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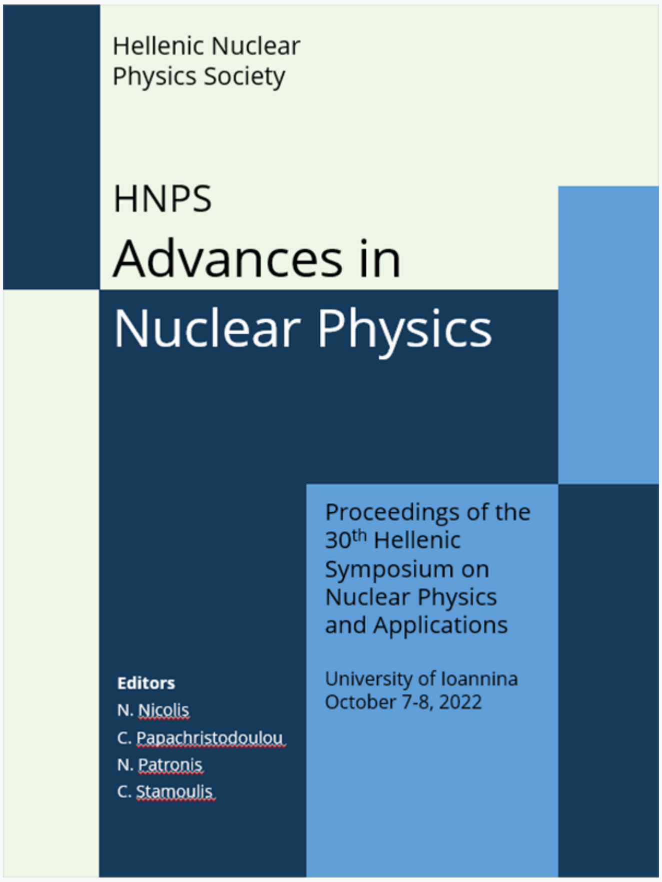

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Cross Section Measurements of (n,x) Reactions In the Energy Range Between 16.4 and 18.9 MeV Using Highly Enriched Ge Isotopes

S. Chasapoglou[1,*], R. Vlastou[1], M. Kokkoris[1], M. Diakaki[1], V. Michalopoulou[1], A. Stamatopoulos[1,5], M. Axiotis[2], S. Harissopulos[2], A. Lagoyannis[2], M.I. Savva[3], I.E. Stamatelatos[3], T. Vasilopoulou[3], C. Lederer-Woods[4]

[1] *Department of Physics, National Technical University of Athens, 157 80 Athens, Greece*
[2] *Tandem Accelerator Laboratory, Institute of Nuclear and Particle Physics, NCSR Demokritos, 153 10 Aghia Paraskevi, Greece*
[3] *Institute of Nuclear and Radiological Sciences, Energy, Technology & Safety, NCSR Demokritos*
[4]*School of Physics and Astronomy, University of Edinburgh, United Kingdom*
[5]*Physics Division, Los Alamos National Laboratory, 87545, NM, USA*

Abstract In this work, the cross sections of the neutron induced reactions ^{70}Ge(n,2n)^{69}Ge, ^{76}Ge(n,2n)^{75}Ge, ^{73}Ge(n,p)^{73}Ga, ^{72}Ge(n,p)^{72}Ga, ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga, ^{74}Ge(n,α)^{71m}Zn, ^{72}Ge(n,α)^{69m}Zn, ^{73}Ge(n,nα)^{69m}Zn have been measured in the energy range between 16.4 and 18.9 MeV via the activation technique with respect to the ^{27}Al(n,α)^{24}Na reference reaction. Most of the existing experimental datasets found in literature for these reactions, were obtained with the use of a ^{nat}Ge target. In this case, however, the residual nucleus produced from some reaction channels could also be produced from neutron induced reactions in neighboring isotopes that exist in the ^{nat}Ge in their natural abundance, acting as a contamination to the measured yield of the reaction of interest. This parasitic contribution should then be subtracted, based on theoretical calculations that bear their own uncertainties. Isotopically enriched targets on the other hand, do not suffer from such contaminations, leading to more accurate experimental cross section results. In this work, five highly enriched targets have been used that helped in the determination of accurate cross section data, especially in the case of the ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga and ^{73}Ge(n,nα)^{69m}Zn challenging reactions, that will be presented in detail in this manuscript. The experiments were carried out at the 5.5 MV Tandem Van de Graaff accelerator of N.C.S.R. "Demokritos", implementing the ^{3}H(d,n)^{4}He reaction for the production of the quasi-monoenergetic neutron beams.

Keywords cross section, neutron activation, enriched targets, Ge

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INTRODUCTION

The study of neutron induced reactions on Ge isotopes is of major importance both for practical applications, as well as from a fundamental research point of view. Practical applications include, among others, dosimetry, nuclear medicine, astrophysical projects and reactor technology and γ-ray spectroscopy applications, thus the investigation of its behavior in a neutron field is very important [1-3]. Regarding fundamental research in the Nuclear Physics field, the residual nucleus through (n,xα) reaction channels, is produced in high spin isomeric states. The population of these sates is very strongly related to the spin distribution of the continuum phase space, as well as the spins of the discrete levels involved [1-3]. The accurate experimental determination of reaction cross sections that lead to the formation of such residual nuclei, can play a very important role in the study of the compound nucleus evaporation. Furthermore, there are a lot of available reaction channels on the five Ge isotopes, produced by neutron induced reactions, that reveal very interesting systematics, crucial for the optimization of input parameters in statistical model calculations. The simultaneous reproduction of a plethora of the available reaction channels yielding residual nuclei either in normal or isomeric states with the same set of input parameters, can act as a very sensitive constraint in

* Corresponding author: sotirischasapoglou@mail.ntua.gr

theoretical calculations. Moreover, the cross section measurements of the present work were carried out in the energy region above 15 MeV, where pre-equilibrium effects in the de-excitation of the compound nucleus become more significant [1], making these measurements very important. Neutron induced reactions have been studied in the past by the Nuclear Physics Group of the National Technical University of Athens, in the energy range of 8-11.4 MeV, with the quasi-monoenergetic neutron beams produced via the ^{2}H(d,n)^{3}He reaction [4, 5]. Despite the fact that the existing experimental data found in literature [6] for (n,x) reactions on Ge isotopes cover a wide energy range, they are discrepant and scarce, especially in the energy region above 15 MeV. Most of these datasets implement a ^{nat}Ge target for the cross section measurements. In this case, the residual nucleus that is produced from the measured reaction, is also inevitably produced from 'parasitic' or 'interfering' reactions from neighboring isotopes that exist in the ^{nat}Ge target in their natural abundance. These parasitic contributions become more significant above 14 MeV. The estimation of the contribution of these parasitic channels, that need to be subtracted from the measured yield, are heavily based on theoretical corrections, that are accompanied by their own uncertainties. The use of highly enriched targets on the other hand, such as the ones used in the present work, does not dictate such corrections since the contribution of parasitic reactions is negligible, rendering the provided data as more accurate and reliable.

EXPERIMENTAL DETAILS

The targets used in the present work were five isotopically enriched GeO_2 pellets of ^{70,72,73,74,76}Ge with enrichment levels of 97.71, 96.59, 96.07, 95.51 and 88.46% respectively, and were provided by the CERN n_TOF collaboration. Each pellet had a mass of ~2 g and was glued on a thin mylar foil which was in turn glued on a Al ring of 6 cm diameter. High purity Al metallic reference foils were also used for the determination of the neutron flux in the Ge targets. The diameter of the Ge targets and the Al reference foils was 2 cm.

The experiments were carried out at the 5.5 MV Tandem Van de Graaf accelerator of N.C.S.R. "Demokritos" [7, 8]. The quasi-monoenergetic neutron beams were produced via the ^{3}H(d,n)^{4}He reaction. The neutron producing target was a solid Ti-tritiated (TiT) target of 373 GBq activity, placed at the end of the beam line and was air cooled during the irradiations. In front of the TiT target, a 10 μm Mo foil was placed, where the deuteron beams lost part of its energy, while the target itself consists of a 2.1 mg/cm^{2} TiT layer placed on a 1 mm thick Cu backing for good heat conductivity.

The duration of the irradiations varied between 5 and 24 hours, taking into account the half-life of the residual nucleus of the measured reaction. The instabilities of the neutron beam were recorded with a BF_3 detector placed at a 3 m distance from the neutron producing target and at an angle with respect to the axis of the neutron beam to minimize neutron scattering. The spectra of the BF_3 detector were saved at regular time intervals of 300 sec, during the irradiations, in a separate ADC. The effect of the beam instabilities could then be considered in the offline analysis to correct for the decay of the nuclei produced during the irradiations. The distance between the neutron producing target and the first reference foil was 2.7 cm, leading to an uncertainty in the neutron beam energy in the order of 0.3 MeV for the Ge targets. The absolute value of the neutron flux was obtained with the use of the ^{27}Al(n,α)^{24}Na reference reaction. In this sense, two high purity Al foils were used in each irradiation, one in the front and one in the back of the measuring Ge targets.

The induced radioactivity of the Ge targets and Al reference foils was measured via HPGe detectors at a distance of 10 cm from the detector window to avoid significant pile-up or true coincidence summing effect corrections.

DATA ANALYSIS

The cross-section calculation for every reaction channel measured in this work was based on the following formula:

$$\sigma = \sigma_{\mathrm{ref}} \cdot \frac{N_{\gamma,\mathrm{tar}}}{N_{\gamma,\mathrm{ref}}} \cdot \left[\frac{(\varepsilon F I_\gamma D f_c N_t)_{\mathrm{ref}}}{(\varepsilon F I_\gamma D f_c N_t)_{\mathrm{tar}}}\right] \cdot \frac{\Phi_{\mathrm{ref}}}{\Phi_{\mathrm{tar}}}, \quad (1)$$

where the subscripts "ref" and "tar" refer to the reference Al foils and measured Ge targets, respectively. Regarding the rest of the factors in eq. (1):

- $\boldsymbol{\sigma}$ is the cross section measured in barns. For the ^{27}Al(n,α)^{24}Na reference reaction, the cross section value was obtained from the ENDF/B-VIII.0 library [9]
- $\mathbf{N_\gamma}$ is the γ-ray peak integral from the spectrum of the HPGe detector. The γ-ray spectra were analyzed via the "Tv" software [10]
- $\boldsymbol{\varepsilon}$ is the absolute efficiency of the HPGe detector at each characteristic γ-ray energy
- $\mathbf{I_\gamma}$ is the γ-ray intensity
- $\mathbf{D}$ is a correction factor for the de-excitation of the residual nuclei during the cooling time (t_1), between the end of the irradiation and the start of the measurement in the HPGe detector, and during the measurement in the HPGe detector (t_2). This correction factor is given by the formula: $D = \exp(-\lambda t_1) - \exp(-\lambda t_2)$
- $\mathbf{f_c}$ is a correction factor for the decaying nuclei produced during the irradiation time (t_b), taking into account the fluctuations of the neutron beam. This correction factor is given by the expression: $f_c = \frac{\int_0^{t_b} e^{\lambda t} f(t)dt}{\int_0^{t_b} f(t)dt} \cdot e^{-\lambda t_b}$, where the factor f(t) represents the counts obtained from the BF_3 detector, stored in equal dt time intervals.
- $\mathbf{N_t}$ is the number of total nuclei of the measured isotope in the target
- $\mathbf{\Phi_{ref}/\Phi_{tar}}$ is the neutron flux ratio for the reference foil and the measured target respectively. This ratio was calculated via the combined use of NeusSDesc [11, 12] and MCNP5 [13] Monte Carlo codes.

In more detail, the propagation of the neutron beam through the consecutive stack of Al reference foils and Ge targets was achieved via the combined use of the NeuSDesc and MCNP5 Monte Carlo codes. Firstly, the NeuSDesc code was used to produce the neutron source, taking into account the physical and geometrical characteristics of the neutron producing target as an SDEF card, which was then fed into the MCNP5 code. In this code, a detailed description of the experimental setup was provided, and the neutron flux was calculated in each Al reference foil and Ge target.

PRELIMINARY RESULTS AND DISCUSSION

In these experimental campaigns, the ^{70}Ge(n,2n)^{69}Ge, ^{76}Ge(n,2n)^{75}Ge, ^{73}Ge(n,p)^{73}Ga, ^{72}Ge(n,p)^{72}Ga, ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga, ^{74}Ge(n,α)^{71m}Zn, ^{72}Ge(n,α)^{69m}Zn, ^{73}Ge(n,nα)^{69m}Zn reaction cross sections have been measured [14]. The half-lives and the most characteristic γ-rays alongside with their intensities, for all the measured reactions, are presented in Table 1. In the present manuscript, the attention will be focused on the ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga and ^{73}Ge(n,nα)^{69m}Zn challenging reactions, that can be most effectively studied with the use of isotopically enriched targets, relative to the ^{27}Al(n,α)^{24}Na reference reaction. The statistical uncertainties ranged from 2 to 6% for the neutron flux obtained from the reference reaction, and for the challenging reactions in the Ge isotopes, from 6 to 18%, due to the low counting rates. The uncertainties in the experimental points shown in Figs 1, 3 and 5, were obtained by summing up quadratically the statistical uncertainties along with the uncertainties of the neutron flux, the detector efficiency, and the γ-ray intensity.

Table 1. *Decay data used for the daughter nuclei of the measured and reference reactions*

Reaction	$t_{1/2}$	Eγ (keV)	Iγ (%)
^{70}Ge(n,2n)^{69}Ge	39.05 h	1106.77 574.11 871.98 1336.60	36.0 13.3 11.9 4.5
^{72}Ge(n,p)^{72}Ga ^{73}Ge(n,np/d)^{72}Ga	14.1 h	834.13 629.97 894.33	95.5 26.1 10.1
^{72}Ge(n,α)^{69m}Zn ^{73}Ge(n,nα)^{69m}Zn	13.76 h	438.63	94.9
^{73}Ge(n,p)^{73}Ga ^{74}Ge(n,np/d)^{73}Ga	4.86 h	297.32 325.70	79.8 11.9
^{74}Ge(n,α)^{71m}Zn	3.96 h	386.28 487.34	91.4 61.2
^{76}Ge(n,2n)^{75}Ge	82.78 min	264.6 198.6	11.4 1.2
^{27}Al(n,α)^{24}Na	14.99 h	1368.6	99.9

THE ^{73}Ge(n,d/np)^{72}Ga REACTION

The preliminary results of the ^{73}Ge(n,d/np)^{72}Ga reaction are presented in Figure 1 with red solid points for neutron energies of 16.4 and 17.9 MeV. The residual nucleus ^{72}Ga decays with a half-life of 14.1 h to ^{72}Ge that de-excites to its ground state with the emission of the 834.1, 629.9 and 894.33 keV γ-rays of 95.45%, 26.13% and 10.14% intensity, respectively. The γ-ray spectrum that was obtained from this reaction is presented in Figure 2 with the aforementioned γ-rays that were used for the calculation of the reaction cross section. Both ^{73}Ge(n,d)^{72}Ga and ^{73}Ge(n,np)^{72}Ga reactions produce the same residual nucleus. Therefore, the contribution of both of these reactions is measured with the activation method. The results of the present work display a very good agreement with the trend of the only dataset (Konno et al. [15]) that can be found for this challenging reaction. The study of this reaction with a ^{nat}Ge target is a very challenging task, considering the low natural abundance of the ^{73}Ge isotope with a combination of the low cross section value.

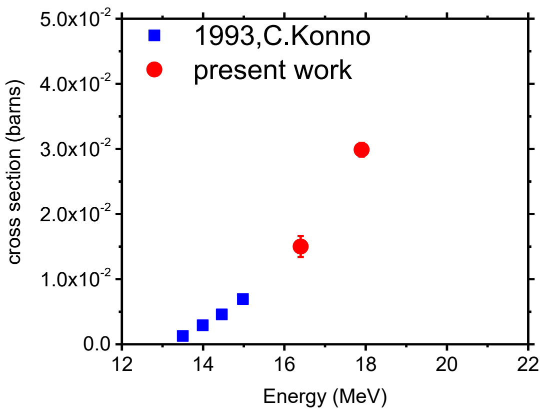

Figure 1. *The ^{73}Ge(n,d/np)^{72}Ga cross section results*

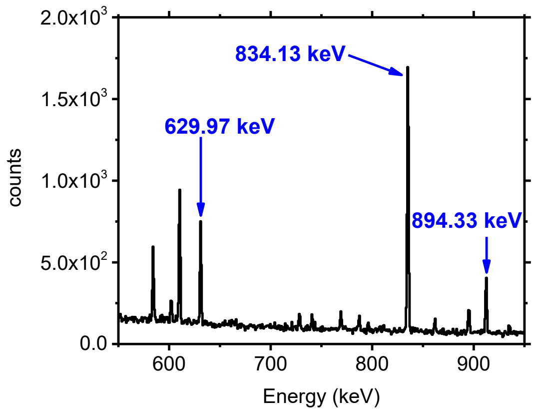

Figure 2. *The γ-ray spectrum obtained for the ^{73}Ge(n,d/np)^{72}Ga reaction*

The ^{74}Ge(n,d/np)^{73}Ga reaction

The preliminary results for the ^{74}Ge(n,d/np)^{73}Ga reaction are presented in Figure 3 with red solid points for energies of 16.4, 17.9 and 18.9 MeV. The residual nucleus ^{73}Ga that is produced from the reaction, decays to ^{73}Ge with a half-life of 4.86 h. Through this process, the characteristic 297.3 and

325.70 keV γ-ray lines are emitted with 79.8 % and 11.2 % intensity respectively. The γ-ray spectrum obtained from this reaction is presented in Figure 4, along with the aforementioned γ-rays. This is another example of a challenging reaction to study with a ^{nat}Ge target due to the significant contribution of the ^{73}Ge(n,p)^{73}Ga reaction to the measured yield, which can be considered negligible in the case of the enriched target. The production of the residual nucleus ^{73}Ga can be achieved from both ^{74}Ge(n,d)^{73}Ga and ^{74}Ge(n,np)^{73}Ga reactions, the contributions of which is indistinguishable with the activation method. The experimental data of the present work are in good agreement with the experimental point of Webber et al. [16] and in fair agreement with the trend of the dataset of Konno et al., slightly lower though than expected even considering the different contamination of the ^{74}Ge monoisotopic target with the ^{73}Ge(n,p)^{73}Ga reaction.

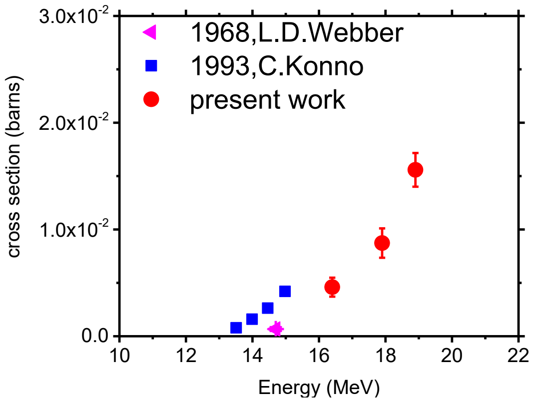

Figure 4. *The ^{74}Ge(n,d/np)^{73}Ga cross section results*

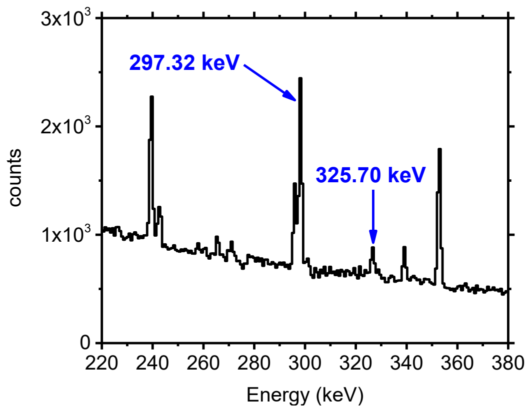

Figure 3. *The γ-ray spectrum obtained from the ^{74}Ge(n,d/np)^{73}Ga reaction*

The ^{73}Ge(n,nα)^{69m}Zn reaction

The preliminary results for the ^{73}Ge(n,nα)^{69m}Zn reaction are presented in Figure 5 with red solid points. The residual nucleus ^{69}Zn is produced both in its ground and metastable state. The ground state of ^{69}Zn decays directly to the ground state of ^{69}Ge, while the metastable state (with a half-life of 13.76 h) decays to the ground state of ^{69}Zn with the emission of the characteristic 438.6 keV γ-ray (with an intensity of 94.85 %), that is presented in the spectrum of Figure 6. No other datasets were found in literature for this challenging reaction, possibly due to the low natural abundance (7.76 %) of the ^{73}Ge isotope, found in ^{nat}Ge targets, that are most commonly used and the very low cross section.

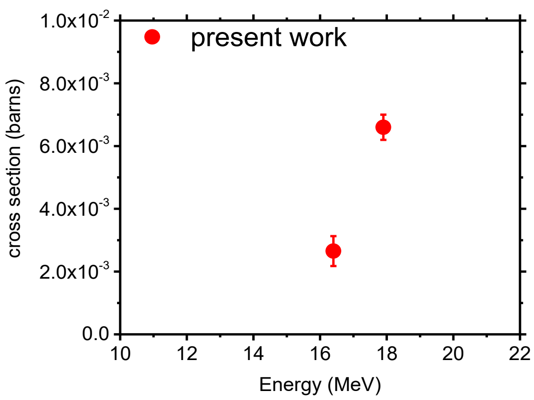

Figure 6. *The ^{73}Ge(n,nα)^{69m}Zn reaction results*

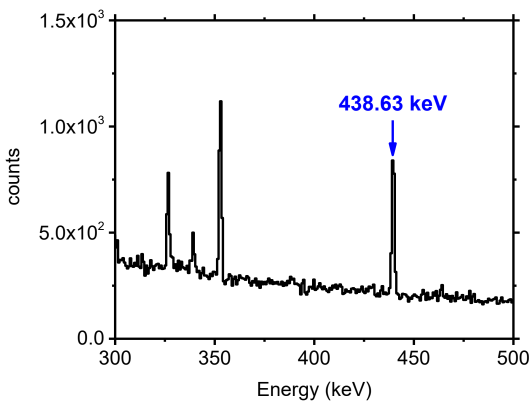

Figure 5. *The γ-ray spectrum obtained from the ^{73}Ge(n,nα)^{69m}Zn reaction*

SUMMARY & CONCLUSIONS

The cross sections of the ^{70}Ge(n,2n)^{69}Ge, ^{76}Ge(n,2n)^{75}Ge, ^{73}Ge(n,p)^{73}Ga, ^{72}Ge(n,p)^{72}Ga, ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga, ^{74}Ge(n,α)^{71m}Zn, ^{72}Ge(n,α)^{69m}Zn and ^{73}Ge(n,nα)^{69m}Zn reactions have been measured at neutron energies of 16.4, 17.9 and 18.9 MeV using highly enriched Ge targets that were provided by the CERN n_TOF collaboration [16]. The results of the present work are focused on the ^{73}Ge(n,d/np)^{72}Ga, ^{74}Ge(n,d/np)^{73}Ga and ^{73}Ge(n,nα)^{69m}Zn challenging reactions, providing cross section points in an energy range where very few datasets exist. Moreover, the use of highly enriched isotopes, yield more accurate experimental results, since no theoretical corrections for the subtraction of parasitic contributions in the measured yield needs to be performed. Most datasets in literature use ^{nat}Ge targets, where parasitic contributions from reactions of neighboring isotopes leading to the production of the same residual nucleus, cannot be avoided. The subtraction of these contributions is heavily based on theoretical calculations that bear their own uncertainties. These accurate cross section results in a plethora of reaction channels could act as a very important constraint in the optimization of input parameters in statistical model calculations, by the simultaneous reproduction of all the reaction channels. These parameters could then be used to describe the behavior of other medium-heavy nuclei in the same mass region. Consequently, despite their cost and difficulty to obtain, measurements with enriched targets in key isotopes (such as Ge) and in specific energies could vastly improve the accuracy and reliability of future evaluations. More experimental cross section measurements will be performed in the high energy region above 15 MeV, and the results of all the available reaction channels will be theoretically reproduced with the use of the EMPIRE-3.2 using the same set of input parameters.

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