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Citation	Nuclear instruments & methods in physics research section b-beam interactions with materials and atoms, 426, 18-21 https://doi.org/10.1016/j.nimb.2018.04.023
Issue Date	2018-07-01
Doc URL	http://hdl.handle.net/2115/78760
Type	article (author version)
File Information	manuscript-In+a-final.pdf

Activation cross sections of alpha-induced reactions on ^{nat}In for ^{117m}Sn production

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Abstract

The production of ^{117m}Sn by charged-particle induced reactions is an interesting topic for medical application. Production cross sections of α-induced reactions on ^{nat}In for ^{117m}Sn up to 50 MeV were measured using the stacked foil technique and activation method. The integral yield of ^{117m}Sn was estimated using the measured cross sections. The results were compared with experimental data investigated earlier and theoretical calculation. Measured cross sections for ^{113}Sn and ^{116m,117,118m}Sb isotopes were also presented.

1. Introduction

The radioisotope (RI) ^{117m}Sn ($T_{1/2}$ = 13.76 d) decays with emissions of conversion electrons (126.82, 129.369 and 151.56 keV) and γ rays (158.56 keV). It is available as a theranostic RI because the electrons and γ rays are appropriate for therapy and imaging, respectively [1,2]. The production reactions of ^{117m}Sn for clinical applications have recently been investigated and discussed in [3,4]. Batch production of appropriate quantities for routine use in hospitals is still of much concern. We focused on one of the reactions, the ^{115}In(α,x)^{117m}Sn reaction. Three reports on the cross sections of the reaction [5,6,7] were found in the EXFOR library [8]. The data show large discrepancy from each other and more reliable and accurate data are required. Therefore, we performed a new experiment to measure the cross sections of the reaction. From the measured cross sections, the integral yield of ^{117m}Sn was estimated. In addition, the cross sections for other Sn and Sb isotopes were also presented.

2. Method

The experiment was performed at the AVF cyclotron of the RIKEN RI Beam Factory by using the stacked foil technique, activation method and high resolution γ-ray spectrometry. Natural In foils (purity: 99.99%, Nilaco Corp., Japan) of which isotopic composition is listed in Table 1 were stacked with ^{nat}Ti monitor foils (purity: 99.6%, Nilaco Corp., Japan) as a target. The thicknesses of the In and Ti foils were estimated from the measured area and weight of large foils (50 × 50 mm^2) and found to be 16.60 mg/cm^2 and 2.44 mg/cm^2, respectively. The stacked target consisted of 11 sets of the In-In-Ti-Ti foils (8 × 8 mm^2) cut off from the large foils. The foils at the downstream side of the beam would be measured to compensate for losses of recoil products. However, the 2 adjoining In foils were partially molten during the irradiation (low melting point at 156.6°C) and could not be separated when dismantling the target. Each pair of In foils was therefore considered as one foil of 33.20 mg/cm^2. The target was irradiated by a 51.6 MeV α beam with an average intensity of 202.1 nA for 2 hours. The intensity and beam energy were measured by a Faraday cup like target holder and the time-of-flight method using a plastic scintillator monitor [9]. The beam energy degradation in the stacked target was calculated using the SRIM code [10]. After adequate cooling time, the irradiated foils were subjected to γ-spectrometry with a HPGe detector. Decay data of reaction products were obtained from online databases, e.g. NuDat 2.7 [11], Lund/LBNL Nuclear Data Search [12], and Q-value Calculator (QCalc) [13] (Table 2).

Table 1: Isotopic composition of natural indium

^{113}In	4.29%
^{115}In	95.71%

Table 2: Reactions and decay data of reaction products

Nuclide	Half-life	Decay mode (%)	E$_\gamma$ (keV)	I$_\gamma$ (%)	Contributing reactions	Q-value (MeV)
^{117m}Sn	13.76 d	IT (100)	156.02	2.113(12)	^{115}In(α,d)	-9.85
			158.56	86.4	^{115}In(α,2p)^{117}In(β$^-$)	-12.75
					^{115}In(α,2n)^{117}Sb(ε)	-14.61
^{113}Sn	115.09 d	ε (100)	391.698	64.97(17)	^{115}In(α,t3n)	-37.95
					^{113}In(α,tn)	-21.64
^{118m}Sb	5.00 h	ε+β$^+$ (100)	253.678	99(6)	^{115}In(α,n)	-7.19
			1050.69	97(5)		
			1229.65	100(5)		
^{117}Sb	2.80 h	ε+β$^+$ (100)	158.562	85.9	^{115}In(α,2n)	-14.61

^{116m}Sb	60.3 m	$\varepsilon+\beta^+$ (100)	542.867	48.1(4)	^{115}In(α,3n)	-24.50
			972.564	74.2(7)	^{113}In(α,n)	-8.19

3. Result

The excitation function of the ^{nat}Ti(α,x)^{51}Cr monitor reaction was used to assess beam parameters, e.g. intensity and energy. The excitation function could be derived from measurements of the γ-line at 320.08 keV (9.91%) from the decay of ^{51}Cr ($T_{1/2}$ = 27.7025 d). The measurements of the Ti foils were performed after a cooling time of 34 hours for background reduction. During the measurements, the dead time was kept below 3.5%. The statistical uncertainty at maximum was about 6% at the lowest cross section of 0.7 mb. The result was compared with the recommended values [14] as shown in Fig. 1. The result is in good agreement with the recommended values and suggests no adjustment required for the beam parameters.

The cross sections of ^{117m}Sn and ^{117,118m}Sb obtained for ^{nat}In were normalized to for 100%-^{115}In target because no or negligible contribution from reactions on ^{113}In are expected. The numerical data were presented in Table 3 and shown in Figs. 2-6 in comparison with the experimental data studied earlier [5,6,7,15,16,17] and the TENDL-2017 data [18].

The total uncertainty was estimated to be less than 25.4% including statistical errors (<24.1%). It was estimated as the square root of the quadratic summation of the components; the beam intensity (5%), target thickness (1%), target purity (1%), detector efficiency (5%), γ intensity (<6%) and peak area determination (3%).

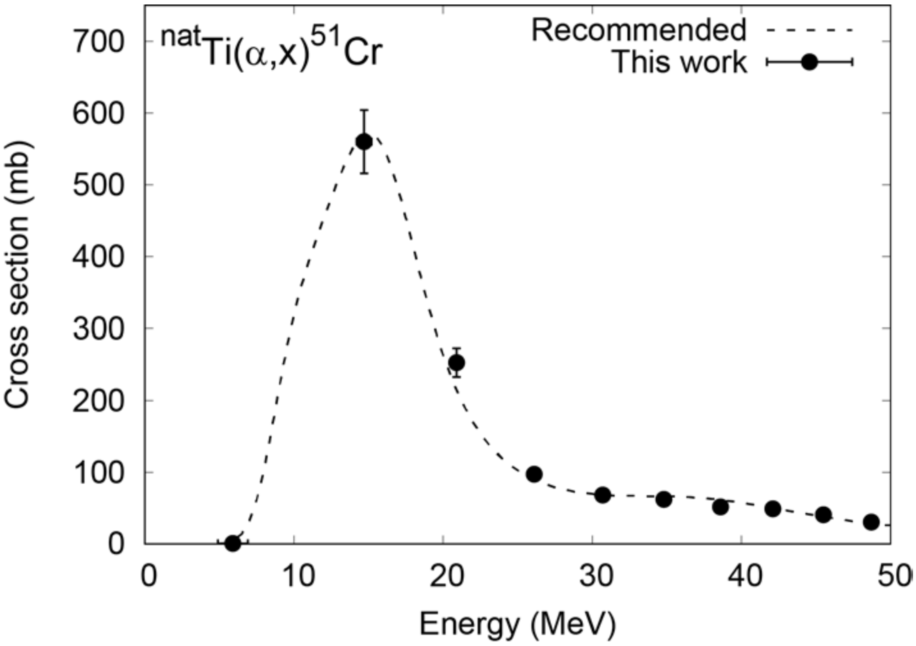

Fig. 1. Excitation function of the monitor reaction ^{nat}Ti(α,x)^{51}Cr.

Table 3. Measured cross sections for Sn and Sb isotope production.

Energy (MeV)	^{115}In(α,x)^{117m}Sn (mb)	^{nat}In(α,x)^{113}Sn (mb)	^{115}In(α,n)^{118m}Sb (mb)	^{115}In(α,2n)^{117}Sb (mb)	^{nat}In(α,x)^{116m}Sb (mb)
50.3 ±1.4	32.1 ±2.5	26.0 ±2.0	5.2 ±0.5	76.5 ±6.6	393 ±31
47.2 ±1.4	35.9 ±2.8	13.7 ±1.1	5.8 ±0.6	95.4 ±8.2	614 ±49
43.9 ±1.5	40.6 ±3.2	4.4 ±0.4	7.1 ±0.7	125 ±11	827 ±65
40.5 ±1.6	48.0 ±3.8	0.65 ±0.08	8.7 ±0.9	196 ±17	949 ±75
36.8 ±1.7	54.3 ±4.3		11.8 ±1.2	386 ±33	782 ±62
32.9 ±1.9	34.4 ±2.7		17.1 ±1.7	861 ±74	386 ±31
28.6 ±2.0	21.0 ±1.7		29.9 ±3.0	1258 ±108	33.7 ±5.0
23.8 ±2.3	5.3 ±0.4		106 ±11	866 ±74	
18.2 ±2.8	0.20 ±0.05		182 ±18	133 ±11	

3.1 ^{117m}Sn production

The measurement of the 156.02 keV γ-line (2.113%) from the ^{117m}Sn decay ($T_{1/2}$ = 13.76 d) was performed after a cooling time of about 45 hours. The cooling time was set to be long enough for the decay of the parent nuclei, ^{117g}In ($T_{1/2}$ = 43.2 min.), ^{117m}In ($T_{1/2}$ = 116.2 min.) and ^{117}Sb ($T_{1/2}$ = 2.80 h). The γ-line at 158.56 keV (86.4%) was not used to avoid the contribution of the 159.377 keV γ-line (68.3%) from ^{47}Sc ($T_{1/2}$ = 3.3492 d) produced in the Ti catcher foils, which were measured together. Cumulative cross sections of the ^{115}In(α,x)^{117m}Sn reaction taking into account the ^{115}In abundance were shown in Fig. 2 together with the previous experimental data [5,6,7] and the TENDL-2017 data [18]. The present result is almost coincident with the data by Qaim and Döhler (1984) [6] among the three earlier experimental data. The data by Fukushima et al. (1963) [5] and TENDL-2017 show much smaller amplitude than ours and the others, while the data by Bhardwaj et al. (1992) are larger than others.

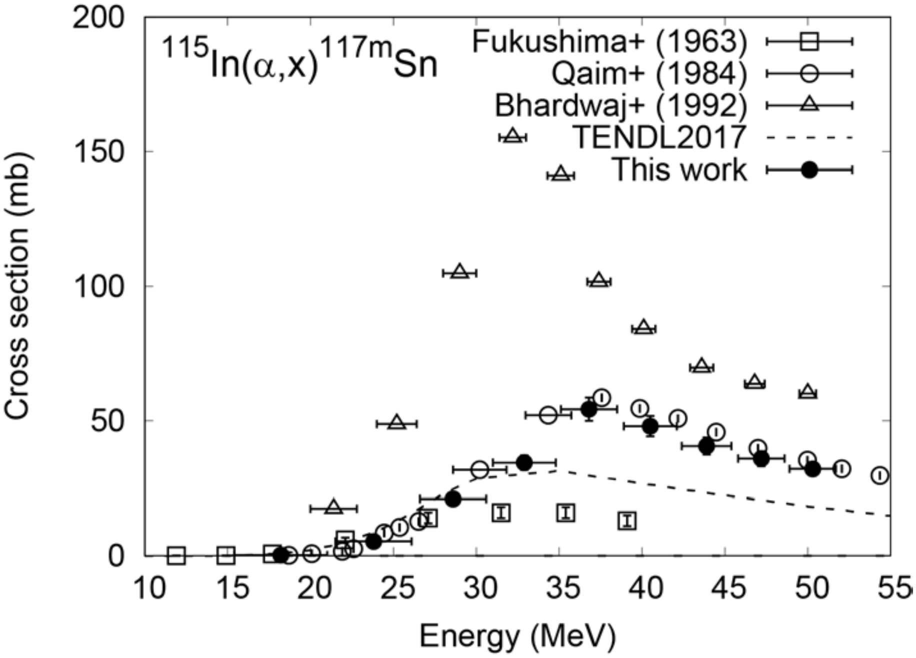

Fig. 2. Excitation function of the ^{115}In(α,x)^{117m}Sn reaction with experimental data measured earlier [5,6,7] and the TENDL-2017 data.

3.2 ^{113}Sn production

The excitation function of the ^{nat}In(α,x)^{113}Sn reaction were derived from the measurement of the γ-line at 391.698 keV (64.97%). Both ^{113}In and ^{115}In can contribute to the cross sections according to the Q-values (Table 2). The measurement of the long-lived ^{113}Sn ($T_{1/2}$ = 115.09 d) was performed after a cooling time of about 45 hours. It is long enough to obtain cumulative ^{113}Sn production from the decay of ^{113}Sb ($T_{1/2}$ = 6.67 min.) and ^{113m}Sn ($T_{1/2}$ = 21.4 min.) and also to reduce disturbing signals from the shorter-lived activation products. The result is shown in Fig. 3 together with the experimental data earlier [6] and the TENDL-2017 data [18]. Our result is in good agreement with the data by Qaim and Döhler (1984) [6] as was the case of ^{117m}Sn. The TENDL-2017 data overestimate the experimental data in the energy region as shown in the figure.

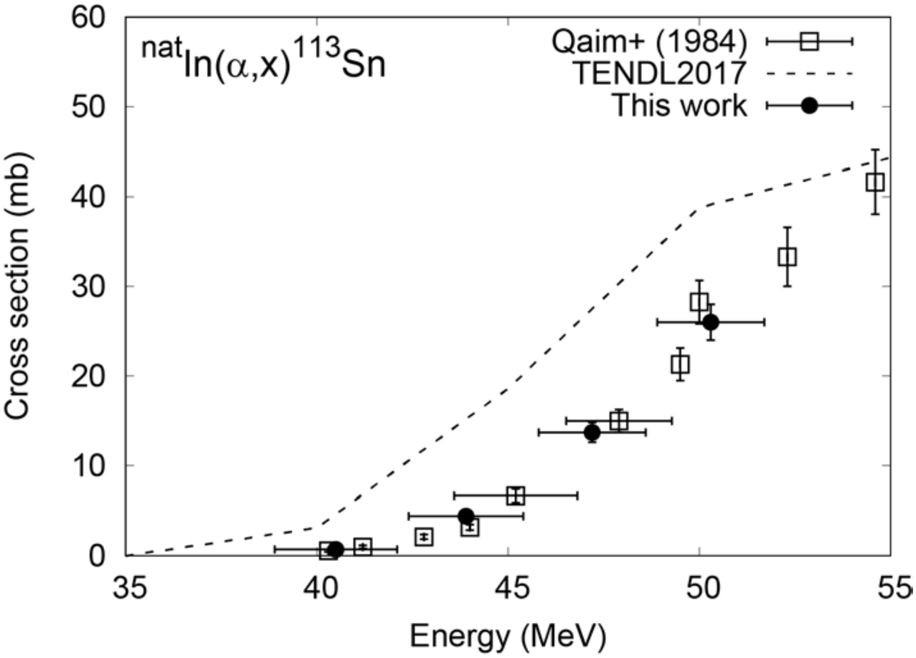

Fig. 6: Excitation function of the ^{nat}In(α,x)^{113}Sn reaction with the experimental data measured earlier [6] and the TENDL-2017 data.

3.3 ^{118m}Sb production

The cross sections of ^{118m}Sb were derived from the measurement of the 253.5 keV γ-line (99%) from the ^{118m}Sb decay ($T_{1/2}$ = 5.00 h). The measurement was performed after a cooling time of about 6 hours. The cross sections of the ^{115}In(α,n)^{118m}Sb reaction could be derived by considering the isotopic ratio of ^{115}In due to no contribution from ^{113}In. The result is shown in Fig. 4 in comparison with the experimental data studied earlier [7,15] and the TENDL-2017 data [18]. The peak position is consistent with the experimental data [15] and the TENDL-2017 data. However, the peak amplitude of our result is lower than the two experimental data and higher than the TENDL-2017 data. One of the reasons for our lower amplitude than the other experimental data may be the thicker In foils used in this study.

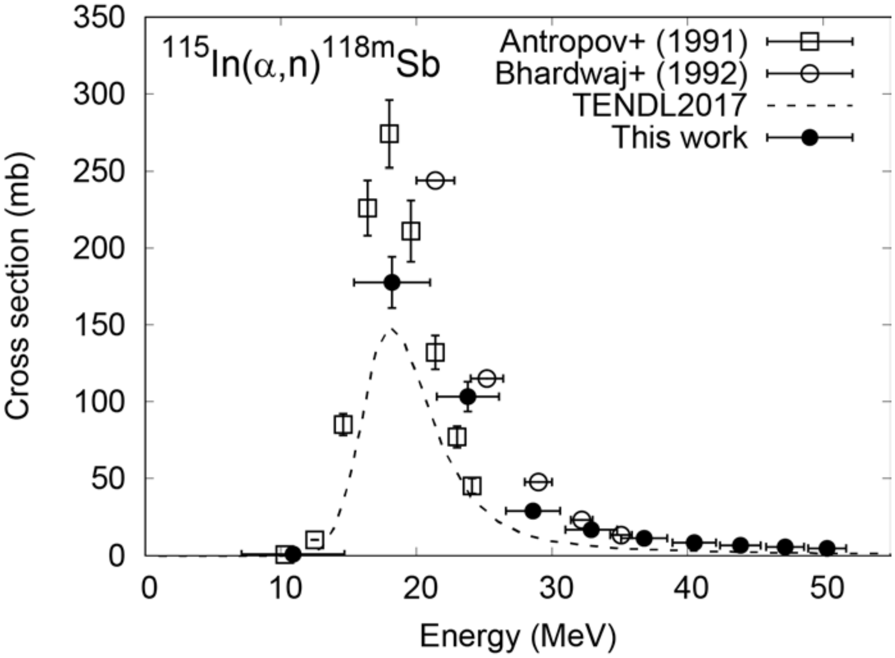

Fig. 4. Excitation function of the ^{115}In(α,n)^{118m}Sb reaction with the experimental data earlier [7,15] and the TENDL-2017 data.

3.4 ^{117}Sb production

The measurements in 6-8 hours after the end of bombardment were used to derive the cross sections of ^{117}Sb ($T_{1/2}$ = 2.80 h). The γ-line at 158.562 keV (86%) from the decay of ^{117}Sb was contributed from the decay of ^{117g}In ($T_{1/2}$ = 43.2 min.), ^{117m}In ($T_{1/2}$ = 116.2 min.) and ^{117m}Sn ($T_{1/2}$ = 13.76 d). Each contribution to the γ-line was estimated independently from other γ-lines. The contributions of ^{117g}In and ^{117m}In were estimated using the more intense γ-lines at 552.9 keV (100%) from ^{117g}In and 315.302 keV (19%) from ^{117m}In. There were no peaks at the energies and therefore the ^{117g,117m}In contributions to the γ-line at 158.562 keV were found to be negligibly small. The contribution from ^{117m}Sn was estimated from the cumulative cross sections of ^{117m}Sn derived in Section 3.1 and found to be small (<2.2%). The net counts of the γ-line at 158.562 keV from ^{117}Sb were obtained by subtracting the contribution of ^{117m}Sn. The derived cross sections of the ^{nat}In(α,x)^{117}Sb reaction were normalized for the monoisotopic ^{115}In because the ^{113}In(α,γ)^{117}Sb reaction less contributes the production of ^{117}Sb.

The result is compared with the experimental data [7,15] found in the EXFOR library and the TENDL-2017 data [18] (Fig. 5). The peak of our result shows a small shift to the higher energy region than the data earlier [7], though its amplitude is nearly consistent. The TENDL-2017 data underestimate the experimental data above 25 MeV.

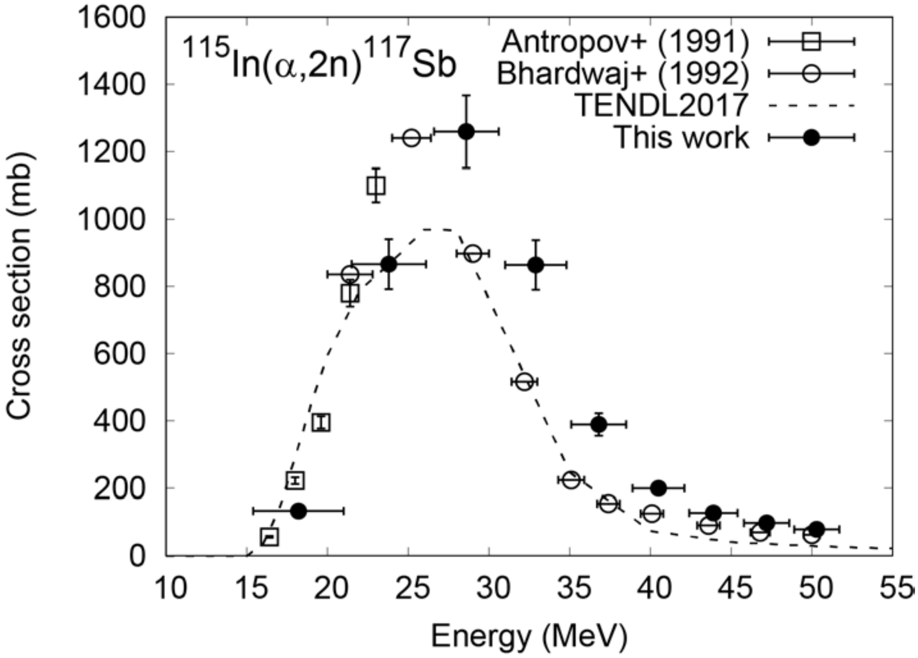

Fig. 5. Excitation function of the ^{115}In(α,2n)^{117}Sb reaction with the experimental data measured earlier [7,15] and the TENDL-2017 data.

3.5 ^{116m}Sb production

The production cross sections of ^{116m}Sb ($T_{1/2}$ = 60.3 min) were derived from the γ-line at 972.564 keV (74.2%) using the same measurement series with the ^{117,118m}Sb production. The result is shown in Fig. 6 together with the experimental data of the ^{113}In(α,n)^{116m}Sb reaction [16,17]. The earlier experimental data were up to 25 MeV below the threshold energy of the ^{115}In(α,x)^{116m}Sb reaction and contributed only from ^{113}In. The cross sections of the ^{113}In(α,n)^{116m}Sb reaction were normalized to the natural indium by considering the isotopic ratio of ^{113}In. The cross sections above 25 MeV were measured for the first time in this work. Our and earlier data were separated in energy, however smoothly connected as the TENDL-2017 data. The TENDL-2017 data reproduce well our data especially below 40 MeV.

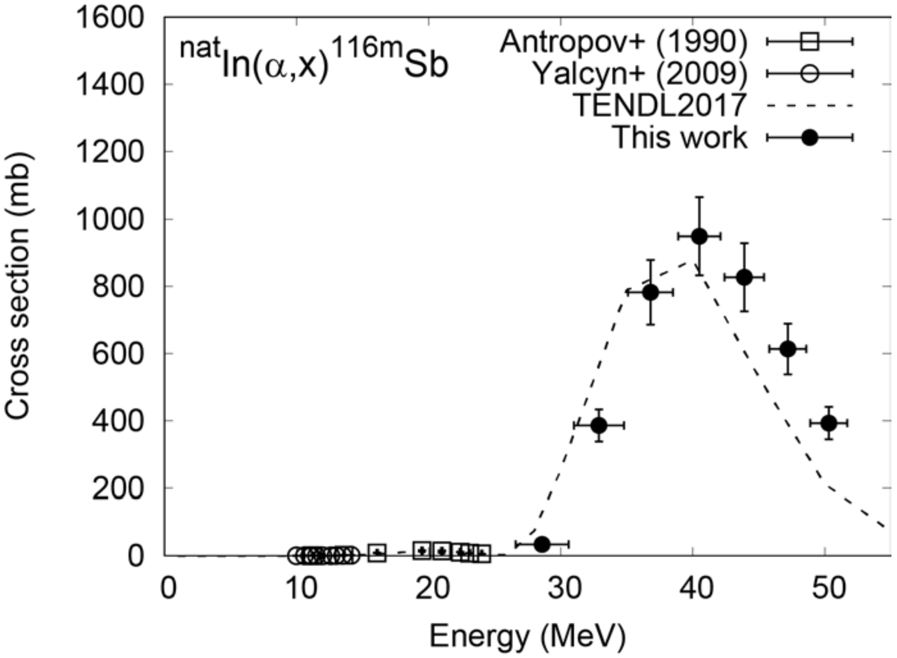

Fig. 6. Excitation function of the ^{nat}In(α,x)^{116m}Sb reaction with the experimental data measured earlier [16,17]. and the TENDL-2017 data.

3.6 Thick target yield of ^{117m}Sn

The integral yield of ^{117m}Sn was estimated using stopping powers calculated by the SRIM code [10] and the excitation function of the ^{115}In(α,x)^{117m}Sn reaction interpolated by the spline fit. The derived integral yield was compared with the experimental [5,19] and calculated data [4] (Fig. 7). The calculated data [4] were based on cross sections derived from EMPIRE and TALYS codes, which were larger than experimental data by Qaim and Döhler (1984) [6]. Our result is smaller than the other data at the whole energy region because the cross sections were consistent with the data by Qaim and Döhler (1984) [6] and lower than the calculated data [4].

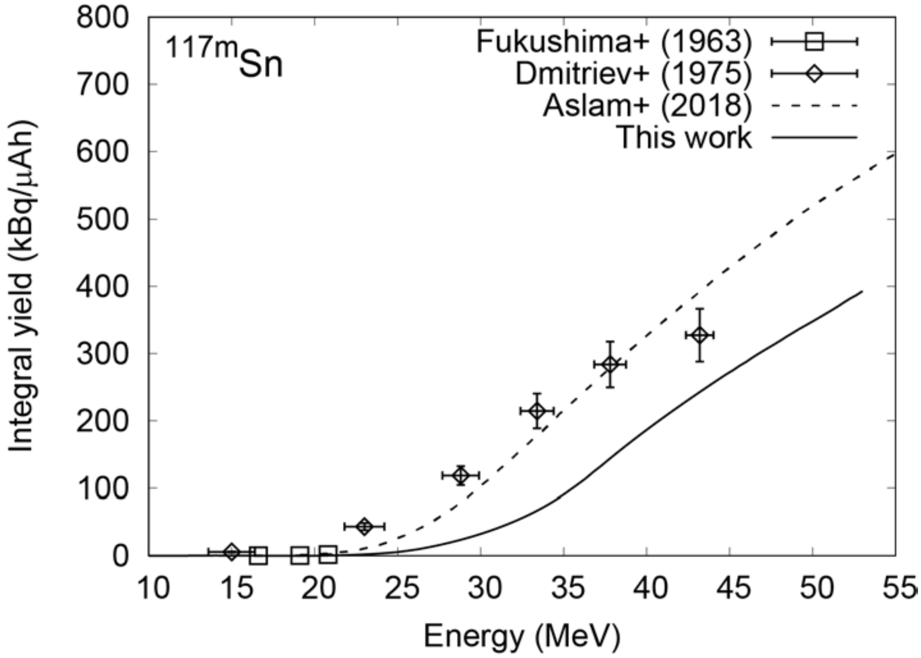

Fig. 7. Integral yield of ^{117m}Sn with the experimental data measured earlier [5,19] and the calculated data [4].

4. Conclusion

Charged-particle reactions to produce a medical radioisotope ^{117m}Sn are worthy investigated. One of the reactions to produce ^{117m}Sn is the α-induced reactions on ^{115}In. We performed an experiment to measure the cross sections of the reaction up to 50 MeV at the AVF cyclotron of the RIKEN RI Beam Factory. The present result is almost consistent with the data by Qaim and Döhler (1984) [6] among the three experimental data earlier. From the measured cross sections, the integral yield of ^{117m}Sn was estimated. The derived yield is smaller than the other data at the whole energy region.

Acknowledgement

This work was carried out at RI Beam Factory operated by RIKEN Nishina Center and CNS, University of Tokyo, Japan. This work was supported by JSPS KAKENHI Grant Number 17K07004.

Reference

[1] S.C. Srivastava, G.E. Meinken, P. Richards, P. Som, Z.H. Oster, H.L. Atkins, A.B. Brill, F.F. Knapp Jr, and T.A. Butler, "The Development and In-Vivo Behavior of Tin Containing Radiopharmaceuticals: I. Chemistry, Preparation, and Biodistribution in Small Animals," *Int. J. Nucl. Med.*, 12, 167, 1985.

[2] Suresh Srivastava, H. William Strauss, Jagat Narula, Renu Virmani, Frank Kolodgie, David Mueller, Nigel Stevenson, and Gilbert Gonzales, "Theragnostic potential of Sn-117m for the molecular targeting and therapy of vulnerable plaque," *J. Nucl. Med.*, 54, Suppl. 2, 461, 2013.

[3] Syed M. Qaim, "Nuclear data for medical radionuclides," *J. Radioanal. Nucl. Chem.*, 305, 233, 2015.

[4] M.N. Aslam, K. Zubia, and S.M. Qaim, "Nuclear model analysis of excitation functions of a-particle induced reactions on In and Cd up to 60 MeV with relevance to the production of high specific activity 117mSn," *Appl. Radiat. Isot.*, 132, 181, 2018.

[5] S. Fukushima, S. Hayashi, S. Kume, H. Okamura, K. Otozai, K. Sakamoto, and R. Tsujino, "The Production of High Specific Activities of Tin," *Bull. Chem. Soc. Jpn.*, 36, 1225, 1963.

[6] S.M. Qaim and H. Döhler, "Production of Carrier-free 117mSn," *Int. J. Appl. Radiat.*

Isot., 645, July 1984.

[7] M.K. Bhardwaj, I.A. Rizvi, and A.K. Chaubey, "EXCITATION FUNCTION STUDIES FOR THE ALPHA INDUCED REACTIONS IN INDIUM," *Int. J. Mod. Phys. E*, 1, 389, 1992.

[8] N. Otuka, E. Dupont, V. Semkova, B. Pritychenko, A.I. Blokhin, M. Aikawa, S. Babykina, M. Bossant, G. Chen, S. Dunaeva, R.A. Forrest, T. Fukahori, N. Furutachi, S. Ganesan, Z. Ge, O.O. Gritzay, M. Herman, S. Hlavač, K. Katō, and Lalremruata, "Towards a More Complete and Accurate Experimental Nuclear Reaction Data Library (EXFOR): International Collaboration Between Nuclear Reaction Data Centres (NRDC)," *Nucl. Data Sheets*, 272, 120, 2012.

[9] T. Watanabe, M. Fujimaki, N. Fukunishi, H. Imao, O. Kamigaito, M. Kase, M. Komiyama, N. Sakamoto, K. Suda, M. Wakasugi, and K. Yamada, "Beam Energy and Longitudinal Beam Profile Measurement System at RIBF," in *Proc. 5th Int. Part. Accel. Conf. (IPAC2014)*, 2014, 3566.

[10] J. Ziegler, J.P. Biersack, and M. Ziegler. SRIM: the Stopping and Range of Ions in. [Online]. http://www.srim.org

[11] National Nuclear Data Center, Brookhaven National Laboratory, *Nuclear structure and decay data on-line library, Nudat 2.7.* [Online]. http://www.nndc.bnl.gov/nudat2/

[12] S.Y.F. Chu, L.P. Ekström, and R.B. Firestone. The Lund/LBNL Nuclear Data Search. [Online]. http://nucleardata.nuclear.lu.se/toi/

[13] National Nuclear Data Center, Brookhaven National Laboratory. Q-value Calculator (QCalc). [Online]. http://www.nndc.bnl.gov/qcalc/

[14] IAEA-TECDOC-1211, *Charged particle cross section database for medical radioisotope production.*, 2001. [Online]. https://www-nds.iaea.org/medical/monitor_reactions.html

[15] A.E. Antropov, V.G. Batij, V.P. Gusev, A.A. Kolozhvari, E.A. Skakun, and A.V. Smirnov, "Excitation Functions of (a,a'), (a,n) and (a,2n) Reactions on 115In," in *41st Ann. Conf. Nucl. Spectrosc. Struct. At. Nuclei*, Minsk, 1991, 330.

[16] C. Yalçın, R.T. Güray, N. Özkan, S. Kutlu, Gy. Gyürky, J. Farkas, G. G. Kiss, Zs. Fülöp, A. Simon, E. Somorjai, and T. Rauscher, "Odd p isotope 113In: Measurement of a-induced reactions," *Phys. Rev. C*, 79, 065801, 2009.

[17] A.E. Antropov, V.G. Batij, V.P. Gusev, A.A. Kolozhvari, E.A. Skakun, and A.V. Smirnov, "Excitation Functions of (a,n) and (a,2n) Reactions on 113In," in *Ann.*

Conf. Nucl. Spectrosc. Struct. At. Nuclei, Leningrad, 1990, 343.

[18] A.J. Koning and D. Rochman, "Modern nuclear data evaluation with the TALYS code system," *Nucl. Data Sheets*, 113, 2841, 2012. [Online]. https://tendl.web.psi.ch/tendl_2017/tendl2017.html

[19] P.P. Dmitriev, M.V. Panarin, G.A. Molin, and Z.P. Dmitrieva, "Yields of 113Sn and 117mSn in nuclear reactions with protons, deuterons, and alpha particles," *Soviet Atomic Energy*, 39, 734, 1975.