

CHAPTER I

INTRODUCTION

The study of nuclear reactions is directed at the exploration of the laws governing the interaction between the nucleons and nuclei. The partial successes of the shell model and the liquid drop model in explaining different properties of nuclei, led to the formation of two scenarios (i) the direct reaction model which assumes weak or no interaction between nuclear nucleons as in the shell model and (ii) the compound nucleus model which assumes nucleons as in the liquid drop model. According to the first view, the nuclear reaction should be completed within a time of the order of 10^{-21} sec, which it takes the incident particle to cross the nuclear diameter. However, the observation of slow neutron resonances indicating a such longer reaction time of the order of 10^{-16} sec, has led to the formulation of the second model by Bohr in 1936 /1/. The compound nucleus model derives its name from the fact that the reaction is assumed to take place in two independent steps, the formation and decay of a “ long-lived ” intermediate stage called the compound nucleus. The direct and compound nuclear reactions could explain the spectral features of the reaction products at rather low energies till the development of Variable Energy Cyclotrons. With the advent of these machines, a third type of reactions, namely the precompound or preequilibrium reactions came to light.

However, a close look at the two traditional mechanisms reveals that they constitute two extreme ends on the “ time of interaction ” scale. Because of the short time of interaction, direct reactions are important as sources of information on nuclear structure /2-4/. The reaction is completed in one step, the reaction amplitudes depend on the overlap of the initial and final states and hence direct reaction cross section tells us directly about the relation between initial and final states. On the contrary, compound nucleus reactions do not reveal anything about the structure. Here, the decay of the compound nucleus is almost, independent of the formation and the direct overlap of initial and final states does not happen. The two

models have different kinematics characteristics too. A direct reaction favours the transfer of only relatively small amounts of energy to the final nucleus and readily feeds the low lying excited states which are well separated. Hence, the emitted particles in direct reactions appear in discrete peaks. In contradistinction, in compound nucleus reactions / 5-8 / particles are evaporated with small energies and therefore feed the higher excited states in the continuum of the residual nucleus. Hence, the evaporation spectra are continuous and Maxwellian. Further due to the small time separation between entrance and exit channels, the direct reactions are coherent and so the angular distributions of the ejectiles are typically diffraction - like. On the contrary, due to the relatively long time separation between initial and final stages of compound nucleus reactions, they are said to be incoherent, with the emitted particle angular distributions being either symmetric or at least fore and aft symmetric. A third distinguishing feature between the two types of mechanisms is provided by the fact that compound nucleus evaporated particles are completely unpolarised where as direct reaction ejectiles are completely or partially polarised.

The advent of accelerator technology and break-throughs in nuclear radiation detectors have brought a sea change in the scenario of nuclear reactions. There was increasing evidence for experimental features deviating far away from any one of the above described traditional reaction characteristics. This pointed out to reaction modes, intermediate in time scale as well as in the number of degrees of freedom, as compared to the compound and direct processes. The new characteristics of these intermediate processes are

(i) The high energy tail in the excitation function of variety of reactions in the energy range 20 - 200 MeV (less rapid decrease of cross section with energy than expected from the compound nucleus theory)

(ii) The continuous, but non-Maxwellian, spectral shapes of angle integrated spectra and lastly

(iii) The forward peaked but not diffraction-like, angular distributions of the emitted particles.

Thus, in the comprehensive view expressed by P.E.Hodgson /9/, a nuclear reaction is not one-step or two-step process but in general a multi-step process, the reaction may end at any time starting from one step direct to many step compound process. If the interaction ends at the first collision, it is a direct reaction or if it ends at the last stage, it is a compound nuclear reaction, and hence the termination at intermediate stages comprises the new and third type of reactions named as “preequilibrium” or “precompound” reactions / 10-13 /.

Thus it became evident that the preequilibrium reactions occupy a place intermediate between direct and compound nucleus reactions. As such their characteristics are also in between the characteristics exhibited by direct and compound nucleus reactions. As the preequilibrium particles are emitted, when the composite system is moving towards statistical equilibrium, the energy and momentum brought in by the incident particle is shared between relatively fewer degrees of freedom and consequently the energy available to each degree of freedom is comparatively large. So the particles will be emitted with higher energy than evaporated particles. This qualitatively explains the non-Maxwellian distribution of preequilibrium particle spectrum and the high energy tail of the excitation function. Also in the initial stages of the precompound process, the composite nucleus retains a partial memory of the incident direction. That is why the emitted particles show a forward peaking in the angular distributions.

To explain the newly observed preequilibrium phenomena, many theoretical models have been proposed. The first model that appeared in 1966 was the Statistical Model of Intermediate Structure (SMIS) of Griffin / 14 /, which explained the shape of the emitted neutron spectrum in $^{117}\text{Sn}(p, n)^{117}\text{Sb}$ reaction /15,16/ at 14 MeV proton energy. In this model the equilibrating system as a whole was considered, envisioning it to pass through increasingly complex intermediate configurations of single particle excitations called “door way” states. This model has been the fore-runner of all subsequent preequilibrium models, the basic idea being that the equilibration of composite system achieved due to the series of two body collisions between projectile and target nucleus. Each intermediate state is characterised by the number of excited particles and holes defined with respect to the Fermi energy. They are together called as excitons. For the sake of simplicity, however, it is assumed that at each stage, all possible single particle configurations are equally likely so that the occurrence of configurations capable of particle emission into continuum may be estimated on a statistical basis. For each exciton number, some fraction of states will have particles that are unbound and thus have a finite probability for emission. The preequilibrium decay probability is computed as the sum over contributions from states from initial exciton number n_0 to a final exciton number \bar{n} . However, this model which is also called Griffin’s Exciton model, did not explicitly treat the competition between particle emission and intranuclear collisions and so it could not predict absolute cross section. Griffin, however made no attempt to calculate the matrix element for two body interaction or study its energy dependence, to make the model more quantitative. The crucial parameter in this model is the initial exciton number which governs the entire cascading process of binary collisions and thereby influence the shape of the particle spectrum.

Another approach to describe the nuclear equilibration is given by Harp, Miller and Berne in 1968 /17/ by proposing “Fermi-gas equilibrium model” which is widely known as HMB model. In this model, the nucleus is considered as a Fermi gas and the available single particle levels are grouped into energy bins of certain width. The fractional occupation of each bin is followed as a function of time. The particle flux is divided in proportion to the rate at which it makes an internal two body collisions. Redistribution of population is governed by a master equation which describes the relaxation process until a steady state condition is reached. The salient feature of the model is that it provides a unified description of the reaction process, with a smooth and natural transition between the preequilibrium and equilibrium phases. Further, since the two body collision rates are calculated from free nucleon - nucleon scattering cross sections, the HMB model makes quantitative predictions. However, because of the computational complexity, this model is not very widely applied and also it does not predict the angular distribution.

In later years, Blann /18/ proposed the Hybrid model in 1971 which combines the advantageous features of HMB model and Griffin’s Exciton model. As in the Griffin’s model the nuclear states are classified according to the number of excitons as far as the equilibration sequence is concerned. But with regard to particle emission, in each class of states, the energy distribution of excited particles is considered and the flux is divided according to the branching ratios of continuum emission relative to the creation of particle-hole pairs such as in the HMB model. The intranuclear transition rates are determined from free nucleon-nucleon scattering cross sections as in the HMB model. A further refinement is named as the Geometry Dependent Hybrid (GDH) model /19/ in which effects of interaction in the diffuse nuclear surface have been considered. In Hybrid model the nuclear matter density is

taken as uniform while in GDH a reduced matter density is taken at the nuclear surface.

Further refinement of the Griffin's Exciton model was done by Gadioli et al /20/ in 1973. They have calculated the intranuclear transition rates within the framework of a two component Fermi gas model, using free nucleon-nucleon cross section. However, the rates had to be multiplied by an arbitrary factor to reach agreement with experiments.

The original Exciton model has been extended by a number of authors Bragg-Marcuzzan /21/, Kalbach /22/ and Gadioli et al /23/. Also there have been some ideological differences between the Hybrid model and Exciton model which were later resolved by several authors Blann /24/, Gadioli et al /25/, Ernst and Rama Rao /26/, Bisplinghoff /27/ and Machner/28/.

All these models have the common feature that they group the many body states of the equilibrating system according to exciton numbers and employ particle hole densities to estimate the occurrence of configurations capable of precompound particle emission. These models treat the competition between particle emission and intranuclear transitions during the equilibration phase explicitly and evaluate the rates at which such intranuclear transitions take place. The particle emission rates into continuum are calculated from reciprocity theorem while the internal transition rates are derived either from free nucleon-nucleon cross sections or from imaginary part of the optical potential. In Kalbach's model, the matrix element for internal collision is treated as a fit parameter in a semi-empirical way.

All the above models, being phase space models, can predict only the angle integrated particle spectra but not the detailed angular distribution of the emitted

particles. However, there are two models based on the Monte Carlo method of calculation, which predicts energy and angle correlated particle spectra (i) the intra nuclear cascade model /29/ originally used for high energy direct interactions, but later applied to preequilibrium reactions, without real justification. (ii) the Generalised Master Equation model of Mantzouranis et al /30/ using the “fast particle” concept. Later on, Random-walk model precompound decay proposed by Akkermans et al /31/ using the “random walk approach”. Further De et al /32/ have explained detailed angular distribution of emitted particles to some extent.

While nucleon emission at high excitation is understable in terms of preequilibrium decay, the emission of complex particles, such as α -particles from excited nuclei remained as an enigma. Even in the case of well understood compound nucleus ‘evaporation’ of α -particles, there remains the unsettled question as to whether the emitted α -particles were pre-existent in the nucleus or not. While some theorists prefer to have a “pre-formation” probability factor, others content that the α -particle could be formed at the time of emission. The model for preformed clusters was proposed by Millazzo Colli and Braga-Marcazzan /33/ in an attempt to describe spectra of (n, α) reactions. In the context of preequilibrium decay, the α -particle emission is considered by some authors /34-35/ as ‘correlated emission’ of four nucleons, two protons and two neutrons having their momenta directed in such a way as to correspond to the Fermi momenta of these nucleons in an actual α -particle, i.e. helium nucleus in ground state. To put it in another way, it may be said that, in momentum space, the momenta of the four nucleons are contained within a sphere of radius P_0 , where P_0 called the “Coalescence radius” is of the order of Fermi momentum for α -particle, i.e. helium nucleus in its ground state.

Machner proposed “Exciton Coalescence model” /36/ in 1979 by combining the ideas of Generalised Exciton model and Coalescence model /37/. However, from a theoretical point of view it is rather difficult to calculate accurately the probability factors involved in such points of view.

Despite various improvements from time to time, all the above preequilibrium models still remained semi-classical or phenomenological, lacking in a quantum mechanical base. To make up for this, in 1980 Feshbach, Kerman, and Koonin / 38/ have proposed a rigorous nuclear reaction theory with quantum mechanical basis known as “ Multistep Direct and Multistep Compound ” formalism.

In 1981, Kalbach and Mann /39/, in the spirit of Feshbach, Kerman and Koonin model, modified the Exciton model to calculate the preequilibrium cross section in two parts namely Multi Step Direct (MSD) and Multi Step Compound (MSC) cross sections. For this purpose, the partial level densities are classified into two categories namely bound and unbound states. Using the angle integrated cross section and appropriate legendre polynomials, they proposed a heuristic formula for the angular distributions, based on the trends of existing experimental data.

Summarising the theoretical developments it may be said that, by and large, the semi-classical preequilibrium models are more often used because of their simplicity and transparency. Although, these models embody few of the details of nuclear structure, they employ more general properties of nuclei such as the mean free path of nucleons in nuclear matter, density of particle-hole states at varying excitations, effects of nuclear geometry and emission of particles from highly excited nuclear systems. In short, these are ‘nuclear matter’ calculations. By means

of closed form analytical expressions they give fairly accurately the angle-integrated particle spectra from which the integral cross section at each bombarding energy, as well as the variation of cross section with energy, can be readily obtained in the form of theoretical excitation function. Further, useful extensions such as multi-particle emission and multiple chance preequilibrium emission which becomes important at high energies can be easily incorporated into these models.

Blann, who is continuously improving his computer codes on Hybrid model /18/ [ALICE (1972), OVER LAID ALICE (1976), ALICE / 82 (1982) etc], in 1983 introduced new algorithms into the code to calculate multiple chance preequilibrium emission of nucleons in an approximate way/40/. This code is known as ALICE/85/300 /41/. If this code is utilised for a multi-particle emission reaction, then, only the first two nucleons are considered to be emitted in preequilibrium phase, rest of the nucleons and particles are treated by evaporation calculation, provided in the code. Later on, Kataria et al /42/ incorporated a shell dependent level density formula due to Kataria, Ramamurthy and Kapoor (KRK) /43/, in the above code. This version of code is known as ALICE/90. This model relates the shell effects in the nuclear level densities to the shell correction term of the nuclear mass surface, which is to be tested against experimental data. It is appropriate to mention here that above code is designed for the emission of two or fewer precompound nucleons from each nuclide in preequilibrium phase. As precompound models have been extended to higher energies, this constraint has become an ever more serious limitation. To overcome this problems a new formulation has been made by Blann in 1996 /44/, which uses only the kinematically justified two and three exciton densities /41/ and which allows unlimited precompound emission from each nuclide. The new approach is a Hybrid Monte Carlo Simulation (HMS) model.

It is valid upto the effective pion threshold, around 280 MeV. It has been installed as an option in the ALICE nuclear reaction code, in a version called "HMS-ALICE".

One of the motivation for the present work is to study experimentally the probabilities for the emission of nucleons and complex particles from excited light, medium and heavy nuclei formed in α -induced reactions and to make a comparative study with the predictions of theoretical models such as the simple Weisskopf - Ewing estimates of the compound nucleus model and of preequilibrium Hybrid model (ALICE/90).

It is appropriate to list here some of the motivations, aside from those given in the preceding paragraphs, which led to the initiation of this experimental programme. Two methods are generally employed for the determination of reaction cross section. These are (i) the integration technique and (ii) the activation technique. In the former method, observations are made on the angular distribution and energy of the particles emitted while irradiating the targets with incident particles (On-line study). The particle spectra are integrated to obtain the cross section. One difficulty of this method is in obtaining spectra at very forward angles which are important from the view point of integration. The activation technique on the other hand, involves the irradiation of a target with incident particle beam, followed by the identification and quantitative estimation of the induced radioactivity of the residual nucleus (Off-line study) using a beta or gamma ray detector. From the measured activity, the cross section is computed. Infact this method is usually preferred because of its convenience coupled with accuracy and is used in majority of the experimental investigations.

During the last four decades, techniques of measuring the activation cross sections have steadily been improved with an eye on accuracy. Thus the earlier Geiger Müller counter for beta rays detection, has given place to gamma detection by NaI (TI) detector, which is subsequently replaced by the more versatile Ge(Li) and HPGe detectors. The earliest beta measurement using a Geiger Müller counter resulted in too high cross sections since the contributions of interfering reactions could not be effectively separated even if enriched targets were used. Some of the difficulties with beta counter could be circumvented by using a NaI(Tl) scintillation spectrometer to measure the induced gamma activities in the irradiated materials. Though the efficiency is good, the NaI (TI) system does not have adequate resolution to resolve closely lying gamma ray peaks. In addition, the analysis of gamma ray spectra recorded on a NaI(Tl) spectrometer is associated with considerable errors due to the presence of large Compton background, back scattering and escape peaks.

The Ge(Li) and HPGe detectors have exceptionally good energy resolution (2.0 keV FWHM for 1332 keV photons of ^{60}Co) and very low Compton background. Hence even closely lying gamma rays appear as prominent and well resolved photo peaks. So, it is possible to sort out the interfering contributions due to unwanted reactions even without resorting to chemical separation technique. It is also noteworthy that with the advent of Ge detector there has been an improvement in the quality of the spectroscopic data needed for the determination of the reaction cross sections. The γ -ray energies, abundances and half-lives of radioactive isotopes have undergone a revision in recent years and extensive and accurate tabulations /45-46/ have become available now. These developments also partly motivated the present work in which high resolution Germanium detectors were used for gamma ray detection and the latest spectroscopic data were employed in deducing the cross

sections. However, this was done only in a few cases as can be seen from the following survey of literature.

The choice of target element is made on the basis of a survey of literature, keeping an eye on the experimental accuracy. A survey of literature /47-60/ reveals that there are large uncertainties or mutual discrepancies among the reported values for the same reaction measured by different investigators. Also in some cases the measurements were done with poor resolution detector such as scintillation detector. Hence in the present work, we have selected target elements gold, antimony, indium and iron for systematic experimental study to improve the quality of data using high resolution HPGe detector and to test the preequilibrium model.

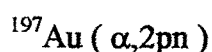
Rather large experimental data are available in literature for alpha induced reactions on gold /47-50/ using stacked foil activation technique. Lanzafame and Blann /47/ used both NaI crystal and Ge(Li) detector for measurement of gamma activities and the reported uncertainty in their measurement was quoted as 20%. Kurz et al /48/ studied reactions up to 43 MeV employing a small volume (7cc) Ge(Li) detector with a resolution of 4.2 keV for 1332 keV photons of ^{60}Co . The experimental errors in the cross section were mentioned as less than 10%, but it is not mentioned whether or not this includes the uncertainty in the monitor cross section used for flux measurement as well as the errors in spectroscopic data. Capurro et al /49/ measured the reactions upto 55 MeV using an intrinsic Ge-detector. The overall error in their measurement varied in the range 24% to 37%. Further the previous measurements, however do not agree with one another in the overlapping energy-region. Bhardwaj et al /50/ measured the reactions upto 40 MeV using HPGe detector. The reported values of Capurro et al and Bhardwaj et al differ by more than 50% around 30 MeV and deviation is large at high energies.

Indeed, rather extensive experimental data are available in literature for alpha induced reactions on natural antimony /51-54/ using Ge detector. Calboreanu et al /51/ studied only $^{121}\text{Sb}(\alpha,n)$ and $^{121}\text{Sb}(\alpha,2n)$ reactions upto an alpha particle energy of 27 MeV. Ismail /52/ reported the alpha-induced reactions on antimony upto 58 MeV with an error of 8%. Singh et al /53/ measured these reactions upto 60 MeV with an error of 10%. Bhardwaj et al /54/ studied these reactions upto 55 MeV with an error of 10 %. In view of the mutual discrepancies in the cross section values for the same reactions, a reinvestigation of the above reactions was under taken to improve the quality of experimental data. Since natural antimony used as the target has two odd mass stable isotopes of abundances 57.3%(^{121}Sb) and 42.7%(^{123}Sb), their activation in some cases gives the same product nucleus through different reaction channels, but with very different Q-values. In such cases, the individual reaction cross sections are separated with the help of theoretical cross sections.

However, only a few experimental data concerning alpha induced reactions on indium exist in the literature in low energy region. A reaction $^{115}\text{In}(\alpha,n)^{118\text{m}}\text{Sb}$ was studied earlier by Hansen and Stelts /55/ in the energy range 12 to 18 MeV using poor resolution detector. Bhardwaj et al /56/ measured α -induced reaction upto 50 MeV using Ge(Li) detector. Mukherjee et al /57/ also studied α -induced reactions upto 50 MeV using Ge(Li) detector with an error of 9-14%. A comparison of experimental data of Bhardwaj et al and those of Mukherjee et al shows that the shapes of their excitation functions are quite similar but absolute cross sections differ by a factor of two or more in high energy region. Hence, in view of large discrepancies, a reinvestigation of alpha induced reactions on indium was under taken to improve the quality of experimental data.

The study of α -induced reactions on iron is a case in point for mutual discrepancies among the earlier results obtained using different detecting system /58-60/. Tanaka et al /58/ studied these reactions upto 40 MeV using a GM counter and a scintillation counter with an error of 16%. Ewart et al /59/ studied these reactions upto 68 MeV employing proportional counter and scintillation detector using enriched ^{56}Fe for $E_\alpha > 43$ MeV and natural iron for $E_\alpha < 43$ MeV with an error of about 20%. Vedoya et al /60/ studied these reactions upto 85 MeV using a Ge detector with an error varying from 3 to 16 %. The reported values of Vedoya et al and Tanaka et al differ by more than 50% and 24 % respectively to Ewart et al. Further in some cases /58,59/, the cumulative yield due to the decay of all isobars of the reaction products was measured and no attempt was made to separate them analytically or otherwise. Hence in the present work a detailed mathematical formalism is developed /61/ from first principles to separate out exactly, point by point the isobaric precursor contributions from the measured inclusive excitation function so as to obtain the individual excitation function relevant to the particular reaction under study. This will enable a direct comparison of the theory and experiment for a single specified reaction.

In the present experimental study, the available maximum energy of 50 MeV alpha particles from an indigenous Variable Energy Cyclotron situated at Calcutta, has been used. The versatile stacked foil activation technique and gamma counting method are employed, using high resolution HPGe detector to identify the characteristic gamma rays of the residual nuclei. The excitation functions of the following twenty three reactions are measured:



$^{197}\text{Au}(\alpha, \alpha n)$
 $^{121}\text{Sb}(\alpha, xn); x=1,2,4$
 $^{123}\text{Sb}(\alpha, xn); x=3,4$
 $^{121}\text{Sb}(\alpha, p3n)$
 $^{121}\text{Sb}(\alpha, \alpha n)$
 $^{123}\text{Sb}(\alpha, \alpha 3n)$
 $^{115}\text{In}(\alpha, xn); x=1-3$
 $^{115}\text{In}(\alpha, 2p)$
 $^{113}\text{In}(\alpha, n)$
 $^{56}\text{Fe}(\alpha, xn); x=3,4$
 $^{56}\text{Fe}(\alpha, pxn); x=1-3$

The experimental results measured in the present work are compared with theoretical predictions based on the Hybrid model of Blann (code ALICE/90) to test the underlying Physics of the model and judge their limits of validity. Details of the work are given in the ensuing chapters.

Chapter II. This chapter contains an outline of the statistical theory of nuclear reactions and later developments in preequilibrium decay formalism.

Chapter III. This chapter deals with the experimental technique, brief description of the apparatus used such as detector system and its calibration, data collection and reduction.

Chapter IV. This chapter embodies the present experimental results of nuclear reaction cross sections and their comparison with literature values wherever available.

Chapter V. This chapter is devoted to a detailed comparison between the experimental results and the theoretical calculations based on preequilibrium Hybrid model and assess the relative merits and limitations of the Hybrid model.

Summary and conclusions are given at the end.

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