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Excitation functions of alpha particle induced reactions on aluminium and copper

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Abstract. Cross-sections for the reactions with product nuclei ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga were investigated over the energy range of 30 to 75 MeV for alpha particle induced reactions on natural aluminium and copper, using stacked-foil activation technique. The measured excitation functions were analysed with special reference to their suitability for monitoring beam energy and intensity. The experimental results were compared with the predictions of hybrid model of Blann. The assumption of initial exciton number $n_0 = 4(4p0h)$ best satisfies the measured excitation functions in the present work.

Keywords. Nuclear reactions; stacked foil activation technique; hybrid model.

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1. Introduction

Nuclear reactions are frequently understood in terms of two extreme models on the basis of their time scale of occurence. These are the fast direct reaction [1] and the slow compound nucleus reaction mechanisms [2].

However, in recent years, there has been an increasing evidence pointing out to new types of processes, known as pre-equilibrium reaction mechanism [3]. Several semi-classical and quantum mechanical models [4–14] have been proposed to explain the emission of energetic light particles by the equilibration process (pre-equilibrium emission) from the nuclear system excited at medium energies. Predictions from these models as to excitation functions and the energy spectra of the emitted particles compared well with the existing experimental data. This has prompted a continued interest in these models as tools both to predict cross sections for a number of practical purposes and to test the adequacy of the underlying physics of the models. There are many data on inclusive energy spectra of light ejectiles [3] but the complimentary information on residual nucleus excitation functions is far from being abundant.

Regarding alpha particle induced reactions, the integral cross sections data exist for large number of target nuclei. However, the present measurement, of the radioactive isotopes ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga are of great importance regarding their possible application as standard monitor nuclides for the determination of beam currents in nuclear reaction studies with low and intermediate energy alpha particles. For optimising the conditions of ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga (targetting, yields, purity etc.) a knowledge of the excitation function was necessary. The alpha particle induced reactions on aluminium were earlier measured by a few groups [15–17] with large mutual disagreement among themselves, with some measurements having errors ranging from 20–40% [15]. Even though large number of experimental data are available in literature [18–25] for the alpha induced reactions producing ^{68}Ga and ^{67}Ga, most of them were obtained using poor resolution detectors. However, some of the measurements have also been done using good resolution Ge (Li) detectors [22–25].

In view of the large uncertainties and mutual discrepancies, precise and accurate measurement of these reactions are still needed. With the advent of high resolution and large volume semiconductor detectors and with the availability of good quality beams, the excitation functions measurement has entered into a new era. Therefore the present work is carried out with the following motivations:

(1) determination and optimisation of the ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga yields via alpha induced reactions on aluminium and copper,
(2) to compare the experimentally obtained data with the theoretically calculated results [7, 26, 27],
(3) to study the reaction mechanism involved in the production of ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga isotopes.

2. Experimental procedure

2.1 *Irradiation and counting*

Stacked-foil activation technique was employed in the present work. Stacks of self supporting metallic foils of aluminium and copper of purity greater than 99.99% and of thicknesses $14\,mg/cm^2$ and $23\,mg/cm^2$, respectively were used for the purpose of irradiation. Copper foils were interspersed with aluminium foils of varying thickness acting as energy degraders, while the aluminium stack was prepared without any degrader foils. The irradiation of the stacks were carried out at the Variable Energy Cyclotron Centre, Calcutta. The primary beam of the alpha particles was kept at 75 MeV for different irradiations. The beam spot on the foil stack was always restricted to 5 mm by a central hole in a 6 mm thick collimator placed in front of the stack. Beam current of the order of (200/250) nA was maintained in each irradiation.

The activation produced in the experimental foils have been measured by a $95\,cm^3$ coaxial Ge (Li) detector, having resolution of 2 KeV for the 1332 keV peak of ^{60}Co in a well-defined reproducible geometry. The residual nuclei were identified using their characteristic gamma-rays as listed in table 1 [28]. The energy calibration and relative detector efficiency were calculated from ^{152}Eu standard spectra.

2.2 *Flux measurements*

During the irradiation of the stack, the counting of the incoming alpha particles was done from an integrated beam charge. Here the beam was totally stopped in the electrically

Table 1. Reaction studied in the present work and the relevant decay characteristics of residual nuclei.

Reaction	Residual nucleus	Q-Value (MeV)	Half-life $T_{1/2}$	E_γ (KeV)	θ_γ (%)
$^{27}Al(\alpha, \alpha 2pn)$	^{24}Na	−31.40	15.05 h	1369	100
$^{27}Al(\alpha, 2\alpha n)$	^{22}Na	−22.50	2.60 Y	1275	100
$^{65}Cu(\alpha, n)$	^{68}Ga	−5.85	68.00 m	1077	3
$^{65}Cu(\alpha, 2n)$	^{67}Ga	−14.10	28.26 h	184.5	23.56
				300.2	19.00

insulated irradiation head serving as a kind of Faraday cup [17–21] where secondary electrons were prevented from escaping. Using this charge, the flux was calculated.

2.3 *Cross-section determination*

The formula used for the determination of reaction cross section is

$$\sigma = \frac{A_\gamma A_{\rm gm} \lambda}{\phi W_i P_i P_\gamma \theta_\gamma N_{\rm Av}[1 - \exp(-\lambda t_{\rm i})][\exp - \lambda t_{\rm w}][1 - \exp(-\lambda \Delta)]},$$

where, σ is the cross section for the reaction, A_γ is the photopeak area of the characteristic gamma ray of the residual nucleus, $A_{\rm gm}$ is the gram atomic weight of the target element, λ is the disintegration constant of the residual nucleus, ϕ is the flux of the incident particles, W_i is the weight per unit area of the target foil, P_i is the fractional abundance by weight of the target isotope of interest, θ_γ is the fraction of the characteristic gamma ray emitted, P_γ is the photopeak efficiency of the gamma rays, $N_{\rm Av}$ is the Avogadro number, $t_{\rm i}, t_{\rm w}, \Delta$ are the periods of irradiation, waiting and counting respectively.

2.4 *Errors*

The total error in the experimental cross section is contributed by 1–4% error in the photopeak areas of characteristic gamma-ray, 1–8% error in their absolute abundances, 3% error in the photopeak efficiency of gamma-ray, 1–2% error due to the non-uniformity of the foil thickness and 1–6% error in the flux measurement.

3. Experimental results

3.1 *Excitation functions for the product nuclei ^{24}Na and ^{22}Na*

The excitation functions for the reactions leading to the formation of ^{24}Na and ^{22}Na are shown in figure 1. These reactions were earlier studied by Bowman and Blann [15] up to 120 MeV using 6 cm^3 and 32 cm^3 Ge (Li) detectors, with an overall error ranging between 20–40%. Probst *et al* [16] measured the above reactions up to 156 MeV, with no measurements below 50 MeV, which forms a major part of the present energy region of interest. Ismail [17] studied the above reactions only up to 65 MeV, but there are no

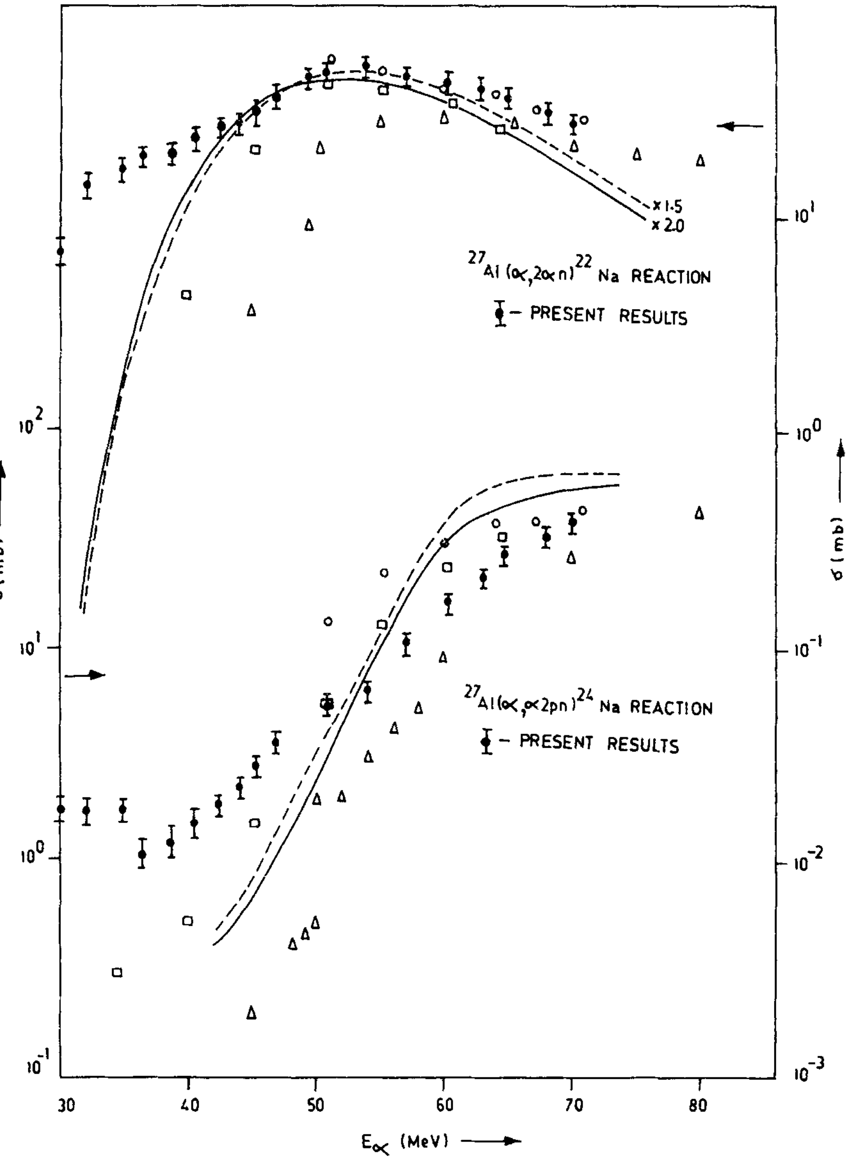

Figure 1. Comparison of the present and previous experimental results along with the theoretical excitation functions of (**a**) $^{27}Al(\alpha, 2\alpha n)$ ^{22}Na, (**b**) $^{27}Al(\alpha, \alpha 2pn)$ ^{24}Na reactions. □ Ismail [17], ○ Probst *et al* [16], △ Bowman and Blann [15], – – – Weisskopf–Ewing estimate (W.E.), ——— hybrid model (ALICE/85/300).

theoretical results to support his data. There are large mutual disagreements among all the previous results, especially those measured by Ismail. Hence the present investigations of these reactions have been carried out up to 75 MeV, using a high resolution 95 cc Ge (Li) detector. It can be seen in figure 1 that in an all important energy region between 40 to 70 MeV, the present results show better systematic trend than those of earlier reported measurements.

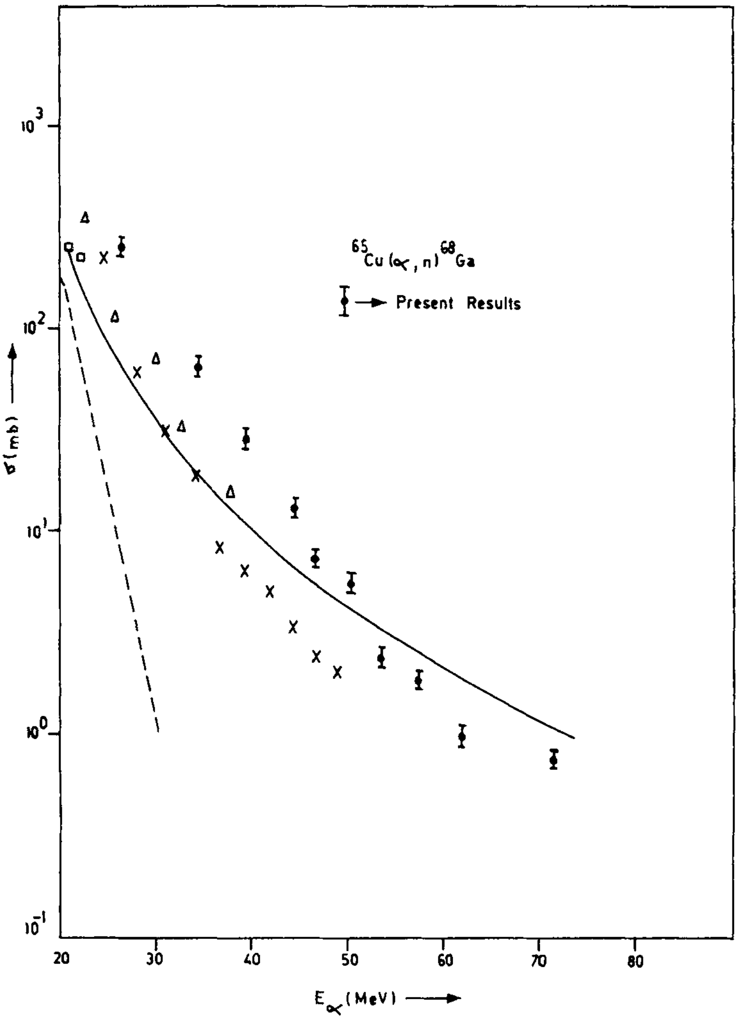

Figure 2. Comparison of the present and previous experimental results along with the theoretical excitation functions of $^{65}Cu(\alpha, n)$ ^{68}Ga reaction X N L Singh *et al* [25], □ Rizvi *et al* [23], △ Bharadwaj *et al* [24]. – – – Weisskopf–Ewing estimate (W.E.), ——— hybrid model (ALICE/85/300).

3.2 *Excitation function for product nuclei* ^{68}*Ga and* ^{67}*Ga*

The above mentioned reactions for the product nuclei ^{68}Ga and ^{67}Ga were earlier studied by several authors [18–21], mostly using poor resolution detectors, thereby resulting in large inconsistencies among the measured data. Recent measurements are also available [22–25] using Ge (Li) detectors. However, a careful look at figures 2 and 3 reveals that there are large uncertainties and mutual discrepancies in the earlier results, especially in the reaction with product nucleus ^{68}Ga. The present study reports new data points in the energy region between 50 to 75 MeV.

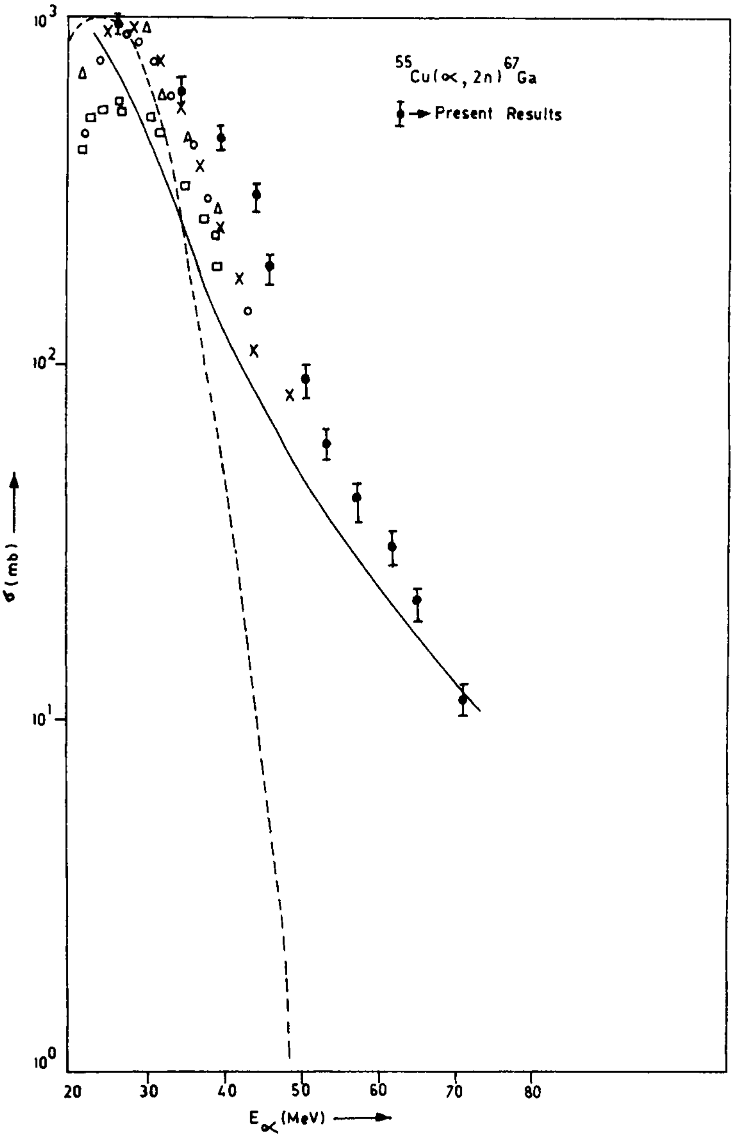

Figure 3. Comparison of the present and previous experimental results along with the theoretical excitation functions of ^{55}Cu($\alpha, 2n$) ^{67}Ga reaction X N L Singh *et al* [25], □ Rizvi *et al* [23], △ Bharadwaj *et al* [24], ○ Graf *et al* [22], - - - Weisskopf–Ewing estimate (W.E.), ——— hybrid model (ALICE/85/300).

From the viewpoint of potential application as monitor reaction, the excitation functions for the production of the above radionuclides appears to be interesting. Out of these ^{68}Ga has a relatively short half-life period, ^{22}Na has a very long half-life (see table 1) and others have half-lives in the hour–day range. The excitation functions for the ^{22}Na and ^{24}Na radionuclides exhibit 'plateau' over broad energy range from 35 to 75 MeV. This can help avoid errors in the flux determination originating from the improper estimation of the middle energy of the bombarding particle in the monitor foil. The excitation functions for the other two product nuclei ^{67}Ga and ^{68}Ga show a systematic trend in the entire region starting from 30 to 75 MeV. Therefore, these reactions may also be used for the determination of flux of the bombarding alpha particle.

4. Comparison with hybrid model

In the present work the excitation functions are calculated on the basis of the hybrid model [7, 26, 27] with and without the inclusion of pre-equilbrium emission of particles. Basically the hybrid model is semi-classical and hence involves a large number of physical parameters along with a few adjustable parameters. A short description of the options chosen is given below. The nuclear masses were calculated from the Myers–Swiatecki mass formula [29] with the liquid drop having shell correction term without pairing, i.e. the level density pairing absorbed in binding energies. The inverse cross-sections were calculated by optical model subroutine included in the code. A level density parameter of $A\backslash 8\,\mathrm{MeV}^{-1}$ was used. For the equilibrium part, the so called S-wave approximation is used which takes into account the full gamma, competition with particle emission. The initial exciton number $n_0(P_0h_0)$ plays an important role because it governs the entire cascading process of binary collisions and thereby influences the shape of hard component in the particle spectra.

We have made the theoretical calculations using the initial exciton configurations $4(4p0h)$, $5(5p0h)$ and $6(5p1h)$. It was observed that the theoretical predictions of the hybrid model with $4(4p0h)$ gives better results, as compared to the predictions of the $5(5p0h)$ and $6(5p1h)$ configurations with respect to initial configuration. Blann and Mignerey [30] used $n_0 = 4(4p0h)$ to calculate (α, p) spectra on cobalt. Gadioli *et al* [31] have also discussed this point in detail and recommended the general application of $n_0 = 4$. Therefore we have shown $4(4p0h)$ configuration in the comparison. Evidently, an initial exciton $n_0 = 4(4p0h)$ configuration which is equivalent to a break up of the incoming alpha particle in the field of the nucleus and the nucleons occupying excited states above the Fermi level gives a better description of the excitation functions compared to other configurations for the alpha particle energies up to 80 MeV.

Figures 1 to 3 shows the observed excitation functions for the reactions with product nuclei ^{24}Na, ^{22}Na, ^{68}Ga and ^{67}Ga. As shown in figure 1 the reactions with product nuclei ^{24}Na and ^{22}Na contain alpha particles in the outgoing channel. Hybrid model [7, 26, 27] on the other hand does not account for the pre-equilibrium emission of complex particles. Gadioli *et al* [32] have shown that the contribution of pre-equilibrium emission of alpha particles is pre-dominant in the valley which follows the first evaporation peak. Therefore, from the shape of the excitation functions for the above two reactions between 40 to 80 MeV, it can be argued that pre-equilbrium alpha particles can not account for the near plateau cross section in the $(\alpha, 2\alpha n)$ ^{22}Na reaction and slow monotonically rising part in the $(\alpha, \alpha 2pn)$ ^{24}Na reaction. Thus one can conclude that, the dominant mechanism for the reaction induced by alpha particles in the energy range 40 to 80 MeV is by statistical evaporation process. Similar observations were also made by Bowman and Blann in their study of mean recoil ranges of alpha induced reactions on ^{27}Al. They found that there is only 20 to 25% of full momentum transfer for ^{24}Na and ^{22}Na, confirming compound nucleus mechanism in the reaction for the production of ^{24}Na and ^{22}Na.

Figures 2 and 3 shows the reactions ^{65}Cu(α, n) and ^{65}Cu$(\alpha, 2n)$, for the production of ^{68}Ga and ^{67}Ga respectively. In (α, n) reaction, as shown in figure 2, the compound nucleus mechanism, given by Weisskopf–Ewing estimate, is dominating in the lower part of the excitation function, while there are considerable pre-equilibrium effects beyond 30 MeV of bombarding energies. Similar arguments can also be made for $(\alpha, 2n)$ reaction, where the pre-equilibrium effects predominate beyond 50 MeV of excitation energy.

It can be seen in figure 2 that there is a clear shift in energy between the theoretical and experimental compound nucleus peaks. Generally, such shifts are prescribed to the complete neglect of angular momentum effects in the Weisskopf–Ewing theoretical calculations of the compound nucleus contributions. More elaborate computing using Hauser–Feshbach theory may bring about a better agreement.

5. Conclusions

Four excitation functions for the production of ^{22}Na, ^{24}Na, ^{67}Ga and ^{68}Ga radionuclides from the alpha bombardment of aluminium and copper were measured for alpha energies up to 75 MeV. The high resolution Ge(Li) detectors were employed and a definite improvement in the quality of data was observed. In all the four reaction products ^{22}Na, ^{24}Na, ^{67}Ga and ^{68}Ga, the excitation functions for the reaction products ^{67}Ga and ^{68}Ga were studied for the first time in the energy range 50 to 75 MeV.

A comparison of the experimental data with the results of hybrid model calculations were performed with and without using the pre-equilibrium emission of particles in the code. The initial exciton configuration was varied between $n_0 = 4(4p0h)$ to $n_0 = 6(5p1h)$. However, $n_0 = 4(4p0h)$ gave the best agreement between the theory and experiment. A pronounced pre-eqilibrium emission of neutron was observed in the reactions with product nuclei ^{67}Ga and ^{68}Ga beyond 30 and 40 MeV respectively. However the reactions with product nuclei ^{24}Na and ^{22}Na mainly proceeds through statistical evaporation of nucleons and alpha particles. So far as their application in the alpha particle flux determination is concerned it can be concluded that ^{24}Na and ^{22}Na are the suitable reaction products because of the presence of a plateau-like structure in the excitation function over a very wide energy range.

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