

Nuclear Reactions of High-Energy C^{12} , N^{14} , and O^{16} with Carbon†

MURRY A. TAMERS,* *Department of Chemistry, University of Texas, Austin, Texas*

AND

RICHARD WOLFGANG, *Department of Chemistry, Yale University, New Haven, Connecticut*

(Received August 25, 1959)

The reactions of 160-Mev O^{16} , 140-Mev N^{14} , and 120-Mev C^{12} with C^{12} to yield radioactive products have been studied and the results compared with those for proton induced reactions giving formally similar compound systems. Excitation functions for products formed at lower energies are similar in the corresponding proton and heavy ion cases, indicating that their formation involved similar low energy, low spin compound nuclei. However, no "tail" due to knock-on cascade processes was observed with heavy ions. Yields for higher energy products, chiefly F^{18} , were much higher in the heavy ion bombardments. This may be attributed partly to higher average energy deposition with heavy ions, due to the absence of knock-on processes, and partly to enhanced alpha emission from the distorted high-spin compound nuclei formed by heavy ions. The contribution of various stripping processes to these yields is discussed.

AN experiment that has often been proposed in the heavy-ion reaction field is to examine and compare the properties of compound systems of similar nucleon and energy content which have been produced using, on the one hand heavy ions and on the other, protons as projectiles. Such an experiment would extend that of Ghoshal¹ who showed that similar proton and alpha produced compound systems decayed similarly, thus indicating the validity of the Bohr compound nucleus assumption that only the excitation energy of the system determined its modes of decay. (The term "compound system" here denotes any kind of interaction between projectile and target; whereas "compound nucleus" carries the Bohr connotation of a long lived quasi-equilibrium intermediate complex.)

Cohen, Reynolds, and Zucker² compared proton reactions on Mg^{25} with the formally equivalent reactions with carbon of 29-Mev N^{14} . This paper reports on a study of the reactions with carbon targets of C^{12} , N^{14} , and O^{16} with higher energies, up to 120–160 Mev. The system was chosen because of its experimental simplicity and because data on proton-induced reactions to form the same compound systems, Mg^{24*} , Al^{26*} , and Si^{28*} , was available.^{2–4} In comparing systems of such relatively low mass as produced by protons and heavy ions at energies up to 80 Mev in the center-of-mass system it should be expected that the differences between them will be emphasized, rather than their similarities as would be the case with heavier compound systems.⁵ With light systems direct interactions such as intra-

nuclear knock-on processes for protons⁶ and stripping reactions for heavy ions^{7,8} may be expected to be prominent. Perhaps even more important are the very high rotational energies that are involved in the impact of high-energy heavy ions on a light target. At the bombarding energies used in this work, formation of a conventional spherical compound nucleus as a result of a peripheral collision actually becomes impossible simply because its rotational energy would exceed the available energy in the system. Thus the effect of the rotational energy as well as the excitation energy of the

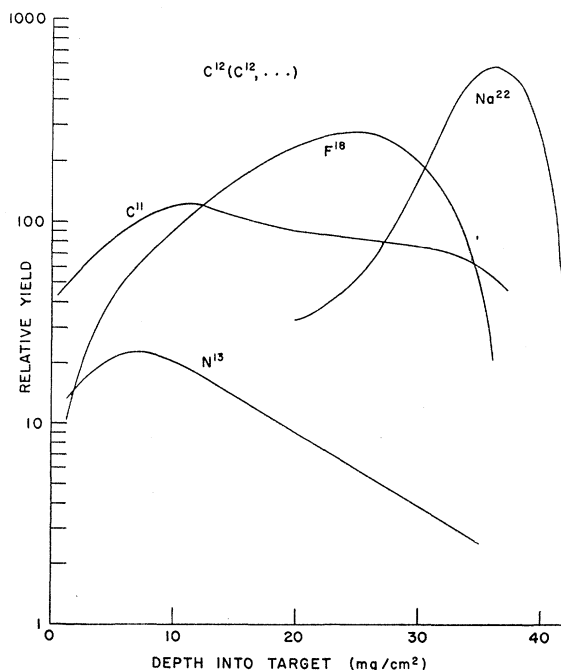


FIG. 1. Distribution of products in polyethylene foil stack irradiated with 120-Mev C^{12} .

† Contribution No. 1563 from Sterling Chemistry Laboratory, Yale University, New Haven, Connecticut. This work was supported by the U. S. Atomic Energy Commission.

* Assistance by the Robert A. Welch Foundation, Houston, Texas is acknowledged.

¹ S. N. Ghoshal, *Phys. Rev.* **80**, 939 (1950).

² Cohen, Reynolds, and Zucker, *Phys. Rev.* **96**, 1617 (1954).

³ J. W. Meadows and R. B. Holt, *Phys. Rev.* **83**, 47, 1257 (1951).

⁴ N. M. Hintz and N. R. Ramsey, *Phys. Rev.* **88**, 19 (1952).

⁵ Sikkeland, Thompson, and Ghiorso, *Phys. Rev.* **112**, 543 (1958).

⁶ N. Metropolis *et al.*, *Phys. Rev.* **110**, 185 (1958).

⁷ M. L. Halbert *et al.*, *Phys. Rev.* **106**, 251 (1957).

⁸ G. Breit and M. E. Ebel, *Phys. Rev.* **103**, 679 (1956).

compound nucleus must become significant in determining its decay modes.

EXPERIMENTAL

Stacks of approximately 15 polyethylene foils each about 3 mg/cm^2 thick were used as targets. [In an attempt to eliminate (p, α) reactions involving hydrogen in the target, carbon foils, prepared by charring paper, were sometimes used. However, this led to difficulties caused by absorbed air on the highly activated carbon and for this reason results from these runs are not included in this paper.]

These targets were irradiated by C^{12} , N^{14} , and O^{16} beams from the Yale Heavy Ion Accelerator at an

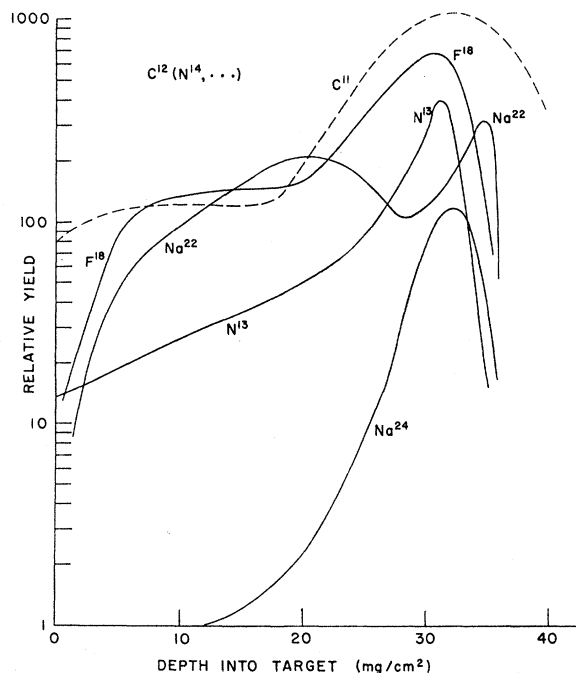


FIG. 2. Distribution of products in polyethylene foil stack irradiated with 140-MeV N^{14} .

average beam intensity of about 3×10^{-9} amp. The beam energy was 10.0 ± 0.2 Mev per nucleon, "satellite energy" components being eliminated by a simple deflecting magnet. Total beam current was monitored by a Faraday cup.

After counting under end window proportional flow counters, decay curves for the individual foils were resolved into 10.0 minute, 20.5 minute, 112 minute, 15 hour, and longer lived components. With corrections for counting efficiencies, this served to establish relative yields of Na^{24} , Na^{22} , and F^{18} with a relative accuracy of about 10% and an estimated absolute accuracy of 25%. Due to the similarity of half-lives, yields of C^{11} and N^{13} were subject to somewhat larger errors. Indeed, the N^{13}

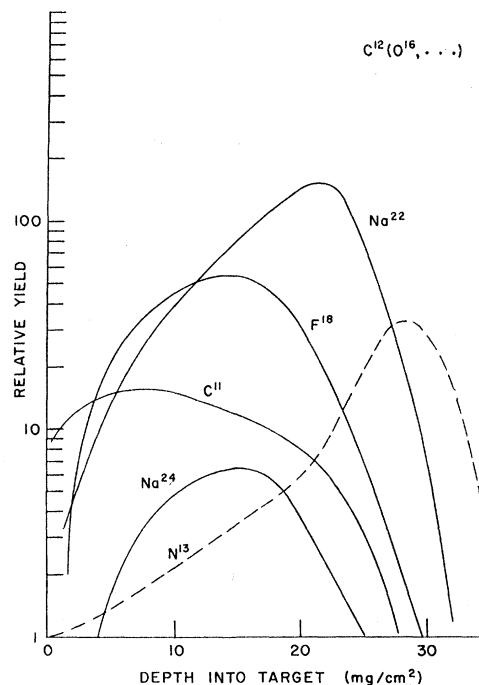


FIG. 3. Distribution of products in polyethylene foil stack irradiated with 160-MeV O^{16} .

data are only useful in indicating general magnitudes and trends. This is also the case for Be^7 .

Na^{22} was chemically separated and identified. After subtraction of Na^{22} activity another long-lived component remained. By comparing β and γ disintegration rates this activity was identified as due to Be^7 . Although activity levels were very low the expected half-life of about 50 days was observed. The identification was further confirmed by chemical means (see Appendix).

RESULTS AND DISCUSSION

"Inverse" (p, α) Reactions

Figures 1-3 give the distribution of activity in the stacked foils due to $C^{12}(C^{12}, \dots)$, $N^{14}(C^{12}, \dots)$ and $O^{16}(C^{12}, \dots)$ reactions. (p, α) reactions involving the hydrogen atoms in the polyethylene also occur. The laboratory system threshold for the $H(N^{14}, \alpha)C^{11}$ reaction is 44 Mev and that for the $H(O^{16}, \alpha)N^{13}$ reaction is 88 Mev. The products of these reactions have approximately the same velocity as the bombarding heavy ion and are therefore found toward the back of the foil stacks, as the C^{11} peak in Fig. 2 and the N^{13} peak in Fig. 3.

Reactions Giving Heavier Products

Table I gives the average thick target cross sections for the products of heavy ion reactions and the corresponding proton induced reactions, all relative to the yield of Na^{24} or Na^{22} . The proton yields are calculated

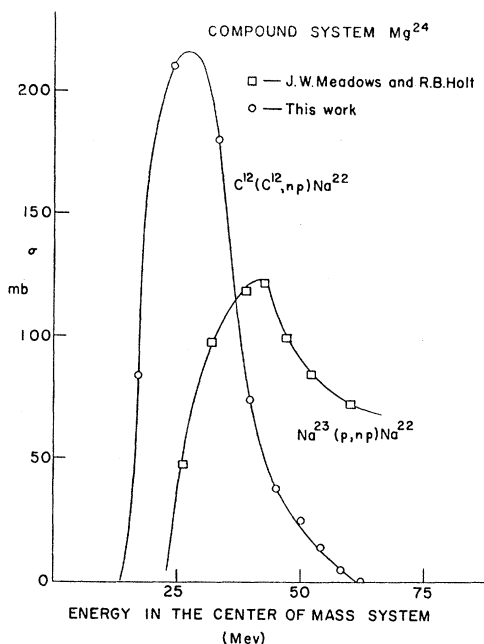


FIG. 4. Excitation functions for Na^{22} produced from the Mg^{24} compound system. Heavy ion curve constructed on the assumption that a compound nucleus is formed.

from the data of Meadows and Holt³ and Hintz and Ramsay.⁴ The lack of similarity between the proton and heavy ion systems is immediately apparent. Not only do the presumptive single nucleon stripping products C^{11} and N^{13} appear in the heavy ion case but furthermore the yields of F^{18} are an order of magnitude larger than with the proton reactions.

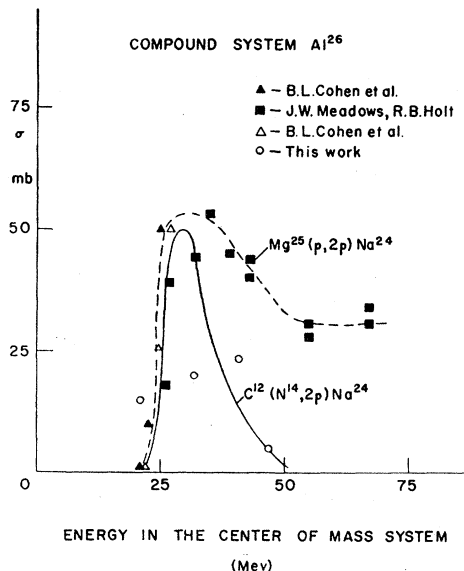


FIG. 5. Excitation functions for Na^{22} produced from the Al^{26} compound system. Heavy ion curve constructed on the assumption that a compound nucleus is formed.

Because of the recoil of the products, Figs. 1-3 cannot be directly translated into excitation functions by a change in abscissa. However, excitation functions can be calculated if the assumption is made that the projectile shares its momentum with all the nucleons of the compound system. Such an assumption will have little if any error, if the observed product has most of the mass of the compound system. The recoil due to evaporation from the compound system is easily shown to be much smaller than the recoil due to the initial interaction and may be neglected as a factor influencing either the magnitude or forward direction of the displacement of the product.

The recoil energy of the heavier products was then calculated as a function of bombardment energy and the corresponding displacements of the products obtained using range energy curves for Na^{24} and Na^{22} calculated using the results of Papineau.⁹ Correcting for such displacement, curves of yield vs position in target stack where reaction occurred were obtained. From these, excitation functions were calculated by the usual procedure.

TABLE I. Cross sections averaged over range projectile (millibarns).

Compound system	Mg^{24} (0-62 Mev)		Al^{26} (0-66 Mev)		Si^{28} (0-69 Mev)	
Reaction	$\text{Na}^{23} + p$	$\text{C}^{12} + \text{C}^{12}$	$\text{Mg}^{25} + p$	$\text{C}^{12} + \text{N}^{14}$	$\text{Al}^{27} + p$	$\text{C}^{12} + \text{O}^{16}$
Product						
Na^{24}	31	5.5	6.8	4.9
Na^{22}	76	19	43	39	26	109
F^{18}	20	21	8.6	70	3.7	45
N^{13}	...	1.7	...	20	...	~3
C^{11}	...	13	...	~19	...	17
Be^7	...	~6	...	~5	...	~3

The excitation functions thus obtained are shown in Figs. 4-7. They are quite approximate, especially in their energy ordinate, partly because of straggling in the recoils but mainly because of the considerable uncertainty in energy and the straggling of beams reduced from over 100 to about 30 Mev. (In Figs. 5 and 6 the more accurate low-energy proton and N^{14} data of Cohen *et al.*² are included.)

A considerable similarity is readily apparent between the excitation functions of the proton and heavy ion induced reactions of those products formed by the loss of two to four nucleons from the compound system. The main difference is the absence of a high-energy "tail" for the heavy ion products. In the case of proton induced reactions this "tail" is ascribed to product formation by a nucleon "knock-on" cascade.⁶ Such a cascade would not be expected to occur in heavy ion bombardment as is now confirmed by the absence of the high-energy tail.¹⁰

⁹ M. A. Papineau, Compt. rend. 242, 2933 (1956).

¹⁰ The fact that the assumption of momentum sharing in the compound system, which is used in deriving the excitation functions, will not hold for knockout reactions does not affect

Making allowance for the high-energy knockout products, the two sets of excitation functions are quite similar in shape. The double peak in the excitation functions of Na^{22} from the Al^{26} compound system is an example; the peak at lower energies presumably corresponding to α emission and that at higher energies to $2p2n$ emission. The displacement of the proton and heavy ion curves on the energy axis while perhaps real, is of the same magnitude as the limits of error. The magnitudes of the cross sections are also similar in the heavy ion and proton cases, except for the $C^{12}(C^{12},pn)-Na^{22}$ reaction which appears to be more probable over a narrow energy range than the corresponding $Na^{23}(p,pn)Na^{22}$ process.

It thus appears that in the formation of products only a few nucleons removed from the compound system, similar compound nuclei are formed by protons and heavy ions. A like conclusion was reached by Cohen, Reynolds, and Zucker² for the $N^{14}(C^{12},2p)Na^{24}$ reaction at low energies. This similarity is presumably a result of the relatively low excitation energy of these compound nuclei. Not only is the compound nucleus model expected to be a more accurate representation at such energies,¹¹ but more important low-energy compound nuclei formed by heavy ions will still have a relatively low average rotational energy.

Simple Stripping Reactions

The C^{11} and N^{13} observed in the heavy ion reactions are evidently simple stripping products^{7,8} produced by pick up or loss of a single nucleon by target or projectile [except in those cases, discussed previously, where they are formed by an inverse (p,α) reaction]. Where such products are made by nucleon addition or subtraction from the target they should have little recoil energy. Thus the distribution of C^{11} in the target bombarded with O^{16} (Fig. 3) shows a peak in the forward foils indicating a steadily increasing cross section with energy. The slight decline in the most forward foils which is observed in this and similar cases probably indicates that some recoil energy (about 5 Mev) is imparted to the stripped target nucleus, since there is no reason to expect a decline in cross section at the highest energies and since no such decline is observed for the similar $Ni^{58}(O^{16},\dots)Ni^{57}$ reaction.¹² Products formed by stripping from, or pickup by, the projectile recoil far into the foil, as is shown for the $C^{12}(N^{14},N^{13})$ reaction in Fig. 2. Where the product is formed from both target and projectile its distribution in the foil stack is approximately uniform (see C^{11} in Fig. 1).

this conclusion. A knockout product, having less momentum than assumed will recoil less and thus give the appearance of having been formed at still higher energy. But, as shown, the excitation functions drop to zero above the region for compound nucleus formation.

¹¹ Nakasami, Tanaka, and Kikuchi, Progr. Theoret. Phys. (Kyoto) 15, 574 (1956).

¹² R. Wolfgang (unpublished).

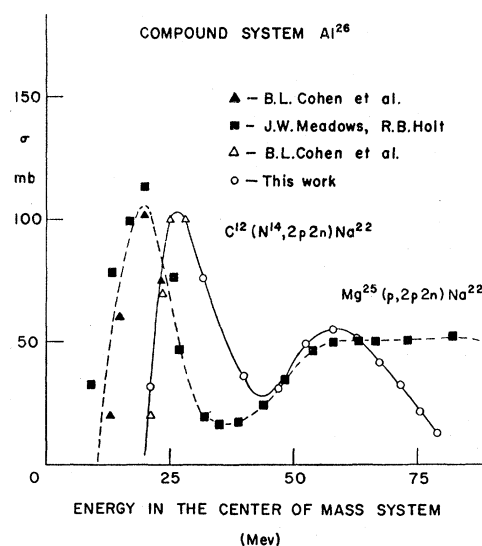


FIG. 6. Excitation functions for Na^{22} produced from the Al^{26*} compound system. Heavy ion curve constructed on the assumption that a compound nucleus is formed.

The yield of neutron stripping reactions to form C^{11} appears to be about 5–10 times that of proton stripping products. This is probably mainly due to the lower barrier for neutrons⁸ but the absence of excited states in N^{13} stable towards proton emission may also be a factor.

Intermediate Mass Products

In the heavy ion bombardments the largest yields are those represented by F^{18} in all three cases and Na^{22} in the O^{16} bombardment. These represent products heavier than the simple stripping products but lighter than

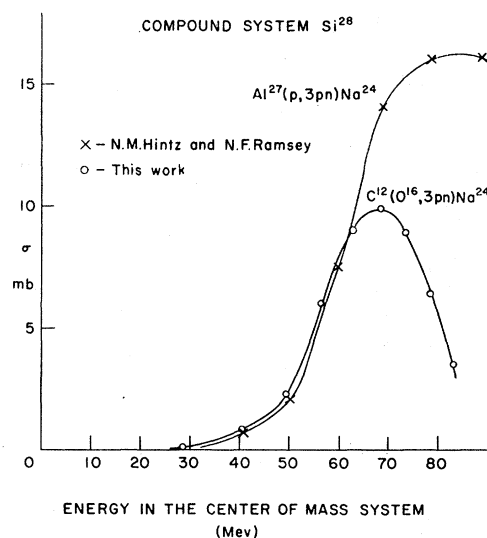


FIG. 7. Excitation functions for Na^{24} produced from the Si^{28*} compound system. Heavy ion curve constructed on the assumption that a compound nucleus is formed.

those discussed earlier which apparently originate in a relatively conventional compound nucleus. As shown in Table I these yields are an order of magnitude higher than those produced in the corresponding proton formed compound systems. Three possible explanations may be advanced for this striking difference in yields:

(1) High excitation energies of the compound system will be required to form F^{18} , etc., by evaporation. However, at higher energies a bombarding proton is less likely to deposit its full energy in a compound nucleus, because of the increasing importance of knock-on cascades in which high-energy particles are ejected from the system. Metropolis *et al.*⁶ have calculated that the average excitation energy (and its root mean square deviation) imparted to an Al^{27} nucleus bombarded with 82-Mev protons is 41 ± 22 Mev. As shown earlier such knock-on cascades appear unimportant in heavy ion reactions and would not interfere with conversion of the full center-of-mass bombarding energy into excitation energy of the compound nucleus.

(2) In a heavy ion bombardment only a part of the projectile may stick to the target (or vice versa). This has been termed the "buckshot" effect by Chackett *et al.*¹³ who have used it to account for yield distributions produced by O^{16} and N^{14} bombardments of aluminum. Thus in this work, F^{18} could be formed in the N^{14} bombardment of C^{12} by transfer of two alphas to the C^{12} followed by evaporation of two nucleons; and by simple alpha transfer to the N^{14} . Stripping of alpha particles from C^{12} to leave the remainder of the projectile as two alpha particles or Be^8 of essentially unchanged velocity was first observed by Miller¹⁴ in emulsions. Recent work of Knox *et al.* shows a similar phenomenon.¹⁵ The existence of a full range of such complex stripping reactions and evidence on the mechanism of their production, is indicated by the observations of Kaufmann and Wolfgang¹⁶ on C^{11} , N^{13} , O^{15} , and F^{18} produced with essentially the velocity of the original ion when O^{16} or N^{14} strike a target. At this time, however, there is still insufficient quantitative data on which to base an estimate on the relative importance of such complex stripping processes in determining the final yield distribution.

(3) Heavy ion collisions are characterized by very high l values and at energies somewhat above the Coulomb threshold will result in nuclei having very high rotational energy. Although a compound nucleus may be formed under these circumstances, its shape will be highly distorted, thus lowering its Coulomb barrier. This will lead to a preferential emission of protons and especially alpha particles. Independent evidence for such processes has been obtained by Knox

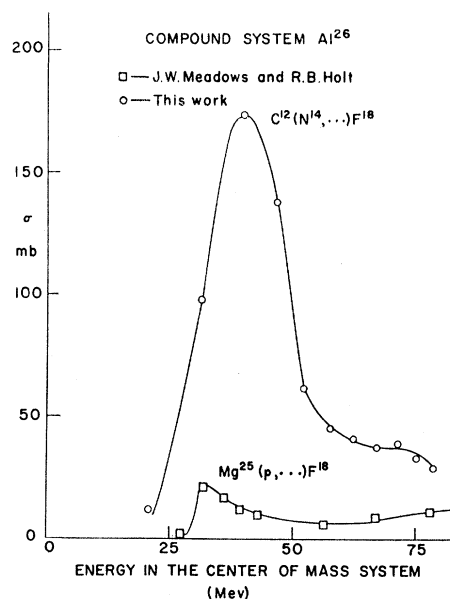


FIG. 8. Excitation functions for F^{18} produced from the Al^{26*} compound system. Heavy ion curve constructed on the assumption that a compound nucleus is formed.

*et al.*¹⁷ This would lead to the same kind of distribution of products as the "buckshot" theory.

De-excitation by fission will also be favored by a high spin in the compound nucleus. However, it would be difficult to explain on this basis, the rather different product distribution observed in the O^{16} and N^{14} bombardments of aluminum.¹³

Factor (1) should reduce F^{18} yields produced by proton reaction by perhaps two to fourfold compared to the heavy ion systems. The rest of the yield difference observed is presumably due to factors 2 and 3. However, the present work gives little indication as to the relative importance of these mechanisms. Thus the F^{18} peak appearing deep in the target of the $C^{12}(N^{14}, \dots)$ system (Fig. 2) shows about the right recoil for formation by capture of an alpha by the N^{14} , as suggested by Alkazov *et al.*¹⁸ On the other hand, if formation of a compound nucleus is assumed the excitation function which can be calculated (Fig. 8) shows a very plausible shape. The yield peak previously referred to is now interpreted as corresponding to a relatively low-energy compound nucleus process involving emission of two alpha particles. The second peak on this excitation curve can be ascribed to $\alpha 2p2n$ emission. The other F^{18} yields and that of Na^{22} from the $C^{12}(O^{16}, \dots)Na^{22}$ reaction give equally plausible excitation functions on the compound nucleus assumption.

It thus appears that yield data of this type can be accounted for by both complex stripping or "buckshot"

¹³ Chackett, Fremlin, and Walker, *Phil. Mag.* **45**, 173 (1954).

¹⁴ J. F. Miller, *Phys. Rev.* **83**, 1261 (1951).

¹⁵ Knox, Quinton, and Anderson, *Phys. Rev. Letters* **2**, 402 (1959).

¹⁶ R. Kaufmann and R. Wolfgang, *Phys. Rev. Letters* **3**, 232 (1959).

¹⁷ W. Knox (private communication).

¹⁸ Alkhozov, Gangrskii, and Lemberg, *J. Exptl. Theoret. Phys. U. S. S. R.* **33**, 1160 (1957) [translation: *Soviet Phys., JETP* **6**, 892 (1958)].

processes, or by the preferential alpha emission which must be expected from the high spin compound nuclei which are formed. It is hoped that experiments now in process will define the relative importance of these mechanisms.

CONCLUSION

(1) Reactions proceeding by intranuclear knock-on cascades such as are observed with protons are negligible in heavy-ion processes. On the other hand, nucleon stripping reactions are important with heavy ions.

(2) Products which may be formed by emission of only a few nucleons from the compound system of projectile and target, show similar excitation functions whether formed by protons or heavy ions. Thus in relatively low energy, low angular momentum collisions, protons and heavy ions appear to form similar compound nuclei.

(3) Yields of nuclides (typified by F^{18} in this work) formed by emission of a larger number of nucleons from the compound system are an order of magnitude higher for heavy ions than for protons. This must, in part, be the result of the absence of knockout cascades in heavy ion events which lead to higher average excitation energies of the compound system. Higher heavy ion yields of these products will also result from preferential alpha emission from high-spin compound nuclei and from any complex stripping "buckshot" reactions which may take place.

ACKNOWLEDGMENTS

The authors are grateful for stimulating discussions with Professors Knox and Beringer, particularly on the problem of the distortion and alpha emission from

high-spin nuclei. It is a pleasure to acknowledge the help of Dr. Malkin and Dr. Anderson and of the heavy ion operating crew. The assistance of Mr. Richard Kaufmann was of the greatest value.

APPENDIX. CHEMICAL VERIFICATION OF Na^{22} AND Be^7 IN POLYETHYLENE TARGETS

The foils were burned in an oxygen atmosphere in platinum crucibles and the long-lived activity washed out with 5 ml 0.001*M* $BeCl_2$ and 0.001*M* $NaCl$ solution and counted at 0.49 to 0.53 Mev with a well scintillation crystal in a low background shield. The activity found (0.5 to 8 cpm) corresponded to a 90% recovery.

The beryllium in the samples was precipitated with NH_4OH and the supernates containing the sodium were again counted in the well counter. The beryllium precipitates were dissolved with 5 ml HCl and this liquid was counted in the well counter.

The supernates containing the Na^{22} long-lived activity were tagged with small amounts of Na^{24} , counted in the well counter, and then put through a small ion exchange column and eluted with 1*M* $NaCl$ solution. The most active sample collected in each elution (containing about one-third the original Na^{24} activity) was counted in the well counter. After two weeks, the Na^{24} activity had died away and the eluates were again counted, this time for the long-lived activity from the original solution. The ratios of the Na^{24} activities put into the ion exchange column to those obtained in the particular fractions taken (corrected for decay) were the same as the fractions of the long-lived activity recovered, within the limits of the accuracy of the counting statistics. It had been shown previously, using various tracers, that this would only be the case for Na^{22} .