Measurement and analysis of isomeric cross section ratios in the reaction ^{93}Nb(α, $2n$)^{95}Tc: Pre-equilibrium reaction mechanism

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Isomeric cross-section ratio and excitation functions have been measured for ^{93}Nb(α, $2n$)^{95}Tc^{m+g} reactions up to 120 MeV. The ratio and the excitation functions were interpreted in terms of the theoretical results obtained by two models COMPLET and STAPRE, which include preequilibrium effects. The experimental results were fairly well reproduced by the COMPLET calculations, which take into account the angular momentum removal in preequilibrium emission in an approximate way. Conversely, STAPRE calculations show a better agreement for isomeric ratio, but the experimental excitation function is underestimated by almost a factor of 2.

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I. INTRODUCTION

The study of excitation functions (EF) and isomeric cross-section ratio (ICR) of nuclear reaction is a significant tool for testing nuclear models. Furthermore, the cross-section ratio for the formation of the same pair of isomeric states of the residual nucleus at various beam energies is of prime importance for studying the effect of nuclear spin [1]. The spin distribution of the compound system is dependent on the angular momentum brought in by the projectile that changes with its energy. The spin distribution finally changes as a result of particle emission followed by γ deexcitation leading to the formation of the residual nucleus.

As the projectile energy increases by several tens of mega-electron-volts, the preequilibrium particle emission becomes a significant factor [2]. As a result the emitted particles in preequilibrium mode have higher energy and carries higher angular momentum as compared to the particles evaporated from a compound nucleus. Thus removal of a substantial amount of angular momentum from the excited composite system will have a strong impact on the yields of isomers having an appreciable amount of spin difference. The EF and ICR have been an object of theoretical estimations with several computer codes. The trustworthiness of these estimations strongly depends on the correct understanding of the underlying nuclear processes. However, the shortcomings of the theoretical models become more clear when compared with the absolute cross sections.

In this scenario, the present investigation is undertaken with two main objectives. (i) To make a careful and systematic measurement of excitation functions of α-particle-induced reactions on niobium up to 120 MeV, leading to the formation of the residues ^{95}Tc^{m} and ^{95}Tc^{g} and thus measuring the ICR. (ii) To make theoretical interpretation based on COMPLET [3,4] and STAPRE [5,6] models for preequilibrium emission leading to the formation of these radioisotopes.

II. EXPERIMENTAL PROCEDURE

The experiments for the measurement of ICR of the α-particle induced reaction on ^{93}Nb ware carried out at the IUCF-USA cyclotron facility and at the Variable Energy-Centre (VECC; Kolkata, India). In the present experiment, an activation technique followed by offline γ-ray spectrometry was used for the measurement of excitation functions and ICR of evaporation residues (ER) ^{95}Tc^{m} and ^{95}Tc^{g}. In this method a stack of foils with energy degraders is irradiated in a fixed geometry in such a way that each successive foil of the stack are irradiated at decreasing incident energies.

Two consecutive α-beam irradiations were carried out for the energy range 120 to 50 MeV on two separate stacks of foils, each of about 2h duration. To check the consistency of the data collected, sufficient overlapping energy region was taken. A third set of irradiation in the energy range 50 to 17 MeV was done at the VECC (Kolkata, India). Spectroscopically pure (99.99%) niobium (Nb) targets (21 mg/cm^2) and aluminum catchers (7 mg/cm^2)/degraders (7 mg/cm^2) were obtained from Goodfellow Metals (Cambridge, UK). The beam energy degradation in target foil was calculated using the stopping power tables of Northcliffe and Schilling [7]. An electron suppressed Faraday cup placed behind the target-catcher assembly was used to measure the integrated beam current every minute to correct for the variation in the beam intensity during the irradiation time. Beam currents of the order of 120 nA were used. For the flux measurement, the ^{27}A1(α, $\alpha 2pn$)^{24}Na reaction was used as a secondary standard [8].

A. Calculation of cross-section and isomeric cross-section ratios

After the irradiation each target-catcher assembly was counted for the γ-ray activity of evaporation residues on an

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TABLE I. Spectroscopic data of the product nuclei $^{95}Tc^{m+g}$ taken from Ref. [9] for α-induced reaction on ^{93}Nb.

Nuclide	Half-life	Spin	E_γ (keV)	I_γ (%)	Reaction	Q value (MeV)
$^{95}Tc^m$	61 d	1/2-	204	66.2	$^{93}Nb(\alpha,2n)^{95m}Tc$	−14.914
			582	32.5		
			835	28.1		
$^{95}Tc^g$	20 h	9/2+	765	99.82	$^{93}Nb(\alpha,2n)^{95m}Tc$	−14.914
			948	2.1		
			1074	4.2		

HPGe detector coupled to a 4K channel analyzer. The energy and efficiency calibration of the detector was performed with a standard 152Eu source. The count rates were corrected for random pileup losses (using a pulse generator as reference) as well as for γ-ray abundance and the efficiency of the detector. In the reaction $^{93}Nb(\alpha, 2n)^{95}Tc^{m+g}$ the most important decay data of the product nuclides $95Tc^m$ and $^{95}Tc^g$ are summarized in Table I. Both the isomers decay almost independently of each other except for a weak 4% isomeric transition (IT) from 61 days isomer to 20 h ground state. Nuclear data on radioisotopes, such as their γ-ray abundances and half-lives, were taken from the compilation of Firestone and Shirley [9]. The cross section (σ) of the production of the radioisotopes was calculated from the measured peak areas, $PA(t)$, using the following equation:

$$PA(t) = N\sigma \sum_{i=1}^{n} I_i(1 - e^{-\lambda\Delta t})e^{-\lambda(T_i - \sum_{i=1}^{n}\Delta T_i)}e^{-\lambda t}a_\gamma\varepsilon_\gamma$$

where N is the number of target atoms per cm^2, I_i is the beam intensity in the i^{th} interval Δt_i, λ is the decay constant of the radioisotope, T_i and t are the time of irradiation and time elapsed after the end of irradiation, and a_γ and ε_γ are the abundance and detector efficiency for particular γ-ray energy. The isomeric cross-section ratio could be determined more simply from the growth and decay curve measurement of the ground-state activity.

B. Experimental uncertainties

The major sources of errors involved were those associated with the measurement of the beam current and determination of the absolute activities of the products. The error in the α-beam flux was around 4–7%. The efficiency of the γ detector was known with in an uncertainty of 3–5%. The error in the initial count rate at the end of the irradiation as determined by the least squares fitting procedure was about 0.5–2.5%. The error in the decay data used was <1% and the error in the target thickness was 0.5–1.5%. The total error in each cross section was obtained by combining the individual errors in quadrature. The absolute errors for the isomeric cross-section ratios are smaller because the errors because of target thickness, beam flux, do not enter into the isomeric cross-section ratio calculation.

III. EXPERIMENTAL RESULTS

In the present study, the excitation functions of the radionuclide ^{97}Rh formed in $^4He + ^{93}Nb$ reaction was measured. The cross sections of $^{95}Tc^m$ and $^{95}Tc^g$ were measured by selecting the most abundant γ-rays from Table I. This reaction was previously studies by Bond and Jha [10] in 1970 up to 40 MeV, Branquinho *et al*. [11] up to 50 MeV in 1979, Ernst *et al*. [12] in 1982, Gadioli *et al*. [13] in 1984 and Agarwal *et al*. in 2002 [14] employing Ge(Li) detectors. In the present work, the excitation function and the ICR for the above reaction were measured up to 120 MeV. To avoid possible interference (from the Compton background of high energy γ rays) from the short lived $^{95}Tc^g$, the long lived $^{95}Tc^m$ was studied after allowing for a decay time of several days. Figure 1 shows that the present results are agreeing well with the previous ones, while providing more data points in the high-energy side, which is of interest from the view point of preequilibrium decay.

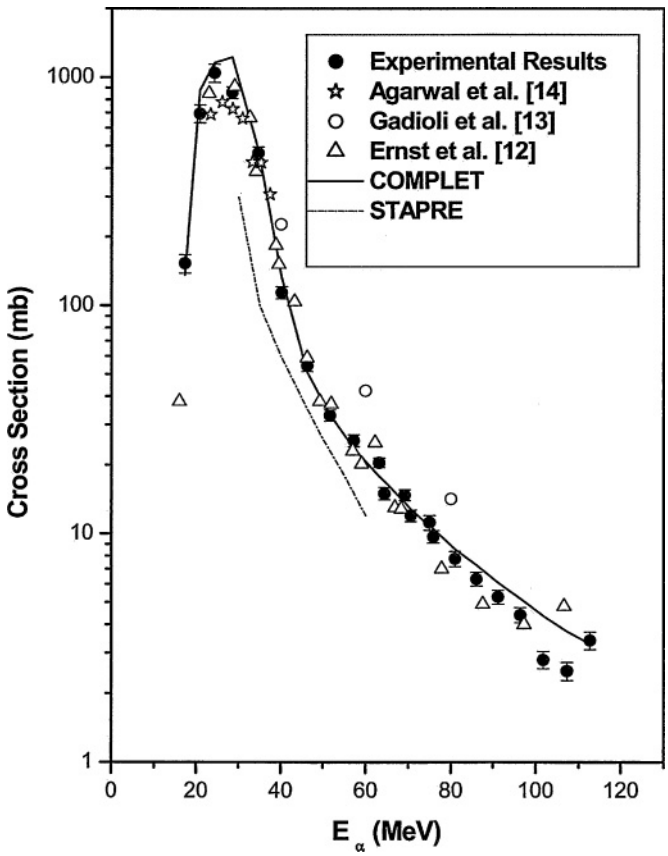

FIG. 1. The total production cross-section residue $^{95}Tc^{m+g}$ for the α-induced reaction on ^{93}Nb up to 120 MeV. The COMPLET and STAPRE model calculations along with previous experimental results are shown.

IV. GENERAL DESCRIPTION OF THE MODELS

The theoretical excitation function and ICR calculations for the preequilibrium process was done using COMPLET [3,4] and STAPRE [5,6] codes.

A. COMPLET Code

The code COMPLET [3,4] is based on the same philosophy as the former code INDEX [4]. It predicts the yield of residual nuclei in nuclear reactions with excitation energy up to 225 MeV taking into account two mechanisms. The first one preequilibrium emission is accomplished in the frames of the model of independently interacting excitons. The following simplifying assumptions have been made: (i) the preequilibrium (further PE) emission of complex particles is neglected, (ii) the equilibrium is reached after the second stage of the evolution, and (iii) maximum two PE nucleons can be emitted. An approximation concerning preequilibrium angular momentum removal is included. The equilibrium part formerly based on the Weisskopf-Ewing evaporation formula [15] is also modified to include full angular momentum decoupling regarding the emission of light particles with $A \leqslant 4$. In COMPLET code, identical to the INDEX model [4] a preequilibrium process in two stages is assumed and up to two preequilibrium particles can be emitted. The particles in the initial exciton configuration (first stage) can be neutron-, proton-, or α-particles, represented by the exciton numbers EX_1, EX_2, and EX_3 respectively. In the present calculations, we have used $EX_1 = 2.0$, $EX_2 = 2.0$, and $EX_3 = 0.2$. These excitons interact independently with particles below the Fermi level, thereby either creating new particle-hole configurations in the second stage or getting emitted into the continuum. In these interactions the original exciton type is assumed to be conserved. Conversely, the newly created exciton (particle-hole pair) may be either a (α-particle, α-hole) state formed with probability ALF or a (nucleon, nucleon-hole) state formed with probability (1-ALF). We varied the value of ALF from 0.1 to 1 after fixing the initial exciton configuration. The variation in the value of ALF produces variation at the compound nucleus peak, which is less than 10%. The value of ALF = 0.2 is found to give the best fit to the compound nucleus peak. Therefore, we have chosen ALF = 0.2 as a global parameter for all the remaining calculations using COMPLET code. Each emitted preequilibrium particle x of kinetic energy E_x carries away the angular momentum L with $L = k_x^* R$, where $k_x = 0.219^*[E_x^* A_x^* (A - A_x)/A]^{1/2}$ is the center-of-mass wave number of the ejectile and $R = 1.55A^{1/3}$. C_{ANG} [3]. The coefficient C_{ANG} has nonzero values $\leqslant 1$. Thus the quantity C_{ANG} adjusts the magnitude of the angular momentum removed. The energy E_x is measured in units of mega-electron-volts and the masses of ejectiles A_x and target nucleus A are used in units of unified atomic mass. In a classical description in terms of particle trajectories R may be interpreted as the most probable distance from the origin, at which a preequilibrium particle leaves the nucleus. In particle induced preequilibrium reactions this distance is strongly correlated to the impact parameter of the incoming partial wave as was shown earlier [16].

The equilibrium calculation in COMPLET considers the following ejectiles: $\gamma, n, p, d, t,{}^3\mathrm{He}$, and ${}^4\mathrm{He}$. Instead of former simple Weisskopf-Ewing approach a modified Hauser-Feshbach ansatz is used with angular momentum-dependent transmission coefficients. The later are derived in a sharp cutoff approximation from usual optical model inverse cross sections neglecting spin-orbit couplings of the ejectile. The $(2s+1)$ multiplicities of the spin s of the ejectiles are taken into account. However, all nuclear spins are assumed to be whole numbers. A simple decoupling scheme is formulated in counting the ways the residual nucleus spin J' can be reached from the preceding nucleus with spin J. Respective coefficients are computed and stored in an array that is precalculated for all combinations of $J \leqslant 99$, $0 \leqslant |J - J'| \leqslant 12$, and $0 \leqslant L \leqslant 12$. Spin-dependent level densities are used where the rotational energy according to Sierk's macroscopic model of rotating nuclei [17] is subtracted from the total excitation energy for spin zero. Within these approximations spin and angular momentum are strictly conserved throughout all evaporation cascades. The level density parameter $a_{\mathrm{CN}} = A/k$ is defined for each nucleus in the decay chain given by its atomic and neutron number. In the present calculation we take $k = 8$ MeV. The reaction thresholds and light particle binding energies are calculated from experimental mass tables [18].

B. STAPRE Code

The present experimental results were also modeled by the code STAPRE [5,6]. In this code the evaporation of particles and γ rays are treated in the frame work of the statistical model with consideration of angular momentum and parity using the Hauser-Feshbach formalism [19]. For the analysis of the preequlibrium part the code employs the exciton model approach proposed by Cline and Blann [20]. It should be noted that the pre-equllibrium effects are taken into account only for the first nucleon emitted in a reaction, where more than one particle is emitted. The calculated value of the preequilibrium contribution was found to be sensitive to (i) the square of absolute value of the average matrix element for two-body residual interactions $|\overline{M}|^2$ and (ii) the choice of the initial configuration, $n_0(P_0, h_0)$, of the composite system. The following relation was used to estimate the value of effective matrix element [21]:

$$|\overline{M}|^2 = FM.A^{-3}E^{-1},$$

where A and E are the mass number and excitation energy of the composite system respectively. In general FM $(\mathrm{MeV})^3$ is treated as an adjustable parameter and values between 95 and 700 $(\mathrm{MeV})^3$ have been recommended for it in the literature [19]. After trying several values of FM in the theoretical calculations in the present work, the best value of FM to give general agreement in this reaction under investigation was found to be 200 $(\mathrm{MeV})^3$.

The theoretical cross section calculations were also carried out taking different values of initial excitation number and configurations such as $n_0 = 4(4p, Oh), 5(4p, lh), 5(5p, Oh)$, and so on. It was generally found that the initial configuration

$n_0 = 4(4p, Oh)$, that is an initial configuration of four particles and zero holes give better agreement with the measured cross section. In Hauser-Feshbach calculations the transmission coefficients for n, p, α, and d were generated from the previously described code—COMPLET. The photon channels and strength functions derived from Brink-Axel theorem [22] were used.

C. Isomeric ratios

In the present work the emphasis was on the calculation of the isomer cross section. Because such calculations are strongly dependent on the input level scheme of the product nucleus [23,24], we have chosen those parameters very carefully. The energies, spins, parities, and branching ratios of the discrete levels were selected from the *Nuclear Data Sheets* [25]. Another important consideration in calculating the isomeric cross sections is the spin distribution of the level density [23,26]. This was characterized by the ratio of the effective moment of inertia Θ_{eff} to the rigid-body moment of inertia $\Theta_{\text{rig}}(\eta = \Theta_{\text{eff}}/\Theta_{\text{rig}})$ and the calculations were performed for $\eta = 1.0$. In the present theoretical calculations, the influence of this parameter was explored by varying the value to as much as half the value of the rigid body moment of inertia in typical cases. It was observed that decrease in the value of moment of inertia by half the rigid body value leads to the underestimation of the cross section by a factor of 2. Hence the rigid body moment of inertia was generally adopted.

For a specified nucleus in the decay chain the algorithm of STAPRE yields a set of cross sections for the isomeric states at discrete energies E_α. Similar to the experimental IRs the computed ones are determined as follows:

$$IR(E_\alpha) = \frac{\sigma_{\text{high}}(E_\alpha)}{\sigma_{\text{low}}(E_\alpha)}, \tag{1}$$

where $\sigma_{\text{high}}(E_\alpha)$ and $\sigma_{\text{low}}(E_\alpha)$ are the cross sections of the high and low isomers, respectively.

The 1-MeV wide excitation energy bins in the code COMPLET do not favor the implementation of an experimental nuclear level structure. Therefore, the Huizenga and Vandenbosch model was applied for calculating final γ cascade and the population of isomeric states. For a specified isomeric nucleus in the decay chain a large number of states exist with excitation energies below the particle-emission threshold. The population of these states depending on spin and bin energy is computed by COMPLET for each incident E_α. The yield distributions are then used as initial data in standard HVM calculations. The IRs obtained is defined exactly the same way as in Eq. (1).

V. RESULTS AND DISCUSSIONS

In an attempt to understand the reaction mechanism, a comparison is made between the theoretical predictions of COMPLET and STAPRE and the experimentally observed excitation function and isomeric cross section ratio for the reaction $^{93}\text{Nb}(\alpha, 2n)^{95m+g}\text{Tc}$.

A. Excitation function

Figure 1 shows the experimentally measured excitation function together with the COMPLET model (solid curve) and STAPRE (dash-dot) predictions. It can be seen that the high energy part (40 to 120 MeV) is dominated by preequilibrium process, whereas the low energy part (below 40 MeV) is dominated by compound nucleus process with its characteristic peak.

It can be seen that in the entire energy range of the present measurement the COMPLET model calculations fit the excitation function reasonably well, taking limitations of the calculations into account. The magnitude of the uncertainties in preequilibrium calculations is dependent on several factors, such as the range of equilibrium and preequilibrium reaction cross sections involved, and also the calculations involving so many free parameters, such as Fermi energy, mean free path multiplier, initial exciton number, and level densities. Within the limitations of the choice of best fitted parameters, the present comparison giving the correct spectral shape and variation of yield with excitation energy is an encouraging result.

It can be seen in the figure that the STAPRE model calculations underestimate the experimentally measured cross section within a factor of 2 in the entire energy region from 30 to 60 MeV where the model calculations are valid. As pointed out earlier, a decrease in the value of moment of inertia from the rigid body value will push down the theoretical estimates. In the light of this, the present calculations for the set FM = 200 $(\text{MeV})^3$ and $\eta = 1$ (rigid body value) gives the best fit results.

B. Isomeric ratio

Figure 2 shows the ratio $\sigma_{\text{highspin}}/\sigma_{\text{total}}$ as a function of incident particle energy, together with the two sets of theoretical predictions, by the COMPLET and STAPRE models. At the outset it can be seen that the experimentally measured ratio first increases with the incident energy up to about 35 MeV and later shows a definite decreasing trend between 35 to 50 MeV. Above 50 MeV, the ratio increases again and becomes stable beyond 60 MeV. The tendency in compound nuclear reactions is to produce residual nucleus at higher and higher spin values in the continuum with increasing bombarding energy. This naturally enhances the population of high spin state and hence the increase of observed ratio up to 35 MeV. However, at higher energies a fraction, called the bypass fraction, of the states with angular momentum (J) greater than a critical value (J_{cr}) of the compound nucleus, may feed the low spin isomer. As a result the observed ratio $\frac{\sigma_{\text{highspin}}}{\sigma_{\text{total}}}$ decreases in the energy region from 35 to 50 MeV. Both the COMPLET and STAPRE calculations reproduce fairly well the isomeric cross-section ratio in this energy region. At energies higher than 50 MeV, preequlibrium process becomes increasingly predominant. As a result the feeding of the high-spin state might be because of decay of low-lying states with high spin with single-particle origin called yrast traps. This keeps the isomeric ratio almost constant in this energy region. This hypothesis is confumed by the COMPLET calculation of the

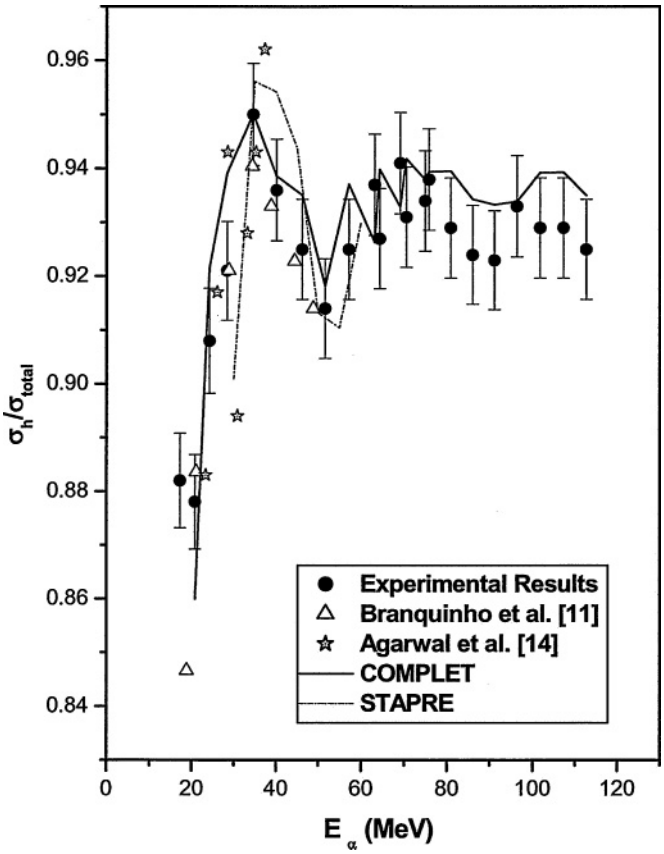

FIG. 2. The isomeric cross-section ratio (ICR) for the reaction ^{93}Nb(α, $2n$)^{95}Tc^{m+g} along with the COMPLET and STAPRE calculations and previous ICR results.

isomeric ratio in the energy region 50 to 120 MeV. The isomeric ratio as obtained from the COMPLET calculations is in good agreement with the experimental ratio in this energy region. It can also be remarked here that, in the COMPLET code, the angular momentum removal is accounted for in the pre-equllibrium calculation, in an approximate way. This is responsible for the better predictions obtained from COMPLET code.

VI. CONCLUSIONS

The excitation function and the isomeric cross-section ratio were measured for the reaction ^{93}Nb(α, $2n$)^{95}Tc^{m+g} up to 120 MeV. The experimental results were interpreted in terms of the preequlibrium models COMPLET and STAPRE. Although the COMPLET calculation fits the excitation function reasonably well in the entire energy region of the present measurement, whereas the STAPRE code gives a fit within a factor of 2. In the present reaction studied the effect of the angular momentum removal in the preequlibrium emission was observed to play a major role in the formation of the isomers and hence in the measurement of ICR. The COMPLET code successfully explains the ICR for this reaction, as the angular momentum removal has been incorporated in the code. The STAPRE code also gives the best fit for this reaction by the calculations with the set of parameters FM $= 200$ (MeV)^3 and $\eta = 1.0$.

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