

Annual Symposium of the Hellenic Nuclear Physics Society

Τόμ. 28 (2021)

HNPS2021



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doi: [10.12681/hnps.3621](https://doi.org/10.12681/hnps.3621)

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Βιβλιογραφική αναφορά:

Chasapoglou, S., Axiotis, M., Gkatis, G., Harissopulos, S., Kokkoris, M., Lagoyannis, A., Lederer-Woods, C., Michalopoulou, V., Savva, M. I., Stamatelatos, I. E., Stamatopoulos, A., Vasilopoulou, T., & Vlastou, R. (2022). Cross Section Measurements of (n,x) Reactions at 17.9 MeV Using Highly Enriched Ge Isotopes. *Annual Symposium of the Hellenic Nuclear Physics Society*, 28, 135–141. <https://doi.org/10.12681/hnps.3621>



Cross Section Measurements of (n,x) Reactions at 17.9 MeV Using Highly Enriched Ge Isotopes

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Abstract The $^{70}\text{Ge}(n,2n)^{69}\text{Ge}$, $^{72}\text{Ge}(n,a)^{69m}\text{Zn}$, $^{72}\text{Ge}(n,p)^{72}\text{Ga}$ and $^{73}\text{Ge}(n,p)^{73}\text{Ga}$ reactions have been measured by means of the activation technique at neutron energy 17.9 MeV. The quasimonoenergetic neutron beam was produced via the $^2\text{H}(d,n)^3\text{He}$ reaction at the 5.5 MV Tandem Van de Graaff accelerator of NCSR "Demokritos." Isotopically highly enriched targets of ^{70}Ge , ^{72}Ge and ^{73}Ge , provided by the nTOF collaboration at CERN, have been used, thus allowing accurate cross section measurements since no corrections are needed to compensate for the parasitic reactions from neighboring isotopes that exist in the case of using natural Ge target. The cross section has been deduced with respect to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction.

Keywords neutrons, activation method, enriched targets, cross section measurements

INTRODUCTION

The study of neutron induced reactions on Ge isotopes is very important both for practical applications and fundamental research. Practical applications include dosimetry, nuclear medicine, astrophysical projects and reactor technology [1–3]. Moreover, Ge as a semiconductor is widely used in γ -spectroscopy detectors. From a fundamental research point of view, (n,x) reactions on Ge isotopes produce residual nuclei in high spin isomeric states. The decay of the produced compound nucleus to isomeric and normal states is heavily dependent on the spins of the discrete levels involved, as well as the spin distribution of the continuum phase space [1,4,5]. In this sense, high accuracy experimental isomeric cross section results are of major importance for the study of the residual compound nucleus. In addition, input parameters in statistical model calculations can also be optimized, via the simultaneous reproduction of many competing reaction channels on different Ge isotopes. New experimental cross section data in the energy range above 15 MeV are very important, due to the fact that in this energy range pre-equilibrium effects in the de-excitation of the compound nucleus become important [1].

Cross sections of (n,x) reactions on ^{nat}Ge targets have been studied in the past by the Nuclear Physics Group of the National Technical University of Athens [6, 7], implementing the $^2\text{H}(d,n)^3\text{He}$ reaction in the energy range $E_n=8-11.4$ MeV. There are only a few existing

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datasets found in literature [8] above 15 MeV with significant discrepancies among them. These discrepancies may stem from the inevitable corrections for parasitic reactions that lead to the production of the same residual nucleus as the measured reaction, in the case of the use of ^{nat}Ge targets. These corrections, that bear their own uncertainties, are not needed in the case of highly enriched targets. For this reason, highly enriched targets that have become available from CERN have been used in this work for the experimental measurement of (n,x) reaction cross sections on Ge isotopes at incident neutron energy $E_n=17.9$ MeV.

EXPERIMENTAL DETAILS

Targets

Three highly enriched Ge pellets provided by the n_TOF collaboration of CERN have been used in the frame of this work, with different enrichment levels. More specifically, the ^{70}Ge , ^{72}Ge and ^{73}Ge targets, were provided in the form of GeO_2 with enrichment levels of 97.71%, 96.59% and 96.07%, respectively. Each pellet was glued on a thin mylar foil, which was in turn glued on an Al ring. Each Ge target was placed between two high purity Al foils, to calculate the neutron flux on the Ge target via the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction. A more detailed presentation of the physical and geometrical properties of the reference foils and the targets is presented in Table 1.

Table 1 Physical and geometrical properties of the highly enriched Ge pellets along with the Al reference foils used in this work. The masses of the Ge targets refer to the mass of the isotope present in the respective pellet.

Isotope	Enrichment (%)	Form	Diameter (mm)	Mass (g)
^{70}Ge	97.71	GeO_2	20.00 ± 0.01	1.81
^{72}Ge	96.59	GeO_2	20.00 ± 0.01	1.79
^{73}Ge	96.07	GeO_2	20.00 ± 0.01	1.79
^{27}Al	99.90	metal	20.00 ± 0.01	0.41

Neutron Beam Facility And Irradiations

The measurements were carried out at the 5.5 MV Tandem Van de Graaf accelerator [9, 10] of the NCSR “Demokritos” at Athens, Greece. The quasi mono-energetic beam was produced via the $^3\text{H}(d,n)^4\text{He}$ reaction at $E_n=17.9 \pm 0.3$ MeV. A 2.9 MeV deuteron beam impinged on a solid Ti tritiated target (TiT) after passing a 10 μm Molybdenum foil, losing part of its energy. At the end of the TiT target, a Cu backing acted as a beam stop for the incident deuteron beam. The TiT target was kept in high vacuum, fixed in a stainless steel flange, and was also air cooled during the irradiations, to avoid any possible thermal damage caused by the impinging deuteron beam.

In each performed irradiation, the Ge targets were placed between two reference Al foils, that were used to obtain the flux via the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction. The samples were placed at a distance of ~ 2.7 cm from the TiT target for each irradiation, to ensure that the neutron beam would be monoenergetic within the angular acceptance of the first target, while the fluctuations of the beam were monitored via a BF_3 detector placed at a ~ 3 m distance from the neutron producing target. This detector was placed at an angle relative to the neutron beam

in order to minimize the scattering of the neutron beam back to the target assembly. Following the irradiations, the induced radioactivity of the Ge targets and reference foils was measured with four HPGe detectors of relative efficiencies between 40% and 80%.

DATA ANALYSIS

Cross Section Calculation And Uncertainties

The cross section was calculated via the activation technique using the equation (1).

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

The subscripts “tar” and “ref” refer to the Ge targets and Al reference foils respectively. The factor:

- σ is the cross section in barns. The σ_{ref} factor represents the cross section of the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction, obtained from the ENDF/B-VIII.0 evaluation library [11]
- N_{γ} is the integral of the γ -ray peak from the experimental spectrum. All spectra were analyzed with the use of the “Tv” software [12]
- ε is the absolute efficiency of the HPGe detector in a specific γ -ray energy
- F is a factor to correct for the γ -ray self absorption within the sample
- I_{γ} is the intensity of each γ -ray
- D is a correction factor for the de-excitation of the nuclei during the measurement in the HPGe detector (t_1) and the cooling time (t_2). It is given by the expression $D = \exp(-\lambda t_1) - \exp(-\lambda t_2)$
- f_c is a correction factor for the decaying nuclei during the irradiation time (t_{irr}), that is divided in small time intervals (dt). This factor is calculated by the expression $f_c = \frac{\int_0^{t_{irr}} e^{\lambda t} f(t) dt}{\int_0^{t_{irr}} f(t) dt} e^{-\lambda t_{irr}}$, where $f(t)$ are the counts obtained from the BF3 detector in arbitrary units.
- N_t is the total number of nuclei in the target
- Φ_{ref}/Φ_{tar} is the neutron flux ratio between the reference foil and the Ge target. This ratio was calculated via the MCNP6 Monte Carlo code [13]

All factors in equation (1) were considered uncorrelated and the uncertainties in the final cross section result were calculated simply as a quadratic summation of the uncertainties of all the factors. In the case however, when two or three γ -rays were used for the calculation of the cross section, equation (27) reported in [14] and the formalism presented in Appendix 2 of [15] was implemented respectively. In both cases, the correlation between all the factors of Eq. (1) was taken into account.

Monte Carlo Simulations

The MCNP6 [13] Monte Carlo code was used, in order to calculate the propagation of the neutron beam through the consecutive reference foils and targets. Firstly, the experimental neutron flux was calculated at the position of the reference Al foil via the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction since its cross section is well known in literature [11]. Then, Monte Carlo simulations were performed, with a detailed description of the neutron producing target, the target-foil assembly and the target holder. The result of the simulated neutron flux is presented in Fig. 1. Apart from the main peak of the neutron beam, a tail of low energy parasitic neutrons also exists, with intensity lower by 2-3 orders of magnitude compared to the main beam.

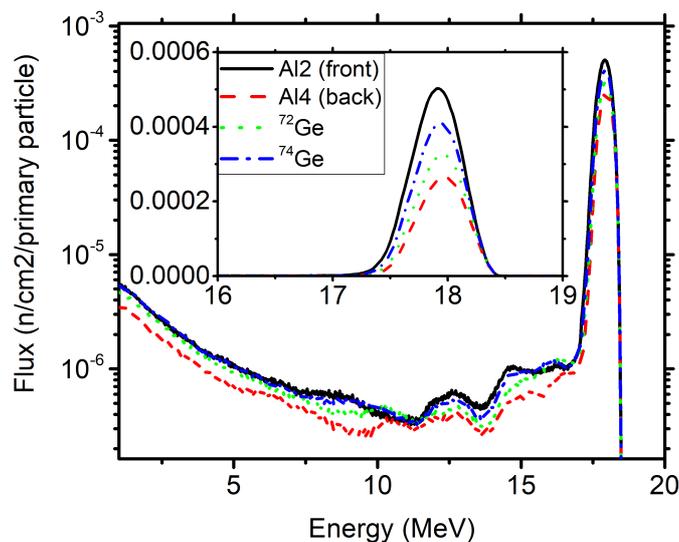


Fig. 1. Typical flux as an MCNP6 output for one of the target assemblies of the experiment. The two Ge targets were placed between two Al foils.

The factor Φ_{ref}/Φ_{tar} of Eq. (1) is calculated from the ratio of the integrals of the main peaks of the flux for the reference foil and the target as shown in the inset of Fig. 1.

The contribution of the tail of low energy parasitic neutrons is very small due to the fact that every measured and reference reaction has a threshold in the MeV region, and is inserted in the analysis as a systematic error that does not exceed the value of 3.5% for every reaction.

RESULTS AND DISCUSSION

In this section the experimental results for the cross section of the $^{70}\text{Ge}(n,2n)^{69}\text{Ge}$, $^{72}\text{Ge}(n,\alpha)^{69\text{m}}\text{Zn}$, $^{72}\text{Ge}(n,p)^{72}\text{Ga}$ and $^{73}\text{Ge}(n,p)^{73}\text{Ga}$ reactions are presented in Fig. 2 along with the experimental datasets found in literature [8] and existing evaluation curves [11, 16–19].

The cross section of the $^{70}\text{Ge}(n,2n)^{69}\text{Ge}$ reaction that was calculated from the weighted average of the 574 and 871.98 keV lines is presented in Fig. 2(a). The two different general trends of the evaluations can be explained due to the large errors of the intensities of the different γ -rays from the ^{69}Ge residual nucleus that reach up to 13% [20].

The cross section of the $^{73}\text{Ge}(n,p)^{73}\text{Ga}$ reaction was calculated from the 297.32 keV line. As presented in Fig. 2(b), the result of the present work seem to be in fair agreement with the dataset of Hoang et al. and JENDL-4.0 and ENDF/B-VIII.0 evaluation curves.

The cross section of the $^{72}\text{Ge}(n,p)^{72}\text{Ga}$ reaction was obtained from the weighted average of the 834.13, 629.97 and 894.33 keV lines. It is presented in Fig. 2(c) along with the existing datasets and evaluation curves found in literature. The result of the present work seem to be in agreement with the general trend of the dataset of Konno et al, that also used a highly enriched target (97.8% enrichment), and the JENDL-4.0 and ENDF/B-VIII.0 libraries.

Finally, for the $^{76}\text{Ge}(n,a)^{69m}\text{Zn}$ reaction, the experimental cross section results are presented in Fig. 2(d), and seem to be in good agreement with the general trend of the only existing dataset in this energy region of Hoang et al.

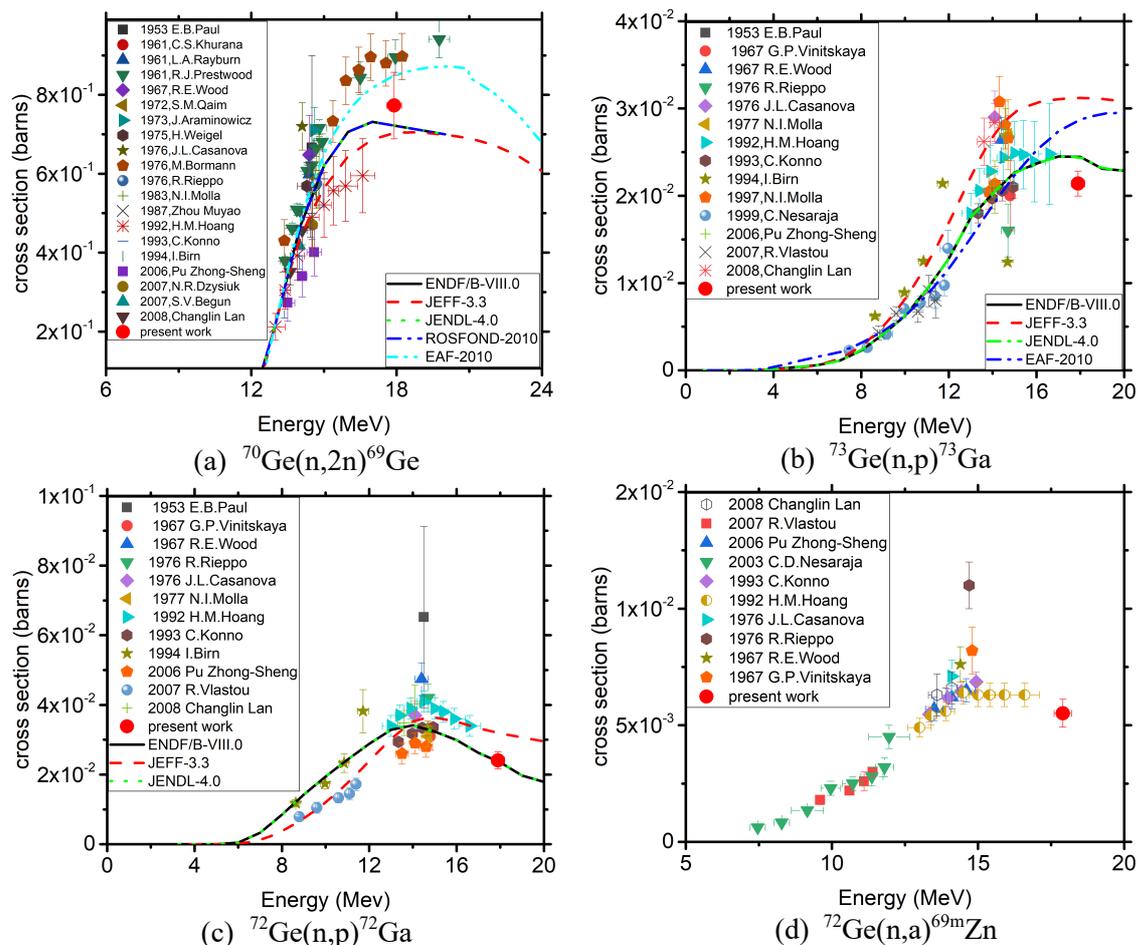


Fig. 2. The experimental cross section results for the different reactions studied in this work implementing highly enriched Ge targets

CONCLUSIONS

The activation technique was used for the measurement of the cross sections of the $^{70}\text{Ge}(n,2n)^{69}\text{Ge}$, $^{73}\text{Ge}(n,p)^{73}\text{Ga}$, $^{72}\text{Ge}(n,p)^{72}\text{Ga}$ and $^{72}\text{Ge}(n,a)^{69m}\text{Zn}$ reactions at incident neutron beam energy of 17.9 MeV using the $^3\text{H}(d,n)^4\text{He}$ neutron producing reaction. In this energy region, only a few, and many times discrepant data exist that most commonly use $^{\text{nat}}\text{Ge}$ targets. These discrepancies could be explained due to the inevitable theoretical corrections that need to be applied in this case, to compensate for the contribution of interfering reactions stemming from neighbouring isotopes in the $^{\text{nat}}\text{Ge}$ target.

The targets that were used in this work were highly enriched ones, and yielded accurate cross section results, since no theoretical corrections, bearing their own uncertainties, are needed. These accurate cross section results could act as a constraint to input parameters in statistical model calculations via the simultaneous reproduction of many competing reaction channels with the same set of parameters. They could also improve the accuracy of the existing evaluation curves in the energy region above 15 MeV.

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