Cross-section measurements for the theranostic radionuclide ^{67}Cu using 69 MeV proton beam

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1. Introduction

^{67}Cu is one of the most promising candidates among the theranostic isotopes of major interest, owing to the specific role of copper in several biochemical processes [1,2]. ^{67}Cu of peculiar physical-chemical characteristics has been considered an excellent nuclide for radioimmuno-therapy for a long time. Relatively long half-life of this nuclide (61.83 h) allows the slow biodistribution of antibodies, additionally its β-emission being of a therapeutic effect of short-medium range on the targeted cells [1,2].

Although, in early days, ^{67}Cu is limited in producing from nuclear reactors, it shifts over particle accelerators over the last decade because of the higher quality of the final product. It meets the specifications for its use in targeted therapy [3]. The accurate knowledge of the cross section is the first important step to optimize of radioisotopes production [2].

Here we provide a new measurement of the cross-sections for the ^{nat}Zn(p,x)^{67}Cu and ^{nat}Zn(p,x)^{67}Ga reactions in the range of 47 to 67 MeV. The ^{67}Cu and ^{67}Ga radionuclides show gamma ray peaks at the same energy and have similar half-life times. The overlapped gamma spectra for ^{67}Cu and ^{67}Ga were separated by employing an analytical method without the support of radiochemical separation. Our data are compared with previous published values and with theoretical estimations provided by the TALYS code [1].

2. Methods and Results

Cross-section measurements for proton-induced reactions on natural Zn foils (purity: 99.9%) have been carried out at an energy of 69 MeV proton beam. The thickness of the Zn foils was 48.799 ± 0.443 μm. The experiments were made by using a 100 MeV proton accelerator installed at KOMAC. To determine the incident proton flux, stacked Zn foils (purity: 99.9%) were activated along with Al monitor foils, as well as the accumulated particles incident on the foils are directly measured. To change of incident energy for each foil, we put the Al degraders onto the foils in between. Each foil was stacked in the order of Al-Zn-Degrader into a collimator.

Figure 1 shows the foils and degraders used in this work. The area and mass of the foils are measured several times using a Vernier calipers and an electronic scale, respectively, to diminish systematical errors.

2.1 Calculation of the cross-section for ^{nat}Zn(p,x)^{67}Cu and ^{nat}Zn(p,x)^{67}Ga reactions

The activity A of the radioactive nuclei that produce and disintegrate during the activation or irradiation time (t_i) is determined [4]

$$A(t) = N_0\sigma\phi\left(1 - e^{-\lambda t_i}\right)e^{-\lambda t}\,, \qquad (1)$$

where N_0 is the number of radioactive nuclei at an initial time, σ reaction cross-section of the nuclide, ϕ the flux of incident particles, and λ the decay constant of half-life ($t_{1/2}$) of radioactive decay.

The flux on incident particles on the reference foils can be expressed,

Table I: Deconvolution analysis for overlapped gamma spectra of ^{67}Cu and ^{67}Ga

Gamma-ray energy	E_0 [keV]	E_1 [keV]	E_2 [keV]	E_3 [keV]	…	E_n [keV]
Absolute γ-ray intensity for nuclide A	I_{A,γ_0}	I_{A,γ_1}	I_{A,γ_2}	I_{A,γ_3}	…	I_{A,γ_n}
Absolute γ -ray intensity for nuclide B	I_{B,γ_0}	I_{B,γ_1}	I_{B,γ_2}	I_{B,γ_3}	…	I_{B,γ_n}
γ -counting efficiency	ϵ_{γ_0}	ϵ_{γ_1}	ϵ_{γ_2}	ϵ_{γ_3}	…	ϵ_{γ_n}
γ -peak area for the counting time $t_{counting}$ for nuclide A	S_{A,γ_0}	S_{A,γ_1}	S_{A,γ_2}	S_{A,γ_3}	…	S_{A,γ_n}
γ -peak area for the counting time $t_{counting}$ for nuclide B	S_{B,γ_0}	S_{B,γ_1}	S_{B,γ_2}	S_{B,γ_3}	…	S_{B,γ_n}
Total corrected count numbers, $N_n = A_n + B_n$	N_0	N_1	N_2	N_3	…	N_n
Corrected count numbers for nuclide A, $A_n = S_{A,\gamma_n}/\epsilon_{\gamma_n}$	A_0	A_1	A_2	A_3	…	A_n
Corrected count numbers for nuclide B, $B_n = S_{B,\gamma_n}/\epsilon_{\gamma_n}$	B_0	B_1	B_2	B_3	…	B_n

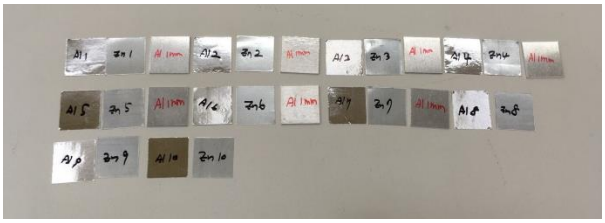

Fig. 1. Image of the Al reference foils, Al degraders, and Zn foils used in this work.

$$\phi = \frac{N\lambda}{N_0 \sigma \epsilon I_\gamma} \frac{1}{\left(1-e^{-\lambda t_i}\right) e^{-\lambda t_w} \left(1-e^{-\lambda t_m}\right)}, \quad (2)$$

where I_γ is the intensity of γ-ray, t_w cooling or waiting time, and t_m measuring time. Reaction cross-section of the nuclide can thus be calculated using ϕ from the reference Al foils,

$$\sigma[cm^2] = \frac{N\lambda}{N_0 \phi \epsilon I_\gamma} \cdot \frac{1}{\left(1-e^{-\lambda t_i}\right) e^{-\lambda t_w} \left(1-e^{-\lambda t_m}\right)}. \quad (3)$$

2.2 Peak convolution

The overlapped gamma-ray spectrum analysis was made by employing a mathematical method, but not with radiochemical separation. From the definition of γ-ray absolute intensity of the nuclides it is written,

$$\frac{A_n}{A_0} = \frac{I_{A,\gamma_n}}{I_{A,\gamma_0}}, \frac{B_n}{B_0} = \frac{I_{B,\gamma_n}}{I_{B,\gamma_0}} \quad (4)$$

$$N_0 = A_0 + B_0 \quad (5)$$

$$N_1 = A_1 + B_1 = \frac{I_{A,\gamma_1}}{I_{A,\gamma_0}} A_0 + \frac{I_{B,\gamma_1}}{I_{B,\gamma_0}} B_0 \quad (6)$$

Combining the Eqs. (5) and (6) gives

$$A_0 = \frac{I_{A,\gamma_0}\left(I_{B,\gamma_0} \cdot N_1 - I_{B,\gamma_1} \cdot N_0\right)}{I_{A,\gamma_1} \cdot I_{B,\gamma_0} - I_{A,\gamma_0} \cdot I_{B,\gamma_1}},$$

$$B_0 = \frac{I_{B,\gamma_0}\left(I_{A,\gamma_1} \cdot N_0 - I_{A,\gamma_0} \cdot N_1\right)}{I_{A,\gamma_1} \cdot I_{B,\gamma_0} - I_{A,\gamma_0} \cdot I_{B,\gamma_1}}. \quad (7)$$

In Table I, we summarize the parameters being of physical meaning on deconvolution process. From this analytical methods we estimated each contribution of ^{67}Cu and ^{67}Ga radionuclides from the same peak, the obtained data being presented in Figs. 2 and 3. The data being obtained from this work show very similar behavior compared with previous experimental results, in which most previous works are made by the support of radiochemical separation. We thus confirmed the previous production results are reproducible for both ^{67}Cu and ^{67}Ga radionuclides in our analytical method. Extending of a larger energy range would be made to maximize the production of the radionuclide of interest. The best energy range should be evaluated for each specific case, considering the cost of the enriched material and the final use of ^{67}Cu radionuclide in future work.

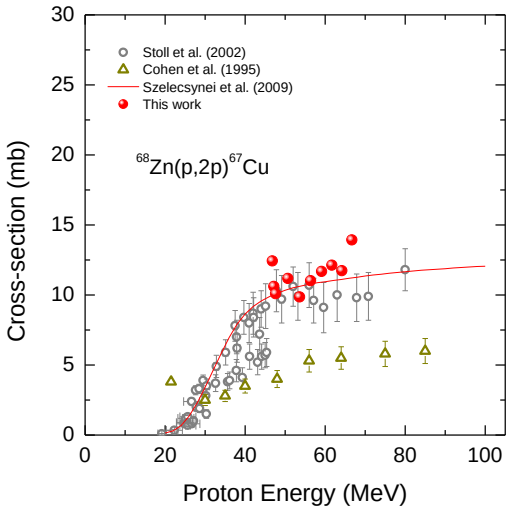

Fig. 2. Comparison of measured cross sections of ^{67}Cu from ^{68}Zn(p,2p)^{67}Cu reaction with those reported previously.

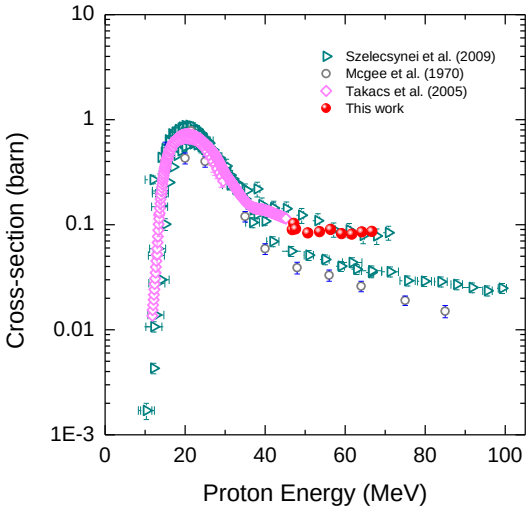

Fig. 3. Comparison of measured cross sections of ^{67}Ga from ^{68}Zn(p,2n)^{67}Ga reaction with those reported previously.

3. Conclusions

In this work, we have obtained the cross-sections for the ^{nat}Zn(p,x)^{67}Cu and ^{nat}Zn(p,x)^{67}Ga reactions in the 47 to 67 MeV energy range with an analytical peak separation method. The experiment data was a good agreement with previously reported cross-section data.

4. Acknowledgments

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