

second $1+$ excited level in Rh^{108} would not be inconsistent with the predictions of these latter coupling rules.

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Compound Nucleus Formation Mechanisms in Reaction of Heavy Ions with Medium-Mass Elements

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Excitation functions and range distributions of heavy products formed by the reaction of O^{16} , N^{14} , and C^{12} with Mn^{55} and Co^{59} have been measured. Results show that the bulk of products, with mass numbers greater than that of the target, are formed exclusively by decay of a compound nucleus formed by complete fusion of target and projectile. One nuclide, Cu^{61} , can also be formed from O^{16} bombardment of Co^{59} (but not from the formally similar compound system $\text{Mn}^{55} + \text{Ne}^{20}$) by a mechanism in which only part of the momentum of the projectile is transferred. It appears to be representative of products with mass near that of the target, which can be formed by nucleon transfer in a grazing reaction. No evidence is found for a general type of "buckshot" mechanism. Small yields of Na^{24} and P^{32} appear to result from a fission or fragmentation mechanism.

INTRODUCTION

SEVERAL mechanisms of heavy-ion reactions have been proposed in recent years. However the validity and relationship of these mechanisms in accounting for the major yields of heavy products (mass comparable to that of target) is not yet clear. At small and at large impact parameters, respectively, the situation is relatively straightforward. In a headon (or near headon) collision a compound nucleus of low angular momentum, de-exciting to the expected spallation products is presumably formed. In distant collisions, in which the Coulomb barrier is not penetrated, tunneling of a nucleon may occur,¹ generally leading to products one mass number removed from target and projectile, respectively.^{2,3}

The role of collisions of intermediate impact parameters in contributing to the observed heavy products is less clear. One view is that any collision in which the Coulomb barrier is penetrated will lead to a compound nucleus containing all nucleons and likely to have high

average angular momentum⁴ (hereafter referred to as a "complete fusion compound nucleus" CFCN). Another view, generally referred to as the "buckshot" hypothesis,⁵ holds that the projectile tends to break up in the field of the target and that, depending on the impact parameter, some fraction of its constituent nucleons is captured. A complete range of "partial fusion compound nuclei" (PFCN) ranging in mass from that of the target to that of the CFCN is thus to be expected. These compound nuclei will be excited to an extent corresponding largely to the kinetic energy of the captured nucleons, and will de-excite by the familiar evaporation process. Product distribution found in early work⁶ originally suggested that such a range of compound nuclei were indeed formed. However, it was later pointed out that preferential alpha emission, which might be expected in complete fusion compound nuclei formed at high impact parameters and possessing many units of angular momentum, can also account for these product distributions.^{6,7} Indications of such preferential alpha

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¹ G. Breit and M. E. Ebel, *Phys. Rev.* **103**, 679 (1956).

² J. A. McIntyre, T. L. Watts, and F. C. Jobs, *Phys. Rev.* **119**, 1331 (1960).

³ R. Kaufmann and R. Wolfgang, *Phys. Rev.* **121**, 206 (1961).

⁴ T. D. Thomas, *Phys. Rev.* **116**, 703 (1959).

⁵ K. F. Chackett, J. H. Fremlin, and D. Walker, *Phil. Mag.* **45**, 735 (1954).

⁶ L. M. Parfanovich, M. V. Rabin, and A. M. Sevachemova, *Soviet Phys.—JETP* **31**, 188 (1956).

⁷ M. Tamers and R. Wolfgang, *Phys. Rev.* **117**, 812 (1959).

emission have been found in heavy-ion bombardment of nickel.⁸

Another mechanism for collision at intermediate impact parameters is the so-called grazing model.⁹ In this mechanism it is postulated that the Coulomb barrier is penetrated, but that because of the relative motion of the two entities normal to their line-of-centers, the zone of contact is frictionally heated and only weakly binding. As a result, centrifugal and Coulomb forces reseparate the system and no compound nucleus is formed. On separation several nucleons may transfer from one nucleus to the other. The observation of products several nucleons removed from the projectile but possessing its approximate velocity and with a strongly forward peaked angular distribution led to suggestion of this grazing mechanism.⁹ At incident energies well over the barrier the grazing mechanism should represent a substantial part of the geometric cross section. Much of this cross section probably represents grazing collisions which result in breakup of the projectile.⁶ Experiments on the interaction of 160-MeV O^{16} and 140-MeV N^{14} with heavy elements show of the order of several hundred millibarns of cross section for fast alpha-particle production.¹⁰ These alphas have a range and angular distribution consistent with the grazing hypothesis.

Grazing reaction may result in the abstraction from, or addition to the target of one or more nucleons. In the sense that partial fusion compound nuclei are thus created, grazing may thus be regarded as a mechanism of the "buckshot" phenomenon. However, it is important to recognize that the "buckshot" hypothesis suggests a more or less equal probability for capture of any number (or all) of the projectile nucleons by the target. On the other hand the grazing mechanism implies transfer of only a few nucleons. The resulting entities are expected to be compound nuclei of mass similar to the target and may have sufficient energy for particle evaporation.

In this study we have attempted to define the validity and relationship of these mechanisms, as encountered in the reaction of heavy ions with medium mass nuclides. With considerable information already available on the lighter products (masses up to about that of the projectile)^{8,9,10} this study has been concerned with the heavier products. Excitation functions of a number of radioactive products from several systems, particularly $O^{16} + Co^{59}$, have been obtained. However, as implied above, excitation functions alone are not always a sufficiently sensitive criterion of the reaction mechanism. The same product may be formed by nucleon and alpha-particle emission from a CFCN containing the entire energy and angular momentum of the system; or by evaporation from a PFCN which is less excited

because it contains only part of the incident nucleons and their kinetic energy. Measurement of the recoil range of the product provides a further criterion of its formation mechanism. From it can be deduced the momentum with which the compound nucleus was formed. If it is a CFCN this momentum must of course correspond to that of the projectile. In PFCN events the momentum will depend upon the fraction of the incident nucleons captured, and the velocity and angle of recoil of the uncaptured projectile residue. At energies well above the Coulomb barrier the velocity and direction of this residue approximates that of the incident particle.⁹ As a result the momentum and range of the PFCN will be less than that of the corresponding CFCN and should be a measure of the number of nucleons transferred.

The systems chosen were C^{12} impinging on Co^{59} , O^{16} on Mn^{55} (both forming an As^{71} compound system), O^{16} on Co^{59} , and N^{14} on Co^{59} . Most of the excitation functions were measured by the usual stacked foil technique. In the range measurements recoils from a thin film of the target were caught in thin aluminum foils. Such range measurements have already been made by Alexander and Winsberg¹¹ in experiments with a number of heavy elements. The present study in the medium-mass range is facilitated by the relatively large recoil range and correspondingly smaller relative straggling of the products. This minimizes ambiguity in the interpretation of the data.

EXPERIMENTAL

Further details on the procedures outlined below may be obtained from the thesis of Read.^{12,13}

Target Assemblies

Three types of targets were used in the experiments: (1) Co foils, thick with respect to the heavy-ion range, were used for determinations of P^{32} and Na^{24} . This was necessary because of the difficulty of obtaining thin Co foils which were still thick enough to stop these products. (2) Targets for excitation functions of nuclides other than Na^{24} and P^{32} , consisted of stacks of Al foils (~ 4.6 mg/cm²) coated with 200 to 800 μ g/cm² of cobalt or manganese oxides. These foils were prepared by spraying the metal nitrates in 50% alcohol solution onto hot aluminum foil and subsequently igniting to 300°C. The resulting coating was measured to be uniform in thickness within 20%. The target-element coated foils were separated by other aluminum foils of appropriate thickness, which served to degrade the energy of the beam as it penetrated the stack. (3) In recoil-range determination experiments a coated foil with 100–200

⁸ W. J. Knox, A. R. Quinton, and C. Anderson, *Phys. Rev.* **120**, 2120 (1960).

⁹ R. Kaufmann and R. Wolfgang, *Phys. Rev.* **121**, 192 (1961).

¹⁰ H. C. Britt and A. R. Quinton, *Phys. Rev.* **124**, 877 (1961).

¹¹ J. Alexander and L. Winsberg, *Phys. Rev.* **121**, 518, 529 (1961).

¹² J. B. J. Read, Ph.D. thesis, Yale University, 1961 (unpublished).

¹³ W. W. Meinke, University of California Radiation Laboratory Report. UCRL-432, 1949 (unpublished).

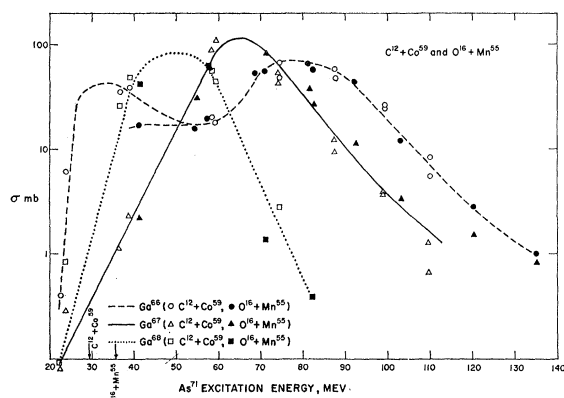


FIG. 1. Excitation functions for products of the As^{71} compound system; as formed by two separate modes. As^{71} excitation energy = $(59/71)\text{C}^{12}$ bombarding energy + 5.6 MeV; or, As^{71} excitation energy = $(55/71)\text{O}^{16}$ bombarding energy + 5.3 MeV. (5.6 and 5.3 MeV represent estimated Q values for reaction of $\text{C}^{12} + \text{Co}^{59}$ and $\text{O}^{16} + \text{Mn}^{55}$, respectively, to form an As^{71} compound system in the ground state.) Arrows show nominal position of Coulomb barrier assuming reduced radius of 1.5×10^{-13} cm. (These values are quite approximate in view of possible error in energy scale of lower energies.)

$\mu\text{g}/\text{cm}^2$ cobalt on the downbeam side was backed by about 20 aluminum foils 100 to 200 $\mu\text{g}/\text{cm}^2$ thick. These catcher foils were individually weighed, to an accuracy of $\pm 10 \mu\text{g}/\text{cm}^2$.

Bombardment

The target stack 1 in. in diameter was clamped by a ring onto a brass cylinder which slipped into a Faraday cup $1\frac{1}{2} \times 6$ in. Particles of energy of about 10 MeV/nucleon, as determined by a deflection system were collimated to a beam $\frac{1}{4}$ to $\frac{1}{2}$ in. in diameter before striking the target. To degrade this energy to values less than 10 MeV/nucleon, appropriate aluminum foils were placed on top of the target stack. The maximum energy spread of the beam was ± 0.2 MeV/nucleon.

Beam intensity was integrated by a calibrated Cary electrometer. The efficiency of the Faraday cup in retaining secondary electrons was checked by showing that a strong magnetic field imposed on it did not affect the charge collected. In cases where the half-life of the isotope sought was not long compared to the bombardment, correction was made for time variation of beam intensity.

Chemical Separation

Target and catcher foils were separately dissolved. Chemical procedures were adapted from procedures described elsewhere.¹² The chemical yields of these procedures were determined by weighing, or by redissolution and analysis after counting had been completed.

Enough time was allowed between bombardment and separation for the precursors of the Ga^{67} and Cu^{61} to decay almost completely. These products, therefore, represent cumulative isobaric yields while those of

Ga^{68} and Cu^{64} are independent yields. The Ga^{66} includes 50 to 90% of that deriving from its Ge^{66} precursor making these cross sections somewhat uncertain in their absolute magnitude.

Counting

Filter papers bearing the radioactive products in a few milligrams of carrier were mounted on Al plates and covered with 1-mg/cm² Mylar foils. They were counted using an end window gas flow proportional counter. Counting efficiencies were taken from Brookhaven data on identical counters¹⁴ or were measured by counting the isotope in question with NaI(Tl) scintillation or gas-filled x-ray counter of known geometry and efficiency. They are Ga^{66} :0.47; Ga^{67} :0.25; Ga^{68} :0.49; Cu^{61} :0.51; Cu^{64} :0.37; Na^{24} :0.49; P^{32} :0.51 (all for samples on top shelf covered with 1-mg/cm² aluminized Mylar).

RESULTS

Excitation Functions

The results of the cross section measurements for production of gallium and copper isotopes are recorded in Figs. 1–3. The excitation energy ordinate gives the internal energy available in the system of the compound nucleus and includes the binding energy of the projectile to the target. Arrows on this ordinate show the classical barrier penetration energy, as calculated using 1.5×10^{-13} cm as the reduced radius parameter and expressed in terms of the excitation energy of the resulting compound system.

The resulting excitation functions are on the whole qualitatively similar to those found by Karamyan and Pleve¹⁵ for O^{16} bombardment of vanadium. Individual

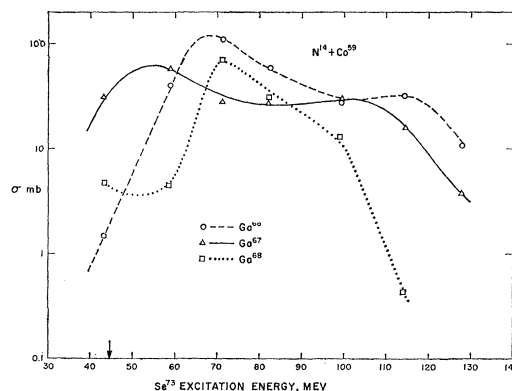


FIG. 2. Excitation functions for products of the Se^{73} compound system. Se^{73} excitation energy = bombarding energy + 8.9 MeV.

¹⁴ We are indebted to Dr. J. B. Cumming for this information.
¹⁵ A. S. Karamyan and A. A. Pleve, Soviet Phys.—JETP **37**, 654 (1959). This article reports the existence of a high energy "tail" in the excitation functions $\text{V}^{51}(\text{C}^{12}, 2n) \text{Cu}^{61}$ and $\text{Nb}^{93}(\text{C}^{12}, 2n) \text{Ag}^{103}$. This appears to be a unique observation insofar that nothing similar was observed in the present and earlier work (see reference 7.). We are inclined to regard it with some reserve since no chemical separations were made and identification depended primarily on analysis of half-lives in complex decay curves.

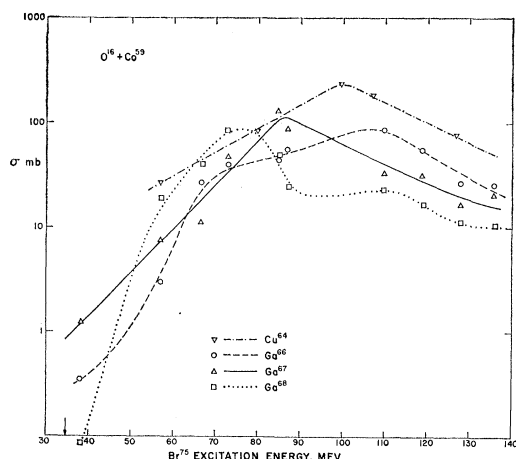


FIG. 3. Excitation functions for products of the As^{75} compound system. As^{75} excitation energy = bombarding energy + 2.4 MeV.

points are accurate to about $\pm 30\%$ on the cross-section scale (this error is larger for cross section $\lesssim 1$ mb) and ± 8 MeV on the energy scale; and the points are too widely spaced to find much detailed structure. Nevertheless the curves show clearly just those features which would be expected from a classic model of evaporation from a compound nucleus formed by fusion of target and projectile (CFCN):

(1) Figure 1 shows excitation functions for product formation from the compound system As^{71} as formed in two ways: O^{16} on Mn^{55} and C^{12} on Co^{59} . Within experimental error it appears that decay of the compound system to form gallium products is independent of the method by which it was formed. (The relatively low values of the cross sections at 41 MeV as formed from $\text{O}^{16} + \text{Mn}^{55}$ probably merely reflect the higher Coulomb barrier for the formation of this system, which would be expected to have a depressing effect on yields in this energy region.) This demonstration of independence of formation and decay is of course a classic test of the compound nucleus hypothesis.¹⁶ It would seem to be a most unlikely coincidence that such a result could be obtained from a "buckshot"-type mechanism.¹⁷

¹⁶ S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).

¹⁷ Some displacement of the two sets of functions might be expected as a result of the higher average angular momentum brought in by the O^{16} relative to the C^{12} . Higher angular momentum might have two consequences which however tend to cancel one another: (1) More energy is tied up in rotation and unavailable for evaporation. (2) Barriers to evaporation may be reduced in a nucleus distorted to a prolate shape by its high spin. In the present example, the O^{16} carries in only $\frac{1}{3}$ more angular momentum than does the C^{12} . In any case it appears that high-impact-parameter events do not lead to formation of a CFCN, but rather to grazing events which will not yield isotopes more than a few mass numbers removed from the target (see subsequent discussion). The fact that no displacement of excitation functions is observed is therefore not particularly surprising. Karamyan and Plevé (see reference 15) report that in O^{16} bombardment of vanadium peaks in the excitation functions occur at excitation energies 10 to 15 MeV higher than with corresponding proton induced processes. Such an effect was not obvious in our work. The approximate energy required per nucleon emitted (Table I) tends to be somewhat lower than found by Karamyan and Plevé,

(2) The width of the peaks is approximately in proportion to the number of nucleons emitted, as would be expected from a statistical evaporation model. As a consequence peaks found at higher excitation energies are invariably broader. In agreement with earlier observations in our laboratory, no high energy "tails" of products peaking at low energies were observed.^{7,15}

(3) In a number of cases the functions show double peaks. (See Figs. 1 and 2 and Table I.) The same product may be formed at a low energy by an emission including an alpha particle, and at about 45 MeV higher when this alpha emerges as four nucleons. An example is Ga^{66} from C^{12} on Co^{59} as formed by (αn) and $(2p3n)$ evaporation. The lower peak is frequently not observed because the energy corresponding to its maximum cross section is below or near the Coulomb barrier cutoff (e.g., for Ga^{67} from C^{12} on Co^{59} a peak would be expected at ~ 20 -MeV excitation energy). At higher energies these double peaks tend to be poorly resolved because of their breadth when many nucleons are emitted.

(4) The excitation energies at which yield peaks occur correspond to the number of particles which must have been emitted from the compound system. (See Table I.) Approximately 15-MeV excitation energy appears to be required for the emission of each additional nucleon or alpha particle.¹⁷ Such behavior is, of course, to be expected from a CFCN, but is hardly obvious from a "buckshot" model.

The above criteria cannot be clearly applied to Cu^{61} from O^{16} on Co^{59} . This is primarily because the large number of ways in which 14 nucleons can be emitted to yield a very broad, featureless excitation function. If conclusions were based on excitation functions alone, although sensitive indications are lacking, there would be little reason to suppose that Cu^{61} was not formed in a manner similar to that of the other products. As will become apparent below, such a guess would be incorrect.

TABLE I. Excitation energies at cross-section peaks.

Compound system	Product	Probable mode of decay	Excitation energy at peak (MeV)
As^{71a}	Ga^{68}	$2pn$	50
As^{71a}	Ga^{67}	$2p2n$	65
As^{71}	Ga^{66}	αn	35
As^{71a}	Ga^{66}	$2p3n$	80
Se^{73}	Ga^{68}	αp	< 40
Se^{73}	Ga^{68}	$3p2n$	75
Se^{73}	Ga^{67}	αpn	55
Se^{73}	Ga^{67}	$3p3n$	~ 100
Se^{73}	Ga^{66}	$\alpha p2n$	70
Se^{73}	Ga^{66}	$3p4n$	~ 115
Br^{75}	Ga^{68}	$\alpha 2pn$	75
Br^{75}	Ga^{68}	$4p3n$	$\sim 115^b$
Br^{75}	Ga^{67}	$\alpha 2p2n$	90
Br^{75}	Ga^{66}	$\alpha 2p3n$	~ 105
Br^{75}	Cu^{64}	$2\alpha 2pn$	100

^a From both C^{12} on Co^{59} and O^{16} on Mn^{55} .

^b Appears only as shoulder, not as separate peak.

TABLE II Upper limits on cross sections from system $O^{16}+Co^{59}$

Product	Bombardment energy (MeV)	Upper limit (mb)
Se ⁷²	159	10
Se ⁷³	159	1
As ⁷⁰	159	2
As ⁷¹	159	2
As ⁷²	159	2
Zn ⁶²	138	5
Ni ⁵⁷	159	10
Co ⁵⁵	159	2
Fe ⁵²	159	10

Cu^{61} from O^{16} on Co^{59} may be formed by non-CFCN mechanisms and provides an example of how excitation function data alone can be insufficient to determine a mechanism.

Unobserved Products

Nine radioactive products of reactions of O^{16} with Co^{59} were sought but not found. These results are recorded in Table II. The selenium and arsenic isotopes would be expected to be absent on the basis of a CFCN model. A compound nucleus containing all the energy of a 159-MeV O^{16} projectile would de-excite by the emission of so many nucleons that only lower mass products (such as Ga^{66-68}) would form. On the other hand not enough nucleons would be emitted to form Ni^{57} , Co^{55} , or Fe^{52} nor do these products seem to be formed by other mechanisms. Enough energy should be available to form Zn^{62} at 138-MeV O^{16} bombardment energy by ($2\alpha p4n$) emission from a CFCN. (Ga^{68} formed by $3p4n$ emission has an energy requirement in

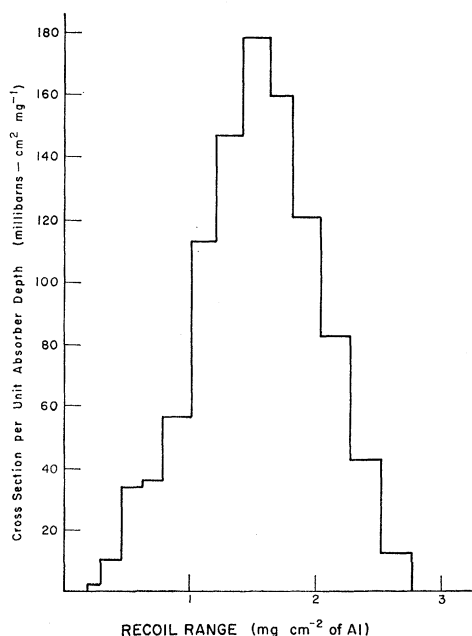


FIG. 4. Example of simple range distribution. Cu^{64} from 129-MeV O^{16} on Co^{59} .

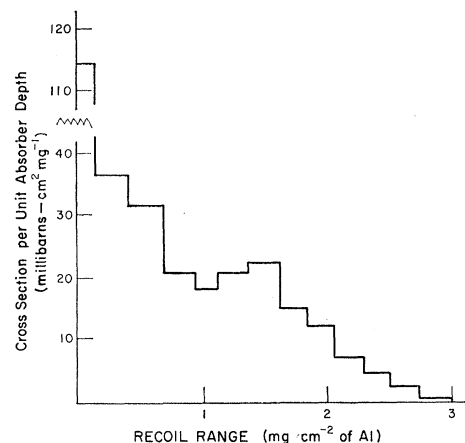


FIG. 5. Example of complex range distribution. Cu^{61} from 129-MeV O^{16} on Co^{59} .

the same range and is formed with a 23-mb cross section.) However, the low relative probability of $2\alpha p4n$ emission and the scarcity of levels in the even-even Zn^{62} make its low cross section appear reasonable.

Data on Momentum Transfer

Typical examples of plots of the recoil range of products vs the range are given in Figs. 4 and 5. These plots fall into two distinct categories: one, which we shall call "simple" (Fig. 4) is typified by a single peak of approximately Gaussian shape; the other, called "complex", shows in addition to this peak an appreciable yield of products with much shorter ranges (Fig. 5). All products, except Cu^{61} , show simple distributions. Even Cu^{61} has the complex pattern only when formed from O^{16} on Co^{59} ; Ne^{20} on Mn^{55} gives a simple distribution for Cu^{61} . The peak of the simple distribution is where it would be expected assuming full transfer of momentum of the heavy ion to a CFCN and using previously available (but admittedly rather crude) range-energy relations.¹⁸ The conclusion immediately presents itself that most products—those having simple range distributions—result nearly exclusively from a complete fusion compound nucleus. Only products near the mass of the target—in this case Cu^{61} from Co^{59} —may also be formed from some other type of interaction. This is presumably a grazing collision, in which only a part of the projectile nucleons add to the target, giving a low-momentum, low-excitation "partial fusion" compound nucleus.

This preliminary conclusion will now be examined in some detail.

The mean ranges and the standard deviations for the straggling in the simple distributions are given in Table III.¹⁹

¹⁸ For example, N. Porile and N. S. Sugarman, Phys. Rev. **107**, 1414 (1957).

¹⁹ These quantities were computed using number-distance curves—integrated forms of the differential yield range curves in

Range-Energy Relation for Recoil Products

No accurate range-energy relation for species in the mass and energy of the products obtained has been available. Attempts have been made to fit experimental mean recoil ranges into power series with dependence on recoil mass and initial recoil energy. This approach is successful only at very low recoil energies where nuclear elastic collision is the sole stopping mechanism. At higher energies, as are encountered in this work, electronic interactions become an important mode of energy loss and the recoil range becomes a very complex function of stopping material, and mass, charge, and initial recoil energy.

We have had good success in empirical fitting of a theoretically derived range-energy relation,

$$\bar{R} = k + R^0/a,$$

where \bar{R} is the range, R^0 is calculated using Bohr's theory²⁰ for stopping by both electronic and nuclear elastic interactions and k and a are empirically fitted constants.

R^0 is calculated using the initial recoil velocity or energy E_r . If the CFCN mechanism is assumed, this will be, for the simple range distributions,

$$E_r = E_b M_i M_p / (75)^2,$$

where E_b is the energy of the bombarding projectile and M_i its mass number. M_p and 75 are the mass numbers of the final product and the compound nucleus, respectively.

The fit obtained is shown in Fig. 6. The evident internal consistency demonstrates that the incident heavy ion must impart a constant fraction, close to unity, of its momentum to the compound system, yielding products with simple range distributions. A mechanistic basis for the nondependence of this fraction on both product and energy is most difficult to con-

Fig. 4. [M. S. Livingston and H. A. Bethe, *Revs. Modern Phys.* **9**, 245 (1937).] The median ranges obtained by this method are in all cases equal within experimental error to the mean ranges.

²⁰ N. Bohr, *Phys. Rev.* **58**, 654 (1940); **59**, 270 (1941); *Kgl. Danske Videnskab. Selskab, Mat-fys. Medd.* **24**, 19 (1952).

$$R^0 = - \int_0^{V_i} \frac{dV}{(dV/dR)},$$

$$\frac{dV}{dR} = \frac{-4\pi N e^4}{M m \mu U^3} \left\{ O_n \left(\frac{V}{U} \right)^2 + \frac{Z^2 z^2 (m+M) \mu}{m M} \left(\frac{U}{V} \right)^3 \right.$$

$$\left. \times \ln \left[\frac{M m (Z+z)}{\mu (M+m) Z^2 z^2} \left(\frac{V}{U} \right)^2 \right] \right\},$$

where the recoil nucleus has charge number Z , mass M , and velocity V , in a stopping medium of charge number z and mass m . μ is the electronic mass and $U = 2.2 \times 10^{-8}$ cm/sec. V_i is the initial velocity. There is a divergence at $V \leq U$ which must be avoided by suppressing the first term below U . Since this will result in ignoring electronic interactions at low velocities, an additive term must be employed in the final expression. This divergence is caused by the approximation used for the effective charge of the recoil, and the additive term k is to be expected to depend only upon Z , which is its observed behavior. The choice of a value for U is fairly arbitrary within a factor of two of the value used. Since the final expression contains U raised to the fourth power, the parameter a is employed. a , as is expected, is independent of both Z and M .

TABLE III Measured properties of range distributions.^a

Product	Bombardment energy E_b (MeV)	Median range R_m (mg/cm ²)	Standard deviation S (mg/cm ²)	Corrected standard deviation $S(\text{corr})$ (mg/cm ²)
Ga ⁶⁶	82	1.27	0.43	0.43
Ga ⁶⁶	108	1.52	0.38	0.37
Ga ⁶⁶	138	1.66	0.40	0.39
Ga ⁶⁶	159	1.79	0.45	0.44
Ga ⁶⁷	82	1.29	0.39	0.38
Ga ⁶⁷	108	1.55	0.38	0.37
Ga ⁶⁷	138	1.63	0.36	0.35
Ga ⁶⁷	159	1.85	0.32	0.31
Ga ⁶⁸	82	1.35	0.35	0.34
Ga ⁶⁸	108	1.54	0.40	0.39
Ga ⁶⁸	138	1.72	0.30	0.30
Ga ⁶⁸	159	1.89	0.31	0.30
Cu ⁶¹	70	complex		
Cu ⁶¹	97	complex		
Cu ⁶¹	129	complex		
Cu ⁶¹	135	complex		
Cu ⁶¹	158	complex		
Cu ⁶⁴	70	1.1	0.4	0.4
Cu ⁶⁴	97	1.28	0.55	0.54
Cu ⁶⁴	129	1.44	0.47	0.46
Cu ⁶⁴	135	1.55	0.50	0.49
Cu ⁶⁴	158	1.60	0.5	0.5
Cu ⁶¹ b	134 ^b	1.53	0.60	0.52
Cu ⁶⁴ b	134 ^b	1.55	0.5	0.5

^a Note. Except where low activity production decreased accuracy, absolute uncertainties are as follows: 2% for the bombardment energies E_b ; 5% for the median range \bar{R} ; and about 10% for the standard deviation S . Errors on relative values are smaller.

^b Denotes neon-20 bombardment.

ceive—unless it is exactly unity, corresponding to complete fusion. The range data thus provide strong evidence for the CFCN mechanism. They may also be used as the basis of a range-energy relation (see Fig. 6 and reference 20) much more accurate than heretofore available for these masses and energies.

Straggling of the Recoil Products

If the products having simple range distributions derive from evaporation from a CFCN, their straggling

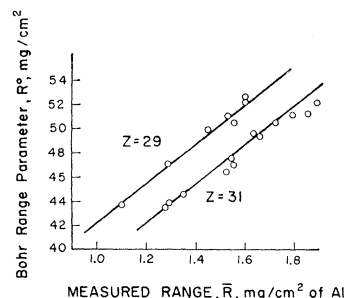


FIG. 6. Plot of experimental ranges vs range parameter calculated from Bohr theory. (See text and reference 20.) Z values given refer to the atomic number of the measured product. In the case of Ga⁶⁶, Ga⁶⁷, and Cu⁶¹ the points, therefore, include some contribution from recoils of the same mass but higher Z , which decay to the observed species. The uncertainty from this and other factors is estimated as $\pm 5\%$.

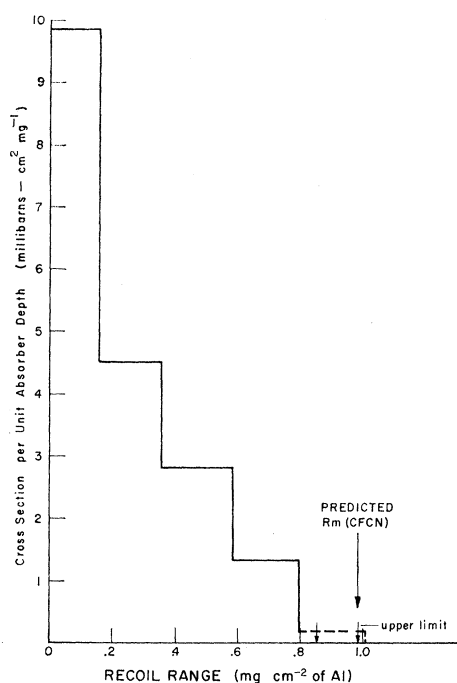


FIG. 7. Range distribution of Cu^{61} from 70-MeV O^{16} on Co^{59} .

will yield the following standard deviation of the range.

$$S = (S_r^2 + S_n^2 + S_e^2 + S_t^2 + S_f^2)^{1/2}.$$

The individual contributions to this standard deviation are due to: (S_r) variation of the recoil momentum due to evaporation from the CFCN; (S_n) stopping by nuclear elastic collisions; (S_e) stopping by electronic interactions; (S_t) the finite thickness of the target; (S_f) nonuniformity in stopping foils.

S_e is readily calculated to be negligible.²⁰ Consideration of the microscopic uniformity of the stopping foils shows that S_f is likely to be also negligible and this is confirmed by the nondependence of S on recoil range with any given CFCN product. Following the procedure of Winsberg and Alexander,¹¹ we have approximated S_t for a target film uniform to $\pm 20\%$, by

$$S_t = 0.35 \text{ (target thickness).}$$

The last column in Table III is corrected for this term,

$$S(\text{corr}) = (S^2 - S_t^2)^{1/2} = (S_n^2 + S_r^2)^{1/2}.$$

The magnitudes of S_n and S_r may be estimated and are sufficient to account for $S(\text{corr})$. However they cannot be calculated with sufficient precision to make a quantitative comparison meaningful. Qualitatively, $S(\text{corr})$ behaves in precisely the manner expected if it is actually the sum of these two components. S_r should increase with decreasing mass number of the product, since at a given excitation energy of the compound nucleus more alpha particles must be emitted to reach a lower residual mass. The effect of the higher recoil of these alpha particles is reflected in the tendency of

$S(\text{corr})$ to increase at constant bombarding energy in the order Ga^{68} , Ga^{67} , Ga^{66} , Cu^{64} , Cu^{61} . S_n is given by²¹

$$S_n = [R/(M+m)] \left(\frac{2}{3} M m \right)^{1/2},$$

where m and M are the masses of the recoil particle and stopping atoms respectively, and R is the range over which stopping is primarily by nuclear elastic collision. R is not well known but is exceeded by the actual range in all experiments giving simple distributions. Hence, S_n should be independent of bombarding energy. For a given recoiling species this is the observed behavior of S .

We conclude that the straggling behavior is fully consistent with a CFCN mechanism for products showing simple range distributions. If as much as 10% of the total activity in a simple distribution had recoiled with a momentum less than 0.7 of that corresponding to CFCN this would have been detectable. By contrast a "buckshot" type mechanism would be expected to give straggling extending to zero range and a dependence on bombarding energy.

Analysis of the Complex Range Distributions

From the above it may be seen that \bar{R} and S for a simple range distribution are predictable [by interpolation using empirically fitted values of R^0 (see Fig. 6) and $S(\text{corr})$], to an accuracy as good as that with which they may be measured experimentally. The range distributions of Cu^{61} from O^{16} bombardment of Co^{59} appear composed of a simple range distribution plus a shorter-range distribution. If this is indeed the case we may subtract the simple distribution, which has a calculable \bar{R} and S , to obtain the short-range component. This is done by means of a graphical technique. This method requires the following assumptions: (1) no appreciable amount of the shorter-range distribution extends to ranges larger than the mean range of the simple distribution, and (2) the simple distribution contained in the complex distribution is, as are all other observed simple distributions, symmetric about its mean.

The validity of these assumptions may be demonstrated: at a bombardment energy of 70 MeV, only 57 MeV of excitation energy would be available in the CFCN to evaporate 14 nucleons to form Cu^{61} . It is, therefore, improbable that at this energy appreciable Cu^{61} can be produced by evaporation from a Br^{76} CFCN. Thus, no simple CFCN component should be present in the Cu^{61} range distribution, and the short-range component should be obtainable without subtraction. This is actually the case as is shown in the range distribution in Fig. 7. No Cu^{61} recoils to ranges near the predicted mean range of CFCN Cu^{61} and the distribution is similar to those obtained by the subtraction procedure at higher energies.

The separation of the two contributions to the yield of Cu^{61} from $\text{O}^{16} + \text{Co}^{59}$ reactions permits the construc-

²¹ R. B. Leachman and H. Atterling, *Arkiv Fysik*, **13**, 101 (1958).

tion of separate excitation functions for long-ranged (CFCN) Cu^{61} and short-ranged (PFCN) Cu^{61} . These are shown in Fig. 8. An experimental point representing Cu^{61} produced by the $\text{Ne}^{20} + \text{Mn}^{55}$ reaction, which yielded a simple range distribution, is included. It is in good agreement with the CFCN component of the excitation function for Cu^{61} from the $\text{O}^{16} + \text{Co}^{59}$ reaction.

The range distributions of the short-range Cu^{61} show considerable higher range "tails" (Fig. 7). Straggling of monoenergetic recoils can account for only a very small fraction of this, indicating that a broad distribution of momenta is imparted to the partial fusion compound nucleus (PFCN) Cu^{61} . The median ranges of the PFCN Cu^{61} at each bombarding energy are shown in Table IV. To estimate median recoil energies corresponding to these median ranges, the range-energy relation

$$E_r(\text{MeV}) = \bar{R}(\text{mg/cm}^2)/0.11$$

was used. This relation was obtained from Harvey²², and checked by measuring the median recoil ranges of Cu^{61} produced by 30 to 40 MeV bombardments of Co^{59} with alpha particles. The recoil energies corresponding to the component of recoil velocity in the beam direction thus calculated are given in the third column of Table IV. In the last column is given Ω , the fraction of the heavy ion's momentum transferred to the Cu^{61} . The decrease of this quantity with energy is interesting.

DISCUSSION

No attempt has been made in this study to account for all the radioactive products of heavy-ion bombardment of a medium mass nucleus. However, a total of 16 possible representative products were sought in the system O^{16} on Co^{59} of which 7 were found. Furthermore, excitation functions of 15 products from related systems were measured. These results cover a sufficiently wide range of product mass number, relative to target and compound system, and neutron deficiency to make some degree of generalization possible.

The bulk of the products, with mass numbers between that of the target and the compound system, appear to result from decay of a compound nucleus formed by complete fusion of projectile and target. As shown in the section on results, both the nature and the

TABLE IV. Fractional momentum transfer to short-range (PFCN) Cu^{61} .^a

E_b (MeV)	\bar{R} (mg/cm ²)	$E(\text{recoil})$ (MeV)	Ω
70	0.17	1.5	0.29
97	0.12	1.1	0.2
129	0.13	1.2	0.19
135	0.09	0.8	0.16
158	0.06	0.5	0.11

^a The uncertainty of \bar{R} and $E(\text{recoil})$ is $\pm 20\%$, due largely to uncertainty in the target thicknesses. The uncertainty of Ω is about 20%.

²² B. G. Harvey, University of California Radiation Laboratory Report UCRL-9151, 1960 (unpublished).

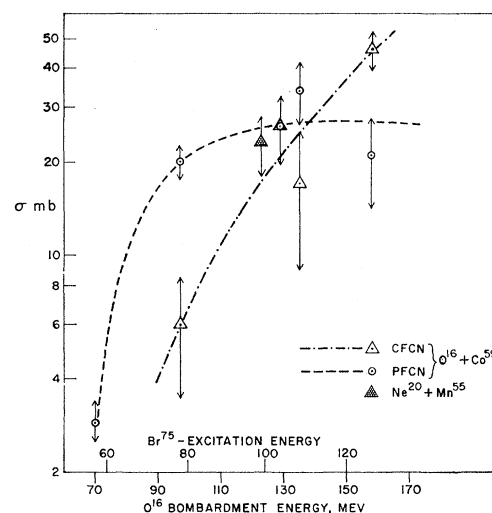


FIG. 8. Excitation functions for production of Cu^{61} from O^{16} bombardment of Co^{59} by complete fusion compound nucleus and grazing mechanisms.

excitation functions of these products strongly indicate that they are produced exclusively by decay of a compound nucleus formed by complete fusion of target and projectile (CFCN). The data on recoil ranges provide definite confirmation of this. In the system $\text{O}^{16} + \text{Co}^{59}$ all major products except Cu^{61} carry with them a momentum corresponding to the complete fusion of projectile and target. A portion of the Cu^{61} seems to be formed by a CFCN mechanism and the other, carrying less momentum, by a direct interaction (discussed below). The Cu^{61} is only two protons removed from the Co^{59} target. In the formally similar compound system Ne^{20} on Mn^{55} , Cu^{61} is 6 nucleons removed from the target. Here the low-momentum, direct-interaction portion of its yield is absent, although the cross section for formation through the full-momentum transfer CFCN is the same as with Co^{59} .

Evidently the bulk of heavy products are formed through a compound nucleus incorporating all of the projectile. Only products near to the target are formed by another mechanism. This is not in accordance with the buckshot mechanism as originally formulated.⁵ However, the grazing mechanism⁹ does correspond to these observations in that it predicts the preferential transfer of only a few nucleons in any collision not leading to complete fusion.

The momentum carried by Cu^{61} from grazing processes can have a wide range, from zero up (see Fig. 7). This is presumably due, at least in part, to fluctuations in the internal momentum vectors of the two protons being transferred. The variation with energy of the average fraction of the momentum transferred from the O^{16} projectile to the Cu^{61} product is more interesting. (See Table IV.) At the highest energy this average momentum transfer corresponds, within the limit of error, to the fraction of the momentum of the O^{16}

carried by two of its nucleons. This suggests that: (1) the Cu^{61} is largely formed by the transfer of two protons to give a compound nucleus which does not have sufficient excitation energy to evaporate a particle, and (2) the Cu^{61} does not receive any appreciable forward momentum from the projectile residue (formally N^{14}). This is consistent with the grazing model since the residue will retain nearly all of its velocity⁹ and will be deflected from its forward direction only slightly at this energy. At lower energies the projectile residue will be deflected sideways or even backwards by the Coulomb field of the target. This accounts for the increasing fraction of projectile momentum transferred as the bombarding energy is decreased. (See Table IV.)

Using data in Table IV and earlier findings that the projectile residue retains, on the average, nearly all of its velocity, it can be calculated that at 70-MeV bombarding energy it will be deflected 30° from the beam axis. This angle is significantly less than the Rutherford scattering cutoff angle, for purely Coulombic scattering in a close collision. The residue thus appears to have been deflected forward of the Rutherford cutoff angle by a nuclear interaction. The nuclear binding during collision necessary to account for such a deflection appears to be of the same order of magnitude—10 MeV—as that found in a previous study of the grazing mechanism.⁹

Relatively crude calculations on the grazing mechanism indicate that it can occur in high yield.⁹ In the case of 160-MeV O^{16} reacting with a target of mass number 60 the grazing reaction cross section should be of the order of half of the geometric cross section. There is evidence for the production of projectile residues from grazing in about this yield. Radioactive residues resulting from nucleon transfer (e.g., C^{11} , N^{13} , O^{15} , and F^{18} from O^{16}) have a cross section of the order of 50 mb and imply the existence of a larger yield of stable or short-lived radioactive products of a similar nature. A major portion of grazing reactions appear to result in projectile disintegration. Alpha particles having the angular distribution and kinetic energy corresponding to such a process have been observed in yields of the order of 0.5 b from 160-MeV O^{16} bombardment of heavy targets.¹⁰

In the systems investigated here, cross sections of the order of hundreds of millibarns would, therefore, be expected for the heavy or target residues of grazing reactions. Unfortunately the targets chosen, while serving to show that the main bulk of products is formed by evaporation from a fused target-projectile compound nucleus, do not suffice to establish the pattern of grazing products in the vicinity of the target. Of these expected products only Cu^{61} , containing two protons more than the target, has decay properties that make its investigation convenient. Cu^{61} has a cross section for formation by grazing reaction of ~ 25 mb (see Fig. 8). Our results show no evidence that transfer in grazing reactions involves more than a few nucleons, but even if the possible products of 1- or 2-nucleon transfer have

TABLE V. Yields of P^{32} and Na^{24} .

System $\text{C}^{12} + \text{Co}^{59}$			System $\text{O}^{16} + \text{Mn}^{55}$		
Energy range (MeV)	Average cross section ^a		Energy range (MeV)	Average cross section ^a	
	Na^{24} (mb)	P^{32} (mb)		Na^{24} (mb)	P^{32} (mb)
0-77	0.05 ^b	0.04 ^b	0-108	0.05 ^b	0.02 ^b
77-95	0.13	0.16	108-140	1.1	0.54
95-107	0.36	0.29	140-168	3.4	0.85
107-126	0.40	0.54			

^a Cross section averaged over range in Co^{59} traversed by projectile between stated energies.

^b Possibly due to impurities. Regard as upper limit only.

cross sections of the magnitude of that of Cu^{61} , total grazing cross sections will be of the order of hundreds of millibarns.

In summary then, while no evidence whatever for a generalized "buckshot" phenomenon has been observed, the grazing mechanism will, as was expected, yield transfer products a few nucleons removed from the target. Note, however, that the mechanism originally proposed by Chackett *et al.*⁵ for the "buckshot" mechanism—Coulomb breakup of the projectile in the field of the target, prior to impact—would be most effective at high atomic numbers. While definitive results in the heavy-target region are rather meager, there is some evidence for transfer events involving more than a few nucleons. Alexander and Winsberg¹¹ have bombarded several elements from cerium to bismuth with heavy ions. Most of their results are consistent with a CFCN mechanism, although with Bi and Pb a considerable cross section for partial momentum transfer is observed. In particular a net two-proton transfer PFCN event yielding At^{211} from Bi^{209} is unambiguously identified. The situation regarding transfer of a larger number of nucleons is not as clear. Partial momentum transfer in the processes $\text{Au}^{197}(\text{O}^{16}, 2p\pi n)$ and $3p\pi n\text{At}$, Po is reported. However, the maximum deficiency in momentum—13% compared to the CFCN—is not large compared to the inherent uncertainties of the method.

More recently momentum transfer in heavy-ion induced heavy element fission has been examined.²³ In addition to the dominant CFCN mechanism, the momentum distribution indicates transfer of an alpha particle leading to U^{238} fission. With gold and bismuth, where transfer of one alpha will not provide enough excitation energy to cause fission, evidence for transfer of the equivalent of about eight nucleons was found. This observation suggests that though the "buckshot" phenomenon is apparently not important with lighter elements, the increased possibility of Coulomb disintegration may make it more significant at high atomic numbers.

²³ T. Sikkeland, E. Haines, and V. Viola, University of California Radiation Laboratory Report UCRL-97151, 1961 (unpublished).

Na²⁴ and P³² Formation

The Na²⁴ and P³² formed do not fall within the scope of the above discussion. Cross sections for their production are given in Table V. Because of the subtraction procedures involved in thick target bombardments, these cross sections are accurate to only $\pm 50\%$.

Energy requirements for the formation of these products are such that it is highly unlikely that they were formed by nucleon or alpha-particle emission. It therefore appears that heavy-ion-induced fission or fragmentation of medium-mass targets takes place with an appreciable cross section. Little more can be said on

the basis of present data, apart from the customary comment that it would be interesting to investigate this phenomenon further.

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Excitation Functions of (p, xp) Reactions*

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Radiochemically measured excitation functions are reported for several (p, xp) reactions, where $x \geq 2$, for target nuclei encompassing a wide range of mass numbers and for incident energies of 100 to 440 MeV. The $(p, 2p)$ reactions were studied using Si³⁰, Zn⁶⁸, and W¹⁸⁶ as target nuclei; the $(p, 3p)$ reactions were studied using Si³⁰, P³¹, V⁵¹, As⁷⁵, W¹⁸⁶, and Re¹⁸⁷; the $(p, 4p)$ reactions were studied using P³¹, S³², As⁷⁵, and Re¹⁸⁷; and a $(p, 5p)$ excitation function was measured using S³² as the target. The data do not seem to exclude important contributions by either the knock-on or evaporation mechanisms in all mass regions studied, although, according to qualitative arguments it seems plausible that knock-on processes predominate in the high-mass region.

INTRODUCTION

NUCLEAR reactions by which both the mass and the charge of the target nucleus are reduced by $x-1$ units, where x is an integer, can be referred to as (p, xp) reactions. Only a few cross sections for $(p, 2p)$ reactions induced by 100 to 400 MeV protons have been reported,¹⁻⁵ and reported values of $(p, 3p)$ cross sections^{6,7} are even scarcer. Only one reported value of a $(p, 4p)$ cross section⁷ and none at all for $x \geq 5$ have been found.

A systematic study of (p, xp) reactions seems to be a logical step in trying to understand what happens

when a high-energy particle interacts with a complex nucleus. These reactions occupy the interval between simple reactions, which are beginning to be understood, at least qualitatively, and the spallation reactions in which the interactions are so complex as to defy all serious attempts at analysis or information extraction. In $(p, 2p)$ reactions only one of the target nucleons participates dominantly. As the value of x increases, the complexity of the problem increases in sufficiently small steps so that an understanding of the simpler reactions may help one to understand the more complex reactions if one studies such a sequence carefully. For those reactions in which only protons are emitted, there are fewer paths by which the product can be formed than in reactions where both proton and neutron emission must occur.

The general mechanism within which high-energy reactions have usually been interpreted was proposed by Heisenberg⁸ and by Serber,⁹ and is referred to in succeeding sections as the Serber process. According to this model, the high-energy reaction is roughly divided into two phases: (i) the knock-on cascade, in which a number of particles are ejected from the nucleus by direct interaction, leaving a residual

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