2021 International Nuclear Atlantic Conference – INAC 2021
Virtual meeting, Brazil, November 29 – December 2, 2021

ISBN: 978-65-594-1256-3

Neutronic Analysis of an Aqueous Homogeneous Reactor for ^{99}Mo Isotope Production Based on an Aqueous Thorium Sulfate Solution

D. Pérez[1], D. Milian[2], L. Hernández[3], D. Lorenzo[3], A. Gámez[1], C. Brayner[4], and A. Antonino[1]

[1]daniel.milian@ufpe.br, abel.rodriguez@ufpe.br, antonio.antonino@ufpe.br
Departamento de Energia Nuclear, Universidade Federal de Pernambuco (UFPE), Cidade Universitária, Avenida Professor Luiz Freire 1000, Recife, PE, Brasil.
[2]dmperez@cubaenergia.cu
Departamento de Energía Renovable y Eficiencia Energética, Cubaenergía, Calle 20 No 4111 / 18a y 47, Playa, Habana, Cuba.
[3]lhernandez@instec.cu, dmilian@instec.cu
Instituto Superior de Tecnologías y Ciencias Aplicadas (InSTEC), Universidad de La Habana, Avenida Salvador Allende y Luaces, Quinta de Los Molinos, Plaza de la Revolución, 10400, La Habana, Cuba.
[4]brayner@cnen.gov.br
Centro Regional de Ciências Nucleares (CRCN-NE/CNEN), Cidade Universitária, Avenida Professor Luiz Freire 200, Recife, PE, Brasil.

1. Introduction

Nowadays, radioisotopes are extensively employed in nuclear medicine, with widespread use in the diagnosis and treatment of diseases in areas such as oncology, hematology, cardiology, and neurology. One of those radioisotopes, Molybdenum-99 (^{99}Mo) is used in hospitals to produce the Technetium-99m (^{99m}Tc) employed in around 80% of nuclear imaging procedures. Globally, a total of 30–40 million of these procedures use ^{99m}Tc as a working substance. Produced in research reactors, ^{99}Mo has a half-life of only 65.94 hours and cannot be stockpiled, and security of supply is nowadays a key concern. Most of the world's supply, to cover an estimated demand of between 9,000 and 10,000 six-days Ci per week, currently relies on a small group of research heterogeneous reactors. Most of these research reactors are more than 50 years old and have an estimated production end date before or in 2030. Recent years have illustrated how unexpected shutdowns at any of those reactors can quickly lead to shortages. Furthermore, in some of these reactors, ^{99}Mo is produced from high-enriched uranium (HEU) targets, which are seen as a potential nuclear proliferation risk [1]–[8].

The use of Aqueous Homogeneous Reactors (AHR) for producing ^{99}Mo could be a promising technology, compared to the traditional method of irradiating targets in heterogeneous reactors, due to their expected low cost (up to US $ 30 million per unit), small critical mass (~ 10kg of Uranium), low thermal power (50-300 kWth), pressure (slightly below atmospheric pressure) and temperature (up to 90 °C), inherent safety and simplified fuel handling, processing and purification characteristics [1]. An AHR conceptual design, based on the Russian reactor ARGUS, was developed for the production of ^{99}Mo and other medical isotopes [9]-[14]. The flexibility of the AHR parameters variation, specifically the fuel selection, allows the use of various mixtures of uranium salts (uranyl sulfate, nitrate, etc.) and various levels of enrichment in ^{235}U [1]. So far, the possibility of replacing the enriched uranium with a non-uranium fuel, specifically a mixture of ^{232}Th and ^{233}U, has not been evaluated in the conceptual design. Although there seems to be no evidence against the viability of using an AHR with an aqueous solution of Thorium salts for the production of ^{99}Mo. Few articles dealing with this issue are identified in the scientific literature. Specifically concerning the production levels of ^{99}Mo when the fuel/target is a mixture of ^{232}Th and ^{233}U. The use of Thorium could be potentially advantageous considering the advantages of this fertile material over the use of Uranium. Therefore, the studies conducted in this article aim to test this hypothesis.

2. Methodology

The AHR conceptual design (Figure 1), previously studied in [9]–[14] using LEU fuel, consists of an aqueous solution located in a steel cylindrical vessel (wall thicknesses of 0.5 cm) with a hemispherical bottom. Placed inside the vessel, there are two coiled-tube heat exchangers and three channels. The central channel has an experimental purpose, while the other two channels are intended for poison rods, with sufficient reactivity worth to compensate the reactivity reserve and to be able to shut down the reactor. Side and bottom graphite reflectors surround the reactor vessel. Neutronic calculations have been carried out using the computational code MCNP6 with the ENDF/B-VII.1 library, evaluated at 70 °C (fuel solution average temperature determined in a previous study [14]). The main reactor core parameters are shown in Table I.

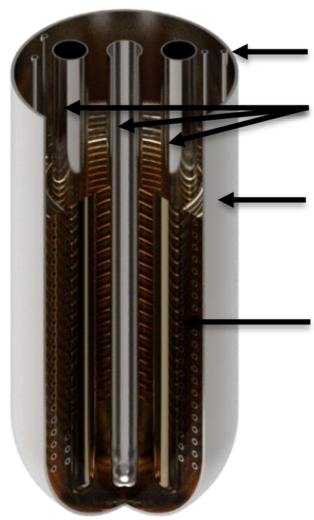

Figure 1: AHR conceptual design model.

Table I: The reactor core parameters.

Parameter	Value
Fuel solution	Thorium sulfate solution
^{233}U in the mixture (%)	14
Metal concentration (g/liter)	380
Inner core diameter (cm)	30.5
Reactor height (cm)	65.6
Reactor vessel	Stainless steel
Vessel thickness (cm)	0.5
Reflector (radial)	Graphite – 60 cm
Solution Density (g/cm^3)	1.67084
Fuel solution height (cm)	52.94*
Amount of ^{233}U in the whole reactor (kg)	1.74
Amount of ^{232}Th in the whole reactor (kg)	10.63
Cold solution volume with no voids (liter)	29.50*
Thermal Power (kWth)	50
Power density (kWth/liter of solution)	1.70
Operating temperature	less than 90 °C

* Considering the effects of the fuel thermal expansion (20 to 70 °C) and the radiolytic gas bubble formation, the fuel solution height and volume increase to 54.45 cm to 30.50 liters, respectively.

3. Results and Discussion

To quantify production levels of ^{99}Mo, a comparison was made against the isotope production in the same AHR conceptual design using Uranium fuel. Figures 2 shows how the choice of fuel to use affects isotope production. It is observed that the production of ^{99}Mo is 24.4% higher with the Uranium fuel than with the Thorium fuel. This behavior is a result of the differences between the thermal and fast fission yields. A ^{232}Th-^{233}U fuel, in comparison with a standard Uranium fuel, produces approximately 18% and 53% less ^{99}Mo due to the thermal and fast fission yield in ^{233}U and ^{232}Th, respectively (Table II) [15]. Therefore, an AHR with Thorium fuel needs to work at a power level 1.244 times higher than an AHR with Uranium fuel, to produce the same amount of ^{99}Mo. This implies that the heat removal system must be improved to take into account the increase in thermal power.

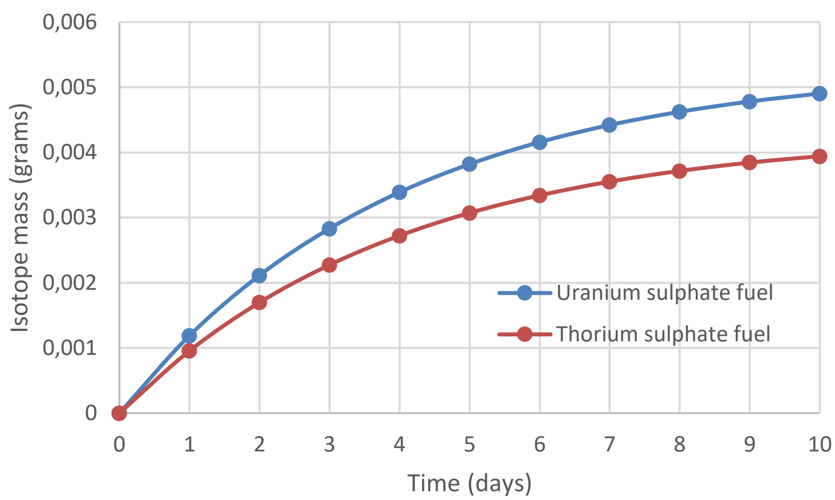

Figure 2: Accumulation of ^{99}Mo for ten days of reactor operation.

Table II. Fission yields for ^{99}Mo.

Isotope	Thermal fission yield (% per fission)	Fast fission yield (% per fission)
^{232}Th	-	2.919
^{238}U	-	6.181
^{233}U	5.03	4.85
^{235}U	6.132	5.80

4. Conclusions

The primary objective of this paper is to evaluate the viability of using an AHR with an aqueous solution of Thorium salts for the production of ^{99}Mo. An AHR conceptual design consisting of the vessel, the core channels, the coiled cooling pipe, the upper air zone, the core reflector, the structural elements, and a fuel solution of ^{232}Th and ^{233}U have been studied. The studies focused in the evaluation of ^{99}Mo production levels. It was determined that the ^{99}Mo production in the AHR conceptual design is 24.4% higher with Uranium fuel than with Thorium fuel. Therefore, an AHR with Thorium fuel needs to work at a power level 1.244 times higher than an AHR with Uranium fuel, to produce the same amount of ^{99}Mo. In conclusion, based only on the factors studied, the use of Uranium fuel seems more advantageous and safer.

Acknowledgments

This research was partially supported by the National Council for Scientific and Technological Development (CNPq), project No. 141270/2016-0, and the Coordination for the Improvement of Higher Education Personnel (CAPES), project No. 88887.518186/2020-00.

References

[1] IAEA, “Homogeneous Aqueous Solution Nuclear Reactors for the Production of Mo-99 and other Short Lived Radioisotopes,” Vienna, 2008.
[2] NEA, “A Supply and Demand Update of the Molybdenum-99 Market,” Paris, 2012.
[3] NEA, “The Supply of Medical Radioisotopes 2015 Medical Isotope Supply Review: 99 Mo/99m Tc Market Demand and Production Capacity Projection 2015-2020,” Paris, 2015.
[4] NEA, “The Supply of Medical Radioisotopes 2018 Medical Isotope Demand and Capacity Projection for the 2018-2023 Period,” Paris, 2018.
[5] NEA, “The Supply of Medical Radioisotopes. 2019 Medical Isotope Demand and Capacity Projection for the 2019-2024 Period,” 2019.
[6] OECD/NEA, “Generation IV International Forum, Technology Roadmap Update for Generation IV Nuclear Energy Systems,” Paris, 2014.
[7] NEA, “The Supply of Medical Radioisotopes Results from the Third Self-assessment of the Global Mo-99/Tc-99m Supply Chain,” Paris, 2017.
[8] NEA, “The Supply of Medical Radioisotopes: Review of Potential Molybdenum-99 / Technetium-99m Production Technologies,” Paris, 2010.
[9] D. M. Pérez et al., “Thermal-Hydraulics Study of a 75 kWth Aqueous Homogeneous Reactor for 99 Mo Production,” J. Thermodyn., vol. 2015, pp. 1–11, 2015.
[10] D. M. Pérez, D. E. M. Lorenzo, C. A. B. De Oliveira Lira, C. R. G. Hernández, M. C. Rodríguez, and L. P. R. Garcia, “Effects of some calculation parameters on the computational modelling of temperature, velocity and gas volume fraction during steady-state operation of an aqueous homogeneous reactor,” Int. J. Nucl. Energy Sci. Technol., vol. 11, no. 1, p. 1, 2017.
[11] D. M. Pérez, D. E. M. Lorenzo, C. A. B. de Oliveira Lira, and L. P. R. Garcia, “Neutronic evaluation of the steady-state operation of a 20 kWth Aqueous Homogeneous Reactor for Mo-99 production,” Ann. Nucl. Energy, vol. 128, 2019.
[12] D. M. Pérez, L. H. Pardo, D. M. Pérez, A. G. Rodríguez, D. E. M. Lorenzo, and C. A. B. de O. Lira, “New advances in the computational simulation of Aqueous Homogeneous Reactor for medical isotopes production,” BRAZILIAN J. Radiat. Sci., vol. 8, no. 3A, p. 18, 2020.
[13] D. M. Pérez et al., “Study of radiolytic gas bubbles formation and behaviour in an aqueous uranyl sulphate solution using ImageJ,” Int. J. Nucl. Energy Sci. Technol., vol. 14, no. 3, pp. 235–251, 2020.
[14] D. M. Pérez, D. M. Pérez, L. H. Pardo, D. E. M. Lorenzo, and C. A. B. de Oliveira Lira, “Multi-physics evaluation of the steady-state operation of an Aqueous Homogeneous Reactor for producing Mo-99 for the Brazilian demand,” Int. J. Thermodyn., vol. 24, no. 1, pp. 9–22, Feb. 2021.
[15] A. L. Nichols, D. L. Aldama, and M. Verpelli, Handbook of Nuclear data for safeguards database extensions, no. August. 2008.