

COMPUTATIONAL ANALYSIS OF RADIO-MEDICAL ISOTOPES' PRODUCTION RATE IN A MODELED ACCELERATOR DRIVEN SUBCRITICAL REACTOR FUELED WITH $\text{UO}_2(\text{NO}_3)_2\text{-Th}(\text{NO}_3)_4$ AQUEOUS SOLUTIONS USING PROTON ENERGY OF 10 – 100 MEV

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Abstract

In the present work, MCNPX 2.6.0 Monte Carlo based computational code was used to simulate an aqueous subcritical reactor fueled with thorium-uranium nitrate solution; containing ^{20%-enriched}UO₂. The core was chosen as cylindrical shape with 36×41 cm dimension. The effective multiplication of the subcritical core was chosen less than 0.94. Fission reaction rate induced by 10-100 MeV accelerated proton particles was determined in the subcritical system respectively. Deposited power was calculated for any incident energy. Radial and axial power densities were calculated using different projectile energies. The proton currents of 10, 30, 150 and 300 μA were investigated. The obtained data showed a 20 MeV proton of current 300 μA resulted in a 1.80kW power, which is referred to fission process. The axial and radial power densities vary in range of ~1-10 W/cm³ corresponded with the incident particles of 10-100 MeV respectively. The subcritical core β and β_{eff} parameters were 738 and 670 pcm respectively. The core void reactivity coefficients were highly negative (-9.4--6.32 mk/% void). Total temperature reactivity coefficient was a little positive (~ +0.009 mk/°C). Burnup calculations showed 20 MeV protons having 300 μA current resulted in production of ~ 550 Ci/Week carrier-free ⁹⁹Mo, ~137Ci/Week of ¹³¹I and ~469Ci/Week of ⁸⁹Sr. ¹³¹I carrier-free product will be obtainable after a delay time for decay of the other radioisotopes to the stable Xe. The subcritical core shows admirable potentials for the radioisotope production with outstandingly enhanced safety operation. Clearly, higher proton energy or beam current utilization increases noticeably the production rate. The subcritical core can be used for a long time (>1 year) without refueling requirements.

Keywords: ADSR, Nitrate aqueous fuel, Proton particles, Radioisotope production

Introduction

The use of solution reactors for producing medical isotopes is potentially advantageous because of their low cost, small critical mass, inherent passive safety, and simplified fuel handling, processing and purification characteristics. Approximately 30 solution reactors have been built and operated in the 1940s and 1950s but most of them are no longer in service. However, in the 1990s a renewed interest in solution reactors for the production of medical isotopes has prompted by several countries, including China, the Russian Federation and the United States of America to initiate programs to assess the feasibility of utilizing AHR technology for medical isotope production applications on commercial basis [1]. Accelerator driven subcritical solution reactor idea is following by some countries such as Belgium and United States of America [2]. B&W is developing the Medical Isotope Production System

(MIPS); in this system, the ^{99}Mo produced in an LEU-fueled aqueous homogeneous reactor (AHR) by the fission of ^{235}U [3]. IBA Company investigates the feasibility of using charged particle accelerators to irradiate lowly-enriched Uranium targets in order to produce $^{99\text{m}}\text{Tc}/^{99}\text{Mo}$ generators. As the obtained results show a 350 MeV proton accelerator coupled to a subcritical reactor, with or without spallation target, can produce half of the ^{99}Mo worldwide demand [4]. For mini-MIPS/SHINE experiments carried out by B&W, a linear accelerator (linac) will drive a photo-neutron source that induces fissions in a uranyl nitrate solution surrounding the neutron target. These photo-nuclear reactions occur in the Giant Dipole Resonance (GDR) region, typically in the range of 12-16 MeV [3]. In sum up, Aqueous Homogeneous Reactor (AHR) offers an alternative way to produce medical isotopes as well it advantageously works at low temperature, pressure and power operations. The smaller, simplified reactor and control systems, large negative temperature coefficients of reactivity, and less waste generation per unit product makes it worthwhile for economic production of the routine radio-medical isotopes [1]. Hence, in the present study the Monte Carlo based computational code is used to model an AHR fueled with nitrate solution. Investigation of potentially advantages of the modeled subcritical aqueous reactor for production of ^{99}Mo , ^{89}Sr and ^{131}I radioisotopes was proposed in this work.

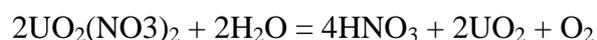
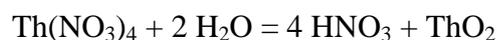
Material and methods

In this work, MCNPX 2.6.0 has been used as a powerful particle transport code with the ability to calculate steady-state reaction rates, normalization parameters, neutronic parameters, as well as fuel burn up using CINDER90 to calculate the time-dependent parameters [5-6]. A cylindrical aqueous reactor was modeled using the MCNPX 2.6.0 code. Light water flowing inside the considered coils was selected as a coolant for the fuel solution. A 3D neutronic model was set up using the MCNPX 2.6.0 code in cold zero power situations by means of ENDF/B-VI continuous-energy cross section. The cross sections of $S(\alpha, \beta)$ was used for fuel solution, heavy water and light water. A 26 cm thick heavy water reflector was used to reflect the emerging neutrons and performs a controlling role for the system shut down requirements. KCODE card was used for neutronic parameter calculations.

Table 1 Core material and dimensions modeled using MCNPX 2.6.0

Core specifications	value	unit
Fuel solution: W%: ^{235}U : 2.995, ^{238}U : 12.173, O: 71.651, H: 7.619, N: 2.522, ^{232}Th : 3.037	1.22	g/cm ³
Stainless Steel cover plate: W%: Fe: 69.5, Cr: 19, Ni: 9.5, Mn: 2.0	6.50	g/cm ³
D ₂ O reflector : W%: D: 33.33, O: 66.67	1.105	g/cm ³
Core dimension	36×41	cm

The Uranium enrichment was 19.75% (^{235}U : 19.75%, ^{238}U : 80.25%) in the nitrate salt. As the following balance equations show, the nitrate solution containing the 0.524 M salt can obtain a PH ~ 7.5.



Fuel solution pH cannot be allowed to rise above pH 3; if it does precipitation of uranium and many fission products will begin. In case of the solution PH adjustment, several drops of 13M HNO₃ can adjust the nitrate solution PH. However, if the pH is too low, the ion exchange resin, which is used for ⁹⁹Mo recovery from the aqueous solution, is the less effective [1].

The nitrate solution involves 180 g/L of enriched U and 36 g/L of ²³²Th. A lower uranium salt concentration in the fuel solution results in a larger Kd for Mo(VI) and therefore a more effective and efficient recovery of ⁹⁹Mo from such solutions [1]. Uranyl nitrate solubility in water is 660 g/L [7].

Light water coils with 3 cm diameter were used to cool the aqueous solution. The cross sectional view of the modeled core in MCNPX is depicted in Fig. 1.

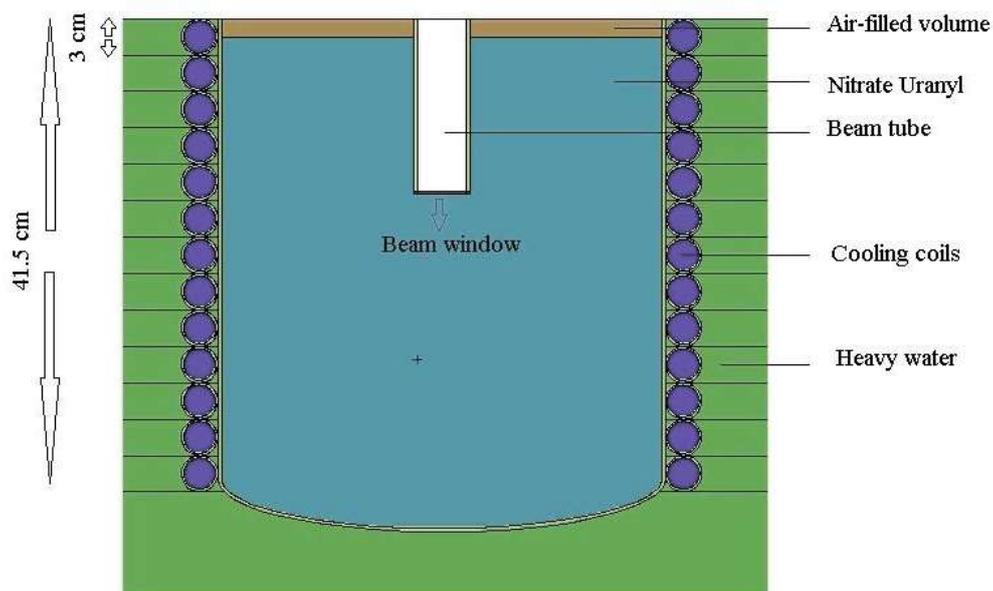


Fig 1. Cross sectional view of the modeled core

The fuel solution of uranium-thorium nitrate was considered for the modeled aqueous homogeneous reactor to investigate ⁹⁹Mo, ⁸⁹Sr and ¹³¹I production efficiency and safety factors of the nuclear reactor. Accelerated proton particle was used to induce fission in the aqueous solution. Energy range of 10 up 100 MeV was investigated. Radial and axial neutron flux distributions were calculated using the mesh tally card of the computational code. Deposited power distributions were calculated using the mesh tally card for the solution fuel loading. Reactivity coefficients of fuel, coolant, and moderator were calculated using the TMP card and temperature-related cross section library of .71c from endf70 in MCNPX. Void reactivity effect of the coolant, reflector and the solution fuel were calculated for the fuel load in the modeled core. Delayed neutron fraction and effective delayed neutron fraction were calculated separately for the core fueled the uranium-thorium nitrate solution. Burn-up calculation for the fuel solution was performed at a power of 1.8 kW for 2 weeks using the BURN card. Production rate of ⁹⁹Mo, ⁸⁹Sr and ¹³¹I radioisotopes after the burn-up time was investigated. Production rate of long half-life alpha emitter radioisotopes after the burn-up time was investigated.

Result and discussion

An large rise of the neutron flux is occurred in the aqueous solution irradiated by projectile proton particles of higher energetic in comparison the low energy beam currents. About 33% of neutrons are thermal in case of all investigated proton energies (Fig.2). Also, energy enhancement of 10 to 20 MeV increases total neutron flux with a factor of 5, the energy rising of 20 to 30 MeV increases the neutron flux with a factor of 3 (Fig.3).

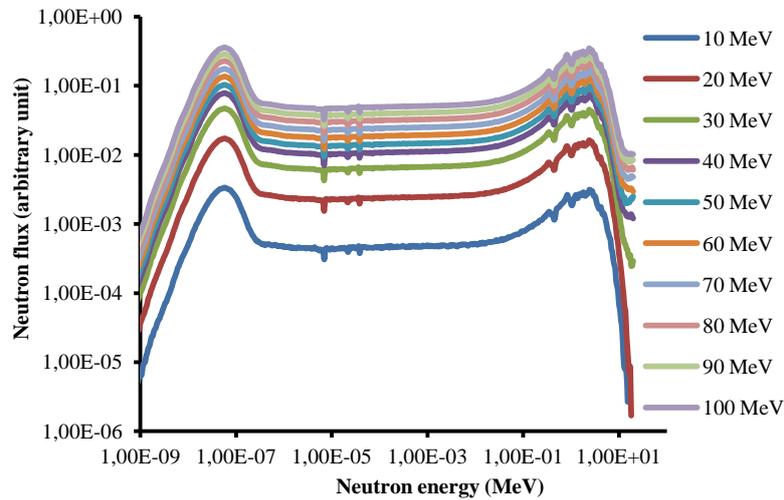


Fig.2 Dependence of neutron spectra to the proton projectile energy

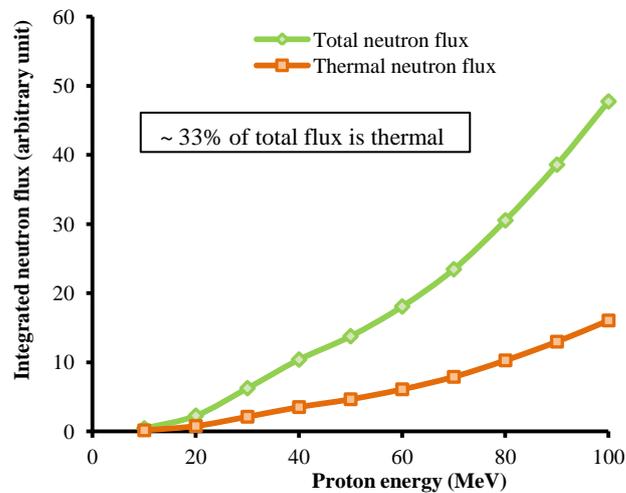


Fig.3 Dependence of integrated neutron spectra to the proton projectile energy

Deposited heat was calculated using F6 tally of the used computational code. The results showed maximum deposited power in axial direction is 17 W/cm^3 using 30 MeV protons which experienced at 5.2 cm position i.e. very closest to beam window; except near the window other positions experience a heat deposition less than 0.5 W/cm^3 . Proton energy enhancement decreases the high discrepancy between the beared deposited heat by the window vicinity area and the used aqueous solution. In radial direction, maximum deposited

heat is experienced by central region of the aqueous homogenous reactor; the region is related to entrance proton beam domain (Figs. 4-5).

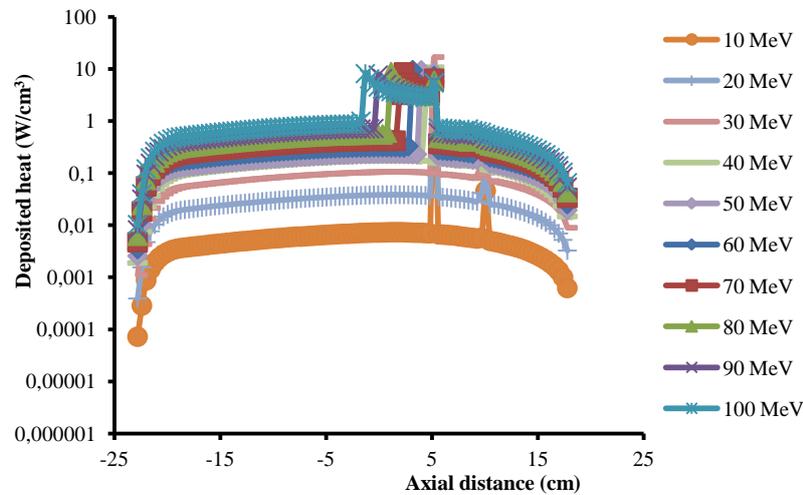


Fig.4 Dependence of axial deposited heat to the proton projectile energy, normalized for 300 μ A current.

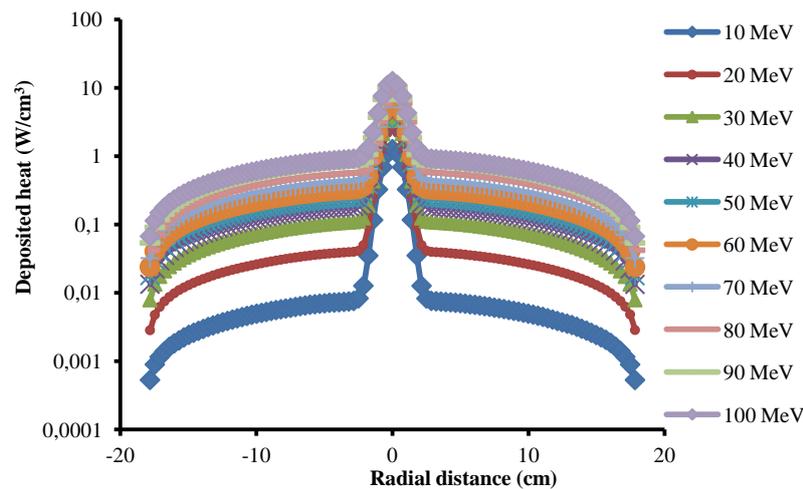


Fig.5 Dependence of radial deposited heat to the proton projectile energy, normalized for 300 μ A current.

Deposited power experienced by the aqueous solution was determined for different projectile energies and beam currents of 10, 30, 150 and 300 μ A. Obviously higher projectile energies produce higher deposited power inside the subcritical core. Hence, using higher beam currents or projectile energies an operation power of several 10 kW order is obtainable (Fig.6).

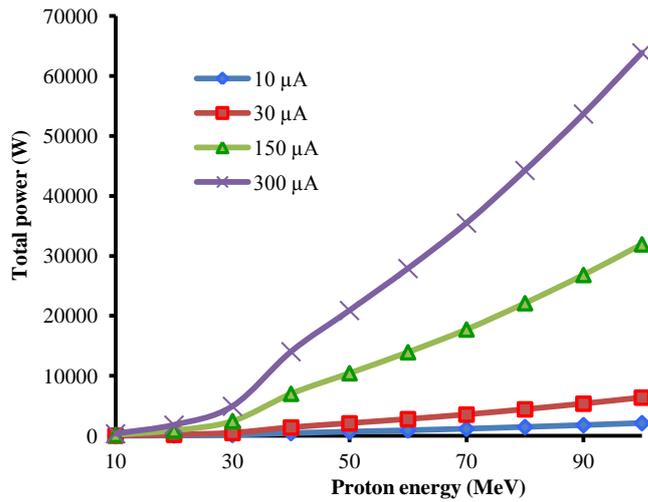


Fig.6 Dependence of deposited heat in the modeled core to the proton projectile energy, normalized for different beam currents.

F7 tally calculates the deposited power as a result of the occurred fissions inside the subcritical core. For 20-40 MeV there was not any noticeable discrepancy between the F6 and F7 tally outputs (<2%). While the other energies resulted in noticeable discrepancies; for $E_p > 40$ MeV deposited power by fission was less than total heat deposition and for $E_p < 20$ MeV the previous result was inverse. F6 tally determines temperature profiles and F7 tally determines the delivered power to the aqueous subcritical solution. The calculations showed 20 MeV protons with 300 μA deliver about 1.8 kW power as a result of the occurred fissions. According to burn-up calculations at 1.8 kW power, about 550 Ci of carrier-free ^{99}Mo and 137 Ci of ^{131}I (can be considered as carrier-free regarding to a proposed time for decay of the other radioisotopes) will be produced every week.

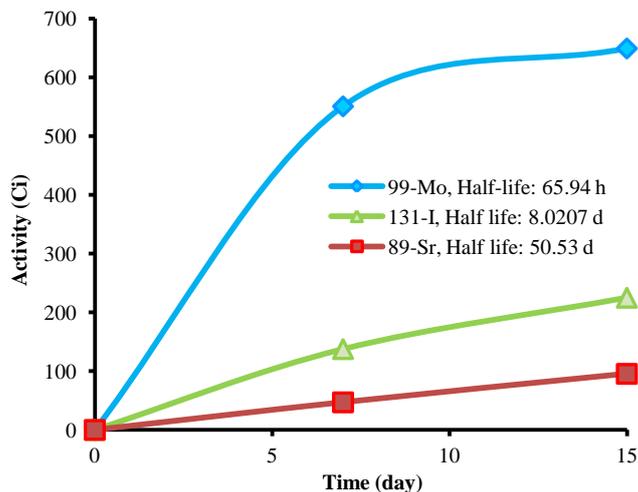


Fig.7 Dependence of the produced radioisotopes' activity on burn-up days at 1.8 kW power.

^{89}Sr is produced with activity of ~ 47 Ci/week, but ~ 0.3 Ci/week of ^{90}Sr with a half-life 28.79 y accompanies the ^{89}Sr product. The burn-up calculations showed the plutonium isotopes and

other high level radiotoxic elements produced in <0.5 g after two weeks burn-up at 1.8 kW power. Also the neutron poison buildups are ignorable to disturb the subcritical core effective multiplication (Table 2).

Table 2 Comparison of actinide inventory in the modeled aqueous solution during a 15-days burn-up at 1.8 kW power

Element	Mass (g)	Activity (Ci)	Half-life	Emitted particle
²³² Th	1.48E+03	1.63E-04	1.405E+10 y	α
²³³ Pa	3.13E-03	6.50E+01	22.3 m	β-
²³³ U	6.43E-04	6.20E-06	26.967 d	β-
²³⁵ U	1.46E+03	3.16E-03	1.592E+5 y	α
²³⁶ U	3.69E-02	2.39E-06	7.038E+8 y	α
²³⁸ U	5.94E+03	2.00E-03	2.455E+5 y	α
²³⁹ Np	1.77E-02	4.11E+03	2.342E7 y	α
²³⁹ Pu	6.22E-02	3.86E-03	23.45 m	β-
	²³⁵ U/ ²³⁸ U	²³² Th	¹³⁵ Xe	¹⁴⁹ Sm
Consumption (g)	1/<0.001	<0.001	—	—
Buildup (g)	—	—	2.732E-04	1.076E-03
Δk/k _{shut down} (pcm)	After 6h:	1.06	After 12h:	-12.76

The neutronic parameters' calculations showed the effective multiplication of the modeled subcritical core is 0.94027± 37 pcm. The subcritical core β and β_{eff} parameters were 738 and 670 pcm respectively. Fuel, coolant and reflector temperature reactivity coefficients were -2.43, +2.36 and +1.03 pcm/°C respectively. Void coefficients of fuel, coolant and reflector were -942, -632 and -652 pcm/% void respectively.

Conclusion

Design and development of such subcritical aqueous reactors driven by low and medium-energy accelerators assures secure and economic production of radiomedical-required isotopes. Some complementary modifications can be applied to increase the production capacity to >1000 Ci/week.

Reference

1. Homogeneous Aqueous Solution Nuclear Reactors for the Production of Mo-99 and other Short Lived Radioisotopes, IAEA-TECDOC-1601, (2008).
2. F. Stichelbaut, Design of New Solutions to Produce ⁹⁹Mo Using Sub-Critical Systems, IBA, Belgium, (2011).
3. S. Chemerisov, A.J. Youker, A. Hebden, N. Smith, P. Tkac, C.D. Jonah, J. Bailey, V. Makarashvili, B. Micklich, M. Kalensky, G.F. Vandegrif, Development of the mini-SHINE/MIPS experiments Mo-99 2011 — Molybdenum-99 Topical Meeting, December 4-7, 2011.

4. F. Stichelbaut, Y. Jongen, Design of Accelerator-Based Solutions to Produce ^{99}Mo Using Lowly-Enriched Uranium. Prog. in Nucl. Sci. and Tech., Vol. 2, pp.284-288 (2011).
5. D.B. Pelowitz (2008) Users' manual versión of MCNPX2.6.0, LANL, LA-CP-07-1473.
6. M.L Fensin (2008) Development of the MCNPX depletion capability: A Monte Carlo depletion method that automates the Coupling between MCNPX and CINDER90 for high fidelity burn up calculations. Florida University.
7. K. Elgin, A study of the feasibility of ^{99}Mo production inside the TU Delft Hoger Onderwijs Reactor, thesis, 2014.