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Experimental cross-sections for proton-induced nuclear reactions on ^{nat}Mo

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ABSTRACT

In the framework of the Co-ordinated Research Project of the IAEA, we measured in detail cross-sections of the nuclear reactions ^{nat}Mo(p,x)^{93g}Tc, ^{93m}Tc, $^{93m+g}$Tc, ^{94g}Tc, ^{94m}Tc, ^{95g}Tc, ^{95m}Tc, $^{96m+g}$Tc, ^{97m}Tc, ^{99m}Tc, ^{90}Mo, ^{93m}Mo, ^{99}Mo, ^{88g}Nb, ^{88m}Nb, ^{89g}Nb, ^{89m}Nb, $^{90m+g}$Nb, $^{90m+g}$Nb^{cum}, ^{91m}Nb, ^{92m}Nb, ^{95g}Nb, ^{95m}Nb, $^{95m+g}$Nb, ^{96}Nb, $^{97m+g}$Nb, $^{88m+g}$Zr^{cum} and $^{89m+g}$Zr^{cum} in the energy range of 6.9–35.8 MeV. The data for formation of ^{97m}Tc, ^{88g}Nb, ^{88m}Nb and ^{89m}Nb are reported for the first time. The obtained results were compared to the prediction of the nuclear reaction model code TALYS adopted from the TENDL-2015 library and to the previously published cross-sections. The thick target yields for all the radionuclides were calculated from the measured data. We suggest recommended cross-sections and thick target yields for the ^{100}Mo(p,2n)^{99m}Tc, ^{100}Mo(p,x)^{99}Mo and ^{nat}Mo(p,x)$^{96m+g}$Tc nuclear reactions deduced from the selected experimental data.

1. Introduction

The current data base contains many experimental data on the proton-induced reactions on natural molybdenum [1–22]. Cross-sections for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,x)^{99}Mo reactions attracted attention of the wider scientific community due to the recent severe crisis in the supply of ^{99}Mo/^{99m}Tc generators [23,24], because direct cyclotron production of ^{99m}Tc represents a promising alternative to reactor-produced ^{99}Mo/^{99m}Tc generators [24–28]. Besides that, elemental cross-sections for the reaction ^{nat}Mo(p,x)$^{96m+g}$Tc are relevant for proton beam monitoring [7,22], the reaction ^{nat}Mo(p,x)^{95m}Tc provides tracer of ^{99g}Tc [29] and data for formation of ^{93g}Tc, ^{93m}Tc, $^{93m+g}$Tc, ^{94g}Tc, ^{94m}Tc, ^{95g}Tc, ^{97m}Tc, ^{90}Mo, ^{93m}Mo, ^{88g}Nb, ^{88m}Nb, ^{89g}Nb, ^{89m}Nb, $^{90m+g}$Nb, ^{91m}Nb, ^{92m}Nb, ^{95g}Nb, ^{95m}Nb, $^{95m+g}$Nb, ^{96}Nb, $^{97m+g}$Nb, $^{88m+g}$Zr^{cum} and $^{89m+g}$Zr^{cum} are of interest for testing the nuclear reaction model codes, estimating activation of parts exposed to proton beams and for the thin layer activation (TLA) [30].

In spite of a large number of experiments focused on proton activation of natural molybdenum, the available data are often scattered and inconsistent. It is true particularly for formation of ^{99m}Tc, but also in the cases, where no problems like interferences or substantial nuclear decay data unreliability occur, e.g. ^{99}Mo [22]. Cross-section data for several relevant radionuclides like ^{97m}Tc are missing at all.

In the framework of the Coordinated Research Project of International Atomic Energy Agency (IAEA) "*Nuclear Data for Charged-Particle Monitor Reactions and Medical Isotope Production*", we were charged with providing new experimental data for formation of ^{99m}Tc and ^{99}Mo on ^{100}Mo, as well as for the ^{nat}Mo(p,x)$^{96m+g}$Tc monitoring reaction. We have, therefore, measured thoroughly elemental activation cross-sections for the ^{nat}Mo(p,x)^{93g}Tc, ^{93m}Tc, $^{93m+g}$Tc, ^{94g}Tc, ^{94m}Tc, ^{95g}Tc, ^{95m}Tc, $^{96m+g}$Tc, ^{97m}Tc, ^{99m}Tc, ^{90}Mo, ^{93m}Mo, ^{99}Mo, ^{88g}Nb, ^{88m}Nb, ^{89g}Nb, ^{89m}Nb, $^{90m+g}$Nb, $^{90m+g}$Nb^{cum}, ^{91m}Nb, ^{92m}Nb, ^{95g}Nb, ^{95m}Nb, $^{95m+g}$Nb, ^{96}Nb, $^{97m+g}$Nb, $^{88m+g}$Zr^{cum} and $^{89m+g}$Zr^{cum} reactions and compared them with the previously published data.

Since new abundances of stable Mo isotopes (^{92}Mo 14.53%, ^{94}Mo 9.15%, ^{95}Mo 15.84%, ^{96}Mo 16.67%, ^{97}Mo 9.60%, ^{98}Mo 24.39% and ^{100}Mo 9.82%) have been recently released [31], we used them for conversion of elemental to isotopic cross-sections for reactions on ^{100}Mo. The converted cross-sections adopted from the publications that used the former abundances (^{92}Mo 14.84%, ^{94}Mo 9.25%, ^{95}Mo 15.92%, ^{96}Mo 16.68%, ^{97}Mo 9.55%, ^{98}Mo 24.13% and ^{100}Mo 9.63% [32]) were also corrected for the new abundance of ^{100}Mo before comparing them with our results.

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2. Experimental

2.1. Target and irradiation

Two stacks of foils, each consisting of ten ^{nat}Mo foils (99.9%, 19.3 μm thick, Goodfellow, Great Britain) interleaved with ten ^{nat}Cu foils (99.9%, 10.6 μm thick, Goodfellow, Great Britain), eleven ^{nat}Ti foils (99.6%, 11.0 μm thick, Alfa Aesar, USA) and a few Cu degraders (55.9 and/or 10.6 μm thick) were irradiated on the external proton beam of the cyclotron U-120 M in the Nuclear Physics Institute of the CAS. Both stacks were located in a Faraday-cup like target holder and irradiated for one hour. The entrance beam energy was set by positioning of an extracting carbon foil and its value was deduced from the precisely measured beam orbit position [33]. The average beam energy in the center of each foil was calculated using the code SRIM [34]. The current of the collimated beam was kept constant during the irradiation, recorded each second and integrated over the bombardment time t_b.

2.2. Activity measurement

After the end of bombardment (EOB), the stacks were immediately dismounted and the activity of radionuclides in each foil was measured using an energy and efficiency calibrated HPGe detector (GMX45-Plus, Ortec) coupled with the DSPEC jr 2.0 integrated gamma spectrometer (Ortec). The energy and efficiency calibration of the spectrometer was performed using a set of standards (^{241}Am, ^{152}Eu, ^{137}Cs, ^{133}Ba and ^{60}Co) provided by the Czech Institute of Metrology with combined standard uncertainties ranged from 0.4% to 1.0%. Detection efficiency was determined for the sample-detector distances 150, 200, 400, 600, 1000 and 1600 mm. The whole energy range was fitted by a polynomial of the 5th degree:

$$\ln \eta = \sum_{n=0}^{5} a_n \ln E^n, \tag{1}$$

where η is detection efficiency, a_n is polynomial coefficient and E is energy of the photopeak. For the gamma ray energies exceeding 240 keV, a linear fit was used providing practically the same efficiencies as the polynomial fit.

Decay data (half-lives, gamma ray energies and intensities) of the radionuclides were adopted from the data base NuDat2 [35] and for ^{99m}Tc and ^{99}Mo from Ref. [36]. Nuclear reaction energies and thresholds were calculated using Q-calc [37]. The data relevant to the formation and quantification of the activation products are summarized in Table 1.

Each Mo foil was measured five to eight times in order to optimize conditions for quantification of radionuclides with various half-lives and activities, and to see consistency of the applied interference corrections (see Section 2.3). The gamma ray spectra were evaluated in the program DEIMOS32 [38].

2.3. Interference corrections

Table 1 indicates several interferences that affect activity measurement of several radionuclides born in the irradiated target foils. The most complex situation is represented by the set of corrections for determining the ^{99m}Tc activity born directly in the ^{100}Mo(p,2n) reaction. As thoroughly discussed earlier [15,22,28], the 140.51 keV net peak area is to be corrected for:

- Contribution of ^{99m}Tc born from ^{99}Mo during and after irradiation.
- Contribution of the 140.51 keV gamma line emitted directly in ^{99}Mo decay.
- Contribution of 141.18 keV gamma line from the decay of ^{90}Nb produced directly and due to the decay of ^{90}Mo (for E_p > 22 MeV).

The corrections were performed as described in Ref. [15]. We also paid attention to other interferences that occurred in the measurements either due to accidental proximity of the two gamma lines or due to emission of the same energy gamma lines from the nuclei decaying to the same product (^{95}Nb and ^{95}Tc, ^{96}Nb and ^{96g}Tc or ^{94m}Tc and ^{94g}Tc). Activity of the interference-affected radionuclide was then calculated with use of other non-interfering gamma line (if available) or of significant difference in the half-lives or in the gamma line intensities of the interfering radionuclides.

If both cumulative cross-section σ_{cum} of the longer-lived daughter radionuclide and shorter-lived parent cross-section σ_1 could be measured, we deduced the cross-section for the direct formation of the daughter radionuclide σ_2 from the following formula:

$$\sigma_{cum} = \frac{\lambda_1}{\lambda_1 - \lambda_2} f \sigma_1 + \sigma_2, \tag{2}$$

where λ_1 and λ_2 are decay constants of the parent and daughter radionuclides and f is transition probability of the parent to the daughter radionuclide. The same formula was used to derive prediction of the cumulative cross-sections from the cross-sections for the parent and the daughter radionuclide formation in various contributing nuclear reaction channels provided in TENDL-2015.

2.4. Calculation of the cross-sections, their uncertainties, thick target yields and prediction of excitation functions

Elemental and cumulative elemental cross-sections were calculated from the activation formula:

$$\sigma = \frac{P_\gamma}{I_\gamma \eta t_m} \frac{\lambda t_r}{1 - e^{-\lambda t_r}} e^{\lambda t_c} \frac{Aze}{d\rho N_A I (1 - e^{-\lambda t_b})}, \tag{3}$$

where σ is cross-section for formation of a radionuclide at the beam energy in the foil's center (cm^2), P_γ is net peak area of the gamma line selected for the quantification, I_γ is its intensity per decay and η its detection efficiency, t_m is real time of the measurement (h), t_r is live time of the measurement (h), t_c is time elapsed between the EOB and start of the measurement (h), A is atomic weight of the foil's material (g/mol), z is proton charge ($z = 1$), e is elementary charge (1.602177×10^{-19} C), d is thickness of the foil (cm), ρ is density of the foil's material (g/cm^3), N_A is Avogadro's number (6.022137×10^{23} mol^{-1}), I is beam current (A), λ is decay constant of the radionuclide (h^{-1}) and t_b is irradiation time (h).

The total uncertainty of the measured cross-section is obtained by summing up the following uncertainties in quadrature:

- Detection efficiency for a gamma line selected for the activity calculation (ca 3%).
- Emission probability of a gamma line selected for activity calculation (usually < 5%).
- Net peak area of a gamma line selected for activity calculation (< 25%, mostly < 2%).
- Beam current (< 6%).
- Foil's thickness (< 2%).

As ^{99m}Tc and ^{99}Mo are produced solely in the reactions on ^{100}Mo [6], the measured elemental cross-sections were converted to isotopic cross-sections on ^{100}Mo.

Thick target yields for all the radionuclides were deduced from the obtained experimental data by integrating the fitted