ISSN: 0067-2904

Proton Induced Nuclear Excitation Functions of $^{100}Mo(p,2n)^{99m}Tc$ Reaction used as a Tracer in Nuclear Medicine

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Received: 21/2/2023 Accepted: 3/8/2023 Published: 10/9/2024

Abstract

Because of the importance of using ^{99m}Tc isotope as a radioactive tracer for nuclear medicine, the production of this isotope from accelerating protons at energies 1-70 MeV is investigated carefully through the evaluation of the calculated excitation functions of the $^{100}Mo(p,2n)^{99m}Tc$ reaction. For accuracy, the proton interaction channels with ^{100}Mo, the excitation functions of reactions,$^{100}Mo(p,2n)^{99m}Tc$, ^{100}Mo (p, pn)^{99}Mo, and ^{100}Mo (p, nonelastic) are calculated, using Kalbach Systematic approach in Exciton model gathered with Feshbach-Kerman-Koonin (FKK) statistical theory. The results are compared with the available published data in the experimental nuclear reaction data (EXFOR) version of November 2022. The comparisons show acceptable results for the maximum cross-section of producing ^{99m}Tc at 16 MeV protons. Also, rare data was searched for the reactions, ^{100}Mo (p, p n)^{99}Mo, and ^{100}Mo (p, nonelastic), to compare the present calculated results.

Keywords: $^{100}Mo(p,2n)^{99m}Tc$ reaction cross-section, Exciton model with FKK statistical theory, ^{100}Mo (p, pn)^{99}Mo and ^{100}Mo (p, non-elastic) reaction cross-sections.

وظائف التهيج النووي المحرض بالبروتونات للتفاعل $^{100}Mo(p,2n)^{99m}Tc$ المستخدم كمتتبع في الطب النووي

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الخلاصة :

نظرا لاهمية أستخدام نظير ^{99m}Tc كمتتبع اشعاعي للطب النووي, فقد تم التحقيق بعناية في حساب وتقييم وظائف الاثارة لانتاج هذا النظير من البروتونات المتسارعة عند الطاقات 1−70 م أ ف وللتفاعل $^{100}Mo(p, 2n)^{99m}Tc$. لغرض الدقة تم حساب وظائف الاثارة لقنوات تفاعل البروتونات مع النظير ^{100}Mo والتي تشمل التفاعلات:

(^{100}Mo (p,NON , و $^{100}Mo(p,2n)^{99m}Tc$, ^{100}Mo (p, pn)^{99}Mo على التوالي . أستخدم نموذج EXCITON مع النظرية الاحصائية FKK في الحسابات . قورنت النتائج مع البيانات التجريبية المنشورة والمتاحة EXFOR أصدار تشرين الثاني 2022 . تظهر المقارنات نتائج مقبولة عند أقصى مقطع عرضي

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لانتاج ^{99m}Tc عند طاقة البرتون 16 م أ ف . أيضا تم البحث عن بيانات نادرة للتفاعلات $^{100}Mo(p, p\ n)$
$^{100}Mo(p, Non)$, ^{99}Mo() ومناقشة النتائج المحسوبة .

1. Introduction

The metastable state of the isotope technetium-99 (^{99m}Tc) is extensively used in medical applications; it has a half-life of 6.01 hours and a convenient photopeak emission of 141keV energy [1].

^{99m}Tc is a product of the ^{99}Mo decay (Figure 1). ^{99}Mo isotope ($t_{1/2}$=65.94hr) is practically produced from the parent isotope ^{100}Mo by two methods. The first is to use thermal reactors with High Enriched Uranium (HEU) or activation analysis techniques, and the second is to use electron, proton or deuterium accelerators [2,3].

In 2016, the IAEA launched a coordinated research project called CRP (referring to the Chalk River and Patten reactors after their shutdown in 2007, causing a shortage in ^{99}Mo supply). The aim of the project was to develop and improve methods for producing ^{99m}Tc via $^{100}Mo(p,2n)^{99m}Tc$ reaction [4]. In studying this reaction, the excitation functions versus the bombardment of protons were measured. The results are convincing compared with theoretical results [5].

Since then, several other studies have been published examining the excitation functions of the reaction $^{100}Mo(p,2n)^{99m}Tc$ with conflicting results regarding the extent of the data reported, including discovering errors in some experiments [6].

On the other hand, experimental results related to the decay of radionuclides ^{99}Mo and ^{99m}Tc were evaluated, which could partly explain the lower excitation functions reported for the $^{100}Mo(p,2n)^{99m}Tc$ reaction. Additional experimental work and data have been published with a significant difference concerning this reaction data [7-13].

After reviewing the results of the publications, including the benchmark declared in the main library of the IAEA [14], which are related to the relationship of proton energies with the values of the interaction excitation functions for evaluating the $^{100}Mo(p,2n)^{99m}Tc$ reaction, a conflict in the experimental and theoretical results was found that prompted us to re-evaluate the theoretical side in the present work. In addition, in the current research work, a Kalbach Systematic approach in the Exciton model gathered with Feshbach, Kerman and Koonin (FKK) statistical theory [15] was used to calculate the excitation functions of ^{100}Mo (p, 2n) ^{99m}Tc, ^{100}Mo (p, p n)^{99}Mo, and ^{100}Mo (p, nonelastic) reactions, and the results were compared with that of the published work.

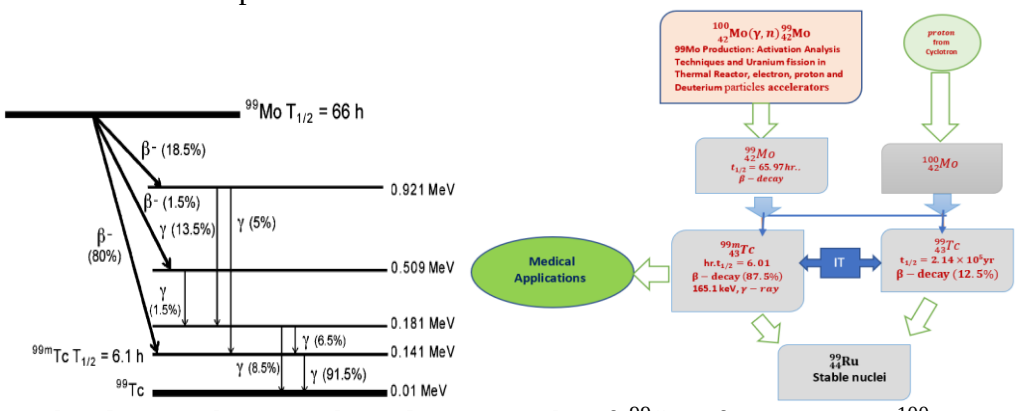

Figure 1: the decay scheme and production cycle of ^{99m}Tc from parents ^{100}Mo and ^{99}Mo nuclei [2,3].

2. Theory

A pre-equilibrium mechanism of the Exciton Model gathered with FKK statistical theory [16-18] were used to calculate the excitation functions of the ^{100}Mo(p,2n)^{99m}Tc, ^{100}Mo(p,pn)^{99}Mo, and ^{100}Mo(p, NON) reactions. For the purpose of identifying the details of the equations used in the present work, one can refer to the study of Jasim and Idrees [15] to find the excitation functions of the above reactions.

For an A (a, b) B reaction, the probability of the Compound Nucleus (CN) decaying into the corresponding outgoing channel b is [19]:

$$\sigma(a,b) = \sigma_c(a)\frac{W_b}{\sum_{\dot{b}} W_b}$$

where: $\sigma_c(a)$ is the cross-section of the CN formed by projectile a , W_b is the probability of the CN decaying into channel b and $\sum_b W_b$ is the sum of the overall possible decay channels. Therefore, the decay probability of the CN is given by Weisskopf-Ewing evaporation formula [19]:

$$W_b(E,\varepsilon_b) = \frac{2s_b+1}{\pi^2\hbar^3}\,\mu_b\,\varepsilon_b\,\sigma_{b,invers}(\varepsilon_b)\,\frac{\Omega(U)}{\Omega(E)} \quad (1)$$

where: s_b is the spin of particle b, μ_b is the reduced mass of particle b, $\sigma_{b,invers}(\varepsilon_b)$ is the inverse reaction cross-section of particle b, $\Omega(U)$ and $\Omega(E)$ are the state densities at excitation energy, respectively, where U=E- ε_b-B_b, E is the interaction energy, ε_b is the channel energy and B_b is the binding energy of particle b.

Here in this work, Equation (1) was characterized in terms of Exciton particle and hole, $(p_\pi, h_\pi, p_\nu, h_\nu)(where\ \pi\ denotes$ proton and ν neutron). The interaction energy and excitation energy are corrected to surface effect, pairing, shell structure and Pauli corrections, and so Equation (1) becomes [15,16-18]:

$$W_b((p_\pi,h_\pi,p_\nu,h_\nu,E,\varepsilon) = \frac{2s_b+1}{\pi^2\hbar^3}\,\mu_b\,\varepsilon_b\,\sigma_{b,invers}(\varepsilon_b)\times$$
$$\sum_{T_B} [C_b(T,T_B)]^2\,\frac{\Omega_{eff}((p_\pi-Z_b,h_\pi,p_\nu-N_b,h_\nu,U)}{\Omega((p_\pi,h_\pi,p_\nu,h_\nu,E)} \quad (2)$$

where, the effective corrected particle-hole state density is:

$$\Omega_{eff}((p_\pi - Z_b, h_\pi, p_\nu - N_b, h_\nu, U) = \sum_{i=i_{min}}^{h_\pi}\sum_{j=j_{min}}^{h_\nu}\Omega((p_\pi - Z_b, i, p_\nu - N_b, j, U)$$

T_B is the isospin quantum number in the residual nucleus B, $C_b(T,T_B)$ is the isospin coupling Clebsch-Gordan coefficient in the exit channel; the sum extends over all the allowed isospins in the residual nucleus, and Ω is the particle-hole state density with different corrections [20-22].

The total energy spectrum of the pre-equilibrium model for the emitted particle b at energy, E, and spin dependent formulations is [16-18,20,23-25]:

$$\frac{d\sigma_{a,b}(E,T)}{d\varepsilon_a}|_{pre} = \sum_T \frac{d\sigma_{a,b}(E,T)}{d\varepsilon_a}|_{pre} = \sigma_a(\varepsilon_a)\sum_T [C_a(T,T_A]^2 \times$$
$$\sum_P \sum_{P_\pi} S_{pre}(p,p_\pi,T)\times\Omega_b(p,p_\pi,E,\varepsilon,T) \quad (3)$$

where: T is the composite isospin, $\sigma_a(\varepsilon_a)$ is the cross-section of the complex nucleus, $S_{pre}(p,p_\pi,T)$ is the average amount of time spent in each class of configuration channel and $p=A_a+1$, A_a =n=p [15-18].

The systematics of the present model expand to calculate the angular distributions with Multistep Direct (MSD) and Multistep compound (MSC), regarding both the necessity of considering the unbound particle degree of freedom at each stage of reaction and that all

particles are bound of reaction. The basic formula suggested by Kalbach for the double differential cross-section for including the mechanism in the reaction A(a,b)B can be written as [12]:

$$\frac{d^2\sigma}{d\omega d\varepsilon_b} = \frac{1}{4\pi}\frac{d\sigma}{d\varepsilon_b}\frac{a_{ex}}{sinhsinh\,(a_{ex})}\,[cosh\,cosh\,(a_{ex}\,cos\theta) + f_{msd}sinh\,(a_{ex}\,cos\,cos\,\theta)] \quad (4)$$

where: a_{ex} is the slope parameter associated with the Exciton model and its related components, $f_{msd}(\varepsilon_b)$ is the fraction of the cross-section at the specified emission energy and θ is the scattering angle in the center of mass system [6].

Equations (3) and (4) were applied to find out the appropriate proton energy for the highest production of ^{99m}Tc from the ^{99m}Tc the ^{100}Mo(p,2n)^{99m}Tc reaction, and to compare the excitation functions of the products of ^{100}Mo(p,pn)^{99}Mo, ^{100}Mo(p,2p)^{99}Nb and ^{100}Mo(p, non-elastic) reactions.

3. Evaluatin and discussion of the results

3.1 ^{100}Mo (p, 2n) ^{99m} Tc Reaction

The increasing demand for the ^{99m}Tc isotope in medical applications has prompted scientists to use the accelerator method instead of using highly enriched uranium in the reactor because of the limitations imposed. Accordingly, proton acceleration was studied theoretically in this work for the direct production of the isotope ^{99m}Tc as one of the alternative production methods using the reaction ^{100}Mo(p,2n)^{99m}Tc on a highly enriched target material such as ^{100}Mo. The calculated excitation functions of this reaction are shown in Figure 2, where the results are compared with the experimental results of other published studies [13,14,16,22,21,24-26]. The comparison distinguished the highest value of the proton energy of 16 MeV, which has the highest probability value for the formation of the isotope ^{99m}Tc with a value close to 300 mb. In addition, the deviation in the cross-section comparisons is definitely due to the accuracy and efficiency of the detection systems for this reaction in the experimental results.

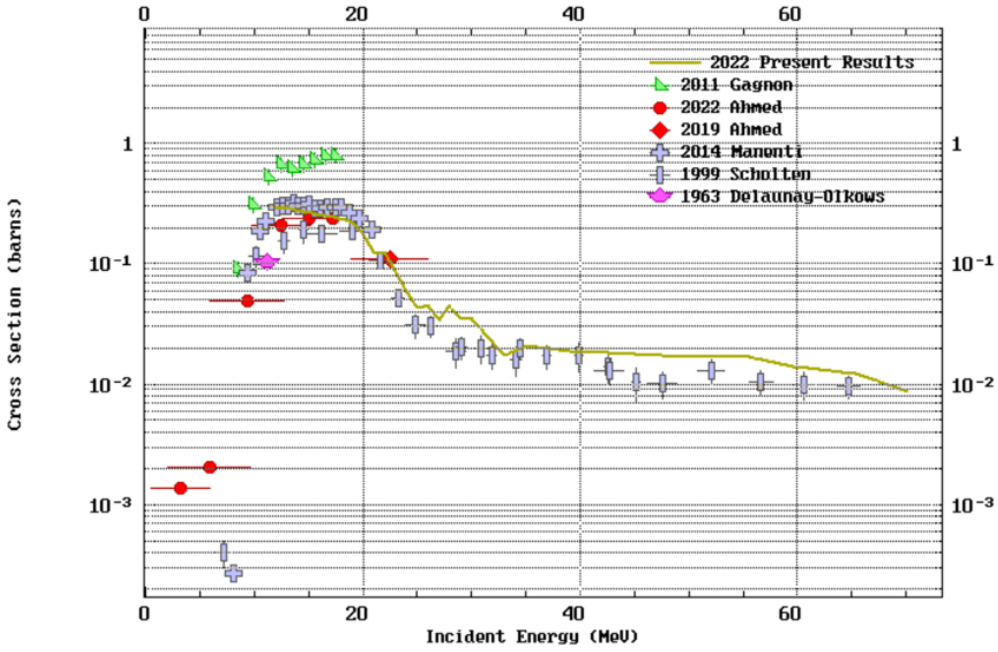

Figure 2: The calculated excitation functions of the ^{100}Mo(p,2n)^{99m}Tc reaction at different proton energies with comparison with experimental and theoretical results [13,14,16,21, 22, 24-26].

For the CN [^{100}Tc] and the primary residual nucleus ^{99m}Tc at excitation energy range between 1-5 MeV, the secondary emission of ^{99m}Tc decay, with 140.5keV gamma, is considered within the energy spectrum behavior of ^{100}Mo(p,2n)^{99m}Tc reaction, as shown in Figure 3. As shown in this figure, the excitation function behavior for the reaction channel is concentrated at the excitation energy range between 1 and 2 MeV for compound ^{100}Mo and residual^{99m}Tc nuclei, which are proportional to the incident proton energy E_p and the threshold energy E_{res} values of the channel.

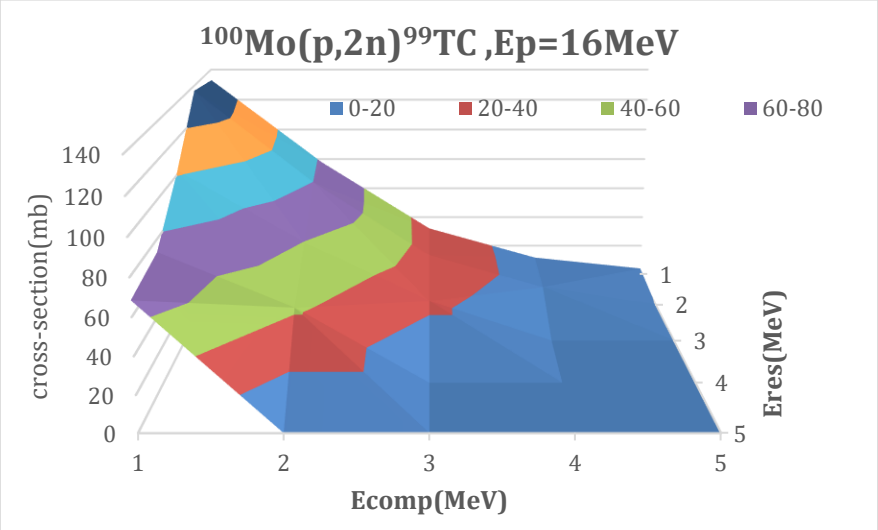

Figure 3: The energy spectrum of the ^{100}Mo (p, 2n)^{99m}Tc reaction at different excitation energies for residual ^{99m}Tc and compound ^{100}Mo nucleuses.

3.2 ^{100}Mo(p,n+p)^{99}Mo Reaction

Figure 4 shows an acceptable agreement with reference [26], noting that the highest cross-section value reaches 0.138mb at proton energy 30 MeV.

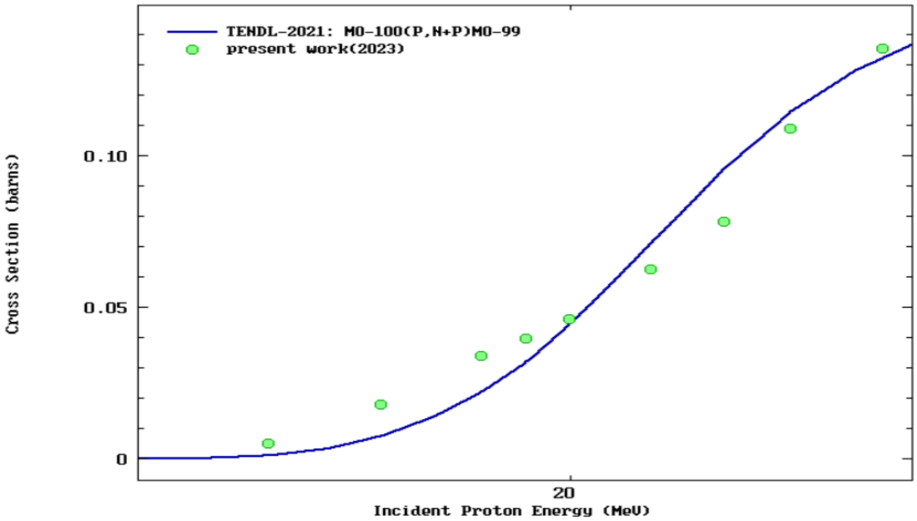

Figure 4: Excitation function of ^{100}Mo(p,n+p) ^{99}Mo reaction at different proton energy 1- 30 MeV, compared with the TALYS-based evaluated nuclear data library [26] where the present model is found appropriate to calculate the particle emission rates at equilibrium stage.

3.3 ^{100}Mo (p, non-elastic) reaction

The calculated excitation functions of the reaction are compared with other published data [26-28], as shown in Figure 5, and the convergence of the results appears at the proton

energy range of 1-70 MeV. The fitted maximum probability of the reaction ^{100}Mo (p, non-elastic) is found at 20-30 MeV proton energy, indicating that this model is appropriate for calculating the transmission coefficients.

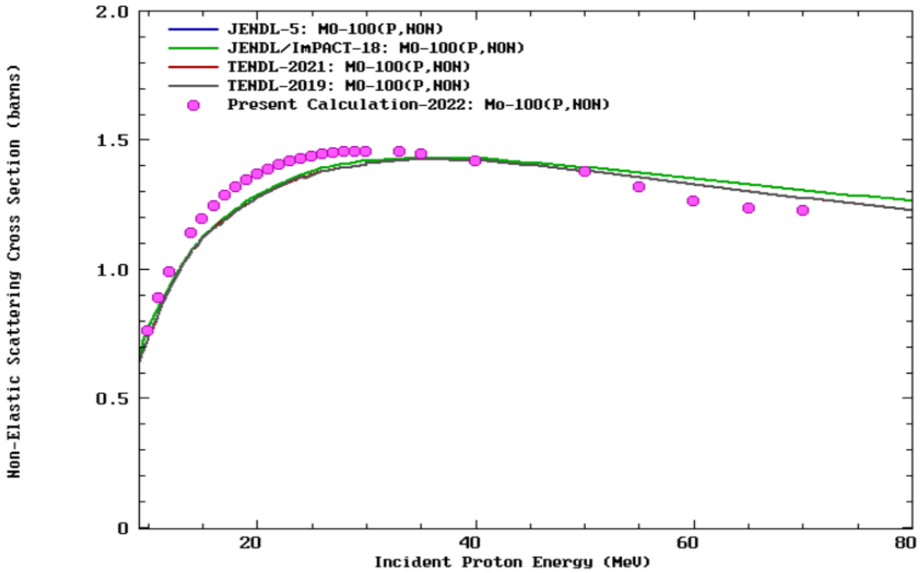

Figure 5: Excitation function of ^{100}Mo (p, NON) reaction at different proton energy compared with different references [26-28], where the present model is appropriate to calculate the transmission coefficients.

4. Conclusion

Due to the importance of the radioactive isotope ^{99m}Tc in medical applications, calculations were made for excitation functions of the reaction ^{100}Mo(p,2n) ^{99m}Tc when protons are accelerated between 1-70 MeV and the search for the energy to achieve the highest probability of producing such isotope. The Exciton model with FKK statistical theory was implemented. Compared with the experimental results, a match was found with the published data in the EXFOR-2022 issue. Where the highest excitation function value of 300mb was determined at 16 MeV proton energy compared to experimental results: 327$\pm$43 mb [6], 307$\pm$26 mb [11] and 292$\pm$13.38 mb [13] for different research work.

The relationship of the excitation functions with the excitation potential of the composite nucleus ^{100}Tc and the primary residual nucleus ^{99m}Tc was investigated. It was found that the maximum probability of the reaction ^{100}Mo(p,2n) ^{99m}Tc was at the excitation energy range 1-2 MeV, at which the secondary emission ($E\gamma = 140.5\ keV$) of the isotope occurs, see Figure 3. The highest value of the reaction's excitation functions for the reactions ^{100}Mo (p, n p) ^{99}Mo was 31mb at 16 MeV proton energy, which is consistent with TALYS calculations [26]. In addition, the results of calculating the excitation functions for the reaction ^{100}Mo (p, NON) were consistent with the results of [26,28], see Figure 5, where the highest probability was found in the proton energy range of 15-30 MeV.

In final conclusion, the most dominant excitation functions at different proton energies were found to be for the reaction ^{100}Mo(p,2n) ^{99m}Tc exceeding the values of other channels, ^{100}Mo (p, n p) ^{99}Mo and ^{100}Mo (p, 2p) ^{99}Nb, which nominates the concept of ^{99m}Tc isotope production cycle using cyclotron rather than irradiation ^{100}Mo by thermal reactor.

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