GEOMETRY OPTIMIZATION OF URANYL NITRATE LIQUID TARGET SYSTEM FOR ^{99}Mo PRODUCTION USING 30 MeV PROTON CYCLOTRON

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Abstract

^{99}Mo radioisotope is widely demanded by world community for both therapy and imaging purposes. Accelerator and reactor-based routine procedures are applied to produce this radioisotope. Newly proton-fission production method has been taken in attention by some research centers. In the present work, computationally investigation of the ^{99}Mo production yield in uranyl nitrate liquid targets irradiated by 30 MeV proton particles was aimed. In the present work, height and radius of the chamber of the liquid target are optimized considering minimum required solution for production of an adequate amount of ^{99}Mo. The obtained results showed uranyl nitrate liquid targets could be efficiently used to produce 18.83 Ci yield of ^{99}Mo using 30 MeV proton irradiation of the optimized-dimension target after 150 μA current application for 24 hours irradiation. Also this accelerator-based procedure using proton-fission induction in the natural uranium dissolved in nitrate solution presents a potentially competitive alternative in comparison with the reactor-based or other accelerator-based methods to produce ^{99}Mo and some other fission products simultaneously.

Keywords: ^{99}Mo production, ^{nat}Uranyl nitrate, MCNPX, Accelerator irradiation

Introduction

Technetium-99m is a principal radioisotope used for medical diagnostic imaging and accounts for approximately 80% of all nuclear medicine procedures. ^{99m}Tc is the daughter product of ^{99}Mo, and it is usually supplied in the form of a ^{99}Mo/^{99m}Tc generator to make diagnostic radiopharmaceuticals [1]. The global supply chain of ^{99}Mo used for generator production has several complex difficulties, which makes the system fragile and unreliable. Aging reactors, limited suppliers and transportation obstacles are all challenges faced with the supply chain. ^{99}Mo has been exclusively produced in 7 nuclear reactors, with an average age of these reactors being more than 40 years old. These 7 nuclear reactors are the Belgian Reactor 2 (BR-2) in Belgium, High Flux Reactor in The Netherlands, LVR-15 REZ Reactor in the Czech Republic, Maria Research Reactor in Poland, National Research Universal (NRU) in Canada, Open Pool Australian Light water reactor in Australia, and South African Fundamental Atomic Research Installation in South Africa [2].

After being produced in nuclear reactors, is ^{99}Mo transported to a processing facility to be chemically separated and purified. There are currently only 5 ^{99}Mo -processing facilities in the world, which are Australian Nuclear Science and Technology Organization (ANSTO) in Australia, the Institute for Radio Elements (IRE) in Belgium, Mallinckrodt in the Netherlands, Nordion in Canada, and Nuclear Technology Products (NTP) Radioisotopes SOC Ltd. in South Africa. The finished ^{99}Mo product material is then isolated and shipped to 1 of 8 generator-manufacturing facilities that supply ^{99}Mo in the form of a ^{99}Mo/^{99m}Tc generator to send users, such as nuclear pharmacies and hospitals [2].

Currently other new methods for production of this important radioisotope are studied by approach of eliminating HEU targets and supplement in small scales for region and central use. In this way solid target plates are used for the production of ^{99}Mo. The targets are generally either miniature Al-clad fuel plates or pins containing U–Al alloy or a thin film of UO_2 coated on the inside of a stainless steel tube [3]. Hence, feasibility and economically study of such liquid target irradiation by proton accelerators in order to ^{99}Mo production was proposed in this work.

Material and methods

In this study, MCNPX 2.6.0 was used for investigation of feasibility of^{99}Mo productionusing proton-fission induction and optimization of geometrical condition of utilized uranyl nitrate liquid target. MCNPX^{TM} is a general purpose Monte Carlo radiation transport code designed to track many particle types over broad ranges of energies. MCNPX 2.6.0 is the latest Radiation Safety Information Computational Center (RSICC) release of the code that includes many new capabilities, particularly in the areas of transmutation, burnup and delayed particle production [4]. ^{99}Mo production yield was determined according to the MCNPX results obtained by HISTP card of the input file. HISTP solves Batman decay chain equations using CINDER90 package involved in the MCNPX code.

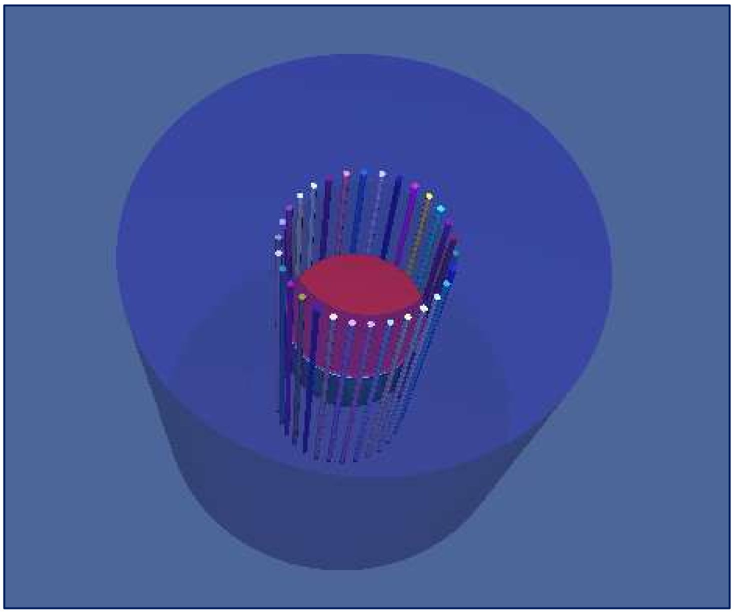

Fig.1. Liquid target layout simulated by MCNP.

In this study, the liquid target system is simulated according to the targetry used at both Duke University Medical Cyclotron and Wisconsin Medical Cyclotron for F-18 production (see Fig.1) [5]. General scheme of the simulated target is shown in Fig. 2. This target system includes the main body which made of ZnCu (target holder), cooling channels, beam line and

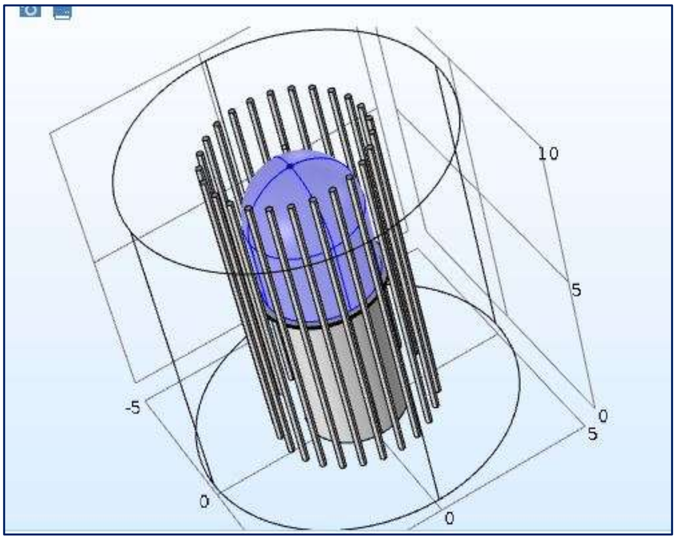

Fig.2. 3D view of liquid target without container (Target holder).

target container (Target chamber). Beam window of the target system is a 500-micron aluminum layer and helium cooling layer with a thickness of 1 mm was considered to cool the lateral surfaces of the container. Next, the ^{99}Mo production has been investigated in various radiuses of cylindrical liquid target, and height changes and an optimized dimension was suggested.

Results and discussion

Our liquid target solution contains 20 grams per liter concentration of natural uranium. It is assumed that this target is irradiated by a 30 MeV proton beam energy with 150 μA current that produced by a cyclotron accelerator. The target radius changed as 2 mm, 4 mm, 6 mm and 8 mm and deposited heat power and molybdenum production yields were investigated in any different situations. According to Fig. 3 it can be seen that ^{99}Mo yield has max value of 193.4898 Mbq/μAh using 2 mm radius. Other results such as deposited heat power and proton flux that obtained by calculations were given in Table1.

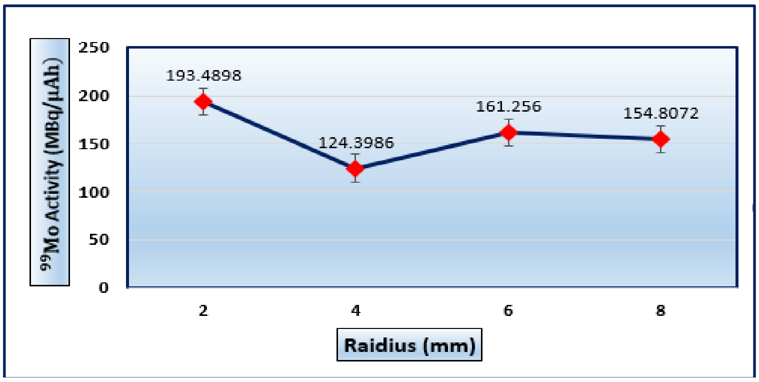

Fig.3. ^{99}Mo production dependence on the radius of target of uranyl nitrate solution.

Table 1. Some of important information in liquid target, the calculation errors <1%.

Radius (mm)	Neutron Flux ($n/cm^2 \cdot s$)	Proton Flux ($p/cm^2 \cdot s$)	Total Deposited Heat Power (W)	Neutron fission heat power (W)	Target Volume (cm^3)
2	1.61E+12	4.18E+14	6.52E+01	1.62E-04	1.42E-01
4	1.24E+12	2.99E+14	9.37E+02	5.52E-04	6.37E-01
6	9.60E+11	1.91E+14	3.71E+03	1.05E-03	1.58E+00
8	7.56E+11	1.17E+14	8.62E+03	1.58E-03	3.08E+00

First, we discuss about the total deposited heat power in liquid target. We know that by increasing the radius of the target, the solution proton-exposure is increased. By volume increasing more fission could occur in the solution and obviously more total and also neutron-fission deposited heat power in solution is experienced so that from 65.2 W in a radius of 2 mm reaches to the extra-large value of 8.62 kW in radius of 8 mm. However, in this case (kW heat power) the jet cooling is applicable, but taken attention to working with radioactive materials we are interested to minimize other problems such as difficulties related to the heat transfer. So by regarding both terms of production yield and heat transfer, the radius of 2 mm is really more desirable than others.

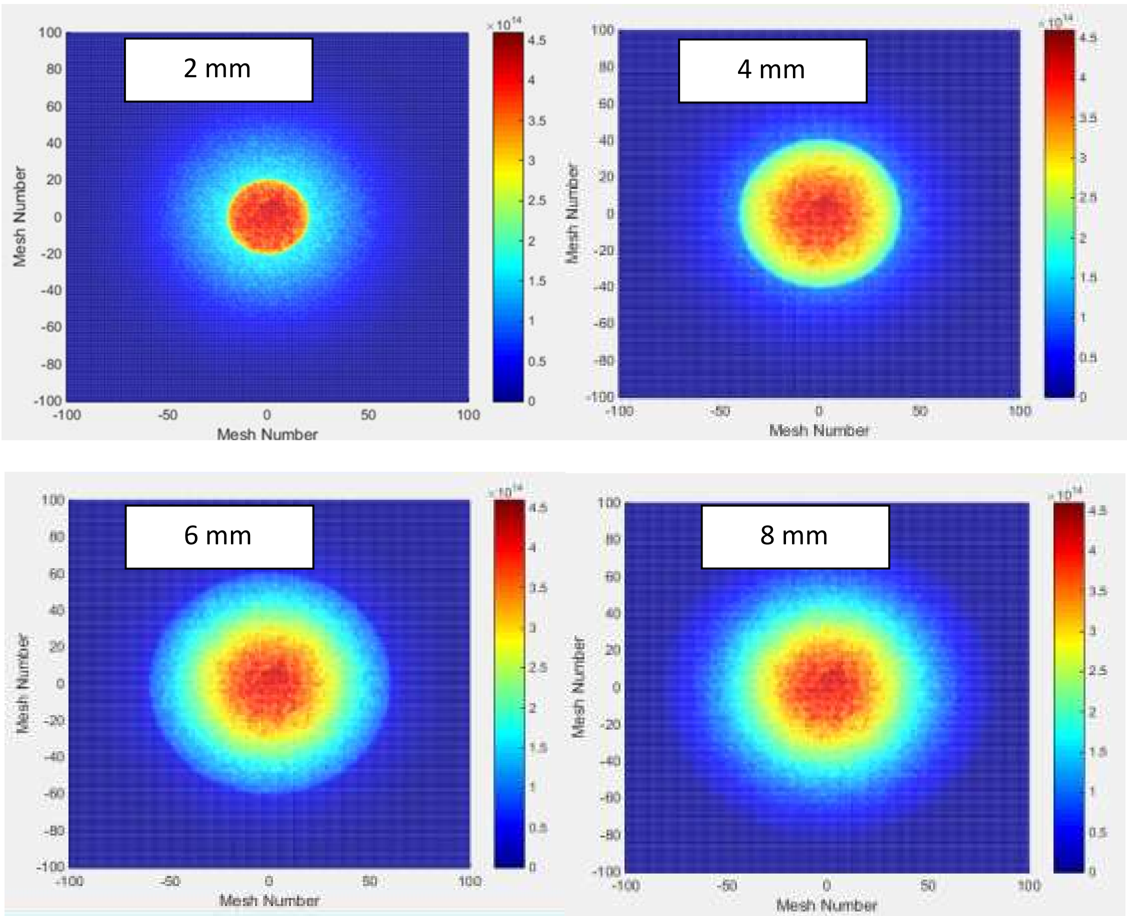

Fig. 4. Proton flux distribution inside the targets with different radii.

It was expected that by the radius increasing, the production yield increases because a Gaussian distribution with a FWHM (Full Width Half Maximum) of 0.8 was used to irradiate the cylindrical liquid target. However it should be illustrated the chain reactions involving absorption, decay and production determine the final yield of the product. Enhancement of neutron flux could decrease ^{99}Mo yield because of n+^{99}Mo reaction rate. According to Fig. 1 clearly higher radius experience higher proton flux availability and thereby higher deposited heat. The carried out calculations showed 2 mm radius is desirable to produce the highest ^{99}Mo yield and the least deposited power.
In the next step different height of the target was investigated. According to our calculation results, height variation does not cause a significant change in the production of ^{99}Mo (Table 2).

Table 2. Some of important information in liquid target, the calculation errors <1%.

Height (mm)	Neutron Flux ($n/cm^2 \cdot s$)	Proton Flux ($p/cm^2 \cdot s$)	Total Deposited Heat Power (W)	Neutron fission heat power (W)	Target Volume (cm^3)
8	1.84E+12	5.08E+14	5.37E+01	1.53E-04	1.17E-01
12	1.43E+12	3.56E+14	7.68E+01	1.69E-04	1.68E-01
16	1.17E+12	2.73E+14	9.98E+01	1.80E-04	2.18E-01
20	9.89E+11	2.22E+14	1.23E+02	1.86E-04	2.68E-01

Height of 8mm, 12 mm, 16 mm and 20 mm have been checked and according to Table 2 by increasing the volume and mass of the irradiated solution, total and neutron-fission deposited heat power enhances. But the protons and neutrons flux is reduced as a result of absorption process. Axial distribution of neutron and proton flux is shown in Fig.5. According to the figure there is an intense distribution at the beginning of the target.

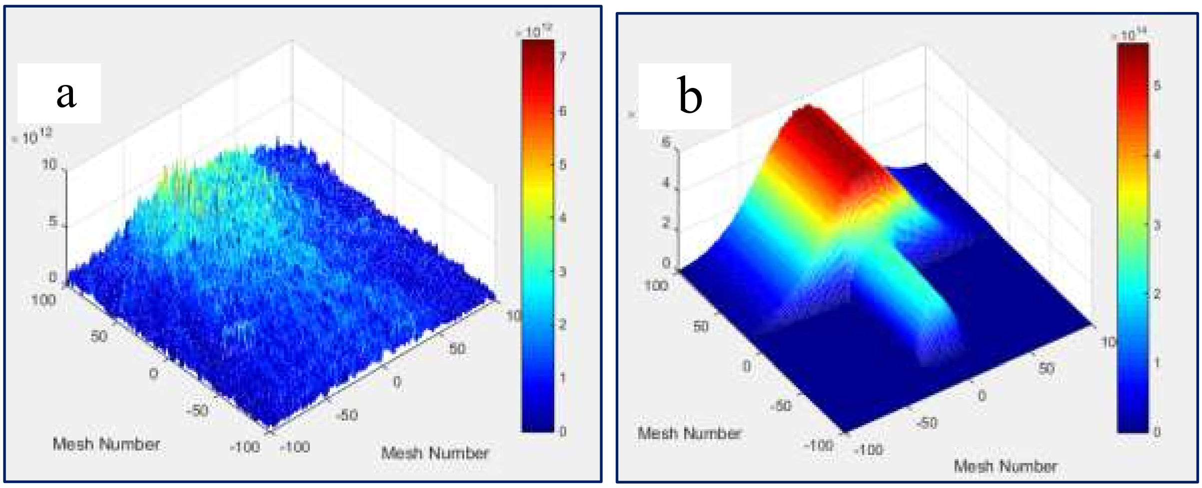

Fig.5. Distribution of a) neutron flux and b) proton flux in liquid target.

According to the results, as well as the fact that none of the investigated heights have no preference in terms of the production yield. The height can be selected arbitrary. Because better heat transfer occurs in bigger target area so we choose a height of 20 mm. For 24 hours irradiation the ^{99}Mo yield is about 18.83 Ci. Therefore, we would account the number of required uranium atoms by A=λN equation. So, the necessary number of atoms is equal to 4E+15 while the available atoms inside the solution are 7.43E+18 for2.68E–1 cm^3 target volume (according to a radius of 20 mm). So this tiny target could be used to produce desirable amount of ^{99}Mo in low-cost condition with minimum nuclear waste and independency to rector-based procedures.

Conclusion

The liquid target containing natural uranium can customize and develop our prospects for the peaceful use of the nuclear industry and also be regarded as excellent replacement of HEU solid target. In the same time this procedure reduces our dependency to high-cost operation nuclear reactors. Our funding showed about 18.83 Ci/week of ^{99}Mo is producible by means of the optimized target.

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