CROSS-SECTIONS FOR PHOTONUCLEAR REACTIONS $^{93}Nb(\gamma,n)^{92m}Nb$ AND $^{93}Nb(\gamma,n)^{92t}Nb$ IN THE END-POINT BREMSSTRAHLUNG ENERGIES 36...91 MeV

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The flux-weighted averaged over the energy range of bremsstrahlung spectrum from reaction threshold up to the maximum energy of γ-ray cross-sections <σ(*E*)> of the $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ photonuclear reactions were determined by the gamma-activation method within the end-point bremsstrahlung energies $E_{\gamma max}$ = 36…91 MeV. Activation of ^{93}Nb targets has been done by a bremsstrahlung flux using an electron beam at the linear accelerator LUE-40 at RDC "Accelerator" NSC KIPT. The γ-ray spectra of irradiated targets were registered using the HPGe detector with an energy resolution of 1.8 keV for the 1332 keV line ^{60}Co. To control the bremsstrahlung flux we used ^{nat}Mo witness-targets and a reaction cross-section of $^{100}Mo(\gamma,n)^{99}Mo$. Obtained experimental cross-sections <σ(*E*)> of the studied reactions are in good agreement with the theoretical values calculated within TALYS 1.9 code and the results of other authors. The averaged cross-sections <σ(*E*)> of the $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ reactions in the energy range 35...45 MeV and > 70 MeV were obtained for the first time.

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INTRODUCTION

Nuclear reactions with various particles in the input channel are an important source of information on both the mechanisms of the reaction and the properties of the excited states of atomic nuclei [1, 2]. Using γ-rays as a projectile has several advantages over other types of incident particles. Photonuclear reactions proceed due to electromagnetic interaction, the properties of which are well studied, and the mechanism of energy transfer from the incident γ-quantum to the studied nucleus is known. Another important factor is the transfer of small spin by γ-quanta into the target nucleus, which is important in the study of angular momenta of exciting levels of nuclei, fission fragments, and the creation of reaction models.

There are no high-intensity monoenergetic sources of γ-rays at present. The use of bremsstrahlung significantly complicates the procedure for determining the characteristics of nuclear reactions. First of all, it is necessary to calculate the density of bremsstrahlung γ-quanta flux corresponding to the actual experimental conditions. In experiments, it is possible to measure the integral characteristics of the reactions. It required additional mathematical processing of the results. Nevertheless, bremsstrahlung of electrons is an important instrument of modern nuclear physics despite the difficulties in the determination of the reaction's cross-sections.

Cross-sections of reactions induced by γ-quanta and neutrons are important for various physical applications, for example, in calculating the absorbed dose, in the technology of nuclear facilities construction, in radioactive waste disposal, and astrophysical nucleosynthesis.

Data on cross-sections of photonuclear reactions with multiple nucleon yields in the region of incident γ-quanta energies above the giant dipole resonance can be used to create power plants based on subcritical systems controlled by an electron accelerator.

At present the information on cross-sections of multi-particle photonuclear reactions and isomeric yield ratios is fragmentary. A review of existing results on these issues can be found, for example, in [3, 4].

As we know, niobium is a material that is used in alloys of zirconium shells of nuclear fuel elements used in the active zone of a subcritical system [5]. Therefore, the study of the cross-sections for multiparticle reactions on niobium is important, since these data are used in the calculation of the neutron flux in the reactor core. However, experimental data for photonuclear reactions on niobium were obtained in limited energy ranges. The experimental values of isomeric ratios in the $^{93}Nb(\gamma,4n)^{89m,g}Nb$ reactions averaged over the bremsstrahlung flux from the reaction threshold energy to the end-point bremsstrahlung energies $E_{\gamma max}$ = 60, 70 MeV can be found in [6]. In [7] authors studied the ^{93}Nb photonuclear reactions with multiple neutron emission with energies 12…16 and 45…70 MeV.

In the present work, we study of the photonuclear reactions $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ using the gamma-activation method at linear electron accelerator LUE-40 at RDC "Accelerator" NSC KIPT. The flux-weighted averaged cross-sections of these reactions were found in a wide range of end-point bremsstrahlung energies $E_{\gamma max}$ = 36...91 MeV.

1. EXPERIMENTAL SETUP AND PROCEDURE

To study the reactions $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ the gamma-activation method was used. This method allows one to the simultaneous study of different channels of photonuclear reactions. The method consists of several stages: irradiation (activation) of the target; registration of γ-ray spectra of reaction products; analysis of γ-ray spectra; calculation of the bremsstrahlung flux in the GEANT4 program code and calculation of cross-sections using the TALYS1.9 code. The experimental complex for photonuclear reactions studies is schematically presented in Fig. 1.

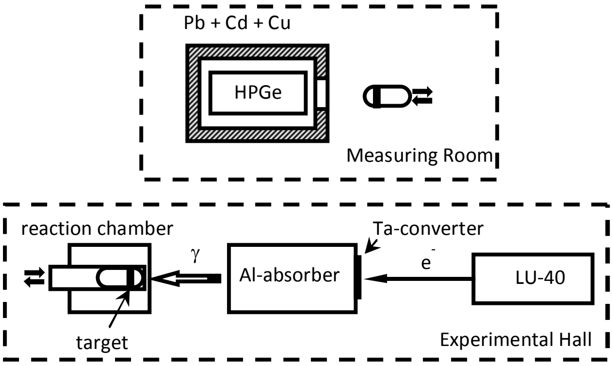

Fig. 1. The scheme of an experimental complex for photonuclear reactions studies by the gamma-activation method

The electron accelerator LUE-40 at RDC "Accelerator" NSC KIPT allows one to obtain electron beam within the energy range of 30...100 MeV with a step of $\Delta E_e \geq 1$ MeV [8, 9]. The average electron current in the experiment was 1...4 μA, and the transverse beam size was 3...7 mm diameter. The energy distribution of electrons depends on the energy of the electron beam and is $\Delta E/E$ = 0.8...1.8% at FWHM. The linac LUE-40 consists of an injector, two accelerating sections, and a beam transport system.

Electrons with an initial energy E_e hit the tantalum target with transverse dimensions of 20×20 mm and a thickness of 1.05 mm. The Ta-converter was fixed on an Al-absorber of electrons, which has a cylindrical shape with dimensions of Ø100×150 mm. The distance between the Ta-converter and the target was 248 mm. The samples of ^{93}Nb and natural molybdenum with Ø8 mm placed in an aluminium capsule were irradiated for 30 minutes. The masses of the high-purity niobium and molybdenum targets were ~ 80 mg and ~ 60 mg, respectively.

To transport the capsule with the sample between the measuring room and the place of irradiation a pneumatic transport system was used. This allowed the capsule to be delivered to the detector and then removed back to the place of irradiation remotely. The time of transportation of the capsule is no longer than 10 seconds, which makes it possible to measure the intensity of the γ-line of unstable nuclei with a half-life of several minutes.

The γ-quanta of the reaction products were detected using a Canberra GC-2018 semiconductor HPGe detector with the relative detection efficiency of 20%. The ratio of the accounts at the full absorption peak to the height of the Compton substrate is 55/1; the resolution FWHM is 1.8 keV for energy E_γ = 1332 keV and is 0.8 keV for E_γ = 122 keV. The dead time for γ-quanta detection varied between 0.1…5%.

The absolute detection efficiency $\varepsilon(E_\gamma,D)$ for γ-quanta of different energies depends on the distance D between the radiation source and the detector (surface of the protective cover). Therefore, for the present experiment, the detector was calibrated for a given distance of D = 64 mm using a standard set of γ-quanta sources: ^{241}Am, ^{133}Ba, ^{60}Co, ^{137}Cs, ^{22}Na, ^{152}Eu. Fig. 2 shows the experimental values of the efficiency ε (E_γ, 64 mm) and an analytical curve in the form of $\ln \varepsilon(E_\gamma) = \sum a_n \times (\ln E_\gamma)^n$, approximating this data [10].

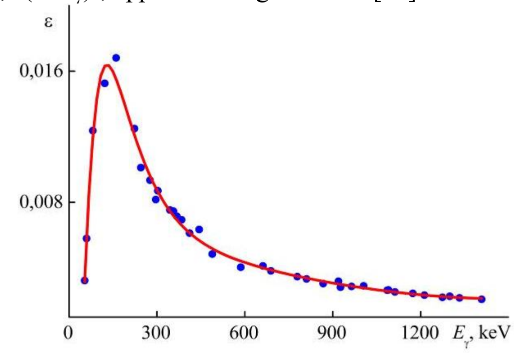

Fig. 2. The experimental values of the absolute detection efficiency $\varepsilon(E_\gamma,D)$ for distance D = 64 mm and the approximation curve in the form of $\ln \varepsilon(E_\gamma) = \sum a_n \times (\ln E_\gamma)^n$

The electron bremsstrahlung spectra were calculated using the open-source software code GEANT4 as in [11, 12]. The real geometry of the experiment was used in calculations also the space and energy distributions of the electron beam were taken into account. Electron energy distributions were approximated by an analytical function. The radius of the electron beam varied within R_e = 1.5…3.5 mm. Fig. 3 shows the calculated bremsstrahlung spectra, which were used in calculations of the gamma-flux irradiating the target.

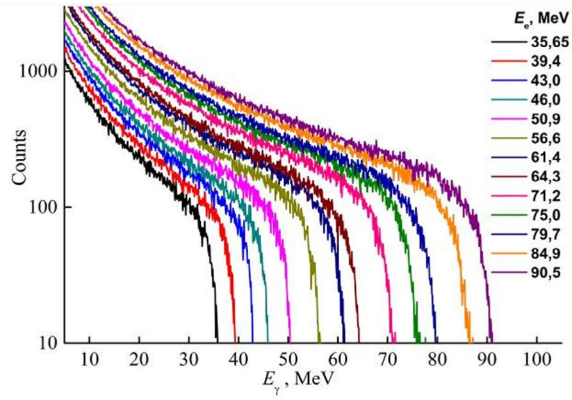

Fig. 3. The bremsstrahlung spectra incident on the targets. The energies of electrons E_e = 35.65…90.5 МэВ

Depending on the transverse dimensions of the target and on the displacement of the centre of the target to the electron beam axis, the bremsstrahlung flux on the target will be different [13]. The experimental error resulting from this can be significant: for example, the shift from centre of the target for 3 mm, decreases the flux of γ-quanta by more than 10%.

To take into account the variations of the flux we placed the ^{nat}Mo witness-target inside the capsule together with the target under investigation. The deviation of the calculated average cross-section from the experimental one found for the ^{100}Mo(γ,n)^{99}Mo reaction is taken into account when evaluating the experimental data for ^{93}Nb.

2. RESULTS AND DISCUSSION

The γ-quanta spectra from irradiated targets represent a complex picture of γ-lines from various reaction products ${}^{N}_{Z}A(\mathrm{r}, xnyp)^{N-x}_{Z-y}B$ together with complex background substrate (Compton scattering). As an example, Fig. 4 shows the γ-spectrum emitted by the activated target ^{93}Nb with a mass of 80 mg after irradiation for 30 min at $E_{\gamma\ max}$ = 84.5 MeV. The measurement time was 30 min.

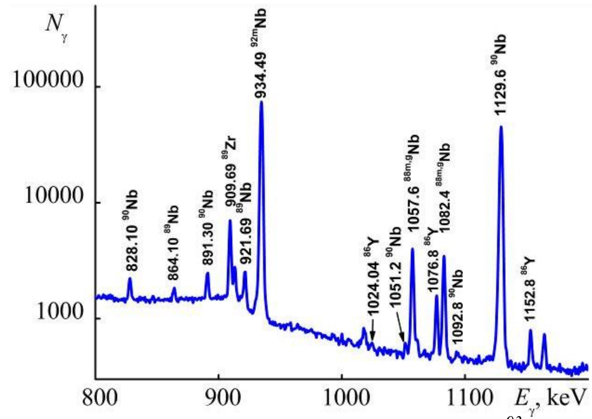

Fig. 4. γ-Spectrum of the irradiated target ^{93}Nb with mass 80 mg. The irradiation and measurement times were 30 min, $E_{\gamma max}$ = 84.5 MeV

Analyzing the γ-spectrum of the irradiated target, we deduced γ-lines from which the experimental activity of the selected reaction channel was estimated [13, 16]. Based on obtained data, the experimental values of the flux-weighted averaged cross-sections were determined as:

$$\langle \sigma(E) \rangle = \frac{\lambda \Delta A}{N_x I_\gamma \varepsilon \sum W(E)(1-e^{-\lambda T_i})e^{-\lambda T_c}(1-e^{-\lambda T_m})}, \quad (1)$$

where ΔA is the number of counts of γ-quanta in the full absorption peak (for the γ-line of the investigated reaction), N_x is the number of target atoms, $\sum W(E)$ is the bremsstrahlung flux summed from the reaction threshold value to the end-point bremsstrahlung energies $E_{\gamma\ max}$, I_γ – the branching 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_i, T_c and T_m are the irradiation time, cooling time and measurement time, respectively.

The calculated values of <σ(*E*)> were obtained by averaging the reaction cross-sections σ(*E*), obtained using the TALYS 1.9 code [13] with the calculated bremsstrahlung flux. Calculation of the average cross-section <σ(*E*)> as a function of energy in a given energy interval was done according to the formula:

$$\langle y(E) \rangle = \sum (y(E)W(E)) \Big/ \sum W(E), \quad (2)$$

where *σ(E)* – reaction cross-section; $\sum W(E)$ – bremsstrahlung flux density incident on the target. The summation is performedt in the energy interval from the reaction threshold to the maximum energy value E_e.

Resulting values of averaged cross-sections were compared to experimental data. In more detail, all calculation procedures for cross-section determination are described in [13-15].

The bremsstrahlung flux was monitored using the natural molybdenum witness-targets. Using the Eq. (1) the experimental averaged cross-section for the ^{100}Mo(γ,n)^{99}Mo reaction was calculated. We used γ-lines with an energy of E_γ = 739.5 keV, $T_{1/2}$ = 65.94 h, I_γ = 12.13% [1, 2]. A part of γ-spectrum emitted by the activated target from ^{nat}Mo with a mass of 57.7 mg is shown in Fig. 5.

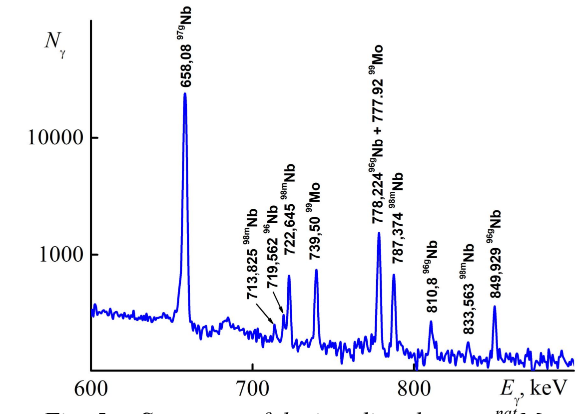

Fig. 5. γ-Spectrum of the irradiated target ^{nat}Mo with mass 57.7 mg. The irradiation and measurement times were 30 min, $E_{\gamma max}$ = 61.4 MeV

Calculated values of the averaged cross-section for the photonuclear reaction ^{100}Mo(γ,n)^{99}Mo were obtained using the Eq. (2) in two versions: using the cross-section of the reaction σ(*E*) calculated in the Talys 1.9 code (with default parameters), and the experimental values for this cross-section from [16], approximated by an analytical curve in the form of Lorentzian [17]:

$$y(E) = y_m \frac{(E \cdot \Gamma)^2}{(E^2 - E_m^2)^2 + (E \cdot \Gamma)^2}, \quad (3)$$

where σ_m is the value of the cross-section at the resonance maximum, E_m is the position of the resonance maximum, Γ is the resonance width. The values σ_m = 163.4 mb, E_m = 14.3 MeV, Γ = 3.9 MeV were taken from [17].

Fig. 6 shows the experimental cross-sections σ(*E*) for the photonuclear reaction of ^{100}Mo(γ,n)^{99}Mo from [16]. The curve in the form of Lorentzian approximating the experimental values is presented together with theoretical calculation performed in TALYS 1.9 code. The calculated cross-sections from TALYS 1.9 show satisfactory agreement with experiment with some underestimation trend, at the same time Lorentzian shows some overestimation for cross-section.

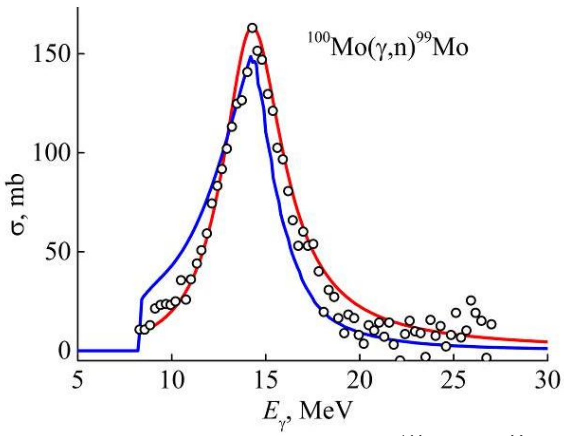

Fig. 6. The cross-section σ(E) of the ^{100}Mo(γ,n)^{99}Mo reaction, the points are experimental data from [16], the blue curve is the calculation result within Talys 1.9 code, the red curve is the Lorentzian approximation

Fig. 7 shows the experimental values of <σ(*E*)> for the $^{100}Mo(\gamma,n)^{99}Mo$ reaction obtained in this work together with the calculations of the <σ(*E*)> with Talys 1.9 code and using the Lorentzian approximation.

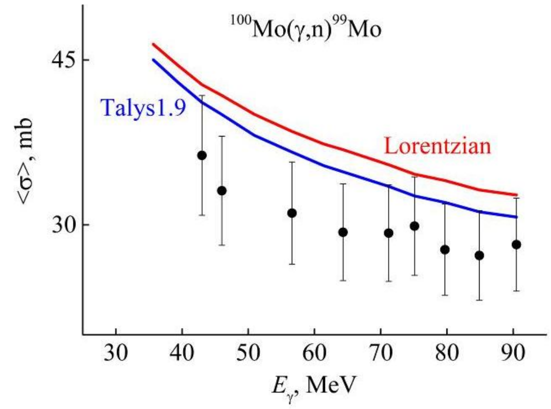

Fig. 7. The flux-weighted average cross-sections <σ(E)> of the $^{100}Mo(\gamma,n)^{99}Mo$ reaction. The points are experimental data, the blue curve is the calculation result with Talys 1.9 code, the red curve – with the Lorentzian approximation

The obtained values of <σ(*E*)> of this reaction are used to normalize the results determined with niobium. Normalization coefficients have the form of:

$k_{Talys} = \langle\sigma(E)\rangle_{Talys}/\langle\sigma(E)\rangle_{exp}$ and

$k_{Lorentz} = \langle\sigma(E)\rangle_{Lorentz}/\langle\sigma(E)\rangle_{exp}$.

The k_{Talys} values vary from 1.08 to 1.18 with an increase of the energy $E_{\gamma max}$, which explains the deviation of the real flux of bremsstrahlung from the calculated one. Results on averaged cross-sections within Talys 1.9 code and Lorentzian have a difference of 3…6% which increases with increasing of energy from 35 to 91 MeV.

Accordingly, the coefficients k_{Talys} and $k_{Lorentz}$, calculated for renormalizing experimental data will also differ for 3…6%. Since the target and the witness-target are of the same size and are in the same position relative to the axis of the electron beam, such consideration is correct. Fig. 8 shows two sets of values of the renormalization of the experimental cross-sections <σ(*E*)> for the reaction $^{93}Nb(\gamma,n)^{92m}Nb$.

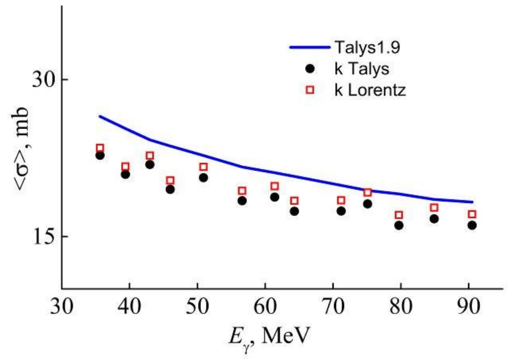

Fig. 8. The flux-weighted average cross-sections <σ(E)> of the $^{93}Nb(\gamma,n)^{92m}Nb$ reaction (points) and the calculation result with Talys 1.9 code (blue curve). The red points were normalized with the $k_{Lorentz}$, the black points – k_{Talys}

Figs. 9 and 10 show the experimentally obtained cross-sections <σ(*E*)> for the $^{93}Nb(\gamma,n)^{92m,t}Nb$ reactions, averaged over the bremsstrahlung spectrum in the energy range starting from the reaction threshold up the end-point bremsstrahlung energies $E_{\gamma max}$ = 36…91 MeV. Normalization was selected based on k_{Talys} values.

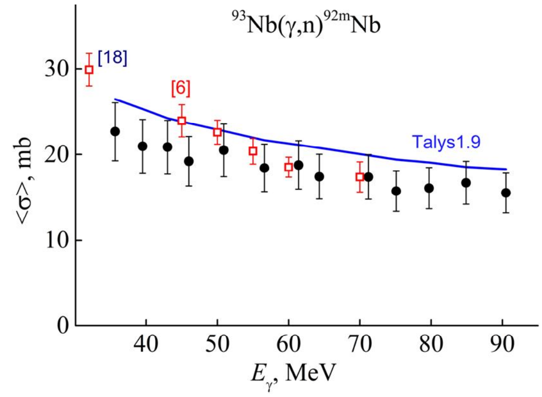

Fig. 9. The flux-weighted average cross-sections <σ(E)> of $^{93}Nb(\gamma,n)^{92m}Nb$ reaction. The blue line is the calculation with the Talys 1.9 code. The black points are data of this work, the red points – from [6, 18]

To determine the <σ(*E*)> of the reaction $^{93}Nb(\gamma,n)^{92m}Nb$, we used the γ-line with E_γ = 934.46 keV, $T_{1/2}$ = 10.15 days, I_γ = 99% [1, 2].

Total cross-section of the $^{93}Nb(\gamma,n)^{92}Nb$ reaction was obtained by multiplying the <σ(*E*)> value for the $^{93}Nb(\gamma,n)^{92m}Nb$ reaction by 0.552. This multiplication factor is based on the assumption of 55.2% contribution of the cross-section of the metastable state to the total cross-section of the $^{93}Nb(\gamma,n)^{92}Nb$ reaction at $E_{\gamma max}$ = 32 MeV [13, 18]. According to the calculations of the cross-sections for the ground and isomeric states made using Talys 1.9, this coefficient is valid in a wide range of γ-quanta energies.

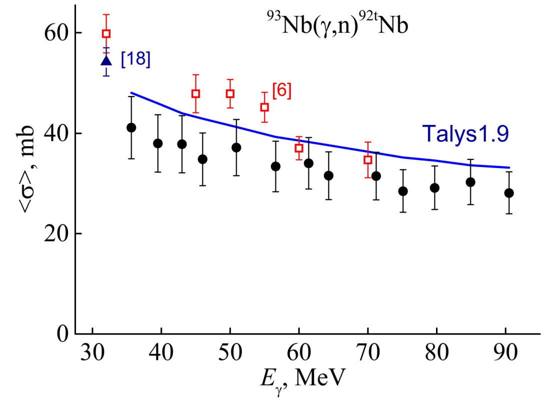

Fig. 10. The flux-weighted average cross-sections <σ(E)> of $^{93}Nb(\gamma,n)^{92t}Nb$ reaction. The blue line is the calculation with the Talys 1.9 code. The black points are data of this work, the red points and triangle – [6, 18]

The measured averaged cross-sections <σ(*E*)> of the $^{93}Nb(\gamma,n)^{92m}Nb$ reaction are in good agreement with the experimental results from [6] in the energy range $E_{\gamma max}$ = 55…70 MeV, still at lower energies the agreement is poor (see Fig. 9). In the case of the $^{93}Nb(\gamma,n)^{92t}Nb$ reaction, data from [6] at $E_{\gamma max}$ = 30...55 MeV are systematically higher than the average cross-sections obtained by our group and higher than results of calculations within Talys 1.9 code with default settings. Results from [6] are also systematically higher than data from [18] for this reaction.

Comparison of averaged cross-sections for the reactions $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ with the theoretical calculation within Talys 1.9 shows a systematic excess of the calculation over experimental values, this difference decreases with increasing energy $E_{\gamma max}$. The excess of the calculation over the experimental data is equal to the value of the experimental error.

CONCLUSIONS

The cross-sections of photonuclear reactions $^{93}Nb(\gamma,n)^{92m}Nb$ and $^{93}Nb(\gamma,n)^{92t}Nb$ averaged over the bremsstrahlung flux from the threshold to the end-point bremsstrahlung energy are determined in the range $E_{\gamma max}$ = 36…91 MeV. The obtained values of the flux-weighted average cross-sections are in good agreement with the estimates calculated within the TALYS 1.9 code using the default parameters and experimental data of other authors [6, 18].

The values $\langle\sigma(E)\rangle$ of the reactions under consideration in the range 35…45 MeV and > 70 MeV were measured for the first time.

The use of a witness-target from natural molybdenum in our measurements of averaged cross-sections made it possible to control the flux of bremsstrahlung during the experiment. The reaction of $^{100}Mo(\gamma,n)^{99}Mo$ was analyzed in the energy range $E_{\gamma max}$ = 36…91 MeV. Obtained values of $\langle\sigma(E)\rangle$ were compared to calculation results from TALYS 1.9 and to calculation results using the $\sigma(E)$ values from [14]. The difference between the normalization coefficients k_{Talys} and k_{Lorenz} was revealed to be in the range of 3...6%.

Similar measurements at the linear accelerator LUE-40 at RDC “Accelerator” NSC KIPT can be performed for a wide range of atomic weights of nuclei, which will allow us to expand the systematics of data on averaged cross-sections of photonuclear reactions with multiple particles yields and isomeric ratios of the reaction in the energy region from the giant dipole resonance to the meson production threshold. These data are essential for testing of parameterization of statistical model of the nucleus, as well as for verifying the software codes used to model reaction cross-sections.

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СЕЧЕНИЯ ФОТОЯДЕРНЫХ РЕАКЦИЙ ^{93}Nb(γ,n)^{92m}Nb и ^{93}Nb(γ,n)^{92t}Nb ПРИ ГРАНИЧНЫХ ЭНЕРГИЯХ ТОРМОЗНЫХ γ-КВАНТОВ $E_{\gamma max}$ = 36…91 МэВ

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Измерены усредненные по тормозному γ-спектру сечения $\langle\sigma(E)\rangle$ фотоядерных реакций ^{93}Nb(γ,n)^{92m,t}Nb в энергетическом интервале от порога реакции до граничных энергий $E_{\gamma max}$ = 36…91 МэВ. Исследования проведены на пучке линейного ускорителя электронов ЛУЭ-40 НИК «Ускоритель» ННЦ ХФТИ с использованием мишеней ^{93}Nb и с применением гамма-активационного метода. Спектры γ-излучения облученных образцов регистрировались HPGe-детектором с энергетическим разрешением 1,8 кэВ по γ-линии 1332 кэВ ^{60}Co. Для контроля потока тормозных γ-квантов использованы мишени-свидетели из натурального молибдена и сечение реакции ^{100}Mo(γ,n)^{99}Mo. Экспериментальные сечения $\langle\sigma(E)\rangle$ исследуемых реакций находятся в удовлетворительном согласии с теоретическими значениями, полученными при использовании программного кода TALYS 1.9 с параметрами по умолчанию. Сечения $\langle\sigma(E)\rangle$ для реакций ^{93}Nb(γ,n)^{92m,t}Nb в области 35…45 МэВ и > 70 МэВ измерены впервые.

ПЕРЕРІЗИ ФОТОЯДЕРНИХ РЕАКЦІЙ ^{93}Nb(γ,n)^{92m}Nb і ^{93}Nb(γ,n)^{92t}Nb ПРИ ГРАНИЧНИХ ЕНЕРГІЯХ ГАЛЬМІВНИХ γ-КВАНТІВ $E_{\gamma max}$ = 36…91 МеВ

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Виміряні усереднені за гальмівним спектром перерізи $\langle\sigma(E)\rangle$ фотоядерних реакцій ^{93}Nb(γ,n)^{92m}Nb і ^{93}Nb(γ,n)^{92t}Nb в енергетичному інтервалі від порога реакції до граничних енергій $E_{\gamma max}$ = 36…91 МеВ. Дослідження проведені на пучку лінійного прискорювача електронів ЛУЕ-40 НІК «Прискорювач» ННЦ ХФТІ з використанням мішеней ^{93}Nb та за допомогою гамма-активаційного методу. Спектри γ-випромінювання активованих зразків реєструвалися HPGe-детектором з енергетичним розрізненням 1,8 кеВ по лінії 1332 кеВ ^{60}Co. Для контролю потоку гальмівних γ-квантів використовувалися мішені-свідки з натурального молібдену і переріз реакції ^{100}Mo(γ,n)^{99}Mo. Експериментальні перерізи $\langle\sigma(E)\rangle$ досліджуваних реакцій знаходяться в задовільному узгодженні з теоретичними значеннями, отриманими за допомогою програмного коду TALYS 1.9 з параметрами за замовчуванням. Перерізи $\langle\sigma(E)\rangle$ реакцій ^{93}Nb(γ,n)^{92m}Nb і ^{93}Nb(γ,n)^{92t}Nb у діапазоні 35…45 МеВ і > 70 МеВ виміряні вперше.