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FEASIBILITY STUDY ON Mo-99 PRODUCTION USING HYBRID METHOD BASED ON HIGH POWER ELECTRON ACCELERATOR

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Abstract

In this study, the idea ^{99}Mo production using hybrid method based on electron accelerator has been presented. Two different main production channels, ^{100}Mo(γ,n)^{99}Mo and ^{98}Mo(n,γ)^{99}Mo, can be used for ^{99}Mo production in this system. By considering high power Linac (30 MeV-1 mA) and one-stage approach, the calculation of ^{100}Mo(γ,n)^{99}Mo reactions in the optimized ^{100}Mo target in two different designs (strip and disc) has been simulated. It is predicted that about 61 and 53 Ci of ^{99}Mo activity per 24-hour irradiation on the strip target and the disc plates can be achieved, respectively. The threshold energy of photoneutron at ^{100}Mo is about 9 MeV, so a large part of bremsstrahlung photons cannot participate in photoneutron reaction. For feasibility study, new hybrid approach has been tested by 10 MeV Rhodotron. Due to the low threshold of photoneutron in deuteron (about 2.2 MeV) and significant low energy photons in ^{100}Mo, photoneutron flux is available. So, Molybdenum target in heavy water Tank increases the production yield of ^{99}Mo using neutron absorption reaction in ^{98}Mo. The total activity of ^{99}Mo has been predicted about 0.23 Ci per 24 hours e-beam irradiation.

INTRODUCTION

^{99m}Tc is the most used radiopharmaceutical in diagnostic nuclear medicine, which is approximately used in 80 percent of nuclear medicine imaging [1, 2]. ^{99m}Tc has a short half-life, $t_{1/2}$= 6 h. Therefore ^{99}Mo with transformation into ^{99m}Tc (β decay mode by $t_{1/2}$= 66 h) is a desirable option. At present, the total need of ^{99}Mo in Iran is 100 Ci with 6-day calibration. This demand annually increases by10% [3].

The major supply of ^{99}Mo is produced in research reactors by irradiating uranium targets. Some shortcomings of reactor-based methods are: (1) shut down of the reactor (suddenly accidents, predetermined repairing programs or expired operating life), (2) unavoidable production of undesirable isotopes in products (with the need to costly separation process) and (3) the need to high-enriched uranium with a more cost and safety challenges to achieve ^{99}Mo with high activity [3-6]. So, the alternative production methods as a short, middle or long-term solution are necessary. By the way, different researches follow the ^{99}Mo / ^{99m}Tc production using non-reactor methods [6-12].

Accelerator-based production is an alternative method. ^{99}Mo production using high power electron linear accelerator (Linac) by ^{100}Mo(γ,n)^{99}Mo and ^{98}Mo(n,γ)^{99}Mo production channels is available, which due to the safety and economic aspects, it is an appropriate method [13-14].

Two main approaches based on ^{100}Mo(γ,n)^{99}Mo reaction, one-stage and two-stage approaches, can be followed using Linac. In two-stage approach, the e-beam is converted to high-energy bremsstrahlung photons in the high-density high-Z target. Then, ^{99}Mo is produced through (γ,n) reactions by photons with energies more than the threshold of (γ,n) in the ^{100}Mo target (more than 9 MeV) [15].

In the one-stage approach, a target is e-γ converter as well as the (γ,n) target simultaneously, so self-absorption of photons in e-γ converter will be prevented and produced photons participate directly in (γ,n) reactions, and subsequently, the production yield of ^{99}Mo will be increased [4].

For isotope production by Linac and through the neutron capture reaction of ^{98}Mo(n,γ)^{99}Mo, variety approaches are available. In this method, the main purpose is producing an adequate flux of thermal neutrons to participate in neutron capture reaction at a target containing ^{98}Mo isotope.

Our final goal is presenting a hybrid system involving both of the mentioned channels simultaneously to increase the production yield. For this purpose, a high energy electron accelerator is needed. In Iran, an 11 MeV electron Linac is under construction at IPM [16]. This Linac has a capability of upgrade to higher energies. Also, other programs to provide or construct new electron accelerators are going on by Atomic Energy Organization of Iran (AEOI) and other related scientific centers in Iran.

But for now, a 10 MeV 100 kW Rhodotron is operational in Yazd radiation processing center [17]. The energy of this accelerator is not proper for production through the (γ,n) reaction on the ^{100}Mo. But, by participating high flux bremsstrahlung photons in (γ,n) reaction in heavy water, proper neutron flux (photoneutron threshold of deuterium is about 2.2 MeV) can be produced. If isotope production using this accelerator and through ^{98}Mo(n,γ)^{99}Mo channel can be proved, it can be concluded that the idea of ^{99}Mo hybrid production via the mentioned channels using high energy and high power electron accelerator is possible.

Beside the nuclear simulations, thermal-mechanical analysis should be considered. Therefore, a part of this study is dedicated to the thermal-mechanical analysis, too. An appropriate mechanical design as well as the ease of production and installation should be considered for the removal of heat from the target.

^{99}Mo PRODUCTION THROUGH PHOTO-NEUTRON REACTION

In this section, two different target designs based on using a high-energy electron accelerator through the photoneutron reaction in ^{100}Mo target (in accordance with one-stage approach) will be discussed. Physical aspects of production and how finding the optimal dimensions are as described on [18,19]. According to these studies, an electron

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Linac (30 MeV-1 mA) is suitable for ^{99}Mo isotope production. Also, the optimum thickness of target has been calculated about 1.8 cm.

The majority of industrial accelerators have a scanning horn at the end-window of the beamline. Depending on the length of horn, the connection between it and target structure has some limitations. Although, in some cases the use of beamline at straight direction on target is available. In addition, due to high cost of ^{100}Mo, melting and corrosion of target should be avoided, two different target designs by considering indirect cooling system has been simulated.

Strip Plates Design (Applying a Beam Scan System on the Target)

The detailed descriptions of optimizing the dimension and thermal analysis for this design have been presented at [19]. Based on the results, 9 strip plates with thickness of 0.2 cm and width of 3 cm is possible (Fig. 1) to have at least length of 10 cm as well as satisfy the proper thermal conditions [18,19].

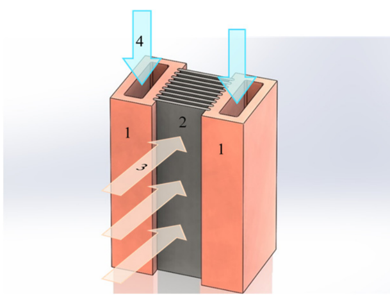


Figure 1: Schematic view of the designed geometry for the stripe plate target. 1: copper clamps as a cooling ducts (hydraulic diameter, D_H = 1.56 cm), 2: target plates (^{100}Mo), 3: e-beam direction, and 4: water inlet.

The ^{99}Mo activity of plates can be calculated based on the reaction rate relation based on Eq. (1). Which is used in Eq. (2).

$$R = N' \int \varphi(E)\, \sigma(E)\, dE \qquad (1)$$

Where, φ is the particle flux (the bremsstrahlung photons, #/cm^2.s), N' is the atomic density (#/cm.barn), and σ is microscopic cross sections (cm^2) [20].

$$A(t) = \lambda N = R(1 - e^{-\lambda t}) \qquad (2)$$

Where, A(t) is the produced activity in irradiation time of t, λ is the ^{99}Mo decay constant and N is the number of radioactive nuclei at the time of t.

The photons flux has been calculated by MCNPX2.6 code, then using the cross-section of ^{100}Mo (γ, n) ^{99}Mo from ENDF / B-VIII.0 library [21], the activity of each plate (Fig. 2) has been obtained until saturation time (330 hours, equal to 5 half-life of ^{99}Mo).

According to Fig. 2, after 24 h irradiation, the 4th plate with 10 Ci is the most activated plate against the first plate with 2.2 Ci as the least activated plate. Also, the activity of 61 Ci will be obtained from all the plates at end of bombarding (EOB) after 24 h and at the saturated situation (t=330 h), total activity of all the plates is equal about 265 Ci.

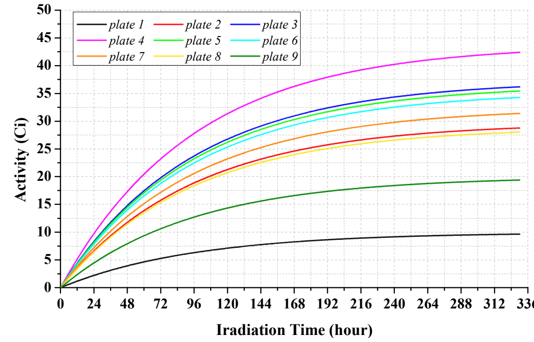
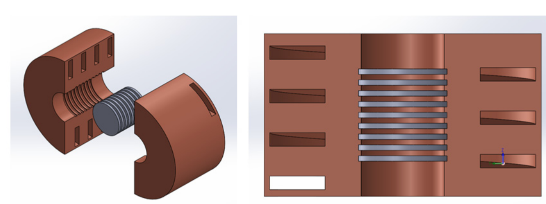


Figure 2: Calculated activity of ^{100}Mo plates during the accelerator irradiation time (for 1mA current at 30 MeV of continues e-beam), e-beam first incidents on plate 1.

Thin Discs Design (with Spiral Coolant Passages)

In this design, according to Fig. 3, the ^{100}Mo target is considered as thin discs (9 disc with 0.2 cm in thickness and 3.2 cm in diameter). The designed copper cooling system consists of a spiral coolant passage that avoids water to contact with the target.

The target cooling system consists of a copper metal body with spiral coolant passages. For ease of installation and separation of targets into this metal body, the body is divided into two parts, with targets in the middle of them. Coolant passages are embedded with a rectangular cross-section, to increase the contact of the coolant with the walls relative to the circular cross-section. The inlet cross-section in this rectangular cooling system has dimensions of 0.2 cm in length and 0.5 cm in width, so the hydraulic diameter for this section is 0.8 cm. The overall height of the structure is 6 cm. In the middle part of the structure, where the targets are located, there are 0.1 cm thick edges that connect the target to the body.



a b

Figure 3: Thin disc target with spiral water channels, a: Isometric view, b: A section of the structure.

Power loss of the targets and structure has been calculated by MCNPX2.6 code. By calculating the convection coefficient, the temperature distribution of the structure has been obtained using ANSYS software. The result of simulation illustrates that the maximum temperature of the structure for the water with inlet velocities more than 4 m/s don't exceed the assurance temperature, 1300°C. It should be noted that 1300°C has been considered for avoiding recrystallization [19].

The ^{99}Mo activity for each disc has been calculated like the strip target. The results are shown in Fig. 4.

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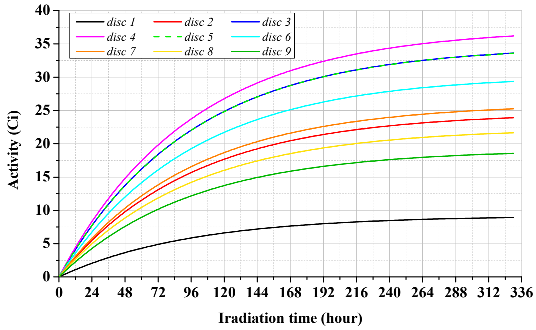

Figure 4: Calculated activity of ^{100}Mo discs during irradiation time (for 1mA current at 30 MeV of continues e-beam), e-beam first incidents on disc 1.

Like to the strip design, first and fourth disc plates are the most and the least activated plates, respectively. After 24 h irradiation, the total activity of target will be about 53 Ci, which in saturated state is growing near 230 Ci.

The possible irradiation time depends on the facilities' capability. In the commercial scale, due to the daily distribution program such as the program that mentioned at reference [22], the 24 hour irradiation is a common period.

^{99}Mo PRODUCTION THROUGH NEUTRON CAPTURE REACTION

The isotope production through $^{98}Mo(n,\gamma)^{99}Mo$ channel is available if the adequate flux of thermal neutrons receives to ^{98}Mo target. ^{98}Mo is the most common isotope, comprising about 24% of ^{nat}Mo, thus providing ^{98}Mo target is more economical than ^{100}Mo target. In this step, the distribution of neutron flux in a heavy water tank by dimensions of 40×40×50 cm^3 has been investigated. A t the deep of 10 cm from top of the tank, a W converter has been located by dimensions of 48×3×0.15 cm^3. The tank itself constructs from a thick carbon layer (as a neutron reflector) that placed between two thin steel layers. The schematic view of this system and the primary results of neutrons distribution are shown in Fig. 5 and Fig. 6, respectively.

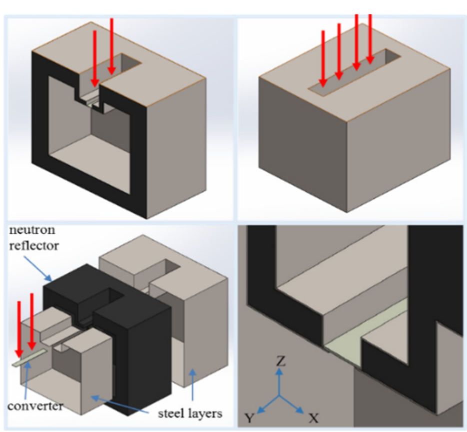

Figure 5: Construction of heavy water tank from different views (red arrows show the direction of e-beam).

It can be expected by optimizing the system, the neutron flux and subsequently production rate will be increased.

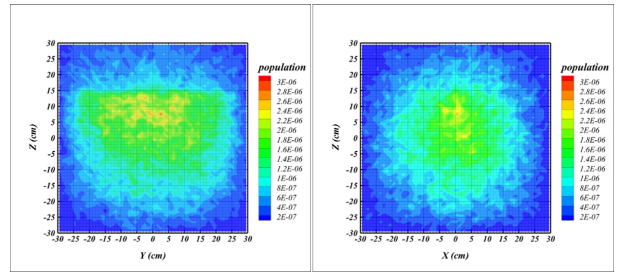

Figure 6: Distribution of neutrons per one particle source, in the heavy water tank. Each mess is 1cm^3.

As initial estimation, the ^{99}Mo activity for number of 32 ^{98}Mo plates has been calculated. The dimension of each plate is 8, 9 and 0.2 cm, respectively in length, width and thickness, in corresponding with high neutron population areas at the heavy water tank. A cording to Fig. 7, the total activity of these plates, through neutron capture reaction using 10 MeV Rhodotron, is about 0.23 Ci after 24 hour e-beam irradiation on ^{98}Mo plates.

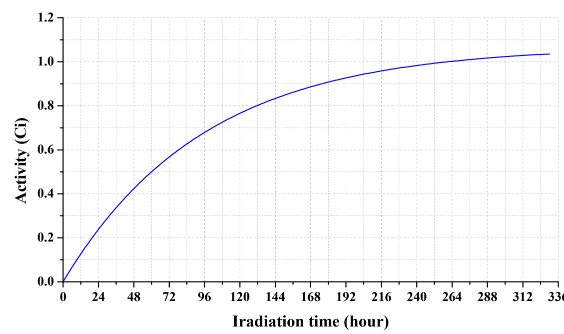

Figure 7: Total ^{99}Mo activity of ^{98}Mo discs during the irradiation time (for 10 MeV e-beam with power of 100 kW).

CONCLUSION

In this study first, production of ^{99}Mo isotope trough (γ,n) reaction using a high energy electron accelerator (30 MeV Linac) have been surveyed. Due to the some limitations on assembling the target to beamlines' end-window, two different designs of target have been suggested and simulated. It is predicted about 61 and 53 Ci of ^{99}Mo activity can be achieved, for strip and disc design, respectively, after 24-hour e-beam irradiation on ^{100}Mo target.

Also because of high cost of enriched ^{100}Mo target, the thermal-mechanical calculation has been performed to be sure about avoiding target melting and corrosion, by considering assurance temperature of 1300 °C as a maximum allowed temperature for molybdenum target.

Then, due to the idea of simultaneous employment of two production channels that can lead to an increase in production yield, production through neutron capture has been studied, too. For proving this idea, the 10 MeV Rhodotron located in Yazd gamma irradiation center, has been considered for simulations. It has been predicted the total activity of ^{99}Mo is about 0.23 Ci per 24 h e-beam irradiation, which it can increase by using high energy accelerator and more optimization of ^{98}Mo target plate's dimension.

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