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Alpha particle induced reactions on copper and tantalum

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Abstract. Alpha particle induced reactions in the target elements copper and tantalum were studied. The excitation functions of ^{63}Cu(α, n) + ^{65}Cu($\alpha, 3n$), ^{65}Cu($\alpha, 2n$), ^{181}Ta($\alpha, 2n$) and ^{181}Ta($\alpha, 4n$) were measured up to 75 MeV. *Eight* new energy point cross-sections were measured for the first time. The experimental results were compared with the predictions of updated hybrid model (ALICE/85/300) as well as with index model using the initial exciton number $n_0 = 4$ (4poh) and level density parameter, $a = A/8$. A general agreement was found for all the reactions with both the models.

Keywords. Hybrid model; index model; stacked-foil activation technique; pre-equilibrium decay; ALICE/85/300 code; index code.

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1. Introduction

Nuclear reactions at medium and high energies have frequently been analyzed in terms of two extreme types of models. They are direct reaction model and compound nucleus model. An ever increasing evidence accumulated during the last two decades concerning reactions induced by light projectiles in the energy region 20–200 MeV, pointed out to some new physics coming into play in this energy region of excitation. The experimental features (Blann 1975; Seidel *et al* 1976; Machner 1985) apparently related to but subtly differing from the well-known characteristics of direct and compound nucleus reactions, seem to provide a linkage between the two extreme models. In the new perspective it is assumed that the projectile-target composite system is characterized by a few degrees of freedom in the first interaction and passes through a series of intermediate stages of increasing complexity and reaches the final stage of a compound nucleus having a statistical equilibrium, with a large number of degrees of freedom. The most noteworthy point is that the reaction may come to an end, by the emission of particles leaving behind the residual nucleus, at any point in the above sequence.

During the last few decades many nuclear reaction models have been developed to treat the pre-equilibrium phase of reaction leading to the formation of the compound nucleus. Although a variety of approaches and computational techniques have been employed, most of these models utilize in one way or another one or both of the two basic concepts ingrained in the intra-nuclear cascade model of Goldberger (1948) and the statistical model of intermediate structure first proposed by Griffin (1966, 1967).

In later years much effort has been devoted to combine the advantageous features of intranuclear cascade model and Griffin's statistical model of intermediate structure into a simple formalism capable of calculating the absolute pre-equilibrium emission cross-sections and yet retaining the simplicity of statistical description of the intermediate structure. As a result, a number of formulations, the hybrid model of Blann (1971), the exciton model of Gadioli *et al* (1973) and many other models (Cline 1973; Betak and Dobes 1976) have emerged as descendents of Griffin's statistical model of intermediate structure. All these models have the common feature that they group the many body states of equilibrating system according to the exciton numbers and particle hole densities to estimate the occurrence of configuration capable of pre-compound particle emission. There are some conceptual differences between the hybrid model and exciton model. The long-standing controversy on the basic viewpoints was resolved by Ernst and Rama Rao (1977). Blann and Vonach (1983) extended the hybrid model to predict in an approximate way, the emission up to two nucleons in pre-equilibrium phase. On the other hand Ernst *et al* (1987) have proposed a model of independently interacting excitons, known as index model in which it is shown that each generation in the frame work of exciton model is characterized by the interaction of exactly one exciton. As such, the index model predicts the exclusive emission spectra which can easily be converted into residual nucleus population probabilities. It is shown that three stages are sufficient to describe single and multinucleon emission to all orders of practical importance.

In both the latest codes of hybrid (Alice 85/300) (Blann 1984) and index (Ernst *et al* 1987) models, at the end of the precompound stages, the equilibrated compound nucleus is calculated using the Weisskopf–Ewing formalism (Weisskopf and Ewing 1949) and added incoherently to the pre-equilibrium contributions, so that the theoretical excitation function can be directly compared with the experimental ones. In this context a careful and systematic experimental study of the excitation functions on typical light and heavy nuclei and to make a comparison with the predictions of these two models, would heip the understanding of the mechanism of pre-equilibrium emission. With this chief motivation, we have undertaken the present work and measured the excitation functions of four reactions up to 75 MeV. The experimental results have been compared with the theoretical predictions based on the updated hybrid model as well as index model.

2. Experimental procedure

The excitation functions of $[^{63}Cu(\alpha,n)+{}^{65}Cu(\alpha,3n)]$, $^{65}Cu(\alpha,2n)$, $^{181}Ta(\alpha,2n)$ and $^{181}Ta(\alpha,4n)$ reactions were measured by means of stacked foil activation technique and Ge(Li) gamma ray spectroscopy method, up to 75 MeV. Spectroscopically pure (99·99%) copper foils and tantalum foils of thickness 25 mg/cm^2 and 57 mg/cm^2 respectively, were used as targets in the stacks together with several aluminium degraders of varying thicknesses to reduce the beam energy to desired levels. The energy of the alpha particles after they had traversed half the thickness of each foil was determined by Williamson *et al* (1966). The irradiations were performed (Mohan Rao 1989) at Variable Energy Cyclotron, Calcutta (India). Beam currents of the order of 200 nA were maintained in each stack so that a few hours of well controlled irradiation was necessary to excite the required activities in the stacks. The flux

Table 1. Reactions investigated and the decay characteristics of the residual nuclei.

Reaction	Residual nucleus	Q-value (MeV)	Half life	E_γ (keV)	I (%)
^{63}Cu(α, n)	$^{66}_{31}$Ga	−7·5	9·45 h	834	6·0
				1039	38·0
				2752	23·0
^{65}Cu(α, 3n)	$^{66}_{31}$Ga	−26·9	9·45 h	834	6·0
				1039	38·0
^{65}Cu(α, 2n)	$^{67}_{31}$Ga	−14·1	78·3 h	93	38·0
				184	24·0
				300	19·0
^{181}Ta(α, 2n)	$^{183}_{75}$Re	−16·35	71·0 d	110	2·9
				162	23·6
				209	3·0
				292	3·2
^{181}Ta(α, 4n)	$^{181}_{75}$Re	−31·75	19·97 h	361	12·3
				365	57·0
				639	6·5
Monitor Reactions					
^{27}Al(α, $\alpha 2pn$)	^{24}Na	−31·4	15·05 h	1369	100
^{27}Al(α, 2αn)	^{22}Na	−22·5	2·60 a	1275	100

monitor reactions used were ^{27}Al(α, $\alpha 2pn$)^{24}Na and ^{27}Al(α, 2αn)^{22}Na for which well measured cross-sections are available in literature (Probst *et al* 1976). Gamma activities produced in each foil were measured at VECC, Calcutta and Physics Department, Banaras Hindu University, Varanasi, using 95 c.c. Ge(Li) and 50 c.c. HPGE detectors having resolution of $\simeq 2$ keV for 1332 keV photons, in conjunction with a 4 K multichannel analyzer. The spectroscopic data necessary for the evaluation of cross-sections are shown in table 1. The energy and efficiency calibration of the detectors were performed by using a standard ^{152}Eu source of known strength. The formula used in the cross-section measurement along with the other details are given in our earlier paper (Rama Rao *et al* 1987)

$$\sigma = \frac{A_\gamma A_{\mathrm{gm}} \lambda}{\phi W P_\gamma \theta_\gamma P(1 - \exp(-\lambda t_i))} \frac{1}{\exp(-\lambda t_w)(1 - \exp(-\lambda \Delta))} \tag{1}$$

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_{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 is the weight per unit area of the target foil; P is the fractional abundance by weight of the target isotope of interest; θ_γ is the fraction of characteristic gamma rays emitted per decay of the residual nucleus; P_γ is the photopeak efficiency of the gamma ray; t_i, t_w, Δ are the periods of irradiation, waiting and counting respectively.

The total error in the present measurements of cross-sections ranges between 8 and 12%, which consists of uncertainities in photopeak area, detection efficiency, uniformity of foil thickness, spectroscopic data and that of monitor cross-section.

3. Experimental results

Consequent on the use of natural copper target, isotopic contributions arise in the production of a given final nucleus such as for example ^{66}Ga formed in ^{63}Cu(α, n) and ^{65}Cu(α, 3n) reactions jointly. However in view of the large difference in the Q-values of both the reactions, there will be energy regions where either of them predominate over the other. On this basis the experimentally measured weighted average cross-sections can be easily interpreted using the formula

$$\langle\sigma\rangle = \bar{A}\left(\frac{P_1}{A_1}\sigma_1 + \frac{P_2}{A_2}\sigma_2\right) \tag{2}$$

where $\bar{A}$ is the average atomic weight of copper, P_1, A_1 and P_2, A_2 are the percentage abundances and mass numbers of the contributing isotopes and σ_1 and σ_2 are the individual cross-sections for ^{63}Cu(α, n) and ^{65}Cu(α, 3n) reaction respectively.

3.1 *^{63}Cu(α, n) + ^{65}Cu(α, 3n) ^{66}Ga and ^{65}Cu(α, 2n) ^{67}Ga reactions*

Figure 1 shows the present experimental results together with the previous measurements by a large number of investigators (Porges *et al* 1956; Porile and Morrison 1959; Bryand *et al* 1963; Graf and Munzel 1974; Rizvi *et al* 1987). Most of the work

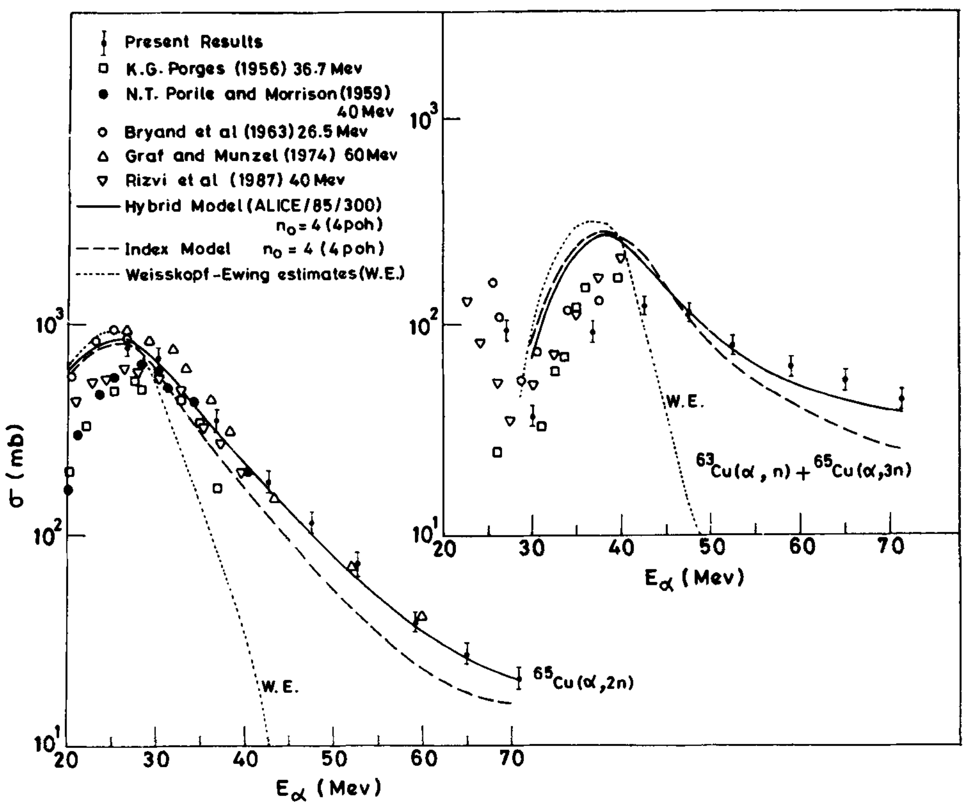

Figure 1. Excitation functions of ^{63,65}Cu(α, xn) reactions.

on this element performed during 1956–74, was carried out employing G. M. counters, scintillation detectors, proportional counters for the measurement of cross sections. Porges (1956) measured these reactions using G. M. counter with an uncertainity of 25%. Porile and Morrison (1959) measured them employing a scintilation counter. A further measurement was done by Bryand *et al* (1963) using a proportional counter. Because of the poor resolution of these detectors, there were many inconsistencies in the data as the closely lying gamma rays of the reaction residues could not be separated. During the past twenty years semiconductor radiation detector has become one of the principal tools in studies requiring high resolution measurements of gamma rays. Thus it was in 1974, the first Ge(Li) measurement was carried out by Graf and Munzel (1974) up to 60 MeV and very recently by Rizvi *et al* (1987) up to 40 MeV. It may be observed in figure 1 that while there is a fair agreement between the present measurements and those of Graf and Munzel (1974), there are some inconsistencies between the present results and those of Rizvi *et al* (1987) who also employed a similar experimental technique as that of ours. The reason for observed anomaly, to our belief, mainly lies in different methods employed in determining the alpha particle flux and possible differences in gamma efficiencies used in the calculation of cross-sections in the two works. Rizvi *et al* (1987) have monitored the alpha particle flux by charge collection method.

Although in principle the Faraday cup is a simple device, several sources of error may be present. For example, the region of the cup in which the beam stops will be a source of secondary electrons which, if they escape will make the beam intensity overestimated. In view of the many problems involved in the direct measurements of the flux through charge collected in a Faraday cup, it is customary to prefix the experimental stack-foils with an aluminium monitor foil and to study the reactions as mentioned earlier in experimental procedure.

It is appropriate here to mention that the present experimental study adds two new energy-point cross-sections in ^{65}Cu(α, 2n) and six new energy-point cross-sections in ^{63}Cu(α, n) + ^{65}Cu(α, 3n) in the energy range 45–75 MeV. Although for the later reaction, the determined values were weighted average cross-sections, in this energy region essentially reflect the behaviour of the dominant ^{65}Cu(α, 3n) reaction.

3.2 ^{181}*Ta*(α, 2n) ^{183}*Re and* ^{181}*Ta*(α, 4n) ^{181}*Re reactions*

These reactions were previously studied by Scott *et al* (1968) employing Ge(Li) as well as scintillation detectors. Later, using Ge(Li) and HPGe detectors, Hermes *et al* (1974) and Ismail and Divatia (1988) studied these reactions. The quoted uncertainties in the measurements of Hermes *et al* were $\simeq 19\%$. But there are discrepancies by a factor of 2–6 among the results of the three groups. It can be seen from the figure 2 that in the case of ^{181}Ta(α, 2n) reaction, there is an excellent agreement between the present results and those of Ismail *et al* all over the region from 30–48 MeV. Beyond that *three* new energy point cross-sections were added in the present work. Similarly, in the case of ^{181}Ta(α, 4n) reaction it can be seen that there is a wide scatter in the measurements between Hermes *et al* and Ismail *et al*. There is an unexplainable overall discrepancy in their results. While the values of Hermes *et al* are lower by at least a factor of two than those of Ismail *et al* for ^{181}Ta(α, 2n) reaction, it is just reverse in ^{181}Ta(α, 4n) reaction. However, our results are in close proximity with those of Ismail *et al*.

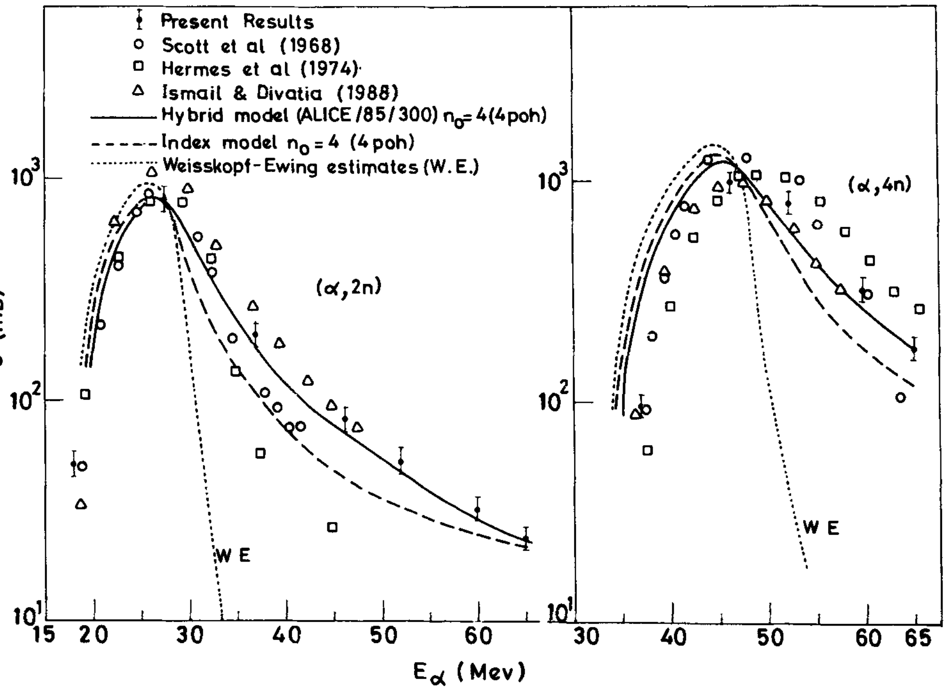

Figure 2. Excitation functions of ^{181}Ta(α, xn) reactions.

4. Comparison with theoretical calculations

The present experimental results are compared with theoretical predictions based on Weisskopf-Ewing estimates giving the compound nucleus contribution as well as with the hybrid and index models in their latest forms. Owing to the semi-classical nature, the hybrid and the index models, involve a large number of physical parameters as well as a few adjustable parameters. The model predictions of course depend on the input values given to these parameters. Also, to enable a relative comparison, on the performance of the two models, the same set of input parameters were applied in the calculations. A short description of the option chosen is given below. The nuclear masses were calculated from the Myers-Swiatecki mass formula (Myers 1977) considering the liquid drop with the 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 codes. A level density parameter of A/8 MeV^{-1} was used. In both the pre-equilibrium models it is customary to use the initial exciton number, $n_0(p_0h_0)$ as a fit parameter to match the theoretical predictions with experimentally observed shapes of spectra and excitation function. The initial exciton number governs the entire cascading process of binary collisions and thereby influences the shape of the hard component in particle spectra. A good guess would be the number of nucleons in the projectile or an additional particle-hole if need be (Ernst *et al* 1989). This view is quite consistent with the basic physics of pre-equilibrium phenomenon that only a small number of degrees of freedom is initially excited in nuclear reactions at moderate energies. So, we have made the theoretical calculations using $n_0 = 4$ (4poh),. $n_0 = 5$ (5poh) and $n_0 = 6$ (5p1h) as the initial configurations. It was found that for both the hybrid and the index models $n_0 = 4$ (4poh) by far gives the good results than the other two in both the models. The predictions of the (5poh) and (5p1h) configurations were lower than those obtained with (4poh). Therefore we have shown (4poh) configuration in both the models for comparison.

Figures 1 or 2 show the comparison of experimental excitation functions of $^{63}Cu(\alpha, n) + ^{65}Cu(\alpha, 3n)$, $^{65}Cu(\alpha, 2n)$, $^{181}Ta(\alpha, 2n)$ and $^{181}Ta(\alpha, 4n)$ reactions with the theoretical calculation based on Weisskopf-Ewing estimates giving compound nucleus contributions as well as with hybrid and index models. It is found that the Weisskopf-Ewing estimates accounted well for the low energy compound nucleus dominated part of the excitation function of all the four reactions, but fail to account for the observed cross-sections at high energies where non-equilibrium effects predominate beyond a few tens of MeV of bombarding energy. The prediction of hybrid model using (4poh) configuration are fairly good in the cases $^{181}Ta(\alpha, 2n)$ as well as $^{65}Cu(\alpha, 2n)$ especially at the high energy tails of the excitation functions of these reactions, whereas the index model predictions are slightly lower than the experimental results, but this cannot be a reflection on their relative abilities to predict pre-equilibrium particle emission. The apparent differences between the two theoretical predictions in all the four multineutron emission reactions, is largely ascribable to the conceptual and computational differences between the two models. Both, hybrid and index models in their present forms take into account the multiple chance pre-equilibrium emission of nucleons only up to two nucleons and there is also a difference in the way this aspect is treated in both the models. While in hybrid model it is assumed that both the nucleons are emitted from the same exciton heirarchy, the index model assumes that the exciton configurations may in general be different. However, since the probability of multiple pre-equilibrium nucleon may not be so high at these energies of the order of 60–70 MeV, the above subtle difference between the model predictions is unlikely to be discerned. Further, as can be seen in the case of $^{63}Cu(\alpha, n) + ^{65}Cu(\alpha, 3n)$ and $^{181}Ta(\alpha, 4n)$ there is a slight shift in the energy between the theoretical and experimental compound nucleus peaks. Generally such shifts are ascribed due to the complete neglect of angular momentum effects in the Weisskopf-Ewing theoretical calculations provided in the codes. More elaborate computations, using Hauser-Feshbach theory may bring about a better agreement. Further, the compound nucleus mechanism dominates up to about 55 MeV. The comparison of present results with the predictions of hybrid and index models as can be seen from figures that given the small role played by the pre-equilibrium effects in these multinucleon emission reactions, the performance of both the models is alike and consistent with the initial exciton number $n_0 = 4$ (4poh) generally.

5. Conclusions

Excitation function for four reactions of (α, xn) type were studied in the present work. The present experimental results are systematic and accurate over many of the previous measurements. From an overall comparison between experimental results and theoretical predictions based on compound nucleus Weisskopf-Ewing estimates as well as pre-equilibrium hybrid and index models, one can infer that the Weisskopf-Ewing estimates accounted well for the low energy compound nucleus dominated part of the excitation functions of all the four reactions, but failed to account for the observed cross-sections at higher energies, where non-equilibrium effects predominate beyond a few tens of MeV of bombarding energy. This is rather indirectly indicated by the "high energy tails" of the experimental excitation functions which signify a less rapid fall of the cross-sections than predicted by compound nucleus model. The shape of the excitation functions in the pre-equilibrium dominated regions of energy is reproduced by the latest improved version of hybrid model as well as the recently proposed index model. The former is a shade better. The basic tenet that only a small number of degrees of freedom in pre-equilibrium reactions is borne out by the consistently reasonable agreement observed between experiment and model predictions for an initial exciton number $n_0 = 4$ (4poh). There is a subtle difference in the viewpoints of hybrid and index models with regard to the possible initial exciton configurations. While the hybrid model tends to fit the experimental data with a single initial exciton configuration for all the reactions induced by same projectile, the index model, harbours the view that several initial exciton configurations are in general possible, for a given projectile, and the experimental results are just the statistical averages over all such possible initial exciton configurations. The physical interpretation of $n_0 = 4$ (4poh) is that, only four excitons initially share the excitation energy and that these are particles above a completely filled Fermi sea. Such an initial configuration is quite consistent with an alpha particle projectile as the four nucleons of the alpha particle are the most likely candidates to share the excitation energy.

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