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Feasibility study for production of ^{99}Mo and ^{99m}Tc by the neutron activation of ^{98}Mo in the MNSR reactor

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Abstract: The calculated weekly specific activities of ^{99}Mo and ^{99m}Tc produced from the irradiation of the MoO_3 targets in the Miniature Neutron Source Reactor (MNSR) are presented in this paper. The productions of the ^{99}Mo and ^{99m}Tc were modelled and calculated through a set of differential equations using the Mathcad program. The resonance self-shielding factor (G_{res}) was calculated using the MATSSF and MCNP4C codes. The effects of the physical parameters such as the neutron flux and irradiation time on the weekly specific activities of ^{99}Mo and ^{99m}Tc have been analysed. It was found that the optimum irradiation scheme was achieved when the MNSR was operated for an extended period of 5 hours a day for 5 days a week at the neutron flux of 7.5×10^{11} n.cm^{-2}.s^{-1}. The weekly specific activities for the ^{99}Mo and ^{99m}Tc which can be produced in the MNSR, filling each of the five inner irradiation sites with 20 g of the MoO_3 targets and using the regular natural convection method to cool the reactor, were 7.20 and 5.05 mCi.g^{-1}, respectively.

Keywords: Mo production; neutron flux; irradiation time; specific activity; MNSR; miniature neutron source reactor.

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1 Introduction

The Miniature Neutron Source Reactor (MNSR) is a tank in pool type research reactor (CIAE, 1993). It is one of the low power research reactors which use highly enriched uranium as fuel, light water as moderator and beryllium as reflector. The beryllium reflector in the reactor can be divided into three sections: the side annulus surrounding the reactor core, the bottom plate and the top beryllium shims. The MNSR reactor has ten irradiation sites: five are called 'inner' and uniformly located inside the annulus beryllium reflector as can be seen in Figures 1 and 2. The other five are called 'outer' and surround the annulus reflector externally. The reactor nominal power is 30 kW, and the reactor nominal neutron flux is 1×10^{12} n.cm^{-2}.s^{-1} at the inner sites (CIAE, 1993). The reactor operating time is 2.5 hours per day, for 5 consecutive days a week at the nominal neutron flux if the heat generated in the core is removed through natural convection method and transferred to the pool through the vessel walls. The reactor operating time is 22 hours a day per week at the nominal neutron flux if the heat generated in the core is removed by operating the auxiliary cooling system of the reactor; the auxiliary cooling system decreases the consumption of the excess reactivity due to the negative reactivity temperature coefficient and therefor extends the reactor operation time.

Figure 1 Vertical cross section of the MNSR reactor

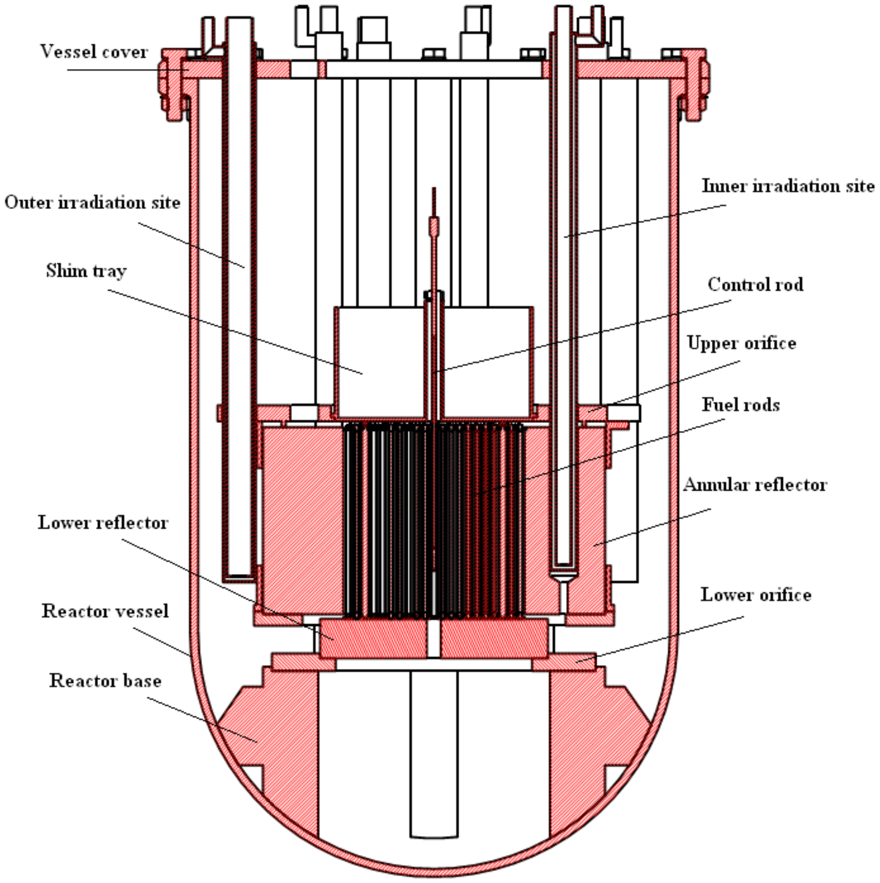
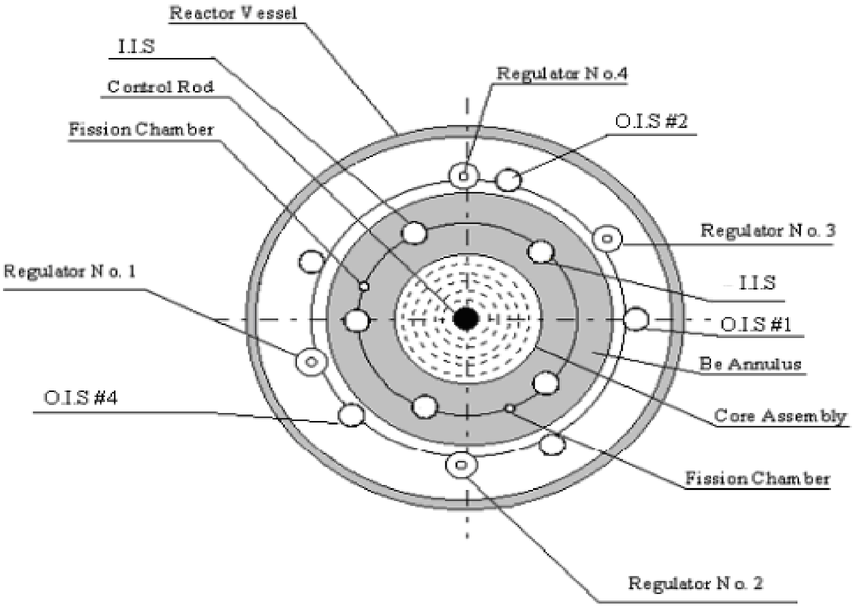


Figure 2 Horizontal cross section of the MNSR reactor: I.I.S: inner irradiation site, O.I.S: outer irradiation site



For many years, the ^{99}Mo ($T_{1/2}$ = 66.02 h) and its decay product the ^{99m}Tc ($T_{1/2}$ =6.006 h) have been produced and supplied to the medical community through the irradiation of uranium targets in the research reactors. The targets are processed after the irradiation using dedicated processing facilities. This has been a mature and effective supply process, which has been capable of supplying the world market and coping with the growth in the market for more than 30 years. The reliability of the industry has allowed widespread use of the ^{99m}Tc as a diagnostic imaging agent and around 30 million procedures are performed each year.

As an alternative, the ^{99}Mo can also be produced by the neutron capture reaction in the ^{98}Mo as in the following reaction: $^{98}\text{Mo}\ (n,\gamma)\ ^{99}\text{Mo}$. In this case, no high-level waste is formed. The thermal neutron cross section, σ_0, of the ^{98}Mo is 0.13 barn. The activation in the ^{98}Mo can be increased by the neutron absorption in the epithermal region. Therefore, the effective cross section, σ_{eff} , will be the sum of the thermal and epithermal neutron cross sections. The MNSR inner irradiation site has both thermal and epithermal neutrons. The ratio of the epithermal to thermal neutron fluxes $r = \frac{\varnothing_{epi}}{\varnothing_{the}}$ in the inner irradiation site was measured before (Khattab and Sulieman, 2009). This ratio was used to calculate the σ_{eff} for the ^{98}Mo in the MNSR and found to be 0.315 barn using the equation mentioned in the literature (Westcott, 1960; Martinho et al., 2003; Ryabchikov et al., 2004):

$$\sigma_{eff} = \sigma_0 + \frac{2}{\sqrt{\pi}} r\, G_{res} \left(I_\gamma - 0.45\sigma_0 \right) \tag{1}$$

According to the Westcott formalism (Westcott, 1960), the neutron activation, *A*, induced in a sample by radioactive neutron capture is given by:

$$A = N\varnothing_{the}\left[g\sigma_0 G_{th} + \frac{2}{\sqrt{\pi}} r\, G_{res}\left(I_\gamma - 0.45\sigma_0\right)\right]\left(1 - e^{-\lambda t}\right) \tag{2}$$

where: N is the number of atoms ${}^{A}X$ present in the sample, σ_0 is the thermal neutron cross-section for the (n,γ) reaction at $v_{0=}2200\,\mathrm{m\,s^{-1}}$, *g* is a parameter representing the deviation of the (n,γ) cross-section from the $1/v$ law in the thermal region, I_γ is the radioactive neutron capture resonance integral (6.7 barn), $\varnothing_{the}$ is the thermal neutron flux, $\varnothing_{epi}$ the epithermal neutron flux, λ is the decay constant of the nuclide ${}^{A+1}X$; *t* is the irradiation time, G_{th} is the thermal neutron self-shielding factor, and G_{res} is the resonance neutron self-shielding factor.

The density of the irradiated MoO_3 target was 4.69g.cm^{-3}. The Mo weight per cent in the irradiated MoO_3 target was 66.7%. The weight per cent of the ^{98}Mo in the Mo was 24.13%. Finally, the weight of the irradiated MoO_3 target used in the calculation was 20 g.

2 Methodology

The rate equations of the ^{99}Mo and ^{99m}Tc in the irradiated MoO_3 targets can be written using the following differential equations (JAEA-Research, 2010):

$$\frac{dN_1(t)}{dt} = -\sigma_{eff}\varnothing N_1(t) \tag{3}$$

$$\frac{dN_2(t)}{dt} = \sigma_{eff}\varnothing N_1(t) - \lambda_1 N_2(t) \tag{4}$$

$$\frac{dN_3(t)}{dt} = \lambda_1 W_1 N_2(t) - \lambda_2 N_3(t) \tag{5}$$

where:

$N_1(t)$, $N_2(t)$ and $N_3(t)$ are the number of the ^{98}Mo, ^{99}Mo and ^{99m}Tc atoms in the target at time *t* respectively. $\varnothing$ is the neutron flux. λ_1 and λ_2 are the decay constants of the ^{99}Mo and ^{99m}Tc respectively. W_1 is the branching fraction to decay to ^{99m}Tc. The σ_{eff} is the effective neutron cross section of the ^{98}Mo (n,γ) ^{99}Mo reaction.

To determinate the σ_{eff} value, the resonance self-shielding factor (G_{res}) was calculated using the MATSSF program (Trkov, 2009) and MCNP4C code (Briesmeister, 2000).

The ^{99}Mo and ^{99m}Tc weekly specific activities were modelled and calculated using the differential equations 3, 4 and 5 implementing the Mathcad program for two different cooling conditions of the reactor:

- Conducting the calculations for different neutron flux and irradiation time values for 5 and 6 continuous operation days per week. The cooling time of the irradiated sample was 24 hours after each irradiation time. The regular natural convection was used to cool off the reactor core in this case.
- The calculation for a continuous 22 hour reactor operation time per week at the neutron flux of 10×10^{11} n.cm^{-2}.s^{-1}. This long operation time can be achieved in the MNSR using the reactor auxiliary cooling system. The cooling time of the irradiated sample was 24 hours as before.

3 Results and discussions

Table 1 shows the resonance self-shielding factor for the irradiated MoO_3 target (60 mm in height and different diameters) using the MATSSF and MCNP4C codes. The maximum relative deference between the MATSSF program and MCNP4C code was 5% due to the simple method approximation used in the MATSSF code. Therefore, the MCNP4C code gave more accurate result of the self-shielding factor than the MATSSF code since there is no approximation in the MCNP4C code.

Table 1 Resonance self-shielding factors calculated by MATSSF and MCNP codes

Sample diameter (mm)	*MCNP reaction rate*	*G_{res} using MCNP4C*	*G_{res} using MATSSF*	*Relative difference, %*
0.000	1.210E-3	0.978	0.995	1.70
0.025	1.178E-3	0.976	0.990	1.41
0.050	1.114E-3	0.945	0.977	3.37
0.125	1.111E-3	0.919	0.957	4.19
0.250	1.068E-3	0.885	0.925	4.55

Table 2 shows weekly specific activities for the ^{99}Mo and ^{99m}Tc using only one inner irradiation site for target irradiation when the heat generated in the core was removed through natural convection method. The weekly specific activities of the ^{99}Mo and ^{99m}Tc for the thermal irradiation, $\sigma = \sigma_0$, and for the thermal and epithermal irradiation, $\sigma = \sigma_{eff}$, in one inner irradiation site were calculated for different neutron flux, irradiation time a day and operation time per week. The neutron fluxes were taken the following values: 10×10^{11}, 9×10^{11}, 8×10^{11}, 7.5×10^{11}, 7×10^{11}, 6×10^{11}, 5×10^{11} and 4×10^{11} n.cm^{-2}.s^{-1} .The irradiation times per day were: 2.5, 3, 4, 5, 6 and 7 hours a day. The operation times were: 5 and 6 days per week.

Table 2 shows as well the good agreements between our results (the weekly specific activities of the ^{99}Mo and ^{99m}Tc for the thermal irradiation only, $\sigma = \sigma_{th}$, and the results published in the literature (Osae, 1995). These good agreements verify the results of our calculations.

As can be observed from this table, the optimum irradiation scheme was achieved when the reactor was operated for an extended period of 5 hours a day for 5 days a week at the neutron flux of 7.5×10^{11} n.cm^{-2}.s^{-1}. The corresponding maximum weekly specific activities of the ^{99}Mo and ^{99m}Tc, for $\sigma = \sigma_{eff}$, were: 1.440 and 1.010 mCi.g^{-1}, respectively, since the inner irradiation site in the MNSR has both thermal and epithermal neutrons.

Table 2 The weekly specific activities of the ^{99}Mo and ^{99m}Tc using one inner irradiation site and different values of neutron flux and irradiation time, (Cooling by natural convection)

Neutron flux, ×10^{11} n.cm^{-2}.s^{-1}	*Irradiation time, hours per day*	*Operational time, days per week*	*The weekly specific activity, mCi.g^{-1}*					
			$\sigma = \sigma_{th}$				$\sigma = \sigma_{eff}$	
			^{99}Mo		*^{99m}Tc*		*^{99}Mo*	*^{99m}Tc*
			this work	*(OSAE, 1995)*	*this work*	*(OSAE, 1995)*		
10.0	2.5	6.0	0.451	0.483	0.314	0.340	1.091	0.760
9.0	3.0	6.0	0.482	0.520	0.338	0.426	1.165	0.817
8.0	4.0	5.0	0.518	0.564	0.358	0.395	1.254	0.867
7.5	5.0	5.0	0.595	0.657	0.418	0.469	1.440	1.010
7.0	5.0	5.0	0.556	0.613	0.390	0.438	1.344	0.943
6.0	6.0	5.0	0.560	0.628	0.398	0.455	1.355	0.963
5.0	6.0	6.0	0.500	0.569	0.363	0.424	1.209	0.879
4.0	7.0	6.0	0.456	0.528	0.335	0.398	1.104	0.811

Figures 3 and 4 show the weekly specific activities for the ^{99}Mo and ^{99m}Tc for one inner irradiation site for different values of neutron flux and irradiation time a day for 5 continuous operation days per week. The cooling time of the irradiated sample was 24 hours after each irradiation time. The heat generated in the core was removed through natural convection and was transferred to the pool through the vessel walls.

Figure 3 The weekly specific activity of the ^{99}Mo using one inner irradiation site and different values of neutron flux and irradiation time, (cooling by natural convection)

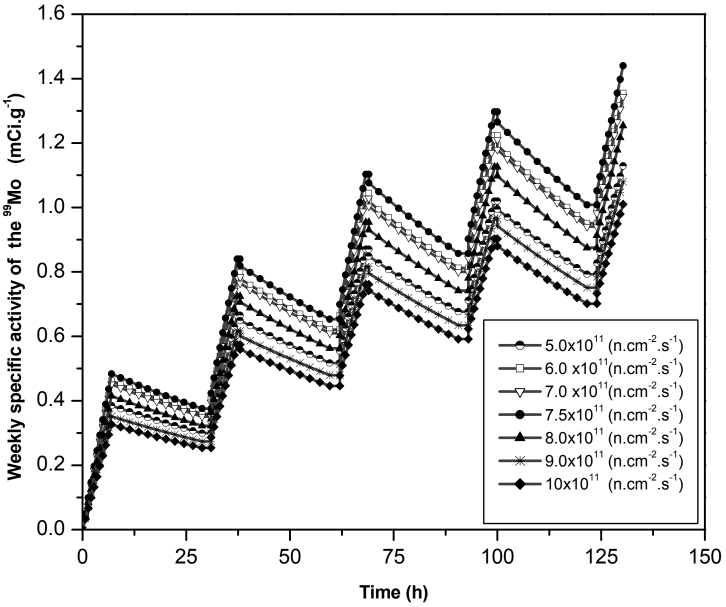


Figure 4 The weekly specific activity of the ^{99m}Tc using one inner irradiation site and different values of neutron flux and irradiation time, (cooling by natural convection)

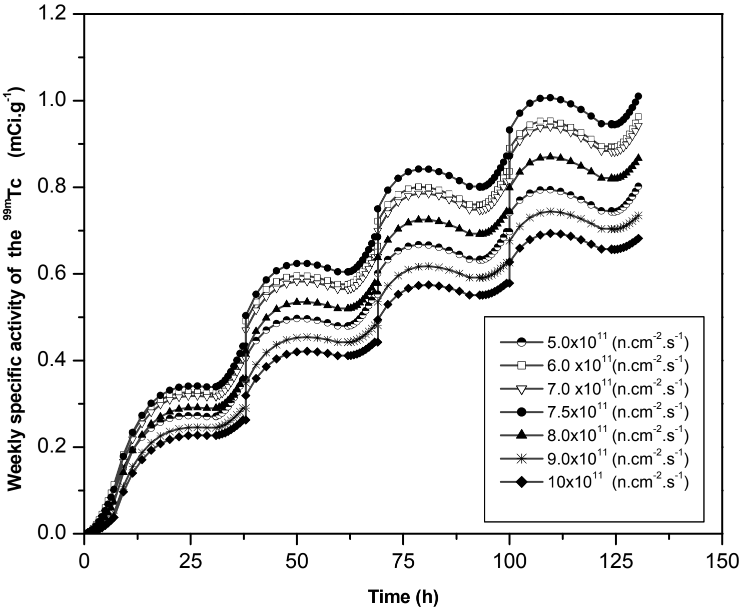


Table 3 shows the weekly specific activities of the ^{99}Mo and ^{99m}Tc produced in 5 and 6 days per week filling the five inner irradiation sites with the MoO_3 targets. As can be seen from this table, the maximum weekly specific activities of the ^{99}Mo and ^{99m}Tc for the five inner irradiation sites were 7.200 and 5.050 mCi.g^{-1}, respectively.

Table 3 The weekly specific activities of the ^{99}Mo and ^{99m}Tc using five inner irradiation sites and different values of neutron flux and irradiation time, (cooling by natural convection)

Neutron flux, ×10^{11} n.cm^{-2}. S^{-1}	*Irradiation time, hours per day*	*Operational time, days per week*	*The weekly specific activity, mCi.g^{-1}*					
			$\sigma = \sigma_{th}$				$\sigma = \sigma_{eff}$	
			^{99}Mo		*^{99m}Tc*		*^{99}Mo*	*^{99m}Tc*
			this work	*(OSAE, 1995)*	*this work*	*(OSAE, 1995)*		
10.0	2.5	6.0	2.255	2.415	1.570	1.700	5.455	3.800
9.0	3.0	6.0	2.410	2.600	1.690	2.130	5.825	4.085
8.0	4.0	5.0	2.590	2.820	1.790	1.795	6.270	4.335
7.5	5.0	5.0	2.975	3.285	2.090	2.345	7.200	5.050
7.0	5.0	5.0	2.780	3.065	1.950	2.190	6.720	4.715
6.0	6.0	5.0	2.800	3.140	1.975	2.275	6.775	4.815
5.0	6.0	6.0	2.500	2.845	1.815	2.120	6.045	4.395
4.0	7.0	6.0	2.280	2.640	1.675	1.990	5.520	4.055

Figure 5 shows the weekly specific activities of the ^{99}Mo and ^{99m}Tc using one inner irradiation site with the MoO_3 target for the optimum irradiation scheme for 5 hours a day for 5 days a week at the neutron flux of 7.5×10^{11} n.cm^{-2}.s^{-1}. The heat generated in the core was removed through natural convection method.

Figure 5 The weekly specific activities of the ^{99}Mo and ^{99m}Tc using one inner irradiation site at the optimum irradiation scheme, (cooling by natural convection)

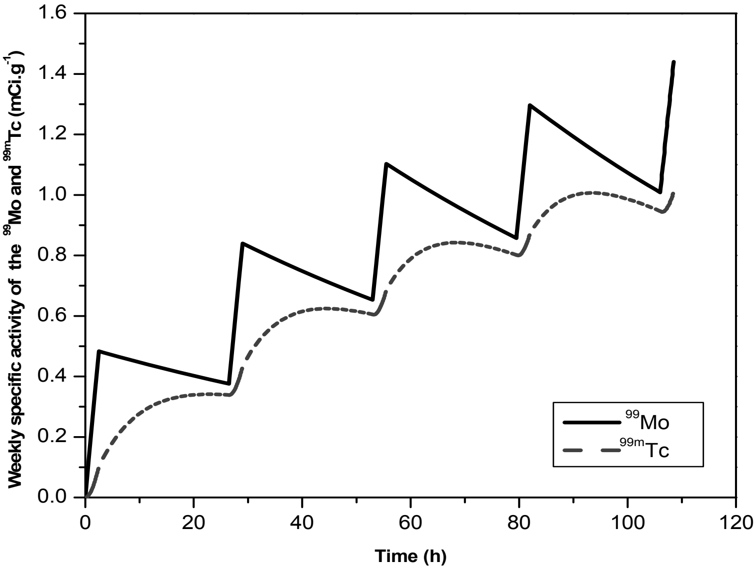


Figure 6 The weekly specific activities of the ^{99}Mo and ^{99m}Tc using five inner irradiation site at the optimum irradiation scheme, (cooling by natural convection)

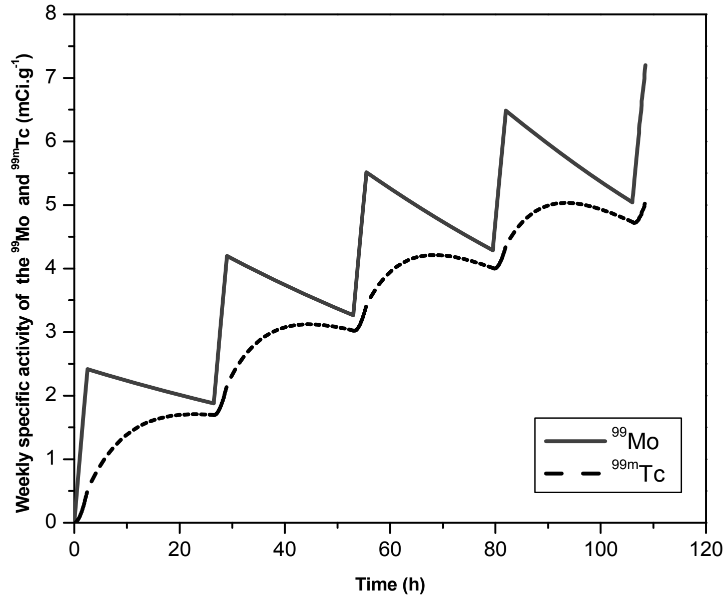


Figure 6 shows the weekly specific activities of the ^{99}Mo and ^{99m}Tc filling the five inner irradiation sites with the MoO_3 targets for the optimum irradiation scheme for 5 hours a day for 5 days a week at the neutron flux of 7.5×10^{11} n.cm^{-2}.s^{-1}. The heat generated in the core was removed through natural convection method.

Tables 4 and 5 show the changes in the specific activities of the ^{99}Mo and ^{99m}Tc filling the five inner irradiation sites with the MoO_3 targets at the optimum irradiation scheme. Table 4 shows the step change in the ^{99}Mo specific activities started from zero to 7.200 mCi.g^{-1}after 5 days of irradiation. Table 5 shows the step change in the ^{99m}Tc specific activities started from zero to 5.050 mCi.g^{-1}after 5 days of irradiation.

Table 4 The step specific activities of the ^{99}Mo using five inner irradiation site at the optimum irradiation scheme, (cooling by natural convection)

Irradiation time per day (5 hours)		
Day	*Specific activity at the start of the irradiation, mCi.g^{-1}*	*Specific activity at the end of the irradiation, mCi.g^{-1}*
1	0.000	2.417
2	1.879	4.2
3	3.265	5.515
4	4.287	6.485
5	5.041	7.200

Table 5 The step specific activities of the ^{99m}Tc using five inner irradiation site at the optimum irradiation scheme, (cooling by natural convection)

Irradiation time per day (5 hours)		
Day	*Specific activity at the start of the irradiation, mCi.g^{-1}*	*Specific activity at the end of the irradiation, mCi.g^{-1}*
1	0.000	0.512
2	1.695	2.164
3	3.025	3.428
4	4.009	4.362
5	4.735	5.050

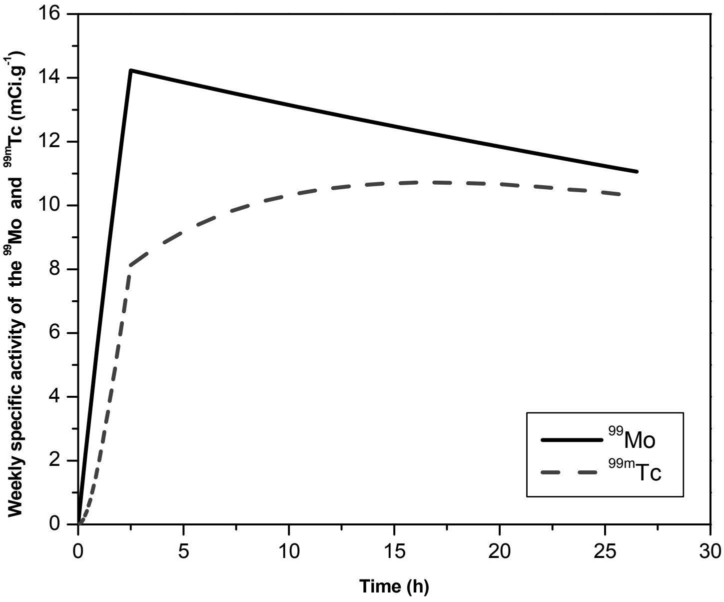
Table 6 shows weekly specific activities of the ^{99}Mo and ^{99m}Tc for thermal and epithermal irradiation filling the five inner irradiation sites with the MoO_3 targets. The reactor was operated for a continuous 22 hours per week at 10×10^{11} n.cm^{-2}.s^{-1} neutron flux. The cooling time of the irradiated sample was 24 hours. The heat generated in the core was removed by operating auxiliary cooling system in the reactor. As can be seen from this table, the weekly specific activities for the ^{99}Mo and ^{99m}Tc for the thermal irradiation were 4.176 and 3.888 mCi.g^{-1}, respectively. The weekly specific activities for the ^{99}Mo and ^{99m}Tc for the thermal and epithermal irradiations were 11.060 and 10.300 mCi.g^{-1}, respectively.

Table 6 The weekly specific activities for the ^{99}Mo and ^{99m}Tc using five inner irradiation sites for 22 continuous hours a day per week, (cooling by auxiliary cooling system)

Neutron flux, 10^{11} n.cm^{-2}.s^{-1}	*Irradiation time, hours per day*	*Operational time, days per week*	*The weekly specific activity, mCi.g^{-1}*			
			$\sigma = \sigma_{th}$		σ_{eff}	
			^{99}Mo	^{99m}Tc	^{99}Mo	^{99m}Tc
10.0	22.0	1.0	4.176	3.888	11.060	10.300

Figure 7 shows the weekly specific activities of the ^{99}Mo and ^{99m}Tc for filling the five inner irradiation sites with the MoO$_3$ targets for a continuous 22 hours a day per week at 10×10^{11} n.cm^{-2}.s^{-1} neutron flux. The cooling time of the irradiated sample after irradiation time was 24 hours. The heat generated in the core was removed by operating auxiliary cooling system in the reactor.

Figure 7 The weekly specific activities of the ^{99}Mo and ^{99m}Tc using five inner irradiation sites for 22 continuous hours a day, (cooling by auxiliary cooling system)



4 Conclusion

To calculate the σ_{eff} for the MoO$_3$, the G_{res} was estimated using the MATSSF and MCNP4C codes. The MCNP4C code gave more accurate result than the MATSSF code. It is observed from the calculated results that the optimum irradiation scheme was achieved when the MNSR was operated, with natural convection cooling method, for an extended period of 5 hours a day for 5 days a week at the neutron flux of 7.5×10^{11} n.cm^{-2}.s^{-1}. The weekly specific activities for the ^{99}Mo and ^{99m}Tc filling the five inner irradiation sites with MoO$_3$ targets were 7.200 and 5.050 mCi.g^{-1}, respectively. The MNSR can be operated as well for a continuous 22 hours a day per week at the neutron flux of 10×10^{11} n.cm^{-2}.s^{-1} using the auxiliary cooling system in the reactor. The weekly specific activities for the ^{99}Mo and ^{99m}Tc filling the five inner irradiation sites with MoO_3 targets in this case were 11.060 and 10.300 mCi.g^{-1}, respectively.

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