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# 14.8 MeV Neutron Activation Cross Section Measurements for Ge Isotopes

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**Abstract**

The cross sections of Ge isotopes were measured with the activation method at 14.8 MeV neutron energy. The quasi-monoenergetic neutron beams were produced via the $^{3}H(d,n)^{4}He$ reaction at the 150 kV J-25 neutron generator of INST, AERE. The characteristics $\gamma$-lines of the product nuclei were measured with a closed end coaxial 17.5 $cm^2$ high purity germanium (HPGe) detector gamma ray spectroscopy. The cross sections were determined with reference to the known $^{27}Al(n,\alpha)^{24}Na$ reaction. Cross section data are presented for $^{72}Ge(n,p)^{72}Ga$, $^{74}Ge(n,\alpha)^{71m}Zn$ and $^{76}Ge(n,2n)^{75m+g}Ge$ reactions. The cross section values obtained for the above reactions were 24.78±1.75 mb, 1.69±0.11 mb and 860±50 mb, respectively. The results obtained were compared with the values reported in literature as well as theoretical calculation performed by the statistical code SINCROS-II. The experimental data were found fairly in good agreement with the calculated and literature data.

*Keywords*: Activation cross section; Neutron induced reaction; Gamma-ray spectroscopy; 14.8 MeV.



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## 1. Introduction

Studies of neutron induced reactions are of considerable significance, both for importance to fundamental research in Nuclear Physics and Astrophysics, and for practical applications in nuclear technology, medicine and industry [1, 2]. The neutron cross section data around 14 MeV is of paramount importance from the view point of fusion reactor technology, especially for the calculation of nuclear transmutation rates, nuclear heating and radiation damage to the materials used in the construction of the core and inner walls of the reactor. Though many measurements of neutron activation cross sections at ~14 MeV were reported in the literature up to now, the new data are still required as documented by the WRENDA 83/74 [3]. There are many reasons for these new

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requirements. The most important one, we think, is the fact that the reported cross sections often exhibit large mutual disagreement greatly exceeding quoted uncertainties [4, 5].

The Neutron Activation Analysis (NAA) [6-8] method was employed in most of the previous cases to measure the reaction cross sections, and data were taken with GM counters for $\beta$-rays and/or NaI(T1) scintillation detectors for $\gamma$-rays. The sources of uncertainty in data of the activation method are considered to be interfering reactions caused by impurities in the samples, uncertainty in the nuclear decay data, efficiency of the separation technique, absolute neutron flux and its variation in time, energy and energy spread of neutrons, and cross section of the monitor reaction. A high resolution HPGe (Canberra) detector was employed as a $\gamma$-ray detector in the present work. HPGe detector is capable to make the detection of $\gamma$-rays from products of the aimed reaction selectively, and can distinguish them from the interfering $\gamma$-rays. This detector, thereby, can reduce the uncertainty due to the interfering reaction, and give accurate amount of reaction products to be measured.

The investigation of fast neutron interaction with atomic nuclei of Ge yields important information to the study nuclear forces, nuclear structure and reaction, and provides a good way of testing the application of nuclear models. Therefore, it is of great importance to measure the cross sections of Ge isotopes induced by neutrons around 14 MeV. A few experimental data for Ge have been reported [5, 9-15], but a survey of available cross section data showed that reported measurement cover a wide energy range but they also show many discrepancies.

In this paper we present our measurements of the neutron activation cross sections for $^{72}$Ge($n$,$p$)$^{72}$Ga, $^{74}$Ge($n$,$\alpha$)$^{71m}$Zn and $^{76}$Ge($n$,2$n$)$^{75m+g}$Ge reactions at 14.8 MeV relative to the $^{27}$Al($n$,$\alpha$)$^{24}$Na reference reaction via the activation method. The cross sections of other three reactions $^{73}$Ge($n$,$p$)$^{73}$Ga, $^{72}$Ge($n$,$\alpha$)$^{69m}$Zn and $^{70}$Ge($n$,2$n$)$^{69}$Ge are reported elsewhere [16]. The comparison of our measured data with the theoretical calculation can improve the systematic development of the model parameters.

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## 2. Experiment

Cross sections were measured via the activation technique and identification of the radioactive products. This technique is ideally suited for investigating low-yield ($n$,$p$) and ($n$,$\alpha$) reaction products, and is almost ideal for studying closely spaced low-lying isomeric states, provided their lifetimes are not too short. The details have been described over the years in several publications [17-21]. Here, we give only some salient features relevant to the present measurements.

### *2.1. Sample irradiation*

For the reactions on Ge isotopes, sample of very high purity (>99.9%) natural Ge in the form of a tablet was used. To prepare the sample, about 0.3 g of Ge powder was pressed at 6 ton/cm$^2$ using a hydraulic press, and a tablet (1.3 cm diameter, 0.05 cm thick) was

obtained. After weighing, the tablet was sealed into a thin polythene bag in order to prevent the air moisture. Two monitor foils (Al, each 200 μm thick) of the same size as the tablet were then attached in the front and at the back of the sample. The foils were initially cleaned carefully with acetone.

Semi-monoenergetic neutrons of energy 14.8±0.34 MeV were generated via the $^{3}H(d,n)^{4}He$ reaction (Q = 17.6 MeV) with the 110 keV deuteron beam delivered from the J-25 neutron generator at the INST, AERE, Dhaka, Bangladesh. J-25 neutron generator is a 150 kV horizontal type positive accelerator manufactured by AID, MEYLAN, FRANCH, and supplied by International Atomic Energy Agency (IAEA) under the technical co-ordinate research program. The acceleration tube of this generator is made of 10 conic electrodes, and an equal division of the high voltage (110 kV) is performed for all the tubes. As a result, the beam receives a kick of 11 kV in passing each electrode, and finally a continuous deuteron beam of 110 keV is delivered. The samples were placed in the $0^{0}$ direction relative to the incident deuteron beam, at a distance of 1 cm from the beam stop. The diameter of the tritium target was 50 mm with active diameter of 30 mm having activity of 10 Ci. The target assembly was so designed that neutron scattering by the assembly itself is reduced as possible. The deuteron beam intensity was approximately 125 μA and the neutron yield was about $2 \times 10^{11}$ *n*/s at the target. Considering the half-life of and gamma ray intensity in the product nuclei, the irradiation time was selected as 2 h. The decay data of the product nuclei are listed in Table 1.

Table 1. Decay data of measured reaction products [22].

| Reaction product | Half-life | Decay mode | Gamma energy $E_\gamma$ (keV) | Isotopic abundance (%) | Gamma intensity $I_\gamma$ (%) |
|---|---|---|---|---|---|
| $^{72}$Ga | 14.1 h | $\beta^-$ | 834.03 | 27.5 | 95.6 |
| $^{71m}$Zn | 3.96 h | $\beta^-$ | 386.28 | 36.4 | 93.0 |
| $^{75m+g}$Ge | 82.78 min | $\beta^-$ | 264.60 | 7.7 | 11.4 |

The neutron flux at the sample position was determined via the monitor reaction $^{27}Al(n,\alpha)^{24}Na$ ($T_{1/2}$ = 14.97 h; $E_\gamma$ = 1369 keV; $I_\gamma$ = 100%). During irradiation, the fluctuation of neutron yield was monitored continuously by counting the associated α-particles with the help of a $BF_3$ long counter. For ascertaining the constancy of the neutron flux, a constant check of the pressure in the acceleration tube and the deuteron beam current on the target was performed. The cross section of the monitor reaction was taken from the IRDP computer file [23]. The flux densities were calculated after correction of monitor product activities from background neutrons. The average flux density on the sample was then obtained by taking the mean value of the calculated flux density for the front and back foils. The average neutron flux was determined to be about $10^{8}$ *n*/cm$^2$/sec.

### *2.2. Gamma-ray spectroscopy and cross section measurement*

After irradiation, the samples were removed from the generator room to nearby gamma spectrometry laboratory for counting. The $\gamma$-ray spectra of product radionuclides in both the target and reference foils were measured with a 17.5 $cm^2$ closed end coaxial HPGe detector associated with the pre-amplifier, amplifier, analogue-to-digital converter (ADC) and multi channel analyzer (MCA). Absolute gamma ray counting was made as precise as possible by selecting the live time for the detector, and taking the count rate after subtracting the corresponding background. The detector was properly shielded by lead blocks in order to reduce the contribution of the natural radioactivity. The samples were placed on the surface of the detector, and spectra were taken for different cooling time to get a good result. Fig. 1 shows a schematic drawing of the measuring geometry.

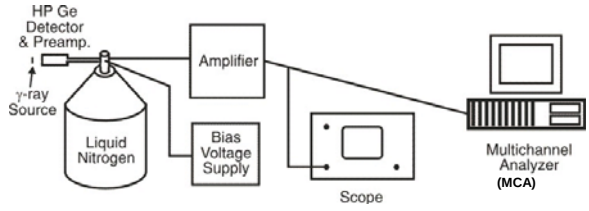


Fig.1. A schematic drawing of the measuring geometry (here gamma-ray spectrometry system includes HPGe detector, pre-amplifier, amplifier, ADC and MCA).

Three important parameters: energy resolution of the detector, energy calibration of MCA and efficiency of the detector were studied using standard gamma-ray sources of $^{60}$Co, $^{57}$Co, $^{22}$Na, $^{133}$Ba and $^{137}$Cs. The energy resolution obtained in this experiment is 1.96 keV FWHM at 1332.51 keV gamma line of $^{60}$C0 point source. The lower value indicates the good resolution of the detector.

The activation cross section was deduced using the following standard activation equation:

$$A = N\sigma\varphi(1-e^{-\lambda t_i}) \tag{1}$$

where, $A$ be the disintegration rate of the reaction product at the end of irradiation, $N$ the total number of atoms in the sample, $\sigma$ the cross section of the reaction to be measured, $\varphi$ the neutron flux, $\lambda = 0.693/T_{1/2}$ the disintegration constant, $T_{1/2}$ the half-life of the reaction product, and $t_i$ the irradiation time. The sources and typical values of the experimental errors are summarized in [16]. The dominant error sources were $\gamma$-ray detector efficiency, counting statistics and the standard cross section for the neutron flux monitor.

## 3. Nuclear Model Calculations

Theoretical calculation of activation cross section for neutron induced reaction has a great importance in nuclear data evaluation. A large number of theoretical codes are considered

for evaluation of activation cross sections. The codes are based on the different theories, approximations and methods of numerical calculations. In the present work, the computer code SINCROS-II [24], which employs the deterministic method, was used to calculate the activation cross sections in the energy region of 13-15 MeV. The SINCROS-II consists of the code ELIESE [25]-GNASH [26] joint program EGNASH2, and simplified input version of DWUCKY [27]. The codes ELIESE and GNASH were joined and the global optical potentials were built into the new code EGNASH, whose input format became simpler than that of GNASH for efficient execution of nuclear cross section calculations.

The key points of the cross section calculation are the determination of level density parameters of daughter nuclei and of the rate of contribution of pre-equilibrium and direct processes to the statistical process. In the code EGNASH2, the pre-equilibrium and direct processes of particle emission are located with the code PRECO developed by Kalbach [28], which is coupled with GNASH and with the code DWUCKY for the inelastic scattering. In the GNASH, free parameters were the single particle state density and the normalization factor excision model. In contrast with this, the single particle state density constant is not free, but dependent upon the level density parameter '*a*' by the relation, $g = (6/\Pi^2)a$. After the parameters for the pre-equilibrium and direct processes were selected, the free parameter to be determined is only the level density parameter '*a*' in the Fermi gas model, because the nuclear temperature used in the constant temperature model can be determined automatically or by the following equation;

$$\rho(E,J) = \rho(E_x)\exp.[(E-E_x)/T]R(J,E) \qquad E < E_x \tag{2}$$

where, $\rho(E_x)$ is the energy term of level density, $\rho(E, J)$ is the spin dependent level density, $R(E, J)$ is the spin term, and $E_x$ is the energy at which both densities are smoothly connected.

Since the total emission of various kinds of particles from the compound nucleus is controlled by a set of level density parameters for daughter nuclei, it is better that the level density parameter of each daughter nucleus is so practically determined that the calculated cross sections for reactions, $(n,p)$, $(n,\alpha)$ and $(n,2n)$ agree with the experimental data of respective reactions. The first step to fix the level density parameters is the calculation of them using the experimental value of mean level spacing for S-wave neutron resonance at the neutron binding energy, $E_B$. The calculation was made by inversely solving the following equation with the spin cutoff factor and the pairing correction quoted by Gilbert and Cameron [29].

$$D_0 = C_0(E_B - \Delta)/(2I+1)\exp.\{[3+(2I+1)^2]/8\sigma^2(E_B) - 2\sqrt{a(E_B-\Delta)}\} \tag{3}$$

where, $D_0$ be the average spacing of S-wave, $I$ the spin of target nucleus, $\sigma^2$ the spin cutoff factor, and $\Delta$ the pairing correction. The next step to determine the level density is the cross section calculation in the mass region where the reliable experimental data exist.

The code EGNASH2 has a few parameters for the equilibrium process, level density and radiative width. Several test calculations of the cross sections in the energy range 13-15 MeV were carried out referring to the experimental data for ($n$,$p$), ($n$,$\alpha$) and ($n$,2$n$) reactions of Ge sample. The cross sections of $^{72}$Ge($n$,$p$)$^{72}$Ga and $^{74}$Ge($n$,$\alpha$)$^{71m}$Zn and $^{76}$Ge($n$,2$n$)$^{75m+g}$Ge reactions depend on the level density parameter and radiative width of $^{72}$Ge, $^{74}$Ge and $^{76}$Ge isotopes, respectively as the target nuclei of the reactions. For comparison the results of calculation and the present experiments along with the literature data are shown in Fig. 2. However, the cross section for the reaction $^{76}$Ge($n$,2$n$)$^{75m+g}$Ge could not be calculated in the present works.

## 3. Results and Discussion

The neutron activation cross sections for $^{72}$Ge($n$,$p$)$^{72}$Ga and $^{74}$Ge($n$,$\alpha$)$^{71m}$Zn and $^{76}$Ge($n$,2$n$)$^{75m+g}$Ge reactions at 14.8 MeV neutron energy measured in the present work are summarized in Table 3 along with the experimental errors. The cross section values listed in the Table are based on three independent measurements. The uncertainties quoted in the values represent the statistical and systematic errors.

Table 3. Measured cross section at neutron energy of 14.8 MeV.

| Reaction | Gamma energy (keV) | Neutron flux, $\varphi$ (n/cm$^2$/sec) | Cross section (mb) |
|---|---|---|---|
| $^{72}$Ge($n$,$p$)$^{72}$Ga | 834.03 | | 24.78±1.8 |
| $^{74}$Ge($n$,$\alpha$)$^{71m}$Zn | 386.28 | $9.20\times10^7$ | 1.69±0.11 |
| $^{76}$Ge($n$,2$n$)$^{75m+g}$Ge | 264.60 | | 859±50 |

The theoretical cross sections as a function of neutron energy were reproduced by the computer code SINCROS-II. Fig. 2 shows the cross section data for the above reactions measured in the present experiments, along with corresponding values from the literature and results from theoretical calculation. A short discussion concerning each reaction will follow.

### 4.1. *$^{72}$Ge(n,p)$^{72}$Ga reaction*

The activation cross section of this reaction was measured by means of 834.03 keV $\gamma$-transitions. The value measured in the present work at 14.8 MeV is 24.78±1.75 mb. The cross section for this reaction reported by Afia Begum [14] is ~30 mb at 14.7 MeV, and that reported by Rehezuddin Miah [15] is ~38 mb at 14.1 MeV. Our results are in good agreement with the above reported data within the quoted uncertainties. The same holds for the calculated value ~25.53 mb using SINCROS-II at 14.8 MeV. We also compared the present data with the ($N$-$Z$)/$A$ dependence of the ($n$,$p$) reactions. ($N$-$Z$)/$A$ dependence

of cross sections of the ($n$,$p$) reactions for the interval $12 \leq A \leq 150$ is presented by Levkovskii [30] as follows:

$$\sigma(n,p) = 45.2(A^{1/3}+1)^2 e^{-33(N-Z)/A} \quad \text{(mb)} \tag{4}$$

where, $A$, $Z$ and $N$ represent the mass number, atomic number and neutron number of the target nuclei, respectively. The cross section for the $^{72}$Ge($n$,$p$)$^{72}$Ga reaction predicted by Eq**. (**4) becomes 30.76 mb, which is very near to our measured value.

### 4.2. *$^{74}$Ge(n,α)$^{71m}$Zn reaction*

Among all isotopes comprising natural Ge, only the $^{72,74}$Ge($n$,$\alpha$)$^{69,71m}$Zn and $^{76}$Ge($n$,$2n$)$^{75m+g}$Ge reactions lead to metastable states which can be measured via the activation technique. The $^{74}$Ge($n$,$\alpha$)$^{71}$Zn reaction leads to the formation of the unstable nucleus $^{71}$Zn both in its $1/2^-$ ground state ($T_{1/2}$ = 2.45 m) and its metastable $9/2^+$ ($T_{1/2}$ = 3.96 h) state. Due to short lifetime of the ground state of $^{71}$Zn, only the activity of the metastable state could be measured via the activation technique. The cross section of metastable state, $\sigma_m$, was determined by analyzing the 386.28 keV transition of $^{71}$Ga which is fed by the deexcitation of the metastable state of $^{71}$Zn.

This reaction was rarely investigated in the past. The $\sigma_m$ measured in the present experiment at 14.8 MeV is 1.69±0.11 mb. Our value is in striking disagreement with the only recent result reported by Miah [15]: ~3.1 mb at 14.71 MeV neutron energy. Though the same decay data were used in both cases, the results differ by a factor of ~2. The calculated value is, however fairly in good agreement with our measured value. The cross section at same neutron energy calculated by SINCROS-II is ~2.15 mb, which is only ~27% higher than the measured value.

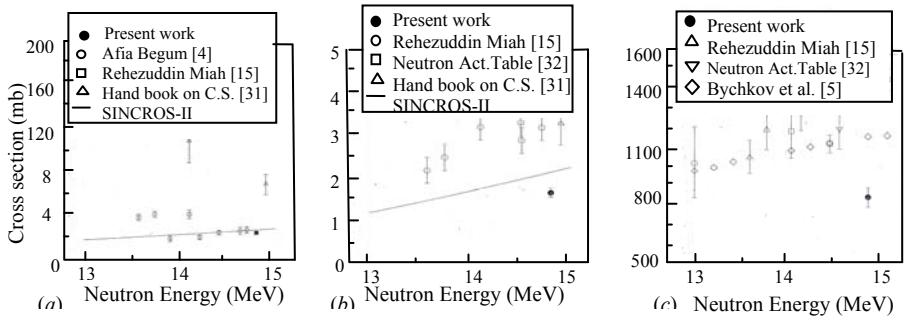


Fig. 2. Cross section data of (*a*) $^{72}$Ge($n$,$p$)$^{72}$Ga, (*b*) $^{74}$Ge($n$,$\alpha$)$^{71m}$Zn and (*c*) $^{76}$Ge($n$,$2n$)$^{75m+g}$Ge reactions.

### 4.3. *$^{76}$Ge(n,2n)$^{75m+g}$Ge reaction*

The high threshold $^{76}$Ge($n$,$2n$)$^{75}$Ge reaction leads to the formation of the unstable nucleus $^{75}$Ge both in its $1/2^-$ ground state ($T_{1/2}$ = 82.78 m) and its metastable $7/2^+$ ($T_{1/2}$ = 47.7 s)

state. The deexcitation of the metastable to the ground state in fraction 99.97% along with its short lifetime lead to the measurement of the total cross section $\sigma_{m+g}$ of the $(n,2n)$ reaction via the 264.60 keV characteristic transition of $^{75}$As.

This reaction was investigated by Bychkov *et al*. [4] and Miah [15]. Their reported values of ~1130 mb at 14.4 MeV and ~1280 mb at 14.2 MeV, respectively, are again in striking disagreement with our result of 859±50 mb at 14.8 MeV neutron energy. We have no reasonable explanation for the observed differences. We note that the theoretical calculation of this reaction could not be performed in the present works. We, therefore, suggest for further measurement of the cross section for this reaction.

## 5. Conclusions

In the present study, neutron activation cross sections of $^{72}$Ge$(n,p)^{72}$Ga, $^{74}$Ge$(n,\alpha)^{71m}$Zn and $^{76}$Ge$(n,2n)^{75m+g}$Ge reactions at 14.8 MeV were measured using HPGe γ-spectroscopy of the reaction products. The measured cross section for the $(n, p)$ reaction agrees with the reported values. There are, however, large discrepancies between present measurement and reported data for $(n,\alpha)$ and $(n,2n)$ reactions. The main sources of discrepancy in the experimental data are attributed to the differences of the experimental conditions (the neutron field characterization, radiation measuring technique, and neutron monitor method), standard cross section data and nuclear data (gamma-ray branching ratio, half-life, and natural abundance). Although in our case all the cross sections were measured in the same laboratory, for the case of second and third reactions gamma-ray energy, gamma-ray branching ratio and detection efficiency were different, also the measurement were taken at different sessions. All these factors may be responsible for the observed discrepancies between the present results and the previous data. These are, however, not sufficiently conclusive and, clearly, still new measurements are needed to improve the situation.

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