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Measurements of ^{67}Ga production cross section induced by protons on ^{nat}Zn in the low energy range from 1.678 to 2.444 MeV

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ABSTRACT

The experimental production cross section for the reaction ^{nat}Zn(p,x)^{67}Ga has been measured in the energy range from 1.678 to 2.444 MeV. The methodology used in this work is based on characteristic X-ray emitted after irradiation by the daughter nuclei that decays by electron capture (EC) and the use of a complementary PIXE experiment. By doing so, expressions needed to determine cross section values are simplified since experimental factors such as geometric setup and an detector efficiency are avoided. ^{67}Ga is a radionuclide particularly suited for this method since it decays by electron capture in 100% and the subsequent characteristic X-ray emission is easily detected.

Natural zinc targets were fabricated by PVD technique and afterwards their thicknesses were determined by Rutherford Backscattering Spectrometry. Cross sections measurements were carried out by using the Van de Graaff accelerator located at Faculty of Sciences, University of Chile. It was found that our data for the ^{nat}Zn(p,x)^{67}Ga reaction are, in general, in good agreement when compared to existing experimental data and to those calculated ALICE/ASH nuclear code. On the other hand, values predicted by Talys-1.6 are showing systematically lower magnitudes than our measured data.

1. Introduction

It is well known that cross section measurements are very important and widely used in the fields of ion beam analysis, industry and biomedical applications [1–3]. Within the latter, Nuclear Medical Diagnostics is a very important tool often utilized for non-invasive determination of physiological processes through the use of radioisotopes usually produced by accelerators. One example is ^{67}Ga ($t_{1/2} = 78.3$ h) which decays 100% by electron capture [4]. This radionuclide is a $\gamma-$emitter that is frequently produced below 18 MeV by using two different nuclear reactions with ^{nat}Zn as target, namely ^{67}Zn(p,n)^{67}Ga and ^{68}Zn(p,2n)^{67}Ga, becoming one of the most employed cyclotron produced radioisotopes [5,6] and is commonly utilized for medical scintigraphy in the diagnosis of infections, lymphomas, granulomatous diseases and temporal arteritis [7]. But besides their practical applications, cross section values can be used to test the prediction capabilities of nuclear reaction models, particularly in energy regions where experimental data are not available or they are very hard to measure due to the experimental difficulty. In these cases, nuclear reaction models are commonly utilized to provide estimates of the particle-induced reaction cross sections and therefore they play a key role in the nuclear data evaluation.

In this work we measured the production cross section of ^{67}Ga by the reaction ^{nat}Zn(p,x)^{67}Ga in the low energy range of 1.678 to 2.444 MeV. To determine these cross section values, the methodology outlined in Ref. [8] was applied since the requirement of a daughter radionuclide decaying by electron capture is fulfilled. Accordingly, the overall uncertainty of the data obtained is significantly reduced because the knowledge of experimental parameters that usually have large uncertainties, like detector efficiency, solid angle sustained by the detector, and attenuation coefficients, are avoided. It is worth mentioning that for this nuclear reaction, previous studies such as those found in Ref. [9,10,5,11–13] attained higher energies and only Ref. [14] reports data below 3 MeV. Following this line of research, the results obtained in this study are then compared to both, data available in the literature and to those derived from both ALICE/ASH [15] and Talys-1.6 [16] nuclear codes. By doing so, it is expected that the obtained data will help to improve nuclear theoretical models so features such as nuclear reaction mechanisms and the properties of the excited states in different energy regimes will be better explained.

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2. Experimental methods

The experimental set-up and procedure used in this work were similar to those described in our previous paper [8]. As established in that reference, if the final product of a particular nuclear reaction is a radionuclide that decays by electron capture, it is feasible to determine the production cross section by measuring the characteristic X-ray emitted during the decaying process. With the aim to reduce the uncertainty budget, a separate PIXE (particle induced X-ray emission) [17] type experiment is performed on a well characterized thin target of the daughter nuclei. Thus, parameters with usually large uncertainties and/or hard to measure (e.g., detector solid angle, efficiency) are not involved in the experimental cross section values but replaced by others easier to determine like charged collected, peak area and time intervals. So far, this methodology has been utilized in our laboratory to successfully measure the production cross section of ^{63}Cu(d,p)^{64}Cu [18] and ^{nat}Zn(p,x)^{67}Ga [19] nuclear reactions. In these references it is demonstrated that, if an ion beam is used to bombard a target of stable nuclei then the radioactive nuclei that decay by electron capture could be eventually produced, and the corresponding production cross section at a beam energy E is given by the following expression:

$$\sigma(E) = \sigma_x(E_p)\left(\frac{N_{exp}}{N_x}\right)\left(\frac{n^*}{n_{Zn}}\right)\left(\frac{\phi_p}{\phi}\right)\Delta \quad (1)$$

$$\Delta = \frac{\lambda\tau}{f_{EC}(e^{\lambda T}-1)(e^{-\lambda t_1}-e^{-\lambda t_2})}, \quad (2)$$

where σ_x corresponds to the K_α emission cross section [20], N_x is the number of K_α X-rays detected in a time interval τ when using a proton beam of energy E_p and flux ϕ_p, λ is the decay constant, n_Z is the atomic surface density of the parent nucleus, ϕ represents the flux of the proton beam, T is the irradiation time, $\Delta t = t_2 - t_1$ is the time interval during which N_{exp} X-rays originated from the E.C. decay were detected, and f_{EC} corresponds to the probability factor for E.C. decay. It is worth mentioning that, since in this work the methodology outlined above was applied to measure the production cross section for the reaction ^{nat}Zn(p,x)^{67}Ga and this radionuclide decays by E.C. to the same element (see Fig. 1), then in Eq. (1) the term $(n^*/n_{Zn}) = 1$.

The cross section measurements were performed at the 3.75 MV Van de Graaff accelerator of the Faculty of Sciences of University of Chile [21]. The energy of the accelerator was calibrated prior to these measurements by using the 872, 934 and 1370 keV resonances of the $^{19}F(p,(\alpha\gamma)^{16}$O reaction, the 1733 keV resonance of ^{12}C(p,p)^{12}C reaction and the 2085 keV resonance of ^{28}Si(p,p)^{28}Si reaction [22]. The irradiations covered an energy range from 1.678 to 2.444 MeV with beam intensities in the range of 0.23 to 1.31 μA. Six thin zinc targets were prepared by physical vapor deposition (PVD) [23,24] at high vacuum conditions using natural zinc provided by GoodFellow in wire shape with a 99.95% of purity [25] having natural isotopic composition (^{64}Zn 48.63%, ^{66}Zn 27.90%, ^{67}Zn 4.10%, ^{68}Zn 18.75% and ^{70}Zn 0.62%). Five targets were evaporated onto an aluminum backing (5 mm thickness) while one of them was a self supporting target. During each measurement the target holder was connected to a cooling system using liquid nitrogen in order to remove the heat produced by the proton beam impact, and hence to keep the target temperature well below its melting point (420 °C) [26]. As can be seen in Fig. 2 this requirement was always met during irradiation with 2.33 MeV protons where target temperature never exceeds −50 °C. Rutherford Backscattering Spectrometry (RBS) [27] was utilized to determine target thicknesses which were obtained from the fitted RBS spectra by using the SIMNRA program [28]. Target thicknesses varied from 2.38 ± 0.12 to 2.68 ± 0.13 μm. Also, gamma spectroscopy analysis was applied to one of these targets as a quality control test. A HPGe ORTEC detector Model GEM-10195 was used to collect a gamma spectrum at proton energy 1307 keV. As shown in Fig. 3, gamma rays emitted by ^{67}Ga after decaying by EC, and depicted as black rows in Fig. 1, are clearly recognized in this spectrum. In addition, the synthesis of both nuclei ^{65}Ga ($t_{1/2} = 15.2$ min.) and ^{68}Ga ($t_{1/2} = 67.8$ min.) is revealed from the 511 keV peak. However, they were neglected during our cross section measurements since their half-lives are much shorter than for ^{67}Ga.

The first stage of every production cross section $\sigma(E)$ measurement consists into determine the term N_x. For that purpose each target was mounted inside the scattering chamber and a PIXE experiment was run for $\tau = 5$ min and using beam currents in the range between 0.24 and 0.31 nA. A Canberra 7333 Si(Li) cryogenic detector having 220 eV FWHM resolution at 5.9 keV was used to obtain the characteristic X-ray spectra. Pulses were then picked up by a Canberra 2008 preamplifier and a Ortec 672 main shaping amplifier. Further signal processing is carried out by an ORTEC PC MCA model Trump-8K. Then, during the second stage, the targets were mounted on the above mentioned cooling system and positioned in an irradiation chamber, with irradiation time intervals T lasting between 180 and 342 min. On average, twenty minutes after the end of irradiations, the irradiated natural zinc

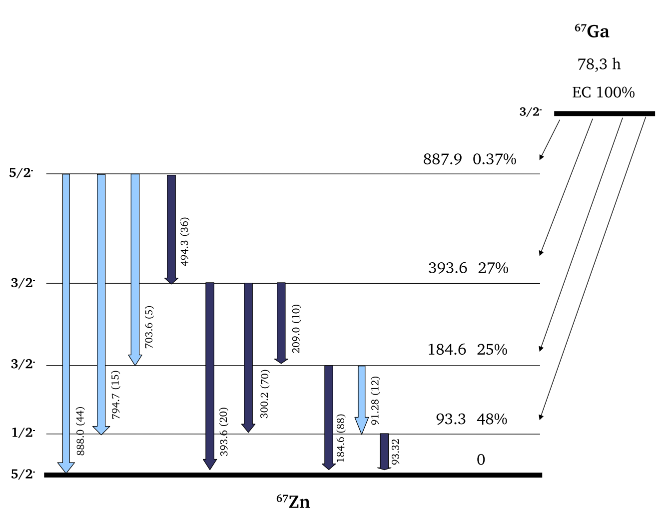

Fig. 1. Decay scheme of ^{67}Ga. Shaded arrows (in black) correspond to gamma rays which were later identified during acquisition of gamma spectra.

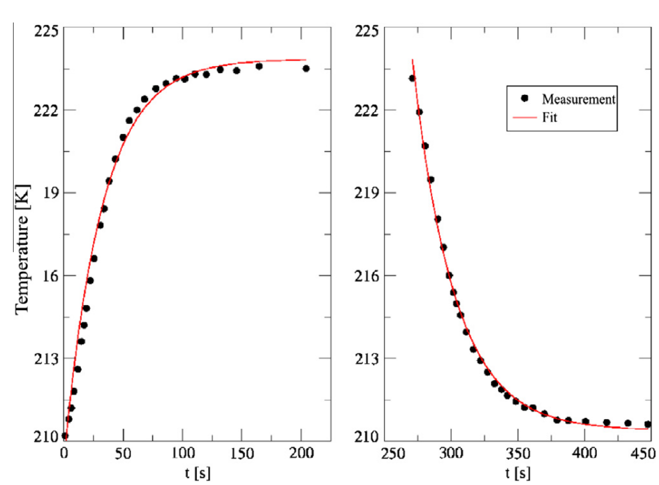

Fig. 2. Target temperature reached during irradiation by using 2.33 MeV protons and a density current of 0.16 μA/mm^2, approximately. (a) A maximum temperature is clearly observed, which was always kept below −50 °C. (b) Temperature curve at end of the irradiation.

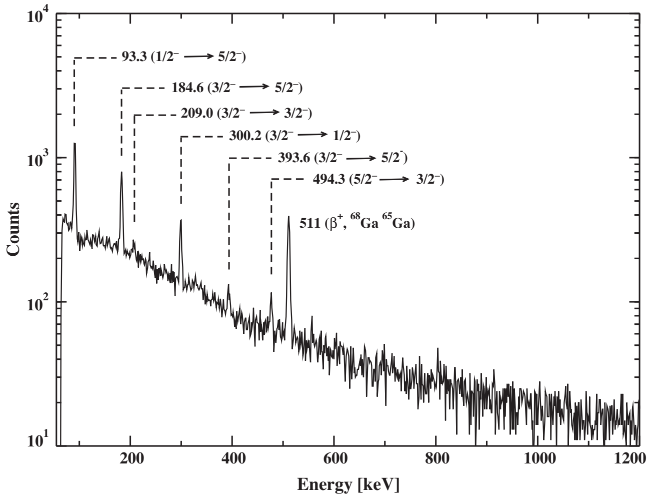

Fig. 3. Spectrum showing gamma rays emitted by ^{67}Ga after decaying by EC and obtained at proton energy 1307 keV.

targets were installed inside the PIXE chamber and the emitted X-rays started to be acquired. The collected X-ray spectra from the ^{67}Ga decay, with collecting time intervals Δt reaching up to 170 h, were saved in periods of 15 min. The total K_α net area counts was obtained by summing every X-ray spectra (see Fig. 4).

3. Results and analysis

Proton induced production cross section $\sigma(E)$ for the reaction $^{nat}Zn(p,x)^{67}Ga$ were measured at four energies between 1.678 and 2.444 MeV, as shown in Table 1. In order to determine the measured uncertainties $\Delta\sigma$, the contribution of experimental parameters such as beam energy, time intervals (0–1%), beam current (5–10%), target thickness (5–9%) and counting statistics (1–10%) has been considered. The uncertainty for σ_X cross section has been estimated around 2% from Ref. [17]. In this way, the total uncertainty added up to 19%. Additionally, the average uncertainties of the proton energies ΔE shown in Table 1 were estimated by considering both the uncertainty of the primary beam energy

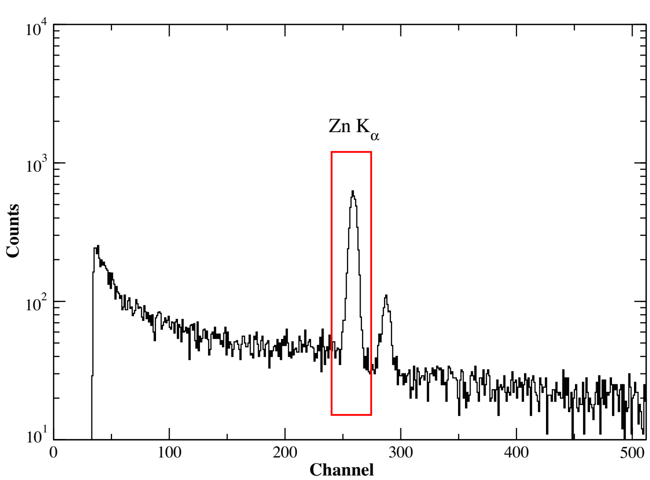

Fig. 4. Characteristic X-ray spectra obtained after irradiation time T. The K_α and K_β peaks corresponding to zinc are clearly seen. After a collecting time interval Δt, the net area of K_α line in this spectra was determined by setting a Region of Interest (ROI), shown as enclosed in a rectangle, which is then used to determine parameter N_{exp}.

Table 1
Experimental production cross section values for $^{nat}Zn(p,x)^{67}Ga$.

Energy (MeV)	Beam current (ϕ) (μA)	$\sigma \pm \Delta\sigma$ (mb)
1.678 ± 0.039	0.23 ± 0.02	0.0196 ± 0.0038
2.212 ± 0.046	1.31 ± 0.13	0.204 ± 0.034
2.328 ± 0.047	0.78 ± 0.08	0.79 ± 0.10
2.444 ± 0.048	0.54 ± 0.05	2.08 ± 0.28

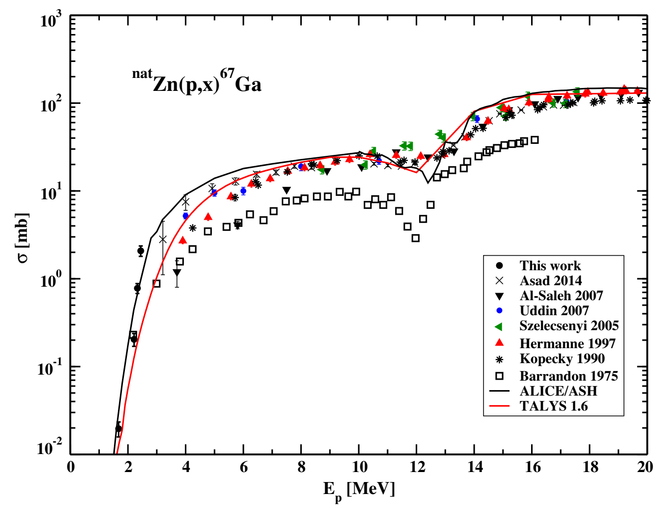

Fig. 5. Production cross section values obtained in this work are compared with other experimental data and also with theoretical predictions based on the Talys-1.6 and ALICE/ASH nuclear codes.

and the contribution arising from the uncertainty of the thickness or the target.

It should be stressed that in this study ^{nat}Zn is being used as target and the range of incident energies surpasses the threshold for neutron emission. Therefore, the experimental cross section $\sigma(E)$ data corresponds to contributions of two open channels multiplied by the respective isotopic abundances, so it can be expressed by the following expression:

$$\sigma(E) = 0.279 \times \sigma_{p,\gamma} + 0.041 \times \sigma_{p,n}, \quad (3)$$

where $\sigma_{p,\gamma}$ and $\sigma_{p,n}$ are the $^{66}Zn(p,\gamma)$ and $^{67}Zn(p,n)$ reaction cross sections, respectively. Besides this two channels, theoretical calculations have been done considering the contribution from $^{68}Zn(p,2n)$ nuclear reaction, which has an energy threshold of 12 MeV, approximately. Predictions of the cross section for the nuclear reaction used for ^{67}Ga production in this study were carried out by using two well known nuclear codes. The ALICE/ASH code [15], which now has been extensively used for nuclear data evaluation during the last decades [29–32], is a modified and more advanced version of the original code. This program uses the geometry dependent hybrid model (GDH) to describe pre-equilibrium particle emission from nuclei while the equilibrium emission of particles is described by the Weisskopf–Ewing model without detail consideration of angular momentum. Three models are used for the calculation of nuclear level density, although as shown in Fig. 5, it was found that the best agreement with experimental data, in the whole energy range, is reached when using the Fermi-Gas with excitation-energy dependent parameter given by $a = A/9$, where A is the mass number for the compound nucleus. For proton projectiles the reaction cross sections are calculated according to the optical model. On the other hand, The Talys-1.6 cross sections presented in this Fig. 5 were calculated using the default parameter set of this program.

Fig. 5 also shows previous measurements which were taken from EXFOR database [33]. It is clear that for proton energies over 3.0 MeV the existing data is rather scattered and they do not seem to follow a common trend. However, it is fair to notice that the data from Ref. [6] not only agrees very well with theoretical calculations, particularly with the ALICE/ASH, but also their data seem to match ours at lower energies. Additionally, there is an excellent agreement between our second data at 2212 keV and the last data from Ref. [14]. On the other hand, predicted values from Talys-1.6 at higher energies are similar to the available experimental data but predicted absolute values are quite lower when compared with our data in the energy region below 3 MeV.

4. Conclusions

In this work, production cross section values for the reaction $^{nat}Zn(p,x)^{67}Ga$ were measured in the energy range between 1.678 and 2.444 MeV and new data were obtained with an overall uncertainty of up to 19%. Data were measured by applying a PIXE methodology which when applied, as established in Ref. [8], is able to reduce significantly the expressions needed to calculate cross section values. Our results are in fair agreement with both, available experimental data in literature and calculations performed with ALICE/ASH nuclear code. Predicted values from Talys-1.6 are quite lower when compared to our data in the energy range covered in this work.

In the higher energy region, where the available cross section values show a rather significant dispersion, our data are close to more recent measurements. On the other hand, at low energies our results show very good agreement not only with ALICE/ASH calculations but also with the only data measured so far below 3.0 MeV [14], despite the experimental methodology mentioned in that reference is quite different to ours.

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