NEUTRONIC ANALYSIS FOR PRODUCTION OF FISSION MOLYBDENUM-99 AT IEA-R1 AND RMB RESEARCH REACTORS

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ABSTRACT

The IEA-R1 reactor of IPEN-CNEN/SP in Brazil is a pool type research reactor cooled and moderated by demineralized water and having Beryllium and Graphite as reflectors. In 1997 the reactor received the operating licensing for 5 MW. A new research reactor is being planning in Brazil to replace the IEA-R1 reactor. This new reactor, the Brazilian Multipurpose Reactor (RMB), planned for 30 MW, is now in the conception design phase. Low enriched uranium (<20% ^{235}U) targets (UAl_x dispersed in Al and metallic U foils) are being considered for the fission Molybdenum-99 (^{99}Mo) production. Neutronic calculations were performed to compare the production of ^{99}Mo for the two types of targets under similar conditions of irradiation (irradiation position, neutron flux and power density) both in the IEA-R1 reactor and RMB.

1. INTRODUCTION

^{99m}TC, product son of ^{99}Mo, is one of the most utilized radioisotopes in nuclear medicine in the world. Annually it is used in approximately 20 to 25 million procedures of medical diagnosis, representing about 80% of all the nuclear medicine procedures. Since 2004, given the worldwide interest in ^{99}Mo production, the International Atomic Energy Agency (IAEA) has developed and implemented a Coordinated Research Project (CRP) [1] to help interested countries start a small-scale domestic ^{99}Mo production in order to meet the requirements of the local nuclear medicine. The purpose of CRP is to provide interested countries with access to non-proprietary technologies and methods for production of ^{99}Mo targets using thin foils of low enriched metallic uranium (LEU), UAl_x-Al miniplates of LEU type, or by neutron activation reaction (n, gamma), for example, using gel generators. Brazil, through IPEN-CNEN/SP, began its CRP participation in late 2009. IPEN-CNEN/SP provides radiopharmaceuticals to more than 300 hospitals and clinics in the country, reaching more than 3.5 million medical procedures per year. The use of radiopharmaceuticals in the country over the last decade has grown at a rate of 10% per year and IPEN-CNEN/SP is primarily responsible for this distribution. ^{99m}Tc generators are the most used ones and are responsible for more than 80% of the radiopharmaceuticals applications in Brazil. IPEN-CNEN/SP imports all the ^{99}Mo used in the country (450 Ci of ^{99}Mo per week or 24,000 Ci per year approximately). In the past, IPEN-CNEN/SP developed the ^{99}Mo production route from neutron activation of ^{98}Mo targets in the IEA-R1. However, the quantity produced does not meet the Brazilian needs of this isotope. Due to the growing need for nuclear medicine in the country and because of the short ^{99}Mo supply observed since 2008 on the world stage, IPEN/CNEN-SP has decided to develop its own project to produce ^{99}Mo through ^{235}U fission. This project has three main goals: 1) the research and development of ^{99}Mo production from fission of LEU targets, 2) the discussion and decision on the best production route technique, and 3) the feasibility study of IPEN/CNEN-SP in reaching a routine production of ^{99}Mo. The main goal of IPEN-CNEN-SP is to accommodate the Brazilian demand for radiopharmaceuticals. Nowadays, this demand is about 450 Ci of ^{99}Mo per week and the future need, after seven years, is estimated at around 1,000 Ci per week. One of the analyses planned in this project is to study the characteristics and specifications of UAl_x-Al targets and of metallic uranium thin foils. The aim of the present work was to perform preliminary neutronic calculations to evaluate the ^{99m}Mo production through fission at the IPEN/CNEN-SP IEA-R1 nuclear reactor and at the Brazilian Multipurpose Reactor (RMB), which is in the conceptual design phase.

2. Target description used in neutronic analysis

The UAl_x-Al targets of LEU type proposed and analyzed in this work are aluminum coated miniplates. Each miniplate measures 52 x 170 mm, 1.52 mm thick, corresponding to a total volume of 13.437 mm^3. The UAl_x-Al core fuel is 40 x 118 mm, 0.76 mm thick, leading to a total volume of 3.587 mm^3. Considering this volume and a ^{235}U mass in the target equals to 2.01 g, the ^{235}U density (ρ_{U-235}) in the target core is 0.58 $g^{235}U/cm^3$. For a 19.9% ^{235}U enrichment, the uranium density in the target is ρ_U = 2.91 gU/cm^3. This corresponds to a UAl_2 volume fraction of 45% and an aluminum volume fraction of 55% in the dispersion. A special Miniplate Irradiation Device (MID) was designed for the UAl_x-Al miniplates irradiation in the IEA-R1 reactor. Figure 1 shows the MID which has the external dimensions of the IEA-R1 fuel element. The miniplates will be allocated in a box with indented bars placed inside the external part of the MID. Figure 2 shows the MID cross section. As seen from Figure 2, up to ten UAl_x-Al miniplates can be placed in the box with indented bars inside of the MID.

Fig 1: Miniplate Irradiation Device – MID.

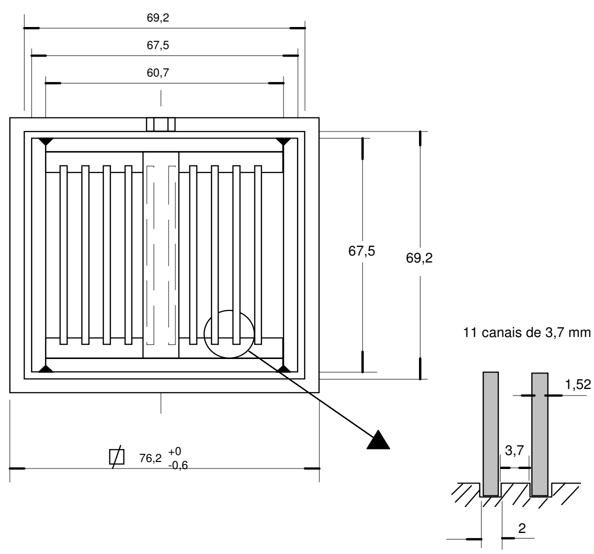
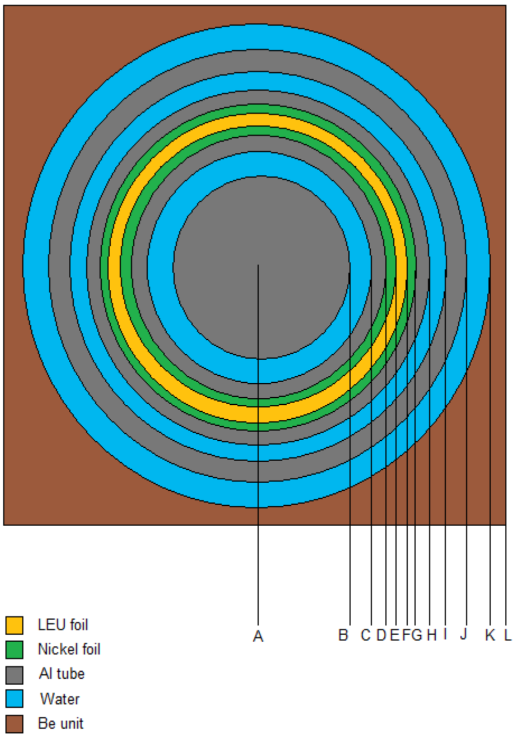


Fig 2: Cross section of the MID (dimensions in mm).

The targets of metallic U foils analyzed at IPEN/CNEN-SP were based on targets that were examined in the Tajoura reactor in Libya to produce ^{99}Mo [2]. The targets were mounted in cylindrical geometry, in a tubular arrangement. The metallic U foil was covered with a Ni sheet before being placed concentrically inside the aluminum tubes. The dimensions of the target are:

1. One foil of uranium (LEU) of 44 cm x 76 mm x 135 µm;
2. Coating nickel foil of 15 µm thickness;
3. Two aluminum cylinder having 44 cm length, outside diameters of 27.99 and 30.00 mm, and inside diameters of 26.21 and 28.22 mm, respectively.
4. ^{235}U mass of 20.1 g, same mass of 10 miniplates irradiated in the MID, with 19.9% enrichment of ^{235}U.

For the performed calculations, the U-Ni target was located in the same irradiation device showed in Figure 2, whose external dimensions are 76.2 mm x 76.2 mm x 88.74 cm (with nozzle). Figure 3 shows the removed Be unit with LEU target and irradiation device horizontal cross section. A mass equals to 20.1 g of ^{235}U in the metallic U foils was considered for the neutronic calculations.



Radius	Lenght (cm)
AB	1.00
AC	1.322
AD	1.394
AE	1.396
AF	1.4095
AG	1.411
AH	1.5
AI	1.75
AJ	1.9
AK	2.2
AL	3.81

Fig 3: The removed Be unit with LEU target and irradiation device horizontal cross section.

4. Target analysis

The IEA-R1 and RMB reactors cores, as well as the UAl_x-Al and the U-Ni targets used for the ^{99}Mo production, were modeled with the HAMMER-TECHNION [3] and CITATION [4] numerical codes. The 1D cross sections for each component of the two reactors and the power distribution for any position r of the reactor core matrix plates were obtained. The SCALE 6.0 [5] code system was used to perform burnup calculations for each target and also to determine the ^{99}Mo activity at the end of irradiation. The target irradiation times for each reactor were defined according to their current and planned operating cycle. The IEA-R1 reactor has a 5x5 configuration, containing 24 MTR-type fuel elements with a beryllium radiator at its central position. The UAl_x-Al and U-Ni targets were modeled and simulated in the core central position using 24 U_3Si_2-Al fuel elements whose density was 1.2 gU/cm^3. The target irradiation time was three (3) days. At the end of irradiation, the total activity obtained for the 10 UAl_x-Al and the U-Ni targets were 581.49 Ci and 1,275.8 Ci, respectively. According to its initial conceptual design, RMB is an open pool type, multipurpose, 30 MW thermal power reactor. The RMB core has a 5x6 configuration with MTR-type U_3Si_2-Al fuel elements with 19.75 wt% uranium-235 enrichment. The reactor core is light water cooled and moderated, using heavy water and beryllium as reflectors. The UAl_x-Al and U-Ni targets were modeled and simulated in a peripheral core position at the heavy water reflector using 30 U_3Si_2-Al fuel elements whose density is 1.9 gU/cm^3. At the end of 7 days of irradiation, the total activity obtained for the 10 UAl_x-Al and the U-Ni targets were 2,070.69 Ci and 4,409.83 Ci, respectively.

5. Conclusion

From the neutronic calculations presented here, we conclude that for the same amount of uranium in the analyzed targets (20.1 g) and the same irradiation conditions, a higher total ^{99}Mo activity was obtained for the U-Ni targets. In the IEA-R1 case, the total ^{99}Mo activity calculated at the end of the irradiation period for U-Ni targets was 1,275.8 Ci, while for the Al-UAl_x targets it was 581.89 Ci. For the RMB, the total ^{99}Mo activity obtained at the end of the irradiation time was 4,409.83 Ci for the Ni-U targets and 2,070.69 Ci for the UAl_x-Al ones. Initially, ^{99m}TC generators will be distributed five (5) days after the end of the irradiation. Consequently, the total ^{99}Mo activity is expected to reach values of 361.35 Ci and 164.7 Ci for the U-Ni and the UAl_x-Al targets irradiated in the IEA-R1, respectively. For the U-Ni and UAlx-Al targets irradiated in the RMB, the total ^{99}Mo activity at the distribution time is expected to be 1,249.06 Ci and 583.51 Ci, respectively. From these values, it is noted that the Brazilian current demand of 450 Ci of ^{99}Mo per week and the future projected demand of 1,000 Ci may only be addressed by the RMB reactor under conception.

6. References

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