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

D. B. DOMINGOS, A. T. SILVA, T. G. JOÃO
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

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 detailed 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 a reactor conception with the same power of 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 is to perform neutronic calculations to evaluate the ^{99m}Mo production through fission in a reactor conception (RC) with the same power of the RMB [3], which is in the detailed 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 analysis

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 1):

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.

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. The geometry of the foil plate target is shown in Figures 2 and 3.

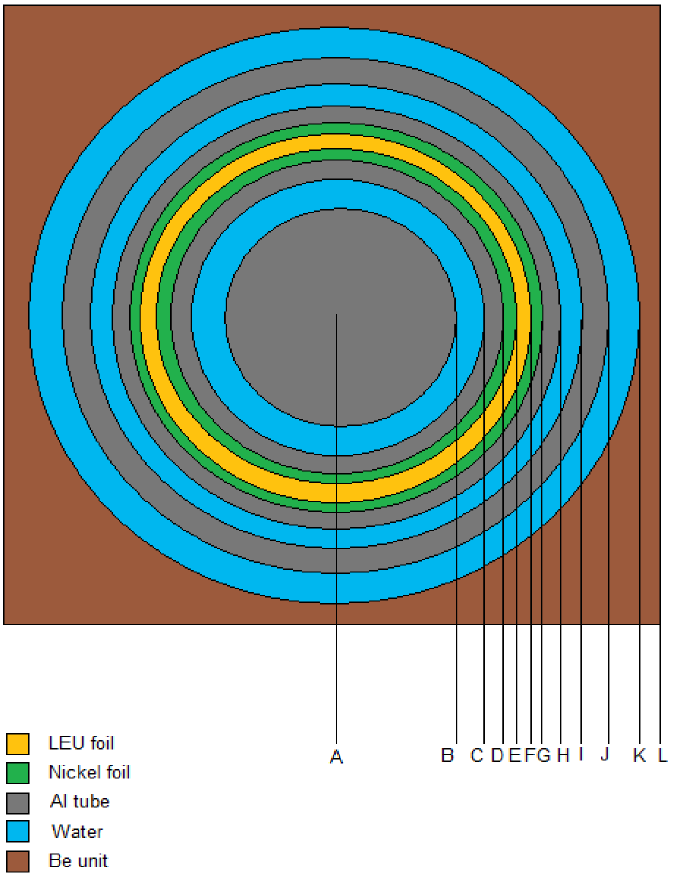
For the performed calculations, the U-Ni targets with cylindrical and plate geometries were modeled in the same irradiation device (ID), whose external dimensions are 76.2 mm x 76.2 mm x 88.74 cm (Fig 4).

For both targets a ^{235}U mass equals to 20.1 g was considered in the neutronic calculations. As seen from Fig 3, ten U-Ni targets with plate geometry were placed in the box with indented bars inside of the ID. Each U-Ni target with plate geometry has a ^{235}U mass equals to 2.01 g. The set of concentric cylinders of the metallic uranium foils with cylinder geometry was positioned in the same ID.

The targets were modeled and simulated in a peripheral core position of the RC, in the heavy water reflector. The target irradiation time was seven (7) days.

3. Neutronic calculations for the irradiation device

The RC 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 [6] and CITATION [7] 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 three-dimensional 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 code system [8] was used to perform burnup calculations for each target and also to calculate 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.



Radius	Length (cm)
AB	1.0000
AC	1.322
AD	1.3940
AE	1.3960
AF	1.4095
AG	1.4110
AH	1.5000
AI	1.7500
AJ	1.9000
AK	2.2000
AL	3.8100

Fig 1: Irradiation device horizontal cross section for the U-Ni target with cylinder geometry.

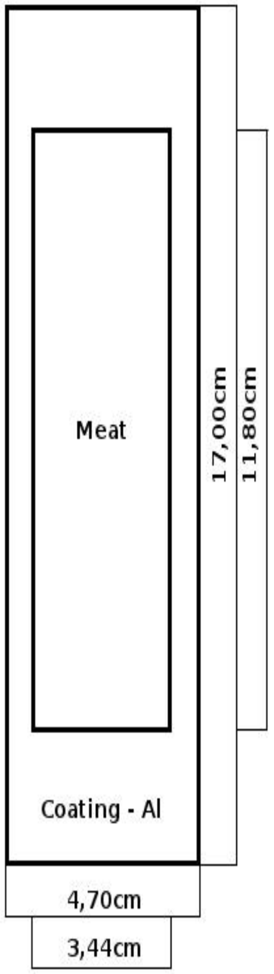


Fig 2: Width and height of the U-Ni plates.

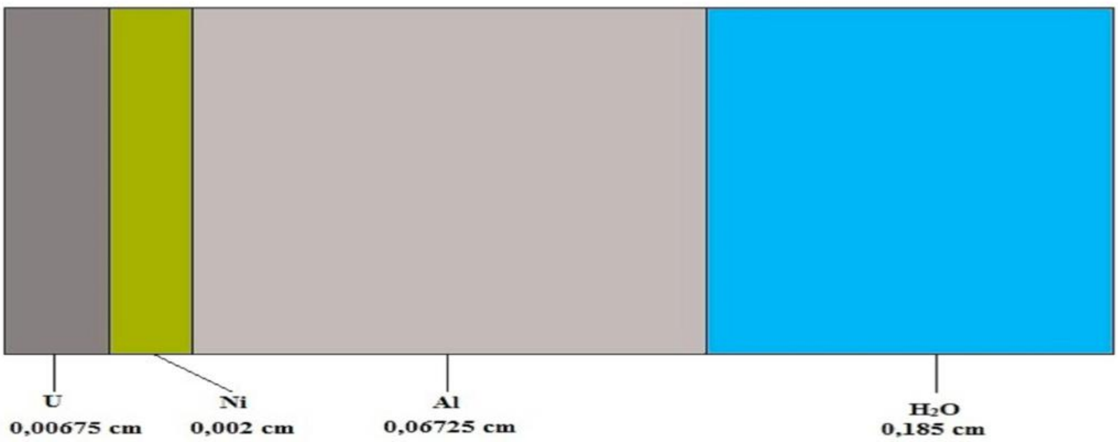


Fig 3: Half the thickness of U-Ni LEU target with plate geometry (67.5 µm), nickel foil, aluminum plate and cooling channel.

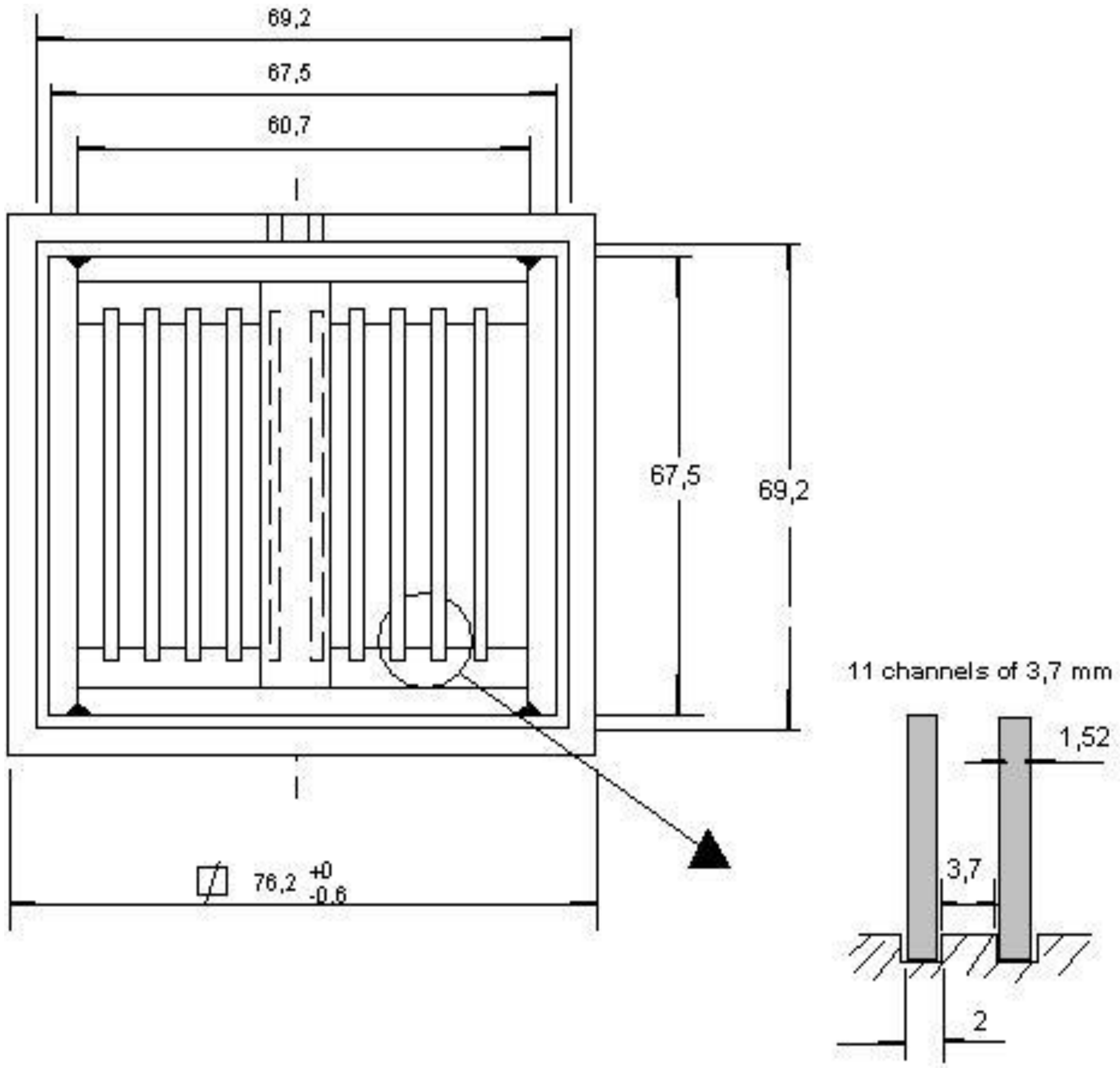


Fig 4: Irradiation device horizontal cross section for the U-Ni targets with plate geometry.

According to its conceptual design, RC is an open pool type, 30 MW thermal power reactor. The core has a 5x6 configuration with MTR-type U_3Si_2-Al fuel elements with 19.75 wt% ^{235}U enrichment. The reactor core, containing 30 U_3Si_2-Al fuel elements with a uranium density of 1.9 gU/cm^3, is light water cooled and moderated, using heavy water as reflector. The U-Ni LEU targets were modeled and simulated in a peripheral core position in the heavy water reflector. At the end of 7 days of irradiation, the total activities obtained for the U-Ni plate and cylinder geometries were, respectively, 3,495.23 Ci and 3,166.6 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 activity obtained for the U-Ni plate and cylinder geometries were, respectively, 597.5 Ci and 541.36 Ci.

4. Thermal-Hydraulics calculation

A thermal-hydraulics model MTCR-IEA-R1 [9] 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 [10].

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 RC core analysis. For the calculation of the temperatures of the U-Ni targets with cylindrical geometry was utilized the software ANSYS CFX [11]. The power density (25 KW/cm³) calculated in the ID position in the RC 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 RC peripheral position respectively for plate and cylindrical geometries.

Tab 1: 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	137
6	171.1	137
7	156.1	137
8	144.5	137
9	135.2	137
10	127.7	137
11	121.4	137
12	118.6	137
13	113.6	137
14	109.3	137
15	105.6	137

Tab 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)	T_{onb} (°C)
5	166	132
6	149	132
7	137	132
8	127	132
9	119	132
10	113	132
11	107	132
12	103	132
13	99	132
14	95	132
15	92	132

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 9 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 137°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. Conclusion

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, 3,495.23 Ci and 3,166.6 Ci was obtained at the end of 7 days irradiation time. 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 values of 597.5 Ci and 541.36, 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 may be addressed for the RC conception addressed in this paper.

Acknowledgments

The authors are grateful for financial support from CAPES and FAPESP.

6. References

[1] G. F. Wienciek, A. B. Vandegrift, A. A. Levya, and A. S. Hebden, "Status and Progress of Foil and Target Fabrication Activities for the Production of ^{99}Mo from LEU", *RERTR 2008 –*

30th International Meeting on Reduced Enrichment for Research and Test, Washington, October 5-9, 2008.

[2] I. N. Goldman, N. Ramamoorthy, and P. Adelfang, “Progress and Status of the IAEA Coordinated Research Project: Production of Mo-99 using LEU Fission or Neutronic Activation”, *RERTR 2007*, September 23-27, 2007, Prague, Czech Republic.

[3] J. A. Perrotta, A. J. Soares, "RMB: The New Brazilian Multipropose Research Reactor", *International Journal for Nuclear Power*, atw vol. 60 (2015), Issue 1, January.

[4] F. M. Bsebsu, F. Abotweirat and S. Elwaer, “Feasibility Study Part-I Thermal Hydraulic Analysis of LEU Target for ^{99}Mo Production in Tajoura Reactor, *RERTR 2007, September 23-27, 2007, Prague, Czech Republic*.

[5] A. Mushtaq, Massod Iqbal, Ishtiaq Hussain Bokhari and Tayyab Mahmood, “Low Enriched Uranium Foil Plate Target for the Production of Fission Molybdenum-99 in Pakistan Research Reactor-I, *Nuclear Instruments and Methods in Physics Research B* 267 (2009) 1109-1114.

[6] J. Barhein, W. Rhotenstein, and E. Taviv, “The HAMMER Code System TECHNION”, *Israel Institute of Technology, Haifa, Israel, NP-565,* 1978.

[7] T. B. Fowler, D. R. Vondy, and G. W. Cunninghan, “Nuclear Reactor Core Analysis Code: CITATION”, *Oak Ridge National Laboratory, ORNL-TM-2496, Rev. 2, Suppl. 3*, July 1972.

[8] SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation, *ORNL/TM-2005/39*, *Version 6.1, Vols. I–III*, November 2006. It is available as CCC-732 from Radiation Safety Information Computational Center at Oak Ridge National Laboratory.

[9] P. E. Umbehaun, "Metodologia para Análise Termo-hidráulica de Reatores de Pesquisa Tipo Piscina com Combustível Tipo Placa". *2000. Dissertação (Mestrado)* – Instituto de Pesquisas Energéticas e Nucleares, São Paulo.

[10] A. E. BERGLES, W. M. ROSENOW, “The Determination of Forced-Convection Surface Boiling Heat Transfers. *Trans, of the ASME 86 (Series C-J. of Heat Transfer), 365-375, Aug.* 1964.

[11] ANSYS CFX Reference Guide, release 12.0, April 2009.