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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