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Production Cross Sections and Induced Activity in Ge Isotopes by 30 MeV Proton Beam

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The excitation functions of ^{70}Ge(p,n)^{70}As,^{72}Ge(p,n)^{72}As, ^{74}Ge(p,n)^{74}As and ^{76}Ge(p,n)^{76}As reactions were studied from reaction threshold to 30 MeV by using EMPIRE-3.2 and TALYS-1.9 nuclear reaction model codes. This study is important because some isotopes produced are important for positron emission tomography (PET). Direct, pre-compound and compound nuclear reactions are considered as main nuclear reaction mechanisms in the codes. The calculated excitation functions have been compared with available experimental data and found to be in fair agreement. Furthermore, the contributions of various reaction mechanisms have been studied in total reaction cross-section that varies with the incident proton energy. The estimation of induced radio activity in thick Ge target due to the primary interaction is carried out for1μA, 30 MeV proton beam.

Keywords: Excitation functions, PCROSS, Hybrid Monte Carlo Simulation, Induced Activity, Pre-equilibrium

1 Introduction

Arsenic, which has only one stable isotope, *i.e.*, ^{75}As is used as a doping agent in semiconductors. The radioisotopes of Arsenic, ^{70,72,74,77}As, find use in different fields of nuclear medicine.^{72,74}As can be successfully employed in Positron Emission Tomography (PET) to get ultra clean images of small tumours while ^{77}As being a β emitter can be used for therapy.^{72}As in combination with ^{77}As finds potential application in the ranostics (therapy combined with online diagnosis).These isotopes can be produced through direct or secondary reaction in nuclear reactor or accelerator[1,2]. In order to plan efficient application of these isotopes in diagnosis, therapy it is essential to estimate the induced activity of the isotopes formed through nuclear reaction[3,4]. Induced activity of a radioisotope is determined from the cross section of the relevant nuclear reaction[5-6]. In the nuclear database available till date experimental data are not plenty in the energy range upto 30 MeV, important for production of ^{72,74,77} As through proton induced reaction on Ge. Secondly, safe handling of the radioisotopes also mandates to determine the cooling time after irradiation of the target. There also exist some discrepancies among the different measurements[7-8]. In present work, we have calculated formation cross-sections of ^{70,72,74,77} As from proton induced reaction on Ge target in the energy range of 1-30 MeV using the nuclear reaction model codes TALYS-1.9 and EMPIRE-3.2. The calculated cross-sections are compared with the available experimentally measured data. The induced radioactivity from the four isotopes of As produced in a thick Ge target by the interaction of 30 MeV, 1 μA proton beam has been determined.

2 Method of calculation

2.1 EMPIRE-3.2

EMPIRE-3.2 is a well-known reaction model code[9] used to calculate pre-equilibrium (PEQ) and equilibrium (EQ) reaction cross sections. We have estimated excitation functions of different radioisotopes of As formed in the thick Ge target by the incident proton beam using EMPIRE-3.2. In calculating the EQ cross sections, the generalized superfluid model and EMPIRE specific level density have been chosen. Total yield of the product isotope and its activity are determined from the calculated excitation function. Attenuation of the projectile flux and the energy at different depths inside the thick target has been calculated using the computer package SRIM[10]. The total yield and activity of a radioisotope

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are determined from the weighted sum of the yield calculated from the production cross-sections at varying projectile energies at different depths in the target[11].The following options are used to carry these calculations:

(a) Empire specific level densities (EGSM) have been used along with Pre-equilibrium model PCROSS, Hybrid Monte Carlo Simulation (HMS) and Multi Step Compound (MSC) options as:

(i) EMPIRE1 (LEVDEN=0) PCROSS
(ii)EMPIRE2 (LEVDEN=0) PCROSS+HMS
(iii) EMPIRE3 (LEVDEN=0) MSC

(b) Generalized Super Fluid Model level densities (GSM) have been used along with Pre-equilibrium model PCROSS, Hybrid Monte Carlo Simulation (HMS) and Multi Step Compound (MSC) options as:

(i) EMPIRE4 (LEVDEN=1) PCROSS
(ii)EMPIRE5 (LEVDEN=1) PCROSS+HMS
(iii) EMPIRE6 (LEVDEN=1) MSC

2.2 TALYS-1.9

In TALYS[12] code direct reactions are computed using giant resonances. The estimation of PEQ particle emission is done by using two component exciton model and angular distribution is calculated using Kalbach systematics. Hauser-Feshbach formalism is used to estimate compound nuclear emission. All the optical model parameters are estimated using ECIS-06 code. Multiple PEQ and EQ emission of the ejectiles are considered. Any one of the five different level density formalisms, namely, Fermi gas model, constant temperature model, back-shifted Fermi gas model, generalised superfluid model and microscopic level densities can be used. In the present work we have used the Fermi gas model.

2.3Induced activity calculation

Induced activity is normally calculated for a thick target. In this thick target proton loses energy as it travels through the target. The energy loss characteristic of the proton is expressed as the stopping power (S). The projectile interacts with the target at each degrading energy with different probability or cross section. The total induced activity in the thick target is the sum of the induced activity at all these energies. Let us now consider that a target ‘X’ is irradiated for a time‘t’ and a radioisotope ‘Y’ is produced.

Activity ‘A’ due to Y after time t is given by:

$$\mathrm{A} = \sum_{\mathrm{i}} \mathrm{A} = \sum_{\mathrm{i}} \mathrm{N_X}^{\mathrm{i}} \sigma_{\mathrm{i}}\, \Phi\left[1 - \mathrm{e}^{-\lambda_{\mathrm{y}} \mathrm{t}}\right] \qquad \text{...(1)}$$

Where,

σ_i = cross section for production of Y at proton energy E_P^i

Φ = proton fluence rate

$\mathrm{N_X}^{\mathrm{i}}$ = number of target atoms available for reaction when proton loses an energy ΔE_P^i MeV.

So with a beam current of one micro ampere the activity A becomes

$$\mathrm{A} = \left(\frac{6.023}{\mathrm{A_T}}\right) \times \left(\frac{\Delta \mathrm{E_P}}{\mathrm{S}}\right) \times \left(\frac{\sigma}{1.602}\right) \times \left(1 - \mathrm{e}^{-\lambda_{\mathrm{Y}} \mathrm{t}}\right)\mathrm{Bq} \qquad \text{... (2)}$$

The energy of beam for present calculations is considered upto 30 MeV and ΔE_P is taken equal to 2 MeV[4, 11].

3 Results and Discussion

We have computed excitation functions for (p,n) reactions on stable isotopes of Ge. Build-up of induced activity due to the product isotopes has also been estimatedfor 1 µA beam current and irradiation periods of 1-120 hours. The reaction model code TALYS 1.9 and EMPIRE3.2 have been used to calculate the production cross-sections. We have considered production of the isotope in its ground state only. The half-life, the radionuclide formed and the induced activity for specified irradiation time are shown in table 1.

Excitation function for ^{70}Ge(p,n)^{70}As is shown in Fig. 1. EMPIRE calculations using EMPIRE specific level density (LEVDEN=0) under predict Levkovskii[13] data up to 12 MeV. Above 12 MeV projectile energy, the measured excitation functions are well reproduced by PCROSS and HMS options. But the MSC model fails to predict the measured cross section even above 12 MeV beam energy. Excitation function of ^{70}As formed in (p,n) reaction on ^{70}Ge is dominated by evaporation of neutrons at low projectile energies. As level density of the residual nucleus strongly influences the evaporation process, it is concluded that Empire specific level density (LEVDEN=0) does not properly predict the level density of the residual nucleus ^{70}As in this case. At higher energies the PEQ emissions are expected to

Table 1 — Induced Activity Values

Reaction	Half-life (hrs.)	Irradiation time (hrs.)	Induced Activity (Bq)	
			EMPIRE-3.2	TALYS-1.9
^{70}Ge(p,n)^{70}As	00.88	48	9862.08	9690.05
^{72}Ge(p,n)^{72}As	26.00	120	18328.61	16668.65
^{74}Ge(p,n)^{74}As	426.48	120	2460.20	2610.00
^{76}Ge(p,n)^{76}As	26.26	120	9119.67	9617.48

be predominant. The PCROSS subroutine in EMPIRE calculates pre-equilibrium cluster emission. Though no cluster emission is involved in the formation of ^{70}As in ^{70}Ge(p,n)^{70}As reaction, it is conjectured that the contribution to PEQ cluster emission channels leading to other products bring down the formation cross section of 70As which is well predicted by the EMPIRE1 and EMPIRE2 calculations above 12 MeV.

The excitation function of ^{70}Ge(p,n)^{70}As calculated by TALYS are consistent with the available data up to 12 MeV and but under predict the measured data at higher beam energies. It is inferred that Fermi gas level density formalism considered for the TALYS calculations could properly predict the level density of the product nucleus ^{70}As and its production cross section.

In the reaction ^{72}Ge (p,n)^{72}As shown in Fig. 2, the EMPIRE computations under predict the experimental data below 11 MeV but are in good agreement with the available data above 14 MeV. In this case also LEVDEN=0 does not properly predict the compound nuclear emissions in the reaction. Both the PEQ models PCROSS and MSC could predict the excitation function of ^{72}As fairly well. The TALYS computed results do not reproduce the experimental cross section in the energy range of 1-25 MeV except over a very narrow interval at 15-16 MeV. The calculations over predict the measured cross sections at 5-15 MeV and under predict them between 16-24 MeV. From these observations it is inferred that Fermi Gas level density option fails to properly predict the neutron evaporation while hybrid model could not assess the higher energy emissions.

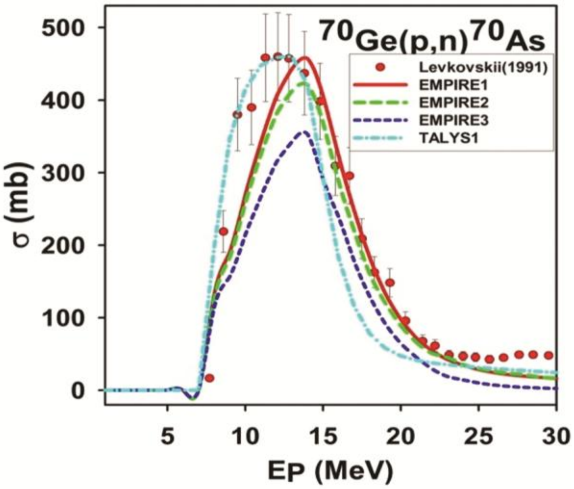

Fig. 1 — Excitation function of ^{70}Ge(p,n)^{70}As reaction.

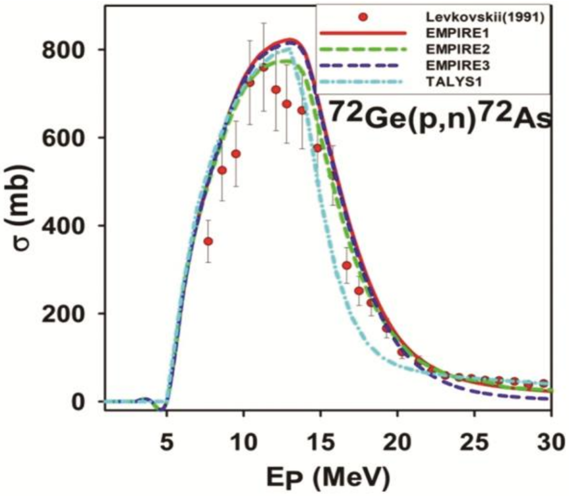

Fig. 2 — Excitation function of ^{72}Ge(p,n)^{72}As reaction.

The reaction channel opens around 4 MeV and experimental data[13-14] are given up to 30 MeV in the Fig. 3 for reaction ^{74}Ge(p,n)^{74}As. TALYS and EMPIRE calculations are in fair agreement with available data up to 15 MeV projectile energy. Above 15 MeV TALYS results give good agreement with the experimental data. EMPIRE4 and EMPIRE5 calculations slightly under predict the measured data; the disagreement is more in the case of EMPIRE6 calculations. EMPIRE (LEVDEN=1) using the Generalized superfluid model give good agreement as compared to EMPIRE specific level density. Since the level density of the residual nucleus plays a crucial role in compound nuclear reaction it is inferred that below 15 MeV projectile energy compound nuclear emissions have significant contribution in the excitation functions of ^{74}Ge.

The excitation functions of ^{76}Ge(p,n)^{76}As is shown in Fig. 4. The results of EMPIRE4, EMPIRE5 and TALYS calculations agree with measured data up to 5 MeV and above 11 MeV. EMPIRE6 results also

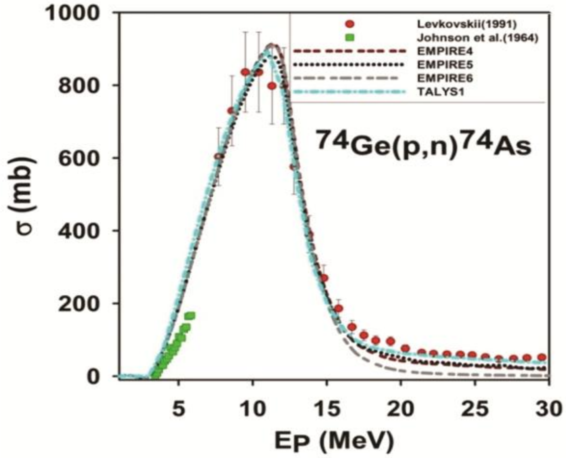

Fig. 3 — Excitation function of ^{74}Ge(p,n)^{74}As reaction.

reproduce the measured cross sections upto 5 MeV. No experimental data are available between 5 to 8 MeV. The formation cross-section of ^{76}Ge increases with the proton energy upto around 9 MeV and then it begins to fall down due to the opening of other reaction channels. Compound nuclear reaction dominates in lower energy range up to 10 MeV. Both the generalised superfluid model (EMPIRE calculations) and the Fermi gas model for level densities could predict the measured data well at low energies. Study of the excitation functions of the four isotopes reveals that as the neutron number increases pre-equilibrium contribution also increases.

The build-up of induced radioactivity, for ^{70,72,74,76}As obtained from ^{70,72,74,76}Ge(p,n) reactions respectively, has been calculated for a thick stopping target and 1 μA beam current upto 120 hours as shown in Figs. 5-8. The EMPIRE2 option is used to calculate induced radioactivity for ^{70}As and ^{72}As.Activity build-up of ^{70}As reaches saturation in 10 hours of irradiation as the half-life of ^{70}As is 0.88 hour. For ^{72}As ($t_{1/2}$ =26 hours) even after 120 hours of irradiation the induced radioactivity shows an increasing trend. The activity calculated by EMPIRE and TALYS nuclear model codes vary within 2 to 9%of each other due to the variation in the excitation functions.

In Figs 7-8 we have shown the induced activities of ^{74}As ($t_{1/2}$ =426.48 hours) and ^{76}As ($t_{1/2}$ = 26.26 hours) for 0-120 hours of irradiation of the targets. EMPIRE5 option is used to calculate induced activity for ^{74}Ge and ^{76}Ge. For ^{74}As and ^{76}As, TALYS and EMPIRE calculated results for induced activity differ about 5%, in each case TALYS giving a higher activity than EMPIRE. These estimates of induced

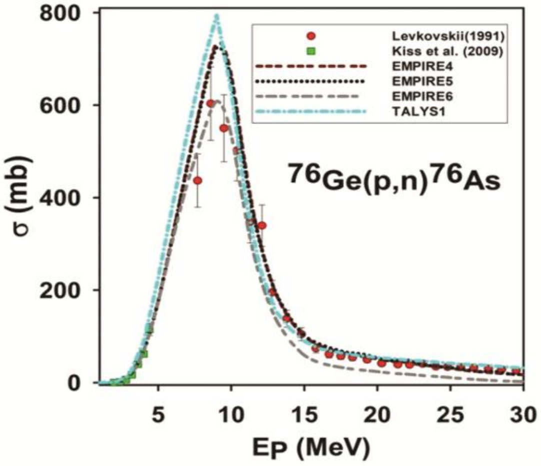

Fig. 4 — Excitation function of ^{76}Ge(p,n)^{76}As reaction.

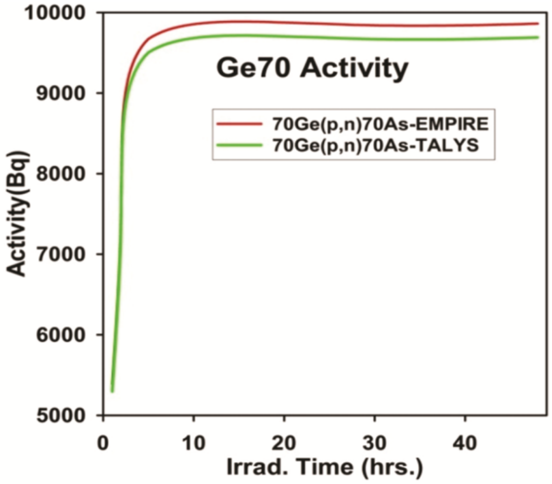

Fig. 5 — Activity build up in a thick Ge target irradiated by 30 MeV proton beam.

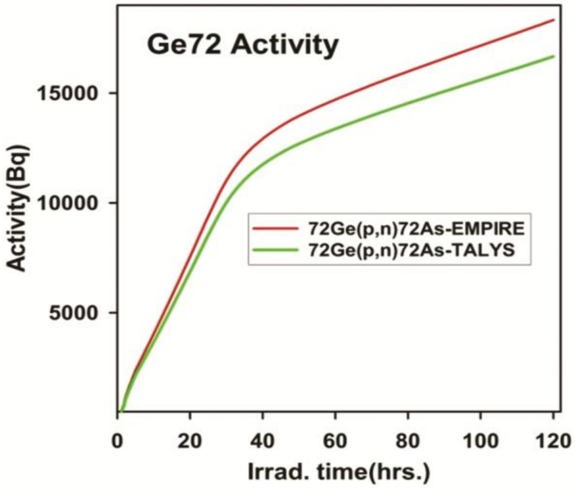

Fig. 6 — Activity build up in a thick Ge target irradiated by 30 MeV proton beam.

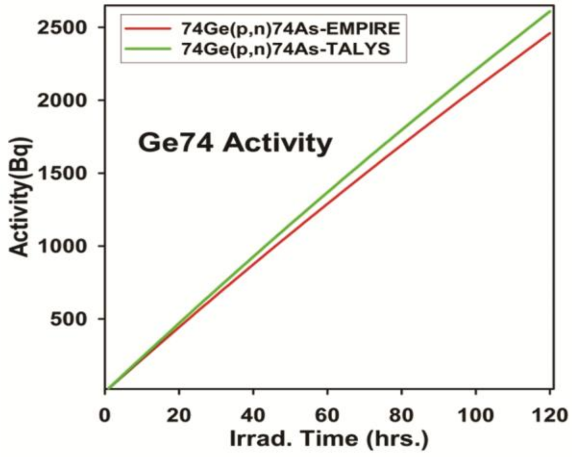

Fig. 7 — Activity build up in a thick Ge target irradiated by 30 MeV proton beam.

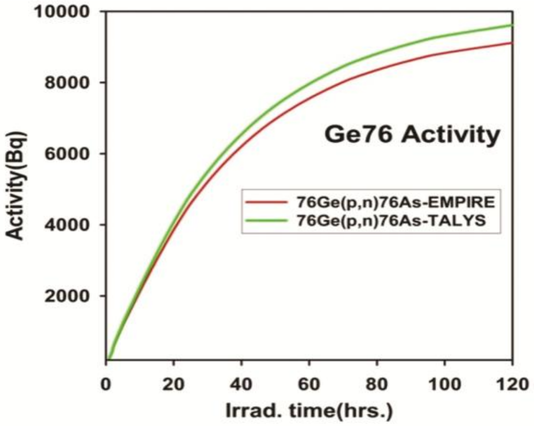

Fig. 8 — Activity build up in a thick Ge target irradiated by 30 MeV proton beam.

activity may be used for proper planning of experiments and to ensure radiation safety where experimental data are not available.

4 Conclusions

EMPIRE 3.2 and TALYS-1.9 nuclear reaction codes are used to estimate the production cross-sections and induced radioactivity of^{70,72,74,76} As due to the primary interaction of 30 MeV proton beam on ^{70,72,74,76}Ge targets. A thick stopping target, 1μA beam current and upto 120 hours of irradiation are considered to calculate the induced radioactivity. The pre-equilibrium models PCROSS along with Hybrid Monte Carlo Simulation in EMPIRE 3.2 predict the measured data well in the energy range above 12-25 MeV. At low projectile energies compound nuclear evaporation dominates. PEQ emissions contribute at energies above 15 MeV. The direct reaction mechanism contribution is found negligibleas compared to the compound nuclear emissions in the total reaction cross-section for the energy range considered. The production cross sections generated from these codes could be used for the estimation of induced activity in the absence of measured induced activity data for specific energy range. Saturation activity of the order of (9.5-9.8)x10^{3} Bq is produced for ^{70}As from thick ^{70}Ge target from a 30 MeV 1 μA proton beam. For ^{72,74}Ge target highest activity produced is ~1.8x10^{4}Bq and ~2.5x10^{3}Bq, respectively for 120 hours of irradiation. In the case of ^{76}Ge target maximum activity produced is ~9.5x10^{3} Bq. The chart of calculated induced activity will help in radiation safety planning as well as in planning of experiment. The computations using well defined and established parameters based on standard nuclear reaction models will be helpful to assess the relative precision of the available measured data and to check the validity of different parameters of the nuclear model codes.

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