

increasing importance of photon emission as more angular momentum is deposited in the reacting system.^{12,13} It would be very informative to be able to determine the dependence of T_γ on total available energy and to extend the analysis to our data for other nuclear reactions. However, neither our straggling parameter measurements nor the theory are sufficiently accurate to permit such an attempt at the present time.

¹³ J. R. Grover, *Phys. Rev.* **127**, 2142 (1962).

ACKNOWLEDGMENTS

We would like to thank Professor John M. Alexander for many stimulating discussions throughout the course of this work. The support of Professor E. Robert Beringer and the cooperation of the HILAC staff are gratefully appreciated. Our thanks are also due to Professor Richard Wolfgang and Dr. Ivor Preiss for making their experimental facilities available to us while our own equipment was being constructed.

PHYSICAL REVIEW

VOLUME 134, NUMBER 1B

13 APRIL 1964

Recoil Properties of Sm^{142} from Nuclear Reactions Induced by Heavy Ions. II. Europium and Gadolinium Compound Systems*

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(Received 28 October 1963)

Using thin-target recoil techniques, we have measured the average ranges and range straggling, in aluminum, of 72-min Sm^{142} produced in heavy-ion induced nuclear reactions. Eight different combinations of target and beam projectile were studied, five leading to Eu compound systems and three leading to Gd compound systems. In all cases the recoil-range distributions could be fitted by Gaussian functions. Comparison of the average ranges with a range-energy curve for Sm^{142} in Al provides evidence for a compound-nucleus mechanism in these reactions. The straggling parameters observed in reactions leading to Eu compound systems are in good agreement with those obtained for (HI, xn) reactions. In reactions leading to Gd compound systems, the straggling parameters are found to be anomalously large. It is suggested that these effects are due to alpha-particle emission from highly excited Gd compound nuclei, and an attempt is made to infer the kinematics associated with this process. The results of a relatively simple analysis of the straggling-parameter data show that the average kinetic energies of the emitted alpha particles are reasonable, but somewhat different for the several reactions investigated.

INTRODUCTION

IN the preceding paper¹ we have described the recoil properties of 72-min Sm^{142} produced from samarium compound systems. (By compound system we simply mean the sum of target atom and beam projectile.) In that work the observed Sm^{142} could only be formed by (HI, xn) reactions; i.e., only neutrons could be emitted. All the reactions studied were shown to occur by means of a pure compound-nucleus mechanism, and a range-energy curve was obtained for Sm^{142} in Al.

The present paper describes experiments in which the constraint on type of particle emitted has been relaxed. As the precursors of Sm^{142} in the radioactive decay chain are unknown, there is some ambiguity introduced into our knowledge of the nuclear reactions which are taking place. Thus, the observed product could arise either by (HI, xn) reactions followed by beta decay or by the emission of charged particles as well as neutrons in the reaction.

We have measured the average ranges and straggling

parameters, in aluminum, for Sm^{142} produced by the interaction of heavy ions with a number of targets. (For a discussion of the significance of these quantities, the reader is referred to the preceding paper¹ and the references given there.) Five different reactions leading to europium compound systems and three reactions leading to gadolinium compound systems have been investigated. The data obtained provide evidence that the observed Sm^{142} is formed by a compound-nucleus reaction mechanism, and an attempt is made to distinguish between competing reactions which would lead to the same product.

EXPERIMENTAL

The experimental procedure has been described in detail in the preceding paper,¹ and hence need only be summarized here. Stacks of thin targets and thin aluminum catcher foils, each of known area and individually weighed, were irradiated with an appropriate beam from the Yale heavy ion linear accelerator. For experiments with Nd^{142} , Ce^{140} , Ba^{136} , and Ba^{137} , the targets were highly enriched in the desired isotope.² The

* This work has been supported by the U. S. Atomic Energy Commission.

¹ Morton Kaplan and Richard D. Fink, preceding paper, *Phys. Rev.* **134**, B30 (1964).

² Obtained from the Isotopes Department, Oak Ridge National Laboratory.

TABLE I. Recoil properties of Sm^{142} in Al, from heavy-ion induced reactions leading to Eu and Gd compound systems.

Reaction	Bombarding energy, E_b (MeV)	Target thickness, W ($\mu\text{g}/\text{cm}^2$)	Average range, R_0 (mg/cm^2 Al)	Straggling parameter, ρ
Reactions leading to Eu compound systems:				
$\text{Nd}^{142} + \text{Li}^6$	62.5	31	0.197	0.374
	61.0	57	0.186	0.415
$\text{Ce}^{140} + \text{B}^{10}$	103.8	26	0.535	0.229
	96.8	25	0.512	0.273
	96.8	25	0.497	0.275
$\text{La}^{139} + \text{C}^{12}$	124.1	112	0.675	0.278
	122.0	105	0.668	0.278
$\text{Ba}^{136} + \text{N}^{14}$	129.9	121	0.808	0.222
	126.3	121	0.839	0.206
$\text{Ba}^{137} + \text{N}^{14}$	144.6	111	0.880	0.198
	139.2	116	0.878	0.199
Reactions leading to Gd compound systems:				
$\text{Pr}^{141} + \text{B}^{10}$	103.8	67	0.422	0.402
	98.3	58	0.421	0.469
	94.2	58	0.400	0.421
$\text{Ce}^{140} + \text{C}^{12}$	124.1	27	0.763	0.372
	122.2	25	0.723	0.394
	115.1	26	0.669	0.350
$\text{La}^{139} + \text{N}^{14}$	144.6	124	0.805	0.281
	144.6	115	0.817	0.302
	144.5	123	0.811	0.272
	132.6	115	0.763	0.283
	129.8	123	0.798	0.274

other targets used are monoisotopic. The Sm^{142} product nuclei recoiled out of the thin target layers and were stopped in the catchers. After bombardment, the stack was disassembled and the foils were counted on a series of intercalibrated proportional counters. Aluminum absorbers, $432 \text{ mg}/\text{cm}^2$ thick, were placed between the samples and detectors to provide energy discrimination, and the Sm^{142} was measured by detecting the high energy positrons (maximum energy 3.80 MeV) emitted in the decay of the 34-sec Pm^{142} daughter.³ Blank corrections were made for activation of the aluminum catchers, and Sm^{142} decay curves were taken over several half-lives to ensure identification of the desired product.

At the bombarding energies available, the cross sections for Sm^{142} production from Eu and Gd compound systems are substantially smaller than those from Sm compound systems. As a result the decay curves obtained in the present experiments did not always yield a single component with the correct half-life. When possible, small corrections were made to remove interfering activities, but if the desired Sm^{142} product was not the major component in the decay curve, the experiment was discarded. In general, the half-lives obtained agreed with the known value to within a few percent. It should also be pointed out, that because the radioactive precursors of Sm^{142} are unknown, the possibility exists that we are actually measuring a combination of

activities of similar, or compensating, half-lives. We believe this to be unlikely, and have analyzed our data as applying to pure Sm^{142} . We shall return to this point in the next section, and briefly discuss its effects on our results. However, in view of the greater uncertainties in these experiments, we feel that the data are probably somewhat less reliable than the corresponding experiments discussed in the preceding paper.

RESULTS AND DISCUSSION

We have studied the combinations $\text{Nd}^{142} + \text{Li}^6$, $\text{Ce}^{140} + \text{B}^{10}$, $\text{La}^{139} + \text{C}^{12}$, $\text{Ba}^{136} + \text{N}^{14}$, and $\text{Ba}^{137} + \text{N}^{14}$ leading to Eu compound systems, and $\text{Pr}^{141} + \text{B}^{10}$, $\text{Ce}^{140} + \text{C}^{12}$, and $\text{La}^{139} + \text{N}^{14}$ leading to Gd compound systems. The Sm^{142} activities obtained from the decay curves were analyzed by means of probability plots^{1,4} and in every case the recoil-range distribution could be fitted by a Gaussian function. Small corrections for finite target thickness were made by adding one-half the target thickness, converted to aluminum equivalent, to the total catcher thickness. The experimental results are presented in Table I. The first two columns list, respectively, the reacting system and the bombarding energy. These energies were computed from the maximum beam energy of 10.5 MeV/amu, the known thickness of aluminum used to degrade the beam, and the range-energy curves of Northcliffe.⁵ The third column gives the target thickness. Columns 4 and 5 show, respectively, the average recoil range and straggling parameter of Sm^{142} in aluminum.

The average range measurements may be used to determine the linear momentum transfer from the incident beam to the reacting system. From the range-energy curve for Sm^{142} in Al, obtained in Ref. 1 from studies of (HI, xn) reactions, we may derive the recoil energies corresponding to our average range measurements in Table I. These values may then be compared with the recoil energies calculated on the basis of a compound-nucleus reaction mechanism. Assuming full momentum transfer from the beam projectile and isotropic emission of particles in the center-of-mass system, the recoil energy is given by

$$E_R = A_b A_R \bar{E}_b / (A_b + A_T)^2, \quad (1)$$

where the subscripts are b for bombarding projectile, T for target nucleus, and R for recoil nucleus. The mass number is A and the kinetic energy is E .

The average ranges measured for reactions leading to Eu compound systems are all in excellent agreement (better than 5%) with the values expected for full momentum transfer, as given by Eq. (1). We believe that this result, along with the Gaussian nature of the range distributions, provides strong evidence for a compound-nucleus mechanism in these reactions. The ranges obtained in the reactions $\text{Ce}^{140} + \text{C}^{12}$ and

³ Thomas V. Marshall, University of California Lawrence Radiation Laboratory Report UCRL-8740, 1960 (unpublished).

⁴ L. Winsberg and J. M. Alexander, Phys. Rev. **121**, 518 (1961).

⁵ L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).

$\text{La}^{139} + \text{N}^{14}$, leading to Gd compound systems, are also in reasonable agreement with the predictions of Eq. (1). The individual values show slightly greater deviations from full momentum transfer than was the case for Eu compound systems, but the deviations are not systematic and amount to about 10% in the least favorable case. We believe that this simply reflects the greater uncertainty in these experiments (due to low cross sections and interfering activities) and that it is very likely that a compound nucleus reaction mechanism is responsible for the observed Sm^{142} product. The reaction $\text{Pr}^{141} + \text{B}^{10}$, which also leads to a Gd compound system, seems to give Sm^{142} recoil ranges which are consistently low by 15–18%, as compared to Eq. (1). We do not understand why this system should be different, and are unable to offer an explanation for the apparent incomplete momentum transfer in the reaction. Still, the average range measurements show that 80% or more of the incident-beam momentum is transferred, and it is difficult to visualize how this could occur if the process were grossly different from a compound-nucleus reaction.⁶

We now turn to a discussion of our straggling-parameter data. The results given in Table I show the striking feature that, for given average range values, the straggling parameters for reactions leading to Gd compound systems are much larger than those for Eu compound systems. A comparison with the extensive measurements in Ref. 1 for (HI, xn) reactions show that the straggling parameters for Sm and Eu compound systems agree very well with each other, and are strongly dependent only on the value of the average range.

The distribution in recoil ranges arises from several contributions: ρ_s , the straggling inherent in the stopping process; ρ_n , the momentum distribution due to effects of the nuclear reaction; ρ_w , the finite target thickness; and ρ_f , inhomogeneities in the catcher foils. These various components combine approximately as the squares, to give the observed straggling parameter:

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

The effect of target thickness may be estimated as $\rho_w = 0.6W/2R_0$, where W is the target thickness, R_0 is the average range, and the factor 0.6 is the approximate relative stopping power of the target material and aluminum. We may subtract out this contribution from the measured straggling parameters:

$$\rho^2 - \rho_w^2 = \rho_s^2 + \rho_n^2 + \rho_f^2. \quad (3)$$

In Fig. 1 we have plotted $\rho^2 - \rho_w^2$ as a function of R_0 for all the differential range experiments reported in this paper and in Ref. 1. As can be seen, the data for Sm

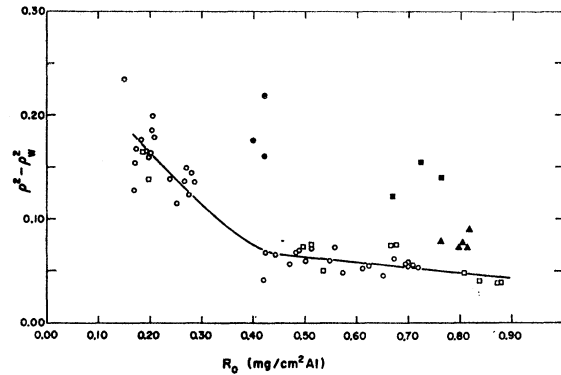


FIG. 1. Representation of the straggling parameter data for Sm^{142} in Al. The squares of the experimental straggling parameters, corrected for the effects of finite target thickness, are plotted against the observed average ranges. The open circles refer to results for (HI, xn) reactions and are taken from the preceding paper (Ref. 1). The open squares are data for reactions leading to Eu compound systems, from the present work. The filled points correspond to reactions leading to Gd compound systems, described in the text, and are as follows: filled circles, $\text{Pr}^{141} + \text{B}^{10}$; filled squares, $\text{Ce}^{140} + \text{C}^{12}$; filled triangles, $\text{La}^{139} + \text{N}^{14}$. The solid curve has been drawn empirically through the data for Sm and Eu compound systems and represents the "normal" relationship for reactions in which alpha-particle emission cannot occur.

and Eu compound systems follow a smooth (within experimental uncertainty) relationship with average range, whereas the points for Gd compound systems are anomalously high. Let us determine which of the terms on the right hand side of Eq. (3) could be responsible for this result. For a given recoil species moving in a given medium, ρ_s^2 is a function only of the recoil energy, and hence is dependent only on the average range.^{7,8} The third term ρ_f^2 should be independent of the reacting system in all our experiments, even though we have little knowledge of its absolute value. Therefore, the unusual behavior observed for Gd compound systems must be due to ρ_n^2 , that is, to the effects of the nuclear reaction.

For the reactions we are considering, ρ_n arises from the vector sum of the momenta imparted to the recoiling nucleus by the emission of particles. In the case of Sm compound systems, only neutrons may be emitted, if we are to observe Sm^{142} , whereas for Eu compound systems, one proton may also be emitted. Because the proton mass is almost the same as the neutron mass, the recoil momenta due to emission of these two particles will not be very different. The presence of a Coulomb barrier will suppress the emission of low-energy protons, and hence any protons which are emitted are likely to have higher average energies than would be associated with neutron emission. This would be expected to result in slightly greater values of ρ_n for Eu compound systems (if protons are emitted) but it is not very likely that we could detect this small difference in our straggling-

⁶ A low average range could also be explained by forward emission of a high-momentum fragment from the compound nucleus. However, to account for the observed effect, the emission would not only have to be strongly forward peaked, but the fragment kinetic energy would have to be unreasonably large.

⁷ J. Lindhard and M. Scharff, Phys. Rev. **124**, 128 (1961).

⁸ J. Lindhard, M. Scharff, and H. E. Schiøtt, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **33**, 14 (1963).

TABLE II. Derived quantities associated with the kinematics of alpha-particle emission from Gd compound systems.

Reaction	E_b (MeV)	$(\rho^2 - \rho_w^2)_{\text{exp}}$	$(\rho^2 - \rho_w^2)_{\text{norm}}$	ρ_α^2	$\frac{\langle V^2 \rangle}{v^2}$	$M_R v^2$	$M_R \langle V^2 \rangle$	$\langle E_\alpha \rangle$ (MeV)
						2 (MeV)	2 (MeV)	
Pr ¹⁴¹ +B ¹⁰	103.8	0.160	0.070	0.090	0.093	5.1	0.47	16.8
	98.3	0.218	0.070	0.148	0.154	5.1	0.79	27.9
	94.2	0.175	0.075	0.100	0.104	4.8	0.50	17.7
Ce ¹⁴⁰ +C ¹²	124.1	0.139	0.050	0.089	0.092	9.9	0.91	32.3
	122.2	0.154	0.052	0.102	0.106	9.3	0.99	35.0
	115.1	0.121	0.056	0.065	0.068	8.6	0.58	20.8
La ¹³⁹ +N ¹⁴	144.6	0.077	0.048	0.029	0.030	10.5	0.32	11.2
	144.6	0.089	0.048	0.041	0.043	10.7	0.46	16.4
	144.5	0.072	0.048	0.024	0.025	10.6	0.27	9.4
	132.6	0.078	0.050	0.028	0.029	9.9	0.29	10.2
	129.8	0.073	0.048	0.025	0.026	10.5	0.27	9.4

parameter data. Because of the somewhat less favorable energetics due to the barrier, it is probable that the Sm¹⁴² produced from Eu compound systems is mainly the result of (HI, xn) reactions followed by β decay.

With Gd compound systems, alpha particle emission becomes a possible competitive mode for Sm¹⁴² production. The recoil momentum due to the emission of an alpha particle would be expected to have a significant effect on ρ_n , due to the fourfold increase in mass as compared with a nucleon. We believe that the anomalously large straggling parameters which we observe for reactions leading to Gd compound systems are due to alpha-particle emission from the compound nuclei. The unfavorable energetics due to the high Coulomb barrier for alpha-particle emission must be considered in light of the high binding energy of the alpha particle, which results in reaction Q values more favorable by about 30 MeV, as compared to the alternative path of (HI, xn) reaction followed by β decay. This is particularly important in the experiments we are considering here, because at the beam energies available the cross sections for the required (HI, xn) reactions are very small.⁹ (This is a consequence of the large values of x needed to yield a mass 142 product.)

We shall analyze our straggling parameter data for Gd compound systems from the point of view that alpha-particle emission does occur. Let us consider the nuclear reaction straggling to be made up of two terms

$$\rho_n^2 = \rho_0^2 + \rho_\alpha^2, \quad (4)$$

where ρ_0^2 represents the contribution from neutron emission and ρ_α^2 is that from alpha-particle emission. From the data in Ref. 1, it appears that nuclear reactions in which different numbers of neutrons are emitted all give the same observed straggling parameters for the same values of the average range. As there is no pronounced dependence on the number of neutrons

⁹ M. Kaplan (unpublished data). Calculations of effective thresholds for these (HI, xn) reactions show that the yields of mass-142 products would be far too low to detect.

emitted, it we compare two equations of the form (3) corresponding to the same average range, their difference should simply yield ρ_α^2 , as indicated from Eq. (4). In Fig. 1 we have drawn an empirical smooth curve through the data for Sm and Eu compound systems. We take this curve as representing, approximately, the "normal" straggling, where no alpha-particle emission can occur. The difference between the experimental values of $(\rho^2 - \rho_w^2)$ for Gd compound systems and the "normal" value, for the same average range, should be just the quantity ρ_α^2 .

We would like to relate ρ_α^2 to the average kinetic energy associated with the emitted alpha particles. A more general relationship between the nuclear reaction straggling parameter ρ_n and the kinematics of the nuclear reaction has been derived in Ref. 4, and we need only specialize the result to apply to the present case. Taking the Sm¹⁴² recoil velocity due to the alpha particle emission to be much smaller than the center-of-mass velocity, and assuming an isotropic angular distribution of alpha particles in the center-of-mass system, we have

$$\rho_\alpha^2 = N^2 \langle V^2 \rangle / 3v^2, \quad (5)$$

where $\langle V^2 \rangle$ is the average square recoil velocity due to alpha-particle emission, v is the center-of-mass velocity, and N is the exponent which appears in the range-velocity relation.^{1,4} We may obtain the value of N from the range-energy curve for Sm¹⁴² in Al, given in Ref. 1. For the recoil energy region we are considering here (5–10 MeV), we have $N = 1.7$. Thus from a derived ρ_α^2 , Eq. (5) yields a value for $(\langle V^2 \rangle / v^2)$. This latter quantity is just the ratio of the average recoil energy from alpha-particle emission to that due to the initial interaction with the beam projectile. From the measured average ranges in Al, and the range-energy curve in Ref. 1, we can easily obtain the energy associated with v^2 and hence also the recoil energy associated with $\langle V^2 \rangle$. Conservation of linear momentum in the process of alpha emission then yields an estimate of the average kinetic energy to be associated with the alpha particles.

The results of the above considerations are given in Table II. The first two columns list the reacting system and the bombarding energy, respectively. Column 3 is the square of the experimental straggling parameter, corrected for effects of finite target thickness. Column 4 gives the same quantity as column 3, but for the condition that alpha-particle emission does not occur. The values in column 4 are obtained from the smooth curve in Fig. 1. The fifth column lists the difference between columns 3 and 4, which we take as representing ρ_α^2 through Eqs. (3) and (4). Columns 6-9 show the kinematic properties derived from ρ_α^2 by means of Eq. (5). They are, respectively, the ratio of recoil energies from alpha emission to that from the beam projectile, the recoil energy due to the impact between beam and target, the kinetic energy due to the alpha emission, and finally, the average energy of the emitted alpha particles.

Our estimates of average alpha-particle energies are admittedly rather crude, and are derived by a simple treatment in an attempt to determine their plausibility as an explanation for the observed large straggling parameters. In view of the approximate nature of the analysis, and considerable scatter in the straggling parameters themselves, the alpha-particle energies given in Table II are probably uncertain by about 50%. Nevertheless, the magnitudes obtained do not seem unreasonable. The Coulomb barrier (computed classically) for alpha-particle emission from Gd nuclei is about 17 MeV, and the derived alpha-particle energies are not too different from this value (within the accuracy involved). It should be pointed out, however, that if we had not assumed the angular distribution of alpha particles to be isotropic in the center-of-mass system, but had instead taken the extreme form $1/\sin\theta$, then the numerical constant in Eq. (5) would be different and the resulting alpha-particle energies would be reduced to two-thirds of the value listed in Table II.

For the three reacting systems in Table II, the derived alpha-particle energies seem to be greatest for $\text{Ce}^{140} + \text{C}^{12}$, lowest for $\text{La}^{139} + \text{N}^{14}$, and in between for $\text{Pr}^{141} + \text{B}^{10}$. We do not understand the significance of these differences, and can only say that there is no obvious correlation with excitation energy or reaction Q value. It would be very useful to study the angular distributions of the Sm^{142} recoils in these reactions, as such measurements can give information similar to

that obtained from the nuclear-reaction straggling parameters.¹⁰

We return now briefly to the effects of uncertainties in identification of the product nuclide in our experiments. It is shown in Ref. 1 that the recoil properties of Sm^{142} and Tb^{149} are the same within an accuracy of about 5%. Consequently, even if our counting data contained contamination from unknown radioactive species with half-lives approximating that of Sm^{142} , our average range measurements would still be valid (i.e., the difference in range between a mass-142 recoil and, say, a mass-144 recoil would be too small to detect). With regard to the range straggling, a contaminant of similar mass formed by a similar reaction mechanism would not significantly change the range distribution attributed to Sm^{142} . In order to account for the large straggling parameters observed with Gd compound systems, a mixture of products would have to be formed in such a way that their individual average ranges straddle the value expected for a single product and their superposition yield an apparent average close to this value. This occurrence would be a very rare coincidence, and could hardly be expected in all three of the reactions studied. Thus, we feel that our experimental data are not much affected by any lack of positive product identification.

Our interpretation of the anomalous straggling parameters in Gd compound systems as arising from alpha-particle emission does, however, depend on Sm^{142} being the product we observe. If unknown Eu or Gd nuclides were contributing to our data, the straggling would still be anomalous but could not be attributed to emission of alpha particles. We have attempted to explore these possibilities by calculations of reaction thresholds and beta-decay energies, with the result that only Gd^{144} or Eu^{143} could be contaminants, if they have an appropriate combination of half-lives. We have seen no evidence in our counting data for the growth of the known radioactive daughters of these species.

ACKNOWLEDGMENTS

The author would like to thank Professor John M. Alexander for many stimulating discussions, and Dr. Richard D. Fink for assistance with the experiments. The support of Professor E. Robert Beringer and the cooperation of the HILAC staff are gratefully appreciated.

¹⁰ G. N. Simonoff and J. M. Alexander, University of California Lawrence Radiation Laboratory Report UCRL-10099-Rev., 1962 (unpublished).