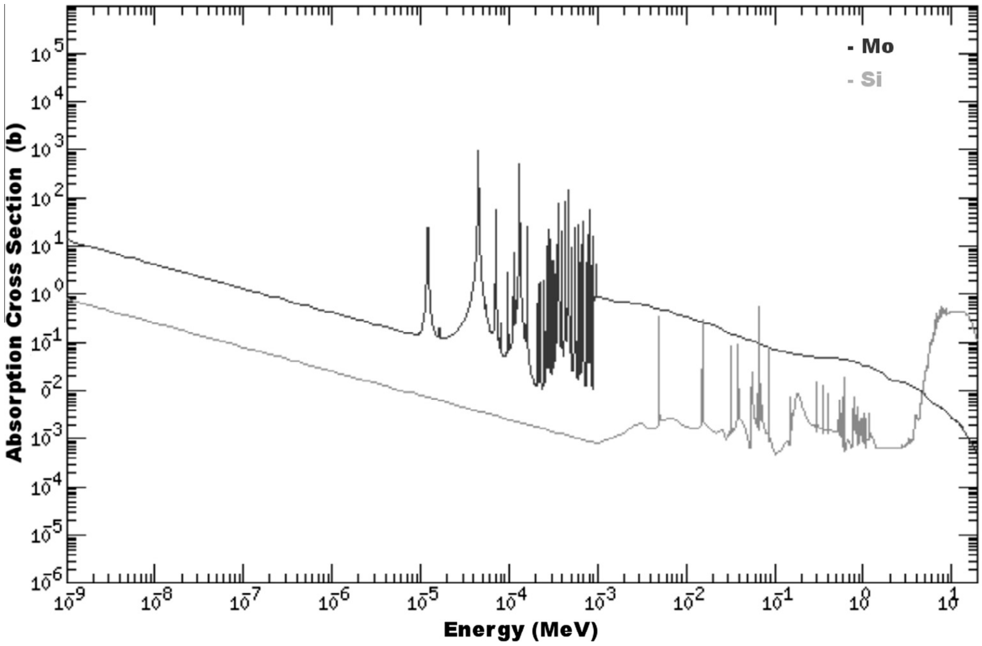

Fig. 1. Comparison of molybdenum and silicon absorption cross sections.

of cycle is of vital importance for the control rod worth safety requirements.

Examples of materials utilized as burnable poison are:

- Gadolinium oxide Gd_2O_3 (applied in PWR's).
- Borated compounds as: B_4C, ZrB_2 and Al–B alloys.
- Cadmium wires inside of the fuel elements (as in OPAL reactor and future RMB).

The U–Mo alloy allows the addition of a metallic element that has properties of burnable poison. Since this burnable poison will be mixed in the meat of the fuel, the desired reaction is radiative capture (n, γ). Table 1 shows the main candidates that pursue these characteristics:

Table 1 shows that the melting point of all elements are close to those of uranium and molybdenum and that the elements that has higher absorption cross section are gadolinium (Gd) and europium (Eu). Due to the thermal neutron flux level of RMB (1.80×10^{14} neutron/cm^2-s) the candidate elements to be used as burnable poison in the study of U–Mo alloys under consideration section are gadolinium (Gd) and europium (Eu).

6. Description of the reactor considered in the analysis

The reactor model for the analyses performed in this work will be based in the reactors mentioned previously. The simulated core contains 23 fuel elements in a square array of 25 positions (5 × 5). Two aluminum blocks simulate the in core irradiation positions. The radial reflector is composed of 50 cm of heavy water (D_2O) and 40 cm of light water. The radial view of the core configuration is shown in Fig. 2.

The fuel element consists of 21 fuel plates coated by aluminum as shown in Fig. 3. The fuel meat is 6.5 cm wide, 61.5 cm long, and 0.061 cm thick. The total thickness of the fuel plates is 0.150 cm and the moderator channel width is 0.245 cm.

The simulations considered a constant power of 30 MW and a cycle length of 30 days. These are the power and cycle length of RMB. Most of the simulation will be performed in time period of 97 days which correspond closely to 3 cycles of the reactor operation. Three cycles is the maximum period of residence of the fuel element in the core. The axial fuel active length was divided into

Table 1
Possible burnable poison (Lamarsh, 1983).

Element	Absorption cross section (σ_a)	Melting point (°C)
Dysprosium	930	1680
Erbium	162	1529
Europium	4600	826
Gadolinium	49,000	1312
Hafnium	102	2233

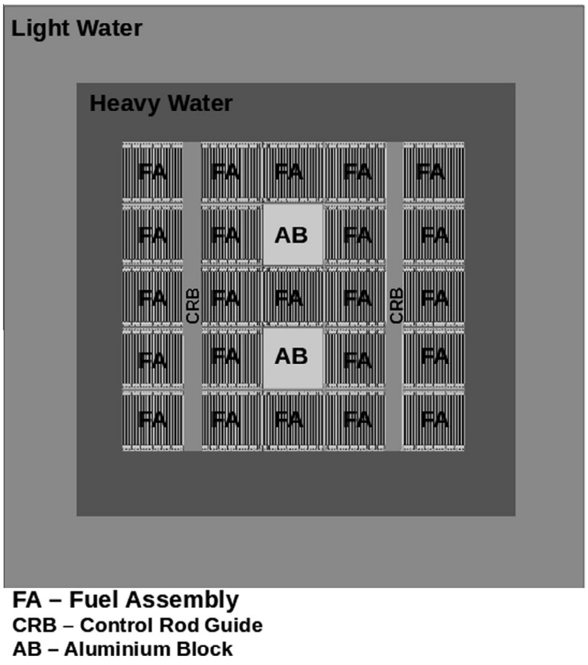

Fig. 2. Reactor core (radial view).

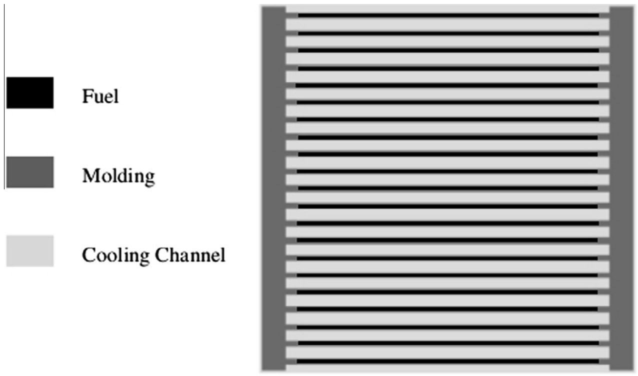

Fig. 3. Fuel element (*XY* view).

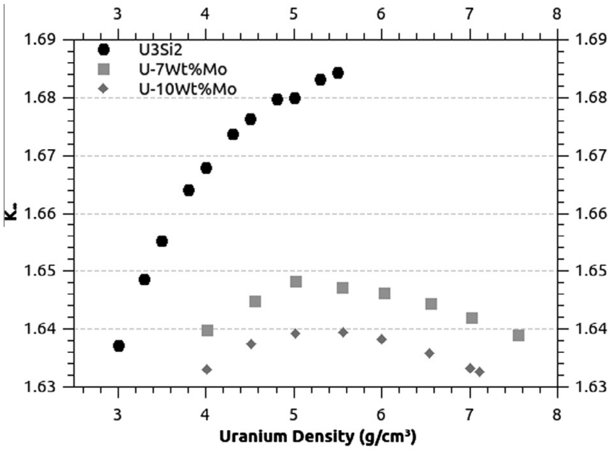

Fig. 4. k_∞ of the U_3Si_2–Al and U–Mo–Al fuels as a function of the uranium density.

four burnup regions in each of which the transmutation analyses of the actinides and fission products were taken into consideration. The ex-core irradiation channels and the structural parts of the core were not considered in the simulations since the neutronic behavior of the fuel elements was of major concern. The axial reflector was considered as pure water.

7. Computer codes utilized in the simulations

The homogenized fuel element cross sections were generated employing the HRC code. This computer program is the result of the coupling of: HAMMER-TECHNION, ROLAIDS and CINDER-2. HAMMER-TECHNION solves the transport equation in an unit fuel cell and an extra region that contains the other components of the fuel element and generates the homogenized few-group cross sections needed for subsequent codes. ROLAIDS performs the mutual shielding of the actinides present in the fuel cell and CINDER performs the transmutation analysis of actinides and fission products. Four group cross sections were employed in the whole analysis.

The computer code CITATION (Fowler et al., 1971) was employed for the selection of the appropriated europium mass content in the U–Mo–Al fuel. The nuclear data in four groups for each burnup step were generated by HRC. The nuclear data for the non-fuel region were generated AMPX-II (Greene et al., 1976).

The computer code SERPENT (Leppänen, 2013) was employed for the final burnup analysis of the U–Mo–Al fuel with europium as burnable poison element. This computer code is being developed at the Technological Research Center (VTT) located in Finland since 2004.

All calculations were performed employing the ENDF/B-VII.0 nuclear data library.

8. The neutronic analyses

8.1. Comparison of the U_3Si_2–Al and U–Mo–Al fuels

The first analysis considers the comparison of the U_3Si_2–Al and U–Mo–Al (with 7% and 10% mass content of Mo) fuels changing the uranium density. These analyses were performed with HRC. Fig. 4 shows the final results for the fuel element infinity multiplication factor (k_∞).

Fig. 4 shows that the k_∞ behavior as a function of the uranium density is very different for the fuel types considered here. Initially, considering the same uranium density, k_∞ for the U–Mo–Al fuel is lower than that of U_3Si_2–Al since the molybdenum absorption cross section is higher than that of silicon. Second, the k_∞ increases steadily for the U_3Si_2–Al case. However, for the U–Mo–Al case, k_∞ increases up to a maximum value and decreases afterwards. This is a striking result since the reactivity for the U–Mo–Al fuel will not increase steadily as verified for the U_3Si_2–Al case. Beyond a certain uranium density, the reactivity will decrease making useless the addition of more uranium.

The burnup behavior of the fuel element infinity multiplication factor (k_∞) for the U_3Si_2 fuel (4 g U/cm^3) and U–Mo–Al with uranium densities of 4, 5, and 7 g U/cm^3 are shown in Fig. 5. These results were obtained once again by HRC.

Fig. 5 shows that considering the same uranium density (4 g U/cm^3) for both fuels, the U–Mo–Al fuel show a systematic underprediction for the fuel element k_∞ due to the Mo presence in the fuel meat. However, when the uranium density is increased, the fuel element k_∞ starts lower than that of the U_3Si_2 and U–Mo–Al with 4.0 g U/cm^3 and ends up with a higher k_∞ after 97 days of burnup. This result is a consequence of the better conversion ration of the U–Mo–Al fuel. Due to that the ^{239}Pu production for the U–Mo–Al fuel will be higher than that of U_3Si_2 fuel. This is a very important result in favor of the U–Mo–Al because although starting with a higher uranium density and a lower k_∞, there is a net gain of reactivity at the end of 97 days relatively to the both cases of both fuels U_3Si_2 fuel and U–Mo–Al with 4.0 g U/cm^3.

8.2. Burnable poison analyses

The burnable poison material was added to the fuel meat. Its mass content is considered relatively to the uranium density. The amount of the burnable poison material inserted is compensated

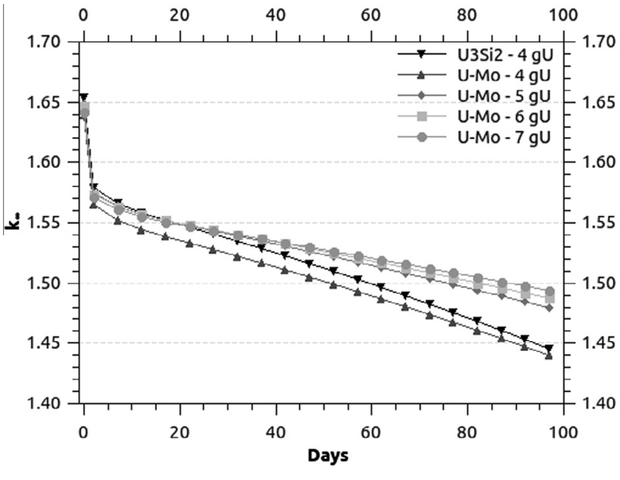

Fig. 5. k_∞ comparison of the U_3Si_2 and that of U–Mo–Al fuels.

by removal of aluminum in the fuel meat so that its total volume is preserved. The goal here is to choose the uranium density and burnable poison concentration so that the beginning of cycle k_{eff} falls around 1.10. The ex-core irradiation channels and the structural parts of the core were not considered in the simulations presented here. As in OPAL, these extra regions will induce a negative reactivity of around 3500 pcm. Taking this aspect into consideration, the initial core reactivity will be close to 6500 pcm. This will abbreviate the control rod requirements. Another desired requirement is to have its reactivity excess as close as possible to 6500 pcm (Xe free) during the core burnup process. As a result of several trials, the chosen uranium density considering gadolinium as a burnable poison is 5.5 g U/cm^3 and for the europium case is 3.5 g U/cm^3.

The first material analyzed as burnable poison was gadolinium (Gd). Gadolinium has a very high absorption cross section; 49,000 b (Lamarsh, 1983). The gadolinium transmutation chain is shown below:

$$^{154}_{64}\text{Gd} \rightarrow {}^{155}_{64}\text{Gd} \rightarrow {}^{156}_{64}\text{Gd} \rightarrow {}^{157}_{64}\text{Gd} \rightarrow {}^{158}_{64}\text{Gd} \rightarrow {}^{159}_{64}\text{Gd} \rightarrow {}^{160}_{64}\text{Gd}.$$

The isotopes that have the major cross sections are $^{155}_{64}$Gd and $^{157}_{64}$Gd.

The gadolinium content in the fuel was changed from 0% to 0.10%. The analysis considered the uranium density 5.55 g U/cm^3 and the molybdenum mass content 7%. The results of the simulations are shown in Fig. 6.

Fig. 6 shows that the initial reactivity of the cycle is strongly affect by the gadolinium content. Only gadolinium mass contents between 0.010% and 0.020% minimize this effect. Other problem in the utilization of gadolinium as burnable poison in the reactor and cycle under consideration is that the gadolinium burnout occurs rapidly and it does not flat the reactivity curve during the cycle length. The reactivity after a few days of operation drops steadily up to the end of cycle. Consequently for the purposes under consideration of this work, gadolinium will not be a good choice as burnable poison material.

The second burnable poison analyzed here is europium. This element has two isotopes ^{151}Eu and ^{153}Eu and has an absorption cross section of 4600 barns (Lamarsh, 1983). Europium has an absorption cross section smaller than that of gadolinium, but still high enough to be considered as a candidate for a burnable poison material. The transmutation chain of europium is shown below:

$$^{151}_{42}\text{Eu} \rightarrow {}^{152}_{42}\text{Eu} \rightarrow {}^{153}_{42}\text{Eu} \rightarrow {}^{154}_{42}\text{Eu} \rightarrow {}^{155}_{42}\text{Eu} \rightarrow {}^{156}_{42}\text{Eu} \rightarrow {}^{157}_{42}\text{Eu} \rightarrow {}^{158}_{42}\text{Eu}.$$

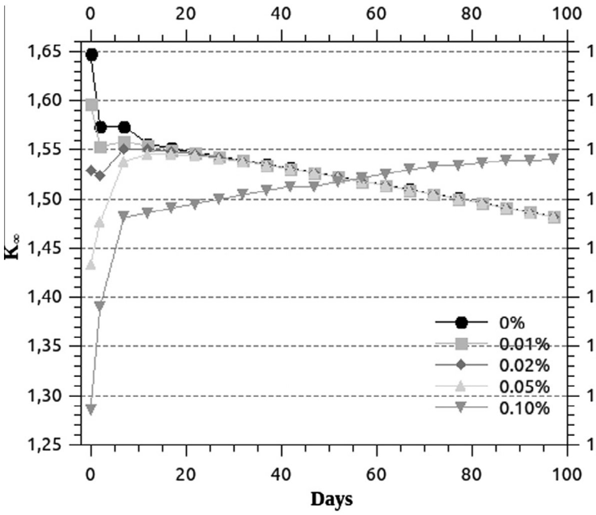

Fig. 6. Behavior of the fuel element k_∞ considering gadolinium as a burnable poison.

For this specific case, the computer code CITATION was employed in a tridimensional simulation for the reactor cycle (30 days) and power (30 MW). The europium mass content varies from 0.0% (no europium) to 0.30%. The uranium density was 3.55 g U/cm^3 and the molybdenum content was 7%. Fig. 7 shows the final results for the effective multiplication factor (k_{eff}) calculated by CITATION for several europium concentrations.

Comparing the cases with and without europium, Fig. 7 shows that there are some residual reactivity loses after nearly 45 days due to the cumulative production of the $_{42}{}^{154}$Eu, $_{42}{}^{155}$Eu, $_{42}{}^{156}$Eu, $_{42}{}^{157}$Eu, and $_{42}{}^{158}$Eu. The residual part of $_{42}{}^{153}$Eu also contributes to this reactivity lose due to its high absorption cross section. Fig. 7 shows that europium attends the desired reactivity characteristics up to 30 days of burnup. The reactivity variation for the first 30 days of operation and after the ^{135}Xe equilibrium is small enough and attends the desired goal of small variation. These results show that from the neutronic point of view and for the reactor with the characteristics shown previously, europium attends the necessary requirements to be considered a good burnable poison material.

SERPENT was employed to calculation first reactor cycle. In this case an europium mass content of 0.25% was adopted. Fig. 8 shows the final results.

The simulations presented here did not take into consideration many reflector components like irradiation positions, beam holes and cold neutron source. These items are of fundamental importance for a multipurpose reactor. These components absorb nêutrons and decrease the reactivity of the reactor. Like in OPAL, here 3500 pcm of reactivity is reserved for these components. The SERPENT reactivity values already discounting 3500 pcm are shown in Fig. 9.

Fig. 9 shows that at beginning of cycle, the reactivity is around 6000 pcm which allows the attendance of the safety requirements of the control banks with a good margin. The minimum reactivity requirement is 1100 pcm which is high to compensate for any uncertainty in the design calculations and for the control of the reactor core. Furthermore, Fig. 9 also shows that it is possible to have an operational cycle beyond 30 days. The analysis made so far is somehow qualitative because one type of fuel element is taken into consideration. The real core will have more than one type of fuel densities. Three types of fuel densities is generally considered enough for flatten the power and optimization of the fuel burnup in the equilibrium cycle.

A final aspect is the isothermal reactivity coefficient for the U–Mo–Al fuel and europium mass content under consideration here.

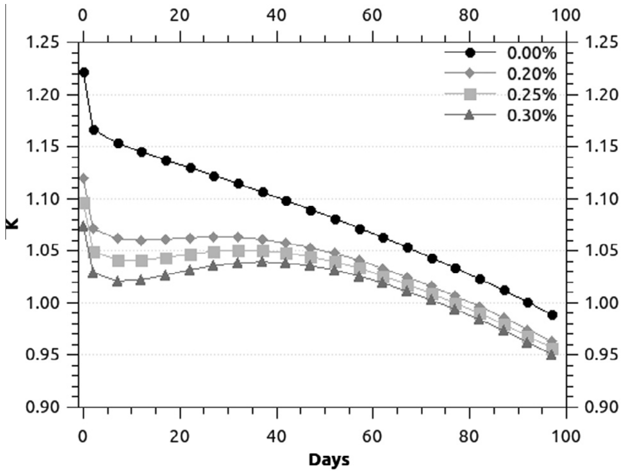

Fig. 7. k_{eff} behavior considering europium as a burnable poison.

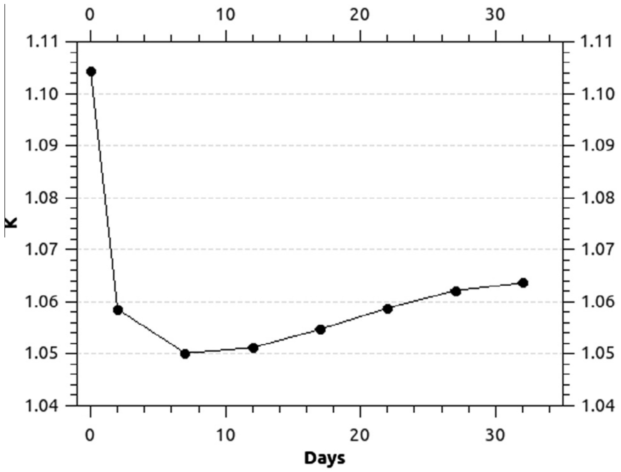

Fig. 8. k_{eff} in the SERPENT for the case U–7 wt% Mo with 3.55 g U/cm^3 and 0.25% of Eu.

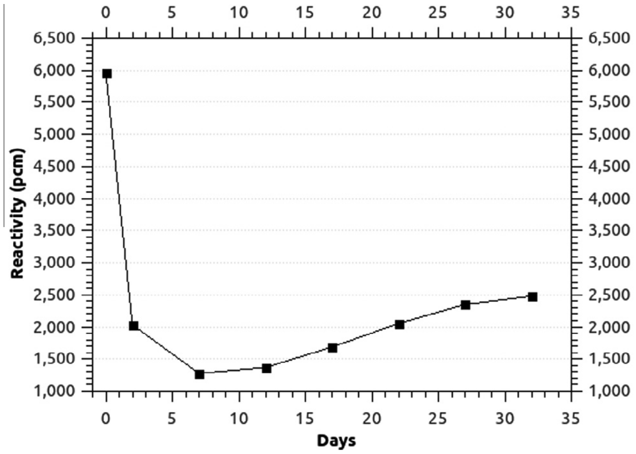

Fig. 9. Reactivity behavior for the U–7 wt% Mo fuel with 3.55 g U/cm^3 and 0.25% of Eu.

A SERPENT analysis at beginning of life show that the average isothermal reactivity coefficient between de 20 and 50 °C is −8.5 ± 0.5 pcm/°C which is a good value for the fuel, burnup poison and core under consideration here.

9. Conclusions

This work shows that from the neutronic point of view, it is possible to utilize U–Mo fuel in a typical multipurpose reactor. The recommended molybdenum mass content is 7% since its absorption cross section is considerable. The analyses reveal that the reactivity behavior of the U–Mo fuel as a function of the uranium density reaches a maximum value beyond of which is not recommended to increase the uranium density since there will not be any gain in the reactivity.

This work also purpose a burnable poison material directly in the U–Mo alloy. The analyses reveal that europium is a good material to cope with this task for the reactor power and cycle length under consideration in this study. The results show that it is possible to have 30 days cycle length in a full power of 30 MW.

There will be a need for some further studies to optimize the reactor core including other fuel elements with different uranium densities to optimize the core neutronic characteristics and fuel cycle economy.

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