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Search for the evidence of ^{209}Bi(γ,p5n)^{203}Pb reaction in 60 MeV bremsstrahlung beams

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Abstract Samples made from natural bismuth were exposed in 60 MeV end-point bremsstrahlung beam. In this paper, a simple model for determination the share of two ways of ^{203}Pb formation: by the decay of ^{203}Bi, produced in ^{209}Bi(γ,6n)^{203}Bi reaction and by ^{209}Bi(γ,p 5n)^{203}Pb reaction is described. The method employs the ratio of ^{203}Pb and ^{203}Bi nuclei numbers and activities at the end of the exposure as the input value. This ratio was estimated from gamma spectra measured after irradiation of natural Bi sample. It was found that the rate of production of ^{203}Pb by ^{209}Bi(γ,p 5n)^{203}Pb reaction is about 6% of the ^{203}Bi production rate in the ^{209}Bi(γ,6n)^{203}Bi reaction. Obtained result is compared with TALYS based estimation.

1 Introduction

The simplest photonuclear reaction (γ,n) usually takes place through the well-known mechanism of giant dipole resonance. For a large number of stable nuclei, the energy differential cross section of this reaction has been successfully measured [1, 2]. Data on photonuclear reactions can be found in available databases [3] also. The experimental evidence for the (γ,2n) reaction is much poorer, while for reactions when three or more neutrons are emitted (usually denoted by (γ,xn)), the reaction cross sections can be obtained by mainly by theoretical calculations [4]. For reactions in which a charged particle, such as proton in the simplest case, is emitted (single or in addition to one or more neutrons) there is a much poorer experimental evidence. In the interactions of high energy photons with nuclei, the probability of emission of a charged particle is significantly lower than the emission of neutrons due to the existence of the Coulomb barrier. It is a reason why experimental data concerning (γ,p xn) are insufficient in literature.

Cross sections for photonuclear reactions on natural bismuth (monoisotope ^{209}Bi) with emission of one and two neutrons can be found in the reference [5]. In several recently published papers [6–11], photonuclear reactions with Bi target exposed in bremsstrahlung photon beams having end-point energies up to 70 MeV were studied. Irradiation of such a heavy element by the high energy photons can give several products of (γ,xn) reactions. In these publications, the relative yield of photonucleated reactions on ^{209}Bi was analyzed, while in reference [12] the cross section results for reactions (γ,n), (γ,2n), (γ,3n) and (γ,4n) were presented. The authors of reference [12] used natural bismuth target and quasimonochromatic laser Compton-scattering γ-ray beams with energies up to 40 MeV. In reference [11], reaction channel (γ,pxn) with emission of one proton along with several neutrons is accounted for.

In this paper, an attempt was made to establish experimental evidence for ^{209}Bi(γ,p 5n)^{203}Pb nuclear reaction by comparison of intensities of gamma lines following EC decay of ^{203}Bi and ^{203}Pb. Lead-203 can be formed by (γ,p 5n) nuclear reaction, but it is certainly created after decay of ^{203}Bi, obtained in ^{209}Bi(γ,6n)^{203}Bi reaction. After activation of the target made from natural bismuth by 60 MeV end-point energy bremsstrahlung beam, several gamma spectra were successively measured. Gamma lines from the measured spectra were selected in order to estimate the ratio of created nuclei numbers of ^{203}Bi and ^{203}Pb in the moment

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Fig. 1 Part of the chart containing relevant nuclides

Bi202 **1.72 h** **5+** **EC,α**	**Bi203** **11.76 h** **9/2-** **EC,α**	**Bi204** **11.22 h** **6+** **EC**	**Bi205** **15.31 d** **9/2-** **EC**	**Bi206** **6.243 d** **6(+)** **EC**	**Bi207** **31.55 y** **9/2+** **EC**	**Bi208** **3.68E5 y** **(5)+** **EC**	**Bi209** **Stable** **9/2-** **100%**
Pb201 **9.33 h** **5/2-** **EC**	**Pb202** **5.24E4 y** **0+** **EC**	**Pb203** **51.873 h** **5/2-** **EC**	**Pb204** **1.4E17 y** **0+** **1.4%**	**Pb205** **1.53E7 y** **5/2-** **EC**	**Pb206** **Stable** **0+** **24.1%**	**Pb207** **Stable** **1/2-** **24.1%**	**Pb208** **Stable** **0+** **52.4%**

when irradiation was stopped. Probability ratio for the occurrence of (γ,6n) and (γ,p 5n) nuclear reactions can be obtained from these data.

For the purposes of this paper, the cross sections for the observed nuclear reactions were extracted using the TALYS code. These cross-sections were used to estimate the ratio of probabilities of nuclear reactions of interest in order to compare them with the values obtained by measurements.

2 Materials and methods

Considering that lead isotopes ^{206}Pb, ^{207}Pb and ^{208}Pb are stable (Fig. 1), ^{209}Bi(γ,p3n)^{205}Pb is the very first photonuclear reaction on ^{209}Bi which can give active lead as a product. However, half life of ^{205}Pb is $1.57\cdot10^7$ y and small amount of produced ^{205}Pb, would be very difficult to detect. Moreover, this isotope does not emit gamma radiation.

The next candidate which can be used to verify if (γ,pxn) on natural bismuth can yield measurable amounts of reaction products is ^{204m}Pb. Half life of ^{204m}Pb is 67.2 min and de-excitation of isomer state takes place through several gamma transitions having high quantum yield. However, ^{204m}Pb can be created by decay of ^{204}Bi, produced in (γ,5n) photonuclear reaction. This means that de-excitation of the isomeric state of ^{204m}Pb created in (γ,p 4n) results in gamma transitions that are almost the same as those occurred after the ^{204}Bi decay. It is very difficult to estimate how ^{204}Bi decay and ^{204m}Pb de-excitation contribute to the total intensity of some of measured gamma lines.

Another possibility to check if irradiation of ^{209}Bi by high-energy photons results in proton emission together with several neutrons is lead isotope ^{203}Pb. This isotope can be created by (γ,p 5n) reaction. Half-life of ^{203}Pb is 51.873 h and after decay two intensive lines in gamma spectra can be observed. A study of gamma photons originating from the decay of ^{203}Pb could be a good way to determine if (γ,p 5n) reaction can give measurable amount of ^{203}Pb during irradiation of the natural bismuth target by high energy photons. But in order to do that, it is necessary to estimate, in some way, how much of the ^{203}Pb activity comes from decay of ^{203}Bi.

2.1 Irradiation

In interactions of ^{209}Bi nuclei with high energy photons, several bismuth isotopes can be created through (γ,xn) reactions. Suppose that a bismuth isotope ^{203}Bi, we have chosen to analyze, is formed at a constant rate q in a photon beam. The change of the number of nuclei of observed bismuth isotope can then be described by the following differential equation:

$$\frac{dN_{Bi}}{dt} = q - \lambda_{Bi} N_{Bi} \tag{1}$$

λ_{Bi} is decay constant of ^{203}Bi, created in (γ,6n) photonuclear reaction. The number of created ^{203}Bi nuclides after irradiation time t_{irr} is:

$$N_{Bi} = \frac{q}{\lambda_{Bi}}(1 - \exp(-\lambda_{Bi} t_{irr})) \tag{2}$$

The dynamics of the number of ^{203}Pb nuclei created exclusively by the decay of ^{203}Bi can be expressed as:

$$\frac{dN_{Pb}}{dt} = \lambda_{Bi} N_{Bi} - \lambda_{Pb} N_{Pb} \tag{3}$$

The very same ^{203}Pb isotope can additionally be produced through the (γ,p 5n) nuclear reaction. In the case when the production of ^{203}Pb occurs, process can be described by the following equation:

$$\frac{dN_{Pb}}{dt} = \lambda_{Bi} N_{Bi} + p - \lambda_{Pb} N_{Pb} \tag{4}$$

where p denotes a constant rate of ^{203}Pb production through the (γ,p 5n) nuclear reaction. It can be considered that at the very beginning ($t = 0$), there were no ^{203}Pb nuclei. The solutions of differential Eqs. 3 and 4 describing time evolution of the number of ^{203}Pb isotopes are:

$$N_{Pb} = q\left[\frac{1}{\lambda_{Pb}}(1 - \exp(-\lambda_{Pb} t_{irr})) + \frac{1}{\lambda_{Pb} - \lambda_{Bi}}(\exp(-\lambda_{Pb} t_{irr}) - \exp(-\lambda_{Bi} t_{irr}))\right] \tag{5}$$

in the case when ^{203}Pb originates from the decay of ^{203}Bi, and

$$N_{Pb} = q\left[\frac{1+\frac{p}{q}}{\lambda_{Pb}}(1-\exp(-\lambda_{Pb}t_{irr})) + \frac{1}{\lambda_{Pb}-\lambda_{Bi}}(\exp(-\lambda_{Pb}t_{irr})-\exp(-\lambda_{Bi}t_{irr}))\right] \quad (6)$$

when production of ^{203}Pb through (γ,p 5n) can not be neglected.

Using Eqs. 2 and 5 ratio $N_{Pb}(t_{irr})/N_{Bi}(t_{irr})$ of produced numbers of ^{203}Pb and ^{203}Bi nuclei, in the case when ^{203}Pb is produced only by the ^{203}Bi decay, can be determined as:

$$\frac{N_{Pb}(t_{irr})}{N_{Bi}(t_{irr})} = \frac{\frac{\lambda_{Bi}}{\lambda_{Pb}}(1-\exp(-\lambda_{Pb}t_{irr})) + \frac{\lambda_{Bi}}{\lambda_{Pb}-\lambda_{Bi}}(\exp(-\lambda_{Pb}t_{irr})-\exp(-\lambda_{Bi}t_{irr}))}{1-\exp(-\lambda_{Bi}t_{irr})} \quad (7)$$

It can be seen from Eq. 7 that the ratio of ^{203}Pb and ^{203}Bi nuclei numbers does not depend on the rate q at which Bi is generated in photonuclear reaction.

In a similar way, using Eqs. 2 and 6, it can be determined the ^{203}Pb/^{203}Bi nuclei number ratio for the case when the ^{203}Pb isotope is formed by decay of ^{203}Bi and the direct ^{209}Bi(γ,p 5n) reaction as well.

$$\frac{N_{Pb}(t_{irr})}{N_{Bi}(t_{irr})} = \frac{(1+\frac{p}{q})\frac{\lambda_{Bi}}{\lambda_{Pb}}(1-\exp(-\lambda_{Pb}t_{irr})) + \frac{\lambda_{Bi}}{\lambda_{Pb}-\lambda_{Bi}}(\exp(-\lambda_{Pb}t_{irr})-\exp(-\lambda_{Bi}t_{irr}))}{1-\exp(-\lambda_{Bi}t_{irr})} \quad (8)$$

From the above Eq. 8, we can see that the ratio p/q, ie. the production rate of ^{203}Bi by the emission of six neutrons and the rate of production of ^{203}Pb by (γ,p 5n) nuclear reaction affects ratio of these two isobars. It can be seen that, if there is no proton emission, ie. if $p = 0$, Eq. 8 turns into Eq. 7.

All the above equations, as well as those that will follow in which the ratios of the nuclei of the observed two isotopes N_{Pb}/N_{Bi} can be written in the form where the ratios of their activities A_{Pb}/A_{Bi} appear.

2.2 Decay measurement

At the moment when the irradiation is completed, there will be some number of both radionuclides in the bismuth sample: $N_{Pb}(t_{irr})$ and $N_{Bi}(t_{irr})$. After the end of the irradiation, it is possible to register gamma spectra of irradiated sample and $N_{Pb}(t_{irr})$ and $N_{Bi}(t_{irr})$ can be considered as the initial numbers of ^{203}Bi and ^{203}Pb. Let's denote them as N^0_{Bi} and N^0_{Pb}. Starting from the end of irradiation, activity of ^{203}Bi will decrease according to a well-known exponential law

$$N_{Bi}(t) = N^0_{Bi}\exp(-\lambda_{Bi}t) \quad (9)$$

while the dynamics of the time change of ^{203}Pb activity will be determined by the rate of its simultaneous decay and creation from ^{203}Bi. It is well known that a number of daughter nuclei in the case of simultaneous decay can be described as:

$$N_{Pb}(t) = \frac{\lambda_{Bi}}{\lambda_{Pb}-\lambda_{Bi}}N^0_{Bi}(\exp(-\lambda_{Bi}t) - \exp(-\lambda_{Pb}t)) + N^0_{Pb}\exp(-\lambda_{Pb}t) \quad (10)$$

Let's assume that the recording of the gamma spectrum started at the moment t_1 after the end of the irradiation, and that the measurement was stopped at the moment t_2. In that time interval, the number of decayed ^{203}Bi nuclei is:

$$N^D_{Bi} = N^0_{Bi}(\exp(-\lambda_{Bi}t_1) - \exp(-\lambda_{Bi}t_2)) \quad (11)$$

By integrating the function that describes the temporal change of ^{203}Pb activity within the same time limits, it is obtained that the total number of ^{203}Pb nuclei that decayed is:

$$N^D_{Pb} = \frac{\lambda_{Bi}}{\lambda_{Bi}-\lambda_{Pb}}N^0_{Bi}(\exp(-\lambda_{Pb}t_1) - \exp(-\lambda_{Pb}t_2)) - \frac{\lambda_{Pb}}{\lambda_{Bi}-\lambda_{Pb}}N^0_{Bi}(\exp(-\lambda_{Bi}t_1) - \exp(-\lambda_{Bi}t_2)) + N^0_{Pb}(\exp(-\lambda_{Pb}t_1) - \exp(-\lambda_{Pb}t_2)) \quad (12)$$

From Eqs. 11 and 12 the values of N^0_{Bi} and N^0_{Pb} can be estimated, but in some cases it is more convenient to analyze the ratio of the number of decays of the observed two nuclides:

$$\frac{N_{Pb}^D}{N_{Bi}^D} = \frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \frac{\exp(-\lambda_{Pb}t_1) - \exp(-\lambda_{Pb}t_2)}{\exp(-\lambda_{Bi}t_1) - \exp(-\lambda_{Bi}t_2)} - \frac{\lambda_{Pb}}{\lambda_{Bi} - \lambda_{Pb}} + \frac{N_{Pb}^0}{N_{Bi}^0} \frac{\exp(-\lambda_{Pb}t_1) - \exp(-\lambda_{Pb}t_2)}{\exp(-\lambda_{Bi}t_1) - \exp(-\lambda_{Bi}t_2)} \tag{13}$$

As can be seen from Eq. 13, if the ratio of the decay numbers of ^{203}Pb and ^{203}Bi is known, it is possible to estimate the ratio of the initial numbers of these two nuclides N_{Pb}^0/N_{Bi}^0.

In order to determine the ratio of the number of decays N_{Pb}^D/N_{Bi}^D in a selected time interval, two gamma lines, one from ^{203}Pb and another from ^{203}Bi should be selected their intensities should be determined. Let's denote registered intensities with N_{Bi}^R and N_{Pb}^R. Ratio N_{Pb}^D/N_{Bi}^D can be determined as:

$$\frac{N_{Pb}^D}{N_{Bi}^D} = \frac{N_{Pb}^R}{N_{Bi}^R} \frac{\varepsilon_{Bi} p_\gamma^{Bi}}{\varepsilon_{Pb} p_\gamma^{Pb}} \tag{14}$$

The quantum yields of the observed gamma transitions of ^{203}Bi and ^{203}Pb are denoted by p_γ^{Bi} and p_γ^{Pb} while ε_{Bi} and ε_{Pb} are the absolute detection efficiency at the selected energies. From Eq. 14 it can be seen that it is sufficient to know the relative efficiency of the detector system for the used geometry.

2.3 Procedure of the results extraction

And finally, let's summarize the procedure that can be used to prove whether a measurable contribution of (γ,p 5n) reaction could be expected:

(a) the ratio of ^{203}Pb and ^{203}Bi nuclei numbers, expected at the end of irradiation, without contribution of (γ,p 5n) reaction, could be calculated using Eq. 7;
(b) intensities of selected gamma lines should be obtained. The N_{Pb}^D/N_{Bi}^D ratio can be calculated using Eq. 14;
(c) Equation 13 should be used to obtain the ratio of initial nuclei numbers N_{Pb}^0/N_{Bi}^0, created during exposition in the photon beam;
(d) obtained N_{Pb}^0/N_{Bi}^0 ratio can be compared with the result derived from Eq. 7. If the obtained values coincide within the interval of experimental error, it can be concluded that the reaction (γ,p 5n) does not give a measurable contribution to the activity of ^{203}Pb. However, if the contribution of the reaction (γ,p 5n) to the total number of ^{203}Pb cannot be neglected, the N_{Pb}^0/N_{Bi}^0 ratio obtained by Eq. 13 should be greater than the ratio acquired from Eq. 7
e) and finally, if the ratios N_{Pb}^0/N_{Bi}^0 differ, it is possible to find the value of the parameter p/q in Eq. 8 which gives the N_{Pb}^0/N_{Bi}^0 ratio experimentally established.

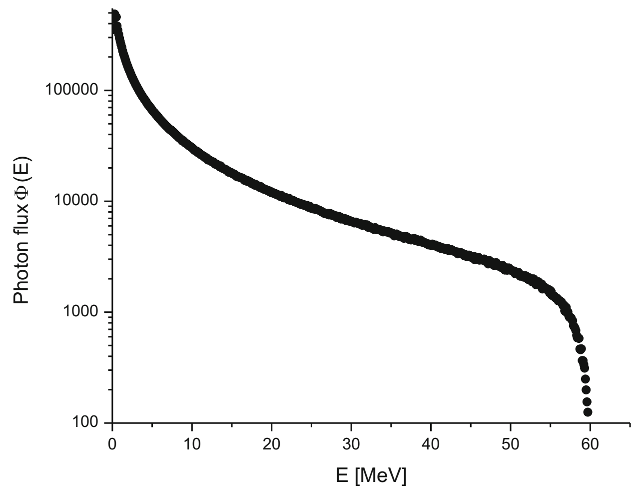

Fig. 2 The shape of the photon spectrum $\Phi(E)$ obtained for 30 M incident electrons

The described procedure can give confirmation about contribution of the (γ,p 5n) reaction to the total activity of ^{203}Pb and to estimate the value p/q.

2.4 Theoretical calculations

The rate of some nuclear reaction at the selected target is directly proportional to the product of the cross section and the number of incident particles. This means that the quantities p and q would be proportional to the saturation activities of observed reactions:

$$q \sim \int_{E_t}^{E_{max}} \sigma_{6n}(E)\Phi(E)dE \tag{15}$$

$$p \sim \int_{E_t}^{E_{max}} \sigma_{p,5n}(E)\Phi(E)dE \tag{16}$$

where $\Phi(E)$ is photon fluency, E_t is energy threshold for observed nuclear reaction and E_{max} is end-point energy of the photon spectra (in our case 60 MeV). By $\sigma_{6n}(E)$ and $\sigma_{p,5n}(E)$ cross-sections for (γ,6n) and (γ,p 5n) nuclear reactions are denoted respectively.

The simplest way to check the obtained result for the p/q ratio would be to calculate the saturation activities for these two reactions using Eqs. 15 and 16 and compare them with the experimental result.

The shape of the photon spectrum $\Phi(E)$ is obtained by using Geant4 software package [13], version v11.1.0, with standard G4 electromagnetic physics option selected. The simulation starts with creating 30 M of 60 MeV electrons in the beam, with very small Gaussian spread in energy of 0.01 MeV. The photon spectrum, obtained at the place of irradiated sample based on the geometry described in the next section is presented in Fig. 2.

There is no experimental evidence of the cross sections of ^{209}Bi (γ,6n) and ^{209}Bi (γ,p 5n) nuclear reactions in literature. Related information can be obtained using some numerical code for evaluation of the cross sections for nuclear reactions. In this experiment, TALYS 1.9 code was used to estimate cross sections for ^{209}Bi(γ,p 5n)^{203}Pb and ^{209}Bi(γ,6n)^{203}Bi reactions. It was decided to use SMLO model for a strength function. It can be expected that the choice of the strength function model has an impact on the estimation of the cross section, but that analysis is beyond the scope of this paper. Six different models of level density were employed in calculations. Cross sections were calculated using phenomenological (1. The Fermi Gas Model + Constant Temperature Model, 2. The Back-shifted Fermi gas Model, 3. The Generalized Superfluid Model) and microscopic (4. Skyrme-Hartree–Fock-Bogoluybov, 5. Gogny-Hartree–Fock-Bogoluybov and temperature-dependent 6. Gogny-Hartree–Fock-Bogoluybov models) of level density [14].

Fig. 3 Measurement setup

3 Measurements

One coin-shaped sample of natural bismuth (high purity 99.999%), 1 cm diameter and mass of 1.1 g, was exposed in bremsstrahlung photon beam having maximal energy of 60 MeV. The source of the photon beams was the linear electron accelerator LUE-75 located at A. Alikhanyan National Science Laboratory in Yerevan, Armenia. Accelerated electrons, after passing through a cylindrical collimator (length of 20 mm and a diameter of 15 mm) strike a pure tungsten converter. The thickness of the converter was 2 mm, and a 30 mm long aluminum cylinder was placed directly behind it. The function of the aluminum was to stop the electrons that passed through the tungsten. At a distance of 60 mm from the tungsten plate, a Bi coin was placed. Duration of exposition was 30 min.

After 22 min., exposed Bi coin was placed 86 mm from the end cap of HPGe detector. Measurement setup is presented in Fig. 3. Activity of measured samples was high enough and no detector shielding was used. Sample was fixed by tape to the holder ring above detector. First 5400 s spectrum was measured and second one was collected during 79,463 s. In the measured spectra, gamma lines from several Bi isotopes were identified. The lightest one was ^{202}Bi. Gamma line intensities were determined using the GENIE software package.

The most intense ^{203}Pb gamma transition of 279.2 keV (quantum yield 81%) was selected for calculation procedure. In both collected spectra, a prominent single 279.2 keV gamma line appeared. For example, in the first spectrum, intensity of this line was 5.87(28)·10^3 counts. In the second spectrum, intensity of this line was 3.71(2)·10^5 detected counts. Several strong gamma lines of ^{203}Bi were identified in spectra, however almost all of them are parts of doublets or even multiplets. In order to avoid errors caused during the numerical procedure in separation of the intensity of individual lines in doublets, several gamma transitions of ^{203}Bi were chosen for calculation. Ratio of decayed nuclei N^D_{Pb}/N^D_{Bi} was calculated using Eq. 14 and intensities of 264.2 keV, 816.2 keV and 1033.8 keV gamma lines of ^{203}Bi and mean average was calculated. For example, in the first spectrum intensity of 264.2 keV gamma line was 3.63(7)·10^4 detected counts. Considering that activity of ^{203}Bi decreased all the time, while the activity of ^{203}Pb increased, intensity 264.2 keV gamma line was smaller than 279.2 keV gamma line of ^{203}Pb in second spectra. Intensity of 264.2 keV gamma line was 1.015(15)·10^4 counts. Two segments of measured gamma spectra are depicted in Fig. 4.

It can be seen from the Eq. 14 that the N^D_{Pb}/N^D_{Bi} ratio can be obtained using the relative efficiency of the detection system. The best way to get relative efficiency is to use gamma lines of ^{206}Bi. Half-life of this isotope is 6.243 days and there are many intensive gamma transitions from 183.98 to 2 MeV. The longer spectrum was used and 16 gamma lines of ^{206}Bi, starting from 183.98 keV to 1878.65 keV were selected to get relative efficiency. Combination of an exponential function and a second order polynomial was used in fit procedure.

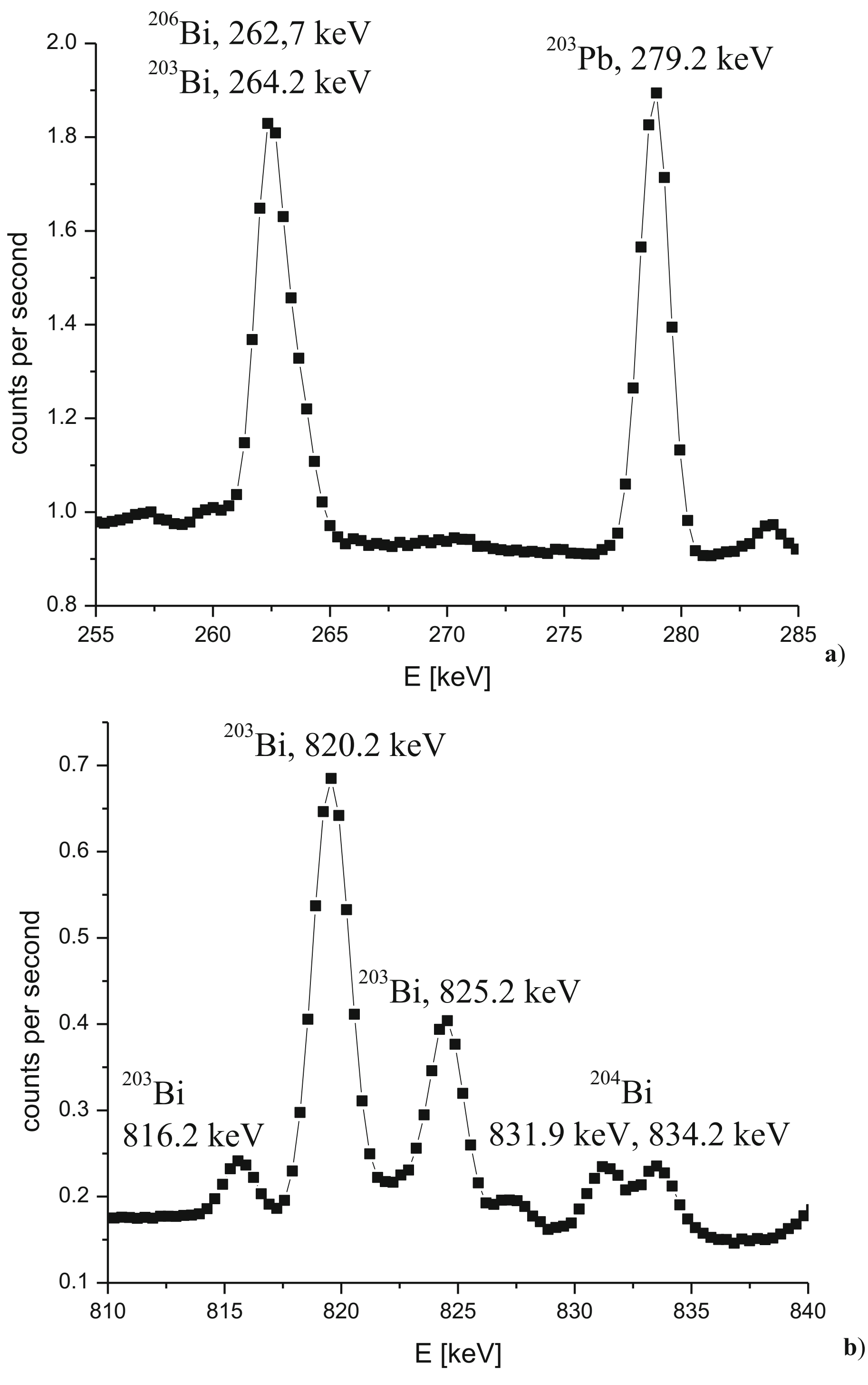

Fig. 4 Two parts of gamma spectra: **a**) low energy part containing ^{203}Pb gamma line (279.2 keV) and ^{203}Bi/^{206}Bi doublet; **b**) multiplet containing three ^{203}Bi lines

4 Results and discussion

4.1 Experiment

The estimation of the $N_{Pb}(t_{irr})/N_{Bi}(t_{irr})$ ratio at the moment when irradiation was stopped can be obtained from the Eq. 7. This equation describes output of the (γ,6n) reaction, without proton emission. It was obtained that the ratio of created ^{203}Pb and ^{203}Bi nuclei is $N_{Pb}(t_{irr})/N_{Bi}(t_{irr}) = 0.0148$. If no (γ,p 5n) reaction occurs, it could be expected that number of ^{203}Pb created nuclei is 1.48% of the number of ^{203}Bi nuclei, after 30 min of irradiation. Ratio of activities of two

Table 1 $^{203}Pb/^{203}Bi$ activity ratio at the end of irradiation

	$N_{Pb}(t_{irr})/N_{Bi}(t_{irr})$	$A_{Pb}(t_{irr})/A_{Bi}(t_{irr})$
Without proton emission ((γ,6n) only), Eq. 7	0.0148	0.00335
Experimental based values, Eq. 13	0.0758(8)	0.0172(2)

mentioned nuclei after irradiation is $A_{Pb}(t_{irr})/A_{Bi}(t_{irr}) = 0.00335$.

The intensities of the selected gamma lines were determined in both measured spectra. Relative efficiency of detection was calculated using gamma lines of ^{206}Bi. Ratio of ^{203}Pb and ^{203}Bi nuclei that decayed in the observed time interval was evaluated as shown in Eq. 14. Obtained N^D_{Pb}/N^D_{Bi} ratio was used to calculate value of N^0_{Pb}/N^0_{Bi} ratios by Eq. 13. The ratio of activities at the moment when irradiation was stopped was calculated as well. Both registered spectra gave ratio results that agreed within experimental error. In the continuation of the paper, the results obtained from gamma lines measured in a longer spectrum will be presented, due to better counting statistics. The calculated and experimental values are presented in Table 1.

It can be seen from the data presented in Table 1 that the ratio of the numbers of created ^{203}Pb and ^{203}Bi nuclei, as well as their activity ratio, at the end of irradiation, extracted from the measured spectra, differs significantly from the estimation based on the assumption that no proton is emitted in photonuclear reaction. This leads us to the conclusion that the production of ^{203}Pb by some other process, probably (γ,p 5n), in addition to the decay of ^{203}Bi can have a significant contribution.

After the experimental confirmation of the possibility that (γ,p 5n) nuclear reaction can make a measurable contribution to total ^{203}Pb activity, the very next step is to estimate *p/q* ratio. Equation 8 gives such a possibility. If the irradiation time was 30 min and experimentally obtained ratio of ^{203}Pb and ^{203}Bi nuclei is 0.0758(8), it was estimated using Eq. 8, that p/q factor is 0.060(1). This means that the rate of production of ^{203}Pb by ^{209}Bi(γ,p 5n) reaction is 6% of the rate of ^{209}Bi(γ,6n) ^{203}Bi (Fig. 5).

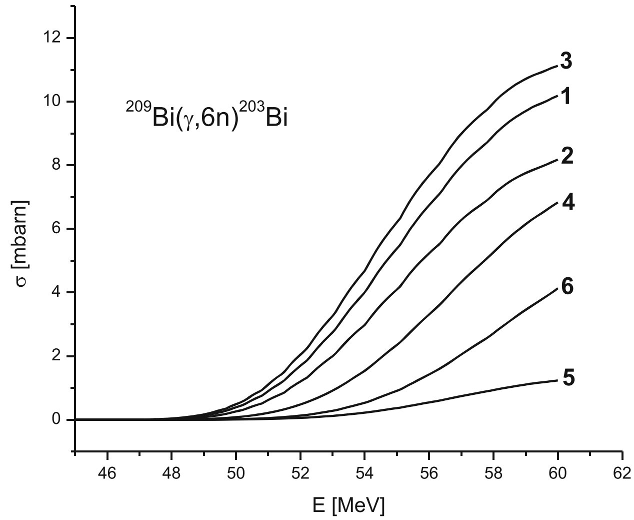

Fig. 5 TALYS estimations for cross section of ^{209}Bi(γ,6n)^{203}Bi reaction. The numbers indicate the level density model as numbered in Sect. 2.4

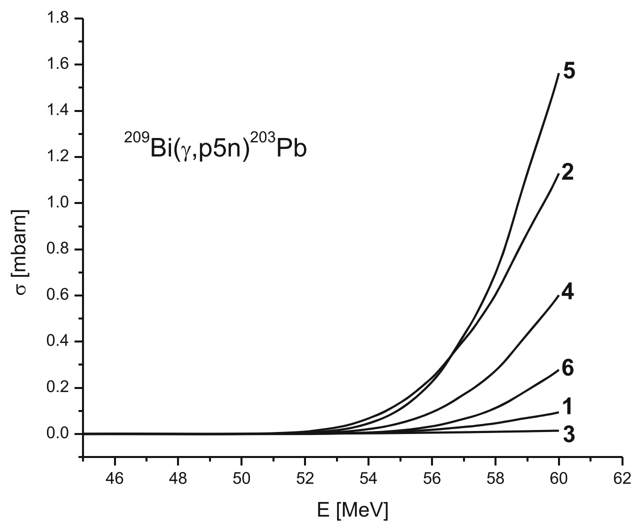

Fig. 6 TALYS estimations for cross section of ^{209}Bi(γ,p 5n)^{203}Pb reaction. The numbers indicate the level density model as numbered in Sect. 2.4

4.2 Calculated ratio of ^{209}Bi(γ,p 5n) and ^{209}Bi(γ,6n) reaction probabilities

And finally, in order to estimate ratio of probabilities of ^{209}Bi(γ,p 5n) and ^{209}Bi(γ,6n) reactions, denoted as p and q, it is necessary to calculate the energy differential cross section for both reactions using the TALYS code. The obtained results, for all six models of level densities are shown in Figs. 5 and 6. As might be expected, the cross section for a reaction without emission of positive charged particle is significantly larger than cross section for emission of one proton in addition to five neutrons. It should also be noted that the

Table 2 Ratio p/q obtained using measured activities and simulation performed with six different models of level densities (in the same order as stated in Sect. 2.4)

	p/q ratio
Estimated using TALYS cross sections	0.00278
	0.0472
	0.000698
	0.0329
	0.5077
	0.0295
Estimated from Eq. 8	0.060(1)

estimated cross sections for both observed reactions can be very different from each other, depending on which density of states model is chosen.

With the obtained values of cross section and chosen bremsstrahlung function it is possible to calculate p/q ratio using both Eqs. 15 and 16. The result of TALYS estimations are depicted in Table 2, together with results of calculations based on measured data.

It is referred that the results obtained in the photo-activation experiments on ^{208}Pb and ^{209}Bi [7, 8] are in good agreement with the calculations performed using the TALYS code. Some examples can be found in the literature [15, 16] that the results of TALYS calculations do not completely match the results of measurements. For the purpose of analyzing the obtained results, it is particularly important to emphasize that one of the conclusions presented in reference [11] is that the shares of ^{209}Bi(γ,p 4n) and ^{209}Bi(γ,p 5n) reaction are not negligible. The authors estimated that the yield of the ^{209}Bi(γ,p 4n) reaction obtained experimentally at 55.6 MeV bremsstrahlung beam was almost 15 times higher than the yield estimated using the TALYS code. In the case of ^{209}Bi(γ,p 5n) reaction, this difference is even greater.

Here, it is particularly important to point out that, based on the results presented in reference [11], it can be concluded that the yield of ^{209}Bi(γ,p 5n)^{203}Pb reaction is about 9.4 times higher than the yield of reaction ^{209}Bi(γ,6n)^{203}Bi. As can be seen from Table 2, in this paper it is estimated that the yield of ^{209}Bi(γ,6n)^{203}Bi reaction is over 16 times higher than the yield of ^{209}Bi(γ,p 5n)^{203}Pb reaction. It is most likely that the Coulomb barrier significantly reduces the probability of reactions in which a charged particle is emitted.

According to the data presented in Table 2, it is difficult to get general conclusion concerning agreement between TALYS estimation and experimental results. As can be seen, TALYS estimations can be almost two orders of magnitude smaller than the experimentally obtained p/q value, as well as one order of magnitude higher.

5 Conclusions

In this study, an attempt was made to check if ^{203}Pb is formed in photonuclear reactions on ^{209}Bi exclusively from the decay of ^{203}Bi formed in (γ,6n) reaction or (γ,p 5n) reaction also plays a part in it, as indicated in reference [11]. It has been shown that this estimation can be made if the ratio of ^{203}Pb and ^{203}Bi nuclei numbers, at the moment when the exposure of the Bi target in the photon beam is completed, is known.

Irradiation of the target from natural bismuth was performed at 60 MeV bremsstrahlung beam. Induced activity was measured in standard off-beam experiment. Registered spectra were used to extract numbers of created ^{203}Bi and ^{203}Pb nuclei. The obtained ratio of nuclei numbers indicates that ^{203}Pb is formed in some other way, other than from the decay of ^{203}Bi. The most likely mechanism is the (γ,p 5n) reaction. Based on the experimentally estimated ^{203}Pb/^{203}Bi nuclei number (or activity) ratio at the moment when the irradiation was stopped, it is possible to estimate how much of the ^{203}Pb activity originates from (γ,p 5n) reaction. It was obtained that the number of produced ^{203}Pb nuclei is about 7.6% of the number of ^{203}Bi nuclei. Based on this value, it was established that the rate of production of ^{203}Pb through (γ,p 5n) reactions is about 6% of the rate of production of ^{203}Bi by (γ,6n) reaction. This is significantly less than presented in reference [11].

In order to verify the obtained results, a TALYS simulation of cross-sections for (γ,p 5n) and (γ,6n) reaction was performed. According to the obtained cross-sections, ratio of reaction rates of both reactions was calculated using one model of strength function and six models of level density function. Due to large scatter of TALYS cross sections for both reactions obtained results of ratios of reaction rates differ significantly.

This approach neither verified nor contradicted certain indications [11] that TALYS code underestimates the probability of photonuclear events in which a proton is realized in addition to neutrons. A new experiment that primarily makes advantage of wider range of energies, preferably higher than 60 MeV and a more detailed analysis of theoretical results and TALYS simulations may eliminate this doubt. The new measurements will also help to resolve the dilemma of whether the yield of ^{209}Bi(γ,p 5n)^{203}Pb reaction is significantly higher than the yield of the ^{209}Bi(γ,6n)^{203}Bi reaction, as stated in reference [11], or whether this ratio is significantly lower, as obtained in the measurements described in this paper. The method described in this study is not limited to the case of ^{209}Bi, and can be applied to other targets and products of photonuclear reactions.

Data Availability Statement This manuscript has no associated data or the data will not be deposited. [Authors' comment: This manuscript has no associated data.]

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