TWO-CHANNEL MODE OF Mo-99 PRODUCTION AT AN ELECTRON ACCELERATOR

A.N. Dovbnya, V.V. Mytrochenko, V.I. Nikiforov, S.A. Perezhogin, V.A. Shevchenko, B.I. Shramenko, A.Eh. Tenishev, A.V. Torgovkin, V.L. Uvarov#, NSC KIPT, Kharkov, 61108, Ukraine

Abstract

High-energy bremsstrahlung radiation is the main source of isotopic target activation at an electron accelerator. The photoneutrons, generated concurrently, are generally considered as a background radiation that contributes insignificantly (mainly, in the form of impurities) to the activation. At the same time, the natural materials entering into photonuclear targets sometimes comprise a mixture of stable isotopes, the atomic-number difference of which equals 2. Thus, if the desired isotope has an intermediate mass, then at certain conditions, it can be produced on two target nuclei at once, via (γ,n) and (n,γ) channels. As an example, we investigate the possibility of increasing the yield of ^{99}Mo by means of its simultaneous production in ^{100}Mo(γ,n)^{99}Mo and ^{98}Mo(n,γ)^{99}Mo reactions.

The technique and the device have been developed to provide measurements of the ^{99}Mo yield from the natural molybdenum target as it is placed inside the neutron moderator and without the latter. Experiments were performed at the NSC KIPT accelerator LU-40m at electron energies ranging from 30 to 60 MeV. It is demonstrated that the use of the moderator gives nearly a 30% gain in the ^{99}Mo yield. The experimental results are in good agreement with the computer simulation data.

INTRODUCTION

In recent years, a lot of attention has been paid to the development of nonreactor methods for production of the main diagnostic isotope ^{99m}Tc. As one of the methods, we consider photonuclear technique with the use of the ^{100}Mo(γ,n)^{99}Mo→^{99m}Tc reaction [1,2]. A special feature of this technology lies in a preliminary conversion of the accelerated electron beam into the bremsstrahlung to be then acting on the target. In this case, the bremsstrahlung converter simultaneously is a quasiisotropic source of photoneutrons [3]. It is also known that natural molybdenum comprises a mixture of stable isotopes, including ^{100}Mo (9.63%) and ^{98}Mo (24.13%). Therefore, during exposure of the natural molybdenum target to a flux of mixed X,n-radiation one can expect, apart from the photonuclear channel, the production of ^{99}Mo owing to the reaction of radiative capture of neutron by ^{98}Mo.

Under ordinary conditions, the additional yield of ^{99}Mo makes no more than 1% due to the smallness of the ^{98}Mo(n,γ)^{99}Mo reaction cross-section for fast neutrons. At the same time, in the case of thermal neutrons, the cross-section substantially increases [4]. The present paper is concerned with the possibility of increasing the ^{99}Mo yield from the natural molybdenum target through the use of a neutron moderator in the target device.

EXPERIMENTAL SETUP

For comparison between the ^{99}Mo yields without and with the neutron moderator, two alternative designs of the target device have been developed.

Thus, to determine the ^{99}Mo yield taken separately in the photonuclear channel, the device shown in Fig. 1 was used. It includes the aluminum tube, which is axially symmetric to the electron beam and accommodates the converter C and the target T. To measure the total yield of ^{99}Mo in (γ,n) and (n,γ) channels under conditions of an increased flux of moderated neutrons, the tube comprising the converter and the target was placed inside the neutron moderator (Fig. 2).

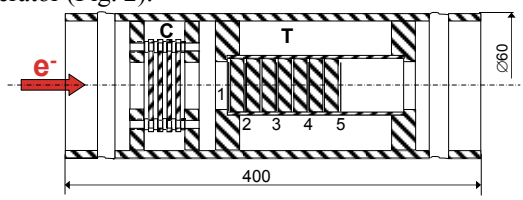

Figure 1: Target device without the neutron moderator

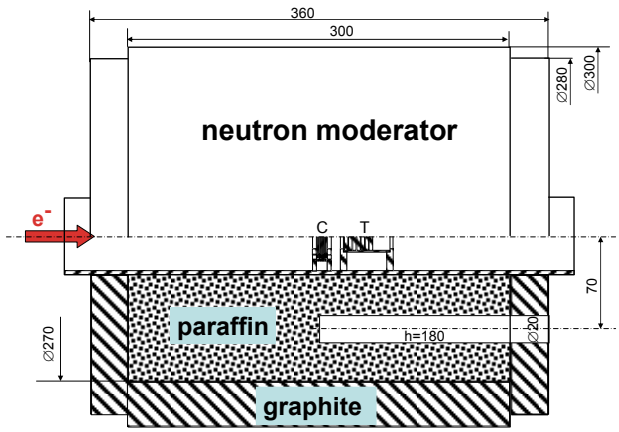

Figure 2: Target device with the neutron moderator

The converter C represents four tantalum plates, each 1 mm thick, separated by same-size air gaps to provide cooling. The target T includes seven molybdenum discs, each 19 mm in diameter and 3 mm in thickness. Between the discs there are five Mo foils, each 0.09 mm thick, intended for spectrometric analysis of isotope yields unaffected by gammas self-absorption (the sites of foil

uvarov@kipt.kharkov.ua

location in the target T are numerated in Fig. 1).

For each electron energy value, the irradiation of targets was carried out in both versions of the output devices. After "cooling" of targets, their activity was measured using the spectrometry complex CANBERRA In Spector 2000. Energy resolution of the spectrometer against 1332 keV-line is no worse than 1.74 keV at a relative uncertainty of activity measurements no more than 9%. A comparison between the yields of ^{99}Mo obtained with each target device version was made by normalizing the measured value to the ^{92}Mo(γ,2n)^{90}Mo reaction yield. Indeed, under given radiation conditions, the ^{90}Mo isotope is produced only in the photonuclear channel. It follows that the ^{90}Mo yield is independent of the spectral distribution of neutrons.

To measure the flux values of fast and moderated neutrons, a channel was bored in the body of the moderator (see Fig.2). A set of activation detectors from Au, Re, In and As was arranged in the channel. The distance from the converter to the detectors made up 70 mm.

SIMULATION

The yields of ^{99}Mo and ^{90}Mo in the photonuclear channel separately were calculated. The step-by-step technique with a modified transport code PENELOPE as the basis has been used to simulate microyield of these isotopes along all the photon trajectories in the target [5,6]. In simulation, real characteristics of the electron beam and the target device were taken into account. The reaction cross-sections (see Fig. 3) employed in the computations were taken from the database of ref. [7].

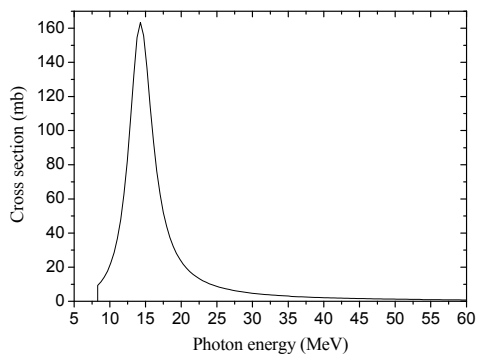

Figure 3: Cross-sections for the ^{100}Mo(γ,n)^{99}Mo (a) and ^{92}Mo(γ,2n)^{90}Mo (b) reactions

RESULTS AND DISCUSSION

In each run of the measurements, the spectra processing data of the first measuring foils (site 1 in Fig. 1) were used to calculate the ^{99}Mo-to-^{90}Mo activity ratio at the EOB. For a beam energy of 40 MeV, the A_0^{99}/A_0^{90} value was measured to be 2.5 in the device without the moderator, and 3.75 - on target irradiation in the device with the moderator (see the spectrum in Fig. 4).

Figure 5 presents the measured data on the distributions of ^{99}Mo and ^{90}Mo specific activities along the target axes at electron energies of 40 and 60 MeV, correspondingly. The solid curves show the simulation data on the production of the mentioned isotopes in the photonuclear channel separately. It can be seen that with an increase in the electron energy up to 60 MeV, the ^{99}Mo activity profile through the target depth changes. This can be explained by the fact that in passing through the converter, a part of beam electrons that has the energy higher than the reaction threshold, generates above-threshold photons immediately in the molybdenum target. This explanation is evidenced, in particular, by the fact that the shape of ^{90}Mo activity distribution has not practically changed with an increasing of electron energy. Really, the reaction of ^{90}Mo generation has the 22.8 MeV threshold, while in the ^{99}Mo case, the reaction threshold equals 8.3 MeV (see Fig. 3). Therefore, at electron energy of 60 MeV, it is the converter that still remains the only source of above-threshold photons for the ^{92}Mo(γ,2n)^{90}Mo reaction.

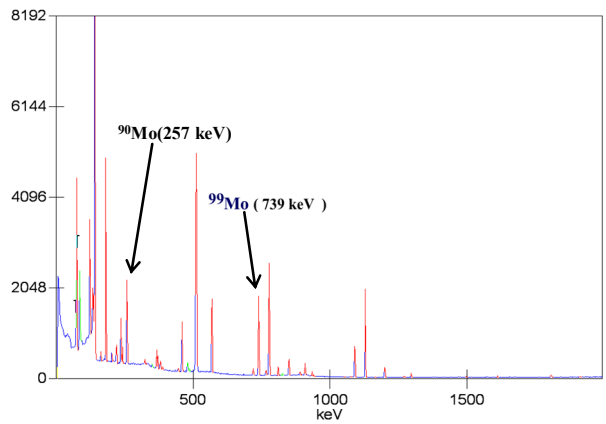

Figure 4: Gamma-spectrum of irradiated molybdenum

The neutron flux densities measured by different detectors in the region of their location appear to range from $3.4 \cdot 10^9$ to $2.8 \cdot 10^{10}$, n/cm^2·s·100μA. This spread in the values can be explained by the fact that in the computations we have used the data on the neutron capture cross-sections without consideration of the real energy spectrum of neutrons. Besides, on application of the moderator of the above-mentioned dimensions, at the site of detector location there is some quantity of epithermal neutrons, which may introduce an additional error.

The thermal neutron flux density at the site of Mo target location can be assessed on the basis of the difference between the ^{99}Mo activities produced at

irradiation with and without the moderator. The estimations have resulted in the following values: ~$1.0 \cdot 10^{11}$ at electron energy of 40 MeV and ~$1.3 \cdot 10^{11}$, n/cm^2·c·100 μA for 60 MeV.

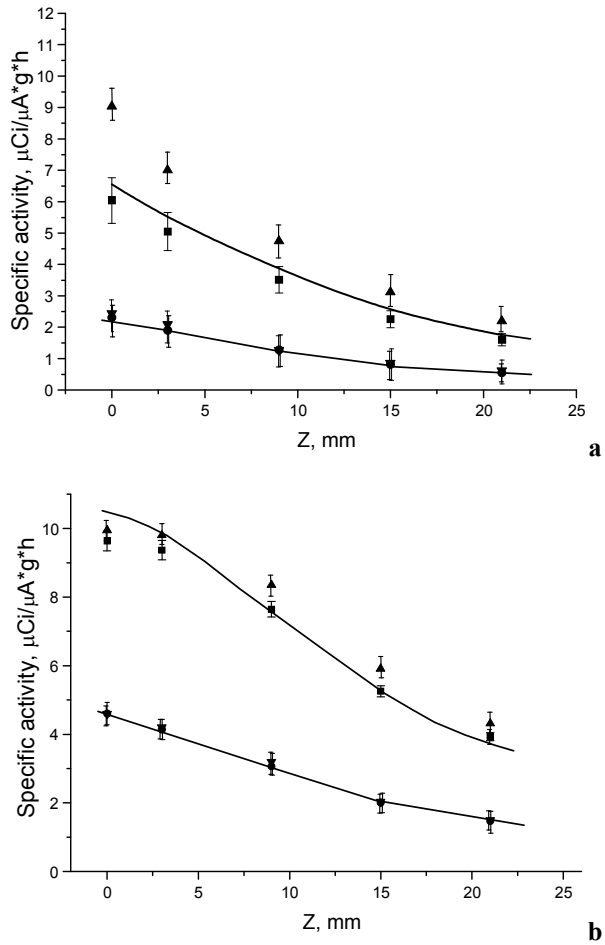

Figure 5: Specific activity distribution along Mo-target: **a** – E_0=40 MeV, **b** - E_0=60 MeV
(■ - Mo^{99} without moderator, ● - Mo^{90} without moderator, ▲ - Mo^{99} with moderator, ▼ - Mo^{90} with moderator)

CONCLUSION

The gain in the ^{99}Mo yield with the use of the given moderator attains 30% for an electron energy of 40 MeV. With a further rise in energy, the relative increase in activity determined by the ^{98}Mo(n,γ)^{99}Mo reaction decreases mainly because of the reduction in the relative fraction of moderated neutrons in the total flux of mixed X,n-radiation. This situation arises as a consequence of insufficient sizes of the employed moderator and reflector that are to provide the maximum thermal neutron flux on the target. So it can be expected that the optimization of the composition and geometry of the moderator would provide an additional increase in the ^{99}Mo yield.

Good agreement between the calculated and measured data on the photonuclear yield of ^{99}Mo and ^{90}Mo gives evidence for a correct description of the cross-sections and an adequate method of simulation of electron-photon cascades.

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