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Experimental and theoretical research of photoneutron reactions in the ^{181}Ta nucleus

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Abstract: Bremsstrahlung fluxes for irradiating tantalum samples were formed by irradiating a tungsten converter with an electron beam having energy up to 130 MeV. The relative yields and flux-averaged cross-sections of multi-nucleon photonuclear reactions that emit up to nine neutrons in ^{181}Ta nuclei were determined. Monte Carlo simulations for studying the yields of photonuclear reactions were performed using Geant4 and TALYS-2.0 codes. The obtained experimental results were compared with available literature data and calculated results. The comparison showed that the values of the relative reaction yield and flux-averaged cross-section coincide with the literature data, considering the different geometries of the experiments. The calculated results coincide with the experimental ones only for reactions that emit up to five neutrons from the nucleus.

Keywords: photonuclear reaction, cross section, yield, neutron, simulation, Geant4, TALYS

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I. INTRODUCTION

In most studies on photonuclear reactions, the region of giant dipole resonance (GDR), in which the nucleus has a highly excited state with the participation of a large number of nucleons, is studied using bremsstrahlung. Additionally, the region lying behind the maximum GDR and extending up to the meson threshold (135 MeV) is interesting, in which the photon predominantly interacts with quasi-deuterons formed inside the nucleus and ends with the emission of several (up to ten) nucleons by the nucleus. This is due to the change in the mechanism of interaction of photons with nuclei from the excitation of GDR to the quasi-deuteron mechanism [1, 2]. To describe the mechanism of multi-particle photonuclear reactions, various theoretical models have been developed and tested through comparison with experimental data in literature. For example, a combined model of photonucleon reactions, which combines a semi-empirical model of oscillations, a quasi-deuteron model of photoabsorption, and an exciton and evaporation model, has been developed [3−5]. A Monte Carlo model of a multi-collision intranuclear cascade, which can describe photonuclear reactions at intermediate energies from 20 to 140 MeV, has also been used [6, 7]. Despite these studies, photonuclear reactions in tantalum nuclei in the energy range above 30 MeV have not been studied adequately. For the correctness of theoretical models of the quasi-deuteron mechanism to be verified, the database on the yields and cross-sections of photonuclear reactions in most stable nuclei, including tantalum nuclei, must be supplemented with new experimental data. In this study, the processes of interaction of bremsstrahlung with ^{181}Ta nuclei in the range of end-point energies from 20 to 130 MeV were investigated experimentally and theoretically.

II. EXPERIMENTAL DESIGN AND DATA ANALYSIS

The experiments were conducted at the LINAC-200 electron accelerator [8]. A tungsten converter with a size of $4.5\times4.5\times0.5$ cm was irradiated with electron beams with energies of 20, 40, 60, 80, 105, and 130 MeV. The diameter of the electron beam incident on the converter was 5.5±0.5 mm. Tantalum samples with masses of 142, 135, 163, 718, and 207 mg, respectively, were placed behind the tungsten converter for experiments with an electron beam with energies of 20, 60, 80, 105, and 130 MeV. The tantalum samples were irradiated with a bremsstrahlung flux generated in a tungsten converter in these experiments, except for the experiment with electrons with an energy of 40 MeV. In the 40 MeV electron

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beam experiment, a set of five samples (Se, Co, Y, Tb, and Ta) were irradiated with a direct electron beam. The mass of the tantalum sample was 184 mg; it was the last one in the set of samples and was irradiated with the bremsstrahlung flux generated in the Se, Co, Y, and Tb samples. The irradiation times were 40, 20, 25, 15, 20, and 15.5 min, and the mean currents were 0.40, 0.96, 0.80, 0.80, 1.16, and 1.00 μA, respectively, with electron energy. The current was measured using an inductive current sensor based on a Rogowski coil, and the measurement uncertainty using the sensor was less than 2%.

After irradiation, the tantalum samples were transferred to the test room, and their gamma spectra were measured using an HPGe detector (model GR1819). More than ten gamma spectra of each sample were measured with different measurement times. The times from the end of irradiation to the start of measuring the first spectrum of the sample were 163, 27, 21, 23, 20, and 34 min, respectively, with electron energy. The gamma spectra obtained were processed using the Deimos32 program [9]. The areas of the identified peaks were determined while the background from the Compton scattering of photons was subtracted. Figure 1 shows the gamma spectrum of a tantalum sample from the experiment with 130 MeV electrons and the background spectrum at the measuring location. The spectrum of the tantalum sample was measured for 1 h, and the time after irradiation until the start of measuring this spectrum was 4 h. The background spectrum was also normalized to a measurement time of 1 h. Figure 1 also shows examples of gamma peak identification. The absolute efficiency of the HPGe detector was measured using standard gamma sources at the same distances from the detector at which the tantalum samples were examined. Figure 2 shows the results of measuring the absolute efficiency for a distance from the detector of 1.3−13.5 cm. When processing the experimental data, we used the interpolation function of the measured values of the detector efficiency.

The yields of photonuclear reactions in the samples can be determined using the following formula:

$$Y_{\rm exp} = \frac{S_p \cdot C_{\rm abs}}{\epsilon_p \cdot I_\gamma} \frac{t_{\rm real}}{t_{\rm live}} \frac{1}{N} \frac{1}{N_\gamma} \frac{{\rm e}^{\lambda \cdot t_{\rm cool}}}{1-{\rm e}^{-\lambda \cdot t_{\rm real}}} \frac{\lambda \cdot t_{\rm irr}}{1-{\rm e}^{-\lambda \cdot t_{\rm irr}}}, \quad (1)$$

where S_p is the full-energy peak area, ϵ_p is the full-energy peak detector efficiency, $C_{\rm abs}$ is the self-absorption correction coefficient, I_γ is the gamma emission probability, $t_{\rm real}$ and $t_{\rm live}$ are the real and live times of the measurement, respectively, N is the number of atoms in a sample, N_γ is the integral number of photons incident on the tantalum sample, λ is the decay constant, $t_{\rm cool}$ is the cooling time, and $t_{\rm irr}$ is the irradiation time. However, because the total number of photons N_γ cannot be measured accurately, we determine the ratio of yields of photoneutron reactions. When determining the ratio of the reaction yields, the number of atoms in the sample and total number of photons incident on the sample cancel. In this study, the ratio of the yield of photoneutron reactions in ^{181}Ta nuclei with the release of two or more neutrons to the yield of a reaction with the release of a single neutron was determined.

The values of the parameters of the nuclear reactions studied in this work according to data from [10] are given in Table 1. $E_{\rm th}$ denotes the reaction thresholds, J, π, and $T_{1/2}$ are the spin, parity, and half-life of the nuclear reaction products, respectively, E_γ denotes the energies of gamma rays emitted by the reaction products, and I_γ de-

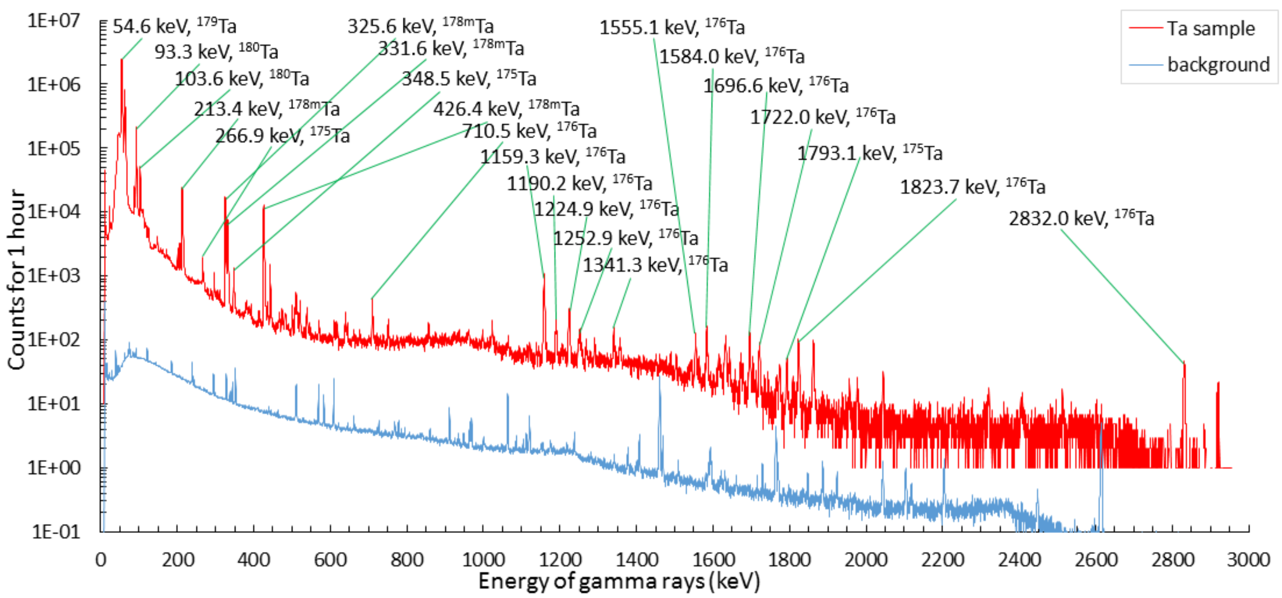

Fig. 1. (color online) Gamma spectrum of a tantalum sample and background spectrum at the measurement location.

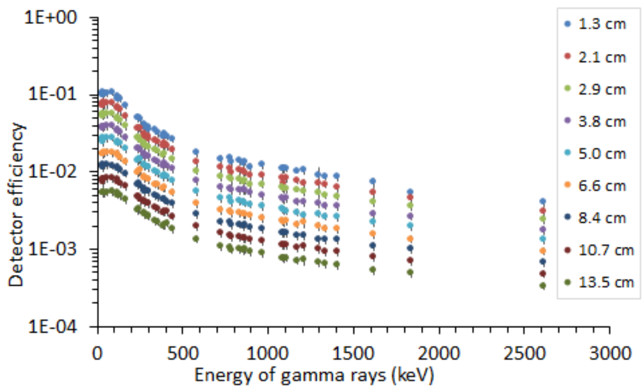

Fig. 2. (color online) Absolute efficiency of the HPGe detector.

notes the intensity of gamma rays. The values of the reaction thresholds $E_{\rm th}$ were obtained from the TALYS-2.0 program [11]. The ratios of the reaction yields were calculated for all identified gamma rays of radionuclides listed in Table 1; if a radionuclide was identified with more than one gamma line, the values of the ratio of their reaction yields were averaged.

III. MONTE CARLO SIMULATIONS

In simulations using the Geant4 code, the classes G4eBremsstrahlung, G4PenelopeBremsstrahlung, and G4LivermoreBremsstrahlungModel calculate the energy loss of electrons and positrons due to the radiation of photons in the nuclear field. The classes G4eBremsstrahlung, G4PenelopeBremsstrahlung, and G4LivermoreBremsstrahlungModel are based on the Seltzer-Berger bremsstrahlung, Penelope, and Livermore models, respectively. In these models, below electron energies of 1 GeV, the cross section evaluation is based on a dedicated parameterization; above this limit, an analytic cross section is used [12]. In our simulations, we used the class G4eBremsstrahlung (default class).

The Seltzer-Berger bremsstrahlung model was developed based on the interpolation of tables of differential cross sections [13, 14], covering electron energies from 1 keV to 10 GeV. Single-differential cross section can be expressed as the sum of the contribution of bremsstrahlung produced in the field of the screened atomic nucleus $\frac{{\rm d}\sigma_n}{{\rm d}k}$ and the part $Z\frac{{\rm d}\sigma_e}{{\rm d}k}$ corresponding to bremsstrahlung produced in the field of the Z atomic electrons,

$$\frac{{\rm d}\sigma}{{\rm d}k}=\frac{{\rm d}\sigma_n}{{\rm d}k}+Z\frac{{\rm d}\sigma_e}{{\rm d}k}.$$

The differential cross section depends on the energy k of the emitted photon, the kinetic energy of the incident electron, and the atomic number Z of the target atom.

Owing to the difficulty of using a sufficient number of electrons to determine the number of photonuclear reactions with a small error, we could obtain only the bremsstrahlung fluence in calculations using Geant4. Furthermore, the yields of the photonuclear reactions were determined using formula (2). Additionally, the reaction cross sections were calculated using TALYS-2.0. Statistical models use nuclear level densities at excitation energies to predict cross sections when information about discrete levels is unavailable or incomplete. Several level density models can be used in TALYS, from phenomenological analytical expressions to tabulated level densities derived from microscopic models [11]. In the cross section calculations, we used ldmodel 1 (Constant Temperature + Fermi gas model, the default model).

$$Y_{\rm calc}=\int_{E_{\rm thr}}^{E_{\rm max}}f(E)\sigma(E){\rm d}E, \tag{2}$$

where $f(E)$ is the bremsstrahlung fluence, and $\sigma(E)$ is the reaction cross section. Figure 3 shows the bremsstrahlung fluence incident on a tantalum sample in experiments with electrons with energies of 20, 60, 80, 105, and 130 MeV. Bremsstrahlung with energies above 7.6 MeV can cause photoneutron reactions in nuclei.

IV. RESULTS AND DISCUSSION

A. Relative yields of reactions

Based on the results of processing the measured gamma spectra, we identified photoneutron reactions with the release of up to nine neutrons from nuclei. Table 2 shows the experimental values of the ratio of photoneutron reaction yields, and Fig. 4 shows the experimental and calculated values of the ratio of yields. Table 2 also provides literature data [4] for comparison.

We can observe from Fig. 4 that, as the reaction threshold increases, the discrepancy between experiment and theory increases. If we do not consider the results of the experiment with 40 MeV electrons, the discrepancies between the experimental results and calculations for the reaction ratios ^{181}Ta(γ,2n)^{179}Ta/^{181}Ta(γ,n)^{180}Ta, ^{181}Ta(γ,3n)^{178}Ta/^{181}Ta(γ,n)^{180}Ta, ^{181}Ta(γ,4n)^{177}Ta/^{181}Ta(γ,n)^{180}Ta, and ^{181}Ta(γ,5n)^{176}Ta/^{181}Ta(γ,n)^{180}Ta are small and less than 50%. Starting from the reaction ratios ^{181}Ta(γ,6n)^{175}Ta/^{181}Ta(γ,n)^{180}Ta to ^{181}Ta(γ,9n)^{172}Ta/^{181}Ta(γ,n)^{180}Ta, the discrepancy reaches up to four times. For the reaction ^{181}Ta(γ,3n)$^{178{\rm m}}$Ta/^{181}Ta(γ,n)^{180}Ta with the formation of isomeric state of ^{178}Ta nuclei, the discrepancy is up to two times. The results from the experiment with 40 MeV electrons also agree with the calculations, except for the ^{181}Ta(γ,4n)^{177}Ta and ^{181}Ta(γ,5n)^{176}Ta reactions, although the calculated yield ratios were calculated based on the bremsstrahlung spectrum from the tungsten converter ex-

Table 1. Spectroscopic data [10] on product nuclei of measured photonuclear reactions.

Nuclear reaction	E_{th}/MeV	J^{π} of nucleus-product	$T_{1/2}$	E_{γ}/keV	I_{γ} (%)
^{181}Ta(γ,n)^{180}Ta	7.64	1^+	8.15 (1) h	93.326 (2)	4.51 (16)
				103.557 (7)	0.87 (16)
^{181}Ta(γ,2n)^{179}Ta	16.73	$7/2^+$	1.82 (3) y	55.786	22.09 (19)
				54.608	12.62 (12)
				63.000	7.29 (13)
^{181}Ta(γ,3n)^{178m}Ta	22.36	7^-	2.36 (8) h	426.383 (6)	97.0 (13)
				325.562 (4)	94.1 (11)
				213.440 (3)	81.4 (11)
				88.867 (1)	64.4 (10)
				331.613 (9)	31.19 (19)
^{181}Ta(γ,3n)^{178}Ta	22.15	$(1)^+$	9.31 (3) min	1350.68 (3)	1.18 (3)
				1340.8 (2)	1.027 (24)
^{181}Ta(γ,4n)^{177}Ta	29.03	$7/2^+$	56.36 (13) h	112.950 (1)	7.2 (8)
^{181}Ta(γ,5n)^{176}Ta	37.70	$(1)^-$	8.09 (5) h	1159.28 (9)	24.7 (18)
				201.83 (3)	5.7 (4)
				1224.93 (7)	5.7 (4)
				710.50 (8)	5.4 (4)
				1584.02 (10)	5.3 (4)
				1696.55 (13)	4.6 (3)
				1190.22 (10)	4.5 (3)
				1823.70 (15)	4.5 (3)
				2832.0 (2)	4.3 (3)
				1555.07 (10)	4.0 (3)
				1341.33 (10)	3.3 (2)
				1722.04 (13)	3.27 (24)
				1252.87 (10)	3.08 (23)
^{181}Ta(γ,6n)^{175}Ta	44.86	$7/2^+$	10.5 (2) h	348.5 (5)	12.0 (6)
				266.9 (4)	10.8 (13)
				1793.1 (3)	4.6 (6)
				436.4 (7)	3.8 (2)
				857.7 (3)	3.2 (3)
				998.3 (4)	2.6 (3)
				393.2 (6)	2.12 (16)
				475.0 (7)	2.04 (20)
^{181}Ta(γ,7n)^{174}Ta	53.39	3^+	1.14 (8) h	206.50 (4)	60 (5)
^{181}Ta(γ,8n)^{173}Ta	61.14	$5/2^-$	3.14 (13) h	172.2 (1)	17.5 (18)
^{181}Ta(γ,9n)^{172}Ta	70.00	$(3)^+$	36.8 (3) min	1109.27 (9)	14.9 (15)
				1330.41 (6)	8.1 (8)

periments. The energy thresholds for the ^{181}Ta(γ,4n)^{177}Ta and ^{181}Ta(γ,5n)^{176}Ta reactions are 29.0 and 37.7 MeV, respectively, and are close to the bremsstrahlung end-point energy. Theoretical models do not accurately calculate the cross section in the region beyond the giant resonance at energies near the reaction threshold. In Fig. 4, we also observe that the value of the reaction yield ratio in the experiment with electrons with an energy of 130 MeV

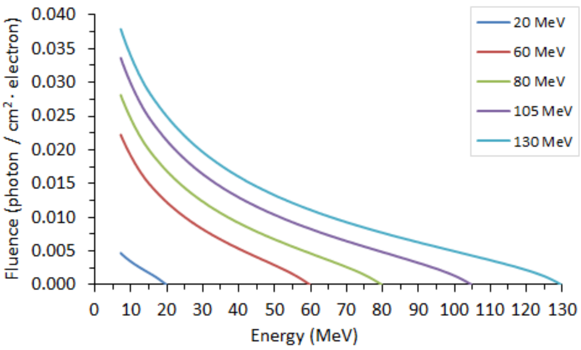

Fig. 3. (color online) Bremsstrahlung fluence.

decreases to 35% relative to the experiment with electrons with an energy of 105 MeV. This is due to the beginning of the birth of mesons. Additionally, in the calculated values of the reaction yield ratio, such a decrease is not observed.

B. Flux-averaged cross-sections

Although in experiments, owing to the impossibility of accurately determining the number of photons, only the relative values of the reaction yields are determined; the absolute values of the reaction yields and flux-averaged cross sections can still be determined. The absolute values of the flux-averaged cross-sections enable us to compare the obtained results with literature data. To determine the absolute values, we must select one well-studied reaction occurring within the sample being studied as a monitor, as in [15]. As a monitor, we selected the

Table 2. Ratios of the yields of photoneutron reactions in ^{181}Ta nuclei. *Reaction with the formation of the isomeric state ^{178m}Ta.

Reactions	Energy of electrons/MeV						
	20	40	60	67.7 [4]	80	105	130
(γ,2n)/(γ,n)	1.77(29)E-01	3.75(60)E-01	3.29(52)E-01	3.4(7)E-01	3.26(52)E-01	4.05(64)E-01	3.72(58)E-01
(γ,3n)*/(γ,n)	–	8.3(12)E-03	7.1(10)E-03	5(1)E-03	8.0(12)E-03	9.5(14)E-03	7.4(11)E-03
(γ,3n)/(γ,n)	–	2.06(38)E-02	1.72(37)E-02	1.8(4)E-02	2.25(42)E-02	3.04(50)E-02	2.93(63)E-02
(γ,4n)/(γ,n)	–	1.17(19)E-02	1.05(17)E-02	1.7(5)E-02	1.31(23)E-02	1.66(28)E-02	1.59(27)E-02
(γ,5n)/(γ,n)	–	3.62(54)E-04	3.08(45)E-03	5(1)E-03	4.32(64)E-03	8.5(12)E-03	6.17(89)E-03
(γ,6n)/(γ,n)	–	–	6.9(11)E-04	1.4(3)E-03	1.58(24)E-03	3.55(53)E-03	3.07(50)E-03
(γ,7n)/(γ,n)	–	–	–	–	3.50(56)E-04	1.52(23)E-03	1.44(21)E-03
(γ,8n)/(γ,n)	–	–	–	–	–	1.17(19)E-03	7.3(12)E-04
(γ,9n)/(γ,n)	–	–	–	–	–	2.82(59)E-04	2.70(52)E-04

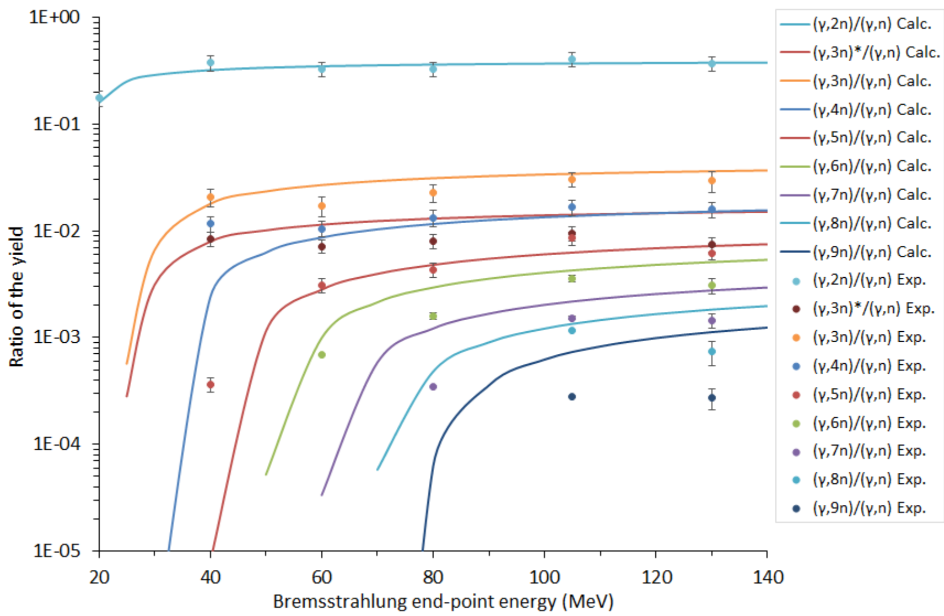

Fig. 4. (color online) Experimental and calculated values of the ratio of the yields of photoneutron reactions in ^{181}Ta nuclei. *Reaction with the formation of the isomeric state ^{178m}Ta.

^{181}Ta(γ,n)^{180}Ta reaction, because several experimental data exist [16–22] on the cross section of this reaction up to a photon energy of 35 MeV, as shown in Fig. 5. Having determined the difference between the experimental value of the yield of ^{181}Ta(γ,n)^{180}Ta reaction and the calculated one, we used it as a correction factor to calculate the integral number of photons for all measured reactions.

The cross sections averaged over the bremsstrahlung flux from the threshold $E_{\rm th}$ of the reaction under study to the end-point energy of the bremsstrahlung spectrum $E_{\gamma\max}$ were calculated using formula (1) only by replacing the integral number of photons incident on the sample to the integral number of photons in the energy range $E_{\rm th}$-$E_{\gamma\max}$. Table 3 shows the flux-averaged reaction cross-section values, and Fig. 6 includes data from [1] for comparison. We observe from Fig. 6 that the obtained experimental data coincide with the data from [1]. Small differences between them are related to the energy spectrum of the bremsstrahlung because the geometry of the experiment (material and thickness of the converter, etc.) was not the same; this is particularly noticeable in the results of the ^{181}Ta(γ,3n)^{178m}Ta reaction.

C. Uncertainty of the results

The measurement uncertainty of experimental values of reaction yield ratios was determined as the squared sum of the standard deviation of the arithmetic mean of the relative yield values and systematical errors. The standard deviation of the arithmetic mean of relative yield values also includes statistical errors. The statistical error of the observed γ-rays is primarily due to the statistical calculation of the total absorption peak of the corresponding γ-line, which varies between 1%–10%. Systematical errors include errors in detector efficiency, half-lives of residual nuclei, and the intensity of γ-rays emitted during the decay of residual nuclei. The uncertainty in the efficiency of detecting γ-radiation by the HPGe detector is up to 5%, and this value is associated with the error in the activity of standard γ-radiation sources. The half-life $T_{1/2}$ of the residual nuclei and the intensity of the analyzed γ-rays have an error of 1 to 20%, as indicated in Table 1 according to data from [10]. In this study, we did not consider the errors in the accelerated electron beam current because we determined the value of the ratio of the yields of photoneutron reactions. The uncertainty of the values of flux-averaged cross-sections is equated to the uncertainty of the values of relative yields.

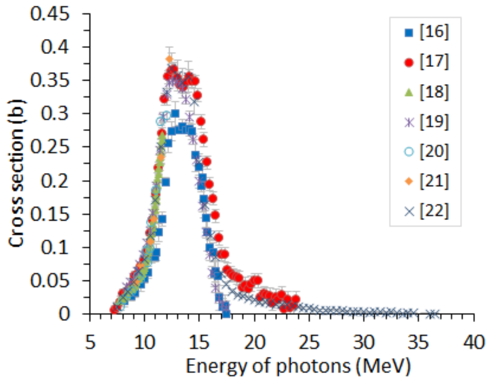

Fig. 5. (color online) Cross section of the ^{181}Ta(γ,n)^{180}Ta reaction.

V. CONCLUSION

The bremsstrahlung flux was formed by irradiating a tungsten converter with an electron beam from the LINAC-200 accelerator. The experiments were performed using electron beams with energies of 20, 40, 60, 80, 105, and 130 MeV. In each experiment, the tantalum sample was irradiated with a bremsstrahlung flux. As a result of processing the obtained data, multinucleon photonuclear reactions with the emission of up to nine neutrons in ^{181}Ta nuclei were identified. For each identified photonuclear

Table 3. Flux-averaged cross sections of photoneutron reactions [mb].

Reactions	Energy of electrons/MeV					
	20	40	60	80	105	130
^{181}Ta(γ,n)^{180}Ta	181(21)	104(11)	101(11)	92.5(99)	87.0(92)	73.9(76)
^{181}Ta(γ,2n)^{179}Ta	420(70)	77(12)	65(10)	54.4(86)	60.5(96)	43.8(68)
^{181}Ta(γ,3n)^{178m}Ta	–	3.76(56)	2.78(41)	2.31(35)	2.30(35)	1.31(20)
^{181}Ta(γ,3n)^{178}Ta	–	9.3(17)	6.7(15)	6.5(12)	7.4(12)	5.2(11)
^{181}Ta(γ,4n)^{177}Ta	–	12.9(21)	7.2(12)	5.9(10)	5.78(98)	3.75(64)
^{181}Ta(γ,5n)^{176}Ta	–	6.9(10)	4.52(66)	3.23(48)	4.44(65)	1.99(29)
^{181}Ta(γ,6n)^{175}Ta	–	–	2.28(36)	1.88(29)	2.59(39)	1.27(19)
^{181}Ta(γ,7n)^{174}Ta	–	–	–	0.80(13)	1.70(26)	0.80(12)
^{181}Ta(γ,8n)^{173}Ta	–	–	–	–	1.88(16)	0.51(16)
^{181}Ta(γ,9n)^{172}Ta	–	–	–	–	0.77(16)	0.26(5)

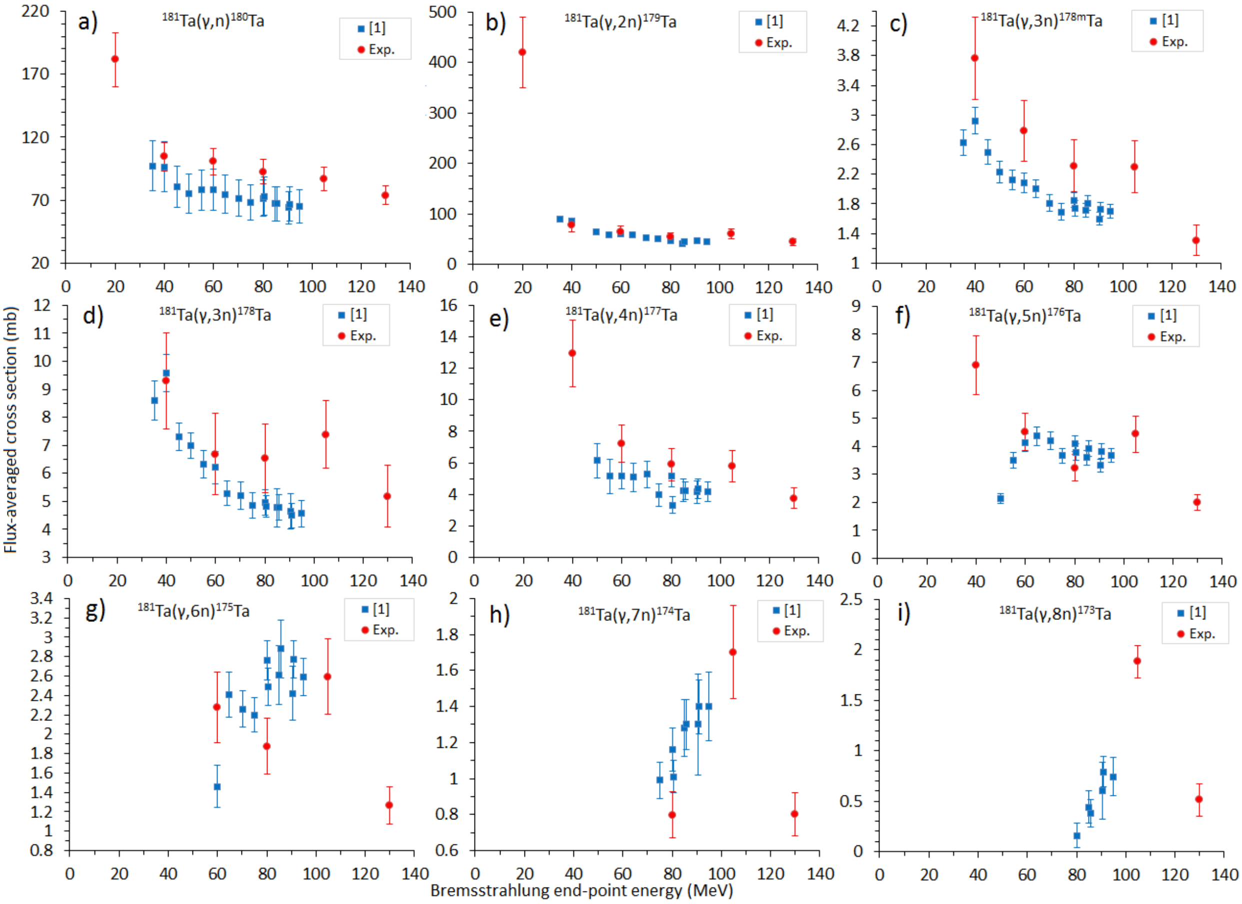

Fig. 6. (color online) Flux-averaged cross sections of photoneutron reactions in ^{181}Ta nuclei.

reaction, relative reaction yields and flux-averaged cross-sections were determined. Calculation studies of the yields of photonuclear reactions were performed using Geant4 and TALYS-2.0 codes. The bremsstrahlung fluxes for tantalum samples were calculated with Geant4, and the photonuclear reaction cross-sections were calculated with TALYS-2.0.

The obtained experimental results were compared with available literature data and calculated results. The experimental values of the relative reaction yield and flux-averaged cross-section coincide with the literature data, considering the different geometries of the experiments. The differences between the experimental and calculated values of the relative yields are less than 50% in reactions with up to five neutrons emitted from the nucleus. With an increase in the number of neutrons emitted from the nucleus, the difference between the experiment and calculation increases. These comparisons show that the used theoretical models cannot correctly describe photonuclear reactions that emit more than 5-6 neutrons from the nucleus.

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