LOW ENRICHED URANIUM FOIL TARGETS WITH DIFFERENT GEOMETRIES FOR THE PRODUCTION OF MOLYBDENUM-99 IN THE RMB

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ABSTRACT

A new research reactor is being planned in Brazil to take care of the demand of radiopharmaceuticals in the country and conduct research in various areas. This new reactor, the Brazilian Multipurpose Reactor (RMB), planned for 30 MW, is now in the conception design phase. Two low enriched ($<20\%$ ^{235}U) metallic uranium foil targets (cylinder and plate 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 and to determine the temperatures achieved in the targets. For the neutronic calculations were utilized the computer codes HAMMER-TECHNION, CITATION and SCALE and for the thermal-hydraulics calculations were utilized the computer codes MTRCR-IEA-R1 and ANSYS CFX.

1. INTRODUCTION

^{99m}TC, decay product 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 metallic uranium thin foils targets. 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 U-Ni targets.

2. U-Ni TARGETS USED IN THE NEUTRONIC AND THERMAL-HYDRAULIC ANALYSES

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 1):

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.

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 Figure 2.

For the performed calculations, the U-Ni targets, cylindrical and plate geometries, were modeled in the same irradiation device, whose external dimensions are 76.2 mm x 76.2 mm x 88.74 cm. Figures 1 and 3 show the irradiation device horizontal cross sections for the U-Ni targets with cylinder and plate geometries, respectively.

For both targets a ^{235}U mass equals to 20.1 g was considered for the neutronic calculations. As seen from Figure 3, ten U-Ni targets with plate geometry were placed in the box with indented bars inside of the irradiation device. Each U-Ni target with plate geometry has a ^{235}U mass equals to 2.01 g.

The targets were modeled and simulated in a peripheral core position, in the heavy water reflector. The target irradiation time was seven (7) days.
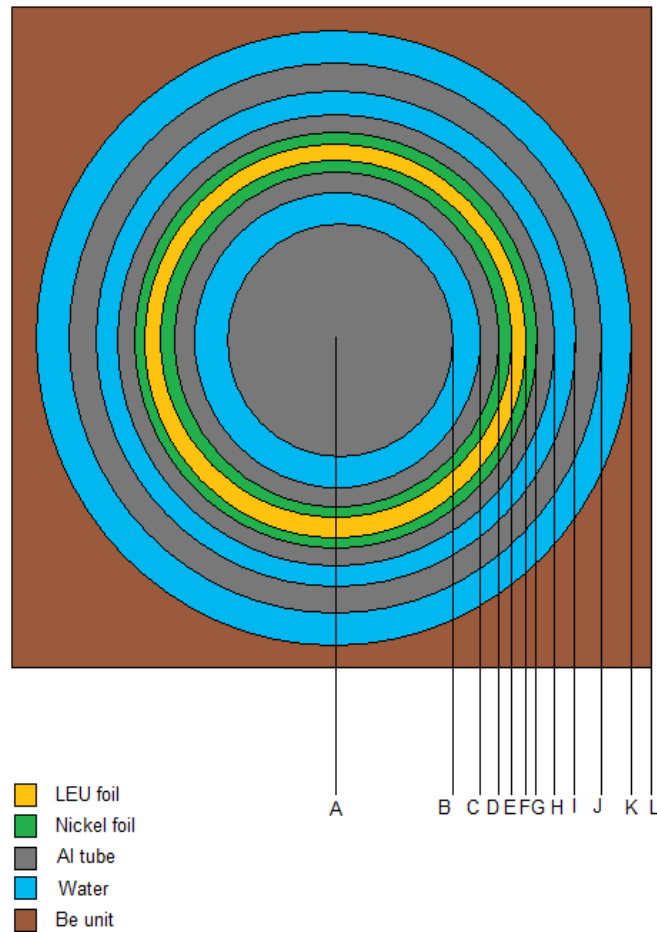
Radius	Length (cm)
AB	1.00
AC	1.322
AD	1.394
AE	1.396
AF	1.4095
AG	1.411
AH	1.5
AI	1.75
AJ	1.9
AK	2.2
AL	3.81

Figure 1: Irradiation device horizontal cross section for the U-Ni target with cylinder geometry.
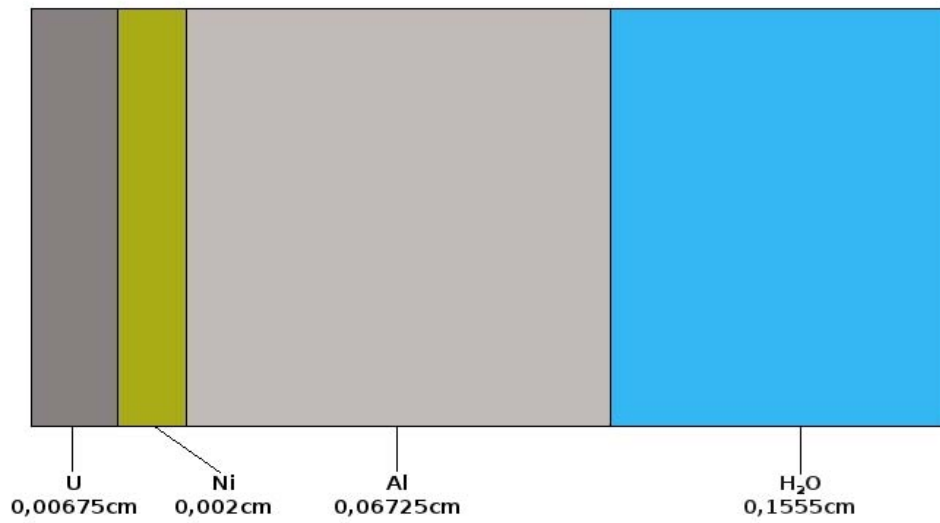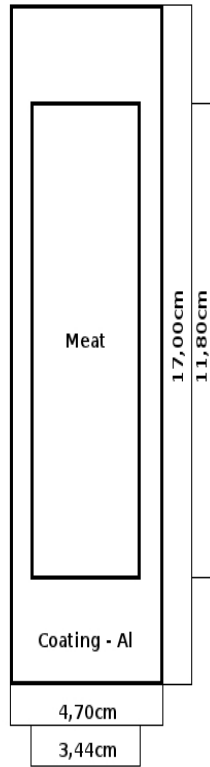
Figure 2: U-Ni LEU target with plate geometry.
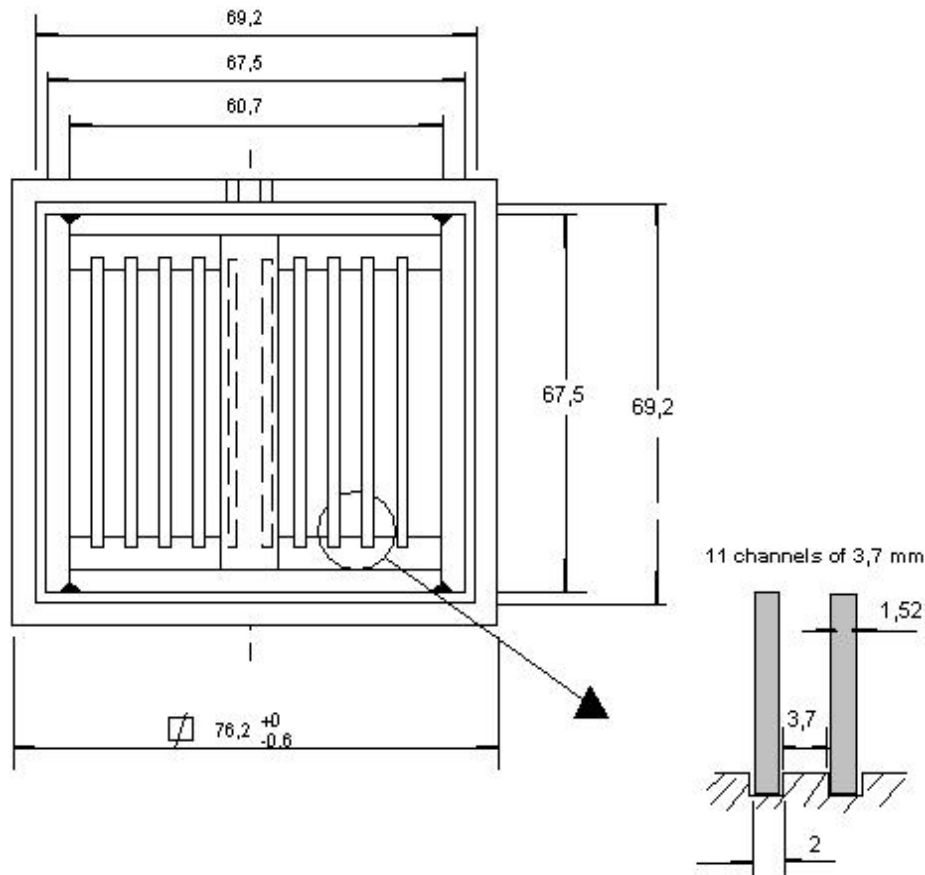
Figure 3: Irradiation device horizontal cross section for the U-Ni targets with plate geometry.

3. NEUTRONIC CALCULATIONS FOR THE IRRADIATION DEVICE

The RMB core, as well as the U-Ni LEU targets (cylinder and plate geometries) 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 was generated by the computer code HAMMER-TECHNION. The computer code CITATION was used for the threedimensional core and radial and axial density curves calculations. These data were used as input data for the thermal-hydraulics irradiation device analysis. The power distribution for any position r of the reactor core matrix plate was 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. The target irradiation time for the reactor was defined according to their current and planned operating cycle. 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 U-Ni LEU 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 activities obtained for the U-Ni plate and cylinder geometries were, respectively, 3,439 Ci and 4,607 Ci.

4. THERMAL HYDRAULICS CALCULATION

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.

Thermal-hydraulics calculations were developed to determine the maximal temperatures achieved in the U-Ni targets during irradiation and to compare the temperature results with the design temperature limits established for the U-Ni targets. 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 U-Ni targets different coolant velocities were tested through the irradiation device (ID). For the temperature calculations of the U-Ni targets with plate geometry the thermal-hydraulics model MTCR-IEA-R1 was used and the results were obtained simultaneously with the RMB core analysis. 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 was utilized as input date to determine the temperatures in the U-Ni target with cylindrical geometry. Tables 1 and 2 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 only the temperatures of the aluminum tube.

Table 1: 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 2: 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 1 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 2 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 8 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, for the uranium amount of 20.1 g in the analyzed U-Ni targets, with plate and cylindrical geometries, a [99]Mo activity of, respectively, 3,439 Ci and 4,607 Ci was obtained at the end of 7 days irradiation time. 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 974 Ci and 1,304 Ci, respectively, for U-Ni targets with plate and cylinder geometries. 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 by the RMB under conception.

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