THE ^{89}Zr PHOTONUCLEAR PRODUCTION ON ^{nat}Mo AT BREMSSTRAHLUNG END-POINT ENERGIES OF 55…95 MeV

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The photoproduction of the ^{89}Zr nucleus on natural molybdenum was studied using the electron beam of the LUE-40 linac RDC "Accelerator" of NSC KIPT. Measurements were performed using the activation method and off-line γ-ray spectrometric technique. The production of ^{89}Zr in photonuclear reactions on ^{nat}Mo is discussed, considering three different accumulation channels. For the ^{nat}Mo(γ,*x*)^{89}Zr reaction, the experimental cumulative flux-averaged cross-section $\langle\sigma(E_{\gamma max})\rangle$ at the bremsstrahlung end-point energy range from 55 to 95 MeV was determined. The theoretical values of the flux-averaged cross-sections $\langle\sigma(E_{\gamma max})\rangle$ for the studied reaction were calculated using the cross-sections σ(*E*) from the TALYS1.96 code with default parameters.

PACS: 25.20.−x, 27.60.+q

INTRODUCTION

Modern theoretical models for describing nuclear reaction mechanisms (including direct, compound-nucleus, pre-equilibrium, quasi-deuteron processes, etc.) are currently under active development. Based on these advanced theoretical concepts, computational codes for simulating nuclear reaction cross sections are being continuously improved. Any modern computational code, such as TALYS, EMPIRE, ALICE, GNASH, and UNF, requires regular validation and updating. This requires reliable experimental data on σ(*E*) and/or $\langle\sigma(E_{\gamma max})\rangle$ cross-sections for multiparticle photoneutron (γ,*x*n) and photonuclear (γ,*x*n*y*p) reactions over a wide range of atomic masses and energies. Unfortunately, such information in international databases is currently unsystematic and fragmentary.

Very few experimental studies have been conducted on photonuclear reactions involving molybdenum isotopes with charged particles or small clusters in the output channel. Several works, for example [1–5], report cross-section data for the photoformation of niobium and zirconium isotopes. Note that studies of the formation of zirconium isotopes have been performed at energies up to 55 MeV.

In this work, a study of the ^{89}Zr production in photonuclear reactions on ^{nat}Mo at the bremsstrahlung end-point energy range of $E_{\gamma max}$ = 55…95 MeV was performed. The obtained experimental results expand the energy range of studies of the zirconium photoproduction from natural molybdenum. The obtained experimental results are compared with theoretical estimates performed with the cross-sections σ(*E*) from the TALYS1.96 code with default parameters.

EXPERIMENTAL PROCEDURE

The experiment was performed using the electron linear accelerator LUE-40 of the Research and Development Centre "Accelerator" of the National Science Center "Kharkiv Institute of Physics and Technology" of the National Academy of Sciences of Ukraine. The linac LUE-40 provides an electron beam with an average current $I_e \approx 4$ μA and full width at half maximum (FWHM) of energy spectrum $\Delta E_e/E_e \approx 1$ %. The range of initial energies of electrons is up to E_e = 100 MeV. A detailed description of the linac and its parameters is given in [6, 7].

An experimental complex for studying multiparticle photonuclear reactions, along with descriptions of the measurements and data processing methods, can be found, for example, in [8, 9]. The LUE-40 linear accelerator (top) and the exposure reaction chamber (bottom) are shown in Fig. 1.

Fig. 1. The LUE-40 linear electron accelerator at the RDC "Accelerator", NSC KIPT (top view). The output of the accelerator, the cylindrical aluminum absorber, and the irradiation chamber with inlet and outlet tubes (bottom view)

On the axis of the electron beam, there are a converter, an absorber, and a reaction chamber. The converter, made of tantalum metal, is a 20 x 20 mm plate with thickness *l* =

1.05 mm, and is attached to an aluminum absorber, shaped as a cylinder, with dimensions ∅ 100 × 150 mm. The thickness of the aluminum absorber was calculated to clean the beam of γ-quanta from electrons with energies up to 100 MeV.

For the experiment, targets were made of natural molybdenum, which were thin discs with a diameter of 8 mm and a thickness of ~0.11 mm, which are corresponded to a mass of ~57…60 mg. Natural molybdenum consists of seven stable isotopes, with isotope abundance, %: ^{92}Mo – 14.84, ^{94}Mo – 9.25, ^{95}Mo – 15.92, ^{96}Mo – 16.68, ^{97}Mo – 9.55, ^{98}Mo –24.13, ^{100}Mo – 9.63.

The target was placed in an aluminum capsule and delivered by a pneumatic transport system to the reaction chamber for irradiation and back to the measurement room to record the induced γ-activity of reaction products in the target substance.

The γ-quanta of the reaction products were detected using a Canberra GC-2018 semiconductor HPGe detector. Its efficiency was 20% relative to the NaI(Tl) scintillator with dimensions 3 inches in diameter and 3 inches in thickness at energy $E_γ$ = 1332 keV. The resolution FWHM is 1.8 keV for energy $E_γ$ = 1332 keV, and 0.8 keV for $E_γ$ = 122 keV. The dead time for γ-quanta detection varied between 0.1 and 5%. The absolute detection efficiency $ε(E_γ)$ for γ-quanta of different energies was obtained using a standard set of γ-ray sources: ^{22}Na, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{241}Am, ^{133}Ba.

The electron bremsstrahlung spectra were calculated using the open-source software code GEANT4.9.2, PhysList G4LowEnergy [10]. The real geometry of the experiment was used in calculations as well as the space and energy distributions of the electron beam were taken into account. The bremsstrahlung flux was monitored by the yield of the ^{100}Mo(γ,n)^{99}Mo reaction (the half-life $T_{1/2}$ of the ^{99}Mo nucleus is 65.94 (1) h) by comparing the experimentally obtained flux-averaged cross-section values $⟨σ(E_{γmax})⟩$ with the computation data $⟨σ(E_{γmax})⟩_{th}$. Details of the monitoring procedure can be found in [8, 9].

To study the ^{nat}Mo(γ,x)^{89}Zr reaction (the half-life $T_{1/2}$ of the ^{89}Zr nucleus is 78.41 (12) h), the yield for the γ-transition with energy $E_γ$ = 909.15 keV and intensity $I_γ$ = 99.04 (3)% was used. The metastable ^{89m}Zr level decays to the ground state of ^{89}Zr in 4.161 (10) min via internal transition *IT* (93.77 (12)%) and to the ^{89}Y nucleus via EC/β$^+$ (6.23 (12)%) processes, respectively.

When ^{nat}Mo is irradiated with high-energy photons, three nuclei-products can be formed in targets: ^{89}Mo, ^{89}Nb, and ^{89}Zr. However, the ^{89}Mo nucleus can lead to the formation of the ^{89}Zr nucleus through a decay chain: ^{89}Mo -β$^+$→ ^{89}Nb -β$^+$→ ^{89}Zr. Analogically, the ^{89}Nb nucleus decays to ^{89}Zr. So, for the studying of the formation of the ^{89}Zr nucleus on molybdenum, it is necessary to take into account three different channels of the accumulation of the ^{89}Zr nucleus. The nuclear spectroscopic data for nuclei produced in reactions from the database [11] are presented in Fig. 2.

The metastable state of the ^{89m}Mo nucleus decays to the ground state via internal transition with $T_{1/2}$ = 190(15) ms, the ground state of ^{89}Mo decays to ^{89}Nb via EC/β$^+$, $T_{1/2}$ = 2.11(10) min. The ^{89}Nb nucleus decays to the ^{89}Zr nucleus (ground state: EC/β$^+$, $T_{1/2}$ = 2.03(7) h; metastable state: EC/β$^+$, $T_{1/2}$ = 66 (2) min). The metastable state of the ^{89m}Zr nucleus decays with $T_{1/2}$ = 190(15) ms to the ^{89}Zr ground state via internal transition (93.77(12)%), and to the ground state of the ^{89}Y nucleus via EC/β$^+$ (6.23(12)%). The ground state of ^{89}Zr decays to stable ^{89}Y via EC/β$^+$ with $T_{1/2}$ = 78.41(12) h. Thus, both nuclei, ^{89}Mo and ^{89}Nb, are precursors of ^{89}Zr in the cascade: ^{89}Mo → ^{89}Nb → ^{89}Zr (the entire chain is converted to ^{89}Zr within several hours after irradiation). The contribution to the yield of the ^{89}Zr production depends on the reaction thresholds, cross-sections, and target cooling time.

The γ-radiation spectrum of a ^{nat}Mo target irradiated by a beam of bremsstrahlung γ-quanta with high end-point energy is a complex pattern. There are emission γ-lines of the reaction products of the ^{nat}Mo(γ,$ypxn$) reactions located on a background, which is formed as a result of Compton scattering of photons. As an example, in Fig. 3 γ-radiation spectrum of a ^{nat}Mo target with a mass of 57.862 mg after irradiation with $E_{γmax}$ = 92.50 MeV is shown.

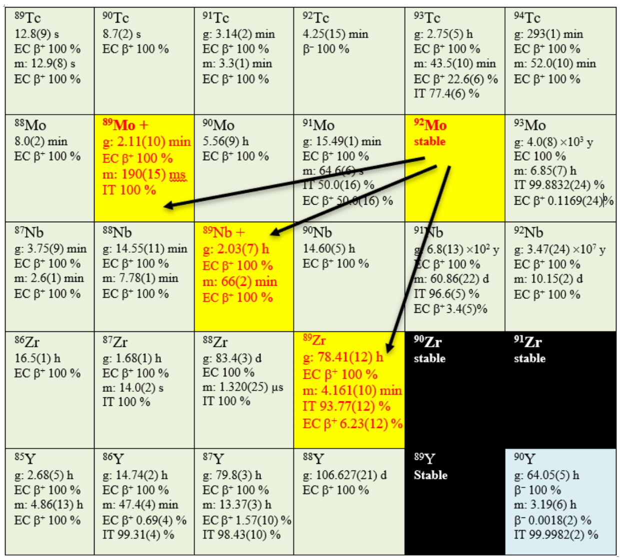

Fig. 2. Nuclear spectroscopic data for nuclei produced in reactions, taken from the database [11]

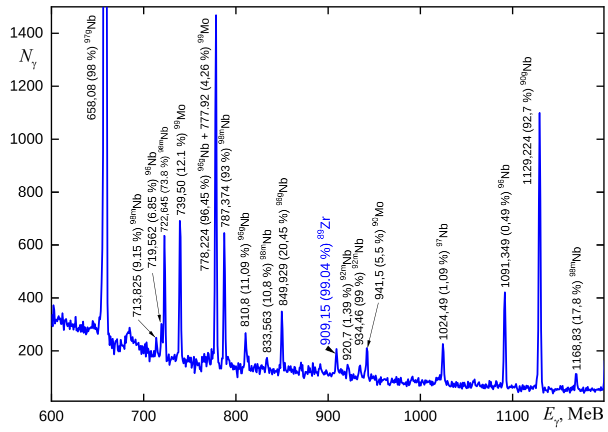

Fig. 3. Fragments of γ-ray spectrum in the energy range $600 \le E_γ \le 1200$ keV from the ^{nat}Mo target of mass 57.862 mg after irradiation of the bremsstrahlung γ-flux at $E_{γmax}$ = 92.50 MeV. Irradiation and measurement time were both 3600 s

CALCULATION OF CROSS-SECTIONS σ(*E*), YIELDS OF PHOTONUCLEAR REACTIONS *Y*($E_{\gamma max}$) AND FLUX-AVERAGED CROSS-SECTIONS ⟨σ($E_{\gamma max}$)⟩

Natural molybdenum consists of seven stable isotopes, all of which contribute to the formation of the final nucleus ^{89}Zr; several reactions with different thresholds are possible.

The ^{89}Zr nucleus can be produced in photonuclear reactions on molybdenum isotopes (reactions with the lowest thresholds are listed below):

^{92}Mo(γ,2pn)^{89}Zr – E_{thr} = 24.59 MeV;
^{92}Mo(γ,dp)^{89}Zr – E_{thr} = 22.36 MeV;
^{92}Mo(γ,3He)^{89}Zr – E_{thr} = 16.88 MeV;
^{94}Mo(γ,αn)^{89}Zr – E_{thr} = **14.04 MeV – minimum**;
^{95}Mo(γ,α2n) ^{89}Zr – E_{thr} = 21.41 MeV;
^{96}Mo(γ,α3n) ^{89}Zr – E_{thr} = 30.56 MeV;
^{97}Mo(γ,α4n) ^{89}Zr – E_{thr} = 37.38 MeV;
^{98}Mo(γ,α5n) ^{89}Zr – E_{thr} = 46.04 MeV;
^{100}Mo(γ,α7n) ^{89}Zr – E_{thr} = 60.26 MeV.

The ^{89}Nb nucleus can also be formed in photonuclear reactions (γ,*x*) on ^{nat}Mo (some reactions with the lowest thresholds include):

^{92}Mo(γ,p2n) ^{89}Nb – E_{thr} = 29.59 MeV;
^{92}Mo(γ,dn) ^{89}Nb – E_{thr} = 27.37 MeV;
^{92}Mo(γ,t) ^{89}Nb – E_{thr} = **21.1 MeV - minimum;**
^{94}Mo(γ,t2n) ^{89}Nb – E_{thr} = 38.88 MeV;
^{95}Mo(γ,t3n) ^{89}Nb – E_{thr} = 46.25 MeV;
^{96}Mo(γ,t4n) ^{89}Nb – E_{thr} = 55.4 MeV;
^{97}Mo(γ,t5n) ^{89}Nb – E_{thr} = 62.2 MeV;
^{98}Mo(γ,t6n) ^{89}Nb – E_{thr} = 70.9 MeV.

The ^{89}Mo nucleus can also be formed in photonuclear reactions (γ,*x*n) on ^{nat}Mo via some reactions:

^{92}Mo(γ,3n) ^{89}Mo – E_{thr} = **36.01 MeV - minimum;**
^{94}Mo(γ,5n) ^{89}Mo – E_{thr} = 53.76 MeV;
^{95}Mo(γ,6n) ^{89}Mo – E_{thr} = 61.1 MeV;
^{96}Mo(γ,7n) ^{89}Mo – E_{thr} = 70.3 MeV;
^{97}Mo(γ,8n) ^{89}Mo – E_{thr} = 77.1 MeV.

The cross-sections σ(*E*) of studied reactions for monochromatic photons were calculated using the TALYS1.96 code [12] for level density model *LD*1 and photon strength function *GSF*9 (default options):

*LD*1: Constant temperature + Fermi gas model, introduced by Gilbert and Cameron,

*GSF*9: Simplified Modified Lorentzian.

The obtained reaction cross-section ^{nat}Mo(γ,*x*)^{89}Zr is calculated as the sum of the cross-sections on all stable Mo isotopes taking into account their abundance. The reaction cross sections ^{nat}Mo(γ,*x*n)^{89}Mo and ^{nat}Mo(γ,*x*)^{89}Nb were calculated similarly.

The cross-sections σ(*E*) are averaged over the bremsstrahlung flux *W*(*E*,$E_{\gamma max}$) in the energy range from the threshold of the corresponding reaction E_{thr} to the maximum energy of the bremsstrahlung spectrum $E_{\gamma max}$. As a result, flux-averaged cross-section values were obtained:

$$\left\langle \sigma\left(E_{\gamma\max}\right)\right\rangle = \frac{\int_{E_{th}}^{E_{\gamma\max}} \sigma(E)\, W\left(E, E_{\gamma\max}\right) dE}{\int_{E_{th}}^{E_{\gamma\max}} W\left(E, E_{\gamma\max}\right) dE}. \qquad (1)$$

To calculate the total flux-average cross-section for the formation of the final nucleus ^{89}Zr on ^{nat}Mo, we summed the total cross-sections for three reactions: ^{nat}Mo(γ,*x*n)^{89}Mo, ^{nat}Mo(γ,*x*)^{89}Nb, and ^{nat}Mo(γ,*x*)^{89}Zr. An averaging over the minimum threshold E_{thr} = 14.04 MeV was then performed. This was necessary for comparison with the experimental data.

Theoretical cross-sections σ(*E*) and ⟨σ($E_{\gamma max}$)⟩ for the formation of the ^{89}Mo, ^{89}Nb, ^{89}Zr nucleus on ^{nat}Mo are presented in Fig. 4,a,b.

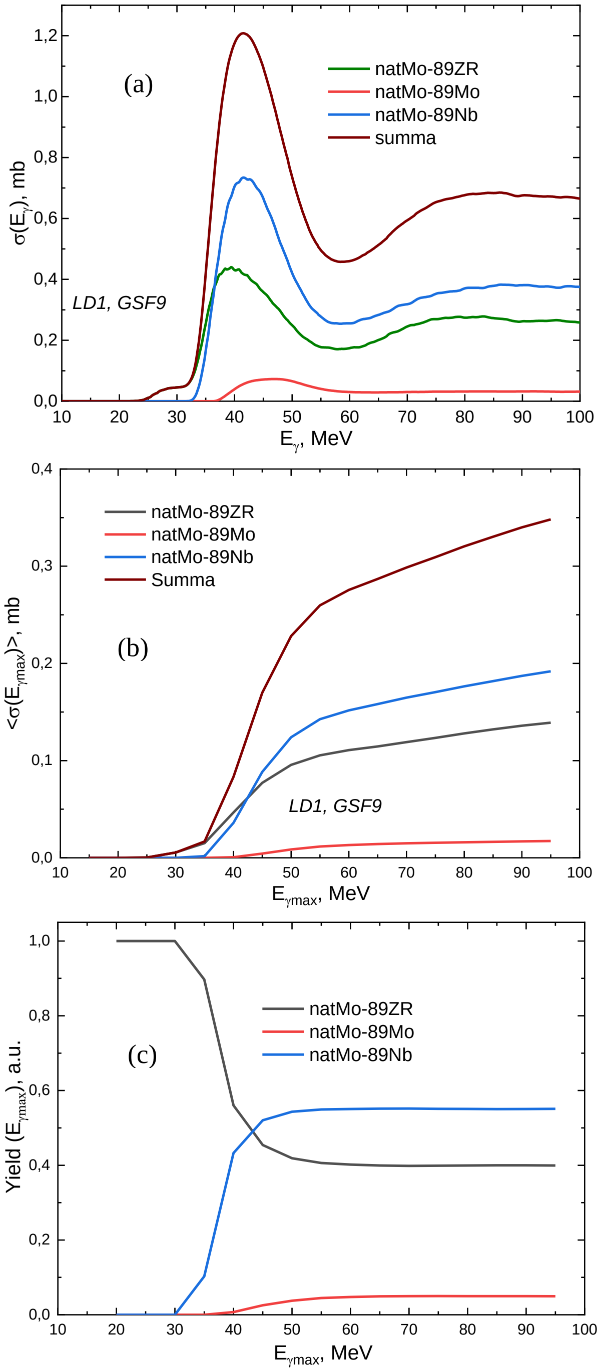

Fig. 4. Theoretical cross-sections σ(E) and ⟨σ($E_{\gamma max}$)⟩ for the formation of the ^{89}Mo, ^{89}Nb, ^{89}Zr nucleus on ^{nat}Mo. Normalized reaction yield Y_i($E_{\gamma max}$) of production of the nuclide ^{89}Mo, ^{89}Nb, ^{89}Zr. The calculations were performed in the TALYS1.96 code for options LD1, GSF9 with a minimum threshold E_{thr} = 14.04 MeV

For the estimation of the contribution of a reaction on natural molybdenum via three different channels (^{89}Mo, ^{89}Nb, ^{89}Zr) in the total production of a studied nuclide

^{89}Zr, the normalized reaction yield $Y_i(E_{\gamma max})$ was used in the same way as [13] (see Fig. 4 (c)).

EXPERIMENTAL RESULTS

The cross-sections for the formation of the ^{89}Zr nucleus in the reaction on ^{nat}Mo(γ,*x*)^{89}Zr can be determined from direct measurements of the number of counts of γ-quanta ΔA in the full absorption peak at an energy of 909.04 keV. To calculate the experimental values $\langle\sigma(E_{\gamma max})\rangle$ the following expression was used:

$$\left\langle \sigma\left(E_{\gamma\max}\right)\right\rangle = \frac{\lambda\Delta A}{\varepsilon N_x I_\gamma \Phi\left(E_{\gamma\max}\right)\left(1-e^{-\lambda t_{irr}}\right)e^{-\lambda t_{cool}}\left(1-e^{-\lambda t_{meas}}\right)}, \quad (2)$$

where $\Phi\left(E_{\gamma\max}\right) = \int_{E_{thr}}^{E_{\gamma\max}} W\left(E, E_{\gamma\max}\right)dE$ – the integrated bremsstrahlung photon flux in the energy range from the reaction threshold E_{thr} = 14.04 MeV to $E_{\gamma max}$, N_x is the number of studied atoms (including seven isotopes – 92-98, and 100), I_γ – the intensity of the analyzed γ-quanta, ε – the absolute detection efficiency for the analyzed γ-quanta energy, λ is the decay constant (ln2/$T_{1/2}$), t_{irr}, t_{cool} and t_{meas} are the irradiation time, cooling time and measurement time, respectively.

In the experiment, the cooling time was 3–6 days. With this cooling time, the nuclei-products ^{89}Mo and ^{89}Nb completely decayed into ^{89}Zr nuclei. In the calculation according to Eq. (2), the half-life for ^{89}Zr (78.41 (12) h) was used. Thus, the value $\langle\sigma(E_{\gamma max})\rangle$ corresponds to the effective cumulative cross-section for the formation of the ^{89}Zr nucleus on ^{nat}Mo.

The experimental values of the flux-averaged experimental cumulative cross-section $\langle\sigma(E_{\gamma max})\rangle$ for the formation of the ^{89}Zr nucleus on ^{nat}Mo were determined at the bremsstrahlung end-point energy of 55…95 MeV and shown in Fig. 5.

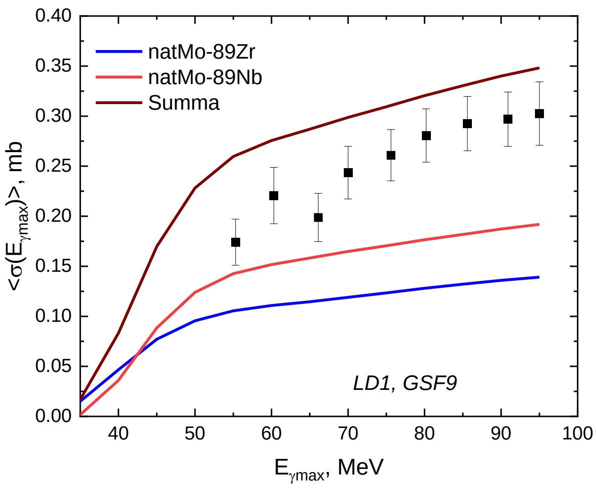

Fig. 5. The flux-averaged cumulative cross-section $\langle\sigma(E_{\gamma max})\rangle$ for the ^{89}Zr formation on ^{nat}Mo. Theoretical cross-section $\langle\sigma(E_{\gamma max})\rangle$ for the formation of the ^{89}Zr nucleus on ^{nat}Mo were calculated with the TALYS1.96 code for options LD1, GSF9, and using a minimum reaction threshold E_{thr} = 14.04 MeV

CONCLUSIONS

In the present work, the experiment was performed using an electron beam from the LUE-40 electron accelerator of RDC "Accelerator" of NSC KIPT and the γ-activation technique. The bremsstrahlung end-point energy range was $E_{\gamma max}$ = 55…95 MeV.

In this work, three different channels for the ^{89}Zr accumulation in photoreactions on ^{nat}Mo are identified and discussed. The value $\langle\sigma(E_{\gamma max})\rangle$ obtained in this way corresponds to the cumulative cross-section for the formation of the ^{89}Zr nucleus on ^{nat}Mo. The obtained cross-section $\langle\sigma(E_{\gamma max})\rangle$ for the ^{89}Zr photoproduction on natural Mo targets was determined for the first time.

The obtained experimental results expand the energy range of studies of the photoformation of zirconium from natural molybdenum.

The calculation of the flux-averaged cross-sections $\langle\sigma(E_{\gamma max})\rangle_{th}$ was performed using the cross-sections σ(*E*) for the studied reactions from the TALYS1.96 code with default parameters. A satisfactory agreement is observed between the experimental data and the TALYS1.96 default estimate.

This work has received funding through the MSCA4Ukraine project, which is funded by the European Union. Views and opinions expressed are, however, those of the author(s) only and do not necessarily reflect those of the European Union, the European Research Executive Agency, or the MSCA4Ukraine Consortium. Neither the European Union nor the European Research Executive Agency, nor the MSCA4Ukraine Consortium as a whole, nor any individual member institutions of the MSCA4Ukraine Consortium can be held responsible for them. This work was also supported by the Slovak Research and Development Agency under Contract No. APVV-24-0516, and the Slovak grant agency VEGA (Contract No. 2/0175/24).

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ФОТОУТВОРЕННЯ ЯДРА ^{89}Zr НА ^{nat}Mo ПРИ КІНЦЕВИХ ЕНЕРГІЯХ ГАЛЬМІВНОГО ВИПРОМІНЮВАННЯ 55…95 МеВ

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Фотоутворення ядра ^{89}Zr на природному молібдені досліджували на електронному пучку лінійного прискорювача ЛУЕ-40 НДК «Прискорювач» ННЦ ХФТІ. Вимірювання проводили з використанням методу наведеної активності та off-line гамма-спектрометрії. Обговорюється отримання ^{89}Zr у фотореакціях на ^{nat}Mo через три різні канали накопичення. Для реакції ^{nat}Mo(γ,*x*)^{89}Zr було визначено експериментальні кумулятивні усереднені поперечні перерізи $\langle\sigma(E_{\gamma max})\rangle$ у діапазоні кінцевої енергії гальмівного випромінювання 55…95 МеВ. Теоретичні значення усереднених поперечних перерізів $\langle\sigma(E_{\gamma max})\rangle$ для досліджуваної реакції розраховували за допомогою поперечних перерізів σ(*E*) з коду TALYS1.96 для параметрів за замовчуванням.