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ORIGINAL PAPER

Energy dependence of pre-equilibrium emission for the (p,xn) reactions in niobium

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Abstract: Proton induced reactions have been studied in the energy region from ≈7.0 to 12.5 MeV for niobium, using the stacked foil activation technique followed by gamma ray spectroscopy. The excitation functions for the production of ^{93m}Mo, ^{93m}Nb and ^{89g}Zr have been determined. The experimental data have been compared with theoretical calculations based on compound reactions in statistical equilibrium as well as on pre-equilibrium reactions.

Keywords: Excitation functions; Nuclear reactions; Niobium; Proton induced reactions; Stacked foil activation technique; Pre-equilibrium emission; GDH model

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1. Introduction

Nuclear reaction cross-section data are generated by nuclear physics experiments and also by nuclear theory model codes [1]. To determine the optimum irradiation condition for the production yield of various radioisotopes more and more experimental nuclear reactions cross-section data are needed. These reaction cross-sections are also in demand in order to know the transmutation probabilities for the proposed accelerator driven systems [2] popularly known as energy amplifiers. Though several investigations [3] are available in literature for the determination of reaction cross-sections related to the production of radionuclides, there are large discrepancies in the cross-sections measured for the same reaction by different authors. Besides, analysis of these excitation functions in the past has been carried out on the basis of compound nucleus (statistical equilibrium) model and in general this mechanism of reaction could not account for the high energy tails of the excitation functions.

The possibility of particles being emitted after the first stage of nuclear interaction but long before the establishment of statistical equilibrium (pre-equilibrium [PE] emission) has been the point of interest for the last several years [4–6]. Many attempts have been made to understand such reactions. Starting from the pioneer work of Griffin [7], which provides the first explanation spectral shapes of the excitation function in the frame work of exciton model, many other semi-classical models have been proposed [8–12]. The hybrid and geometry dependent hybrid (GDH) models proposed by Blann [10, 11] have been found to be relatively simple and closed form models for the successful reproduction of the experimental data.

In this context, a careful and systematic study of excitation function and a comparison with the predictions of some of these models would help the understanding of the intricate mechanism of PE emission. With this motivation, the present work was undertaken to measure the excitation functions for residues ^{93m}Mo, ^{93m}Nb and ^{89g}Zr in ^{93}Nb proton interaction. Though present measurements were done up to 12.5 MeV beam energy but comparison of theory with literature values [13–20] has been made up to 30 MeV, to see the PE effect. As a matter of confidence the relative intensities of the identified γ-rays have also been measured.

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2. Experimental details

2.1. Target preparation

A spectroscopically pure niobium target was made of thickness ≈ 10.5 mg/cm^2, with purity ≈ 99.99 %. The niobium foil was cut into pieces of size 1.5 cm × 1.5 cm, and each of them was glued to an aluminum frame, having a circular hole of diameter 1.2 cm in its centre. Energy degrader aluminum foils of thickness ≈ 13.5 mg/cm^2 were sandwiched between the niobium foils whenever required, so as to get the wide range of energy of desired proton beam incident on each foil.

2.2. Irradiation

The stack comprising of eight target and degrader foils was irradiated for about 30 min with a ≈ 12.5 MeV diffused proton beam of diameter ≈ 5 mm, at the Variable Energy Cyclotron Centre, Kolkata, India. The beam energy was determined from a curve that related to the cyclotron RF with energy constructed from experimental data on elastic scattering. The energy of the proton particle after traversing half of the thickness of each target foil was computed from the stopping power table of Northcliffe and Schilling [21]. A typical experimental set up for the stack irradiation is shown in Fig. 1.

2.3. Flux measurements

During the irradiation of the stack, the counting of the incoming proton particles was done from an integrated beam charge. The beam was totally stopped in the electrically insulated irradiation heads serving as a kind of Faraday Cup [22] where electrons were prevented from escaping. Using this charge, flux was calculated. Copper foil of thickness ≈ 10.68 mg/cm^2 was used as a flux monitor [23] for checking the flux and good agreement was found with <10 % discrepancy.

2.4. Efficiency and energy calibration

The efficiency and energy calibration of the HPGe detector was employed using various standard sources, i.e., ^{22}Na, ^{57}Co, ^{60}Co, ^{133}Ba and ^{152}Eu of known strengths. The geometry dependent efficiency (ε_G) of the detector for different source detector distances was computed using the relation [24]

$$\varepsilon_G = Ce^{\lambda t}/S_o\theta \tag{1}$$

where C is the number of counts per second under the photo peak, θ is the absolute intensity of the relative gamma ray, λ and S_o are the decay constant and strength of the source at the time of its manufacture respectively, t is the time lapse between the date of manufacture of the source and the time of experiment. The values of θ and λ were taken from the Table of Radioactive Isotopes, by Browne and Firestone [25]. The values of ε_G thus obtained were plotted as a function of energy using the program origin 6.0. A polynomial of degree 4 having the following form was found to give the best fit for these curves.

$$\varepsilon_G = a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4 \tag{2}$$

where a_0, a_1, a_2, a_3 and a_4 are the coefficients having different values for different source detector distances. x Being the energy of the characteristic γ-ray. A typical geometry dependent efficiency curve of the 100 cm^3 HPGe detector obtained at a particular distance from the detector surface is shown in Fig. 2.

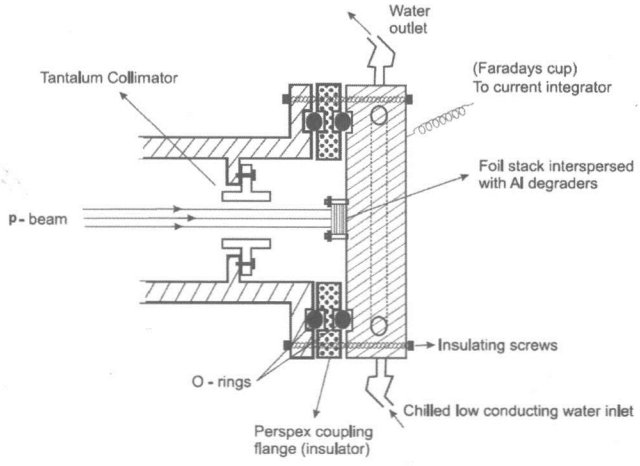

Fig. 1 Typical experimental setup for stack irradiation with proton beam

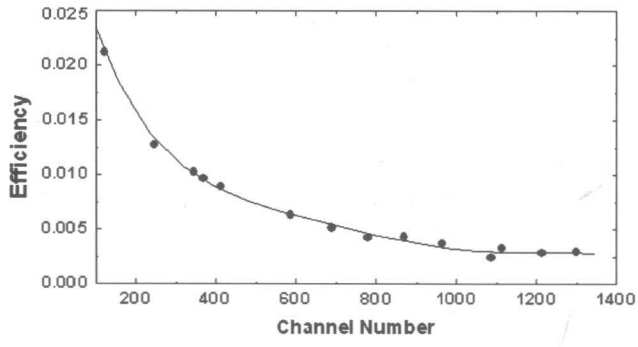

Fig. 2 Geometry dependent efficiency *curve* of the 100 cm^3 HPGe detector at a particular source to detector surface distance

Table 1 Spectroscopic data and measured relative intensities of gamma rays

γ-Rays Energy (kev)	Absolute abundance (θ) [15]	Normalized relative intensity	
		Present measurement	Literature value [15]
Reaction ^{93m}Nb(p,n)^{93}Mo, $t_{1/2}$ of product nucleus 6.85 h, Q value −1.20 MeV			
114	0.0067	7.1 ± 0.5	6.8
263	0.5676	627.1 ± 2.6	569.0
685	0.9968	1000.0 ± 4.6	1000.0[a]
1363	0.0078	7.9 ± 1.0	7.9
1477	0.9900	999.6 ± 5.6	994.0
Reaction ^{93}Nb(p,pn)^{92m}Nb, $t_{1/2}$ of product nucleus 10.15 days, Q value −8.816 MeV			
934	0.9900	...	100.0
Reaction ^{93}Nb(p,αn)^{89g}Zr, $t_{1/2}$ of product nucleus 3.268 days, Q value −5.498 MeV			
909	0.9901	...	100.0

[a] Normalization has been done with respect to this value from literature

2.5. Recording of gamma ray spectra and identification of reaction residue

After the irradiation of the stack, the characteristic γ-activities induced in the individual foils were recorded with a high resolution ($\approx$2 keV for 1,332 keV γ-ray of ^{60}Co) HPGe detector coupled to the ORTEC PC based multi-channel analyzer. The counting geometry was chosen by adjusting the target detector surface separation in such a way that the dead time remains less than 10 %. Evaporation residues were identified using their characteristic gamma rays adopted from the Table of Radioactive Isotopes by Browne and Firestone [25].

2.6. Formulation

The activation cross-section was computed using the following expression [26].

$$\sigma(E) = \frac{A\lambda \exp(\lambda t_2)}{N_o \cdot \phi \cdot (\varepsilon_G) \cdot \theta \cdot K \cdot [1 - \exp(-\lambda t_1)][1 - \exp(-\lambda t_3)]} \quad (3)$$

where $K = [1 - \exp(-\mu d)]/(\mu d)$ is the correction factor for the self-absorption of gamma rays in the sample of thickness d (g/cm^2) and of absorption coefficient μ (cm^2/g). A is the counts under the photo peak of the characteristic gamma ray, λ is the decay constant of the residual radioisotope, N_o is the number of nuclei in the sample, θ is the absolute intensity of the characteristic γ-ray, ε_G is the geometry dependent efficiency of the HPGe detector, ϕ is the average flux of the incident proton beam, t_1 is the irradiation time, t_2 is the time lapse between stopping the beam and start of counting, and t_3 is counting time.

3. Experimental results

The spectroscopic data of identified gamma rays are given in Table 1. Other details viz. residual nucleus, Q value, half life, gamma ray energies, and corresponding intensities are also given in Table 1. The Q values of different reactions were taken from Atomic Data Nuclear Data Tables by Wapstra and Bos [27] and other decay data from Table of Radioactive Isotopes by Browne and Firestone [25]. To check the identification of the gamma rays, the relative intensities of detected gamma rays have been calculated. It can be seen from Table 1 that the measured relative intensities are in good agreement with their respective literature values [25].

The activation cross-section for the same reaction has been calculated from the intensities of the various identified γ-rays emitted from the same residual nucleus. The reported value has been taken as the weighted average [28] of the various cross-section values so obtained. The overall error in the present measurement is estimated to be ≤38 % including the statistical errors. The measured cross-sections for the population of residues ^{93m}Mo, ^{93m}Nb and ^{89g}Zr are presented in Table 2.

4. Model calculations

The excitation functions have been evaluated theoretically using the computer code ALICE-91 [29]. This code employs the Weisskopf–Ewing model [30] for statistical component and GDH model of Blann [11] for the PE emission. Since several authors have already discussed the code and the theories involved, we restrict ourselves here by referring only to the review of Blann [12] on PE decay.

Table 2 Experimental cross-sections for (p,n) (p,pn) and (p,αn) reactions

Cross-section (mb)			
Projectile energy (MeV)	$(p,n)^{93m}MO$	$(p,pn)^{92m}Nb$	$(p,\alpha n)^{89g}Zr$
7.2	5.7 ± 0.6		
8.0	9.9 ± 0.6		
8.7	15.1 ± 0.9		
9.8	18.6 ± 1.1		
10.6	22.9 ± 1.0		
11.2	26.7 ± 1.4		
11.9	30.4 ± 2.0	1.2 ± 0.4	0.4 ± 0.08
12.5	31.2 ± 1.4	1.3 ± 0.5	0.2 ± 0.05

In this code, the level density parameter constant K may be varied to match the experimental data. In the present calculations, a value of $K = 8$ has been found to reproduce experimental data satisfactorily. For the PE calculations the initial exciton number (n_0) was taken to be 3 (1p + 1n + 1h) as it was derived from the investigation of nucleon spectra [31].

5. Results and discussion

The measured excitation functions together with the literature values [13–20] and ALICE-91 [29] calculations are shown in Figs. 3, 4 and 5. The excitation function of the $^{93}Nb(p,n)^{93m}Mo$ reaction, measured in this work (Fig. 3), was found to be in good agreement with the values reported by Levkovskij [15], Avila-Rodriguez et al. [16] and Ditroi et al. [17]. The data of Albert [13], Chodil et al. [14], Singh et al. [18] and Kiselev and Faizrakhmanova [19] for this nuclear reaction shows considerable discrepancies in the magnitude of cross-section values. A better agreement in the trend of the excitation functions of our measured data and

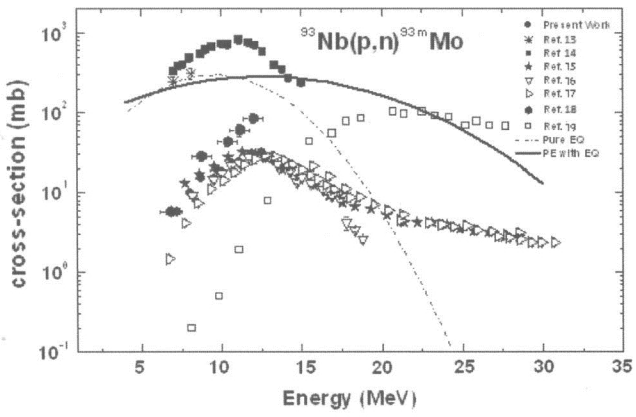

Fig. 3 Experimental and theoretical excitation function for the $^{93}Nb(p,n)^{93m}Mo$ reaction

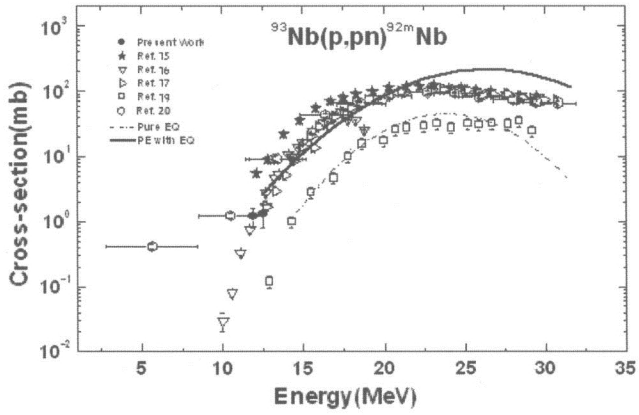

Fig. 4 Experimental and theoretical excitation function for the $^{93}Nb(p,pn)^{92m}Nb$ reaction

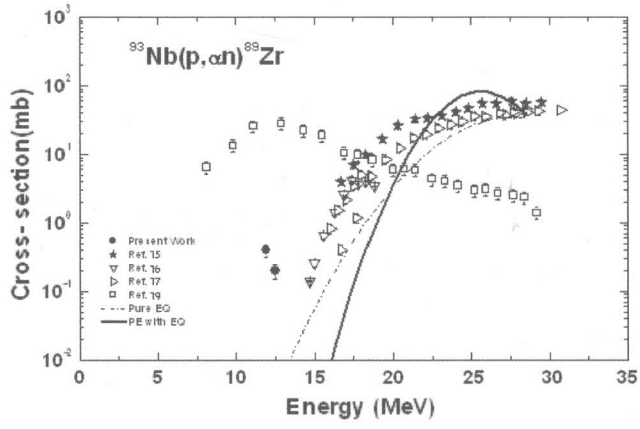

Fig. 5 Experimental and theoretical excitation function for the $^{93}Nb(p,\alpha n)^{89g}Zn$ reaction

the literature values was found when executing theoretical calculations using GDH model (solid line) [11]. The theoretical and the experimental data for the $^{93}Nb(p,pn)^{92m}Nb$ reaction is shown in Fig. 4. The good agreement between the experimental data and the theoretical values was found when executing GDH model (solid line) calculations using the ALICE-91 code. However, discrepancies in the magnitude of cross-section values are evident between the data reported by Levkovskij [15] and Kiselev and Faizrakhmanova [19]. Figure 5 shows the excitation function of $^{93}Nb(p,\alpha n)^{89}Zr$ reaction. As can be seen from Fig. 5, discrepancies in the magnitude of cross-section values are evident between the data reported by different groups [15–17, 19] Again, the trend of the excitation functions of literature values and theoretical values in the high energy range agree with the ALICE-91 [29] GDH Model calculations (solid line) except with data of Kiselev and Faizrakhmanova [19].

The present analysis indicates clearly the presence of significant PE contributions in proton induced reactions. The PE fraction (f_{PE}) is a measure of the relative weight of

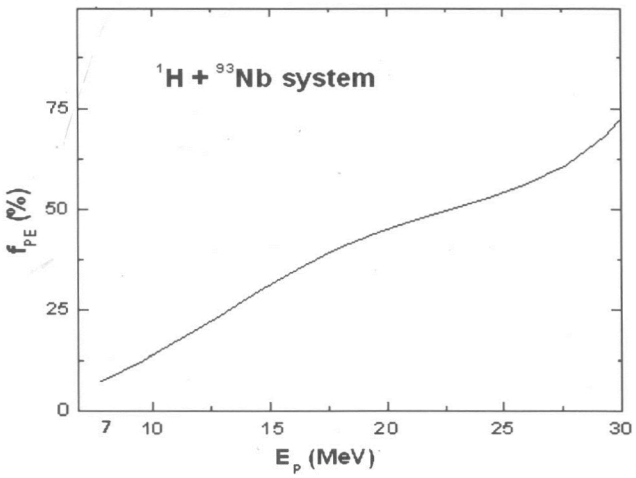

Fig. 6 PE fraction (f_{PE}) of the total reaction cross-section as a function of proton particle energy

the PE contribution needed for the reproduction of excitation functions and it reflects the relative importance of PE and equilibrium processes. It is more meaningful to look for the total PE fraction of all type of emitted particles [32]. In a given target nucleus the total PE fraction, for all types of reactions like (p,xn) reactions, are calculated using the ALICE-91 [29] code. Because of the considerable contributions to the PE fraction from the PE emission of charged particles, the calculated total PE fraction are not directly comparable with the measured excitation functions for (p,xn) type reactions. However, no definite trend for the variation of the PE fraction with the excitation function energy or compound mass number and changes in initial exciton number are reported [32], yet it is reasonable to assume that f_{PE} depends on the excitation energy of the compound system [12]. In the present calculations, the f_{PE} is inherently energy dependent. This dependence is derived from consideration of the internal transition rates and of the continuum decay rates. The f_{PE} has been taken to be proportional to the cumulative sum of the probability of finding the particle in the continuum for every possible configuration during the process of equilibrium. The calculation f_{PE} for the system ^{93}Nb is shown in Fig. 6, as a function of bombarding energy in the energy range $\approx$7–30 MeV. It can be seen that the f_{PE} increases with incident proton beam energy.

6. Conclusions

In general, it is evident from Figs. 3, 4 and 5, that PE emission of multi-particles is necessary before the system is equilibrated and hence the experimentally observed high energy tail of the excitation function can be explained only when the combination of semi-classically treated PE emission GDH model followed by particle evaporation from the equilibrated system (Weisskopf–Ewing model) is taken into account. The pure equilibrated reaction in its decay is unable to explain the experimental data in the high energy tail portion of the excitation function. It is clear from Figs. 3, 4 and 5 that calculated values shown by broken lines (based on the pure equilibrium model) do not reproduce the experimental data. These data are reproduced only when the PE emission is also taken into account, as shown by solid lines.

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