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# Investigations of Production Cross Sections and Induced Activity of $^{94,95,96,98}$Tc Produced in Mo Target by 30 MeV Proton Beam

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Theoretical models often fill significant gap exists in measured data and radioisotopes produced may be used in research, medical and safely disposal of radioactive waste. The excitation functions of $^{94}$Mo(p,n)$^{94}$Tc, $^{95}$Mo(p,n)$^{95}$Tc, $^{96}$Mo(p,n)$^{96}$Tc and $^{98}$Mo(p,n)$^{98}$Tc reactions were investigated using nuclear reaction model codes TALYS-1.95 and EMPIRE-3.2.3 from reaction threshold to 30 MeV. These codes incorporate three major nuclear reaction mechanisms including direct, compound and pre-compound nuclear reactions. Theoretical results obtained from TALYS-1.95 and EMPIRE-3.2.3 codes were compared with existing literature data and found to be in reasonable agreement. Additionally, induced radioactivity estimation is also conducted for a thick Mo target due to the primary interaction of 1μA, 30 MeV proton beam.

**Keywords:** Induced radioactivity; Excitation functions; Pre-compound; TALYS; EMPIRE; Generalized super fluid model

## 1 Introduction

Induced radioactivity is a significant phenomenon observed in certain materials after irradiation of external sources of radiation. This intriguing concept lies at the heart of nuclear physics and has far-reaching implications for different applications and basic science domains. The radioactive isotopes can be produced through bombardment of stable atoms of materials with high-energy particles or photons. This process leads to the emission of ionizing radiation, which can persist for varied durations depending on the half-life of the radioactive isotopes formed[1-5].

The study of induced activity is important to analyze and understand the behavior of nuclear materials. It also plays a crucial role in assessing safety and designing particle accelerator facilities, nuclear reactors and similar facilities that utilize release of radiation. Moreover, induced activity has become vital for medical applications, e.g., radioisotope production for diagnostic imaging and cancer treatment, making it an essential area of research in nuclear medicine[6-9].

Technetium radioisotopes produced in the current study have a wide range of applications in various fields due to their unique nuclear properties and relatively short half-lives. Some technetium radioisotopes are valuable tools in scientific research and used as tracers in various studies to understand biological, chemical, and physical processes. For $^{94,96,98}$Tc (in their ground states), the energies of gamma rays (with significant intensity) are quite high and are not very suitable for diagnosis compared to standard diagnostic isotope $^{99m}$Tc. These isotopes may be more suited to other non-diagnostic applications where higher energy emissions are less of a concern. $^{95}$Tc (20 hr), with a comparatively longer half-life, facilitates the tracking of slower processes such as studies involving proteins, metabolic pathways for heart and brain, anti-bodies, etc. Additionally, radioisotopes like $^{94m}$Tc and $^{96}$Tc (102.72hr) find utility as proton beam monitor. It's important to note that the production, handling, and use of technetium radioisotopes are regulated due to their radioactive nature. In order to work with these isotopes, the proper safety protocols and radiation protection measures need to be ensured[1-2, 9-11].

The cross section data for the relevant nuclear reactions are crucial for determination of the induced radioactivity in the system, arising from the interaction of the proton beam and secondary neutrons in the target. In the IAEA-EXFOR[12] database, such experimental data are not always available for the Tc isotopes studied in this work for certain energy range. Discrepancies also exist among different measurements for several cross-sectional data. So, for complete knowledge of the nuclear reactions, the

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present study carried in this energy range may be helpful to understand the dependency of activation cross-sections on energy in detail. In the current study, formation cross-sections of $^{94}$Mo(p,n)$^{94}$Tc, $^{95}$Mo(p,n)$^{95}$Tc, $^{96}$Mo(p,n)$^{96}$Tc and $^{98}$Mo(p,n)$^{98}$Tc as well as induced radioactivity in a thick Mo target were computed using the nuclear reaction model codes TALYS-1.95 and EMPIRE-3.2.3. Proton energy of 30 MeV and current of 1 μA were used for the purpose. The results obtained for reaction cross-section from these codes were compared with the available experimental data.

## 2 Method of calculation

### 2.1 EMPIRE-3.2.3

EMPIRE-3.2.3[13] is designed to predict the outcomes of nuclear reactions involving neutrons, protons, alpha particles, and heavy ions. EMPIRE-3.2.3 is a nuclear reaction model code that serves as a fundamental tool in the field of nuclear physics research and applications. As an extension of the original EMPIRE code, version 3.2.3 offers significant improvements and enhanced capabilities for simulating and analyzing nuclear reactions induced by various particles. Covering an energy range from a keV to several GeV, EMPIRE can handle both low energy nuclear astrophysics reactions and high energy nuclear reactions making it a flexible tool, for investigating phenomena across different energy levels.

In the determination of induced activity and total yield of the product isotope comprehensive analysis of nuclear reactions is crucial, which is facilitated by the use of computed excitation functions. Utilizing the computer package SRIM[14], attenuation of the projectile energy and flux at various depths within the thick target has been computed. The total yield and activity of a radioisotope were derived through a weighted summation of the yields computed from the production cross-sections at projectile energies. The level densities of Generalized Super Fluid Model (GSM) (LEVDEN=1) were used for the compound nuclear reaction in addition to various Pre-equilibrium models like PCROSS, Hybrid Monte Carlo Simulation (HMS) and Multi Step Compound (MSC) as follows: (i) EMPIRE1-PCROSS (ii) EMPIRE2-PCROSS+HMS(iii) EMPIRE3–MSC

### 2.2 TALYS-1.95

TALYS-1.95[15] is a sophisticated, powerful and widely used nuclear reaction model code that plays a vital role in nuclear physics research and applications. TALYS-1.95 is used for simulating and evaluating nuclear reactions induced by various particles - neutrons, protons, alpha particles, and other light ions. TALYS-1.95 enables the investigation of various reaction mechanisms, including the Hauser-Feshbach statistical model, pre-equilibrium and direct reactions, and fission and fusion reactions. Direct reactions are determined within the framework of giant resonances. The estimation of pre-equilibrium (PEQ) particle emission dynamics relies on a two-component exciton model. The angular distribution of emitted particles is analyzed through the application of Kalbach systematic. Determination of optical model parameters, crucial for describing particle-nucleus interactions, is meticulously conducted using advanced computational tools such as the ECIS-06 code. Equilibrium (EQ) and multiple PEQ emissions of ejectiles are also accounted for. The code offers various level density models, including constant temperature model, Fermi gas model, back-shifted Fermi gas model, Generalized Superfluid model (GSM) and microscopic level densities. The present calculations were carried out by using the Constant temperature Fermi gas model.

### 2.3 Induced activity calculation

Induced activity is typically computed for a thick target where protons lose energy while traversing through it. The proton's energy loss property is quantified as the stopping power (S). To calculate the total radionuclide production from a thick target for continuously degrading proton energies inside the target, the target thickness perpendicular to the incident beam is divided into thin slices such that the proton loses energy of 2 MeV in each slice. This may be considered as a reasonably approximate discretization of the continuous energy degradation process. At each slice the reaction yield is calculated for the proton energy corresponding to that particular slice. Finally, the total yield is calculated by summing the contributions from all slices.

The total activity within the thick target is the sum of induced activity across all these energy ranges. Now, let's assume that a target 'X' is irradiated for a small time 'dt' resulting in the production of a radioisotope 'Y'.

Thus we get

$$dN_Y^0 = N_X \sigma \Phi dt \qquad \ldots (1)$$

where $N_X$,$N_Y$ is numbers of atoms of X and Y respectively. σ is cross section for production of Y and Φ represent fluence rate. After a time 't' the number of atoms Y remaining is

$$dN_Y^t = dN_Y^0 \exp(-\lambda_y t) = N_X \sigma \Phi dt \exp(-\lambda_y t) \quad \text{... (2)}$$

Here $\lambda_y$ is decay constant of residual nucleus. Now if irradiation is carried out for a period t, the number of atoms of Y at time

$$T \text{ is} \int_0^t dN_Y^t = \int_0^t N_X \sigma \Phi \exp(-\lambda_y t) dt \quad \text{... (3)}$$

or

$$N_Y = \frac{N_X \sigma \Phi [1 - e^{-\lambda_y t}]}{\lambda} \quad \text{... (4)}$$

Activity due to Y after time t

$$A = N_X \sigma \Phi [1 - e^{-\lambda_y t}] \quad \text{... (5)}$$

where activity of Y is $A = N_Y \lambda$. $N_X$-total number of target atoms available for reaction when proton loses an energy $\Delta E_P$ MeV. If S is given in MeV/mg/cm$^2$, then

$$N_X = \left(\frac{N_A}{A_T}\right) \times \left(\frac{\Delta E_P}{S}\right) \times 10^{-3} \frac{\text{atoms}}{\text{cm}^2} \quad \text{... (6)}$$

$N_A$ is Avogadro number and $A_T$ atomic mass of target. Now activity with a beam current of one micro ampere becomes

$$A = \left(\frac{6.023}{A_T}\right) \times \left(\frac{\Delta E_P}{S}\right) \times \left(\frac{\sigma}{1.602}\right) \times 10^6 (1 - e^{-\lambda_Y t}) \text{Bq} \quad \text{... (7)}$$

The energy interval $\Delta E_P$ is taken equal to 2 MeV out of total beam energy 30 MeV for present calculations[5,16].

## 3 Results and Discussion

Theoretical models often fill significant gap exists in measured data and radioisotopes produced may be used in safely disposal of radioactive waste. The excitation functions and build-up of induced activity for stable isotopes of Mo have been investigated for product isotopes formed by the interaction of 1 μA proton beam with target for irradiation period upto 960 hours. The reaction model code TALYS-1.95 and EMPIRE-3.2.3 were employed to compute the total production cross-sections. In such cases, the production of the isotope is considered only in its ground state. The induced activity, half-life and radionuclide formed at specific irradiation time are shown in Table 1.

Table 1 — Induced Activity Values.

| Reaction | Half-life (hrs.) | Irradiation time (hrs.) | Induced Activity(Bq) | |
|---|---|---|---|---|
| | | | TALYS-1.95 | EMPIRE-3.2.3 |
| $^{94}$Mo(p,n)$^{94}$Tc | 4.88 | 120 | 11500 | 12500 |
| $^{95}$Mo(p,n)$^{95}$Tc | 20.00 | 240 | 9750 | 9730 |
| $^{96}$Mo(p,n)$^{96}$Tc | 102.72 | 960 | 9520 | 9610 |
| $^{98}$Mo(p,n)$^{98}$Tc | 36792x10$^6$ | 960 | 1.46x10$^{-4}$ | 1.94 x10$^{-4}$ |

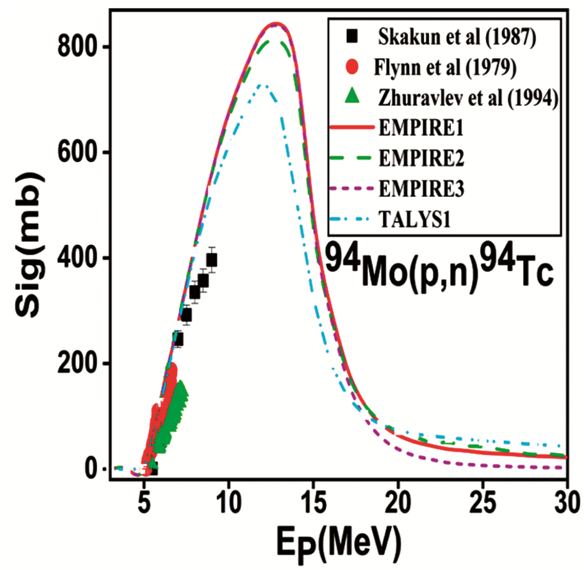


Fig. 1 — Excitation function of $^{94}$Mo(p,n)$^{94}$Tc with proton energy up to 30 MeV.

The excitation function for the formation of $^{94}$Tc is presented in Fig. 1 along with the available literature data which is only upto 9 MeV[17-19]. The computed results show a maximum cross-section of 824 mb at incident energy about 13 MeV. The results obtained using EMPIRE and TALYS nuclear codes agree with the data of Skakun *et al.*[17] and Flynn *et al.*[18] from threshold to 9 MeV within experimental errors while over predict Zhuravlev[19] data. The compound evaporation yield works in low energy range and pre-equilibrium starts around 16 MeV.

The calculated excitation functions and experimental data for the $^{95}$Mo(p,n)$^{95}$Tc nuclear reaction are plotted in Fig. 2. The reaction channel opens at around 4 MeV. The experimental data measured by Skakun *et al.*[17], Trufanov[20] and Flynn *et al.*[18] in entire energy range exhibit the same trend as the excitation functions obtained from the codes EMPIRE and TALYS. The computed data by EMPIRE and TALYS codes overpredict the data reported by Izumo *et al.*[21] up to 12 MeV. This part is contributed from compound nuclear emissions which is strongly dependent on the level density parameter. The over prediction may be attributed to the choice of level density considered in the calculation. EMPIRE3 using MSC options agrees well in energy range 16-28 MeV. EMPIRE 1,2 using Generalized super fluid model agree with Levkovskii[22] in energy 7.7 MeV to 18.3 MeV except peak value. The compound nucleus process is dominated in low energy portion of excitation functions while the pre-equilibrium contribution arises at only about 18–30 MeV.

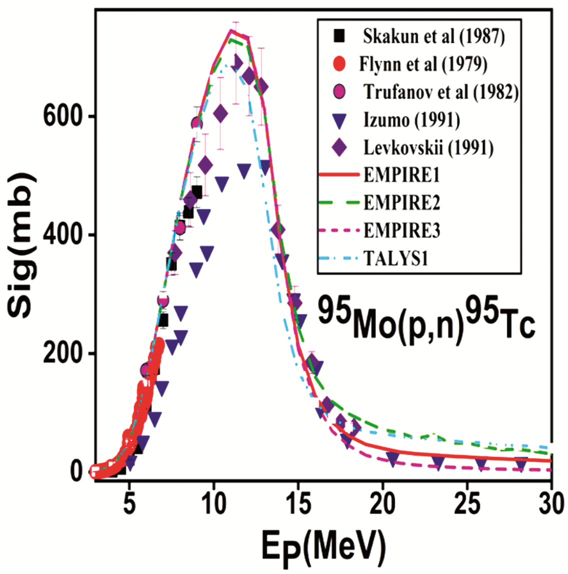


Fig. 2 — Excitation function of $^{95}$Mo(p,n)$^{95}$Tc with proton energy up to 30 MeV.

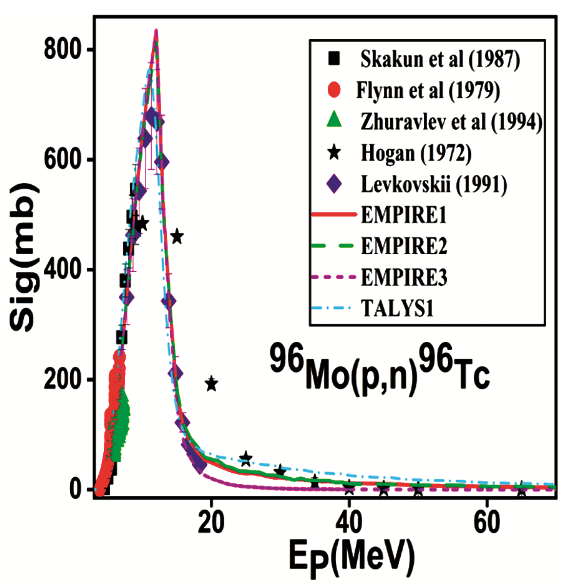


Fig. 3 — Excitation function of $^{96}$Mo(p,n)$^{96}$Tc with proton energy up to 70 MeV.

The excitation functions for $^{96}$Mo(p,n)$^{96}$ Tc reaction are shown in Fig. 3 using EMPIRE and TALYS codes. The reaction threshold is 4 MeV and measured data[17-19, 22-23] are available up to 65 MeV. All computed data reproduce measured data very well. EMPIRE & TALYS results are consistent with experimental data of Skakun *et al.*[17], Zhuravlev *et al.*[19] & Flynn *et al.*[18] in entire energy range. The results of EMPIRE 2, 3 are good with the data of Hogan[23] between 25-65 MeV within experimental error while EMPIRE 1 is good in 35 to 65 MeV energy range. The experimental data of Hogan *et al.* are underpredicted by the model calculations between ~14 MeV and 20 MeV neutron energy range. Uncertainty in the measured data is not shown for the data set which might be a reason for the mismatch.

EMPIRE data are consistent with the reported data of Levkovskii[22] except peak value while TALYS shows good agreement only within error bar limits. In this reaction, formation of $^{96}$Tc is well predicted by theoretical model codes in pre-equilibrium and compound nucleus reaction mechanism.

Figure 4 illustrates the theoretically calculated and available experimental data for the $^{98}$Mo(p,n)$^{98}$Tc reaction. The reported data points of Skakun*et al.*[17] & Flynn *et al.*[18] are in agreement with EMPIRE and TALYS results up to 9 MeV and after that no literature data points are available. The data of TALYS under predict between 9-14 MeV and over predict above 17 MeV EMPIRE results. Above 10 MeV proton energy, no definite conclusion can be

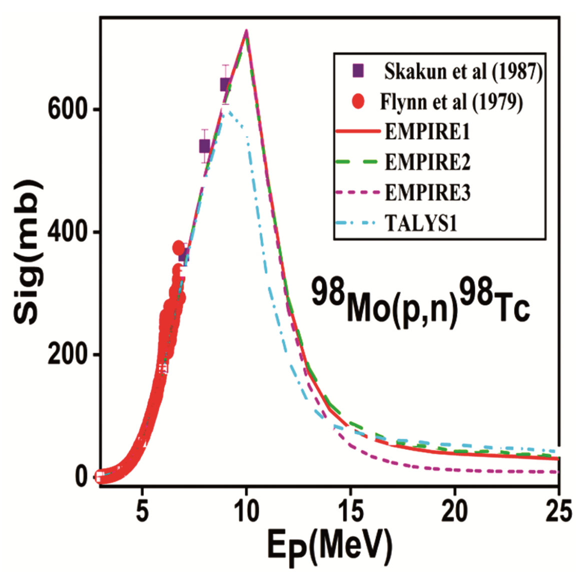


Fig 4 — Excitation function of $^{98}$Mo(p,n)$^{98}$Tc with proton energy up to 25 MeV.

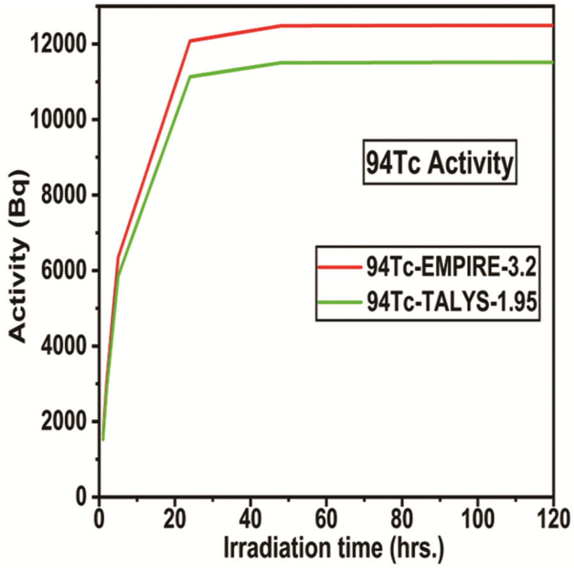


Fig. 5 — Activity build upof $^{94}$Tc as a function of irradiation time for 1 μA proton beam and a thick stopping target.

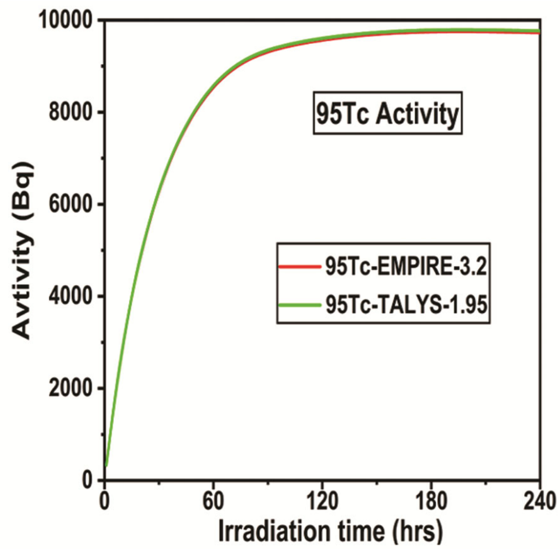


Fig. 6 — Activity build up of $^{95}$Tc as a function of irradiation time for 1 μA proton beam and a thick stopping target.

drawn due to non-availability of experimental data and here theoretical model codes come in to picture and produce data to understand nuclear reaction mechanism. The decline of the excitation curve post 14 MeV proton energy indicates the rise in pre-compound contribution in the total reaction cross-section.

For the reactions $^{95}$Mo(p,n)$^{95}$Tc and $^{96}$Mo(p,n)$^{96}$Tc both the codes reproduce experimentally measured data well except for the Hogan's data in $^{96}$Mo(p,n)$^{96}$Tc reaction. The Empire and TALYS codes use Hausher-Feshbach compound evaporation

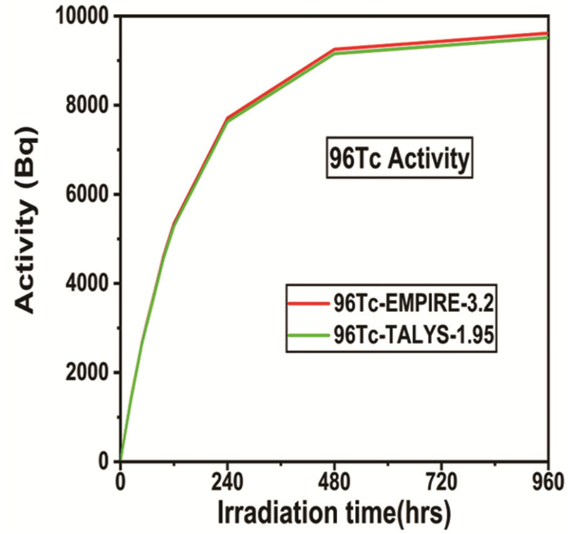


Fig. 7 — Activity build up of $^{96}$Tc as a function of irradiation time for 1 μA proton beam and a thick stopping target.

theory along with different pre-equilibrium models. These model simulations could predict the data well for the above two reactions. In the reactions $^{94}$Mo(p,n)$^{94}$Tc and $^{98}$Mo(p,n)$^{98}$Tc limited experimental data is available only up to 9 MeV. For $^{94}$Mo(p,n)$^{94}$Tc reaction the measured data are over predicted by both the simulations in the energy range of 7-9 MeV. This may be attributed to the level density of the residual nucleus. In the case of $^{98}$Mo(p,n)$^{98}$Tc reaction both EMPIRE & TALYS results are in good agreement with the experimental data. For this reaction the calculations of TALYS under predicts EMPIRE results between energy range ~7-15 MeV due to different level densities used.

The induced radioactivity, for $^{94,95,96,98}$Tc resulting from $^{94,95,96,98}$Mo(p,n) reactions has been computed by 1 μA beam current for a thick stopping target upto960 hours, as depicted in Figs. 5-8. The activity of $^{94}$Tc reaches saturation within 50 hours of irradiation, given its half-life of 4.88 hours. About 8% deviation has been observed among the induced activities calculated using TALYS and EMPIRE model codes due to the variation in the excitation function. For $^{95}$Tc ($t_{1/2}$ =20 hours) the induced activity shows an increasing trend, even after 120 hours of irradiation. The induced activities for $^{96}$Tc ($t_{1/2}$ =102.72 hours) and $^{98}$Tc ($t_{1/2}$ = 36792x10$^6$hrs) is shown in Figs 7 & 8, after 960 hours of irradiation for the $^{96}$Mo and $^{98}$Mo target, respectively. $^{98}$Mo shows very small activity after 960 hours of irradiation time due to highly stable product isotope $^{98}$Tc and the activity calculated by TALYS and EMPIRE nuclear model codes vary

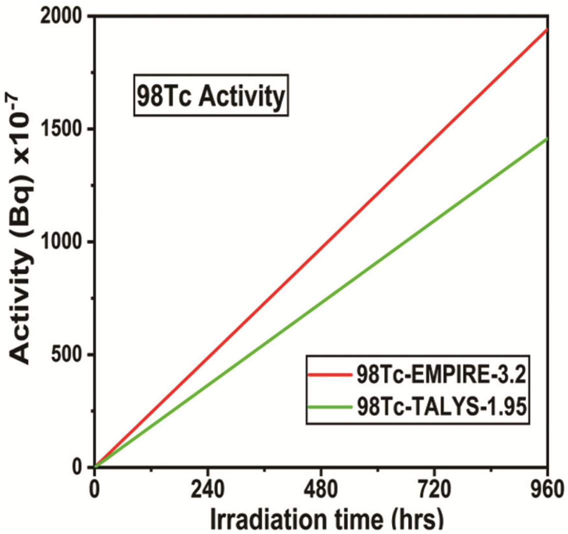


Fig. 8 — Activity build up of $^{98}$Tc as a function of irradiation time for 1 μA proton beam and a thick stopping target.

within 24% due to the variation in the excitation function. The calculation of induced activity for all the radioisotopes is carried using EMPIRE 2 option. The activity values of $^{94}$Mo and $^{98}$Mo calculated using TALYS and EMPIRE codes are differ to some extent, in each case EMPIRE gives a higher value of activity than TALYS. These estimates of induced activity can aid in proper experiment planning and ensuring radiation safety in situations where experimental data is lacking.

## 4 Conclusion

The inventory of radioisotopes produced in a nuclear reaction is important for safe disposal of radioactive waste and reaction cross section is an important input to this inventory. In the estimation of production cross-sections and induced radioactivity by a 30 MeV, 1μA proton beam impinging on Mo targets, the model codes EMPIRE-3.2.3 and TALYS-1.95 serve as invaluable assets. The pre-equilibrium models, PCROSS and Multi step compound, predict measured data very well across the energy range considered. The compound nuclear evaporation predominates in the low-energy region, while significant contribution from the pre-equilibrium process emerges around ~13–30 MeV. It is generally true that out of total reaction cross-sections; the contribution of the direct reaction mechanism is negligible compared to the compound nucleus contribution. Induced radioactivity of approximately $10^4$ Bq is produced for $^{94}$Tc from a thick $^{94}$Mo target in the present study. For $^{95,96}$Tc targets, the highest activity produced are $9.7 \times 10^4$ Bq and $9.6 \times 10^4$ Bq respectively for the irradiation time considered. The maximum activity for the $^{98}$Mo target is $1.9 \times 10^{-4}$ Bq after a 960-hour irradiation period. Simulated results from nuclear reaction models are useful to check the validity of various parameters used and for evaluating the precision of reported literature data.

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