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COMPARISON OF LOW ENRICHED URANIUM (UAlx-Al AND U-Ni) TARGETS WITH DIFFERENT GEOMETRIES FOR THE PRODUCTION OF MOLYBDENUM-99 IN THE RMB

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ABSTRACT

The Brazilian Multipurpose Reactor (RMB), now in the conception design phase, is being designed in Brazil to attend the demand of radiopharmaceuticals in the country and conduct researches in various areas. The new reactor, planned for 30 MW, will replace the IEA-R1 reactor of IPEN-CNEN/SP. Low enriched uranium (<20% ^{235}U) UAl_x dispersed in Al (plate geometry) and metallic uranium foil targets (plate and cylinder geometries) are being considered for production of Molybdenum-99 (^{99}Mo) by fission. Neutronic and thermal-hydraulics calculations were performed to compare the production of ^{99}Mo for these targets in the RMB. For the neutronic calculations were utilized the computer codes HAMMER-TECHNION, CITATION and SCALE and for the thermal-hydraulics calculations were utilized the computer code MTRCR-IEAR1 and ANSYS CFX.

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 and of metallic uranium thin foils. The first aim of the present work was to perform neutronic calculations to evaluate the ^{99m}Mo production through fission 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 e uranium thin foils targets.

2. UAL$_X$-AL AND URANIUM THIN FOILTARGETS 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 Irradiation Device (ID) was designed for the irradiation of the UAl_x-Al targets in the RMB (Figure 1), whose external dimensions are 76.2 mm x 76.2 mm x 88.74 cm. The miniplates will be allocated in a box with indented bars placed inside the external part of the ID. Figure 2 shows the ID 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 ID.

The targets of metallic Uranium foils with cylinder geometry analyzed at IPEN/CNEN-SP were based on targets that were examined in the Tajoura reactor in Libya to produce ^{99}Mo [3]. 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 (see Figure 3):

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, with 19.9% enrichment of ^{235}U.

Figure 1: Miniplate Irradiation Device – ID.

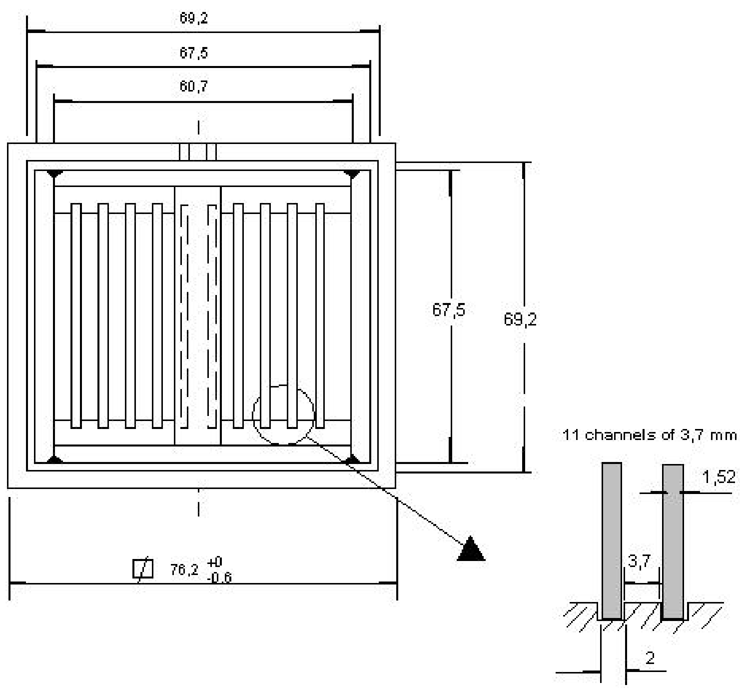
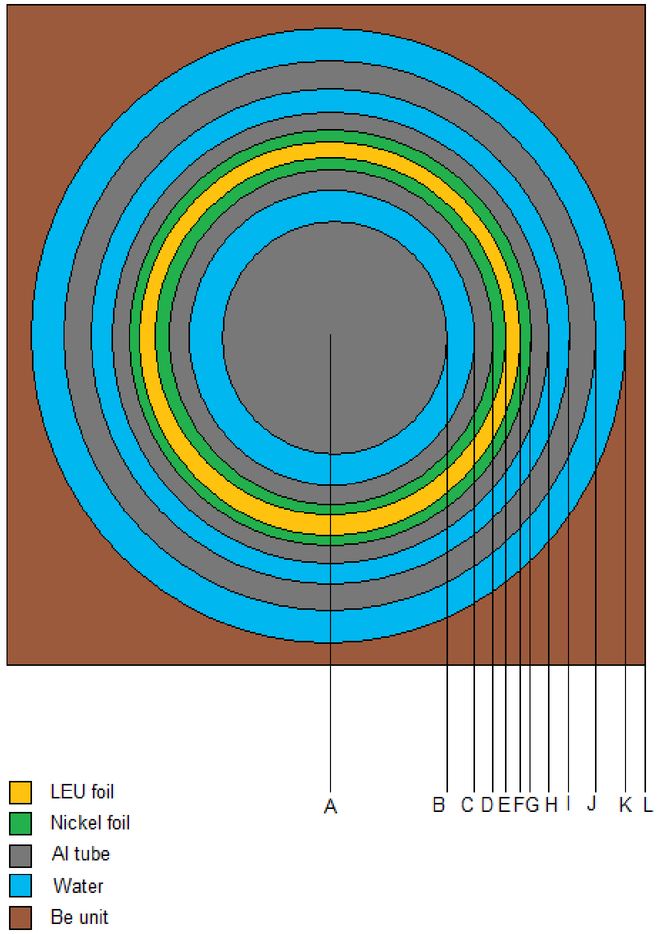


Figure 2: Cross section of the ID (dimensions in mm).



Radius	Length (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

Figure 3: Irradiation device horizontal cross section for the U-Ni target with cylinder geometry.

The targets of metallic Uranium foils with plate geometry were based on targets that were examined in the Paskitan research reactor [4] and consists of a uranium foil (19.99% ^{235}U) with a thickness of 125 µm enveloped in 15 µm thick nickel foil and placed between two aluminum plates that are welded from all sides. The geometry of the foil plate target is shown in Figures 4 and 5.

For the performed calculations, the U-Ni targets, cylindrical and plate geometries, were modeled in the same irradiation device utilized for the calculations of the UAlx-Al targets. . Figures 2 shows, respectively, the ID horizontal cross sections for the U-Alx-Al targets and for U-Ni targets with plate geometry. Figure 3 shows the ID cross section for the U-Ni target with cylinder geometry.

The targets were modeled and simulated in peripheral core position, in the heavy water reflector. The target irradiation time was defined according to their current and planned operating cycle.

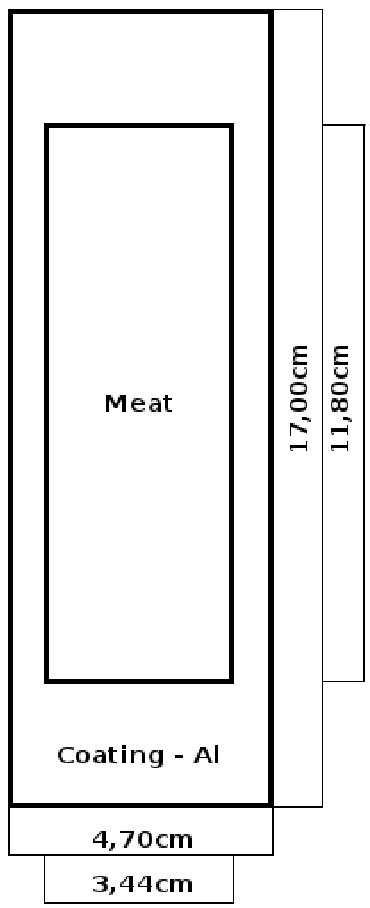


Figure 4: U-Ni LEU target with plate geometry.

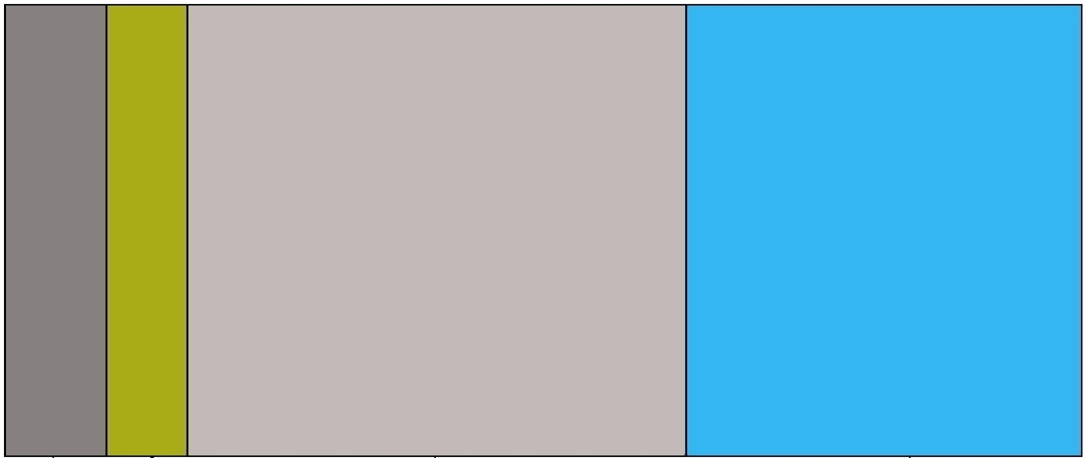


Figure 5: U-Ni LEU target with plate geometry.

3. NEUTRONIC CALCULATION FOR THE IRRADIATION DEVICE

The RMB core, as well as the UAlx-Al and U-Ni cylinder and plate geometries targets used for the ^{99}Mo production, were modeled with the HAMMER-TECHNION [5] and CITATION [6] numerical codes. The 1D cross section for each component of the reactor and the power distribution for any position r of the reactor core matrix plate were obtained. The SCALE 6.0 [7] code system was used to perform burnup calculations for each target and also to determine the ^{99}Mo activity at the end of irradiation. 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 UAlx-Al and U-Ni 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 was 1.9 gU/cm^3. At the end of 7 days of irradiation, the total activity obtained for the UAlx-Al targets were 5,987 Ci. For the U-Ni targets with plate and cylinder geometries the total activities were, respectively, 3,439 Ci and 4,607 Ci.

4. THERMAL HYDRAULICS CALCULATION FOR THE IRRADIATION DEVICE

A thermal-hydraulics model MTCR-IEA-R1 [8] 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 [9].

In order to evaluate the temperatures achieved in the targets different coolant velocities were tested through the irradiation device (ID). For the temperature calculations of the UAlx-Al targets the thermal-hydraulics model MTCR-IEA-R1 was used and the results were obtained simultaneously with the RMB core analysis. The same procedure was used to calculate the temperatures achieved in the U-Ni target with plate geometry. For the calculation of the temperatures of the U-Ni targets with cylindrical geometry was utilized the software ANSYS CFX [10]. The power density (25 KW/cm³) calculated in the ID position in the RMB reflector with the code MTCR-IEAR1was utilized as input date to determine the temperatures in the U-Ni target with cylindrical geometry.

Table 1 provides the calculated UAlx-Al target temperature results for different coolant velocities through the ID placed in the peripheral core position in the heavy water reflector. The simulations considered the ID with ten identical UAlx-Al miniplates. Table 1 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 1: 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

Tables 2 and 3 provide the calculated U-Ni target temperatures for different coolant velocities through the ID in the RMB peripheral position respectively for plate and cylindrical geometries. Table 2 presents for the U-Ni target with cylindrical geometry the temperature of the aluminum tube only.

Table 2: Calculated temperatures for the U-Ni target with plate geometry versus different coolant velocities through the ID.

Coolant velocity (m/s)	U-NI meat central temperature (°C)	Aluminum cladding temperature (°C)	T_{onb} (°C)	Coolant temperature (°C)
5	165	160	132	67
6	149	144	132	63
7	137	133	132	60
8	128	124	132	57
9	122	117	132	55
10	115	110	132	54
11	110	106	132	53
12	105	101	132	52
13	103	98	132	51
14	100	96	132	51
15	98	93	132	50

Table 3: Aluminum tube temperatures for the U-Ni target with cylindrical geometry versus different coolant velocities through the ID.

Coolant velocity (m/s)	Aluminum cladding temperature (°C)
5	166
6	149
7	137
8	127
9	119
10	113
11	107
12	103
13	99
14	95
15	92
16	90

Table 2 provides the calculated target temperature results for different coolant velocities through the ID placed in the peripheral core position in the heavy water reflector. A velocity of 8 m/s is necessary to cool the targets. For this velocity no design limit was achieved for the analyzed irradiation device. The calculated aluminum cladding temperatures are below the value of 132°C, indicating one-phase flow through the U-Ni targets with plate geometry.

Table 3 provides the calculated U-Ni aluminum tube temperatures for different coolant velocities through the ID placed in the peripheral core position in the heavy water reflector . A velocity of 9 m/s is necessary to cool the target. For this velocity no design limit was achieved for the analyzed irradiation device. The calculated aluminum tube temperatures are below the value of 132°C, indicating one-phase flow through the U-Ni target with cylinder geometry.

5. CONCLUSIONS

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 UAlx-Al targets. The total ^{99}Mo activity obtained at the end of the 7 days irradiation time was 5,987 Ci. For the U-Ni targets with plate and cylindrical geometries the calculated total ^{99}Mo activity was, respectively, 3,439 Ci and 4,607 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 a value of 1,695.65 Ci for UAl_x-Al targets irradiated in the RMB. For the U-Ni targets with plate and cylinder geometries the total ^{99}Mo activity is expected to reach values of 974 Ci and 1305 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 be addressed using either UAlx-Al or U-Ni targets.
Through the thermal-hydraulics calculations it was determined a minimum flow for cooling the targets. No design limit was achieved for the analyzed irradiation devices. The calculated cladding temperatures are below the value of 95°C, and the coolant temperatures are below the ONB temperature, indicating one-phase flow through the irradiation devices.

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