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Fission of Medium Weight Elements*

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Evidence is presented here which indicates that large fragments (much larger than alpha-particles) are emitted among the competitive products of transmutation throughout the entire range of atomic numbers of the elements. Threshold considerations for the observed nuclear reactions show that the reactions are observed with small cross sections well below the threshold for spallation reactions in which the maximum number of alpha-particles are considered as being emitted from the excited nucleus. The calculated thresholds include the mass difference between the reactants and the products and the excitation energy which the product particles or fragments must have in order to pass over the coulombic barrier. Preliminary experiments on the ranges of recoil fragments from copper irradiated with 340-Mev protons give additional evidence for the emission of heavy fragments. It is suggested that the term "fission" is proper for such reactions, throughout the entire range of atomic numbers, in which the nucleus is split essentially into pieces of comparable weight.

I. INTRODUCTION

THE fission reaction has been observed with high energy accelerator projectiles for elements as light as tantalum,[1] but has not been reported for medium weight elements. Evidence is presented here for occurrence of reactions which are probably most properly described by the term "fission" and which seem to occur, with very small yield, throughout the entire region where this type of reaction is only slightly exoergic or even endoergic with respect to mass balance.

In the course of the detailed investigation of the spallation of copper and the variation of the product yields with energy of the bombarding particle, the threshold for formation of radioactive Cl^{38} (39-minute half-life) from elemental copper was studied. The most energetically economical way in which Cl^{38} might be formed by spallation reactions is by emission from the bombarded copper nucleus of nucleons in groups, such as alpha-particles, instead of single nucleons. The energetic requirement for the spallation reaction $Cu^{63}(p,pn6\alpha)Cl^{38}$, in which the maximum number of alpha-particles is emitted, is roughly 110 Mev. This threshold includes (1) the mass difference between the reactants and the products and (2) the excitation energy which the alpha-particles must have in order to pass over the coulombic barrier.

The production of Cl^{38} was definitely observed at proton bombardment energies beginning at about 60 to 70 Mev with a cross section of some 10^{-32} cm^2. The cross section increases rapidly to a value of about 10^{-30} cm^2 at 105 Mev and then increases gradually to a value of about 10^{-28} cm^2 at 340 Mev. In order to explain the low threshold, it must be assumed that substantially larger particles than alpha-particles are emitted from the excited nucleus, and the reactions are therefore of a type which might more properly be termed fission. As an example, the extreme reaction $Cu^{63}+p\rightarrow Cl^{38}+Al^{25}+n$, which is energetically most economical but still endoergic, has a threshold of about 50 Mev.

This result made it seem worthwhile to investigate another such reaction in copper and to extend the threshold studies to other elements in the middle portion of the periodic system in order to see whether analogous reactions might occur as a general rule. The variation with energy of the yields of radioactive Na^{24} from copper and radioactive $Sc^{43,44}$ from bromine were studied. The formation of radioactive Co^{61} from silver, radioactive Na^{24}, Ga^{66}, and Ga^{72} from tin, and radioactive Ga^{66} and Ga^{72} from barium were also studied. In all cases the observed thresholds were well below the thresholds calculated for the reactions in which the maximum number of alpha-particles are emitted from the nucleus. Additional evidence for fission of the medium weight elements was found from some preliminary experiments on the ranges of recoil fragments from copper irradiated with 340-Mev protons.

II. PROCEDURE

Irradiations with high energy protons were carried out in the circulating beam of the 184-inch frequency modulated cyclotron, and the proton energy was adjusted by varying the radial distance of the target from the origin of the beam. Foil targets 5 to 10 mils thick were used in the cases of copper, silver, and tin. The bromine was bombarded in the form of ammonium bromide powder wrapped in special purity aluminum foil, and the barium was bombarded in the form of barium carbonate wrapped in the special purity aluminum foil. Since the degradation of the energy of the high energy protons is small in traversing the targets used, all targets can be considered thin targets.

The special purity ammonium bromide was synthesized by forming hydrogen bromide from very pure phosphorous tribromide and mixing the gas with very pure gaseous ammonia in a cooled plastic container. The barium carbonate was prepared from special purity

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* This work was performed under the auspices of the AEC.

[1] Perlman, Goeckermann, Templeton, and Howland, Phys. Rev. 72, 353 (1947).

barium chloride received from Dr. A. J. Keyes and Dr. A. J. King of Syracuse University.

Determinations of the intensity of the internal, proton beam for the silver bombardments and the tin bombardments, in which the yields of Ga^{66} and Ga^{72} were studied, were made by bombarding aluminum foils in conjunction with the regular target foils. This monitor foil undergoes a nuclear reaction $Al^{27}(p,n2p)Na^{24}$ for which the cross section has been determined as a function of the energy of the bombarding proton.[2] The cross sections for the formation of the Na^{24} and the Cl^{38} from copper were determined by comparing the amounts of the isotopes formed with the amounts of radioactive Cu^{64} formed by the $Cu^{65}(p,pn)Cu^{64}$ reaction in a given bombardment. The cross section for the reaction $Cu^{65}(p,pn)Cu^{64}$ as a function of the energy of the bombarding proton has previously been determined.[3] The cross sections for the formation of the $Sc^{43,44}$ from bromine, the Na^{24} from tin, and the Ga^{66} and Ga^{72} from barium were calculated using an estimated value of $\frac{1}{2}$ microampere for the beam current.

After irradiation, the targets were dissolved, and the elemental fractions for the various nuclides were separated by the chemical procedures to be discussed in detail in Sec. V. Separation procedures were designed to give a radiochemical purification factor of about 10^8 from all other activities formed in the target.

Counting of the activities was done on an end-window, alcohol-quenched, argon-filled, Geiger counter tube with a mica window of $\sim$3-mg/cm^2 thickness, used in conjunction with a scale of 64 counting circuit. The nuclides were characterized by half-life determinations, absorption measurements, and sign of particulate radiation emitted.

A crude beta-ray spectrometer was used in the determination of the sign of the beta-particles and was especially useful where nuclides with similar half-life, but differing in sign of particle, had to be resolved.

Thresholds for the various reactions include the mass difference between the reactants and products and the excitation energy which the nuclear fragments or alpha-particles must have in order to pass over the potential barrier. Where available, measured mass values for the reactants and products were used in making the mass balance. The masses of radioactive fragments formed, or considered as being formed as products of the nuclear reaction were calculated by adding the mass equivalent of the decay energy of the radioactive nuclide to the mass of the stable daughter nuclide.

The excitation required for the passage over the potential barrier was calculated assuming that the fragments are spherical and tangent at the nuclear radii (taken as 1.48×10^{-13} $A^{\frac{1}{3}}$ cm). Calculations of the coulombic energy requirements for reactions in which a number of alpha-particles are emitted were made on the basis that the alpha-particles come out consecutively. The coulombic requirement for each alpha-particle was calculated on the basis of the alpha-particles being tangent to the daughter nucleus; and, as an example, for the reaction $Cu^{63}(p,pn6\alpha)Cl^{38}$, the first alpha-particle is assumed to be tangent to Co^{59} at the nuclear radius, the second tangent to a Mn^{55} nucleus, etc.

The experiments on the determination of the apparent average ranges of the recoil fragments from copper irradiated with 340-Mev protons were done in the circulating beam of the 184-inch cyclotron. Pieces of Nylon (0.5×2.5 cm) were placed on both sides of the copper foil (0.5×2.5 cm) to catch the recoil fragments. After irradiation, the pieces of copper and Nylon were dissolved, carriers added, and the elemental fractions were separated and measured.

The ranges of the recoil fragments were calculated on the basis of an isotropic distribution and constant range for the recoils. The copper foils were thick relative to the range of the fragments observed. The ratio of the number of atoms of a nuclide appearing in the Nylon catcher foils to the number of atoms in the copper were related to the ranges by the following treatment of the data.

By integration, the expression

$$0.5\int_0^R [(R-x)/R]dx=\tfrac{1}{4}R,$$

where R is the range of the fragments in the copper, and x is the distance from the surface of the copper, shows that $\frac{1}{4}$ of the fragments formed in a thickness R of the surface of the copper will appear in the catcher foils. The relationship $R=(4A_R/A_{\mathrm{tot}})T$, where R is the range of the recoil fragment, A_R is the activity collected in the catcher foil, A_{tot} is the activity in the copper foil plus that caught on the Nylon, and T is the thickness of the copper, gives the range R of the recoils in terms of the information determined experimentally.

III. RESULTS

A. Radioactive Products from Copper

Radioactive Cl^{38}

The calculated threshold for the spallation reaction $Cu^{63}(p,pn6\alpha)Cl^{38}$ is about 110 Mev, while the threshold for the extreme reaction $Cu^{63}+p\rightarrow Cl^{38}+Al^{25}+n$ is about 50 Mev.

A plot of the cross section for formation of Cl^{38} from copper is given in Fig. 1. The cross section rises rapidly from 70 Mev to 105 Mev and then increases gradually to a value of 4×10^{-29} cm^2 at 346 Mev. Below 70 Mev the level of activity formed was so low that identification of the Cl^{38} was not possible; but above 70 Mev the nuclide was identified in all cases through chemical separation, measurement of half-life with a Geiger

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[2] R. L. Folger and P. C. Stevenson, unpublished work (1950).

[3] R. E. Batzel and G. T. Seaborg, unpublished work (1950).

counter, and observation of the sign of the beta-particles with a simple beta-ray spectrometer.

With cross sections so low, an immediate question is that of impurities, since a small amount of impurity with a high cross section for formation of Cl^{38} might be a source of the observed activity. Special purity copper foil was used; and a spectrographic analysis submitted with the copper shows less than 0.00007 percent iron, less than 0.0001 percent nickel, less than 0.00005 percent chromium, and less than 0.0001 percent sulfur (the sulfur was chemically analyzed). As an additional check, radioactivation methods were also used to check for all possible impurities which might account for the observed yield of Cl^{38}. The results of the activation experiments are listed in Table I.

Except for the value listed for scandium, the cross sections for the formation of the Cl^{38} listed in column two are experimental results from irradiations of the corresponding elements in column one. The cross-section value for scandium was estimated from the observed cross sections for calcium and titanium. Column three lists the amounts of the different elemental impurities which must be present in the special purity copper to produce the number of atoms of the Cl^{38} observed in the chlorine fraction isolated from the irradiated copper. Column four lists the limits set on the actual amounts of the elemental impurities present in the special purity copper, as determined by spectrographic and radioactivation methods. Column five gives the ratios of the amounts of the Cl^{38} to the amounts of Cl^{34} observed in the chlorine fractions separated from the respective elements after irradiation with 85-Mev protons. The ratios listed may be compared with the ratio of two observed in the chlorine fraction separated from the special purity copper irradiated with 85-Mev protons.

Potassium as a possible impurity in the copper, which might be a source of the observed Cl^{38}, can be eliminated on the basis of the ratio of the number of atoms of Cl^{38} formed compared with the number of atoms of Cl^{34} formed in bombardment of potassium with 85-Mev protons as shown in Table I, aside from the fact that potassium is not a likely impurity in electroplated copper. Calcium impurity was eliminated as a possible source of the activity by considerations shown in the above table: amounts of calcium present, cross section for formation of Cl^{38}, and ratio of Cl^{38} to Cl^{34}. Scandium and titanium were eliminated as possible impurities in the copper by cross-section considerations and results of radioactivation analyses performed by irradiating the copper with 30-Mev protons and determining the amount of radioactive Sc^{44} formed. This separation of Sc^{44} also served as a check on the calcium, since Sc^{44} could be formed by a (*p*,*xn*) reaction on calcium. The radioactivation analysis for vanadium was performed by comparing yields of radioactive Cr^{51} formed in 30-Mev proton bombardments of vanadium with the amount of Cr^{51} formed in the copper foil under similar bombardment conditions. The very small cross section for formation of Cl^{38}, as listed in Table I, makes it unlikely for chromium or any of the heavier medium weight elements to be present in amounts which would contribute to the Cl^{38} observed, since their cross sections for formation of Cl^{38} in bombardments with 85-Mev protons would be lower than that obtained for chromium.

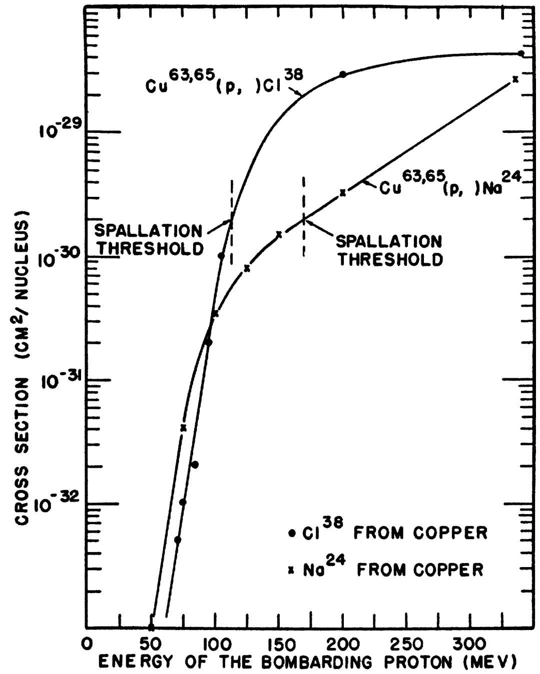

FIG. 1. Cross section for formation of Cl^{38} and Na^{24} from copper *vs* energy of the bombarding proton.

Radioactive Na^{24}

The threshold for the reaction $Cu^{63}(p,p3n9\alpha)Na^{24}$ should be about 170 Mev, of which 100 Mev is due to mass difference and 70 Mev to the coulombic requirement. For the extreme reaction $Cu^{63}+p\rightarrow Na^{24}+K^{39}+n$, the calculated threshold is about 50 Mev.

The Na^{24} was characterized by absorption and half-life measurements. A plot of the cross section for forma-

TABLE I. Examination of possible impurity contribution to observed Cl^{38} activity from copper.

Element	Cross section for formation of Cl^{38} at 85 Mev	% Impurity necessary to explain observed activity	% Impurity determined	Ratio of Cl^{38} to Cl^{34} formed in bombardment of element[a]
K	$<10^{-27}$ cm²	>0.001	<0.01 (spect.)	0.06
Ca	$<10^{-27}$	>0.001	<0.0001 (spect.)	0.10
Sc	$<10^{-27}$	>0.001	<0.0001 (rad.)	
Ti	$<10^{-28}$	>0.0016	<0.0001 (rad.)	10.0
V	$<10^{-28}$	>0.01	<0.0001 (rad.)	
Cr	$<10^{-29}$	>0.1	<0.01 (spect.)	

[a] The ratio Cl^{38} to Cl^{34} formed from the copper at 85 Mev is about 2.

tion of the Na^{24} against the energy of the bombarding proton is shown in Fig. 1. In addition to the fact that no amounts of impurities were found in the copper which could explain the activity observed, the fact that the cross section drops so rapidly with decreasing energy of the bombarding proton is also a good indication that the Na^{24} is not formed from an impurity. The cross section determined at 50 Mev is an upper limit, since the observed activity could not be characterized definitely.

B. Radioactive Sc^{44} from Bromine

The threshold for the reaction $Br^{79}(p,p7n7\alpha)Sc^{44}$ should be about 190 Mev, of which 70 Mev is due to mass difference and 120 Mev is required for the potential barrier. For the extreme reaction $Br^{79}+p\rightarrow P^{34}+Sc^{44}+2n$, the calculated threshold is about 80 Mev.

The characteristic 3.9-hr, 1.5-Mev positron disintegration of Sc^{44} was observed in bombardments of bromine with 125- and 140-Mev protons. The chemically separated activity was characterized by absorption and half-life measurements; and the observed activity would include any Sc^{43} formed, since resolution of the two 3.9-hr activities was not possible. An attempt was made to find the characteristic activity of Sc^{44} in bromine bombarded with 100-Mev protons, but the small amount of activity could not be identified definitely. In a bombardment at 70 Mev, the scandium fraction showed only 90 disintegrations per minute of an $\sim$4-hr activity. Comparable bombardments of bromine with 125- and 140-Mev protons yielded about 10,000 and 30,000 disintegrations per minute, respectively, of the 3.9-hr Sc^{44}. The cross section for formation of the Sc^{44} at 125 Mev is on the order of 10^{-32} cm^2 and is in line with the cross section observed for the formation of Cl^{38} from copper.

The fact that the cross section for formation of Sc^{44} falls off so rapidly as the energy of the bombarding proton is lowered from 140 Mev to 70 Mev rules out the possibility that the observed Sc^{44} is formed from small amounts of calcium, scandium, titanium, or vanadium impurities. The cross sections for formation of Sc^{44} from the elements above vanadium would become increasingly smaller in this energy range, so such impurities are not likely. Spectrographic analysis of the ammonium bromide used as target material showed less than 0.001 percent calcium. Scandium, titanium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, zinc, and gallium were not detectable in the spectrographic analysis. The limit set on the amounts of any of these elements present is 0.01 percent. A limit of less than 0.00015 percent iron impurity was set by colorimetric methods.

C. Radioactive Co^{61} from Silver

The production of the nuclide Co^{61} from silver was studied as a function of energy, but the results must be termed borderline owing to a small amount of copper impurity present in the silver. Since the Co^{61} is formed as a spallation product of copper, it was necessary to determine the amount of copper impurity present in the silver, and the radioactivation method was used. Results of the experiments showed 1×10^{-3} percent copper. The variation with energy of the cross section for formation of the Co^{61} from copper was determined, and the contribution of the Co^{61} formed from the copper impurity was subtracted from the total activity of Co^{61} formed during bombardment of the silver. Resolution of the activity showed that about equal amounts were formed from the silver itself and from the 0.001 percent copper impurity in bombardments at 180 Mev. The observed cross section for formation of Co^{61} from silver at this energy is about 10^{-32} cm^2.

For the reaction in which the maximum number of alpha-particles are emitted, $Ag^{107}(p,p6n10\alpha)Co^{61}$, the calculated threshold is about 210 Mev, of which about 90 Mev is brought about by mass difference; and for the reaction $Ag^{107}+p\rightarrow Co^{61}+Sc^{45}+2n$, the calculated threshold is about 60 Mev. Attempts were made to continue the cross section work at energies lower than 180 Mev; but the problem of resolving the Co^{61} activity formed from the small amount of copper impurity and that formed from the silver alone becomes increasingly difficult, since the cross section for formation of Co^{61} from silver becomes smaller as the energy of the bombarding proton is lowered, while the cross section for formation of Co^{61} from the copper impurity is relatively constant in this energy range.

D. Radioactive Products from Tin

Radioactive Na^{24}

The calculated threshold for the reaction in which the maximum number of alpha-particles are emitted is about 425 Mev, of which 230 Mev is due to mass difference. The extrene reaction ${}_{50}Sn^{118}+p\rightarrow Na^{24}+{}_{40}Zr^{94}+n$, which is exoergic by about 8 Mev with respect to mass difference, has a calculated threshold of about 50 Mev.

The plot of the cross section for formation of the Na^{24} *versus* energy is shown in Fig. 2. The cross-section values plotted are good to a factor of two and possibly better, since all the irradiations were carried out under conditions where the beam intensity should have been the same.

The point at 75 Mev represents an upper limit for the yield, since the activity in the sodium fraction was too small to be identified positively as Na^{24}. The shape of the excitation function almost precludes the possibility of impurities just above sodium in the periodic table. The spectroscopic analysis showed the tin to be 99.999 percent pure with no impurities detectable which could explain the observed activities.

Radioactive Ga^{66} and Ga^{72}

The calculated thresholds for the nuclear reactions $Sn^{118}(p,7n10\alpha)Ga^{72}$ and $Sn^{118}(p,13n10\alpha)Ga^{66}$ are about 230 and 280 Mev, respectively; and for the extreme reactions $Sn^{118}+p\rightarrow Ga^{72}+Ca^{45}+2n$ and $Sn^{118}+p\rightarrow Ga^{66}+Ca^{49}+4n$ the thresholds are about 70 and 90 Mev, respectively. The thresholds for formation of Ga^{66} and Ga^{72} from some of the other tin isotopes may be lower by several Mev, but the examples calculated give approximate values on which to base our reasoning.

The purified gallium fraction isolated from the bombarded tin showed the characteristic activities of Ga^{66}(9.5-hr half-life) and Ga^{72}(14.3-hr half-life), and the absorption curve of Fig. 3 shows the presence of the $\sim$3-Mev positron characteristic of Ga^{66}. The gallium activities were identifiable at energies of 150 and 180 Mev with cross sections based on elemental tin of about 10^{-32} cm^2 and with the value at the higher energy a factor of approximately two times higher than that at the lower energy. Amounts of gallium activity formed in bombardments of tin at 100 Mev were too small to identify definitely, and no gallium activity was observed in the gallium fraction separated from tin bombarded with 80-Mev protons.

The spectrographic analysis presented with the special purity tin showed less than 0.0001 percent arsenic; and no zinc, gallium, or germanium were detectable. The variation of the yield of Ga^{66} with energy of the bombarding particle rules out the probability of the observed activity coming from zinc or gallium, since the cross section for formation of Ga^{66} from zinc or gallium would rise as the energy is lowered from 180 to 80 Mev, while the yield of Ga^{66} from the tin falls off as the energy is lowered.

E. Radioactive Ga^{66} and Ga^{72} from Barium

The calculated threshold for the reaction $Ba^{137}(p,20n13\alpha)Ga^{66}$ is about 370 Mev (200-Mev mass difference and 170-Mev potential barrier). For the extreme reaction $Ba^{137}+p\rightarrow Ga^{66}+Fe^{60}+12n$, the calculated threshold is about 150 Mev. The threshold for formation of Ga^{72} by a similar extreme reaction is some 40 Mev lower.

The gallium fraction isolated from barium irradiated with 335-Mev protons showed the characteristic activities of Ga^{66} and Ga^{72}. The cross sections for formation of these nuclides are about equal and are around 10^{-31} cm^2. The special purity barium contained no spectroscopically detectable impurities, with the exception of a trace of iron. Only the single irradiation was performed, because of the lack of sufficient special purity barium; but the observed cross section is in line with the others reported and seems to indicate that the gallium isotopes are probably formed by a fission reaction.

F. Evidence from Recoil Experiments

Some recent experiments on the range of recoil fragments from copper bombarded with 340-Mev protons have been done in conjunction with this work. Preliminary results indicate that larger fragments than alpha-particles must be emitted from the bombarded nucleus in order to explain the observed energies of the recoils.

Assuming an isotropic distribution for the recoil fragments from the irradiated copper (as discussed in Sec. II), it was found that Cr^{49} nuclei have an average apparent range of about 0.3 mg/cm^2 of copper against the direction from which the incident protons approach the target, and a range of about 2.5 mg/cm^2 of copper with the direction of the beam. Using the formulas for stopping power developed by Knipp and Teller[4] for calculating the energy losses brought about by electronic and nuclear interactions, the energy of the recoils against the beam was found to be about 1 Mev and with the beam, about 13 Mev.

There is a distortion of the recoil distribution, owing to the momentum imparted to the nucleus by the impinging proton, causing more recoil nuclei to appear with the beam and fewer against the beam. Since the ranges were calculated on the basis of an isotropic distribution for the recoils, the calculated range with the beam is larger and that against the beam is smaller than the actual range. The range distribution was corrected, assuming 5 Mev as the kinetic energy that the Cr^{49} fragment received when the excited copper nucleus split, and assuming that the excited nucleus has had momentum transferred to it by the impinging proton equivalent to 1 Mev of kinetic energy for the excited nucleus. The corrected ranges were about 0.6 mg/cm^2 of copper against the beam and about 2.2 mg/cm^2 of copper with the beam, and they are equivalent to energies of about 2.0 Mev and 10 Mev, respectively. These energies correspond to 5.2 Mev of kinetic energy imparted to the Cr^{49} fragment when the excited nucleus breaks up and correspond to a momentum transfer to the copper nucleus equivalent to about 1 Mev of kinetic energy.

A reaction which seems to explain the observed recoil energies is $Cu^{63}+p\rightarrow Cr^{49}+C^{12}+3n$. The C^{12} would leave the excited nucleus with a kinetic energy of about 20 Mev, which is just equivalent to the height of the potential barrier, and would give a recoil energy of about 5 Mev to the Cr^{49}. Explanation of the observed results in terms of the emission of smaller fragments, such as neutrons, protons, or alpha-particles would require a great deal of asymmetry in both the energy and distribution of the particles in the process of the break down of the excited nucleus. The observed transfer of momentum to the copper nucleus corresponds to an excitation of the nucleus which would be consistent with the reaction written. In line with the

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[4] J. Knipp and E. Teller, Phys. Rev. **59**, 659 (1941).

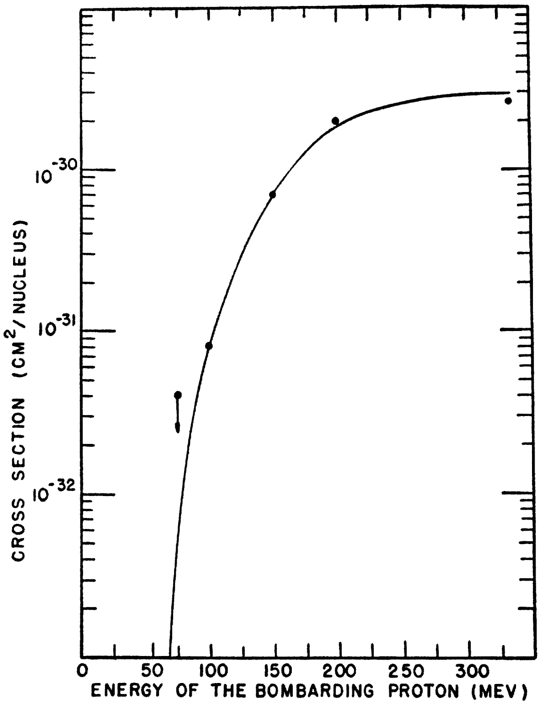

FIG. 2. Cross section for formation of Na^{24} from tin *vs* energy of the bombarding proton.

results of the previous sections, it seems reasonable to assume that a reaction of the type written for the formation of Cr^{49} is correct.

IV. DISCUSSION

The coulombic energy requirement for the emission of the charged particles in the above reactions gives rise to a large contribution to the calculated threshold, and the possibility of the alpha-particles being emitted at energies well below the top of the coulombic barrier should be considered, since in some cases the observed thresholds exceed the values of the mass requirements alone. Calculations of the transmission coefficient[5] for the coulombic barrier show that the time required for a succession of alpha-particles to be emitted with excitation energies well below the top of the barrier is too long to allow this type of nuclear reaction to compete with other reactions in which neutrons and protons (coming out over the barrier) are emitted. It is possible, however, that the coulombic requirement is lowered somewhat because of the effective lowering of the potential barrier at high excitation energies.[6] The only observed thresholds which are borderline are those of the Co^{61} from silver and Ga^{66} and Ga^{72} from barium. The other thresholds are well below the values which could be explained by penetration or an effective lowering of the barrier at the high excitation energies.

The shapes of the excitation curves for the formation of Na^{24} and Cl^{38} from copper are quite similar in the low energy region. This similarity could be attributed to the fact that they are formed by reactions which require about the same amount of excitation energy. The extreme fission reactions have about the same threshold, and the Na^{24} fragment corresponds to a very reasonable complementary fragment corresponding to Cl^{38}. The excitation curves for the formation of Na^{24} and Cl^{38} from copper probably represent sums of several reactions contributing to the total observed cross section. In the low energy range, the main contribution is probably that of the pure fission reaction, while, as the energy is raised, reactions in which smaller fragments are given off begin to contribute to the total reaction. At the high energy end, spallation reactions probably begin to contribute markedly. The difference in the two curves at the higher energies is probably due to the larger contribution of the direct formation of Cl^{38} by spallation reactions.

It is interesting to note that the emission of nuclear fragments a little larger than alpha-particles has been observed in high energy nuclear reactions on medium weight elements. The "hammer tracks" of Li^8 are very well known in reactions induced by cosmic radiation and have also been identified as a reaction product of 340-Mev proton and 190-Mev deuteron bombardments of some of the medium weight elements.[7] Evidence has also been found for the existence of lithium isotopes as a reaction product of 50- to 340-Mev proton and helium

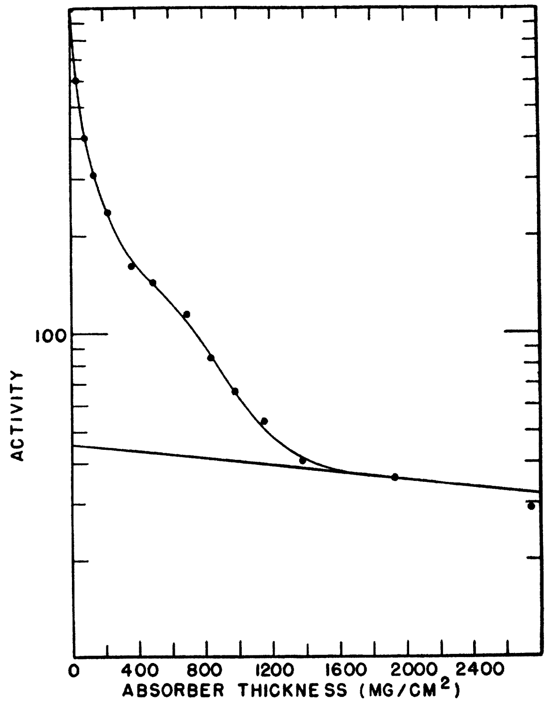

FIG. 3. Aluminum absorption curve for gallium fraction from tin bombarded with 180-Mev protons.

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[5] G. Gamow, Z. Physik **51**, 204 (1928).

[6] K. J. Le Couteur, Proc. Phys. Soc. (London) **A63**, 259 (1950).

[7] S. Wright, Phys. Rev. **77**, 742 (1950).

ion bombardments of tin.[8] Radioactive Be^7 has also been identified as a product of 340-Mev proton bombardments of some of the medium weight elements.[8] Heavy fragments have also been observed in reactions induced by cosmic radiations.[9–11]

Apparently, when the energy threshold requirements are met, large fragments are emitted among the competitive products of nuclear reactions throughout the entire range of atomic numbers of the elements. This is certainly not surprising, and the measured yields reported here seem to be quite reasonable. It seems certain that the size of the fragments varies continuously from those (neutrons, protons, and alpha-particles) which accompany what are conveniently called spallation reactions, through intermediate sizes (for example, Li^8, etc.), on up to sizes such that the nucleus is split essentially into several pieces of comparable weight. Apparently, a number of reactions in which there occurs the latter type of nuclear splitting have been observed in the present investigation, and perhaps the term "fission" is as proper a name as any to apply to the process. As a result of this work it seems that fission reactions occur in all nuclei if the necessary excitation energy is available.

V. EXPERIMENTAL DETAILS

A. 184-Inch Cyclotron Bombardments

All targets were bombarded in the internal beam of the 184-inch cyclotron. Foils were used as target materials in the cases of the copper, silver, and tin; for the bromine bombardments, very pure ammonium bromide powder wrapped in special purity aluminum foil was used; and for the barium bombardments, very pure barium carbonate wrapped in the special purity aluminum foil was used. These targets were mounted on the end of a movable probe, and the energy of the bombarding proton was selected by an accurate adjustment of the radial distance from the origin of the beam to the leading edge of the target.

The maximum energy of the bombarding proton is a function of the radial distance from the origin of the beam to the leading edge of the target; and since the variation of the energy with radius is known precisely, the precision with which the bombarding energy can be selected depends on the measurement of the distance from the leading edge of the target to the origin of the beam. The measurements are probably good to within 5 Mev at energies of 100 Mev. The spread in energies of the particles for a given radial setting varies from about 3 percent at 348 Mev to 15 to 20 percent at energies around 60 to 70 Mev. This spread in the energy of the particles does not affect the maximum energy of the particles for a given radial setting, since the spread is due to the fact that some of the paths of the particles, though circular, have centers which oscillate about the origin of the beam, and the fixed circular path gives the maximum energy to the particle for a given radial setting of the target.

B. Chemical Separation Procedures

The chemical separation procedures must meet severe requirements, since the amount of interfering radioactivities must be reduced to such a degree that the nuclides of interest can be accurately measured by their characteristic radiations. The cross sections for formation of the nuclides produced in the targets bombarded in this series of studies vary from about 10^{-24} cm^2 down to the observed cross sections of about 10^{-32} cm^2. Since the number of atoms of the radio-nuclides which were studied here were extremely small compared with the number of atoms of the interfering radionuclides, it was necessary that the chemical separation procedures give a radioactive decontamination of at least 10^8 from all other radioactivities formed in the targets.

Sodium from Copper

The copper target was dissolved in concentrated hydrochloric acid and hydrogen peroxide, and 5 mg of sodium chloride was added as carrier. Carriers for the elements zinc through potassium were added, the solution adjusted to $1.5N$ in hydrochloric acid, and the copper precipitated as the sulfide. The solution was boiled to dryness to remove the excess acid, the residue dissolved, and then the solution made alkaline with ammonium hydroxide. Hydrogen sulfide was added, and the sulfides and hydroxides were precipitated. Additional 3-mg portions of carriers zinc through scandium were added and precipitated. This scavenging process was repeated twice. Excess hydrochloric acid was added to the alkaline sulfide solution, the acidified solution boiled to dryness, and the ammonium chloride driven off. The residue was dissolved in water, the solution checked to make sure it was neutral, and the sodium precipitated as the sodium zinc uranyl acetate. Two 5-ml portions of zinc uranyl acetate were used to wash the precipitate, and the precipitate was then dissolved in absolute ethyl alcohol saturated with hydrogen chloride gas. The resulting solution was cooled in an ice bath and the sodium chloride separated by centrifugation. The sodium chloride precipitate was washed with a 5-ml portion of the alcohol-hydrogen chloride solution. The sodium chloride was dissolved in water and the solution neutralized with potassium hydroxide. The sodium was again precipitated as sodium zinc uranyl acetate, and the precipitate was then washed with additional zinc uranyl acetate and dissolved in absolute alcohol saturated with hydrogen chloride. An additional portion of the alcohol-hydrogen chloride solution was added to wash the precipitate, and then the precipitate was transferred to a plate for counting.

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Chlorine from Copper

The copper target was dissolved in concentrated nitric acid and 5 mg of chlorine as sodium chloride added to the solution. The chlorine was distilled and trapped in a solution made up of 3 to 4 ml of water containing 2 to 3 drops of concentrated nitric acid and sufficient silver nitrate to precipitate the 5 mg of chlorine as silver chloride. The resulting solution was heated almost to boiling to coagulate the precipitate. The precipitate was separated and washed with dilute nitric acid. The silver chloride was dissolved in ammonium hydroxide and carriers for zinc, copper, nickel, cobalt, and iron added. The ferric hydroxide precipitate was removed by centrifugation and a slight excess of iodine added as sodium iodide. The silver iodide precipitate was removed, the solution made $0.5N$ in nitric acid, 5 to 10 mg of bromine added as sodium bromide, the bromide and iodide oxidized with persulfate, and the bromine and iodine distilled. Silver nitrate was added to the solution and silver chloride was precipitated again. The precipitate was washed with dilute nitric acid and redissolved in dilute ammonium hydroxide. Carriers for zinc, copper, nickel, cobalt, and iron were again added and the ferric hydroxide precipitate removed. The solution was acidified with nitric acid and the silver chloride reprecipitated. The steps for separation of bromine should not be necessary, but were used in order to provide for the eventuality of a very small amount of selenium impurity in the copper which might form some interfering bromine radioactivities during the bombardment of the copper.

Scandium from Bromine

The ammonium bromide powder was dissolved in water, 5 mg of scandium carrier added, the solution adjusted to a *p*H of 3.0, and then transferred to a separatory funnel (consisting of a 40-ml calibrated centrifuge cone with a stopcock sealed to the bottom). Ten ml of $0.5N$ thenoyltrifluoroacetone in benzene was added and the mixture stirred for 5 minutes to extract the chelate compound which scandium forms with the thenoyltrifluoroacetone. The organic layer containing the scandium was washed 3 times with 10-ml portions of water, and then the scandium was extracted from the organic phase with 10 ml of $1N$ hydrochloric acid. Five mg each of arsenic, selenium, and germanium carriers were added. The solution was made alkaline with ammonium hydroxide; and hydrogen sulfide was added to form the polysulfides of selenium, arsenic, and germanium. The scandium precipitates as the hydroxide from this alkaline solution. The precipitate was washed with ammonium sulfide and the scandium dissolved in $6N$ hydrochloric acid. Iron and gallium carriers were added and extracted from the acid solution with ethyl acetate. Approximately 5 mg of arsenic carrier was added and arsenic sulfide precipitated. Another 5 mg of arsenic carrier was added and the arsenic again precipitated as the sulfide. The solution was made alkaline with ammonium hydroxide to precipitate the scandium as the hydroxide. The precipitate was dissolved in hydrochloric acid and the solution adjusted to a *p*H of 3.0. The scandium was extracted with 10 ml of $0.5M$ thenoyltrifluoroacetone, the organic layer washed with water, and the scandium extracted from the organic layer with $1N$ hydrochloric acid. The solution containing the scandium was adjusted to $0.5N$ in hydrochloric acid, and 0.2 ml of $27N$ hydrofluoric acid was added to precipitate scandium fluoride. The precipitate was washed with 5 ml of $0.5N$ hydrochloric acid containing 2 drops of $27N$ hydrofluoric acid. The scandium fluoride precipitate was dissolved in sulfuric acid, the hydrofluoric acid distilled, and the scandium precipitated as the hydroxide.

Cobalt from Silver

The silver target was dissolved in nitric acid, 5 mg of cobalt added, and the silver precipitated as silver chloride. Approximately 5 mg of iron carrier was added and the solution made alkaline with ammonium hydroxide. The ferric hydroxide precipitate was washed with hot ammonium chloride and the wash added to the supernatant solution. Approximately 5 mg of strontium carrier was added and strontium carbonate precipitated by adding sodium carbonate. The strontium carbonate precipitation was repeated and the supernatant solution adjusted to $0.3N$ in hydrochloric acid. Five-mg portions of cadmium, palladium, and copper carrier were added to the solution and the sulfide precipitated with hydrogen sulfide. The supernatant solution was made alkaline with ammonium hydroxide and the cobalt precipitated as cobalt sulfide. The precipitate was washed with water to remove the ammonium sulfide and then dissolved in nitric acid. Silver carrier was added to the solution and silver chloride precipitated to remove any silver which might not have been removed. The solution was made alkaline with potassium hydroxide, and the cobalt hydroxide precipitate was removed. The cobalt hydroxide was dissolved in acetic acid and potassium cobaltinitrite precipitated by adding an equal volume of a hot saturated solution of potassium nitrite. The precipitate was washed and then dissolved in concentrated hydrochloric acid. The solution was adjusted to $0.1N$ in hydrochloric acid, and 3 g of sodium thiocyanate was added. The thiocyanate complex ion of cobalt was extracted into a one to one mixture of amyl alcohol and diethyl ether. The cobalt was extracted from the alcohol-ether layer with $6N$ aqueous ammonium hydroxide and the cobalt precipitated as the sulfide. The sulfide precipitate was dissolved in concentrated nitric acid, the solution neutralized with potassium hydroxide and then acidified with acetic acid. The cobalt was precipitated in the final form as potassium cobaltinitrite.

Sodium from Tin

The separation of the sodium from the tin is almost the same as that for the separation from copper. Different carriers, ruthenium, palladium, molybdenum, silver, strontium, rubidium, lanthanum, iron, and calcium were added and then precipitated several times to remove the interfering activities. The subsequent sodium precipitations were carried out in the manner given in the section on sodium from copper.

Gallium from Tin

The tin target was dissolved in concentrated hydrochloric acid with just sufficient concentrated nitric acid added dropwise to speed the dissolving of the tin. Five mg of gallium carrier and "hold-back" carriers for iron, antimony, ruthenium, and molybdenum were added to the solution. The solution was adjusted to $7.75N$ in hydrochloric acid and the gallium extracted with isopropyl ether. The ether layer was washed six times with 10 ml portions of $7.75N$ hydrochloric acid. The gallium was extracted from the ether layer with water. The water layer was adjusted to $1N$ in hydrochloric acid and the sulfides precipitated with hydrogen sulfide. Milligram amounts of molybdenum, antimony, and palladium were added to the supernatant solution and the sulfides precipitated. Molybdenum, antimony, and palladium carriers were again added and the sulfides precipitated. Three mg of antimony carrier was added to the supernatant solution and antimony sulfide precipitated. The antimony sulfide precipitation was repeated three more times. The supernatant solution was boiled to remove the hydrogen sulfide and then adjusted to $1N$ in sodium hydroxide. The ferric ion, which follows the gallium through the chemical separations, precipitates here as the hydroxide. Three mg of strontium was added and the strontium precipitated as the carbonate. Five mg of iron carrier was added and the hydroxide precipitated. The solution was adjusted to $7.75N$ in hydrochloric acid and the gallium extracted with isopropyl ether. The ether layer was washed three times with 10-ml portions of $7.75N$ hydrochloric acid, and the gallium was extracted from the ether layer with water. Five mg of ruthenium carrier and 5 ml of concentrated perchloric acid were added and the resulting solution evaporated to dryness. Five ml of concentrated perchloric acid was again added and the solution again evaporated to dryness. The residue was taken up in $1N$ hydrochloric acid, 2 mg of antimony carrier added, and the antimony precipitated as the sulfide. Two mg of antimony was again added to the solution and the sulfide precipitated. The solution was boiled to remove the hydrogen sulfide and adjusted to a *p*H of 5.5 with sodium hydroxide to precipitate gallium hydroxide. The hydroxide precipitate was dissolved and adjusted to $7.75N$ with hydrochloric acid. The gallium was extracted with isopropyl ether and the ether layer washed twice with 10-ml portions of $7.75N$ hydrochloric acid. The gallium was extracted from the organic layer with water and the water layer adjusted to a *p*H of 5.5 with sodium hydroxide to precipitate the gallium hydroxide.

Gallium from Barium

The procedure used for the chemical separation of gallium from the irradiated barium was essentially the same as that described in the section "sodium from tin."

Chromium from Copper

The irradiated copper or the Nylon catcher foils were dissolved in concentrated nitric acid and the nitrate converted to the chloride by boiling with hydrochloric acid. Carriers for the elements calcium through zinc were added, the solution adjusted to $2N$ in hydrochloric acid, and the copper precipitated as the sulfide. The supernatant solution was made alkaline with ammonium hydroxide and the alkaline sulfides precipitated. The precipitate was dissolved in concentrated nitric acid and manganese dioxide precipitated by adding potassium chlorate. The supernatant solution was adjusted to about $0.1N$ in nitric acid, and cooled in an ice bath. Hydrogen peroxide was added to form the blue peroxychromic acid, which was extracted into diethyl ether. The ether layer was washed four times with 5-ml portions of water acidified with two drops of concentrated nitric acid. Five ml of water made alkaline with sodium hydroxide was added to extract the chromium from the ether layer. The ether was removed by evaporation and the chromium oxidized completely with hydrogen peroxide in the alkaline solution. After boiling to remove the excess peroxide, the solution was made slightly acid, and the chromium precipitated in the final form as barium chromate.

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