Original article:

Toward for production of Molybdenum-99 by irradiation of MoO3 target in a neutron flux

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Abstract

Background: Molybdenum- 99 is a parent isotope of Technetium-99m (^{99m}Tc) as intermediate to diagnosis and radiation treatment. This production is made according to irradiation of Molybdenum-98 with thermal neutrons. The cycle comprises a complex of MoO_3 or the percentage of Mo-98 is 24.13%, this compressed mixture in an irradiation capsule of aluminum; the latter is disposed in the central thimble of a nuclear reactor so that the thermal neutron flux is at a maximum in order to generate ^{98}Mo to ^{99}Mo by nuclear reaction of (n, γ) where the cross-section of molybdenum is (0.13 ± 0.0013) barn***. Method***: The purpose of this study is to validate of MoO_3 target that will be used for the production of Molybdenum-99 in the central thimble irradiation position of the TRIGA Mark II research reactor at CNESTEN (Morocco National Center for Nuclear Energy, Sciences and Techniques), The thermal neutron flux used for activity calculation of Mo-99 used reactor of research TRIGA Mark-II is 3.1 10^{11} n/cm²s at 250 KW, 2.4E+13 n/cm²s at 1.1 MW and 3.01 10^{13}n/cm²s at 2MW, we are using Fortran-90 code for calculate the activity***. Result:*** The result's finding was validated by other studies.

Keywords: CNESTEN; central thimble; TRIGA Mark-II; Molybdenum-99; thermal neutron flux; Activity

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Introduction

Radioactive isotopes are usually used in industry[1], medicine[2] and the life sciences[3]. the field of nuclear medicine uses radiation to give of diagnostic and treatment[4].

Nuclear reactors permit the production of radioisotopes in greater quantities, with a lower cost. It uses the bombardment of a target with neutrons, which cause nuclear reactions. Produced radioisotopes must be radioactive long enough to allow their transport.

Tens of millions of nuclear medicine events are performed every time and the order for radioisotopes as molybdenum used for medical applies is growing quickly and is having a half-life of 65.94 hours used as the 'parent' in a generator to generate technetium-99m[5-8]. The production of Technetium-99m is a multi-step process that begins with Molybdenum-98 (Mo-98), a radioactively stable isotope of Molybdenum. Mo-98 is irradiated with thermal neutrons, neutrons that are in equilibrium with their surrounding environment at room temperature, which are absorbed by the Mo-98, turning it into

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Molybdenum-99 (Mo-99).

Technetium-99m is the radioisotope a large amount widely used in medicine, engaged in a quantity of 80% of all nuclear medical events, [9-11]. ^{99m}Tc is a serious medical isotope used in a diagnostic and practical study before organs and anatomical systems[12]. With such wide-reaching clinical applications, Tc-99m is the most important isotope to consider for domestic production. It is used, for the most part, to locate tumors in the body[13]; examine cardiac purpose following heart attacks, map blood flux into the brain and guide operation. The in sequence as of these studies is used by many medical specialists, including radiologists, cardiologists and oncologists to well again diagnose and take care of patients. It is furthermore used for brain, brain perfusion, liver, bone and bone marrow, blood pool, and pulmonary perfusion imaging, etc.,[14,15].

Molybdenum trioxide is found frequently in nature, but always in the form of molybdate. Furthermore, known as molybdenum trioxide, this product is a crystalline powder of white color - yellow, which is sparingly soluble in water colder. MoO_3 is produced industrially by roosting molybdenum disulfide and as natural molybdenum contains 24.13% of mixture MoO_3.

$$2MoS_2 + 7O_2 \longrightarrow 2MoO_3 + 4SO_2$$

The laboratory grouping entails the acidification of aqueous solutions of sodium molybdate with perchloric.

$$Na_2MoO_4 + H_2O + 2HClO_4 \longrightarrow MoO_3(H_2O)_2 + 2NaClO_4$$

Molybdenum trioxide dissolves, to some extent, in water to provide molybdic acid in base it dissolves to afford the molybdate anion.

Methods and materials

Theoretical study for production of Molybdenum -99

Molybdenum-99 is produced by irradiation of MoO_3 targets in a thermal neutron flux according to nuclear reaction (n, γ). The nuclear equations for the reactions to produce molybdenum-99 from molybdenum-98 can be written like this in reaction 1: Absorbing a neutron: Beta decay to produce technetium-99 reaction 2:

$$^{98}_{42}\mathrm{Mo} + ^{1}_{0}\mathrm{n} \qquad ^{99}_{42}\mathrm{Mo} + \gamma \qquad (1)$$

$$^{99}_{42}Mo \qquad ^{99}_{43}TC = {}^{0}_{-1}\beta \qquad (2)$$

A reaction of Radiative capture of a neutron by an isotope can be written as follows 3:

$$^{98}_{42}Mo + ^{1}_{0}n \qquad ^{99}_{42}Mo + \gamma \qquad (3)$$

Usually we write this equation in the form: $^{98}_{42}Mo(n,\gamma)^{99}_{42}Mo$

The number **N(Mo-99)$_f$** of the isotope $^{99}_{42}Mo$ forms during irradiation for a period of time **dt** is proportional to the number of isotopes of the target (**N_0**), a cross section of radiative capture (**$\sigma_{n\gamma}$**) and the flux of incident neutrons **φ** in equation 4:

$$N(\text{Mo-99})_f = N_0\sigma_{n\gamma}\varphi dt \qquad (4)$$

The number **N(Mo-99)d** of $^{99}_{42}Mo$ destroyed by radioactive decay at the same time, for a number of naturally occurring isotopes at the moment **t** explain in equation 5:

$$N(\text{Mo-99})_d = N(t)\,\lambda\,dt \qquad (5)$$

Where,

λ constant of the radioisotope decay of $^{99}_{42}Mo$

The radioisotopes decay rate in the target at time **t** is given by the following equation 6:

$$dN = N(\text{Mo-99})_f - N(\text{Mo-99})_d = (N_0\sigma_{n\gamma}\varphi - N(t)\,\lambda)\,dt \qquad (6)$$

In laying at **t = 0** s, **N = 0** find the fo

$$N(t) = \frac{N_0\varphi\sigma_{n\gamma}}{\lambda}(1 - e^{-\lambda t}) \qquad (7)$$

The induced activity which is expressed in Bq (counts per second) is determined by multiplying the above equation by **λ** take eq. 8:

$$A(t) = N(t)_{\lambda=} N_0 \varphi \sigma_{nr}(1 - e^{-\lambda t}) \quad 8$$

It is best to express this relation based on the mass of the sample, since it is the mass that is the parameter searched by **Neutron activation analysis** (NAA)[16-21].

At **t = 0s**, the number of nucleus in the targe $N_0 = \frac{N_a m(^{98}_{42}Mo)}{A}$ 10

Or,

$$m(^{98}_{42}Mo) = m(MoO_3) * \%(^{98}_{42}Mo) \quad 11$$

$\%(^{98}_{42}Mo) = 24{,}13\%$

So, the activity equal eq. 12: $A(t) = \frac{N_a m(MoO_3) * \%(^{98}_{42}Mo)}{A} \varphi \sigma_{nr}(1 - e^{-\lambda t})$ 12

A(t) – Activity of molybdenum-99 (Bq) in time (t)

N_a – Number Avogadro = 6.02 E23 (atom/mole)

σ_{nr} = 0.13 E-24, Radiative capture cross section of Mo-99 (cm^2/atom)

φ Maximum thermal neutron flux (n/cm^2/s)

A = 98, Molar mass of Mo-98 (g/mc

λ constant of the radioisotope decay $\frac{\ln(2)}{t_{\frac{1}{2}}(^{99}_{42}Mo)}$

λ = decay constant of Mo-99 (s^{-1})

2.2 Materials

Mass MoO_3 compacted into a capsule, figure 3, following dimension (2.54 cm radius and height of 15.24 cm), The target is irradiated in the central thimble (position A1) for access to the point of maximum neutron flux of the TRIGA Mark II research reactor [22-24] figures 3 and 4, where the measured thermal neutron flux at 250 KW are Φ_{th} = 3.10E+11 n/cm^2s, thermal neutron flux of 2.4E+13 n/cm^2s at 1.1 MW and 2 MW power are Φ_{th} = 7.0110[13] n/cm^2s[22-27]. Thermal study of MoO_3 target was performed using three thermal transfer modes: convection, conduction and radiation, using approximation one dimension, calculates the neutron was directed by a validated model. Numerical model used to determine temperature of each material to see that the temperature does not come to the melting point during MoO_3 to target irradiation in the central thimble (central thimble)**,** figure **1**.

For the numerical calculus of temperature [28] (Ozisik, 1985) explained an equation that gives heat in a cylindrical radius R geometry differential form is related to thermal conductivity λ and the generation rate q table 1.

$$\frac{1}{R}\frac{\partial}{\partial R}\left(R\frac{\partial T}{\partial R}\right) + q_1''' = 0 \qquad 0 < R < R_1 \quad 13$$

$$T(R) = -\frac{q_1'''}{4\zeta_1}R^2 + t_1 + \frac{q_1'''}{4\zeta_1}R^2 \qquad 0 < R < R_1 \quad 14$$

$$\frac{1}{R}\frac{\partial}{\partial R}\left(R\frac{\partial T}{\partial R}\right) + \frac{q_2'''}{\zeta_2} = 0 \qquad R_1 < R < R_2 \quad 15$$

$$T(R) = -\frac{q_2'''R^2}{4\zeta_2} + \frac{R_1^2}{2}\left(q_1''' - q_1'''\right)\ln(R) + t_2 + \frac{q_2'''R_2^2}{4\zeta_2} - \frac{R_1^2}{2}\left(q_2''' - q_1'''\right)\ln(R_2) \qquad R_1 < R < R_2 \quad 16$$

Since the surface heat flux from equation at R=R_1 serves as, equation 17:

$$q_2'' = \frac{q_1'''\pi R_1^2 + q_2'''\pi(R_2^2 - R_1^2)}{2\pi R_2} \quad 17$$

Then the linear transfer q_2' can be written as, equation 18

$$q_2' = q_1'''\pi R_1^2 + q_2'''\pi(R_2^2 - R_1^2) \quad 18$$

Or the linear heat transfer rate is constant throughout each layer of the heat flow path,

$$q_2' = q_3' = q_4' = q_5' \quad 19$$

On the other hand, the surface heat flux decreases along the radial direction of the heat flow path. The surface heat flux in each surface of each layer can be calculated as by equation 20,

$$q_2'' \times R_2 = q_3'' \times R_3 = q_4'' \times R_4 = q_5'' \times R_5 \quad 20$$

Using the linear transfer rate (q_2), the temperature drop across the Al canal can be derived as, equation 21:

$$T_4 - T_5 = \frac{q_2'}{2\pi\zeta_{Al(DCT)}}\ln\left(\frac{R_5}{R_4}\right) \quad 21$$

Table 1: The physical and chemical properties of the target and the components of the capsule

Material	Density (g/cc)	Thermal conductivity (W/m/K)	Emissivity (ε)	Melting point (°C)
Water	0.992	0.67	–	–
MoO_3	4.69	126	…….	795
Aluminum	2.73	230	0.18	658
Quartz	2.65	4	0.89	1,710

TRIGA MARK II

Training, Research, Isotopes, General Atomics (TRIGA GA's) reactor is the large amount generally used in the world. General Atomic has installed 66 TRIGA reactors at industrial laboratories, universities, and medical centers in 24 countries in what is in the Morocco. TRIGA reactors are used in various diverse applications; include production of radioisotopes for medicine and industry,

Figure 1 represents the design of the capsule used for the irradiation of MoO_3 (2.54 cm radius and height of 15.24 cm) [24], the target will be sealed in a quartz bulb as the first barrier to prevent an exothermic reaction between al and MoO_3, empty space containing the product gas during irradiation. To prevent the gap between the quartz and the aluminum layer the target will be surrounded by an aluminum foil at the end to avoid any mechanical risk and facilitate the transfer of heat.

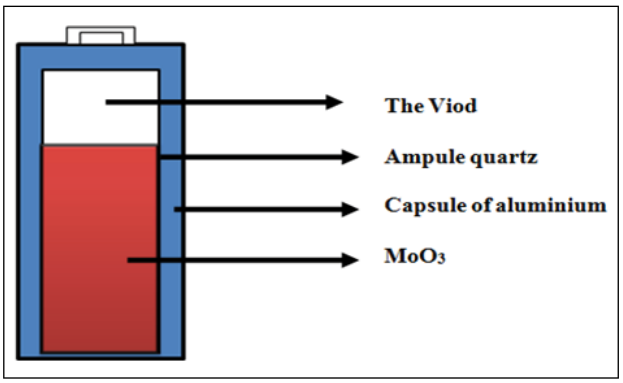

Figure 1: Irradiation capsule of MoO_3

Using the heat transfer equation based on the three heat transfer modes, the thermal study Molybdenum-99 production by irradiation of the target in the central thimble showed that the materials of the temperatures do not exceed their corresponding melting points, and as was confirmed by the Figure shown in Figure 2[22-24].

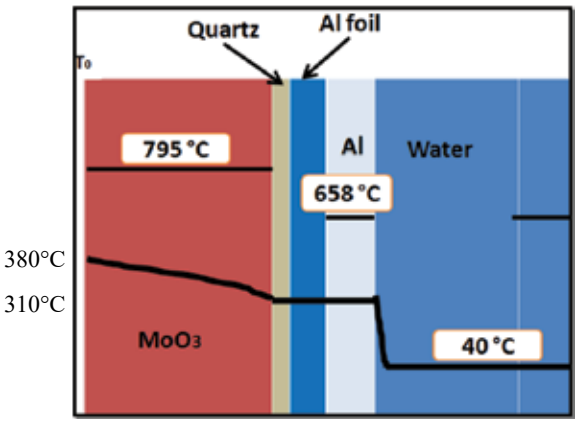

Figure 2: Cross-sectional view of the target vial components

Using conventional analytical methods, and heat transfer correlations, one-dimensional stationery the state, taking into account the transfer of heat, can be two important modes, conduction and thermal convection, radiation mode does not take place because all the material contains no empty. after all made calculates the temperature in the sample MoO_3 is 380°C lower than the melting point and then followed a decreasing in material (quartz, aluminum channel) until the water with a temperature 40°C, after all made calculated using the equation explain we find almost equal temperature values the same value tested experimentally[24,27] the results were schematized in the figure 4.

Result and discussion

Profile of activity in TRIGA MARK II

Table 2: Evolution of activity of Mo-99 in neutron flux (3,10E11 n/cm²s) TRIGA Mark II (250KW power)

Weight (MoO_3) (g)	Weight Mo-98 (g)	Thermal neutron flux n/cm²s	Cross section (barn)	irradiation time (j)	Activity of Mo-99 (Bq)	Activity of Mo-99 (Ci)
1,5	0,362	3,10E11	0,13	7	7,42656E7	0,002007
10	2,413				4,95104E8	0,0134
30	7,239				1,4853E9	0,0401
60	14,478				2,9706E9	0,0803
100	14,13				4,95104E9	0,1338
150	36,195				7,42656E9	0,2007

MoO_3 irradiation targets in the central thimble nuclear TRIGA MARK II reactor. With 250 kW power or neutron flux value is 3,10E11 n / cm²s gives the following results 0.002007, 0.0134, 0.0401, 0.0803, 0.1338 and 0.2007 Ci, respectively, for the amounts of MoO_3 1.5g, 30g, 100g and 150g, with 24.13% of Mo-98, Table 2, and Figure 3.

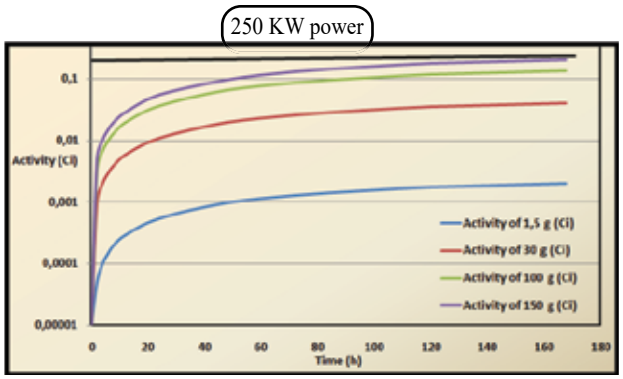

Figure 3: Evolution of activity evolution of Mo-99 in neutron flux (3,10E11 n/cm²s)
TRIGA Mark II (250KW power)

If the amount of MoO_3 more intense the activity of molybdenum-99 is more intense. Analysis of activity of shapes given during irradiation explains that in the early hours of irradiation a rapid increase, then a slight increase during the 5 days after finally stabilization of activity until the end of irradiation.

Table 3: Evolution of activity of Mo-99 in neutron flux (2.4E+13 n/cm²s) TRIGA Mark II (1.1 MW power)

Weight (MoO_3) (g)	Weight Mo-98 (g)	Thermal neutron flux n/cm²s	Cross section (barn)	irradiation time (j)	Activity of Mo-99 (Bq)	Activity of Mo-99 (Ci)
1,5	0,362	2.4E+13	0,13	7	5.7495E+9	0.15532453
10	2,413				3.83E+10	1.03549684
30	7,239				1.15E+11	3.10649052
60	14,478				2.30E+11	6.21298104
100	14,13				3.83E+11	10.3549684
150	36,195				5.75E+11	15.5324526

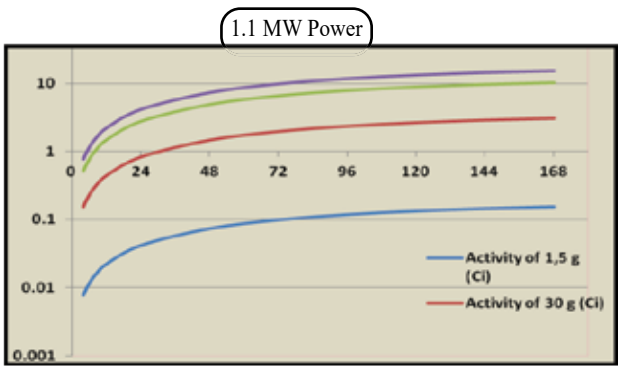

Figure 4: Evolution of activity of Mo-99 in neutron flux (2.4E13 n/cm²s) TRIGA Mark II (1.1 MW power)

MoO_3 irradiation targets in the central thimble. With power of 1.1 MW and neutron flux is 2.4E13 n/cm²s gives this results 0.155, 1.03, 3.10, 10.35 and 15.53 Ci, respectively, for 1.5g, 30g, 100g and 150g of MoO_3, Table 2, and Figure 4.

For one week of irradiation if the MoO_3 more important the activity of molybdenum-99 is more intense.

Table 4: Evolution of activity of Mo-99 in neutron flux (7,01E13 n/cm²s) TRIGA Mark II (2MW power)

Weight (MoO_3) (g)	Weight Mo-98 (g)	Thermal neutron flux n/cm²s	Cross section (barn)	irradiation time (j)	Activity of Mo-99 (Bq)	Activity of Mo-99 (Ci)
1,5	0,362	7,01E13	0,13	7	1,68E+10	0,4537
10	2,413				1,12E+11	3,0245
30	7,239				3,36E+11	9,0735
60	14,478				6,71E+11	18,147
100	14,13				1,12E+12	30,245
150	36,195				1,68E+12	45,368

After a week of irradiation target of MoO_3 in a neutron flux 7.01E13 n/cm²s for a power of 2MW, one sees clearly the increase in activity depending on time and

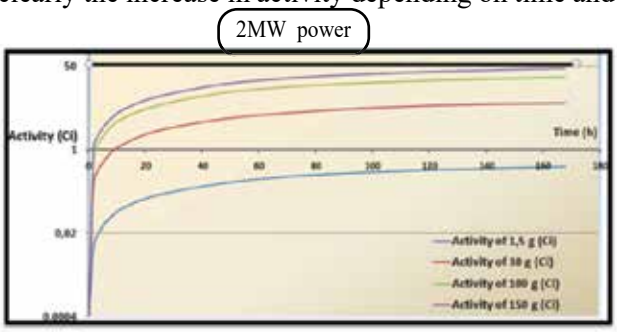

Figure 5: Evolution of activity of Mo-99 in neutron flux (7,01E13 n/cm²s) TRIGA Mark II (2MW power)

the amount of MoO_3 more intense than the activity and important, for example in amounts of 1.5g, 30g, 100g and 150g MoO_3 or Mo-98 is 24.13%, found a bitter radiation week 0.4537, 3.0245 activity, 18.147, 30.245 and 45.368 Ci respectively, Table 4, Figure 5. The analysis of the graph remarks that the activity increases rapidly in the first 12 hours of irradiation, followed by a slight increase during the first 5 days, eventually continuing stabilized within two days.

Conclusions

As the demand for medical isotopes increases in coming years and the age of foreign reactors necessitates their closure, new supply lines will become increasingly necessary. The domestic production of radioisotopes is an alluring prospect due to the accompanying independence and convenience of having a production facility; it was the main purpose of this study, is to produce of Molybdenum in order to produce the Technetium-99, it is playing a role in the field medical remarkable,

However, given that radioisotope demand is ever-increasing and foreign reactors are becoming a more unstable supply,

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