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Neutronic Evaluation of Using a Thorium Sulfate Solution in an Aqueous Homogeneous Reactor

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ABSTRACT

Radioisotope ^{99}Mo is one of the most essential radioisotopes in nuclear medicine. Its production in an Aqueous Homogeneous Reactor (AHR) could be potentially advantageous compared to the traditional technology, based on target irradiation in a heterogeneous reactor. An AHR conceptual design using low-enriched uranium for the production of ^{99}Mo has been studied in depth. So far, the possibility of replacing uranium with a non-uranium fuel, specifically a mixture of ^{232}Th and ^{233}U, has not been evaluated in the conceptual design. Therefore, the studies conducted in this article aim to evaluate the neutronic behavior of the AHR conceptual design using thorium sulfate solution. Here, the ^{232}Th-^{233}U composition to guarantee ten years of operation without refueling, conversion ratio, medical isotopes production levels, and reactor kinetic parameters were evaluated, using the computational code MCNP6. It was obtained that 14 % ^{233}U enrichment guarantees the reactor operation for ten years without refueling. The conversion ratio was calculated at 0.14. The calculated ^{99}Mo production in the AHR conceptual design resulted in 24.4 % higher with uranium fuel than with thorium fuel.

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 generate 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 [1-3].

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 (~ 10 kg of uranium), low thermal power (50-300 kWth), low operating pressure (slightly below atmospheric pressure) and temperature (up to 90 °C), inherent safety, and simplified fuel handling, processing and

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purification characteristics [1,4,5]. An AHR conceptual design, based on the Russian reactor ARGUS, was developed for the production of ^{99}Mo and other medical isotopes [6-14]. Presently, the ARGUS reactor stands out as the only large-scale successful experiment on the use of an AHR in steady-state operation. This reactor, a 20 kWth, HEU fuel solution reactor is in operation at the "Kurchatov Institute" since 1981 with great economic and safety aspects. In July 2014, after feasibility calculations and a conversion period, the ARGUS reactor reached the first criticality with low-enriched uranium (LEU) fuel. This conversion to LEU fuel meets the purposes of the Reduced Enrichment for Research and Test Reactor (RERTR) program, which aims to replace HEU into LEU or with non-uranium targets/fuels [15].

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]. 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, 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. Few articles dealing with this issue are identified in the scientific literature, specifically concerning the production levels of ^{99}Mo and other radioisotopes when the fuel/target is a mixture of ^{232}Th and ^{233}U. The use of thorium could be potentially advantageous considering the benefits of this fertile material over the use of uranium. Therefore, the primary objective of this paper is to evaluate the neutronic viability of using an AHR with an aqueous solution of thorium salts for the production of ^{99}Mo. For this, different ^{232}Th-^{233}U mixtures will be evaluated to guarantee 10 years of normal operation without refueling. In addition, the reactor conversion ratio (CR) will be calculated as well as the production levels of ^{99}Mo, ^{89}Sr, ^{131}I, and ^{133}Xe. In the same way, the effective delayed neutron fraction (β_{eff}) and mean neutron generation time (Λ) will be calculated. All computational calculations will be carried out using the computational code MCNP6.

METHODOLOGY

The AHR conceptual design (Fig. 1) previously studied in [6-11,14] 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 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. The steel channels are 4.8 cm in outer diameter, and their wall thickness is 0.2 cm. The two coiled-tube heat exchangers use 34.5 m of tubing, 0.60 cm inner diameter, and 1.0 cm outer diameter. Side and bottom graphite reflectors surround the reactor vessel. The main reactor core parameters are shown in Table 1.

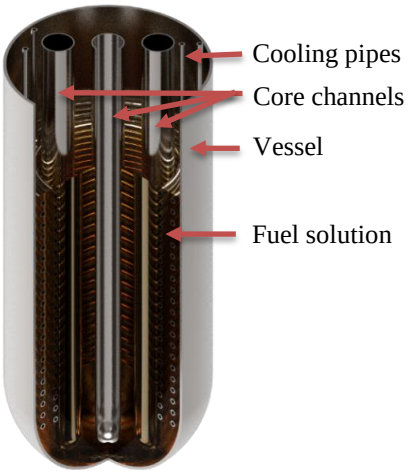

Fig. 1. AHR conceptual design model.

Table 1. The reactor core parameters.

Parameter	Value
Fuel solution	Thorium sulfate solution
^{233}U in the mixture (%)	14
Heavy 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*
Total ^{233}U mass (kg)	1.74
Total ^{232}Th mass (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.

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 [11]). MCNP is a general-purpose, continuous-energy, generalized-geometry, time-dependent, Monte Carlo radiation-transport code designed to track many particle types over broad

ranges of energies. The MCNP version used in this work offers a group of new capabilities beyond its predecessors used in this work such as the depletion/burnup and the FMESH card, which enables to determine the energy deposition and neutrons flux profiles [16]. Figure 2 shows the geometrical model of the AHR conceptual design on the MCNP Visual Editor. As the helical pipes cannot exactly be modeled in MCNP, they were represented by equally spaced toroids.

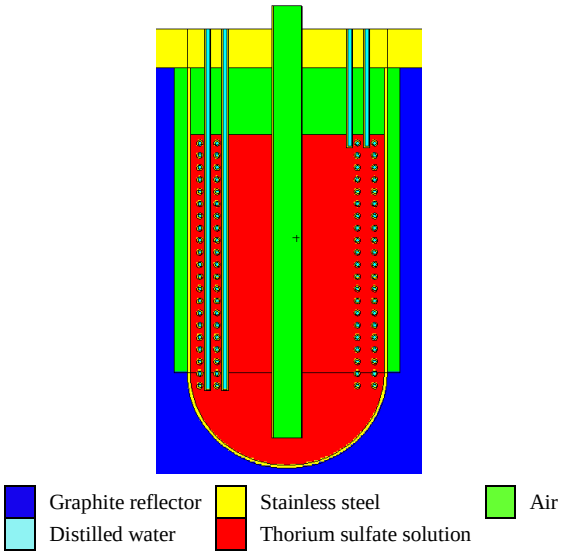

Fig. 2. Longitudinal section of the AHR geometrical model in the MCNP Visual Editor.

Four studies were conducted during the investigation:

First, the determination of the k_{eff} dependence on various enrichments in ^{233}U. The objective of this study was to determine the ^{233}U fraction in the initial mixture (^{233}U + ^{232}Th) which allows the reactor to operate without refueling for at least ten years. The five enrichments in ^{233}U that were evaluated were 12, 14, 16, 18, and 20 %. The calculations were carried out without control rods during ten full-power years.

The second was the determination of the reactor conversion ratio (CR). This parameter, as defined in Eq. (1), measures the ability to convert fertile isotopes into fissile isotopes. In the fuel solution, the fissile atoms (^{233}U) are produced by neutron capture in fertile atoms (^{232}Th) as indicated in Eq. (2), while the consumption includes fission and capture.

$$CR = \frac{Fertile\ atoms\ converted}{Fissile\ atoms\ consumed} = \frac{Fissile\ atoms\ produced}{Fissile\ atoms\ consumed} \quad (1)$$

$$^{232}_{90}Th + n \rightarrow {}^{233}_{90}Th \rightarrow \frac{\beta}{23.2\ minutes} \rightarrow {}^{233}_{91}Pa \rightarrow \frac{\beta}{27.4\ days} \rightarrow {}^{233}_{92}U \quad (2)$$

Third, the determination of the production levels of medical isotopes. This study was carried out considering fresh fuel and an operating time of ten days.

Four, the determination of the most important kinetic parameters in a nuclear reactor, which are the effective delayed neutron fraction (β_{eff}) and the mean neutron generation time (Λ). The β_{eff} for the AHR conceptual design was calculated with MCNP code using the prompt method [8]. The procedure of the prompt method includes the calculation of the multiplication factor with delayed neutron (k_{eff}) and without delayed neutron (k_p), then approximating the β_{eff} according to in Eq. (3).

$$\beta_{eff} = 1 - \frac{K_p}{K_{eff}} \quad (3)$$

The mean neutron generation time was determined using the pulsed neutron technique. In this method, a burst of neutrons is injected into a slightly subcritical system and the decay of the neutron population is observed as a function of time. Figure 3 shows the location of the two detectors used to evaluate the decay of the neutron population as a function of time. A neutron source with a fission spectrum was placed at the center of the reactor core, inside the central experimental channel. A detailed description of the pulsed neutron technique for the mean neutron generation time calculation with MCNP in an AHR can be found in [8].

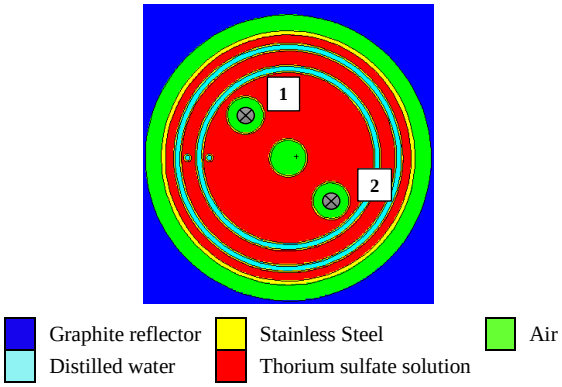

Fig. 3. Location of the two detectors used to evaluate the decay of the neutron population.

To conduct these studies, two types of MCNP calculations were performed, namely KCODE and NPS calculations. The KCODE criticality calculations were performed using 50 and 3,000 inactive and active cycles respectively, with 10,000 neutron histories per cycle. In the other type of calculation, the NPS history cutoff card was used to run 30,000,000 neutron histories using the SDEF card to define the neutron source. Tally F5 was used to determine the neutron flux. Both prompt and delayed neutrons were taken into account for criticality and NPS calculations using the TOTNU card. Benchmarking exercises to evaluate the

prediction capability of the developed models of AHR with MCNP code and the available libraries have been carried out in previous works [8,11].

RESULTS AND DISCUSSION

As a first step, the dependence of k_{eff} on various ^{233}U enrichments was studied. Figure 4 shows the k_{eff} evolution over ten full-power years, reaching a discharge fuel burnup of 14.76 GWd/tonU. As seen in Fig. 4, ^{233}U enrichment of 14 % is sufficient for the reactor to operate without refueling for ten years.

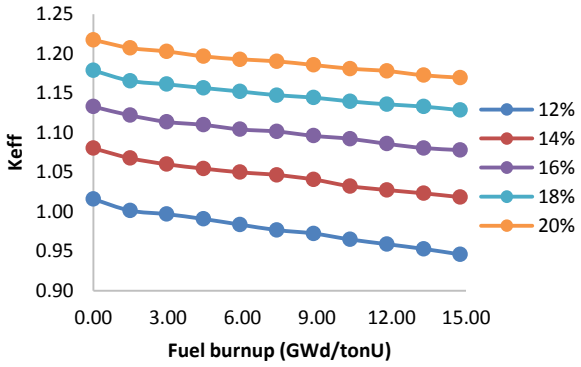

Fig. 4. K_{eff} vs. fuel burnup for five ^{233}U enrichments.

The next step was the determination of the reactor conversion ratio. For that, the masses of the fertile and fissile isotopes obtained from the burnup calculation after ten full-power years were used. A CR of 0.14 was determined, which is consistent with values reported in the literature for this type of system using thorium fuel [17,18].

The third study consisted of determining the production levels of medical isotopes using thorium fuel. For quantifying the production levels, a comparison was made against the isotope production in the same AHR using uranium fuel. Figures 5 to 8 show the comparison for the isotopes ^{99}Mo, ^{89}Sr, ^{131}I, and ^{133}Xe, respectively, and demonstrate how the choice of fuel affects the isotope production. In the case of ^{99}Mo, it is observed that its production is 24.4 % higher with uranium fuel than with 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 2) [19]. 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.

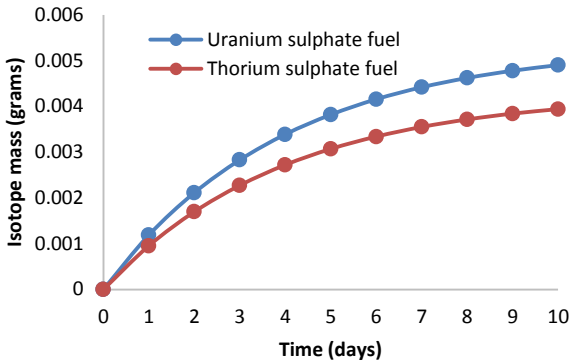

Fig. 5. Accumulation of ^{99}Mo for ten days of reactor operation.

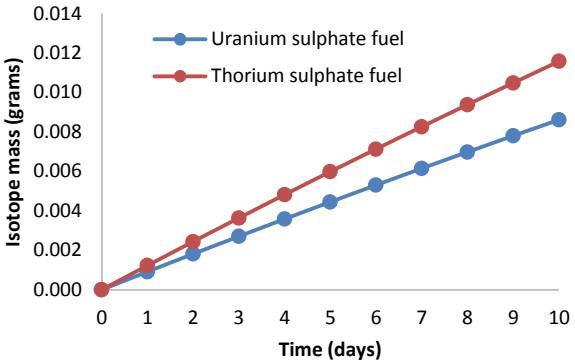

Fig. 6. Accumulation of ^{89}Sr for ten days of reactor operation.

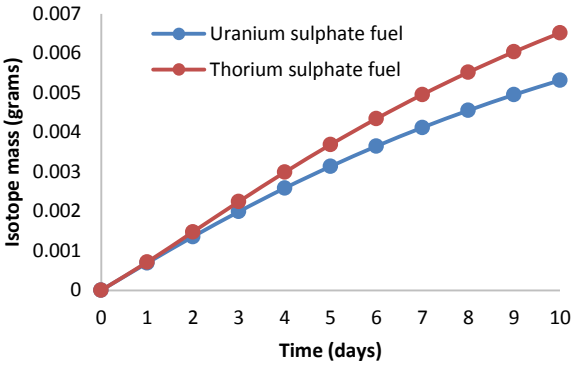

Fig. 7. Accumulation of ^{131}I for ten days of reactor operation.

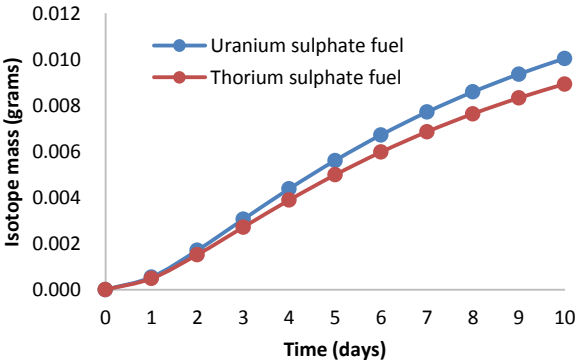

Fig. 8. Accumulation of ^{133}Xe for ten days of reactor operation.

Table 2. Thermal and fast 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

As observed in Figs. 6 and 7, the production of ^{89}Sr and ^{131}I is higher with thorium fuel than with uranium fuel. Meanwhile, in the case of ^{133}Xe, its production with uranium fuel is higher. As with the ^{99}Mo, the thermal fission yields of the ^{233}U and ^{235}U are the main responsible for these behaviors.

The final study was the determination of the most important kinetic parameters in a nuclear reactor, which are β_{eff} and Λ. Table 3 shows the k_{eff}, k_p, and the result of β_{eff} calculations. The β_{eff} with the thorium fuel represents only 45 % of the effective delayed neutron fraction determined for the same system using uranium fuel (726 pcm) [20].

Table 3. Total (k_{eff}) and prompt (k_p) multiplication factors and effective delayed neutron fraction (β_{eff}).

K_{eff}	K_p	β_{eff}	β_{eff} (pcm)
1.07934 ±0.00014	1.07579 ±0.00014	0.00329 ±0.00018	329 ±18

Figure 9 shows the averaged neutron flux in relative units, over the two detectors, after thirty million source pulses. Figure 9 also shows the exponential fitting used. The decay constant (α_0) was obtained by fitting the prompt drop with the first-order exponential decay function, $n(t) = n_0 e^{-\alpha_0 t} + N$; where n denotes the average number of neutrons tallied and N is a constant. As the parameters reactivity (ρ), α_0, and β_{eff} are known, the mean neutron generation time can be determined by solving in Eq. (4).

$$\alpha_0 = \frac{1}{n}\frac{dn}{dt} = \frac{\rho - \beta_{eff}}{\Lambda} \quad (4)$$

Table 4 shows the results of the Λ calculations (for each detector and the average value). As expected, the Λ was found to be relatively independent of the location in which the neutron flux was measured. Combining β_{eff} and Λ, a β_{eff}/Λ ratio of 40.21 s^{-1} was obtained.

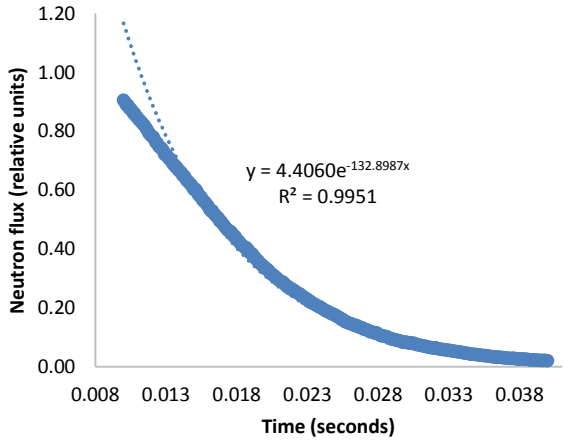

Fig. 9. Averaged neutron flux as a function of time.

Table 4. Mean neutron generation time (Λ) results.

Parameter	Value
Λ_1 (μs)	87.97
Λ_2 (μs)	88.02
Λ_{ave} (μs)	88.00
β_{eff}/Λ (s^{-1})	40.21

The calculated average effective neutron generation time (including the delayed ones - Λ_{eff}) is 0.063 seconds. This demonstrates the effect that delayed neutrons have on neutron generation time and thus on reactor control. If the AHR conceptual design were operated in a self-sustained chain reaction using only prompt neutrons (β_{eff} = 0), the generation time would be 8.8*10^{-5} seconds (Table 4). On the other hand, when operating the reactor so that a fraction of 0.00329 neutrons is delayed, the generation lifetime is extended to 0.063 seconds (more than 700 times higher). The average effective neutron generation time determined with the thorium fuel represents 66 % of the Λ_{eff} determined for the same system using uranium fuel (0.096 seconds) [20]. Therefore, from the point of view of these kinetic parameters of the conceptual design (β_{eff} and Λ_{eff}), the use of uranium fuel seems to be safer.

CONCLUSION

The primary objective of this paper was to evaluate the neutronic viability of using AHR with an aqueous solution of thorium salts for the production of ^{99}Mo. The neutronic calculations demonstrated that 14 % ^{233}U enrichment in the mixture of ^{232}Th-^{233}U is sufficient for the reactor to operate without refueling for ten years. The discharge burnup after ten years reaches 14.76 GWd/tonU. The conversion ratio calculated (0.14) is consistent with values reported by other authors for this type of system using thorium fuel. It was determined that ^{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. The effective delayed neutron fraction, the average mean neutron generation time, and the average effective neutron generation time were 329 pcm, 88.00 μs, and 0.063 seconds, respectively. In conclusion, based only on the neutronic factors studied, the use of uranium fuel in AHR seems more advantageous and safer than a mixture fuel of ^{232}Th-^{233}U. However, many other physical, economic, and safety factors need to be studied and analyzed for a more complete conclusion.

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AUTHOR CONTRIBUTION

D. Pérez conceived the presented idea, developed the theory, and wrote the manuscript. D. Milian, L. Hernández, and A. Gámez performed the computational calculations. D. Lorenzo and C. Brayner supervised the findings of this work. All authors read and approved the final version of the paper.

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