Recoil Range Evidence for the Compound-Nucleus Mechanism in Reactions between Complex Nuclei

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We have measured the recoil-range distributions of 4.1-h Tb^{149} produced in a variety of complex nuclear reactions. The targets were Pr^{141}, Ce^{140}, La^{139}, and Ba^{138}; projectiles were C^{12}, N^{14}, N^{15}, O^{16}, O^{18}, F^{19}, and Ne^{20}. In every case the range distributions could be fitted to a Gaussian function. The average range values give evidence for total momentum transfer, and the range straggling is consistent with nuclear evaporation. We conclude that these reactions are pure compound-nucleus reactions. Excitation energies were approximately 50 to approximately 150 MeV, corresponding to incident energies of 5 to 10.4 MeV per nucleon.

I. INTRODUCTION

IN 1936, Niels Bohr proposed the concept of the "compound nucleus"—a relatively long-lived excited system formed by the union of projectile and target nuclei.[1] The decay of the compound nucleus has been described in most detail for situations in which the statistical model is valid.[2] The simplest form of the statistical model demands a high density of states in the residual nuclei and the vanishing of interference terms in the reaction amplitude—usually attributed to randomness of phases of the relevant matrix elements.[3] Under these conditions a compound nucleus decays by emitting particles with angular distributions symmetric about 90 deg.[4] The term "compound-nucleus reaction" has often been used, and is used in this paper, to denote the complete amalgamation of target and projectile, followed by emission of particles with symmetric angular distributions.

In many studies, the energy spectra of emitted particles and excitation functions for final products have been analyzed by assuming simple compound-nucleus reactions. However, with the exception of fission studies, measurements which verify this assumption are rare and usually indicate a very narrow region of applicability. An angular distribution symmetric about 90 deg in the center-of-mass system is usually taken as a sufficient condition (this is not a necessary condition for small excitation energies[3]) for the applicability of the statistical model. The direct observation of angular distributions of emitted particles would thus seem to be the obvious approach to testing the model. Measurements of this type usually indicate that those emitted particles of higher energy are predominantly emitted in the forward direction.[5] It has been often assumed without further verification that the statistical model is valid for particles observed at backward angles.

The statistical assumption has been clearly verified by observations of selected reaction products. Bodansky *et al.* have made observations of the two protons in coincidence from Ni^{58}(32-MeV α, $2p$) reactions.[6] These experiments give strong evidence that the coincidence requirement screens out the non-compound-nucleus reactions. Hence the properties of the excited compound nucleus were observed without interference from non-compound-nucleus reactions. Similar studies for higher incident energies will be very difficult because of the more complex coincidence requirements.

Recoil properties of the final products of specific nuclear reactions have provided another test of the statistical assumption.[7,8] These studies can furnish a simple, direct test of the model even for very large excitation energies. The measurement of the average range of the products provides a measure of the average momentum transfer. The average range provides a test of (a) momentum transfer in the initial impact and/or (b) symmetry of particle emission.[7] If particles are emitted with angular distributions symmetric about 90 deg (in the c.m. system) then the average recoil velocity of the final product will be equal to the velocity of the center of mass. The term "total momentum transfer" will be used to indicate that the average recoil velocity of the final product is equal to that of the center of mass.

In this study, we report range distributions for the product 4.1-h Tb^{149}. We studied a variety of reactions between complex nuclei over a wide range of incident energies. In every case, the range data are consistent with total momentum transfer, and thus give evidence for the validity of the compound-nucleus and statistical models. The compound nuclei so formed have excitation energies from approximately 50 to approximately 150 MeV, and presumably include angular momentum states of many tens of $\hbar$ units.

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TABLE I. Summary of the Tb^{149} recoil data.

Reactions	Bombarding energy, E_b (MeV) (lab)	Target thickness, W ($\mu g/cm^2$)	Average range, R_0 (mg/cm^2)	Measured straggling parameter, ρ	Nuclear reaction straggling parameter, ρ_n [a]
Leading to $_{65}Tb$ compound nuclei					
$Pr^{141}+C^{12}$	85.1	124	0.491	0.298	0.17
	80.3	125	0.465	0.295	0.16
	75.2	123	0.461	0.255	0.15
	69.8	124	0.430	0.273	0.14
	64.6	121	0.405	0.270	0.12
	58.3	123	0.360	0.273	0.10
$Ce^{140}+N^{14}$	84.4	34	0.566	0.240	0.11
	67.6	30	0.482	0.230	0.09
$Ce^{140}+N^{15}$	108.3	32	0.735	0.220	0.12
	102.2	38	0.696	0.242	0.11
	95.8	33	0.671	0.233	0.11
	88.5	31	0.654	0.237	0.09
$La^{139}+O^{18}$	131.0	134	0.955	0.208	0.10
	122.4	136	0.932	0.203	0.09
	113.9	134	0.892	0.175	0.08
$Ba^{138}+F^{19}$	137.6	136	1.059	0.177	0.10
	125.8	127	1.010	0.181	0.08
	111.9	134	0.90	0.19	0.07
Leading to $_{66}Dy$ compound nuclei					
$Pr^{141}+N^{14}$	142.8	81	0.833	0.25	0.17[b]
	137.2	79	0.810	0.233	0.14
	132.0	82	0.810	0.222	0.12
	126.4	81	0.808	0.197	0.07
	120.8	80	0.789	0.216	0.12
$Pr^{141}+N^{15}$	153.0	82	0.942	0.190	0.09
	147.9	79	0.917	0.193	0.09
	142.8	81	0.926	0.199	0.11
	137.4	81	0.856	0.203	0.10
	132.0	80	0.856	0.209	0.11
	113.1	70	0.779	0.207	0.08
$Ce^{140}+O^{16}$	163.0	36	1.028	0.213	0.15
	155.5	34	1.042	0.204	0.13
	148.6	30	0.992	0.219	0.15
	141.3	32	0.964	0.189	0.10
	133.6	35	0.889	0.187	0.07
	126.2	80	0.863	0.199	0.09
	118.4	32	0.830	0.177	0.00
$La^{139}+F^{19}$	192.9	139	1.312	0.166	0.11
	182.4	134	1.248	0.174	0.11
	170.8	136	1.236	0.163	0.09
	160.7	134	1.169	0.161	0.08
Leading to $_{66}Dy$ compound nuclei—*Continued*					
$Ba^{138}+Ne^{20}$	202.6	136	1.402	0.155	0.10[b]
	188.8	127	1.381	0.153	0.10
	175.6	134	1.199	0.169	0.10
	162.0	135	1.251	0.144	0.06
	148.0	132	1.158	0.160	0.07
Leading to $_{67}Ho$ compound nuclei					
$Pr^{141}+O^{16}$	162.9	81	1.035	0.180	0.09[b]
$Pr^{141}+O^{18}$	179.8	124	1.169	0.202	0.14
	172.4	125	1.162	0.181	0.11
	164.3	123	1.145	0.183	0.12
	156.2	124	1.100	0.188	0.12
	139.0	121	0.976	0.205	0.13
$Ce^{140}+F^{19}$	192.9	36	1.293	0.179	0.13
	182.6	34	1.258	0.177	0.12
	172.1	30	1.228	0.170	0.10
	160.2	32	1.173	0.171	0.10
	148.4	35	1.106	0.196	0.13
$La^{139}+Ne^{20}$	202.6	139	1.371	0.162	0.11
	188.8	134	1.325	0.163	0.11
	174.6	136	1.222	0.162	0.09
	162.0	134	1.179	0.164	0.09
Leading to $_{68}Er$ compound nuclei					
$Pr^{141}+F^{19}$	191.7	150	1.227	0.220	0.17[b]
	182.0	142	1.152	0.230	0.18
	171.8	146	1.126	0.240	0.19
	161.5	147	1.094	0.229	0.18
	151.2	144	1.096	0.205	0.14
	138.9	143	1.033	0.191	0.11
	126.7	143	0.975	0.178	0.07
$Ce^{140}+Ne^{20}$	203.2	36	1.308	0.201	0.16
	188.8	34	1.251	0.236	0.20
	174.0	30	1.209	0.202	0.15
	160.0	32	1.175	0.213	0.16
	145.2	35	1.148	0.191	0.13
	129.6	32	1.017	0.188	0.11
Leading to $_{69}Tm$ compound nuclei					
$Pr^{141}+Ne^{20}$	202.8	124	1.295	0.213	0.17[b]
	188.6	125	1.270	0.206	0.16
	175.6	123	1.200	0.207	0.16
	161.8	124	1.189	0.166	0.09
	147.2	121	1.078	0.173	0.09

[a] The values of ρ_n for the reactions leading to Tb compound nuclei are calculated values based on assumption of isotropic neutron emission. See reference 11. All other values of ρ_n were obtained from Eq. (2) as described in the text.

[b] The estimated standard errors for these values of ρ_n are approximately 0.04.

II. EXPERIMENTAL PROCEDURES

We have made differential range measurements for 4.1-h Tb^{149} using thin Al catcher foils. The techniques were essentially the same as previously described[7]; however, several improvements have been made. Catcher foils were punched from the central areas of commercially available sheets of Al leaf (approximately 150 $\mu g/cm^2$). The foils were visually inspected by looking through them into a lamp. Only the better foils were accepted. All targets were prepared by evaporation of thin layers onto 0.00025-in. Al backing foils. Rare-earth metals and $BaCl_2$ were evaporated. Rare-earth oxides were evaporated for earlier work.[7] The rare-earth metals were volatilized much more readily, and significantly lower amounts of heavy-element impurities were observed with these targets.[9]

We used separated isotopes of Ce^{140} and Ba^{138} from the

[9] A few of the experiments reported in reference 7 indicated complex Tb^{149} range distributions. These experiments were repeated in this work and were found to be due to heavy-element impurities in the target layers.

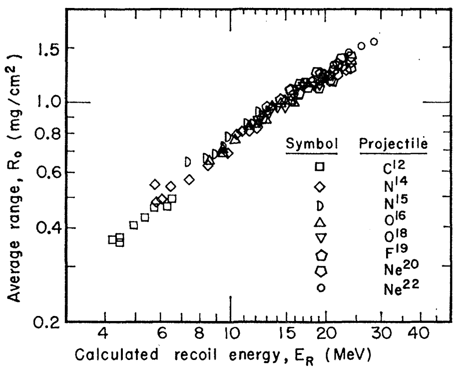

FIG. 1. Average range as a function of calculated recoil energy for reactions leading to 4.1-h Tb^{149}.

Isotopes Division of the Oak Ridge National Laboratory. The isotopic purities were 99.7 and 98.0%, respectively.

The α radioactivity from 4.1-h Tb^{149} in the various foils was measured with 10 to 14 2π ionization chambers gated by a single switch. The counters were set to equivalent sensitivity by intercalibration with thick uranium standards. Backgrounds of all counters were between 0.2 and 0.5 count/min.

III. RESULTS AND DISCUSSION

The experimental data were fitted to a Gaussian function by probability plots as described previously.[7] In every case a very good fit was obtained for about 95% of the range distribution.[9] Therefore, we can describe the range distribution by two parameters, the average range R_0 and the straggling parameter ρ. The standard deviation of the range distribution is given by the product ρR_0. In Table I we show the results of this study. Columns 1 to 3 give the reaction studied, the beam energy, and target thickness. Columns 4 and 5 give the measured values of R_0 and ρ. The last column gives the nuclear reaction straggling parameter, which is discussed later. Beam energies are based on initial energies of 10.38 MeV per amu for the Berkeley Hilac and the range-energy curves of Northcliffe.[10]

The Average Range

The average range values can be used as a measure of the average recoil energy or momentum if a range-energy curve is known independently. However, no independent range-energy data for Tb^{149} are available now. Thus we turn to an internal-consistency argument to test for the possibility of full momentum transfer. As stated in the introduction, if the final product recoils with the momentum of the incident projectile [decreased, of course, by the mass factor $A_R/(A_T+A_b)$; see Eq. (1) below], then the particle emission must be symmetric about 90 deg in the center-of-mass system. The emission of particles with angular distribution symmetric about 90 deg contributes directly to the range straggling, but affects the average range very slightly. (This statement is not general, but applies to the reactions of interest in this study. This is because the projectile momentum is so much greater than that of the evaporated particles—even for evaporated protons and He nuclei. See reference 7.) If a reaction proceeds by full momentum transfer followed by emission of particles with symmetric angular distributions, then the average recoil energy E_R is given by

$$E_R=E_bA_RA_b/(A_b+A_T)^2. \qquad (1)$$

Kinetic energy and mass are denoted by E and A with subscripts R for the recoil, b for the projectile, and T for the target.

In Fig. 1 we plot the measured average range values versus calculated values of E_R. It is clear that the data can be very well described (standard deviation $\approx 4\%$) by one smooth curve. This figure shows all measurements, including those from previous work.[7] From Table I we see that compound systems of atomic number 65 to 69 were possible, and that each reaction was studied at several widely spaced energies. The single-valued relationship of all the measurements of R_0 with E_R demands that the fractional momentum transfer be the same for all these reactions at all energies [or that the true value of E_R be related to Eq. (1) by a constant factor]. It is difficult to propose mechanisms involving partial momentum transfer that predict this result. Therefore, we conclude that the momentum transfer is complete, i.e., that particles are emitted symmetrically. This conclusion implies, in turn, that these reactions involve the formation and decay of a compound nucleus.

It is essential to inquire how sensitive the measurement of R_0 is to deviations from symmetrical particle emission. This question can be answered only by referring to specific examples. Suppose, for instance, that one nucleon is emitted along the incident beam direction with the incident velocity and that all other particles are emitted symmetrically. Such a process would lead to a fractional momentum transfer of $(A_b-1)/A_b$. Since these ranges are proportional to momentum to the power ~ 1.3 to ~ 1.9, the resulting range would be depressed by $[(A_b-1)/A_b]^{1.3 \text{ to } 1.9}$, or about 7–18%. If such a process occurred with equal probability for all reactions at all energies, it would not be evident from these results. However, if such a process occurred with increasing probability for higher incident energies, then deviations from the single-valued R_0-vs-E_R curve would be very likely.

As a second example, suppose that initially all the momentum is transferred. Then imagine that particles are emitted in such a way that the angular distribution of the final product, in the c.m. system, is given by $1+0.1\cos\theta$. The resulting average range would be depressed by approximately 1% by this slightly asymmetric angular distribution.

It is interesting to compare Fig. 1 with Fig. 2, which shows the R_0 measurements for alpha-emitting species

[10] L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).

produced from reactions of complex nuclei with Bi^{209}. Most data in Fig. 2 were taken from reference 7. The Ne^{20} measurements were repeated and found to be in error, and have been corrected. In this plot there is no simple relationship between R_0 and E_R, and this fact gives evidence for non-compound-nucleus reactions. We conclude that measurements of R_0 do furnish a severe test of compound-nucleus formation, provided that a sufficiently accurate and extensive study is performed.

Range Straggling

The observed range-straggling parameter ρ is the result of a combination of effects. Following a previous discussion,[7] we assume that the various sources of range straggling can be approximated by Gaussian functions. We denote the various individual straggling parameters as follows: (a) range straggling inherent in the stopping process, ρ_s; (b) velocity distribution of the nuclear reaction products, ρ_n; (c) catcher foil inhomogeneities, ρ_f; and (d) target thickness, ρ_w. Then we have

$$\rho^2=\rho_s^2+\rho_n^2+\rho_f^2+\rho_w^2. \qquad (2)$$

We would like to unravel the various effects. Since these straggling parameters combine in quadrature, it is quite likely that several of the effects make only minor contributions to the observed straggling. Our target thicknesses were very small compared with the average range values. Therefore the effect of ρ_w is small and can be subtracted accurately.[7] It has been found that Monte Carlo calculations of the nuclear evaporation process give very good agreement with measured angular distributions for Tb^{149} recoils formed from Tb compound nuclei.[11] (Such agreement was not found for reactions leading to Dy^{156} compound systems.) We can infer that the range straggling due to nuclear evaporation can be

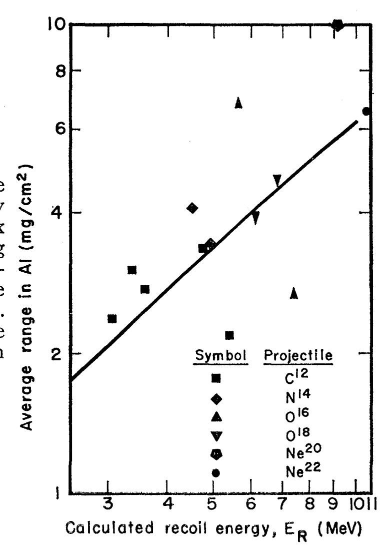

FIG. 2. Average range versus calculated energy for reactions of complex nuclei with Bi^{209} leading to alpha-emitting nuclides. Symbols denote the various projectiles. The solid line is the range-energy curve from reference 7.

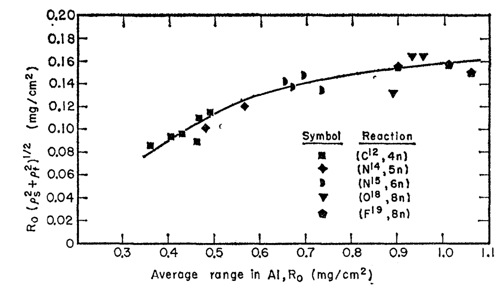

FIG. 3. Range straggling of Tb^{149} due to foil inhomogeneities and stopping phenomena as a function of average range. These data are from Tb compound nuclei only.

adequately calculated for reactions involving Tb compound nuclei. Such calculations have been performed and will be described elsewhere.[12] The results show that a Gaussian function gives a good representation of the range distribution that results from the velocity distribution. Calculated values of ρ_n are given in Table I for those reactions leading to Tb compound nuclei. The calculated values of ρ_n^2 are all less than 4/10 the measured values of ρ^2, and for this reason do not constitute the major source of the observed straggling. We have subtracted these values of ρ_n^2 from $(\rho^2-\rho_w^2)$ to obtain the range straggling due to foil inhomogeneities and the stopping process. The results are shown in Fig. 3 in terms of standard deviations.

These values of $\rho_s^2+\rho_f^2$ have, in turn, been subtracted from $\rho^2-\rho_w^2$ to obtain ρ_n^2 for the reactions leading to compound systems of $Z=66$ to 69. The resulting ρ_n values are given in Table I. The error limits for these values of ρ_n are too large to warrant any quantitative discussion. However, these values are in qualitative agreement with the expectations of nuclear evaporation theory,[13] and therefore give additional evidence that all the reactions are compound-nucleus reactions. The occurrence of other mechanisms would presumably give rise to larger range straggling.

From our data it is not possible to determine the relative magnitudes of ρ_f and ρ_s. However, we did perform one series of experiments that strongly suggests that indeed $\rho_f \ll \rho_s$. In these experiments La targets were irradiated with F^{19} and three catcher foils were used. The first catcher foil was commercially available 0.00025-in. Al ($\approx$1.8 mg/cm²) and the last two foils were from Al leaf ($\approx$0.15 mg/cm²) used for the range measurements. The macroscopic inhomogeneities of the thicker Al are very much less than those of the Al leaf. Yet the measured fraction of the Tb^{149} recoils penetrating the thick Al was equivalent to that obtained from the experiments using only Al leaf for catcher foils. This agreement demands that either (a) the range straggling is not predominantly due to foil inhomogeneities, or (b) microscopic inhomogeneities of both

[11] Gabriel N. Simonoff and John M. Alexander, Lawrence Radiation Laboratory Report UCRL-10099, 1962 (unpublished).

[12] J. M. Alexander, L. Altman, and S. Howry, Lawrence Radiation Laboratory, University of California, Berkeley, California (unpublished).

[13] See Eq. (16) of reference 7.

kinds of Al are equivalent. The former alternative seems more likely.

CONCLUSIONS

Measurements of the average range and range straggling of the product 4.1-h Tb^{149} provide strong evidence for production of this nuclide (and its parents by radioactive decay) via pure compound-nucleus reactions. The reactions studied include compound nuclei of atomic numbers 65 to 69. Therefore the nuclide $_{65}Tb^{149}$ was formed by reactions involving only neutron emission and also by reactions involving charged particle emission. Seventeen different reactions were studied. The initial excitation energies varied from approx 50 to 150 MeV. Further studies of these reactions should provide information about the properties of nuclei with high excitation energies and angular momenta.

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Studies of 15-MeV Inelastic Deuteron Scattering*

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Angular distributions of inelastically scattered deuterons from Ni leading to many individually resolved states of the final nucleus were measured. The collective 2^+ levels have identical angular distributions, but these are considerably different from those leading to other 2^+, 0^+, and 4^+ levels. Many (but not all) of the latter are strongly similar to each other. The 3^- states in Ni^{60} and Ni^{62} have similar angular distributions, but the 3^- state in Ni^{58} is appreciably different. The predictions of the Blair phase rule on relative phases of elastic scattering angular distributions and those leading to states of positive and negative parity are not valid in Ni, but they apply fairly well to the collective states in Zr and Sn. While the other states in these nuclei have somewhat different angular distributions, the phase rule works well enough to make tentative parity assignments for a large number of levels. The method breaks down badly only for the two phonon levels; all attempts at systematizing data from the latter fail. A survey of energy spectra from 27 elemental and isotopic targets is reported. Corrections to a previous survey are listed.

I. INTRODUCTION AND EXPERIMENTAL METHOD

In a previous paper,[1] a study of inelastic deuteron scattering was presented, including a survey of energy spectra from a large number of nuclei through the periodic table, a few angular distribution measurements, and correlations with stripping and Coulomb excitation experiments. Unfortunately, that work was interrupted by an extended cyclotron breakdown, so that the survey of energy spectra was not as complete as had been intended, and the question of the utility of (d,d') angular distribution measurements for nuclear structure studies was left largely unanswered. It is the purpose of this paper to fill these gaps.

A most useful tool for analyzing direct interaction inelastic scattering experiments is the Blair phase rule,[2] which states that under certain approximations, the angular distribution of the inelastically scattered particle is oscillatory, and the phase of the oscillations is determined only by whether Δl, the angular momentum transferred to the nucleus in the collision, is even or odd; in particular, if Δl is odd the oscillations should be in phase with the oscillations in the elastic scattering angular distributions, and if Δl is even the phase should be shifted relative to these by 180°. Since Δl determines the parity change, this method is generally considered to be a technique for determining parities of nuclear levels. It has been used quite successfully[3] in studies of (α,α') reactions, but has been somewhat less successful in (pp') reactions,[4] probably because the interactions are not concentrated at the nuclear surface as postulated in the theory.[2] This objection is certainly not valid for (d,d') reactions, but there are the additional problems in that the deuteron does not fulfill the assumptions in the theory that the spin of the scattered particle be zero, and its size be much smaller than the radius of the target nucleus. It is thus an open question as to whether (d,d') reactions obey the Blair phase rule, and we here take an experimental approach to answering it by

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