Excitation function of ^{nat}Cu(^{3}He,x)^{65}Zn nuclear reaction for ^{3}He beam monitoring purpose

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Abstract. The excitation function of the ^{nat}Cu(^{3}He,x)^{65}Zn nuclear reaction was measured from its threshold up to 53 MeV incident energy relative to the ^{nat}Ti(^{3}He,x)^{48}V monitor reaction by using the conventional stacked foil activation technique combined with HPGe γ-ray spectrometry. Our result is systematically higher than the IAEA recommendation, and more experimental works are desired especially above 25 MeV.

1 Introduction

Production of medical radionuclides via accelerator pathway and/or via charged-particle induced nuclear reactions shows great attention in recent time, and their production cross-sections are important nuclear data to identify efficient production routes. The accuracy of the charged-particle induced nuclear reaction cross-sections largely depends on the precise determination of the accelerated particle beam parameters such as beam intensity. Monitoring of beam intensity can be done in several ways, and some well-studied nuclear reactions (monitor reactions) could be used with high accuracy in this purpose. Understanding the importance of beam parameter monitoring during irradiation, a number of monitor reactions are recommended by the International Atomic Energy Agency [1], where the ^{nat}Cu(^{3}He,x)^{65}Zn is included as one of the recommended reactions to be used for ^{3}He beam monitoring purpose. However, the recommended data of ^{nat}Cu(^{3}He,x)^{65}Zn reaction was obtained based on few experimental data sets above 20 MeV, where they are contradicting each other including the peak region around 30 MeV. In connection to this, we decided to verify the IAEA recommended cross-sections for the ^{nat}Cu(^{3}He,x)^{65}Zn beam monitoring reaction and to enrich the literature database by contributing new data for this reaction of interest. The present work concerns the measurement of the ^{nat}Cu(^{3}He,x)^{65}Zn excitation function from its threshold up to 53 MeV incident energy by using the conventional stacked foil activation technique combined with HPGe γ-ray spectrometry.

2 Experimental

The experimental technique and data reduction procedure of this work are similar to those adopted in our earlier works [2–9]. However, some important information relevant to the present work is given as follows:

A high purity (99.9%) natural copper foil (9.88-μm thick; Nilaco, Japan) was used as the target material, while metallic foils of titanium (5.20-μm thick; 99.6% purity; Nilaco, Japan) and tungsten (10.75 and 14.14-μm thick; 99.95% purity; Nilaco, Japan) with natural isotopic compositions were used for beam monitoring and additional excitation function measurements. All foils were individually cut into square sizes (12 mm × 12 mm), and a typical stack was prepared by placing them to an order of Ti→Cu→W in a regular fashion. A few extra foils were placed at the back side of the target stack to evaluate the probable activities generated by the secondary particles. The stacked samples were then placed in a target holder, which served as a Faraday cup. The exit point of the cyclotron beam line was collimated to 4-mm diameter to ensure that the monitor and target foils received the same number of ^{3}He beam particles. The stacked foils were then irradiated by a ^{3}He beam for 1.5 h with an average beam current of ~100 nA. The energy at the exit point of the cyclotron beam was determined to 55.36 MeV by using the beam energy measurement system installed in the cyclotron via ToF method [10]. After sufficient cooling time, the stacked foils were removed from the target holder, and measured non-destructively by using a high resolution HPGe γ-ray spectrometer (EURISYS MESURES; EGC-15-185-R, 76 cm^3, France). A computer program (MCAwin2000 software; RZMCA, Laboratory Equipment Corp., Ibaraki, Japan) was used to analyze the acquired γ-

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ray spectra. The detection efficiencies for each measured distance were evaluated using a multi-nuclide γ-ray point source (MX402; JIS Z4821-2) obtained at the time of the experiment [2]. The degradation of the ^{3}He beam energy along the stacked samples was calculated by using a computer program described by Williamson et al. [12]. The ^{3}He beam energies along the stacked samples were further adjusted to reduce the deviation from the IAEA recommended ^{nat}Ti(^{3}He,x)^{48}V cross-sections [2].

The IAEA recommended monitor reaction cross-section of ^{nat}Ti(^{3}He,x)^{48}V (σ =163 mb at E_{3He}= 55.27 MeV) [1, 13] was used to determine the beam intensity at the front positioned foil, and we considered that the beam flux was a constant throughout the whole stack. The cross-section of the ^{nat}Cu(^{3}He,x)^{65}Zn reaction was computed using the well-known activation formula available elsewhere [5, 14, 15]. The decay data of the ^{65}Zn nuclide were retrieved from the NuDat-2.6 interface [16]. The Q-values and threshold energies of the reaction of interest were calculated by the Q-tool system [17] on the basis of the Atomic Mass Evaluation [18, 19].

The uncertainties in the measured cross-sections were evaluated in the range of 6.1–27.3% by using the quadratic sum of the following contributing partial uncertainties: γ-ray counting statistics (0.8–27%), beam flux ($\sim$5%), detector efficiency ($\sim$3.5%), sample thickness ($\sim$2%), and γ-ray intensity ($\Delta I_\gamma/I_\gamma = 0.20\%$). On the other hand, the uncertainty/spread of the beam energy in the middle of each irradiated foil was estimated by considering those originated from the incident beam energy ($\sim$5%), target foil thickness ($\sim$2%) and path length straggling. The estimated uncertainty/spread of a representing energy point range from ±0.60 MeV to ±1.58 MeV.

3 Results and discussion

The production cross-sections and their total uncertainties of ^{65}Zn radionuclide are presented in Table 1, and the excitation function is plotted in Fig. 1 together with the earlier reported experimental data compiled in the EXFOR library [20], the evaluated data in the TENDL-2019 library [21] and the data calculated by us with the model code EMPIRE-3.2.3 [22].

The neutron deficient long-lived ^{65}Zn ($T_{1/2}$=243.93 d) radionuclide was populated via the contribution of several processes within our investigated energy region: (i) ^{63}Cu(^{3}He,p) (E_{thr}=0.0 MeV), (ii) ^{65}Cu(^{3}He,2n+p) (E_{thr}=10.31 MeV), and (iii) EC+β^+ decay (100%) of simultaneously produced short-lived ^{65}Ga ($T_{1/2}$=15.2 min). The ^{65}Zn decays to the stable ^{65}Cu via EC+β^+ decay (100%). The 1115.539 keV γ-ray (I_γ=50.04%) [23] emitted following the EC+β^+ decay of ^{65}Zn is the only characteristic and intense γ-line to identify ^{65}Zn, and it was adopted in this work. A general survey of the literature revealed that three earlier works [24–26] are available on the production cross-sections of ^{65}Zn via helion activation on natural copper target, and detailed information on the experimental parameters adopted by the respective authors are summarized in Table 2. The table shows that the three experimental works used a monitor reaction and/or

Table 1. Measured cross-sections of ^{nat}Cu(^{3}He,x)^{65}Zn reaction.

Helion Energy (MeV)		Cross-section (mb)	
E	ΔE	σ	$\Delta\sigma$
52.85	0.60	128.5	7.8
49.96	0.62	148.3	9.7
46.99	0.63	165.0	10.7
43.94	0.64	187.0	12.0
40.79	0.66	212.9	13.6
37.53	0.68	248.1	15.7
34.14	0.70	289.3	18.2
30.61	0.72	314.4	19.7
27.56	0.75	316.3	19.7
22.74	0.80	232.9	15.8
17.48	0.86	98.8	6.8
11.59	0.97	29.6	1.9
6.18	1.58	3.3	0.9

Faraday cups. The numerical values of the monitor cross-sections adopted in these works are absent in their publications, thus no attempt was made to renormalize their published cross-sections by using the latest IAEA recommended monitor cross-sections. On the other hand, all authors report the intensity of the 1115 keV γ-line used for ^{65}Zn identification, and their excitation functions were renormalized by using the latest intensity adopted by us (*i.e.,* 50.04%) for comparison.

Figure 1 shows our result is slightly lower than Kondratyev et al., [25] above 25 MeV, and systematically higher than the IAEA recommended cross-sections [1, 13], which agrees with Lebowitz and Greene (1970) [24] above 25 MeV. The TENDL-2019 library is close to our excitation function below 20 MeV but does not predict the peak around 30 MeV, while EMPIRE-3.2.3 predicts a quite low excitation function.

4 Conclusion

Activation cross-sections for the ^{nat}Cu(^{3}He,x)^{65}Zn reaction were measured using the stacked-foil activation technique with an overall uncertainty of better than 7% in general. The beam flux was determined from the front Ti foil activity and the IAEA recommended ^{nat}Ti(^{3}He,x)^{48}V monitor reaction cross-section, while the incident energies of the other foils were adjusted to reduce the deviation from the recommended cross-section. The IAEA recommended cross-section for the ^{nat}Cu(^{3}He,x)^{65}Zn reaction is systematically lower than our excitation function above 25 MeV, and more experimental works are desired to resolve the discrepancies between the measurements and recommendation in this energy region.

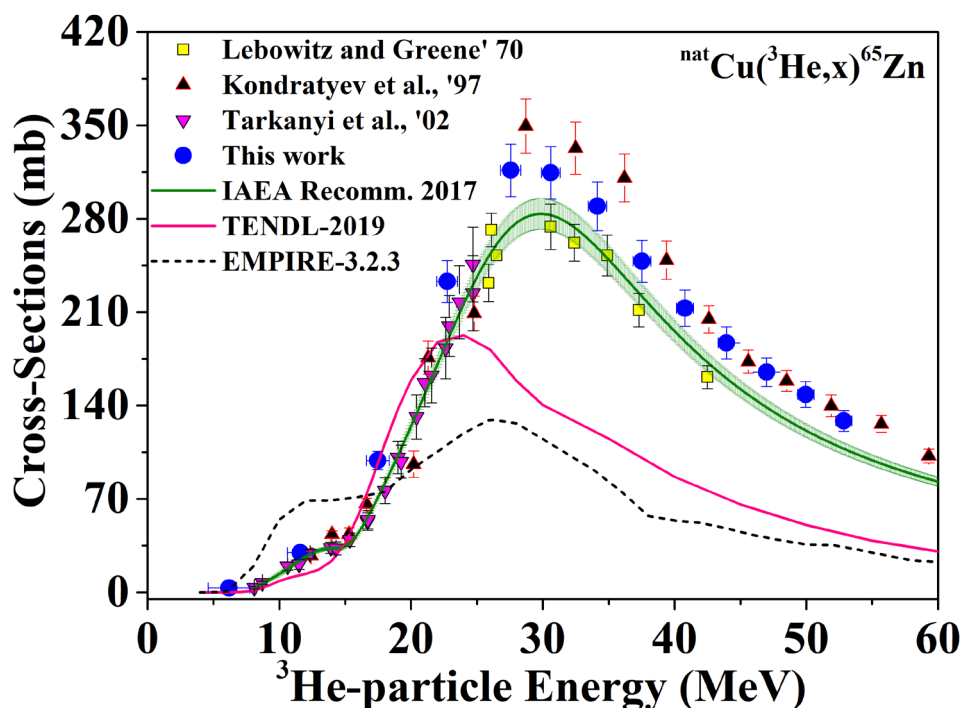**Figure 1.** Excitation function for the $^{nat}Cu(^3He,x)^{65}Zn$ reaction. The excitation functions in the literature are corrected for the renormalization factor in Table 2. The TENDL-2019 and EMPIRE-3.2.3 excitation function curves include the contribution of $^{nat}Cu(^3He,x)^{65}Ga$ reaction.

Table 2. Measured $^{nat}Cu(^3He,x)^{65}Zn$ excitation functions in the literature. All works use the 1115 keV γ-line and I_γ is the intensity adopted in each work. In Fig. 1, the cross-sections in the literature are multiplied by the renormalization factor which is the ratio of the I_γ value adopted by the authors to the I_γ value adopted by us (=50.04%). Lebowitz and Greene (1970) reported isotopic cross-sections for $^{65}Cu(^3He,x)^{65}Zn$ reaction from a measurement with a natural sample, and their data renormalized to the natural abundance are plotted in Fig. 1.

Authors	Incident energy (MeV)			Beam monitoring	I_γ (%)	Renormalization factor
Lebowitz et al. (1970) [24]	25.9	-	42.5	Faraday cup	49	0.97
Kondratyev et al. (1997) [25]	12.4	-	91.6	$^{27}Al(^3He,x)^{24,22}Na$, Faraday cup	50.75	1.01
Tárkányi et al. (2002) [26]	8.1	-	24.7	$^{nat}Ti(^3He,x)^{48}V$, Faraday cup	50.75	1.01

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