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Production of ^{117m}Sn and ^{119m}Sn by photonuclear reactions on natural antimony

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ABSTRACT

Natural antimony targets were irradiated in a 60 MeV bremsstrahlung beam and gamma spectrometric measurements were performed. The goal was to establish the yield of ^{117m}Sn, a radionuclide with great potential for application in medicine. Considering that ^{117m}Sn is predominantly produced through a photonuclear reaction in which an charged particle is emitted (^{121}Sb(γ,p3n)), the yield of this tin isotope is much lower than the yields of several antimony isotopes produced in (γ,xn) reactions. It has been estimated that photonuclear reactions on natural antimony could produce ^{117m}Sn activities needed for therapeutic applications, with accelerators having electron currents of the order of mA. For the used bremsstrahlung energy of 60 MeV, it was estimated how much ^{119m}Sn activity can be expected when exposing the antimony target.

1. Introduction

Photonuclear reactions represent a very interesting field in which the nucleus, as a system determined by a strong interaction, is subjected to electromagnetic forces. These reactions are conducted by the interaction of high-energy electromagnetic radiation (10 MeV and more) with nuclei of the selected target. The process of the highest probability is emission of one neutron. If the energy of the electromagnetic radiation is high enough, the excited nucleus can emit two or more neutrons, with lower probability. Protons and other charged particles can leave the nucleus, but with a much lower probability due to the effect of the Coulomb barrier.

Electromagnetic nature of interaction makes photonuclear reactions a suitable method for studies of the nucleus and some of its properties. Photonuclear reactions have become a convenient method in number of basic research (Pietralla et al., 2019; Zilges et al., 2022). Besides that, they could be quite acceptable way for production of radionuclides. The need for the production of radionuclides used in medicine is particularly important (Qaim, 2017). In currently established practice, a certain number of neutron-deficient radionuclides and positron sources are usually produced by proton cyclotrons (IAEA radioisotopes and radiopharmaceuticals reports No, 2021). However, some of them can be obtained through photonuclear reactions (Starovoitova et al., 2015; Inagaki et al., 2019; Kazakov et al., 2021).

The main objective of this work is to check the feasibility of producing ^{117m}Sn by photonuclear reactions. The tin isotope ^{117m}Sn has shown extremely promising properties in the process of theranostics (Lewington, 2005). The 158.562 keV gamma radiation is emitted after de-excitation of the ^{117m}Sn isomeric state. Moderate attenuation in soft tissues and large cross sections for interaction with NaI (XCOM, 2010) make 158.562 keV photons very suitable for SPECT diagnostic purposes. The large number of conversion electrons (113% emission probability), (Blachot, 2002) can provide a high local dose at some specific location where the radiopharmaceutical labeled with this isotope would be accumulated.

To obtain high specific activities of ^{117m}Sn, various nuclear reactions using charged particles were taken in consideration (Stevenson et al., 2015). The two most important directions are using proton beams (Ermolaev et al., 2009) and alpha particles (Maslov et al., 2011; Ditrói et al., 2016; Duchemin et al., 2016; Aikawa et al., 2018; Aslam et al.,

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2018). Photonuclear reactions for the production of ^{117m}Sn are not frequently studied. A couple of attempts have been performed to get ^{117m}Sn by (γ,γ`) reaction on enriched ^{117}Sn target (Aksenov et al., 1992).

In the present paper, the possibility of producing ^{117m}Sn by photonuclear reactions on a target made of natural antimony with a 60 MeV bremsstrahlung beam has been investigated experimentally. One of the main goals is to estimate the yield of this radionuclide.

Natural antimony contains two isotopes, ^{121}Sb and ^{123}Sb. For the purposes of this study, the reactions ^{121}Sb(γ,p3n) and ^{123}Sb(γ,p5n) are considered. Photonuclear reactions in which no charged particles are emitted have higher cross sections than reactions with emission of charged particle (EXFOR Data Tables), due to the absence of a Coulomb barrier. Unfortunately, ^{121}Sb(γ,4n) and ^{123}Sb(γ,6n), do not lead to the creation of ^{117m}Sn after beta decay (EC) of produced ^{117}Sb (Blachot, 2002).

It is inevitable that some other active radionuclides are formed during the exposure of a natural antimony target to a beam of high-energy photons. It is particularly interesting to see to what extent the produced ^{117m}Sn would be contaminated by the activity of other products of possible photonuclear reactions. In this work, special attention is paid to ^{119m}Sn, which would be created through ^{121}Sb(γ,pn) and ^{123}Sb(γ, p3n) photonuclear reactions.

2. Materials and methods

2.1. Expected nuclear reactions

Natural antimony consists of two isotopes: ^{121}Sb (57.36%) and ^{123}Sb (42.64%) (Chu et al., 1999). Relevant part of the isotope chart is presented in Fig. 1. During exposure of a natural antimony target to a flux of high-energy photons, several types of nuclear reactions occur.

a) (γ,n) and (γ,xn) reactions

Several antimony isotopes, (depending on the endpoint energy of the bremsstrahlung beam used in experiment) could be produced in reactions with emission of one or more neutrons. Both ground state and Sb isomers are created. Besides ^{122}Sb, which decays mostly (97.6%) to ^{122}Te through the emission of a beta particle, all other Sb isotopes decay through electron capture (Chu et al., 1999). It can be seen (Fig. 1) that in this way stable tin nuclei will be formed. Two of the stable tin isotopes have isomers ^{117m}Sn and ^{119m}Sb, however decays of ^{117}Sb and ^{119}Sb do not populate their metastable states. This means that no Sn activity should be expected after decay of antimony isotopes created in (γ,n) and (γ,xn) reactions. If Sn fraction is chemically extracted from the irradiated antimony target, the output of (γ,n) and (γ,xn) reactions can estimate how much non-active Sn would be obtained together with ^{117m}Sn, produced in some other reactions.

b) (γ,p) and (γ,pxn) reactions

If one proton (with or without neutrons) is emitted during the irradiation of the Sb target, tin isotopes will be formed. All of Sn isotopes created in this way are stable, with the exception of ^{121}Sn. This isotope is formed in the ^{123}Sb(γ,pn) reaction in the ground state, but also as an isomer. After decay of the ground state of ^{121}Sn, beta radiation of relatively low energy (Q value: 390.1 keV) is emitted without the emission of gamma radiation. Considering that the time dynamics of the creation of a product of a nuclear reaction can be described by a function $(1 - e^{-\lambda t_{irr}})$ and that ^{121m}Sn has a half-life of 55 years, low activity would be created in photon beam in some realistic case when the irradiation time is of the order of hours or shorter. Additionally, it could be expected that ^{119m}Sn and ^{117m}Sn isomers will be created (half lives 293.1 days and 13.6 days, respectively). ^{119m}Sn can be formed in ^{121}Sb(γ,pn) and ^{123}Sb (γ,p3n) reactions. Another isomer, ^{117m}Sn can be produced in ^{121}Sb(γ, p3n) and ^{123}Sb(γ,p5n) reactions, if the endpoint energy of bremsstrahlung is high enough.

^{117m}Sn has very suitable characteristics for use in medicine. The scheme of de-excitation of the metastable state is shown in Fig. 2. The gamma radiation energy emitted (158.56 keV) is almost ideal for SPECT diagnostic procedures (Duchemin et al., 2016). The transitions of 314.3 keV and 156.2 keV are almost entirely realized by the emission of conversion electrons. It was determined that 1.13 conversion electrons are emitted per one decay of ^{117m}Sn (Ermolaev et al., 2009; Aslam et al., 2018). Due to the low penetrating power, these electrons in the tissues

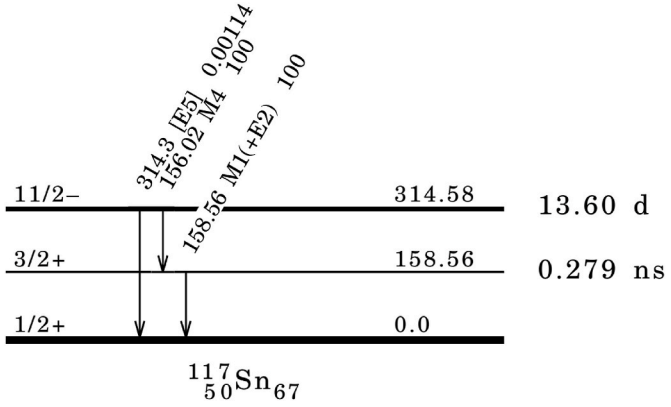

Fig. 2. Decay scheme of ^{117m}Sn. For absolute intensities of transitions, multiply by 0.864 (Firestone 1996).

Sb116 15.8 m 3^+ EC	**Sb117** 2.80 h $5/2^+$ EC	**Sb118** 3.6 m 1^+ EC	**Sb119** 38.19 h $5/2^+$ EC	**Sb120** 15.89 m 1^+ EC	**Sb121** Stable $5/2^+$ 57.36 %	**Sb122** 2.7238 d 2^- EC, β^-	**Sb123** Stable $7/2^+$ 42.64 %
Sn115 Stable $1/2^+$ 0.34 %	**Sn116** Stable 0^+ 14.53 %	**Sn117** Stable $1/2^+$ 7.68 %	**Sn118** Stable 0^+ 24.23 %	**Sn119** Stable $1/2^+$ 8.59 %	**Sn120** Stable 0^+ 32.59 %	**Sn121** 27.06 h $3/2^+$ β^-	**Sn115** Stable $1/2^+$ 0.34 %
In114 71.9 s 1^+ EC, β^-	**In115** 4.41E+14y $9/2^+$ β^-, 95.7 %	**In116** 14.10 s 1^+ EC, β^-	**In117** 43.2 m $9/2^+$ β^-	**In118** 5.0 s 1^+ β^-	**In119** 2.4 m $9/2^+$ β^-	**In120** 3.08 s 1^+ β^-	**In121** 23.1 s $9/2^+$ β^-

Fig. 1. Part of the isotope chart presenting Sb and Sn isotopes of interest (taken from Nuclear charts in (Chu et al., 1999)). Listed are, from top to bottom: isotope, half-life, ground state spin, and decay mode, or natural isotope abundance for stable ones.

deposit their energy in a small space around the location where the radiopharmaceutical labeled with ^{117m}Sn accumulates (Lewington, 2005; Stevenson et al., 2015). In this way, a therapeutic dose can be deposited in a controlled area.

The decay scheme of ^{119m}Sn is shown in Fig. 3. During the de-excitation of the metastable state of 89.53 keV, gamma radiation of energy 23.87 keV is emitted. It is determined that 0.161 photons are emitted per decay (Symochko et al., 2009). Such low energy photon radiation has very little chance of leaving the patient's body, so it cannot be used for diagnostic purposes in nuclear medicine (half-value thickness of 23.98 keV gamma radiation in water is 1.25 cm according to (XCOM, 2010)). It was measured that the transitions shown in Fig. 3 are realized to the greatest extent by the emission of conversion electrons. It is determined that 1.83 conversion electrons are emitted per ^{119m}Sn decay (Symochko et al., 2009). These electrons could also play a significant role in radionuclide therapy.

c) (γ,α), (γ,αn) and (γ,αxn) reactions

Additional interesting outcome of the photoreactions, which could be expected during exposition of antimony target in high energy photon beams, is emission of alpha particle, with or without neutrons. Reactions (γ,α), (γ,αn), (γ,α2n), etc. will produce indium isotopes. In the simplest case, when just an alpha particle is emitted after interaction of high energy photons with ^{123}Sb, the ^{119}In isotope would be created. It is the heaviest isotope of indium that can be formed in this way. Considering that the half-life of ^{119}In is 2.4 min, it is unlikely that it will be identified in an off-line gamma radiation measurement. The same is with ^{118}In, which has a half-life of 5 s. Just ^{117}In and lighter isotopes could be detected in some standard off-line gamma spectroscopy measurements. The decay of the ground state of ^{117}In populates the isomeric state ^{117m}Sn to a very small extent (0.344%), while the decay of the isomer ^{117m}In completely bypasses it (Blachot, 2002).

It should be emphasized that after the decay of ^{117}Sb, as well as the decay of both the ground and isomer states of ^{117}In, the emission of photon radiation of 158.56 keV occurs. This is also the only gamma radiation emitted after de-excitation of the 314.58 keV isomeric state of ^{117m}Sb, which complicates the interpretation of gamma spectra recorded immediately after the irradiation of the antimony target. The half-life of ^{117}Sn is 2.8 days, while ^{117}In and ^{117m}In have half-lives of 43.2 min and 116.2 min, respectively, which is significantly shorter than the half-life of ^{117m}Sn. In repeated measurements after a sufficiently long time interval, the presence of ^{117m}Sn in the irradiated sample can be determined.

d) (n,γ) reactions

The production of bremsstrahlung radiation with endpoint energies higher than the binding energy of neutrons in the nuclei of irradiated materials leads to the emission of neutrons. The sources of neutrons are usually a bremsstrahlung converter, collimators, filters and all other materials affected by the photon beam. These neutrons can be captured by the nuclei of the target material. It is most likely that ^{122}Sb and ^{124}Sb are formed in the interactions of neutrons with Sb target nuclei. The easiest way to assess the presence of neutrons at the location of the irradiated target is to identify the gamma lines of ^{124}Sb. The half-life of this radionuclide is 60.20 d, so for its identification purposes, the spectrum recorded three days after irradiation was used. Only two gamma lines of 602.73 keV and 1690.98 keV were observed in the spectrum. The quantum yields of these two lines are 97.8% and 47.3% respectively (Chu et al., 1999). The intensities of these gamma lines in the measured spectrum are four orders of magnitude lower than the ^{120m}Sb gamma lines, which have a similar quantum yield. In addition, in the spectrum of the gold foil exposed together with the Sb target, a gamma line of 411.8 keV can be observed. This gamma transition originates from ^{198}Au produced by the capture of neutrons on ^{197}Au. Based on these three very weak lines, it was estimated that the neutron capture does not give activities that can be compared with the activities obtained through photonuclear reactions and that the contribution of (n, γ) reactions can be neglected.

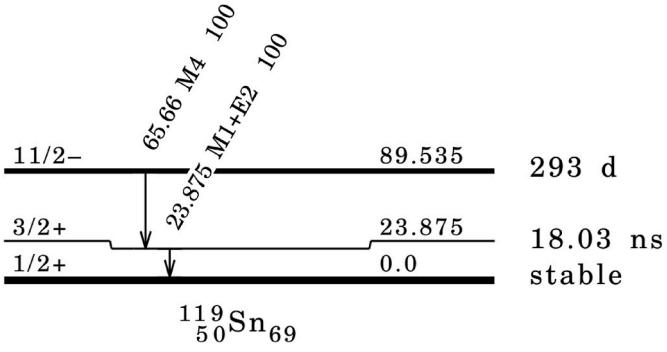

Fig. 3. Decay scheme of ^{119m}Sn (Firestone 1996).

2.2. Reaction yields

Considering that natural antimony consist of two stable isotopes, ^{121}Sb (57.36%) and ^{123}Sb (42.64%), in some cases, one product of photonuclear reaction can be formed in two different ways. Total activity of observed product can be result of two reactions, whose probabilities are defined by the energy differential cross sections $\sigma_i^{121}(E)$ and $\sigma_i^{123}(E)$ for an observed i-th reaction. The atomic number of parent nuclei is denoted in superscript. The frequently used equation for calculating the saturation activity of the product obtained in a nuclear reaction (Krmar et al., 2004) can be somewhat corrected in the case of irradiation of an antimony target to express the yield of produced isotope:

$$\frac{m_t}{M}N_{av}\left(0.5726\int_{E_t^i}^{E_{max}}\sigma_i^{121}(E)\cdot\Phi(E)\cdot dE+0.4264\int_{E_t^i}^{E_{max}}\sigma_i^{123}(E)\cdot\Phi(E)\cdot dE\right)=\frac{N_\gamma\,\lambda}{\varepsilon\,p_\gamma e^{-\lambda\Delta t}\,(1-e^{-\lambda t_{irr}})(1-e^{-\lambda t_m})} \tag{1}$$

where m_t and M are the mass of the exposed target used in experiment and the atomic mass number, N_{av} is Avogadro number, E_t^i is the energy threshold for observed nuclear reaction and E_{max} is the maximal energy of photons, $\Phi(E)$ is flux of photons, N_γ is the number of detected gamma photons of chosen energy, λ is the decay constant, ε is absolute peak efficiency of the detector at the chosen energy, p_γ is the quantum yield of detected photons, Δt, t_{irr} and t_m are cooling, irradiation and measurement time, respectively.

For brevity, the above equation can be represented as:

$$\frac{m_t}{M}N_{av}\left(0.5726\,R_i^{121}+0.4264\,R_i^{123}\right)=\frac{N_\gamma\,\lambda}{\varepsilon\,p_\gamma e^{-\lambda\Delta t}\,(1-e^{-\lambda t_{irr}})(1-e^{-\lambda t_m})} \tag{2}$$

Integrals denoted by symbol R are called saturation activities. A common way is to divide Equation (1) or Equation (2) by the term $\frac{m_t}{M}N_{av}$. In this way, the right-hand side of the equation yields the total saturation activity that can be determined from gamma spectroscopic measurements. In our case, it would be the sum of two saturation activities weighted by the abundances of isotopes ^{121}Sb and ^{123}Sb in the natural element.

Immediately after irradiation of the antimony target, several gamma spectra should be recorded, to identify short-living products of photonuclear reactions. It is very important to record gamma spectra of antimony target several days after irradiation, to get evidence about

long-living radionuclides. The next step would be to determine the intensities of the characteristic gamma lines in the spectrum, in order to determine the experimental values of the yields (or saturation activity) of the identified products based on the right side of Equation (1).

In this way, the output of all those photonuclear reactions that produce an unstable nucleus that emits gamma radiation of sufficiently high energy and intensity can be determined. There are several photonuclear reaction products whose yields cannot be determined from measured gamma spectra. Such is, for example, the isomer ^{119m}Sn reaching the ground state by the emission of low-energy gamma radiation of 23.87 keV, which could not be measured with the available detector. In this case, the yield of ^{119m}Sn can estimated computationally, as shown in the left side of Equation (1). For the purposes of this evaluation, it is necessary to have the energy differential cross sections $\sigma_i^{121}(E)$ and $\sigma_i^{123}(E)$ of the reaction that produces the observed nucleus, as well as the value of the photon flux $\Phi(E)$.

2.3. Ratio of ^{117m}Sb and ^{119m}Sb activities

The aim of this work is to check the possibility of producing ^{117m}Sb through photonuclear reactions. Based on Section 1.1, it can be expected that in the case of complete chemical separation of the tin fraction from the irradiated Sb target, the activity of ^{117m}Sn and the gamma line at 158.56 keV would remain. This certainly does not mean that ^{117m}Sn is the only active isotope of tin that can be expected in this experiment. During the exposure of the antimony target, ^{119}Sb is formed as a product of ^{121}Sb(γ,2n) and ^{123}Sb(γ,4n) reactions. The half-life of this antimony isotope is 38.19 h. It decays through an electron capture and populates the 23.87 keV excited state in ^{119}Sn. The gamma radiation emitted by the de-excitation of this state is not noticeable in the measured spectra due to the low efficiency of the available detector in the low-energy region. The ^{119}Sn nucleus has a metastable state of 89.53 keV, but it is not populated by the decay of ^{119}Sb.

There is a possibility that the creation of ^{119m}Sn also occurs through ^{121}Sb(γ,pn) and ^{123}Sb(γ,p3n) reactions. Considering that the cross section for photonuclear reactions decreases very sharp with the number of emitted particles (Dietrich and Berman, 1988; Ermakov et al., 2010) it can be considered that the probability of creation of ^{119m}Sn is higher than the probability of creation of ^{117m}Sn.

The low energy of the photon radiation emitted by ^{119m}Sn did not provide the possibility to determine the yield of this isomer in the current experiment. The only possibility is to determine the activity ratio of the isotopes ^{117m}Sn and ^{119m}Sn with the help of theoretical estimates of the cross sections for the relevant nuclear reactions.

Equation (2) for the example of production ^{117m}Sn can be written in a slightly different form:

$$\frac{m_t}{M}N_{av}\left(1-e^{-\lambda_{117}t_{irr}}\right)\left(0.5726\,R_{\gamma,p3n}^{121}+0.4264\,R_{\gamma,p5n}^{123}\right)=\frac{N_\gamma\,\lambda_{117}}{\varepsilon p_\gamma e^{-\lambda_{117}\Delta t}\left(1-e^{-\lambda_{117}t_m}\right)} \tag{3}$$

where the subscript of saturation activity R denotes the nuclear reaction producing the observed radionuclide. Both the left and right sides of Equation (3) give the activity of ^{117m}Sn at the end of the irradiation. The left-hand side of Equation (3) provides a way to estimate the activity from known cross-section and photon flux values, while the right-hand side of Equation (3) allows the activity to be calculated using data obtained from the gamma spectrum.

An analogous equation can be written for the production of ^{119m}Sn. The activity ratio of ^{117m}Sn and ^{119m}Sn can be represented as:

$$\frac{A_{119}}{A_{117}}=\frac{\lambda_{119}}{\lambda_{117}}\,\frac{0.5726\,R_{\gamma,pn}^{121}+0.4264\,R_{\gamma,p3n}^{123}}{0.5726\,R_{\gamma,p3n}^{121}+0.4264\,R_{\gamma,p5n}^{123}} \tag{4}$$

The above Equation (4) was written after the exponential function on the right-hand side of Equation (3) was developed into a series, discarding all but the linear terms. Considering that the sample irradiation time $t_{irr} = 30\ m$ is significantly shorter than the half-life of the observed photonuclear reaction products (13.6 d and 293 d) second, the quadratic term is three orders of magnitude smaller than the linear one.

In order to obtain an estimate of the activity ratio, it is necessary to calculate the saturation integrals of the relevant nuclear reactions. For the bremsstrahlung production geometry used in this experiment, the photon flux is obtained by a GEANT4 simulation. Considering that it is necessary to determine the ratio of ^{119m}Sb and ^{117m}Sn activities, it is quite sufficient to know only the shape of the photon spectrum. The shape of the photon spectrum $\Phi(E)$ is obtained by using the GEANT4 software package [13], version v11.1.0, with standard G4 electromagnetic physics option selected. The simulation starts with creating $30{\cdot}10^6$ of 60 MeV electrons in the beam, with a Gaussian spread in energy of 0.01 MeV. The photon spectrum is obtained at the place of irradiated sample based on the geometry used in this experiment.

There are no experimental values of the differential cross section for the observed nuclear reactions in EXFOR database, so the only possibility is to use theoretical estimates.

3. Measurements and results

A pure natural antimony target was exposed to a beam of bremsstrahlung radiation with a maximum energy of 60 MeV. Only one endpoint energy was used in this experiment. The experiment was carried out using the linear electron accelerator LUE-75 located at A. Alikhanyn National Science Laboratory in Yerevan, Armenia. Accelerated electrons, after passing through a cylindrical collimator (length of 20 mm, diameter of 15 mm) strike a pure tungsten convertor. The thickness of the convertor was 2 mm. A 30 mm long aluminum cylinder was placed directly behind it. The function of the aluminum was to stop the electrons that penetrated the tungsten. At a distance of 60 mm from the tungsten plate, a disc-shaped antimony target (diameter of 1 cm and a mass of 0.5772 g) was placed. Duration of irradiation was $t_{irr} = 1800\ s$. The current of accelerated electrons, monitored by Faraday cage was 1.2 μA in average.

Exposed Sb disc was transferred to the room where the single vertical dipstick HPGe detector was located. Measurements were performed in geometry where Sb was placed 86 mm from the top of the detector. The spectrum used in this work was recorded for $t_m = 53599$ s, and cooling time was $\Delta t = 1200\ s$. In the measured spectra, gamma lines from several Sb isotopes were identified, as well as several gamma lines of indium isotopes. Some relevant gamma lines used in further calculations are presented in Fig. 4.

The relative efficiency of the detector was determined using the ^{206}Bi gamma lines. For the purposes of the experiment, simultaneously with the antimony target, a cylindrical bismuth sample of the same dimensions, weighing 1.1 g, was exposed in the photon beam, behind the Sb sample. Bismuth irradiation conditions are not overly significant since the only goal was to obtain measurable activity of the isotope ^{206}Bi.The photonuclear reaction ^{209}Bi(γ,3n)^{206}Bi gave sufficient activity to obtain a satisfactory number of very intense gamma lines in the spectrum recorded the next day for 24526 s. Nineteen gamma lines from the spectrum of ^{206}Bi in the interval from 183.98 keV to 1878.65 keV were used to calculate the relative photo-peak efficiency of the HPGe detector for the used counting geometry. The relative efficiency curve was obtained by fitting a function that was a combination of an exponential and a third-order polynomial ($\varepsilon_{rel} = \exp(a + b \bullet E + c \bullet E^2 + d \bullet E^3)$). With the ^{137}Cs calibration source, it was established that the absolute photopeak efficiency at the energy of 661.66 keV for the given geometry is 0.00147(1). This value was used to normalize the relative efficiency obtained with ^{206}Bi.

Three days (264960 s) after the first measurement, the second one was performed for a 65125 s. The time between the first and second measurements was long enough for ^{117}Sb, ^{117}In and ^{117m}In to completely decay. In that case, all photons of energy 158.56 keV originate from the

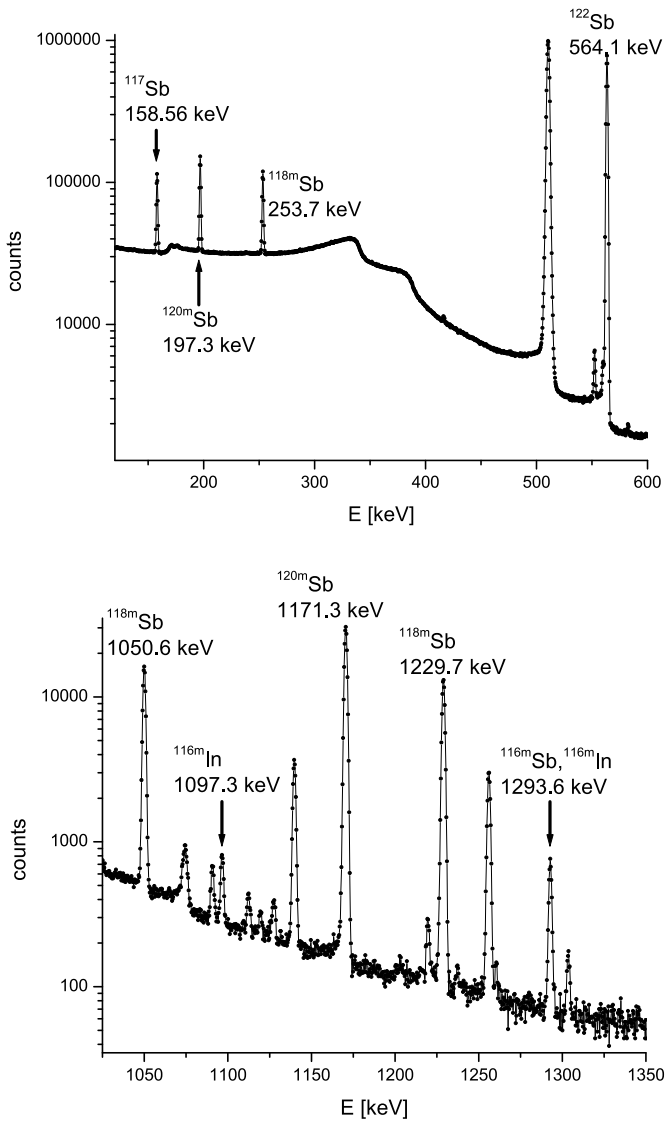

Fig. 4. Two segments of gamma spectra containing gamma lines used in calculations.

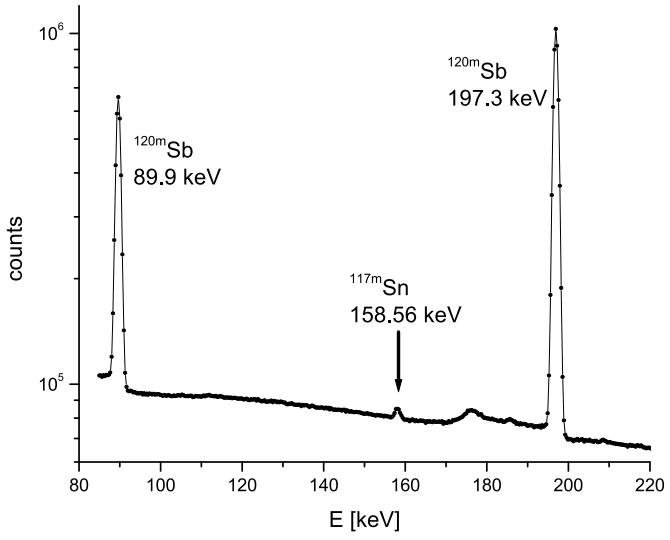

Fig. 5. Part of the gamma spectra recorded three days after irradiation of Sb target.

decay of ^{117m}Sn only. Part of the spectra containing this line is depicted in Fig. 5.

The intensities of selected gamma lines were determined using the GENIE (Genie2000 Spectroscopy Software, 2004) software package. The yields of identified radionuclides were calculated as shown on the right side of Equation (1). Obtained results are depicted in Table 1. Uncertainties are given in parentheses. The main contribution to them is from the statistical uncertainty of the intensity of the observed gamma lines N_γ and from the uncertainty of the absolute detection efficiency ε. The exception is ^{117}Sb, for which the gamma line intensity is determined by subtracting the estimated contribution of ^{117m}Sn, as explained later.

As could be expected, the highest yield among Sb isotopes has reaction products formed after the emission of the smallest number of neutrons. Table 1 shows that ^{122}Sb and ^{120m}Sb have the highest yield. With the increasing number of emitted particles, the probability of a photonuclear reaction drops sharply, so the yield of lighter antimony isotopes is also lower.

The ^{116m}In yield was determined based on the obtained gamma line intensity of 1097.33 keV. Considering that both ^{116m}In and ^{116m}Sb populate the same excited states of ^{116}Sn, 1293.56 keV gamma photons are emitted after the decay of both of these radionuclides. The yield of ^{116m}Sb was determined based on the intensity of the gamma line of 1293.56 keV, but the contribution of ^{116m}In was subtracted. Mentioned gamma lines can be seen in Fig. 4.

The yields of ^{117}Sb and ^{117m}Sn were determined based on the intensity of the gamma line of 158.56 keV. In the first measurement, started 3 h and 20 min after the end of the irradiation, the largest contribution to this gamma line is from ^{117}Sb. After three days, this radionuclide completely disappeared and all detected photons of this energy can be attributed to ^{117m}Sn. Based on the intensity of the gamma line of 158.56 keV from the second spectrum, it was determinate that count rate of ^{117m}Sn was 0.374 s^{-1}. The count rate of ^{117m}Sn three days earlier was calculated in order to assess the contribution of this tin isotope to the total intensity of the gamma line. It was obtained that the intensity of the gamma line of 158.56 keV came predominantly from the decay of ^{117}Sb in the first spectrum. The contribution of ^{117m}Sn to the total intensity of the 158.56 keV gamma line is estimated at 0.7%, which is still an order of magnitude higher than its statistical uncertainty. Based on the intensity of the corrected 158.56 keV gamma line, the yield of ^{117}Sb shown in Table 1 was determined.

The ratio of ^{119m}Sn and ^{117m}Sn activities was determined as shown in Equation (4). It is not necessary to know the absolute value of the photon flux. Only the shape of the photon spectrum obtained by GEANT4 simulation for a given geometry of bremsstrahlung production is necessary.

Cross sections for observed reactions were estimated using the

Table 1
Yields and sum of saturation activities of identified products of photonuclear reactions.

	Half-life	Ways of production	Yield $\cdot 10^6$ [s^{-1}]	Sat. Act. $\cdot 10^{-15}$ [s^{-1}]	Activity10^4 [s^{-1}]
^{122}Sb	2.70 d	^{123}Sb(γ,n) +^{121}Sb(n, γ))	75.9(11)	26.6(5)	38.5(5)
^{120m}Sb	5.76 d	^{121}Sb(γ,n) + ^{123}Sb(γ,3n)	6.05(9)	2.12(4)	1.53(2)
^{118m}Sb	5.00 h	^{121}Sb(γ,3n) + ^{123}Sb(γ,5n)	0.456(9)	0.160(3)	3.55(7)
^{117}Sb	2.80 h	^{121}Sb(γ,4n) + ^{123}Sb(γ,6n)	1.26(6)	0.44(2)	14.7(7)
^{116m}Sb	60.3 min	^{121}Sb(γ,5n) + ^{123}Sb(γ,7n)	0.026(1)	0.0091(4)	0.75(3)
^{116m}In	54.4 min	^{121}Sb(γ,αn)	0.0027 (2)	0.00095(6)	0.14(1)
^{117m}Sn	13.60 d	^{121}Sb(γ,p3n) + ^{123}Sb(γ,p5n)	0.092(4)	0.0322(14)	0.0100(5)

TALYS 1.96/2.0 code (Koning et al., 2023). It has been shown (Gyürky et al., 2014) in the case of nuclear reactions in which isomers are created that TALYS estimates can differ from experimental results. Some scatter in the values of physical quantities determined using TALYS estimates can also be expected (Jovančević et al., 2024). But for a first estimate of the activity ratio A_{119}/A_{117} given in Equation (4), until the result of experimental measurements is obtained, the prediction based on TALYS can serve well. It could be expected that the choice of the strength function model has an impact on the estimation of the cross section, but this analysis is beyond the scope of this paper. It was decided to use SMLO model for a strength function. On the example of the creation of ^{113m}In in photonuclear reactions, it was shown that this model gives good results (Versteegen et al., 2016). 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 (6) Temperature-dependent Gogny-Hartree-Fock-Bogoluybov models) of level density. Default OMP was used in calculations. Different models for the densities of states give estimates of cross sections that can differ significantly from each other. In Figs. 6 and 7 are depicted the energy differential cross sections for the two reactions at ^{121}Sb which give ^{119m}Sn and ^{117m}Sn as a result.

The estimation of the activity ratio of ^{119m}Sn and ^{117m}Sn at the end of the irradiation was made using all six cross sections obtained by the TALYS code. The smallest value of 0.124 is obtained when the model of Fermi Gas + Constant Temperature Model is chosen for the density of states, while The Generalized Superfluid Model gives the highest ratio estimate of 0.268.

4. Discussion

Based on the results shown in Tables 1, it can be seen that ^{121}Sb(γ, p3n) and ^{123}Sb(γ,p5n) nuclear reactions, give a low yield of ^{117m}Sn comparing to the activities of the produced antimony isotopes. The right side of Equation (3) is used to calculate the activities of radionuclides identified in the measured spectra. Obtained results are presented in Table 1, last column. During the irradiation of the antimony target, an activity of ^{117m}Sn of about 100(5) Bq was obtained. This is three orders of magnitude less than the activity of Sb isotopes that have been identified. Considering that the formation of ^{117m}Sn occurs with the emission of charged particles, such a low level of activity could be expected.

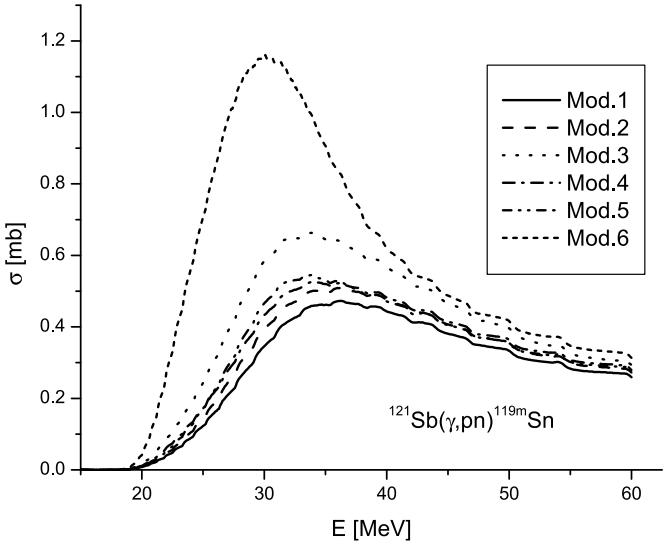

Fig. 6. Cross sections for ^{121}Sb(γ,pn)^{119m}Sn reaction estimated using TALYS code.

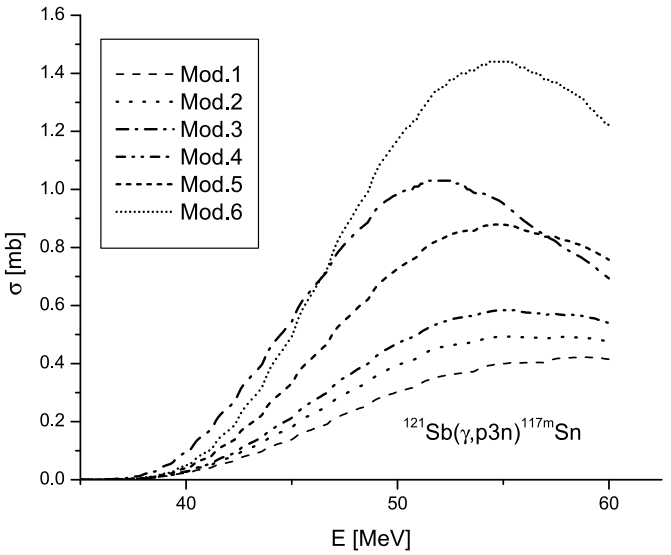

Fig. 7. Cross sections for ^{121}Sb(γ,p3n)^{117m}Sn reactions estimated using TALYS code.

The method currently considered the most promising is the production of ^{117m}Sn by interactions of accelerated alpha particles with cadmium (natural Cd or ^{116}Cd enriched) or indium targets (Maslov et al., 2011; Ditrói et al., 2016; Duchemin et al., 2016; Aikawa et al., 2018; Aslam et al., 2018), using. Since the range of alpha particles in these materials is very small, order of microns, the most common way to show the output of reaction is through the integral yield or Thick Target production Yield (TTY), as it is called by some authors. This quantity is expressed in units of kBq/μAh. A very complete review of theoretical predictions as well as known experimental results of ^{117m}Sn yields can be found in refs. (Rebeles et al., 2008; Ditrói et al., 2016; Aslam et al., 2018). The experimental results of ^{117m}Sn yields (Maslov et al., 2011) of 37.5 kBq/μAh and 410 kBq/μAh obtained on natural and enriched ^{116}Cd targets, respectively, can be added to this review. The mechanism of photon interaction with metal targets is significantly different and they take place throughout the entire depth of the target and reaction yield depends on the target mass and thickness. However, from the estimated activity of ^{117m}Sn, a numerical value can be obtained that could somehow be equivalent to TTY, at least for the geometry used in experiment. With the experimental parameters listed in Section 2, it can be estimated that the production of ^{117m}Sn in photonuclear reaction is 0.174(8) kBq/(μAh).

Such an assessment could be expected taking into consideration the large difference in the values of the cross section for ^{116}Cd(α,3n)^{117m}Sn and ^{121}Sb(γ,p3n)^{117m}Sn reactions. The maximum cross-section for the ^{116}Cd(α,3n)^{117m}Sn reaction is about 1 b in the region of 35 MeV (Montgomery and Porile, 1969; Rebeles et al., 2008; Ditrói et al., 2016; Duchemin et al., 2016). From Fig. 6 it can be seen that the theory for the ^{121}Sb(γ,p3n)^{117m}Sn reaction predicts a cross section of less than 1 mb.

The experiment showed that with an antimony target weighing slightly more than half a gram, in a photon beam produced with an electron current of 1.2 μA during 30 min of exposure, a very low activity of ^{117m}Sn is obtained, the order of magnitude being 0.1 kBq. For the purposes of medical application of this radionuclide, significantly greater activities are required. Standard therapy dose of ^{117m}Sn is usually 1 GBq for 70 kg patient (Srivastava et al., 1998). More ^{117m}Sn activity can be obtained by increasing: target mass, irradiation time and electron current. The obtained activity depends linearly on the mass and electron current and in this case also on the irradiation time, since the half-life of ^{117m}Sn is 13.6 days. It can be expected that an increase in the maximum bremsstrahlung energy can give a growth of the obtained

activity of the product of the photonuclear reaction as well. In any case, there is room for increasing the yield of ^{117m}Sn, which still needs to be investigated.

According to the estimates based on Equation (4), it can be expected that the activity of the long-lived ^{119m}Sn will be even lower and that in the bremsstrahlung beam of maximum energy of 60 MeV it will be up to 26% of the activity of ^{117m}Sb. This ratio can be expected to be smaller at higher bremsstrahlung energies. It can be seen in Fig. 7, that some significant cross section component at energies higher than 60 MeV for the reaction (γ,p3n) exist. Considering that the dominant way of production of ^{117m}Sn is through this reaction, a higher yield of this radionuclide can be expected at higher energies.

If there is an effective method to chemically separate the tin fraction from the antimony target, the activity of the ^{117m}Sn and ^{119m}Sn isotopes would be present in the obtained material only. This can be a very convenient way to produce two radionuclides of high specific activity (activity per unit mass), since a source of radiation without a carrier would be obtained. In photonuclear reactions on natural antimony, several isotopes of Sb are created. All of them, after decay give Sn nuclei as product. In this way, stable isotopes of tin with two active isomers ^{119m}Sn and ^{117m}Sn are created.

The mass of one of the isotopes of tin accumulated over time t by the decay of an unstable isotope of antimony created in a nuclear reaction could be expressed as:

$$m = \frac{M}{N_{av}} \frac{A_0}{\lambda} \left(1 - e^{-\lambda t}\right) \quad (5)$$

where A_0 is activity of observed Sb isotope at the end of irradiation. This activity can be calculated using right side of Equation (3).

The measured activities of ^{122}Sb, ^{120m}Sb, ^{118m}Sb, ^{117}Sb and ^{116m}Sb were used for the approximate estimation of the amount of tin that is created from the decay of Sb isotopes. This estimation do not include all the ways of creating tin from the decay of antimony isotopes, but only those whose activity could be determined based on the measured gamma spectra. However, it can be enough to estimate the order of magnitude of the mass of tin. Relevant gamma lines in registered spectra can be seen in Fig. 4.

In a completely arbitrarily chosen case where the decay of the created antimony isotopes lasted 24 h after irradiation, it was found that the total mass of tin that was created by decay of ^{122}Sb, ^{120m}Sb, ^{118m}Sb, ^{117}Sb and ^{116m}Sb is 8.1(4) • 10^{-12} g. This means that by photonuclear reactions on natural antimony, very high specific activities of ^{117m}Sn can be obtained, order of magnitude up to 10^7 MBq/g.

5. Conclusions

In the experiment described in this paper, photoactivation of a target made of natural antimony was performed in a bremsstralung beam with a maximum energy of 60 MeV, followed by gamma spectrometric measurements. As could be expected, the highest activity in the activated target was registered by neutron-deficient isotopes of antimony, obtained in (γ,xn) reactions.

These isotopes of antimony decay mainly through electron capture and give stable isotopes of tin as a product. The only tin activity detected in the experiment came from ^{117m}Sn. In the experiment ^{119m}Sn was produced also, but the available detector could not detect it due to low-energy of emitted gamma radiation.

In the gamma spectroscopic measurement, which was performed after a few days, when all sources of interference disappeared, the activity of ^{117m}Sn, which was created in the irradiation of the target, was determined. In the described experiment, the yield of ^{117m}Sn that was obtained (0.174(8) kBq/(μAh)) is about 2500 times lower than the yield of the same isotope obtained by (α,3n) reaction on enriched ^{116}Cd.

Such a large difference in yield can be explained by the fact that the cross section for the ^{116}Cd(α,3n)^{117m}Sn reaction is significantly higher than the cross sections of ^{123}Sb(γ,p5n)^{117m}Sn and ^{121}Sb(γ,p3n)^{117m}Sn of photonuclear reactions. However, this still does not mean that photonuclear reactions on antimony should be left out of consideration as a possible commercial source of ^{117m}Sn. There is still significant room to increase the yield of these photonuclear reactions. The yield can be significantly increased in photon accelerator beams that have higher electron currents.

In addition to ^{117m}Sn, some amount of ^{119m}Sn is also obtained in photonuclear reactions. Some of its characteristics, such as a large number of conversion electrons can contribute to the therapeutic effect. It was estimated that in the bremsstrahlung photon beam with a maximum energy of 60 MeV, the activity of ^{119m}Sn is obtained, which would less than 27% of the activity of ^{117m}Sn.

CRediT authorship contribution statement

M. Krmar: Writing – original draft, Supervision, Conceptualization. **N. Jovančević:** Investigation. **Ž. Medić:** Software, Formal analysis. **D. Maletić:** Software, Formal analysis. **Yu Teterev:** Project administration, Investigation. **S. Mitrofanov:** Resources, Funding acquisition. **K.D. Timoshenko:** Visualization, Data curation. **S.I. Alexeev:** Resources, Methodology. **H. Marukyan:** Investigation, Formal analysis. **I. Kerobyan:** Writing – original draft, Software, Investigation. **R. Avetisyan:** Resources, Investigation, Formal analysis. **R. Dallakyan:** Investigation. **A. Hakobyan:** Methodology. **L. Vahradyan:** Methodology. **H. Mkrtchyan:** Supervision, Investigation. **A. Petrosyan:** Resources, Data curation. **H. Torosyan:** Resources, Project administration, Resources, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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