Investigation on the feasibility of $^{99m}$Tc production from $^{100}$Mo(p,2n) reaction at existing Medical Cyclotron Facilities


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Introduction

$^{99m}$Tc (T$_{1/2}$= 6 h) in various chemical forms is the most commonly used radiotracer in nuclear medicine, covering more than 70% of all nuclear medical diagnosis due to its multiple advantages. It decays with a suitable half-life and emits a 140.5 keV γ-ray, which is ideally suited for imaging via single photon emission computed tomography (SPECT) and causes minimal radiation dose to the patient. It is popular due to its convenient availability through a $^{99}$Mo/$^{99m}$Tc generator system and its capability to form a wide variety of complexes with organic ligands, which can be obtained instantaneously as kit formulations [1]. The parent radionuclide $^{99}$Mo (T$_{1/2}$=66 h) is generally produced via neutron-induced fission of highly enriched $^{235}$U in a nuclear reactor. Two reactors, NRU Canada and HFR Netherlands, which together meet about 70% of the world demand of $^{99}$Mo. Owing to various problems from these suppliers, an alternative method for the production of $^{99}$Mo and $^{99m}$Tc is suggested using the proton induced reaction of highly enriched $^{100}$Mo target. Further, there are large discrepancies in published reaction cross section data for $^{100}$Mo(p,2n)$^{99m}$Tc reaction[2].

Experimental details:

In the present work, generation of $^{99}$Mo and $^{99m}$Tc were carried out via 24 hours irradiation of the enriched $^{100}$Mo targets at an incident proton energy of 17.6 MeV using the stacked foil activation technique at the 14 MV Pelletron, TIFR, Mumbai. Stacks of highly enriched foils of molybdenum $^{100}$Mo of thickness 1mg/cm$^2$ alternating with thin foils of high purity aluminum of 5 mg/cm$^2$ were used. The aluminum foils degrade the proton energy from 17.6 to 17.2 MeV. A copper foil of 2 mg/cm$^2$ thickness was used as beam current and energy monitor. The induced activity was measured by using energy and efficiency calibrated HPGe detector. The first measurements of the samples was started after cooling a few hours after end of bombardment (EOB) and measurements continued periodically. The $^{62}$Cu(p,xn)$^{65}$Zn reaction cross section data recommended by IAEA was used as flux monitor.

Data analysis

There are competing reactions, which occur during the proton irradiation of $^{100}$Mo producing $^{99}$Mo from the (p,pn), (p,d) reactions and $^{99m}$Tc from the (p,2n) reaction, respectively. The $^{99m}$Tc is identified by its 140 KeV γ-line, however the total measured net peak area, $C_{Net}^{140}$, is a combination of counts from both $^{99}$Mo and $^{99m}$Tc. $C_{Net}^{140} = C_{Mo}^{140} + C_{Tc}^{140}$, where $C_{Mo}^{140}$ includes the direct $^{99m}$Tc and indirect $^{99}$Mo to $^{99m}$Tc decay contributions during irradiation and post irradiation [3,4]. The contribution of $^{99}$Mo decay to 140 KeV line intensity is estimated from...
measurement of the intensity of 181 KeV $^{99}$Mo line.

$$C_{140}^{\text{Mo}} = [A_{\text{MO}}^{\text{EOB}} e^{-\lambda_1 t_\epsilon} (1 - e^{-\lambda_2 t_\epsilon})] *$$

$$e_{140}^{\text{MO}} \frac{1^{\text{MO}}_{\text{wTc}} t_L}{t_R} / \lambda_2 t_R$$

$$[A_{\text{MO}}^{\text{SOC}}]_{\text{Total}} = (C_{\text{Total}}^{\text{MO}} - C_{\text{MO}}^{140}) *$$

$$\lambda_2 t_R (1 - e^{-\lambda_1 t_\epsilon}) e_{140}^{\text{MO}} l_{\text{wTc}}^{\text{MO}} t_L$$

$$[A_{\text{MO}}^{\text{SOC}}]_{\text{Total}} = [A_{\text{MO}}^{\text{SOC}}]_{\text{Indir}} + [A_{\text{MO}}^{\text{SOC}}]_{\text{Dir}}$$

$$[A_{\text{MO}}^{\text{SOC}}]_{\text{Indir}} = (\lambda_2 / \lambda_2 - \lambda_1) f A_{\text{MO}}^{\text{EOB}} (e^{-\lambda_1 t_\epsilon} - e^{-\lambda_2 t_\epsilon}) + [A_{\text{MO}}^{\text{SOC}}]_{\text{Indir}} e^{-\lambda_2 t_\epsilon}$$

$$[A_{\text{MO}}^{\text{SOC}}]_{\text{Indir}} = f [A_{\text{MO}}^{\text{EOB}} (1 - e^{-\lambda_1 t_\epsilon})] * [1 - (\lambda_2 / \lambda_2 - \lambda_1) e^{-\lambda_1 t_\epsilon} + (\lambda_2 / \lambda_2 - \lambda_1) e^{-\lambda_2 t_\epsilon}]$$

Where, $f=0.87$, $A_{\text{MO}}^{\text{EOB}}$ is the EOB activity of $^{99}$Mo calculated from the 181 KeV line of $^{99}$Mo, $\lambda_1$ and $\lambda_2$ are the decay constants of $^{99}$Mo and $^{99m}$Tc, respectively. $t_\epsilon=$ elapsed time between EOB and start of counts (SOC), $t_R=$ real time of detector, $t_L=$ live time of detector, $t_i=$ irradiation time, $e_{140}=$ detector efficiency at 140 keV, $l_{\text{wTc}}^{\text{MO}}=\gamma$-ray intensity at 140 keV for $^{99}$Mo, $l_{\text{wTc}}^{99m}=\gamma$-ray intensity at 140 keV for $^{99m}$Tc. In equation 4, the first term corresponds to $^{99}$Mo $\rightarrow^{99m}$Tc production post EOB, and the second term corresponds to $^{99}$Mo $\rightarrow^{99m}$Tc production during irradiation.

Fig 1 shows the relative contribution to the total peak area of the 140 keV $\gamma$-line as a function of cooling time at 17.48 MeV, with symbols from present work and lines from Takacs et al.[4]. As can be seen from the Fig 1, the contribution from the direct produced $^{99m}$Tc drops down steadily, and after a long cooling time, the measured counts will no longer contain information about the cross section of the $^{100}$Mo(p,2n)$^{99m}$Tc reaction. Preliminary calculations of cross sections of the $^{100}$Mo(p,2n)$^{99m}$Tc and $^{100}$Mo(p,x)$^{99}$Mo reactions are 320±20 mb and 79±6 mb at 17.6 MeV respectively.

**Conclusion**

Experimental measurements of the cross sections of $^{100}$Mo(p,x)$^{99}$Mo and $^{100}$Mo(p,2n)$^{99m}$Tc reactions have been carried out. Typically the medical cyclotrons have proton beam energies in the range 16 to 18 MeV and the Cyclotron at Radiatıon Medicine Centre, Mumbai has an $E_p=16.5$ MeV. The $^{100}$Mo(p,2n)$^{99m}$Tc reaction has appreciable cross section at these energies and $^{99m}$Tc can be produced in quantities sufficient for local use.

**References**