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Proton Induced Reactions on Natural Se and Ag Targets for Producing $^{76, 77, 82}$Br and ^{107}Cd Radioisotopes Using Low Energy Cyclotron

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Proton beam of energy 14.7MeV has been used to study the reactions cross-section and the excitation functions for the three radioisotopes of bromine namely, ^{76}Br, ^{77}Br, ^{82}Br produced through the reaction of proton with natural selenium and ^{107}Cd formed through the reaction of proton with natural silver targets. Stacked foil technique was applied to measure the excitation functions. The energy degradation through the foils was calculated using a computer program "STACK". The reactions cross-sections were studied and discussed in the energy range from the reactions threshold energy up to 14.3 MeV. Proton induced activation cross-sections are needed to maximize the yield of the required product and to minimize the yields of the radioactive impurities. These studies are also important for the verification of the nuclear models which explain the reactions mechanism. Two theoretical codes namely, EMPIRE 3.2.2 and ALICE-IPPE as well as the data obtained from TENDLE-2017 library were used to obtain the cross-section values which were compared with the experimental results. Integral yields were also calculated and presented.

Keywords: Nuclear reaction / Selenium / Silver/ Excitation function/ Cyclotron/ Integral yield

Introduction

Isotopes produced by cyclotrons mainly decay through EC or β+ which makes it suitable for medical applications. Hence, there is a need for experimental results leading to the formation of some radioisotopes relevant to these applications. Proton reactions with ^{nat}Se and ^{nat}Ag were studied and the cross-section values at different energies were obtained.

The radioisotopes of bromine are of special interest because of their suitable nuclear and chemical characteristic. Recently, through the developments of Positron emission tomography (PET) and high energy gamma rays cameras, bromine isotopes received some more attention.

Isotopic composition of natural selenium is ^{74}Se (0.89%), ^{76}Se (9.36%), ^{77}Se (7.63%), ^{78}Se (23.78%), ^{80}Se (49.61%) and ^{82}Se (9.2%). The reaction of proton with both natural and enriched selenium isotopes for producing Br radioisotopes were studied previously by several authors [1-12]. The low energy part of the excitation functions for some of Br radioisotopes still needs further investigations. In the present work, the authors studied the possibility of producing $^{76,77and\ 82}$Br through the reaction of proton with natural selenium in the energy range from the reactions threshold and up to 14.3MeV. Natural silver with isotopic composition ^{107}Ag (51.84%) and ^{109}Ag (48.16%) was used as a target to study the possibility of producing ^{107}Cd ($T_{1/2}$ = 6.5 h) which decays totally through EC to ^{107}Ag. Proton reactions on natural and enriched silver targets have been studied previously by different authors [13-19].

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Nuclear Models Calculation

Recent nuclear data evaluation methodology relies on the experimental data as well as the nuclear model calculations. Model calculations are useful in validation of experimental data and resolution of data discrepancy among literatures. They are useful also for thorough description of reaction channels and understanding of the complex physical processes. Complementarily, precise and consistent experimental data are necessary for extraction of optimal parameters improving the results of model calculations. Two Codes have been used in the present work to calculate the reactions cross-section namely, ALICE-IPPE [20-21] and EMPIRE 3.2.2 [22] as well as the obtained data from TENDLE2017 [23-24] which were compared with the experimental results.

Experimental Technique

The excitation functions of the investigated reactions were measured using standard stacked foil technique [25]. The energy degradation through the foils was calculated using a computer program "STACK" based on tabulated data for stopping power of charged particles [26]. Two stacks were prepared**.** The first contains natural selenium of purity (99.5%) prepared by sedimentation technique [2] and the second contains high purity natural Ag foils of purity (99.99%) and thickness 10 μm supplied by good-Fellow. Cu foils of thickness 10μm and purity (99.99%) supplied by Good–Fellow used as a beam current monitor and degrader were arranged in each stack and irradiated at a proton beam energy of 14.7 MeV and a nominal current of 150 nA for 1.5 h.

A high resolution gamma ray spectrometer consists of a Canberra GC6020 high purity germanium detector (HPGe) of efficiency 60% coupled to 2026 Canberra spectroscopy amplifier and TRUMP 8k multichannel analyzer was used in the measurements. The energy resolution of the detector was found to be 2.2 keV at 1332.5 keV gamma ray line of ^{60}Co.

The energy spectra were measured after a cooling time of 30 minutes and followed through one month for Br radioisotopes and for four days for ^{107}Cd.

The gamma ray lines used to calculate the cross-section data for the selected radioisotopes were chosen carefully to avoid any interference from other radioactive isotopes. The uncertainty was estimated by summing in quadrate the uncertainties originated from foil thickness, incident proton current, the calculated detector efficiency and the photo peak area of the gamma ray lines considering that the beam intensity along the stack was not changed. The nuclear data for the studied reactions were taken from NuDat database [27] and presented in Table (1).

Table (1): Decay characteristics of the investigated activation products [27]

Nuclide	Half-life	Decay mode %	Eγ (keV)	Iγ (%)	Contributing reactions	E_{th} (MeV)
^{76}Br	16.2 h	EC (57) β+ (43)	559.00 657.00 1853.71 2391.25 2510.79	74.00 14.70 15.90 4.70 1.95	^{76}Se(p,n) ^{77}Se(p,2n)	5.82 13.34
^{77}Br	57.04 h	EC (100)	297.23 303.76 439.47 817.79	4.16 1.18 1.56 2.08	^{77}Se(p,n) ^{78}Se(p,2n)	2.18 12.81
^{82}Br	35.282 h	β$^-$ (100)	619.00 698.30 776.50 1044.00	43.40 28.50 83.50 27.20	^{82}Br(p,n)	0.80
^{107}Cd	6.5 h	EC (100)	93.1	4.80	^{107}Ag (p,n)	2.22

Results and Discussions

^{nat}Se (p,xn) $^{76(m,g)}$Br reaction

The main route for the formation of ^{76}Br ($T_{1/2}$ =16.2 h) from the reaction of proton with ^{nat}Se in the low energy range (5.82MeV and up to 13.33 MeV) is the (p, n) reaction on ^{76}Se of natural abundance 9.36%. At energies higher than 13.5 MeV, ^{77}Se (p, 2n) starts to contribute in the formation of ^{76}Br. Two states can be populated, ^{76}Br ground state which decays by EC (100%) to ^{76}Se and meta-stable state of ($T_{1/2}$=1.31s) decays totally by isomeric transition to the ground state. Production of ^{76}Br from either natural or enriched Se metal has been previously studied by some authors [1-4]. The γ-ray lines used for calculating the reaction cross-sections are presented in Table (1). The experimental reactions cross-sections obtained in the present study are shown in Table (2).

Fig. (1) shows a comparison between the current results and those obtained previously on natural and enriched targets (after normalization) as well as with the theoretically calculated curves. Although there is a fairly good agreement between the current results and those obtained before by Al-Azony et al. [1] and the normalized data of Kovacs et al. [3], still there is a clear discrepancy between the present results and those obtained previously by Hassan et al. [2] and Levkoveski [4]. The theoretically calculated cross values are higher than the present experimental values except for those measured in the energy range 8.0-9.0 MeV. Theoretical excitation function curves obtained by EMBIRE-3.2.2, ALICE-IPPE codes and TENDLE 2017 are also presented in the Fig. (1). The three curves are showing the same trend, but they are far from explaining the measured excitation functions.

Table (2): Experimental cross-sections for the ^{nat}Se(p,x)$^{76, 77, 82}$Br reactions

E (MeV)	σ(mb) $^{76(m+g)}$Br	σ(mb) $^{77(m+g)}$Br	σ(mb) $^{82(m+g)}$Br
14.1	36.4±7.4	20.4±3.8	3.7±0.8
13.4	41.4±5.7	31.5±5.7	5.5±0.9
12.8	53.7±9.8	25.4±3.9	8.2±1.1
11.9	30.7±5.2	32.8±5.1	19.1±2.3
10.4	27.8±4.6	34.2±5.4	27.3±2.1
9.0	66.7±10.8	31.8±4.8	41.3±6.2
8.6	70.2±9.7	34.6±4.4	60.2±6.9
7.03	60.0±6.8	26.3±5.2	37.9±6.5
6.8	19.1±4.1	33.7±6.1	25.4±4.6
4.7	--------	4.9±2.1	7.7±1.2
3.9	--------	6.3±1.9	3.4±0.7

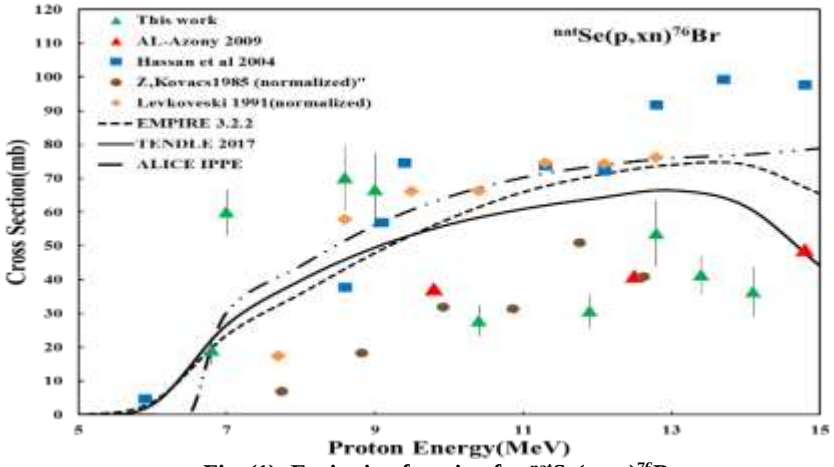

Fig. (1): Excitation function for ^{nat}Se(p,xn)^{76}Br

^{nat} Se (p,xn) ^{77}Br reaction

Two routes are contributing in the formation of ^{77}Br ($T_{1/2}$=57h) radioisotope, these are ^{77}Se (p,n) and ^{78}Se (p,2n) with threshold energies 2.18 and 12.81 MeV, respectively. Two states can be populated, the ground state with spin and parity 3/2$^-$ and the meta stable state ($T_{1/2}$=4.2m) with spin and parity 9/2$^+$. ^{77m}Br decays totally by IT (100%) to the ground state while the ground state decays through EC (100%) to ^{77}Se. The decay data are presented in Table (1). The production of ^{77}Br from enriched ^{77}Se target was investigated previously by Levkoveski [4], Janssen et al. [5] and Johnson et al. [6, 7]. The present results as well as those previously measured on natural targets by Al-Azony et al. [1] and Hassan et al. [2] as well as those obtained after normalization for the natural abundance of the ^{77}Se isotope along with the theoretical calculations are presented in Fig.(2). Considering the experimental results, it is clear from the Figure that the cross-section values obtained by Janssen et al. [5] are very far from any of the shown excitation functions. Although our measured excitation function shows nearly the same behavior as those calculated using the three codes, the cross-section values are lower than those calculated theoretically as well as some of the previously measured data.

^{nat} Se(p,x)$^{82(m+g)}$Br reaction

Proton reaction with ^{nat}Se leads to the formation of ^{82}Br radioisotope in its ground and meta-stable states with spins and parities 5$^-$ and 2$^-$ and half-lives ($T_{1/2}$=35.28h) and ($T_{1/2}$= 6.13m), respectively. The meta-stable state of ^{82}Br decays by (97.60%) IT to the ground state and (2.40 %) β^- to ^{82}Kr, while the ground state decays through β^- (100 %) to ^{82}Kr. The gamma ray lines used for calculating the cross-section values are shown in Table (1) while the measured cross-section values for the ^{nat}Se(p,x)$^{82(m+,g)}$Br reaction are presented in Table (2). Fig. (3) shows the measured excitation function as compared with those obtained previously as well as with the theoretically calculated curves.

Measured and calculated excitation functions show a pronounced peak in the energy range 5.0-13.0MeV. At energies lower than 9.0 MeV, an acceptable agreement between the experimental results is observed. On the other hand, there is a discrepancy between the measured excitation functions at energies higher than this value. The present results agree well with those obtained by AL-Azony et al. [1] while Levkoviski [4], Hassan et al. [2] and Gyurky et al. [8] show a higher cross-section values at energies higher than 9.0 MeV. The maximum cross-section value of the theoretically calculated excitation function obtained by ALICE-IPPE is shifted by about1.0 MeV towards a higher energy range as compared with those calculated using EMBIRE 3.2.2 as well as the values obtained from TENDLE 2017. It should be mentioned that there is a good agreement between the experimental results and the calculated cross-section values in the low energy range of the curve.

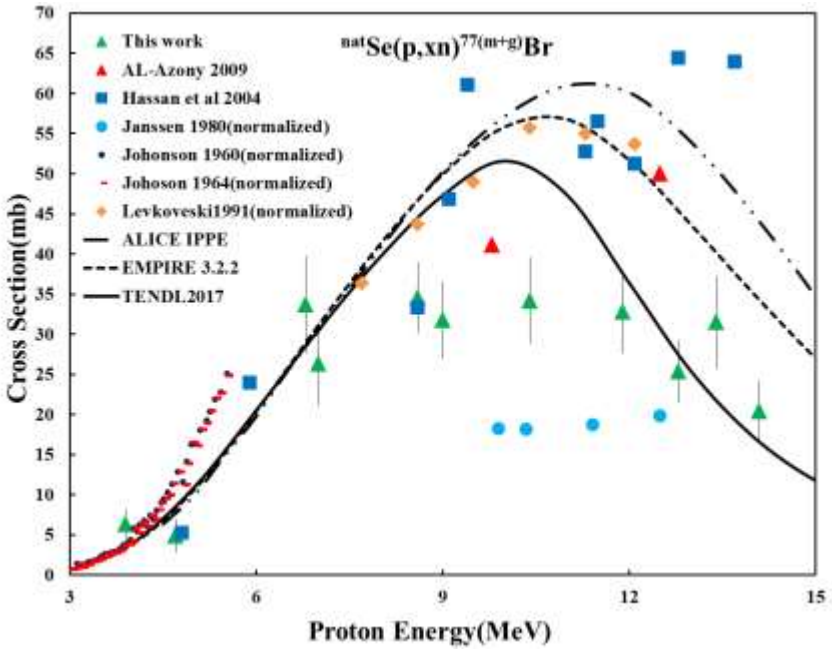

Fig. (2): Excitation function for ^{nat}Se(p,xn)^{77m,g}Br

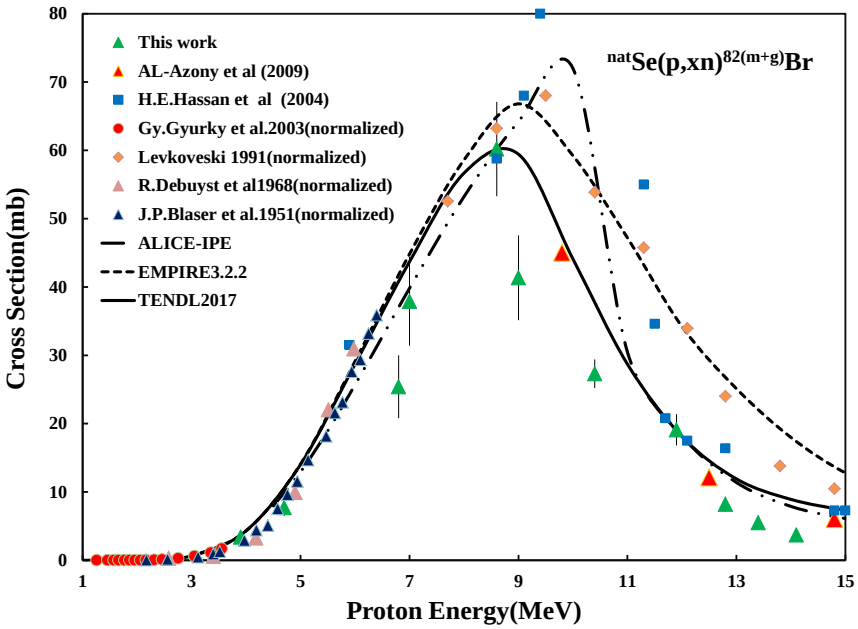

Fig. (3): Measured excitation function for the reaction $^{nat}Se(p,xn)^{82m+g}Br$

$^{nat}Ag(p,n)^{107}Cd$ reaction

High purity ^{nat}Ag of thickness (10µm) along with ^{nat} Cu foils of thickness (10 µm) were arranged in a stack and irradiated by a proton beam of energy 14.3 MeV to study the possibility of producing the ^{107}Cd radioisotope ($T_{1/2}$=6.5h). Natural Ag is composed of two stable isotopes namely, ^{107}Ag (abun.=51.839%)and ^{109}Ag(abun.=48.161%). The only route for the formation of ^{107}Cd in the present energy range is through the reaction ^{107}Ag (p,n), since the threshold energy for the reaction ^{109}Ag(p,3n) is high (18.82 MeV). $^{107}Ag(p,xn)^{107}Cd$ was studied previously using natural and enriched ^{107}Ag target by Hershberger et al. [14], Colle et al. [15], Wing and Huizenga [16], and Blaser et al. [17] in the low energy region. On the other hand. Uddin et al. [18] measured the excitation function of proton induced reactions on natural silver target for both the short and long–lived isotopes. Khandaker et al. [13] measured the production cross-section of residual radionuclides from proton–induced reaction on ^{nat}Ag up to 40 MeV. Recently, Tarkanyi et al. [19] studied the production routes of $^{107,109}Cd$ radioisotopes via charged particle induced nuclear reactions in the energy range from 11.6 MeV to 78.4 MeV.

Fig.(4) shows the measured excitation function as compared with the previously reported data as well as with the theoretically calculated curves where a pronounced peak is observed in the energy range between 5.0-13.8 MeV. Generally, there is a satisfactory agreement between the present results and those obtained previously, only the results of Colle et al. [15] show higher cross-section values at energies higher than 10 MeV with a shift of the maximum to the higher energy range. Table (3) shows the experimentally obtained cross-section values as a function of proton energy.

Integral Yields

The integral yield for the studied reactions were calculated for a beam current 1µA and irradiation time 1h using the measured excitation function curves and presented in Figs. (5 & 6). Integral yields of 20.81, 5.94 and 11.42 MBq/µA.h were obtained for the three isotopes ^{76}Br, ^{77}Br and ^{82}Br, respectively in the energy ranges 14.3 → 6, 14.3 →3.4 and 14.3 → 3.0 MeV. From Fig. (5) it is clear that there is no possibility to produce any of the three isotopes with a high radionuclidic purity from natural Se targets since all of them can be formed nearly in the same energy range, and their half-lives are comparable with each other. From the measured excitation function for the reaction ^{nat}Ag (p, n)^{107}Cd, the integral yield was calculated and an amount of 212.34 Mbq/µA.h can be obtained in the energy range E_p= 14.3 → 2.5 MeV. Fig. (6) shows the integral yield of ^{107}Cd plotted as a function of incident proton energy. The formation of other radioisotopes of Cd could be neglected because of the very long half-life of ^{109}Cd ($T_{1/2}$ = 468 d) as compared with that of ^{107}Cd half-life ($T_{1/2}$=6.5h) which means a longer irradiation time is required to have a measurable quantity of ^{109}Cd.

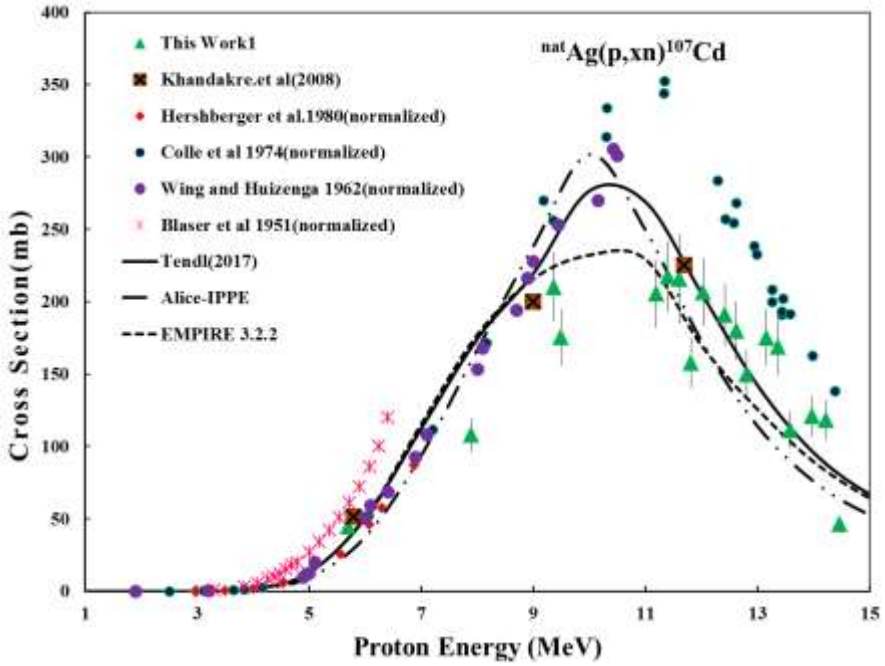

Fig. (4): Measured excitation function for the reaction $^{nat}Ag(p,xn)^{107}Cd$ as compared with the previously reported data and theoretical calculations

Table (3): Experimental cross-sections for the $^{107}Ag(p,n)^{107}Cd$ reaction

E (MeV)	cross section (mb)
14.47	46.3±5.8
14.22	118.3±13.7
13.97	120. ±13.8
13.59	111.5±12.7
13.37	168.8±19.1
13.16	175.2±19.8
12.81	149.8±16.9
12.62	180.1±20.3
12.42	191.1±21.5
12.03	206.6±23.2
11.81	157.6±17.7
11.61	215.9±31.1
11.40	217.7±24.5
11.19	205.5±23.1
9.37	210.2±23.6
9.49	175.5±19.7
7.89	108.1±12.1
5.69	44.7±5.0

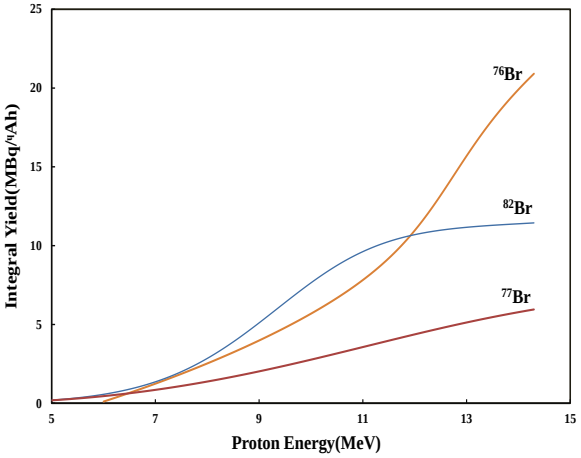

Fig. (5): Calculated integral yields of $^{76, 77\&82}$Br plotted as a function of incident proton energy

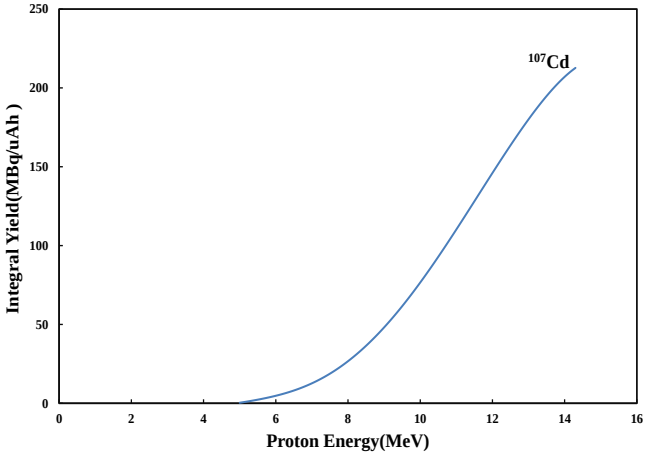

Fig. (6): Calculated integral yields of ^{107}Cd plotted as a function of incident proton energy

Conclusion

The present work included an investigation of the possibility of producing ^{76}Br, ^{77}Br and ^{82}Br in a suitable quantity and purity in the low energy range, as well as obtaining some of the nuclear data related to the reaction of proton with natural Se targets which were prepared by sedimentation technique. The excitation functions in the proton energy range from the reactions threshold and up to 14.3 MeV were measured using the STACK foil technique. A comparison between the present results and those obtained previously show that there is a discrepancy between all of the available data. The excitation functions were compared also with the theoretically calculated curves using the codes ALICE-IPPE, EMBIRE-3.2.2 and the data obtained from TENDLE 2017. The present results reveal that the production of the three isotopes ^{76}Br, ^{77}Br and ^{82}Br can be formed with an amount of 20.81, 5.94 and 11.42 MBq/ µAh in the energy ranges 14.3→6.0 MeV , 14.3→ 3.4 MeV and 14.3→3.0 MeV, respectively.

Regarding ^{107}Cd which was studied using high purity silver foils an amount of 212.34 MBq/ µAh, it can be produced in the energy range 14.3→ 2.5MeV. Because of the high threshold energy for some of the reactions leading to the formation of other Cd radioisotopes or because of the short half-lives of the others, ^{107}Cd can be produced with an acceptable impurity level using highly pure silver targets.

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