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

D. B. DOMINGOS, A. T. SILVA, T. G. JOÃO, R. O. R. MUNIZ

Instituto de Pesquisas Energéticas e Nucleares - Comissão Nacional de Energia Nuclear (IPEN/CNEN-SP)

Av. Professor Lineu Prestes, 2242, Cidade Universitária, 05508-000 São Paulo, SP - Brazil

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 detailed design phase. Low enriched uranium (<20% ^{235}U) targets (UAl_x dispersed in Al and metallic U foils with different geometries) are being considered for the production of Molybdenum-99 (^{99}Mo) by fission in Brazil. Neutronic and thermal-hydraulics calculations were performed to determine the production of ^{99}Mo for the UAlx-Al targets irradiated in the IEA-R1 reactor core and for three different types of targets (UAlx-Al, U-Ni cylindrical and U-Ni plate) irradiated in a reactor conception with the same power of the RMB. The neutronic analyses showed that the total activity obtained for ^{99}Mo for 10 UALx-Al miniplates with a mass of 20,1 g of ^{235}U irradiated in the IEA-R1 reactor core was 1406.63 Ci. Considering that the time needed for the chemical processing and recovering of the ^{99}Mo will be seven days after the irradiation, the total ^{99}Mo activity available for distribution will be 240.48 Ci. No thermal-hydraulics design limit was overtaken. The same calculations were performed for three targets (UAl_x-Al, U-Ni cylindrical and U-Ni plate) irradiated in a reactor conception of 30 MW with a ^{235}U mass of 20.1 g. The ^{99}Mo activities produced were, respectively, 2,980.62 Ci, 3,166.6 Ci and 3495.23 Ci for the three targets. At the end of 7 days of irradiation, the total activity obtained for the targets were, respectively, 509.57 Ci, 541.36 Ci and 597.5 Ci. The thermal hydraulics analyses show that a minimum coolant speed of 7 m/s for the UAxl-Al target, 8 m/s for the U-Ni cylindrical target and 9 m/s for the U-Ni plate target will be necessary through the irradiation device to cool the targets and not exceeding the thermal-hydraulics design limits.

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 this 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) research and development of ^{99}Mo production from fission of LEU targets, 2) discussion and decision on the best production route technique, and 3) 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 and 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 IEA-R1 reactor and at a reactor conception with the same power of the RMB [3], designate in this paper as RC. 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 foil targets used in the neutronic and thermal-hydraulics analysis

The UAl_x-Al targets of LEU type proposed and analyzed in this work are aluminum coated miniplates (Fig 1). Each miniplate measures 4.7 cm x 17 cm, 0.152 cm thick, corresponding to a total volume of 12.2 cm^3. The UAl_x-Al meat is 4.0 cm x 11.8 cm, 0.076 cm thick, leading to a total volume of 3.59 cm^3. Considering this volume and a ^{235}U mass in the target equals to 2.01 g, the ^{235}U density ($\rho_{U\text{-}235}$) 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_x 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 and in the reflector part of the RC (Fig 2), 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 MID. Fig 3 shows the MID cross section. As seen from Fig 3, up to ten UAl_x-Al targets can be placed in the box with indented bars inside of the MID.

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 [4]. 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 Fig 4):

1. One foil of uranium (LEU) of 46.05 cm x 87.7 mm x 135 μm;
2. Coating nickel foil of 20 μm thickness;
3. Two aluminum cylinder having 46.05 cm length, outside diameters of 27.88 and 30.00 mm, and inside diameters of 26.44 and 28.22 mm, respectively;
4. ^{235}U mass of 20.1 g, with 19.9% enrichment of ^{235}U.

Fig 5 shows the set of concentric cylinders (Fig 6) positioned in a device with the same dimensions of the MID.

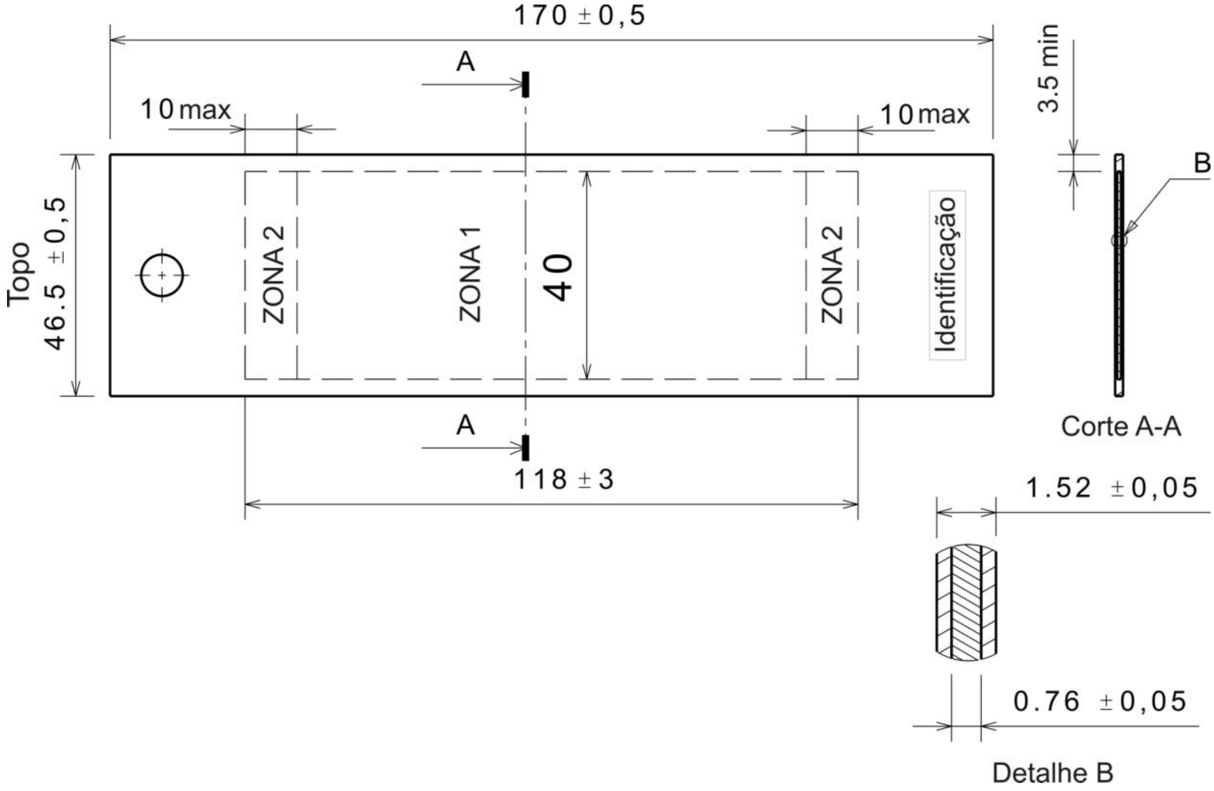


Fig 1: UAlx-Al miniplate dimensions.

Fig 2: Miniplate irradiation device – MID.

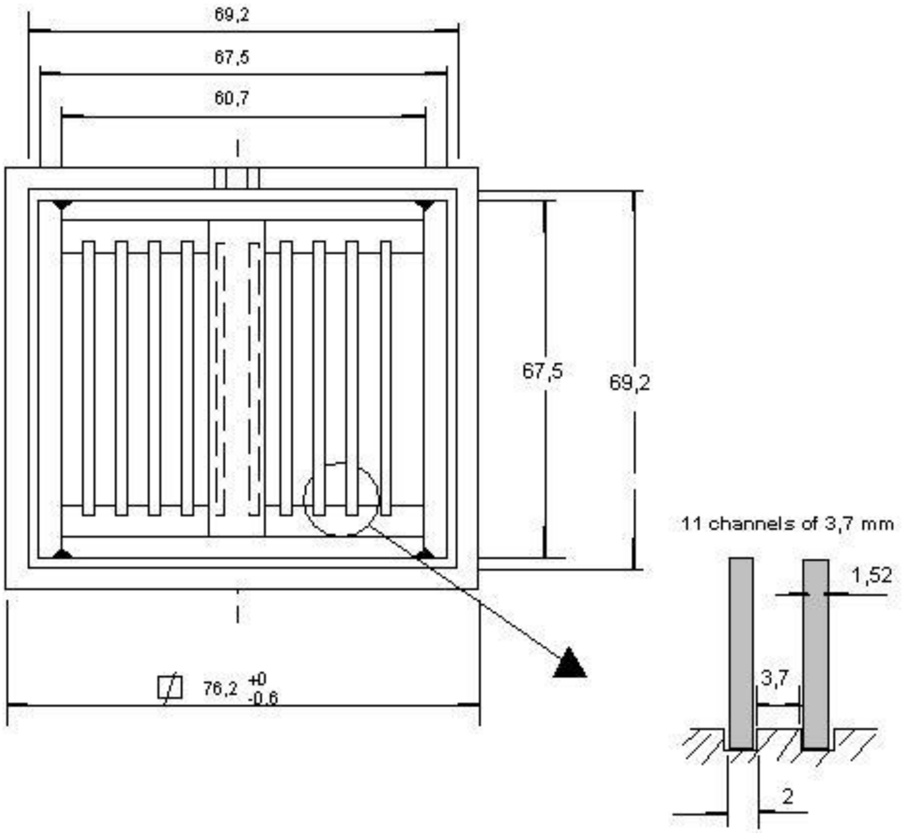
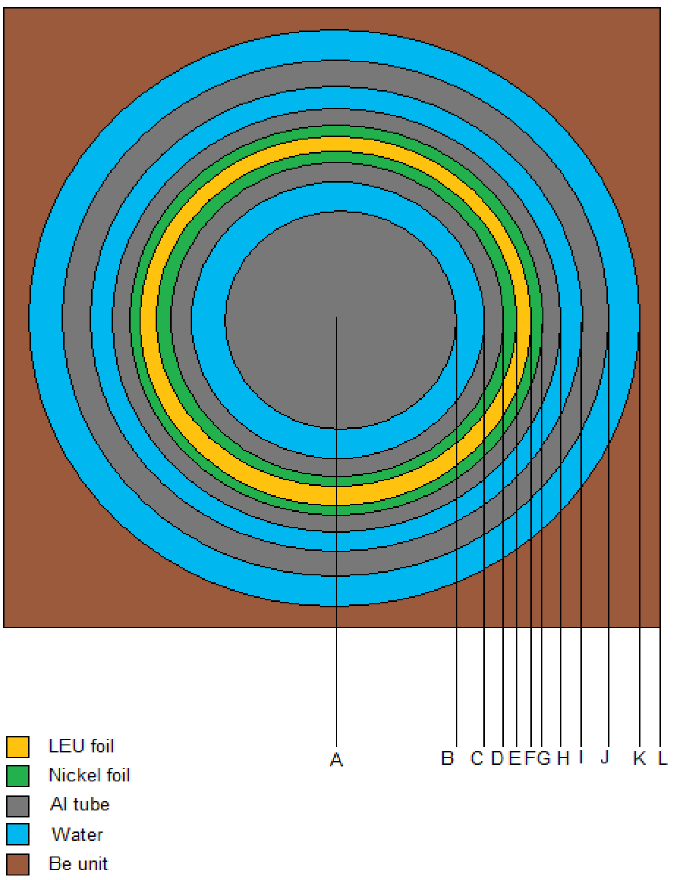


Fig 3: Cross section of the MID (dimensions in mm).



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

Fig 4: Irradiation device horizontal cross section for the U-Ni target with cylinder geometry.

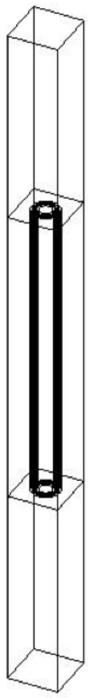


Fig 5: Set of concentric cylinders positioned in the MID.

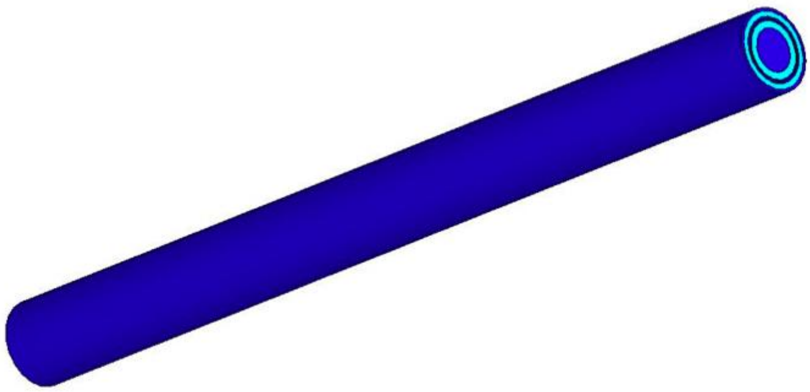


Fig 6: Set of concentric cylinders of U-Ni foil target.

The targets of metallic uranium foils with plate geometry were based on targets that were examined in the Paskitan research reactor [5] and consists of a uranium foil (19.99% ^{235}U) with a thickness of 135 μm enveloped in 20 μm thick nickel foil and placed between two aluminum plates that are welded from all sides. Each U-Ni plate has a uranium density of 2.01 g. The geometry of the foil plate target is shown in Figures 7 and 8.

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.

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

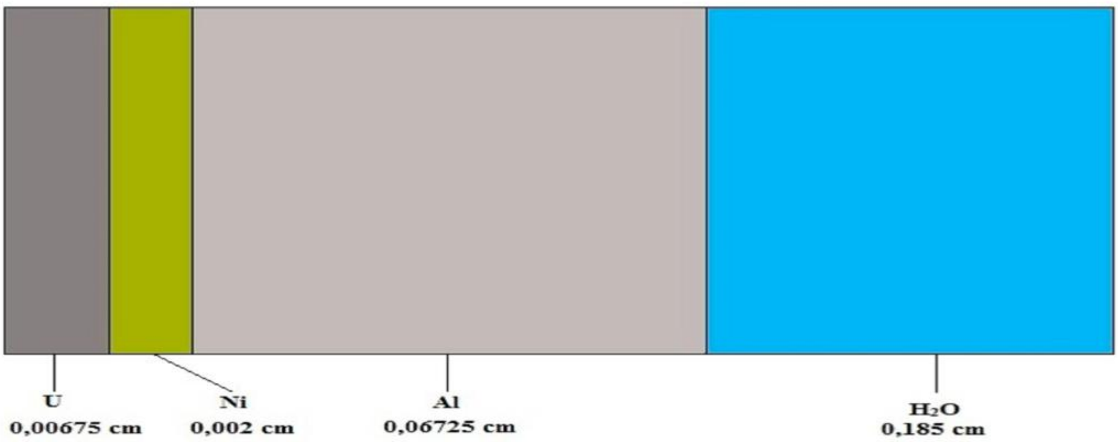


Fig 7: Half the thickness of U-Ni LEU target with plate geometry (67.5 μm), nickel foil, aluminum plate and cooling channel.

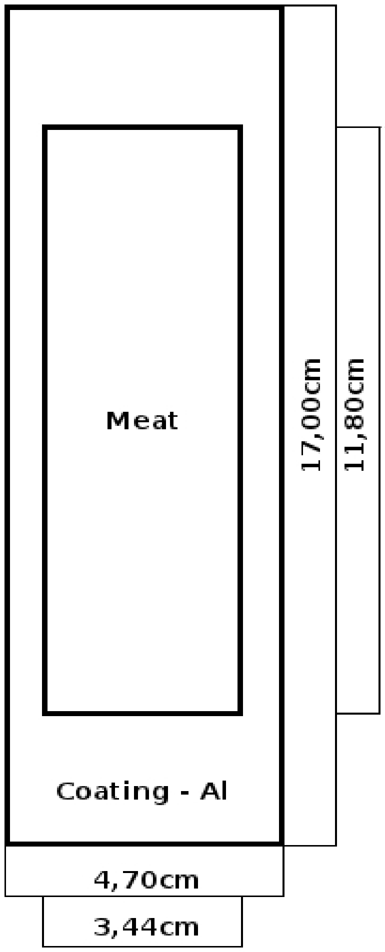


Fig 8: Width and height of the U-Ni plates.

3. Neutronic calculation for UALx-Al and U-Ni targets

The cores of the IEA-R1 and RC reactors as well as the UAlx-Al and the U-Ni targets used for the ^{99}Mo production were modeled with the HAMMER-TECHNION [6] and CITATION [7] numerical codes.

To simulate the targets in the IEA-R1 reactor, it was created a fictitious core, reflected with Beryllium, composed of 24 fuel elements of U_3Si_2-Al, 4 control elements, with density of 1.2 gU/cm^3. All fuel and control elements were taken as new and the adopted power operation was 5 MW. The cross sections of all elements were generated with HAMMER-TECHNION. The code CITATION was used to create the 3D model of the core and to determine parameters such as K-effective, neutron flux and power density. The SCALE 6.0 code system [8] 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 UAl_x-Al targets were modeled and simulated in the IEA-R1 core central position. The target irradiation time was three (3) days. At the end of irradiation, the total activity obtained for the 10 UAl_x-Al was 1,406.63 Ci. Considering that the time needed for the chemical processing and recovering of the ^{99}Mo will be seven days after the irradiation, the total ^{99}Mo activity available for distribution will be 240.48 Ci [9].

The RC conceptual design used was an open pool type, 30 MW thermal power reactor. The RC 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 as reflector. 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 was 1.9 gU/cm^3. The total activity obtained for the 10 UAl_x-Al minplates and for the U-Ni cylindrical and plate type targets were, respectively, 2,980.62 Ci, 3,166.6 Ci and 3,495.23 Ci. Considering that the time needed for the chemical processing and recovering of the ^{99}Mo will be seven (7) days after the irradiation, the total activity obtained for the 10 UAl_x-Al miniplates and for the U-Ni cylindrical and plate types targets were, respectively, 509.57 Ci, 541.36 Ci and 597.5 Ci.

4. Thermal Hydraulics Calculation for the Irradiation Device

A thermal-hydraulics model MTCR-IEA-R1 [10] 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 [11].

In order to evaluate the temperatures achieved in the targets different coolant velocities were tested through the MID. For the temperature calculations of the UAlx-Al targets the thermal-hydraulics model MTCR-IEA-R1 was used and the results were obtained for the analysis of the IEA-R1 and RC cores. 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 [12]. The power density (25 KW/cm^3) calculated in the ID position in the RC reflector with the code MTCR-IEAR1 was utilized as input date to determine the temperatures in the U-Ni target with cylindrical geometry.

The placement of the MID in the core central position of IEA-R1 reactor will deviate part of the reactor flow rate to cool the UAlx-Al targets. The flow rate in the core of the IEA-R1 reactor is 3,400 gpm which provides a flow rate of approximately 23 m^3/h per fuel element, and sufficient to cool a standard fuel element. The insertion of the MID in the IEA-R1 reactor core will divert part of the reactor core coolant to cool the UAlx-Al miniplates. Thus, a MID thermo-hydraulic analysis was developed to determine the required coolant velocity to cool the miniplates, but without damaging the fuel elements in the reactor core. Coolant velocities from 5 to 15 m/s were tested through the MID. Table 1 provide the calculated UAlx-Al target temperatures for different coolant velocities through the MID in the IEA-R1 reactor core. The simulations considered the MID with ten identical UAlx-Al miniplates. Table 1 show that coolant velocities equal or higher than 5 m/s through the MID are sufficient to cool the targets without achieving ONB temperatures. The calculated cladding temperatures are below the value of 128.5 °C, indicating one-phase flow through the targets. As calculated in the reference 13, even coolant velocities of 1.78 m/s will be sufficient to cool the targets and a coolant flow restrictor (see Fig 1) was fabricated in order to maintain a MID flow rate of 12 m^3/hr in the reactor core during target irradiation.

Tab 1: Target temperatures versus DIM coolant velocities in the IEA-R1 reactor.

Coolant velocity (m/s)	UAlx-Al meat central temperature (°C)	UALx-Al aluminum cladding temperature (°C)	ONB Temperature (T_{ONB}) (°C)	Coolant Temperature (°C)
5	111.2	99.06	128.5	45.00
6	103.2	91.07	128.5	44.48
7	97.38	85,21	128.5	44.11
8	92.89	80.71	128.5	43.84
9	89.32	77.14	128.5	43.63
10	86.42	74.24	128.5	43.46
11	85.15	72.98	128.5	43.39
12	82.93	70.75	128.5	43.27
13	81.04	68.86	128.5	43.16
14	79.40	67.23	128.5	43.08
15	77.98	65.80	128.5	43.00

Table 2 provides the calculated UAlx-Al target temperature results for different coolant velocities through the MID placed in the peripheral RC core position in the heavy water reflector. The simulations considered the MID with ten identical UAlx-Al miniplates. Table 2 shows that a velocity of 7 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 134.7 °C, indicating one-phase flow through the targets.

Tab 2: UAlx-Al target temperatures versus different MID coolant velocities in the peripheral core position of the RC.

Coolant velocity (m/s)	UAlx-Al meat central temperature (°C)	Aluminum cladding temperature (°C)	T_{onb} (°C)	Coolant temperature (°C)
5	189.0	162.6	134.7	48.51
6	172.5	146.1	134.7	47.38
7	160.3	134.0	134.7	46.58
8	151.0	124.6	134.7	45.99
9	143.5	117.1	134.7	45.53
10	137.4	111.0	134.7	45.17
11	132.3	105.9	134.7	44.87
12	130.0	103.6	134.7	44.75
13	126.0	99.6	134.7	44.52
14	122.5	96.2	134.7	44.33
15	119.5	93.1	134.7	44.17

Tables 3 and 4 provide the calculated U-Ni target temperatures for different coolant velocities through the ID in the RC core peripheral position, respectively, for plate and cylindrical geometries. Tab 4 presents for the U-Ni target with cylindrical geometry the temperature of the aluminum tube.

Tab 3: Calculated temperatures for the U-Ni target with plate geometry versus different coolant velocities through the ID.

Coolant velocity (m/s)	Aluminum cladding temperature (°C)	T_{onb} (°C)
5	191.4	132
6	171.1	132
7	156.1	132
8	144.5	132
9	135.2	132
10	127.7	132
11	121.4	132
12	118.8	132
13	113.6	132
14	109.3	132
15	105.6	132

Tab 4: Aluminum tube temperatures for the U-Ni target with cylindrical geometry versus different coolant velocities through the ID.

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

Tab 3 provides the calculated target temperature results for different coolant velocities through the MID 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 4 provides the calculated U-Ni aluminum tube temperatures for different coolant velocities through the MID 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 137°C, indicating one-phase flow through the U-Ni target with cylinder geometry.

5. Conclusion

From the neutronic calculations presented here, for a uranium amount of 20.1 g in the analyzed targets a ^{99}Mo activity of 1406.63 Ci was obtained for 7 days irradiation time in the IEA-R1 core. For the UAlx-Al target and for the U-Ni targets with plate and cylindrical geometries the calculated total ^{99}Mo activity was, respectively, 2,980.62 Ci, 3,166.6 Ci and 3.495.23 Ci. Initially, ^{99m}TC generators will be distributed seven (7) days after the end of the irradiation. Consequently, the total ^{99}Mo activity is expected to reach a value of 240.48 Ci for UAl_x-Al targets irradiated in the IEA-R1 core. For the UAlx-Al target and U-Ni targets with plate and cylinder geometries irradiated in the peripheral core position of the RC the total ^{99}Mo activity is expected to reach values of 509.57 Ci, 541.36 Ci and 597.5 Ci, respectively. From these values, it is noted that the Brazilian current demand of 450 Ci of ^{99}Mo per week may be addressed irradiating the targets in a peripheral core position of the RC.

Through the thermal-hydraulics calculations it was determined the minimum flow necessary to cool the targets. No design limit was achieved for the analyzed targets. 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.

Acknowledgments

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6. References

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