

Recoil Ranges of Products from Reactions of Copper with 11–43-MeV He⁴ Ions*

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Average projected ranges have been determined for the following reactions of copper with 11–43-MeV He⁴ ions: Cu⁶⁵(α, n), Cu⁶⁵($\alpha, 2n$), Cu⁶⁵($\alpha, 3n$), Cu⁶⁵($\alpha, \alpha n$), and Cu⁶³(α, n). The results have been compared with calculations based on the assumption of compound-nucleus formation. Good agreement has been obtained at all energies for the (α, n) reactions. On the other hand, the ranges of Cu⁶⁴ are considerably smaller than those predicted by the theory, indicating that there is a significant contribution of a direct process to the ($\alpha, \alpha n$) reaction.

I. INTRODUCTION

MEDIUM-energy nuclear reactions are thought to involve either compound-nucleus formation or a direct-interaction process. Considerable interest exists in ascertaining which of these two mechanisms is of importance for a particular reaction at a given bombarding energy.

A relatively direct method of distinguishing between these processes involves the determination of the momentum transferred to the product nucleus. In the case of a compound-nuclear reaction the projectile transfers its entire momentum to the compound nucleus. The subsequent evaporation of particles is symmetric about 90° in the center-of-mass system and, in the common case where the recoil range of the product is proportional to its energy, increases the momentum of the recoiling nucleus. On the other hand, in most direct interactions the momentum brought in by the projectile is to a large extent immediately removed by the predominantly forward emission of particles. The momentum of the product will in most instances thus be considerably smaller than in the case of compound-nucleus formation.

A valuable technique for the determination of the momentum transferred to the product nuclide is the measurement of its recoil range. The thick-target method, by which the average projected range of the recoils in the target material can be determined, is useful for this purpose. This technique has been used in a number of recent instances^{1–6} for the study of He⁴-induced reactions. In order to relate the measured ranges to the momentum transferred to the struck nucleus, a range-energy relation is needed. The theo-

retical relation due to Lindhard *et al.*⁷ may be used for this purpose. Its applicability to experiments of this type has been demonstrated⁸ in a study in which recoils of known energy were produced by radiative capture reactions.

The present study is an application of the thick-target recoil technique to the reactions of copper with He⁴ ions. We report results for the (α, n), ($\alpha, 2n$), ($\alpha, 3n$), and ($\alpha, \alpha n$) reactions of Cu⁶⁵ and also for the Cu⁶³(α, n) reaction in the energy range of 11–43 MeV. The excitation functions of these reactions have been previously studied by Porile and Morrison⁹ and a comparison with the statistical theory¹⁰ gave evidence for compound-nucleus formation. The present work in the main corroborates this conclusion.

II. EXPERIMENTAL PROCEDURE

The targets used in this work consisted of copper foils having a thickness of 2.4 mg/cm². A number of experiments were also performed in which the targets consisted of highly enriched¹¹ (99.7%) Cu⁶⁵. In these cases self-supporting target foils were prepared by electrodeposition of Cu⁶⁵ onto nickel and subsequent mechanical removal of the latter. These enriched foils had a thickness of 4–6 mg/cm². The catcher foils consisted of 0.001-in.-thick aluminum of high purity (99.999%). All foils were carefully cleaned prior to irradiation. The target foils were inspected to ascertain that their surface was free of a visible oxide layer and other imperfections.

The target stack consisted of 5–10 targets and forward catcher foils as well as of appropriate aluminum degrading foils. In one irradiation a few additional 0.001-in.-thick aluminum foils were included in order to determine the activity due to impurities. In all cases the activation correction was found to be less than 3%. The bombarding energy at a given position in the target

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² T. Matsuo and T. T. Sugihara, *Can. J. Chem.* **39**, 697 (1961).
T. Matsuo, J. M. Matuszek, N. D. Dudey, and T. T. Sugihara, *Phys. Rev.* **139**, B886 (1965).

³ N. T. Porile, A. M. Poskanzer, and M. Rho, *Phys. Rev.* **128**, 242 (1962); N. T. Porile, *ibid.* **127**, 224 (1962).

⁴ L. Bryde, N. O. Lassen, and L. O. R. Poulsen, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **33**, No. 8 (1962).

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⁶ A. Ewart, C. Valentine, and M. Blann, *Nucl. Phys.* **69**, 625 (1965).

⁷ J. Lindhard, M. Scharff, and H. E. Schiott, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **33**, No. 14 (1963).

⁸ N. T. Porile, *Phys. Rev.* **135**, A1115 (1964).

⁹ N. T. Porile and D. L. Morrison, *Phys. Rev.* **116**, 1193 (1959).

¹⁰ I. Dostrovsky, Z. Fraenkel, and G. Friedlander, *Phys. Rev.* **116**, 683 (1959).

¹¹ Obtained from Oak Ridge National Laboratory.

stack was obtained from the incident energy with the aid of a range-energy relation based on that of Bichsel *et al.*¹² for protons.

Irradiations were performed with the He⁴-ion beam of the 60-in. cyclotron at Argonne National Laboratory. The duration of the bombardments ranged from 2 to 40 min and the beam current varied between 1 and 2 μ A. The target stack was mounted in an evacuated target holder that also served as a Faraday cup.

After irradiation the appropriate foils were separated from the stack. Gallium and copper were radiochemically separated by procedures given elsewhere.⁹ Chemical yields were determined gravimetrically. In the experiments designed to determine the recoil ranges of Ga⁶⁸ it proved desirable to determine directly the activity of the foils without prior chemical separation.

The activity measurements were performed with the following two instruments. A γ - γ coincidence spectrometer was used to detect the annihilation radiation resulting from the positron decay of 68-min Ga⁶⁸, 9.5-h Ga⁶⁶, and 12.9-h Cu⁶⁴. A 3 in. \times 3 in. NaI(Tl) detector coupled to a 400-channel pulse-height analyzer was used to measure the 90- and 182-keV γ rays of 78-h Ga⁶⁷.

III. RESULTS

In a thick-target integral range experiment the average range in the target material, projected along the beam direction, is given by

$$R_p = FW, \quad (1)$$

where F is the fraction of the total activity due to a given nuclide found in the forward catcher foil and W is the target thickness. This expression is based on the assumption that the formation cross section for the nuclide of interest is constant throughout the target. This is often not the case for reactions having steep excitation functions when the targets are sufficiently thick to appreciably degrade the energy of the beam. This situation has been considered by Porile^{8,3} and more recently by Ewart *et al.*⁶ It can be shown^{8,3} that the recoil range is given by

$$R_p = FW \left[1 + \frac{1}{2} (\sigma_B - \sigma_F) \right], \quad (2)$$

if a linear variation of cross section over an energy range corresponding to the target thickness is assumed. In this expression σ_F is the cross section for a particular reaction at the forward side of the target and σ_B that at the backward side. It is to be understood that the beam is incident on the target at the backward side.

The experimental FW values were corrected according to Eq. (2) by use of the measured⁹ excitation functions for the reactions of interest. The experimental and corrected values are summarized in Table I. In most instances the correction was less than 3% and

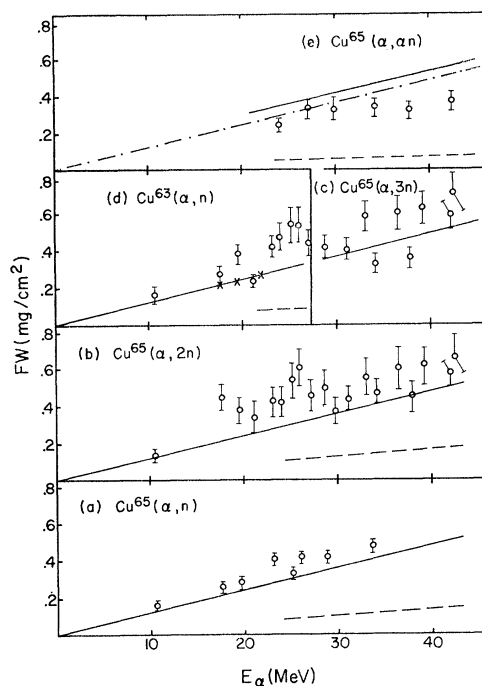


Fig. 1. Average projected ranges of reactions of Cu⁶⁵ and Cu⁶⁸ with He⁴ ions. Solid lines—calculation assuming compound-nucleus formation. Dot-dashed line—compound-nucleus calculation neglecting the effect of evaporation. Dashed lines—calculation assuming a particular type of direct interaction. The \times symbols in 1(d) are data from Ref. 4.

was not made. However, in the case of the $(\alpha,2n)$ reaction, the correction amounted to as much as 33%.

The corrected recoil ranges are plotted as a function of bombarding energy in Fig. 1. The errors in the data range up to 25% and are based on the estimated non-uniformity of the targets, the uncertainty in the chemical-yield determinations, errors in the activity measurements, and uncertainty in the correction given by Eq. (2). The scatter of the individual measurements is in most cases consistent with these estimates.

The formation of Ga⁶⁶ from natural copper is exclusively due to the Cu⁶³(α,n) reaction up to 27 MeV. Above this energy the Cu⁶⁵($\alpha,3n$) reaction is also energetically possible and its contribution to Ga⁶⁶ formation quickly overshadows that of the Cu⁶³(α,n) reaction. The experimental points shown in Fig. 1(c) have therefore been attributed to the Cu⁶⁵($\alpha,3n$) reaction.

Recoil ranges for the Cu⁶³(α,n) reaction have previously been measured by Bryde *et al.*⁴ Their results have been included in Fig. 1(d) and are seen to agree with the present data. The results at 11–16 MeV may also be compared with the Cu⁶³(α,γ) recoil ranges.⁸ The latter are on the average about 25% lower than the present values but this difference is within experimental error. As the (α,γ) experiment was performed with a much lower beam current the discrepancy may be

¹² H. Bichsel, R. F. Mozley, and W. A. Aron, Phys. Rev. **105**, 1788 (1957).

TABLE I. Experimental and corrected average projected ranges.

Bombarding energy (MeV)	Cu ⁶⁵ (α, n)		Cu ⁶⁵ ($\alpha, 2n$)		Cu ⁶⁵ ($\alpha, 3n$)		Cu ⁶⁵ ($\alpha, \alpha n$)		Cu ⁶⁸ (α, n)	
		FW	FW	FW _{corr} ^a	FW	FW	FW _{corr}	FW	FW	FW _{corr}
42.5			0.654		0.702					
42.3			0.574 ^b		0.585 ^b		0.366 ^b			
39.3			0.615		0.628					
37.9			0.447 ^b		0.363 ^b		0.321 ^b			
36.6			0.601		0.601					
34.2			0.458 ^b		0.329 ^b		0.338 ^b			
33.6	0.471									
33.1			0.547		0.582					
31.1			0.435		0.402					
29.9			0.375 ^b				0.320 ^b	0.336		
28.8	0.423		0.495		0.418					
27.1			0.450				0.326 ^b	0.342	0.468	0.440
26.0	0.423		0.609						0.577	0.537
25.3	0.327		0.534						0.570	0.532
24.0			0.418				0.237	0.254	0.477	0.453
23.2	0.399		0.424						0.412	
21.0			0.300	0.333					0.229	
19.6	0.281		0.320	0.374					0.382	
17.6	0.262		0.333	0.443					0.277	
10.6	0.164		0.131						0.164	

^a Corrected values of *FW* are only given in those instances where they differ by more than 3% from the measured values. The *FW* values are given in mg/cm².

^b Enriched Cu⁶⁵ targets.

indicative of a small spurious enlargement of the present ranges due to heating.

IV. DISCUSSION

The recoil ranges are in most cases seen to increase with bombarding energy. This trend is a manifestation of the increasing momentum of the incident He⁴ ion. In order to determine if this trend indicates compound-nucleus formation a quantitative comparison must be made. We have taken a somewhat different approach than has heretofore been used. The momentum of the compound nucleus is first corrected for the effect of particle evaporation and then converted to the corresponding range in the target material. The range is finally projected along the beam direction for comparison with experiment. The correction for evaporation is made on the basis of calculated energy spectra instead of on that of average energies.

The calculation consists of an adaptation^{13,14} of the Monte Carlo evaporation code of Dostrovsky *et al.*¹⁰ In this adaptation the velocity and direction of motion of the residual nucleus is computed. The starting nuclide for the evaporation calculation is the compound nucleus with an excitation energy corresponding to a particular bombarding energy. The velocity of the compound nucleus $V_z(\text{CN})$ is directed along the beam direction and is obtained from the incident energy by conservation of momentum, i.e.,

$$V_z(\text{CN}) = \frac{(2M_\alpha E_\alpha)^{1/2}}{M_T + M_\alpha}, \quad (3)$$

where M_α and E_α are, respectively, the mass and energy in the laboratory of the incident particle and M_T is the mass of the target.

The velocity of the recoiling nucleus is computed at each step of the evaporation process by the formula

$$V(n) = \left[\frac{2M_p(n)E(n)}{M(n)(M(n) + M_p(n))} \right]^{1/2}, \quad (4)$$

where $M(n)$ and $M_p(n)$ are, respectively, the masses of the residual nucleus and the particle evaporated in the n th evaporation step and $E(n)$ is the calculated evaporation channel energy. The choice of two random numbers determines the direction of recoil on the assumption of isotropic evaporation. The resulting velocity components are algebraically added to those of prior steps and the velocity of the product nucleus obtained after N_{max} evaporation steps is then given as

$$V_F = \left[(V_z(\text{CN}) + \sum_{n=1}^{N_{\text{max}}} V_z(n))^2 + \left(\sum_{n=1}^{N_{\text{max}}} V_Y(n) \right)^2 + \left(\sum_{n=1}^{N_{\text{max}}} V_X(n) \right)^2 \right]^{1/2}. \quad (5)$$

The kinetic energy of the product follows immediately and the recoil angle relative to the beam is given by

$$\cos\theta_L = (V_z(\text{CN}) + \sum_{n=1}^{N_{\text{max}}} V_z(n)) / V_F. \quad (6)$$

The evaporation program incorporated a range-energy relation based on the work of Lindhard *et al.*⁷ This relation was obtained from the reduced range-

¹³ N. T. Porile and S. Tanaka, Phys. Rev. **135**, B122 (1964).

¹⁴ N. T. Porile, Phys. Rev. **135**, B371 (1964).

energy (ρ - ϵ) plot of Lindhard *et al.*⁷ for an appropriate value of the electronic stopping parameter k . The effect of scattering was taken into account by converting the total path lengths to ranges projected along the initial direction of motion in the manner suggested by these authors. It was found that over the energy range of interest the range-energy relation for gallium nuclides in copper could be accurately approximated by the relation $R(\text{mg}/\text{cm}^2) = 0.186E(\text{MeV})$, while that for Cu^{64} was given by $R = 0.213E$. The kinetic energy of the final product was thus converted to a range by the appropriate relation. Finally, the projected range was obtained by the relation

$$R_p = FW = R \cos\theta_L. \quad (7)$$

One thousand iterations were performed for a given bombarding energy and the resulting FW values for each product were averaged for comparison with experiment. It should be noted that the kinematics of the reactions require that $\theta_L < 90^\circ$, so that all FW values are positive. The calculation was performed at 5-MeV intervals over the energy range of interest. The level density parameter was chosen as $a = A/20$.

The results of the calculation are given by the solid lines in Fig. 1. In the case of the (α, xn) reactions the effect of evaporation is practically negligible and the calculated curves essentially coincide with those obtained from Lindhard *et al.*⁷ On the other hand, evaporation increases the projected ranges of the $(\alpha, \alpha n)$ product by about 15% above the Lindhard values. The latter are given by the dot-dashed line in Fig. 1(e). This difference between these two types of reactions is due to the much larger momentum of an evaporated alpha particle.

The effect of evaporation has previously been estimated on the basis of the formalism developed by Winsberg and Alexander.¹⁵ Their treatment is based on the use of an estimated value of the average recoil velocity due to evaporation. On the basis of this formalism we find that the effect of evaporation on the projected ranges amounts to about 3–8% for the (α, xn) reactions and to 30% for the $(\alpha, \alpha n)$ reaction at 40 MeV. These values are somewhat larger than those obtained from the present Monte Carlo calculation.

The comparison with experiment for the (α, xn) reactions shows that most of the measured values lie significantly above the curve. Since no reasonable mechanism for (α, xn) reactions will lead to this effect we ascribe the discrepancy to possible heating or scattering effects as well as to the uncertainty in the range-energy relation. It seems reasonable to conclude that the (α, xn) reaction ranges are consistent with compound-nucleus formation.

The ranges of Cu^{64} lie significantly below the calculated line, particularly at the higher energies. This fact suggests that a direct process contributes at least partially to the $(\alpha, \alpha n)$ reaction. A similar conclusion was obtained by Blann and Ewart⁵ for the $\text{Ni}^{58}(\alpha, \alpha n)$ reaction at 46–68 MeV.

In order to test the sensitivity of the results to a contribution from a direct process we have estimated the ranges to be expected for the latter. The following simple models for the direct interaction have been assumed for this purpose. The (α, xn) reactions were assumed to involve the direct emission of a single neutron at an average angle of 20° to the beam. The kinetic energy of the prompt neutron was chosen so that enough energy was left behind to evaporate $(x-1)$ neutrons. The $(\alpha, \alpha n)$ reaction was assumed to involve the inelastic scattering of the incident He^4 ion followed by the evaporation of one neutron. A scattering angle of 20° was chosen on the basis of the angular-distribution data obtained by Broek *et al.*¹⁶ for the $\text{Ni}^{58}(\alpha, \alpha')$ reaction. The recoil energies obtained on the basis of the above assumptions were converted to projected ranges by use of the relation of Lindhard *et al.*⁷

The results are given by the dashed lines in Fig. 1. It is seen that the ranges are as much as a factor of 8 smaller than the values expected for compound-nucleus formation. The magnitude of this difference depends, of course, on the assumed scattering angle and on the exact form of the direct mechanism. In the absence of detailed information about these factors the calculation is highly tentative. Nonetheless, it may be concluded that a direct process is unimportant for the (α, xn) reactions in the energy range of interest. This is to be expected for the $(\alpha, 2n)$ and $(\alpha, 3n)$ reactions since our measurements correspond to the early portions of their excitation functions. On the other hand, the (α, n) data correspond to the high-energy tail of the excitation function where a direct process might have seemed more likely. Evidently, the compound-nuclear mechanism can in some instances account for the formation of a product a good 20 MeV past its peak energy. By contrast, in the case of the $(\alpha, \alpha n)$ reaction direct inelastic scattering could account for perhaps 30% of the cross section at 40 MeV.

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¹⁵ L. Winsberg and J. M. Alexander, Phys. Rev. **121**, 518 (1961).

¹⁶ H. E. Broek, T. H. Braid, J. L. Yntema, and B. Zeidman, Phys. Rev. **126**, 1514 (1962).