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UCRL-10098

For Phys. Rev.

UNIVERSITY OF CALIFORNIA
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Berkeley, California
Contract No. W-7405-eng-48

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ABSTRACT

We have measured the recoil-range distributions of 4.1-hr 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 approx 50 to approx 150 Mev, corresponding to incident energies of 5 to 10.4 Mev per nucleon.

RECOIL RANGE EVIDENCE FOR THE COMPOUND-NUCLEUS MECHANISM IN REACTIONS BETWEEN COMPLEX NUCLEI*

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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 for situations in which the statistical model is valid.[2] The simplest form of the statistical model demands the participation of a large number of states in the decay process and a randomness of their phases.[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 fusion 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] Total momentum transfer is the average result if the ejected particles are emitted symmetrically in the center-of-mass system.

In this study, we report range distributions for the product 4.1-hr 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 approx 50 to approx 150 Mev, and presumably include angular momentum states of many tens of $\hbar$ units.

II. EXPERIMENTAL PROCEDURES

We have made differential range measurements for 4.1-hr 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 (approx 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 Isotopes Division of the Oak Ridge National Laboratory. The isotopic purities were 99.7% and 98.0% respectively.

The α radioactivity from 4.1-hr 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 counts/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

$$E_R = \frac{E_b A_R A_b}{(A_b + A_T)^2} \quad . \qquad (1)$$

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 and 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 approx 1.3 to approx 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 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 adequately calculated for reactions involving Tb compound nuclei. Such calculations have been performed and are 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.

The range straggling due to the stopping process is very interesting. It reflects the relative importance of energy loss to electrons and to nuclei of the stopping medium. Lindhard and Scharff calculate that the range straggling due to nuclear stopping, $\langle (R-R_0)^2 \rangle / R_0^2$, is given by

$$\frac{\langle (R-R_0)^2 \rangle}{R_0^2} \approx \frac{2}{3} \frac{A_R A_S}{(A_R + A_S)^2} , \qquad (3)$$

where A denotes mass number with subscripts R for the recoil atoms and S for the stopping atoms.[14] Let us assume $\rho_f << \rho_s$. Then from Fig. 3 we get a measure of the range straggling due to the stopping process. For 4-Mev Tb^{149} recoils this straggling is in agreement with Eq. (3). For 15-Mev recoils the observed straggling parameter, ρ (which equals $[\langle (R - R_0)^2 \rangle / R_0^2]^{1/2}$), is about one-half the value from Eq. (3). We conclude that essentially all the energy loss of 4-Mev Tb^{149} recoils is to Al nuclei, but an appreciable amount of electronic stopping occurs for the 15-Mev recoils.

From our data it is not possible to determine the relative magnitudes of ρ_f and ρ_s. Therefore the inferences of the last paragraph are open to question. However, we did perform one series of experiments that strongly suggests that indeed $\rho_f << \rho_s$. 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 (≈ 1.8 mg/cm^2) and the last two foils were from Al leaf (≈ 0.15 mg/cm^2) 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 kinds of Al are equivalent. The former alternative seems more likely.

CONCLUSIONS

Measurements of the average range and range straggling of 4.1-hr Tb^{149} give strong evidence for pure compound-nucleus reactions. The reactions studied include compound nuclei of atomic numbers 65 to 69 and excitation energies of approximately 50 to 150 Mev. The projectiles employed were C^{12}, N^{14}, N^{15}, O^{16}, O^{18}, F^{19}, and Ne^{20}, and the targets were Pr, Ce, La, and Ba. These reactions furnish a most promising tool for exploring the properties of nuclei with high excitation energies and angular momenta.

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_o (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

Table I. (cont)

Reactions	E_b	W	R_o	ρ	ρ_n
$La^{139} + F^{19}$	192.9	139	1.312	0.166	0.11[b]
	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
$Ba^{138} + Ne^{20}$	202.6	136	1.402	0.155	0.10
	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

Table I. (concl.)

Reactions	E_b	W	R_o	ρ	ρ_n
		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.

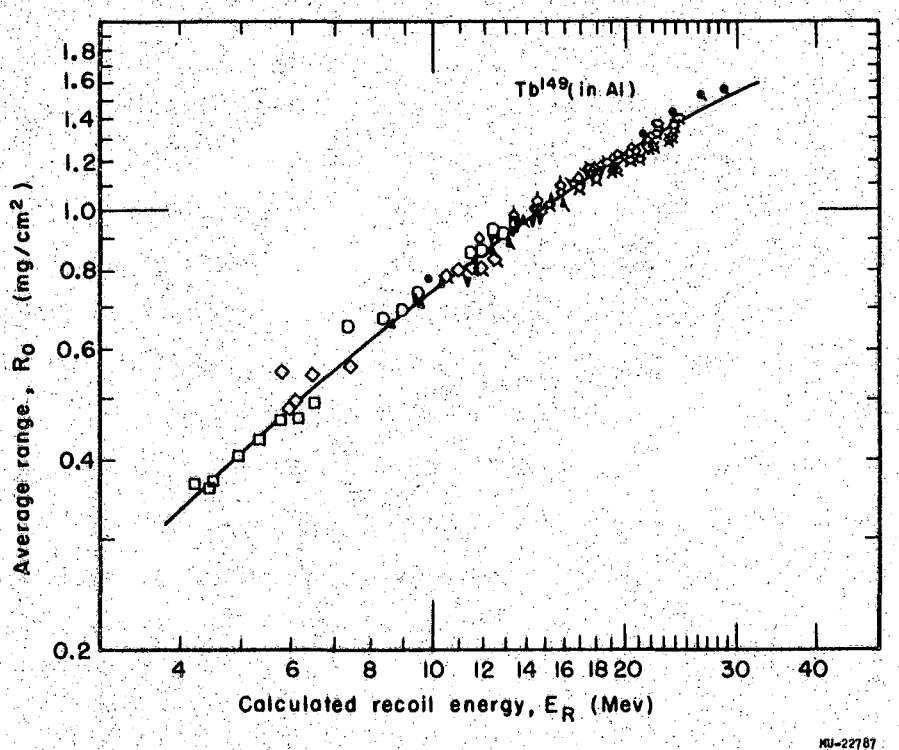

Fig. 1. Average range as a function of calculated recoil energy for reactions leading to 4.1-hr Tb^{149}. The symbols are as follows: C^{12}, □; N^{14}, ◇; N^{15}, ⬠; O^{16}, △; O^{18}, ▽; F^{19}, ⬠; Ne^{20}, ⬡; Ne^{22}, ○. Four bars are for reactions forming compound nuclei for Z = 69; three bars for Z = 68, two bars for Z = 67; one bar for Z = 66; no bars for Z = 65.

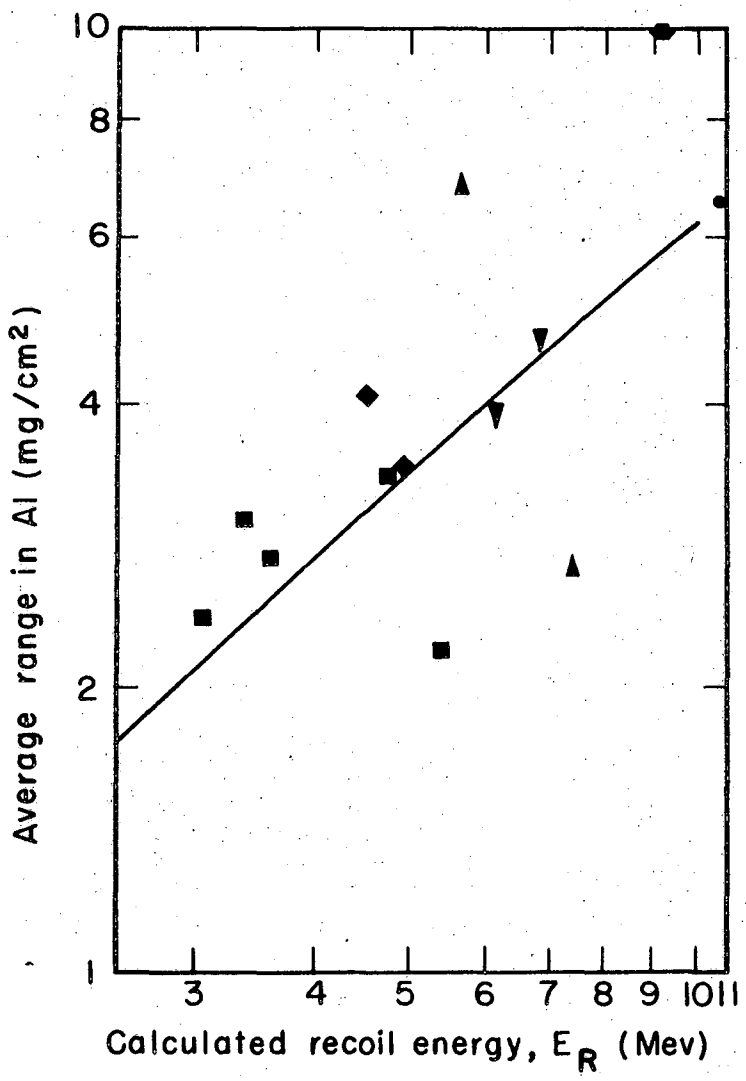

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Fig. 2. Average range versus calculated energy for reactions of complex nuclei with Bi^{209} leading to alpha-emitting nuclides. Symbols denote the various projectiles as in Fig. 1. The solid line is the range-energy curve from reference 7.

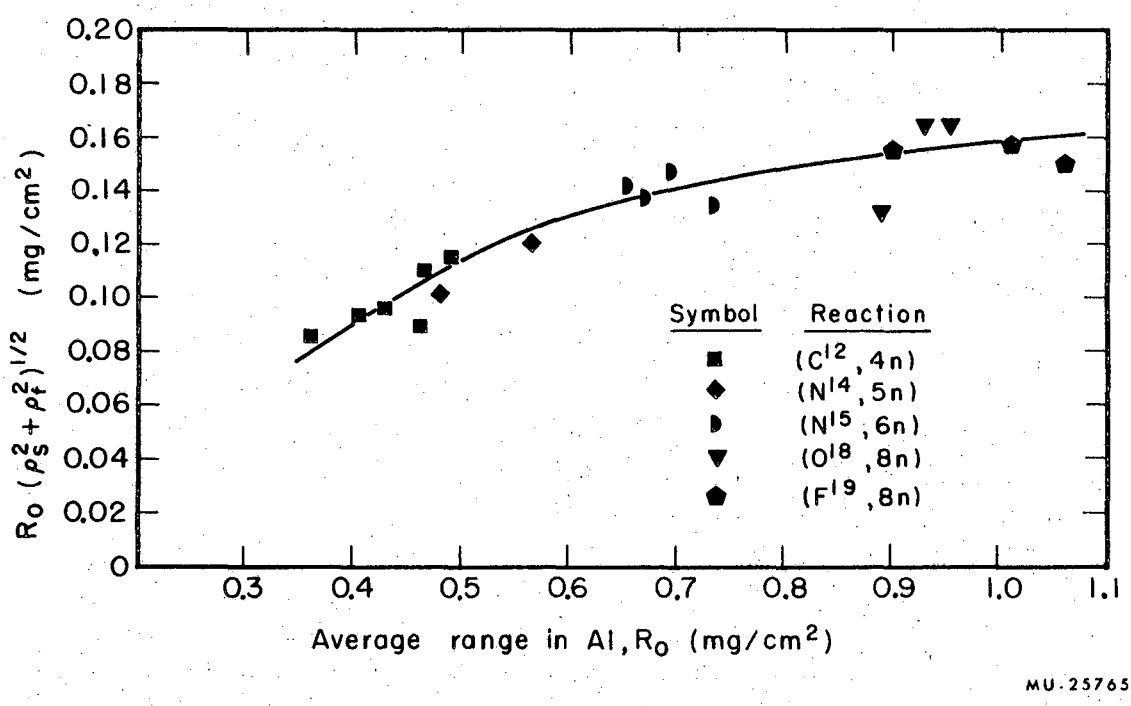

Fig. 3. Range straggling of Tb^{149} due to foil inhomogeneities and stopping phenomena as a function of average range. Symbols are as in Fig. 1. These data are from Tb compound nuclei only.

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* Work done under the auspices of the U. S. Atomic Energy Commission.

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ACKNOWLEDGMENTS

We are indebted to Dan O'Connell and Gordon Steers for their tireless efforts in the preparation of targets. We appreciate the help of Ken Russell with the counting equipment. The operating crew of the Hilac furnished the projectiles.

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