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LOW ENRICHED URANIUM UAL_X-AL TARGETS FOR THE PRODUCTION OF MOLYBDENUM-99 IN THE IEA-R1 AND RMB 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 planned 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 (LEU) (<20% ^{235}U) UAl_x dispersed in Al targets are being considered for production of Molybdenum-99 (^{99}Mo) by fission. Neutronic and thermal-hydraulics calculations were performed, respectively, to compare the production of ^{99}Mo for these targets in IEA-R1 reactor and RMB and to determine the temperatures achieved in the UAlx-Al targets during irradiation. For the neutronic calculations were utilized the computer codes HAMMER-TECHNION, CITATION and SCALE and for the thermal-hydraulics calculations was utilized the computer code MTRCR-IEAR1.

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 [1]. 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) [2] 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 using targets of thin foils of metallic low enriched 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 six 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. The first aim of the present work was to perform neutronic calculations to evaluate the ^{99m}Mo production through fission at the IPEN/CNEN-SP IEA-R1 nuclear reactor and at the RMB, which is in the conceptual design phase. The second aim of this work is to perform thermal-hydraulics calculations to determine the maximal temperatures achieved in the targets during irradiation and compared them with the design temperature limits established for UAlx-Al targets.

2. UAL_X-AL TARGETS USED IN THE NEUTRONIC AND THERMAL-HYDRAULIC ANALYSES

The UAl_x-Al targets of LEU type proposed and analyzed in this work are aluminum coated miniplates. Each miniplate measures 52 mm x 170 mm, 1.52 mm thick, corresponding to a total volume of 13.437 mm^3. The UAl_x-Al meat is 40 mm 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 meat 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 irradiation of the UAl_x-Al targets in the IEA-R1 reactor and in the RMB. 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 targets can be placed in the box with indented bars inside of the MID.

The UAl_x-Al targets were modeled and simulated, respectively, in the core central position in the IEA-R1 reactor, and in a peripheral core position, in the heavy water reflector, in the RMB. The target irradiation times for each reactor were defined according to their current and planned operating cycle.

Figure 1: Miniplate Irradiation Device – MID.

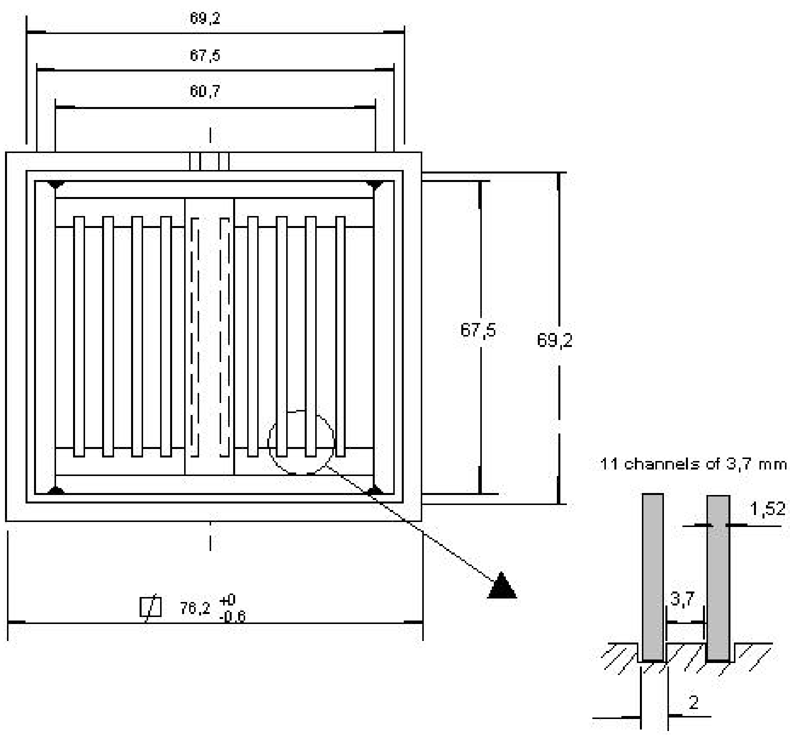

Figure 2: Cross section of the MID (dimensions in mm).

3. NEUTRONIC CALCULATION FOR THE IRRADIATION DEVICE

The IEA-R1 and RMB reactor cores, as well as the UAl_x-Al targets used for the ^{99}Mo production, were modeled with the HAMMER-TECHNION [3] and CITATION [4] numerical codes. The 1D cross section for each component of the two reactors and the power distribution for any position r of the reactor cores 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 IEA-R1 reactor has a 5x5 configuration, containing 24 MTR-type fuel elements with a beryllium irradiation device at its central position. The UAl_x-Al 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. At the end of 3 irradiation days, the total ^{99}Mo activity obtained for the 10 UAl_x-Al was 632.58 Ci.

According to its first 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% ^{235}U enrichment. The reactor core is light water cooled and moderated, using heavy water and beryllium as reflectors. The UAl_x-Al targets were modeled and simulated in a peripheral core position in 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 irradiation days, the total ^{99}Mo activity obtained for the 10 UAl_x-Al targets were 5,987 Ci.

4. THERMAL HYDRAULICS CALCULATION FOR THE IRRADIATION DEVICE

A thermal-hydraulics model MTCR-IEA-R1 [6] was developed in 2000 at IPEN-CNEN/SP using a commercial program Engineering Equation Solver (EES). The use of this computer model enables the steady-state thermal and hydraulics core analyses of research reactors with MTR fuel elements. The following parameters are calculated along the fuel element channels: fuel meat central temperature (T_c), cladding temperature (T_r), coolant temperature (T_f), Onset of Nucleate Boiling (ONB) temperature (T_{onb}), critical heat flux (Departure of Nucleate Boiling-DNB), flow instability and thermal-hydraulics safety margins MDNBR and FIR. The thermal-hydraulics safety margins MDNBR and FIR are calculated as the ratio between, respectively, the critical heat flux and the heat flux for flow instability and the local heat flux in the fuel plate. Furthermore, the MTCR-IEA-R1 model also utilizes in its calculation the involved uncertainties in the thermal-hydraulics calculation such as: fuel fabrication uncertainties, errors in the power density distribution calculation, in the coolant flow distribution in the core, reactor power control deviation, in the coolant flow measures, and in the safety margins for the heat transfer coefficients. The calculated thermal-hydraulics core parameters are compared with the design limits established for MTR fuels: a) cladding temperature < 95°C; 2) safety margin for ONB > 1.3, or the ONB temperature higher than coolant temperature; 3) safety margin for flow instability > 2.0; and 4) safety margin for critical heat flux > 2.0. For the targets, it was considered the following design limits: 1) no material may experience a temperature greater than ½ any target material melting temperature. The lowest melting temperature for any of the proposed target materials is that of the aluminum cladding, whose melting temperature is 660°C. Therefore 330°C is the maximum allowable temperature for the LEU target; 2) the pool coolant must be kept below its saturation temperature. In this work it was adopted as target design limit the cladding temperature that initiated the coolant nucleate boiling (T_{ONB}) for a given coolant pressure and superficial heat flux given by Bergles and Rosenow correlation [7].

The placement of the MID in the core central position of IEA-R1 reactor will deviate part of the reactor coolant flow to cool the UAlx-Al targets. The flow rate in the core of the reactor IEA-R1 is 3,400 gpm which provides flow rates of approximately 20 m^3/h per fuel element, sufficient to cool a standard fuel element. In order to evaluate the temperatures achieved in the UAlx-Al targets, different coolant velocities were tested through the MID. Table 1 provide the calculated target temperatures for different coolant velocities through the MID in the IEA-R1 reactor core. The simulations have considered the MID with ten identical UAlx-Al miniplates. Table 1 shows that even velocities smaller than 5 m/s in the MID is sufficient to cool the targets and represent only a small deviation of the total coolant flow rate in the reactor core. For this velocity no design limit was achieved. The calculated cladding temperatures are below the value of 123°C, indicating one-phase flow through the targets. It will be necessary to fabricate a coolant flow restrictor in order to maintain the desired coolant velocity in the MID (see Figure 1).

Table 1: Target temperatures versus different coolant velocities for the IEA-R1 reactor.

Coolant velocity (m/s)	UAlx-Al meat central temperature (°C)	Aluminum cladding temperature (°C)	T_{onb} (°C)	Coolant temperature (°C)
5	81	73	123	49
6	76	69	123	48
7	73	66	123	47
8	71	63	123	46
9	69	61	123	46
10	67	60	123	45
11	66	58	123	45
12	65	57	123	45
13	64	56	123	45
14	63	55	123	44
15	62	55	123	44

The same calculation was repeated for the RMB. Table 2 provides the calculated target temperature results for different coolant velocities through the MID placed in the peripheral core position in the heavy water reflector. The simulations have also considered the MID with ten identical UAlx-Al miniplates. Table 2 shows that a velocity of 12 m/s is necessary to cool the targets. For this velocity no design limit was achieved for the analyzed irradiation device. The calculated cladding temperatures are below the value of 136°C, indicating one-phase flow through the targets.

Table 2: Target temperatures versus different coolant velocities for the RMB.

Coolant velocity (m/s)	UAlx-Al meat central temperature (°C)	Aluminum cladding temperature (°C)	T_{onb} (°C)	Coolant temperature (°C)
5	259	212	136	86
6	238	190	136	78

7	222	174	136	73
8	209	162	136	69
9	199	152	136	66
10	190	143	136	63
11	183	138	136	62
12	177	134	136	60
13	172	125	136	58
14	170	121	136	57
15	166	118	136	56

5. CONCLUSIONS

From the neutronic calculations presented here, for the uranium amount of 20.1 g in the analyzed targets a ^{99}Mo activity of 632.58 Ci was obtained for the 3 days irradiation in the IEA-R1 core. For the targets irradiated in the RMB, the ^{99}Mo activity obtained at the end of 7 days irradiation time was 5,987 Ci. 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 179 Ci and 1,695.65 Ci for UAl_x-Al targets irradiated in the IEA-R1 and RMB, 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 under conception.

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