NEUTRONIC AND THERMAL-HYDRAULICS CALCULATIONS FOR THE PRODUCTION OF MOLYBDENUM-99 BY FISSION IN LOW ENRICHED URANIUM UALx-AL TARGETS

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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. 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 evaluate the production of ^{99}Mo for these targets in the IEA-R1 reactor and to determine the temperatures achieved in the UAlx-Al targets during irradiation. For the neutronic calculations were utilized the computer codes HAMMER-TECHNION and CITATION, and for the thermal-hydraulics calculations was utilized the computer code MTRCR-IEAR1. The analysis demonstrated that the irradiation will occur without adverse consequences to the operation of the reactor. The total amount of ^{99}Mo produced was calculated with the program SCALE. Considering that the time needed for the chemical processing and recovering of the ^{99}Mo will be five days after the irradiation, the total ^{99}Mo activity available for distribution will be 176 Ci for 3 days of irradiation, 236 Ci for 5 days of irradiation, and 272 Ci for 7 days of targets irradiation.

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. 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 analysis

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 2.06 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 = 2.89 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. 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. The target irradiation time was defined according to their current and planned operating cycle.

Fig 1: Miniplate irradiation device – MID.

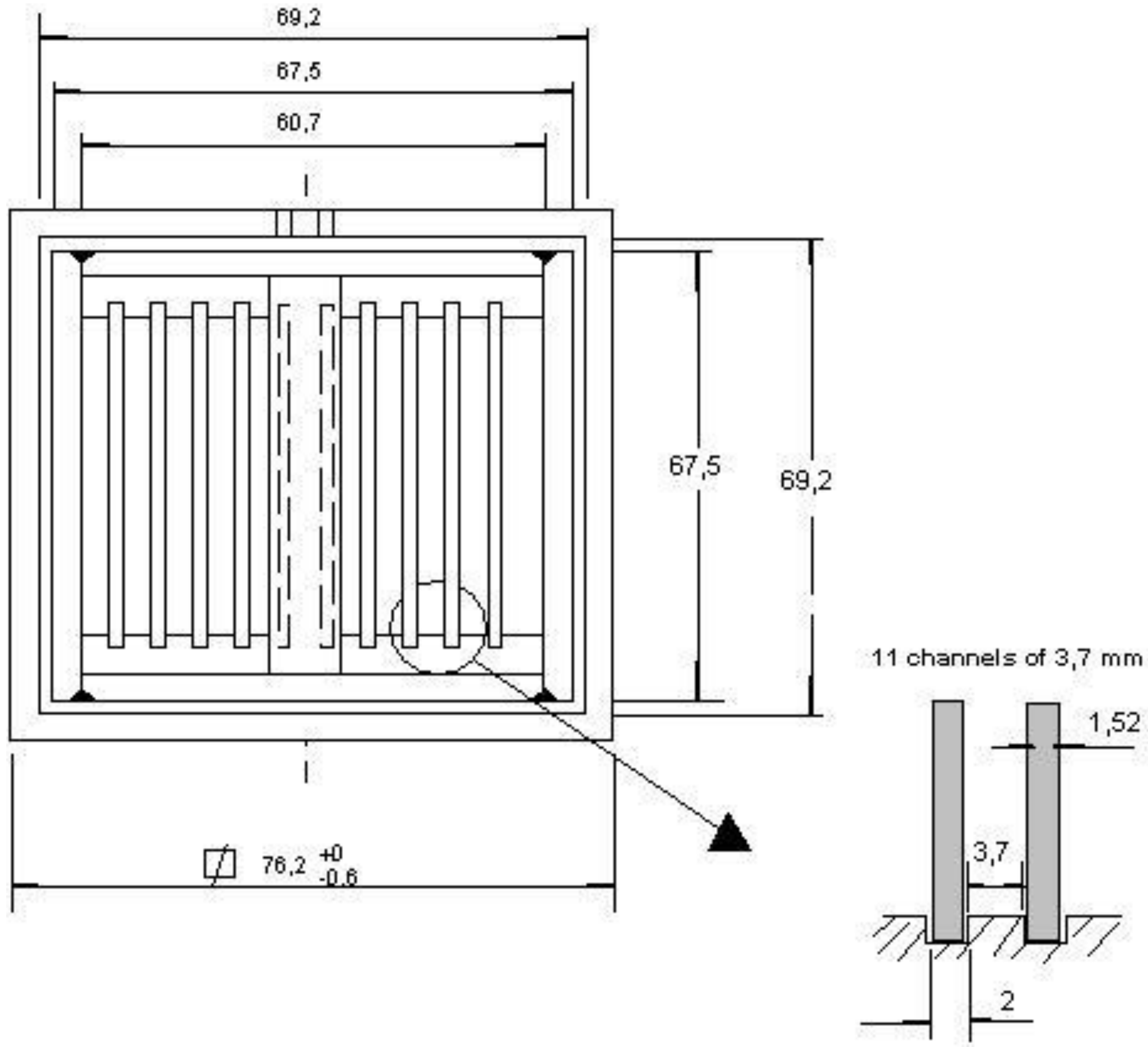

Fig 2: Cross section of the MID (dimensions in mm).

3. Neutronic calculation for the irradiation device

The IEA-R1 reactor core, 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, 5 MW, 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. The calculations were developed for three irradiation periods: 3, 5 and 7 days. At the end of 3 irradiation days, the total ^{99}Mo activity obtained for the 10 UAl_x-Al miniplates was 620 Ci. After 5 irradiation days, the total ^{99}Mo activity obtained was 832 Ci, and after 7 irradiation days the total ^{99}Mo activity obtained was 958 Ci. Considering that the time needed for the chemical processing and recovering of the ^{99}Mo will be five days after the irradiation, the total ^{99}Mo activity available for distribution will be 176 Ci for 3 irradiation days, 236 Ci for 5 irradiation days, and 272 Ci for 7 irradiation days of the targets [6].

4. Thermal-Hydraulics calculation for the irradiation device

A thermal-hydraulics model MTCR-IEA-R1 [7] 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 FIR and MDNBR. 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, in reactor power control deviation, in the coolant flow rate 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 (T_{onb}) 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 reactor core coolant temperature 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 [8].

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 flow rate to cool the miniplates, but without damaging the fuel elements in the reactor core. Flow rates from 1 to 20 m^3/hr were tested through the MID. Table 1 provide the calculated UAlx-Al target temperatures for different flow rates through the MID in the IEA-R1 reactor core. The simulations considered the MID with ten identical UAlx-Al miniplates. Table 1 show that flow rates higher than 10 m^3/h through the MID are sufficient to cool the targets without achieving ONB temperatures. The calculated cladding temperatures are below the value of 123.1°C, indicating one-phase flow through the targets. A coolant flow restrictor was fabricated in order to maintain a MID flow rate of 12 m^3/hr in the reactor core during target irradiation (see Figure 1).

Tab 1: Target temperatures versus DIM flow rates and coolant velocities.

Flow rate (m^3/h)	Coolant velocity (m/s)	UAlx-Al meat central temperature (°C)	UALx-Al aluminum cladding temperature (°C)	ONB Temperature (T_{ONB}) (°C)	Coolant Temperature (°C)
1	0.18	478.4	470.4	123.1	92.3
2	0.36	301.6	293.6	123.1	67.0

3	0.53	239	231	123.1	58.6
4	0.71	203.2	195.2	123.1	54.5
5	0.89	179.8	171.8	123.1	52.0
6	1.07	163.1	155.1	123.1	50.3
7	1.24	150.5	142.5	123.1	49.1
8	1.42	140.7	132.7	123.1	48.2
9	1.60	132.8	124.8	123.1	47.6
10	1.78	126.3	118.3	123.1	47.0
11	1.95	120.8	112.8	123.1	46.6
12	2.13	116.2	108.2	123.1	46.2
13	2.31	112.1	104.1	123.1	45.9
14	2.49	108.6	100.6	123.1	45.6
15	2.66	105.5	97.5	123.1	45.3
16	2.84	102.8	94.8	123.1	45.1
17	3.02	100.3	92.3	123.1	45.0
18	3.20	98.1	90.1	123.1	44.8
19	3.37	96.1	88.1	123.1	44.6
20	3.55	94.2	86.2	123.1	44.5

5. Conclusion

From the neutronic calculations presented for ten targets of UAlx-Al dispersion fuel with low enriched uranium (LEU) and density of 2.889 gU/cm^3, ^{99}Mo activities of 620 Ci, 832 Ci and 958 Ci were obtained, respectively for three (3), five(5) and seven (7) irradiation days in IEA-R1 reactor core at a reactor power of 5 MW. 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, respectively, values of 176 Ci, 236 Ci and 272 Ci for UAl_x-Al targets irradiated during three (3), five (5) and seven (7) irradiation days in the core central position of the IEA-R1 reactor. 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 will not be achieved with the proposed UALx-Al targets in the core central position of IEA-R1 reactor.

Acknowledgments

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

[4] 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.

[5] 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. The code is available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-732.

[6] P. J. B. O. Nishiyama, "Análises Neutrônica e Termo-hidráulica de um Dispositivo para Irradiação de Alvos Tipo LEU de UAlx-Al para Produção de ^{99}Mo no Reator IEA-R1". *2012. Dissertação (Mestrado)* – Instituto de Pesquisas Energéticas e Nucleares, São Paulo.

[7] 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.

[8] 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.