Benchmark experiment for the cross section of the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions

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Abstract

As nuclear medicine community has shown an increasing interest in accelerator produced ^{99m}Tc radionuclide, the possible alternative direct production routes for producing ^{99m}Tc were investigated intensively. One of these accelerator production routes is based on the ^{100}Mo(p,2n)^{99m}Tc reaction. The cross section of this nuclear reaction was studied by several laboratories earlier but the available data-sets are not in good agreement. For large scale accelerator production of ^{99m}Tc based on the ^{100}Mo(p,2n)^{99m}Tc reaction, a well-defined excitation function is required to optimize the production process effectively. One of our recent publications pointed out that most of the available experimental excitation functions for the ^{100}Mo(p,2n)^{99m}Tc reaction have the same general shape while their amplitudes are different. To confirm the proper amplitude of the excitation function, results of three independent experiments were presented [1]. In this work we present results of a thick target count rate measurement of the E_γ=140.5keV gamma-line from molybdenum irradiated by E_p = 17.9 MeV proton beam, as an integral benchmark experiment, to prove the cross section data reported for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions in [1].

Keywords: ^{99m}Tc, ^{99}Mo, thick target count rate, cross section, cyclotron

1. Introduction

As an alternative to reactor produced ^{99}Mo/^{99m}Tc generator technology, the direct production of ^{99m}Tc on cyclotrons is considered. The possible reactions and their cross sections, the achievable production yields, specific activity and purity problems were discussed, refer to some selected publications [2, 3, 4, 5, 6]. Cross sections of proton induced nuclear reactions on natural and enriched molybdenum have been studied extensively. Several experimental and evaluated data-sets and evaluation are published for the activation cross sections of different reactions regarding the production of ^{99m}Tc and ^{99}Mo radionuclides. Studies on measuring the cross sections of the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions as a function of the bombarding proton energy were carried out by many research groups with conflicting results regarding the amplitude of the reported data [1], [7], [8], [9], [10], [11], [12], [13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23] by using both natural Mo and enriched ^{100}Mo targets. Selected data-sets of the available cross sections for

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the $^{100}Mo(p,2n)^{99m}Tc$ reaction are collected in Fig. 1 and for the $^{100}Mo(p,pn)^{99}Mo$ reaction in Fig 2. In these figures Levkovskij's data (1991) [8] are renormalized by a factor of 0.82 to be consistent with the latest $^{nat}Mo(p,x)^{96g}Tc$ monitor cross section by Takács (2003) [24] Fig. 1 clearly shows the amplitude differences among the available data-sets, and corrections (reevaluation) are attempted in both Figs. 1 and 2.

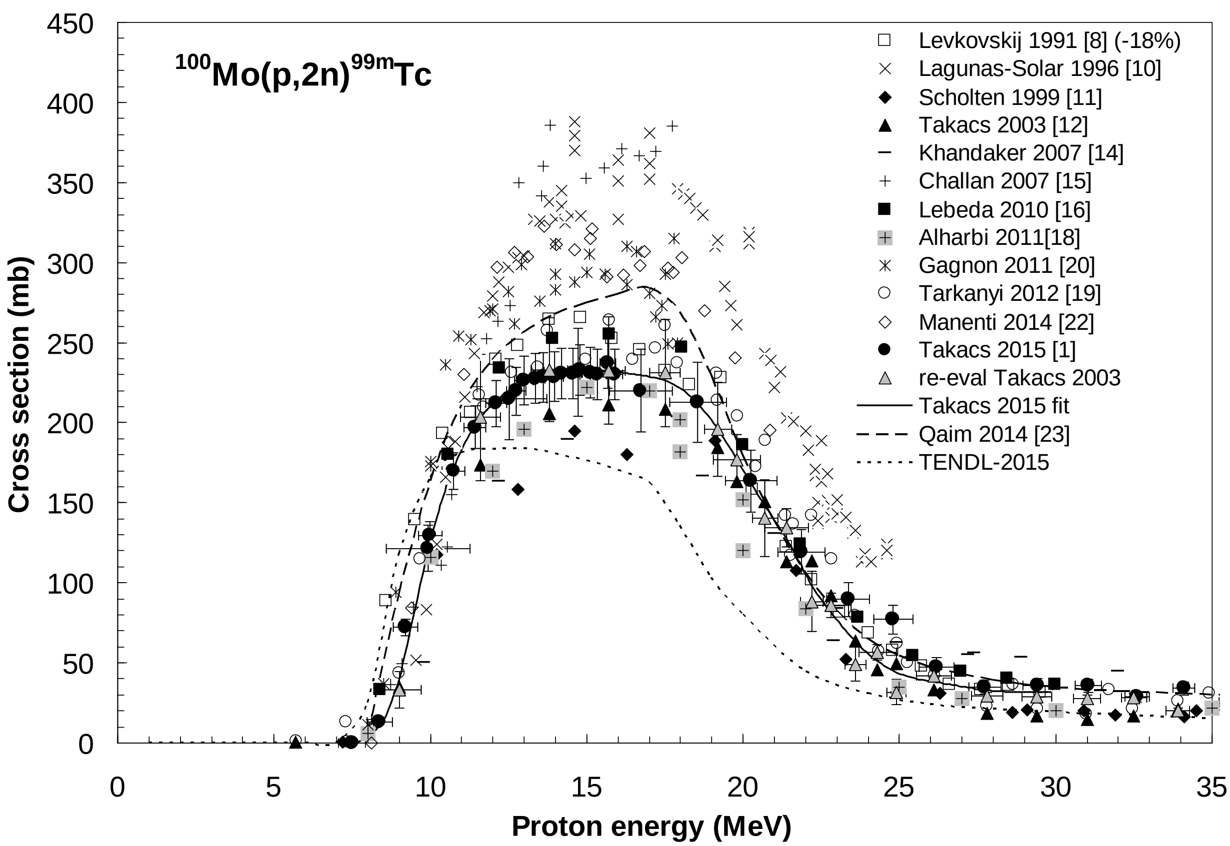

Figure 1. Available experimental and evaluated cross section data for the $^{100}Mo(p,2n)^{99m}Tc$ reaction.

In our recent work [1] three independent experiments were performed with the aim to determine the amplitude of the excitation function of the $^{100}Mo(p,2n)^{99m}Tc$ reaction. Three experiments were carried out at E_p = 16, 36.4, and 38 MeV bombarding proton energies. New experimental cross section data were provided on a Mo target with natural isotopic composition, to clarify the existing discrepancies among the available data-sets. Determination of the cross section of the $^{100}Mo(p,2n)^{99m}Tc$ reaction does not require a ^{100}Mo enriched target material, since only two reactions contribute to direct production of ^{99m}Tc, the $^{100}Mo(p,2n)^{99m}Tc$ main reaction and the $^{98}Mo(p,\gamma)^{99m}Tc$ reaction with negligible contribution. As it was pointed out, the three new data-sets measured in independent experiments have a very good overall agreement among each other both in shape and in amplitude. The excitation functions of the $^{100}Mo(p,x)^{99}Mo$ and $^{100}Mo(p,2n)^{99m}Tc$ reactions were determined experimentally by using analytically derived equations in the data evaluation, avoiding various approximations in the data analysis. The good agreement among the results of the three independent irradiations proves that the main discrepancy among the earlier published experimental cross section data for the $^{100}Mo(p,2n)^{99m}Tc$

reaction could originate from large uncertainty of the used outdated decay data and probably from the applied data deduction methods.

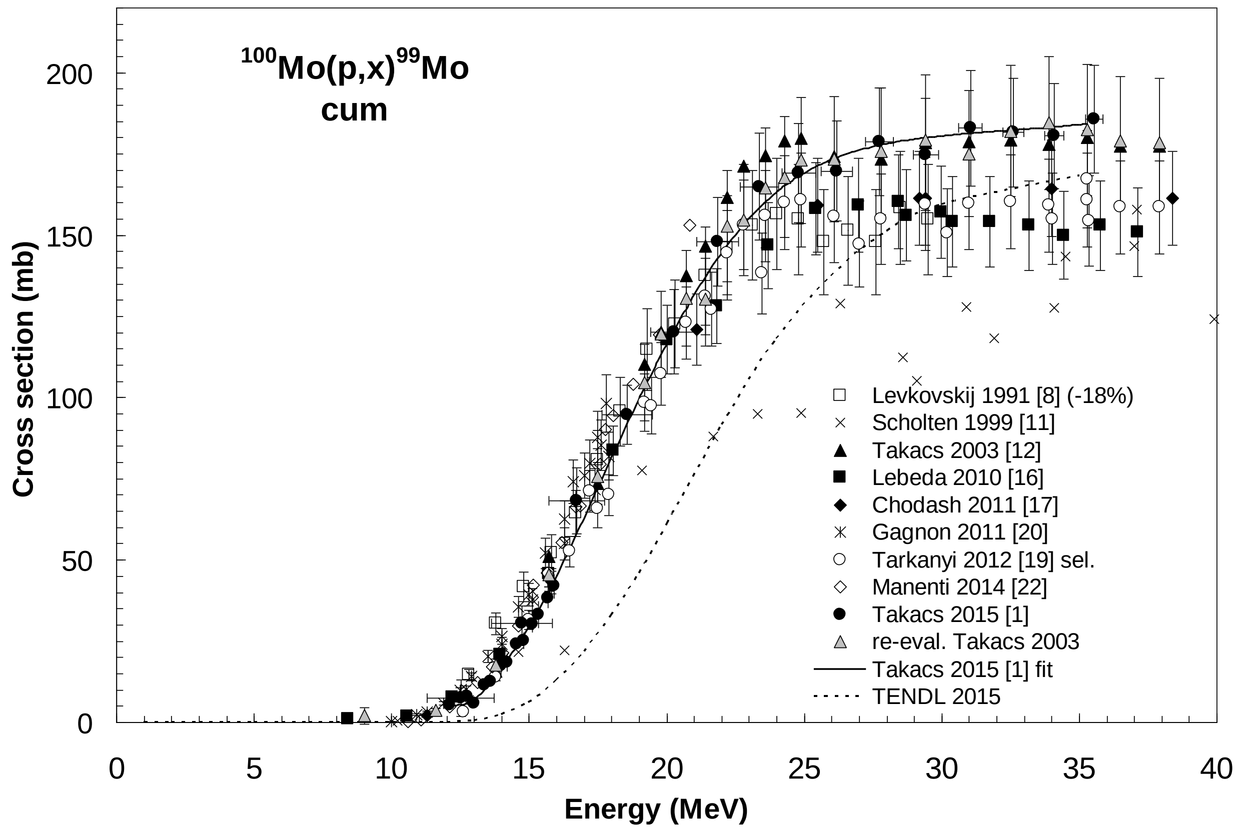

Figure 2. Selected experimental and evaluated cumulative cross section data (including production by decay of the short lived ^{99}Nb) for the ^{100}Mo(p,x)^{99}Mo reaction.

The aim of this work was to give a further confirmation of the results presented in our recent paper [1] on cross section data of the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions by measuring thick target count rates of the reactions and comparing them with values calculated by using the cross section data presented in [1].

Count rate determination

A simple experiment was performed in which the intensity of the E_γ = 140.5 keV gamma-line originating from decay of ^{99m}Tc and ^{99}Mo isotopes was measured. These radionuclides are produced in the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions. A 1 mm thick Mo target with natural isotopic composition was irradiated with an E_p = 17.9 MeV proton beam and the activity of the produced ^{99m}Tc and ^{99}Mo was measured through the common E_γ = 140.5 keV gamma-line. The standard activation method and high resolution HPGe-gamma-spectrometry were used for determining the activity of the irradiated sample. Since the E_γ = 140.5 keV gamma-line can originate from the decay of both the ^{99m}Tc and ^{99}Mo radioisotopes, and ^{99}Mo decays partially into

^{99m}Tc, the counting was repeated several times to follow the decay of the two radionuclides to be able to identify properly the sources of the radiation. The count rate was also determined by calculation, using a simple model according to the well-known activation and decay laws. In the calculation the cross sections presented in [1] for the $^{100}Mo(p,2n)^{99m}Tc$ and $^{100}Mo(p,pn)^{99}Mo$ nuclear reactions and the latest nuclear decay data [25] regarding the decay of the ^{99m}Tc and ^{99}Mo radionuclides were involved. The model calculation is based on the equations presented in [1] and repeated here below. The equations include all possible sources of the E_γ = 140.5 keV gamma-line that can originate from the complex activation and decay processes of the ^{99}Mo-^{99m}Tc mother-daughter radionuclide pair. The count rate determined by model calculation was then compared to the experimentally determined count rate to validate the used cross sections.

Irradiation

A target composed of a Ti foil (10.9 microns, from Goodfellow, UK) and a Mo disc (chemical purity 99.9%, 1 mm thick with natural isotopic composition) was irradiated for 5 minutes with an E_p = 18 MeV and I_p = 50 nA proton beam from the ATOMKI cyclotron. The irradiation took place in a Faraday-cup under vacuum which assured the proper charge integration. The Ti foil was placed in front of the Mo target to monitor the beam intensity. For monitoring the beam energy, another stack, which composed from Ti foils with the same thickness as the one placed in front of the Mo target and Al energy absorber foils, was irradiated by using the same proton beam provided by cyclotron without changing its tuning parameters. The 1 mm thick Mo target was thick enough to stop the proton beam completely. The effective range (from the on-surface bombarding energy, E_p = 17.9 MeV, down to the respective threshold energy of the two reactions) of the proton beam in the Mo target was about 487 and 466 micro-meters [26], regarding production of ^{99m}Tc and ^{99}Mo isotopes, respectively. The particle flux was normalised to the upgraded web version of the recommended values of the $^{nat}Ti(p,x)^{48}V$ monitor reaction provided by IAEA [27]. Comparing the direct measured beam intensity value and the beam intensity deduced from the monitor reaction, a difference of 2% was found. In the model calculation for determining the count rate, the beam current derived from monitor reaction was adopted.

Activity measurement

Proton activation technique and high resolution gamma-spectrometry method were used for measuring the count rate of the E_γ = 140.5 keV gamma-line from the complex decay of ^{99m}Tc and ^{99}Mo isotopes produced in the Mo target. For the gamma-measurement a high resolution HPGe gamma-spectrometer was used, the same which was used for the cross section measurement [1]. The irradiated Mo sample was fixed in an efficiency calibrated detector-sample position with the irradiated surface facing toward the detector.

Series of spectra were recorded to follow the intensity change of the E_γ = 140.5 keV gamma line due to decay and growth processes in a time period of about one week. Each spectrum was analysed and the net peak area was determined, from which an average count rate of the E_γ = 140.5 keV gamma-line was calculated, including the necessary corrections for dead time loss and isotopic abundance of the ^{100}Mo isotope of the target. For determining the experimental count rates only the irradiation and measuring parameters (irradiation time, cooling time and measuring time for the given beam energy and beam intensity) were used to evaluate the spectra and no any decay data or cross section data were involved in the data deduction process; therefore the experimental count rates can be considered as absolute values for this given experiment. The time dependent count rates as primary experimental data are presented in Fig. 4 together with the calculated values (solid line). In the calculation the cross sections reported in [1] for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions and the latest decay data published in [25] were used.

Estimating the count rate by calculation

In the experiment we have followed the decay of the activated Mo target by measuring the count rates of the E_γ = 140.5 keV gamma-line, which has a complex time dependent origin according to the formation and decay process of the ^{99m}Tc and ^{99}Mo radionuclides. It can be considered as four independent sources as follows.

- Decay of ^{99m}Tc produced directly in the ^{100}Mo(p,2n)^{99m}Tc reaction during irradiation, (Eq. 1)
- Decay of ^{99m}Tc produced during irradiation exclusively by decay of ^{99}Mo, (Eq. 2)
- Decay of ^{99m}Tc produced after end of bombardment (EOB) from decay of ^{99}Mo, (Eq. 3)
- Prompt gamma radiation that follows the decay of ^{99}Mo (Eq. 4).

The corresponding contributions to the T_γ total peak area acquired during a t_m measuring time after a t_b irradiation and a t_c cooling time for the Mo target can be described by the following equations for the above four processes, respectively.

$$T_\gamma(direct)_D = \varepsilon_d \varepsilon_{\gamma_2} \varepsilon_t N_t N_b \overline{\sigma_2}\left(1-e^{-\lambda_2 t_b}\right)\frac{1}{\lambda_2} e^{-\lambda_2 t_c}\left(1-e^{-\lambda_2 t_m}\right) \tag{1}$$

$$T_\gamma(decay)_x = \varepsilon_d \varepsilon_{\gamma_2} \varepsilon_t \frac{f N_t N_b \overline{\sigma_1}}{(\lambda_1-\lambda_2)}\left[\lambda_1\left(1-e^{-\lambda_2 t_b}\right)-\lambda_2\left(1-e^{-\lambda_1 t_b}\right)\right]\frac{1}{\lambda_2} e^{-\lambda_2 t_c}\left(1-e^{-\lambda_2 t_m}\right) \tag{2}$$

$$T_\gamma(decay)_y = \varepsilon_d \varepsilon_{\gamma_2} \varepsilon_t \frac{f N_t N_b \overline{\sigma_1}}{(\lambda_1-\lambda_2)}\left(1-e^{-\lambda_1 t_b}\right)\left[e^{-\lambda_2 t_c}\left(1-e^{-\lambda_2 t_m}\right)-\frac{\lambda_2}{\lambda_1} e^{-\lambda_1 t_c}\left(1-e^{-\lambda_1 t_m}\right)\right] \tag{3}$$

$$T_\gamma(direct)_M = \varepsilon_d \varepsilon_{\gamma_1} \varepsilon_t N_t N_b \overline{\sigma_1}\left(1-e^{-\lambda_1 t_b}\right)\frac{1}{\lambda_1} e^{-\lambda_1 t_c}\left(1-e^{-\lambda_1 t_m}\right) \tag{4}$$

Where

N_t	surface density of target atoms, [atom/cm^2],
N_b	number of bombarding particles per unit time, [proton/sec],
$\overline{\sigma_i}$	thick target activation cross sections, [cm^2], a mean cross section, averaged over the effective bombarding energy with weighting factor of the stopping power [26],
f	decay branching ratio of ^{99}Mo to ^{99m}Tc,
ε_d	detector efficiency,
$\varepsilon_{\gamma\iota}$	corresponding gamma intensity,
ε_t	dead time correction,
λ_i	decay constant, [1/sec],

t_b bombarding time, [sec],
t_c cooling time, [sec],
t_m acquisition time, [sec],
i the i = 1 index refers to the ^{99}Mo parent radionuclide and i = 2 refers to the ^{99m}Tc daughter radionuclide.

The corresponding expected mean count rates were also estimated by calculation for each of the measured points, using the above four equations, by taking the beam energy, beam intensity, irradiation time, cooling time and measuring time, the mean activation cross section, decay data and detector efficiency. The necessary corrections were also applied to the calculated values such as detector efficiency for the E_γ = 140.5 keV gamma-line, gamma branching ratios and absorption of the relatively low energy gamma photons in the thick Mo target. The standard atomic weight A_{Mo} = 95.96(2) [28] and the isotopic composition of molybdenum, ^{92}Mo: 14.525(15)%, ^{94}Mo: 9.1514(74)%, ^{95}Mo: 15.8375(98)%, ^{96}Mo: 16.672(19)%, ^{97}Mo: 9.5991(73)%, ^{98}Mo: 24.391(18)% and ^{100}Mo: 9.824(50)%, were taken from the IUPAC Technical Report 2009 [29] and were used in the calculations. Figure 3 shows the simplified decay scheme of ^{99m}Tc and ^{99}Mo. Table 1 contains information about the actual gamma branching ratios of the E_γ = 140.5 keV gamma-photon used in the calculations.

Eq. 1 contains information about the direct production cross section of ^{99m}Tc, the other three equations depend on the cumulative production cross section of ^{99}Mo (including production of ^{99}Mo by decay of the short-lived ^{99}Nb). Using the count rate versus cooling time curve, the excitation functions for both of the reactions, ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo, can be verified. At short cooling times the count rate curve is mainly determined by the decay of the shorter-lived ^{99m}Tc, while after a long cooling time the curve depends only on the activity of the ^{99}Mo isotope. More details on the roll of the equations can be found in [1].

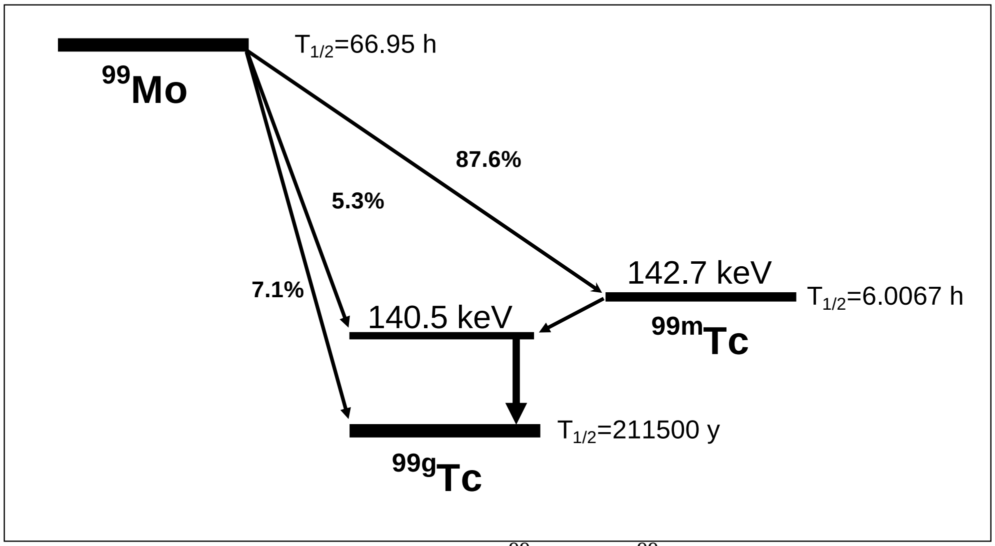

Figure 3. Simplified decay scheme of ^{99}Mo and ^{99m}Tc.

Table 1. Intensities of the E_γ = 140.5 keV gamma photons emitted after prompt and delayed population of the E = 140.5 keV excited level of ^{99}Tc [25].

Intensities of the E_γ = 140.5 keV gamma-photons from different sources	$\varepsilon_{\gamma i}$ (%)
Prompt gamma intensity of the decaying ^{99}Mo, $\varepsilon_{\gamma 1}$	4.72
In equilibrium of the decaying ^{99}Mo and ^{99m}Tc	89.6
Decay of pure ^{99m}Tc (no decaying ^{99}Mo is present), $\varepsilon_{\gamma 2}$	88.5

Results and Discussion

We choose the simplest experimental quantity, the count rate, to measure. Comparing the result of the model calculation based on the cross sections presented in [1] and the measured experimental count rates an excellent agreement was found (Figure 4. open symbol: experimental values, solid line: calculated value). This agreement can give strong confirmation of the excitation functions for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions reported in [1]. The solid curve, in Fig. 4, representing the calculated count rates for the E_γ = 140.5 keV gamma-line, obviously depends on the cross sections of the two reactions involved in the process. The higher/lower the value of the cross section used in the calculation, the higher/lower the calculated count rate is. This effect can be used for validation of the cross section data and to predict the possible error sources according to the observed difference between the calculated and measured count rates.

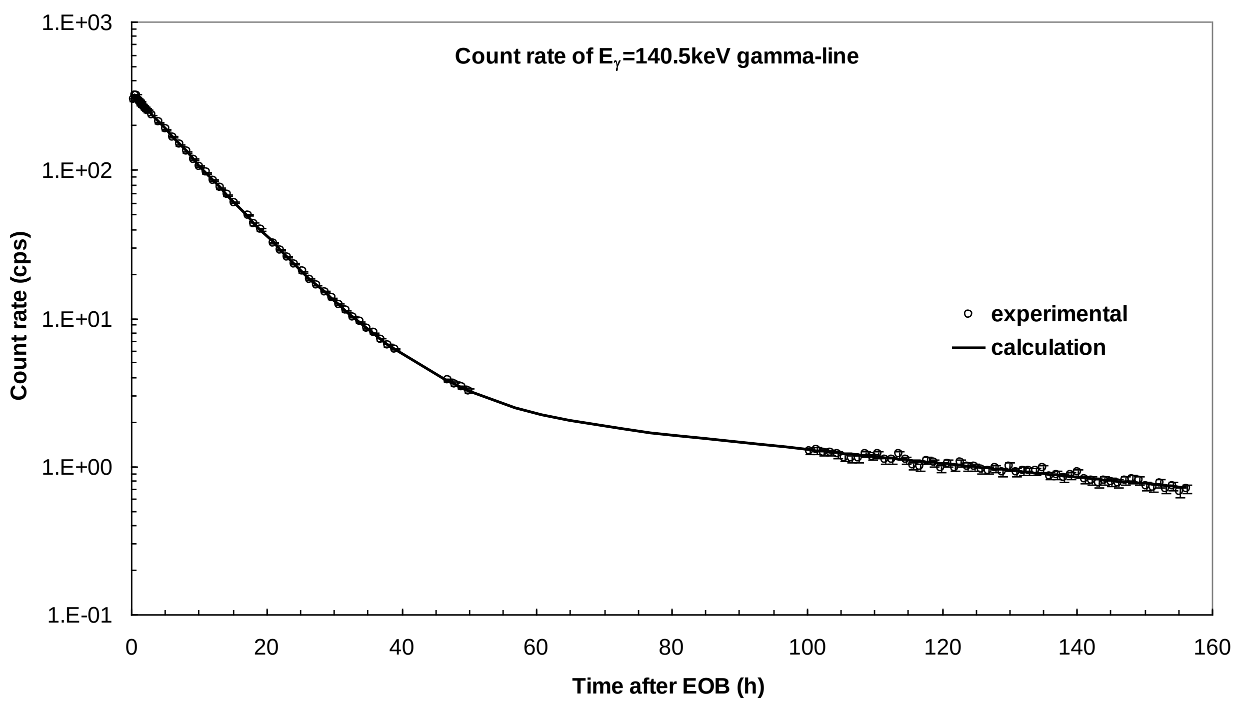

Figure 4. Count rate determined for the E_γ = 140.5 keV gamma-line in a thick Mo target irradiated with a 17.9 MeV proton beam. Open symbols: experimental values, solid line: calculated curve based on the cross sections published in [1].

Since determination of the experimental count rate depends only on the irradiation and measuring parameters and effectively does not require knowledge of the decay parameters of the two isotopes; therefore this experiment also can confirm the decay data used in the model calculation of the count rate. Fig. 4 has logarithmic scale, which would make difficult to see properly the actual shifts among the tested different data series. Instead the figure was divided into two parts for short (Fig.5) and long (Fig.6) cooling time where the contribution of ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo are expected to be dominant, respectively and linear-linear scale was applied to show the differences among the tested cross section data-sets.

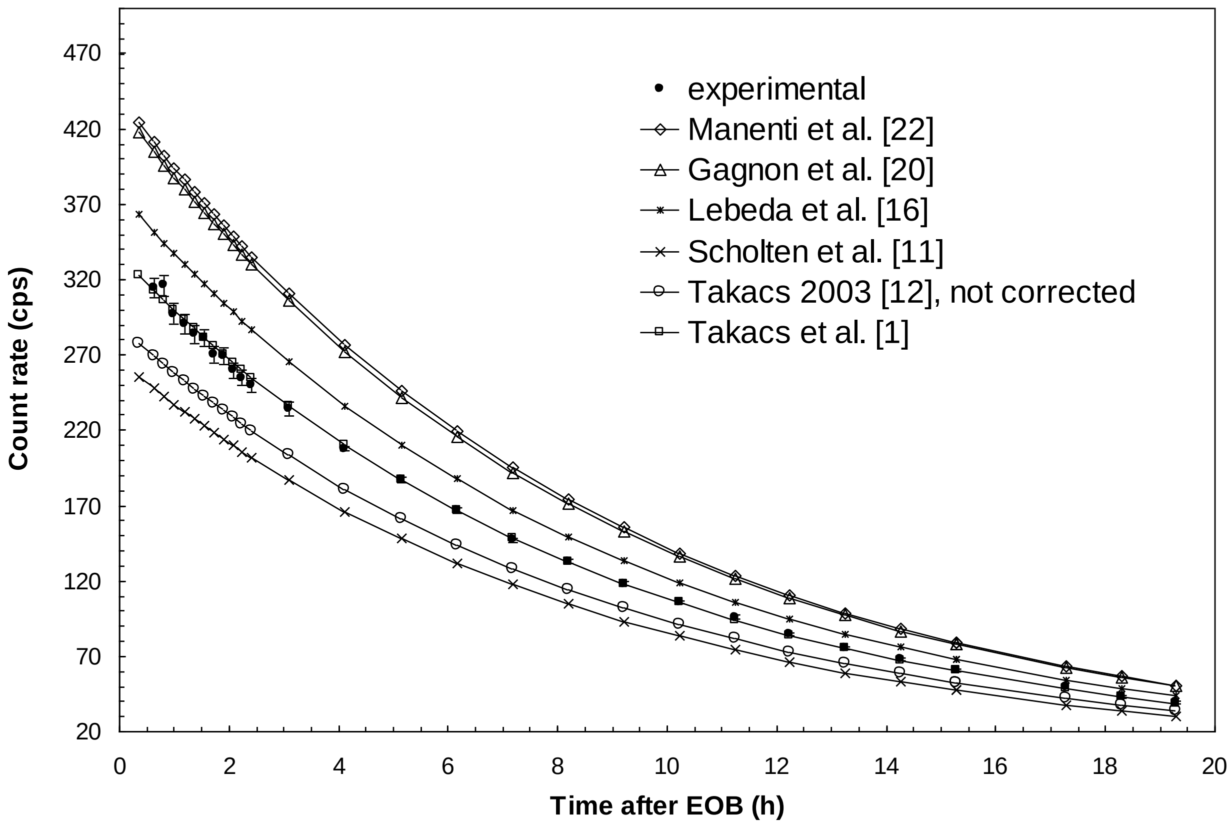

Figure 5. Experimental count rate measured for short cooling time for the E_γ = 140.5 keV gamma-line in a thick Mo target irradiated with a 17.9 MeV proton beam and calculated count rates using different experimental cross section data-sets.

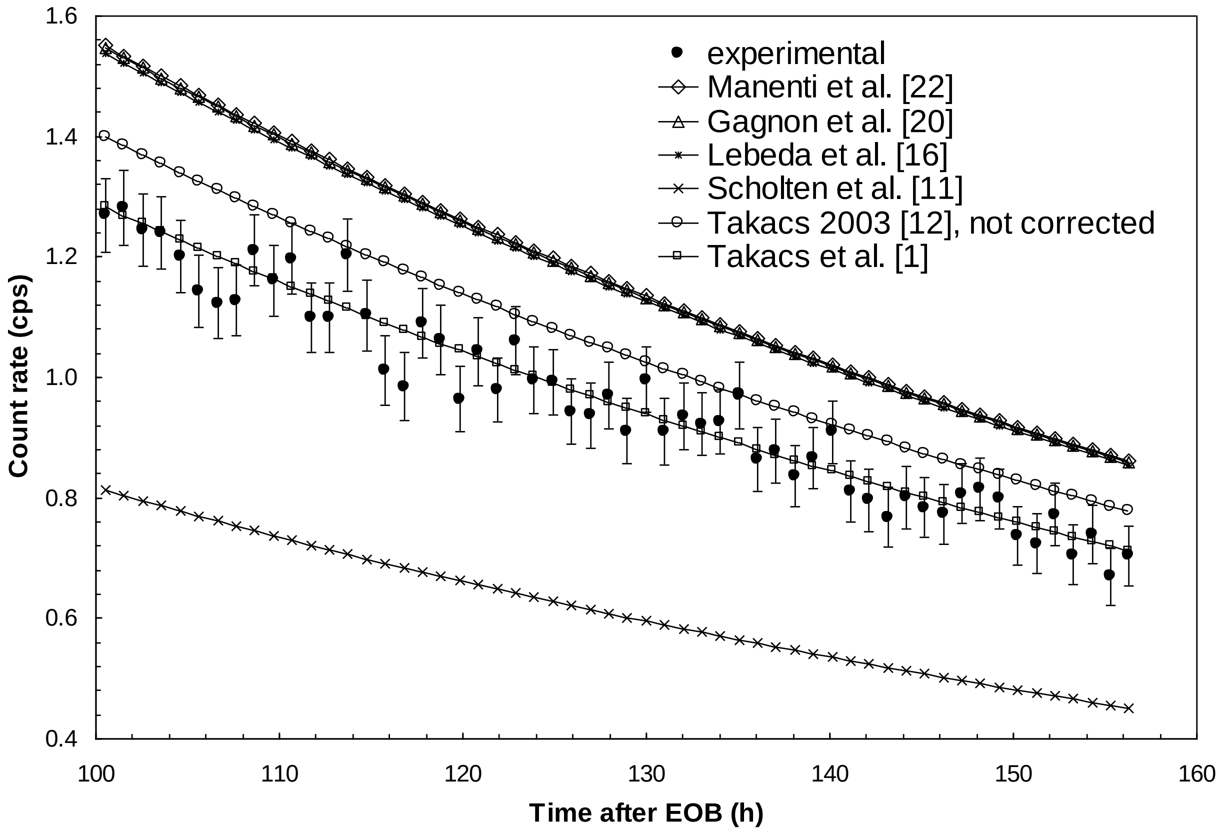

Figure 6. Experimental count rate measured for long cooling time for the E_{γ} = 140.5 keV gamma-line in a thick Mo target irradiated with a 17.9 MeV proton beam and calculated count rates using different experimental cross section data-sets.

In Fig. 7, the calculated count rates are presented relative to the experimental one, for the tested different experimental cross sections data-sets. Since the calculated points are normalised to the corresponding experimental count rate data points, they seem to be scattered. This behaviour is explained by the statistical uncertainty of the experimental data which is reflected as scattering of the normalised points. The scattering is more pronounced for points measured after long cooling time because the counting time was kept for 1 hour for each of the measured spectrum.

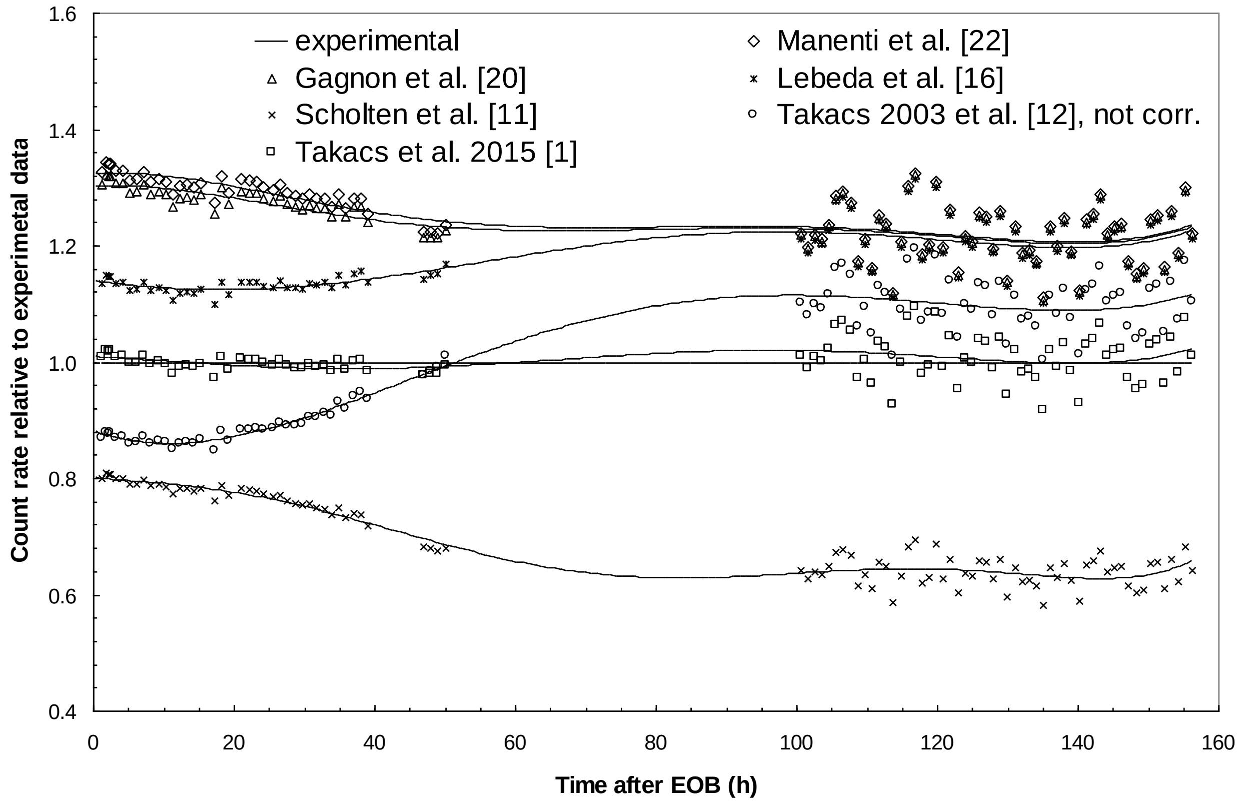

Figure 7. Calculated count rates relative to the experimental one measured for the E_γ = 140.5 keV gamma-line in a thick Mo target irradiated with a 17.9 MeV proton beam. Symbols: relative calculated count rates using different experimental cross section data-sets, dashed lines: polynomial trend lines of the date series.

Determining the direct cross section for the ^{100}Mo(p,2n)^{99m}Tc reaction only the E_γ = 140.5 keV gamma-line is available in the gamma-spectrum, which includes contribution from decay of ^{99}Mo. Therefore, separation of ^{99}Mo contribution is unavoidable. However, cross section of the ^{100}Mo(p,pn)^{99}Mo reaction can be determined by using the E_γ = 739.5 keV independent gamma-line. When the detector efficiency curve is determined properly and the right decay parameters are used for the cross section data deduction, the calculated relative count rate should represent a straight line with the value around 1.0 in Fig. 7. Testing the uncorrected cross section data of Takács et al. 2003 [12], one can find different deviation for ^{99m}Tc (short cooling time, Fig. 5) and ^{99}Mo (long cooling time Fig. 6). Since those data were evaluated by using early decay data and the deduced cross section for the ^{100}Mo(p,pn)^{99}Mo reaction was too high, after separating the ^{99}Mo contribution, the cross section for the ^{100}Mo(p,2n)^{99m}Tc reaction becomes too low. Accordingly, shifts of the calculated relative count rates are reflected in Fig. 7, but the results are symmetrical for the value 1.0. Testing the cross section data reported by Gagnon et al. [20] and Manenti et al. [22] similar shifts can be observed for both datasets. Their cross section for the ^{100}Mo(p,pn)^{99}Mo reaction is a little low therefore after subtraction of the contribution to the E_γ = 140.5 keV gamma-line the deduced cross section for the ^{100}Mo(p,2n)^{99m}Tc reaction becomes high. The calculated relative count rate behaves accordingly. Additionally, their values are symmetrical around 1.3 in Fig. 7, which indicates presence of systematic error that influences the derived cross sections both for ^{99m}Tc and ^{99}Mo in the same way. These can be systematic deviation of the beam intensity (may originate from use of other monitor reaction), target thickness, target composition or detector efficiency. Cross section data of Lebeda et al. [16] also result in a slightly different shifts for the two parts of the calculated count rate curve, which can be observed in Fig. 5 (deviation from the measured count rate is smaller than those for Gagnon et al. and Manenti et al.) and Fig. 6 (deviation from the measured count rate is similar to those for Gagnon et al. and Manenti et al.), indicating the presence of different sources of systematic errors in the measurement for the ^{99m}Tc and ^{99}Mo cross sections. Their calculated relative count rate data in Fig. 7 are around 1.2 and also indicating higher cross section for the ^{100}Mo(p,pn)^{99}Mo reaction and consequently lower values for the ^{100}Mo(p,2n)^{99m}Tc reaction. The consequence of the too low cross sections for both of the reactions reported by Scholten et al. [11] is clearly seen in Fig. 7 as a shift of the calculated relative count rate curve to around 0.7. It also can be concluded that the reported cross section for the ^{100}Mo(p,pn)^{99}Mo reaction was too low, therefore after separating its contribution to the E_γ = 140.5 keV gamma-line the deduced cross section for the ^{100}Mo(p,2n)^{99m}Tc reaction becomes relatively too high.

Conclusion

The primary experimental data, the count rate of the E_γ = 140.5 keV gamma-line, was measured on a thick Mo target irradiated with E_p = 17.9 MeV nominal proton beam and compared with the values estimated theoretically using the latest nuclear decay data and experimental cross section data for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions. Both the measurements and the calculation were repeated several times to be able to follow the decay of both ^{99m}Tc and ^{99}Mo radionuclides. Comparison of the experimental data set with the results of model calculation can validate the cross section data used in the calculation upon agreement. The trend of possible deviation from the experimental values can be used to judge the type of the possible systematic errors involved in the cross section data used in the calculation.

Since determining the experimental count rates effectively does not require any decay data; therefore this type of experiment can also give confirmation of the decay parameters used in the model calculation of the count rate.

Agreement between the experimentally determined and theoretically estimated count rates can confirm both the cross section values for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions and the applied decay data for the two radionuclides used in the calculation.

Acknowledgements

This work was carried out in the frame of the standing HAS-JSPS (Hungary–Japan) bilateral exchange agreement. The authors acknowledge the support of the respective institutions in providing technical support and use of experimental facilities. (Contract No.: NKM-89/2014).

The work also was partly supported by the International Atomic Energy Agency as part of a Co-ordinated Project: "Accelerator-based Alternatives to Non-HEU production of Mo-99/Tc-99m" Contract No.:17018/R0

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