Observations on Lithium and Beryllium Nuclei Ejected from Heavy Nuclei by High Energy Particles

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The yields of Be^7 produced by 335-Mev protons on nuclei of different atomic number have been determined and found to decrease with increasing atomic number. Reactions are described in which the irradiation of an element with protons or alpha-particles results in products higher by three in atomic number. These reactions are interpreted tentatively as second-order processes in which the initial step is the production of high energy lithium nuclei, which in turn cause nuclear reactions of the type (Li,xn). From the observed over-all yields, deductions are made as to the cross sections for the separate steps.

I. INTRODUCTION

THE conditions and mechanisms by which nuclear fragments of mass number greater than four are ejected from heavy nuclei are beginning to receive consideration. The fission of elements in the region of uranium is a special reaction which does not fall into the category under discussion.

The appearance of "hammer" tracks in photographic emulsions struck by cosmic rays is an indication that Li^8 can be produced in a nuclear disintegration. Under better controlled conditions of projectile energy, Li^8 has been observed with high energy protons and deuterons on a variety of gases[1] and with 26.7-Mev x-rays in a photographic emulsion.[2] Evidence that a wide variety of reactions can take place with medium-light elements in which aggregates of nucleons are split off has been obtained by Batzel and Seaborg.[3] How such reactions differ from heavy element fission is not clear; but there probably are differences stemming from the fact that they are endoergic, while heavy element fission is strongly exoergic.

There is also mounting evidence from cosmic-ray studies[4–8] that a variety of charged fragments may be ejected from nuclei and that some of these may have kinetic energies of the order of 100 Mev. The mechanism by which a complex of nucleons can be given such high kinetic energy is not understood. The phenomenon appears to be not unlike that which is the subject matter of the present studies. The question of cross section for such reactions is probably a crucial point in the consideration of the mechanism, but at this stage no significant data are available.‡

The present studies were introduced by some observations of abnormal charge increase in nuclear reactions. It was found that if bismuth (element 83) is irradiated with high energy deuterons, isotopes of astatine (element 85) are produced.[9] It was suggested that high energy deuterons eject alpha-particles from the bismuth, which in turn react with bismuth by well-known reactions of the type (α,n) to produce astatine. Subsequently, other reactions have been observed in which irradiation with alpha-particles resulted in products with an increase of *three* in atomic number. In such a case it would be inferred that a second-order reaction has occurred with *lithium* nuclei formed in the primary reaction.

The present report deals partly with reactions of this type in which it was found that an increase of three in atomic number could be obtained with high energy alpha-particles or protons. The energy of the projectile, rather than its type, is most important in determining the yield. Measurements have also been made on the yields of Be^7 produced in high energy nuclear encounters as this nucleus is one of the few of this size which can be identified.

II. Be^7 FORMATION

The lightest nucleus above helium which can be identified by its radioactive properties is Be^7, which was chosen for investigation of conditions for production of such fragments. The disadvantage in the use of Be^7 for this purpose is its low sensitivity for detection because of the relatively long half-life (52.9 days)[10] and low counting efficiency (one gamma-ray of 0.48 Mev in 10 disintegrations).[11,12]

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[1] S. C. Wright, Phys. Rev. **77**, 742 (1950).
[2] C. H. Millar and A. G. W. Cameron, Phys. Rev. **79**, 182 (1950).
[3] R. E. Batzel and G. T. Seaborg, Phys. Rev. **79**, 528 (1950).
[4] C. F. Powell and G. P. S. Occhialini, *Nuclear Physics in Photographs* (Oxford University Press, London, 1947).
[5] A. Bonetti and C. Dilworth, Phil. Mag. **40**, 585 (1949).
[6] S. O. C. Sörenson, Phil. Mag. **40**, 947 (1949).
[7] P. E. Hodgson and D. H. Perkins, Nature **163**, 439 (1949).
[8] J. B. Harding, Nature **163**, 440 (1949).
‡ *Note added in proof:* A recent paper by D. H. Perkins [Proc. Roy. Soc. (London) **203**, 399 (1950)] describes extensiveo bservations on the emission of heavy fragments in cosmic-ray stars. The author has identified fragments with charges up to ten units and has found them to have energies in excess of that which would result from coulombic repulsion. The present paper contains deductions in agreement with Perkins' observations and others cited, as is the conclusion that a simple evaporation model cannot explain such high particle energies.
[9] D. H. Templeton and I. Perlman, Abstracts of Papers (Meeting of Am. Chem. Soc., Portland, Oregon, September 13–17, 1948), pp. 55–60.
[10] E. Segrè and C. E. Wiegand, Phys. Rev. **75**, 39 (1949).
[11] R. M. Williamson and H. T. Richards, Phys. Rev. **76**, 614 (1949).
[12] C. M. Turner, Phys. Rev. **76**, 148 (1949).

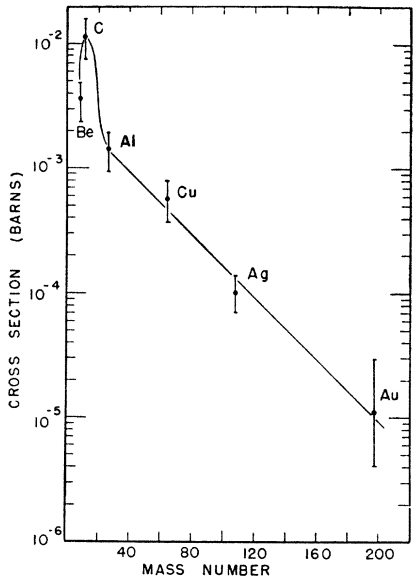

FIG. 1. Yield of Be^7 with 335-Mev protons.

(A) Methods

Beryllium, carbon, aluminum, copper, silver, and gold were irradiated with protons of 335-Mev incident energy. Stacks of foils, 2×4 cm, were used and aligned so that the circulating proton beam penetrated in the direction of the 2-cm dimension. The beam traversing the different targets was therefore degraded in energy by different amounts; for example, the energy range in aluminum was 335–320 Mev, in silver 335–290 Mev, and in gold 335–270 Mev. The accuracy of the cross sections determined and the inferences drawn are not such as to warrant corrections for energy loss nor for attenuation of the beam through nuclear reactions, which becomes appreciable for the heavier elements. The foils were held in place against a copper block and the two foils closest to the block were discarded to avoid contamination from recoil atoms originating in the holder. In some cases thin aluminum foils were wrapped around the target foils to serve as a beam monitor, since Na^{24} from the reaction $Al^{27}(p,3pn)Na^{24}$ is known to be produced with 0.010-barn cross section with protons in the energy range 100–350 Mev.[13]

After irradiation for one to five hours, the targets were worked up and the beryllium fraction isolated with a known weight of added beryllium used to check the yield. In the case of the carbon target, extremely high purity graphite was used; and since no conflicting activities were produced, there were no chemical separations made.

From the aluminum target the only conflicting long-lived isotope produced is Na^{22}. The target was dissolved in HCl, diluted to about 1 liter, some sodium and 10 mg of Be(II) added, and the mixed aluminum and beryllium hydroxides precipitated with ammonia. The precipitate was washed, dissolved in dilute HCl, and the process repeated twice. Only Be^7 could be detected after this treatment.

The Be^7 was separated from the copper, silver, and gold targets by the use of its amphoteric properties. The targets were dissolved in suitable acids, 10–30 mg of Be(II) carrier added and precipitated with ammonia from the copper and silver or, in the case of the gold target, the gold was extracted into ethyl acetate. The resulting beryllium concentrates were dissolved in dilute acids, and sulfide-insoluble substances removed by precipitating a mixture of sulfide-insoluble carriers. Several precipitations were then made from sodium hydroxide solution with iron and other hydroxide insoluble substances used as scavangers. Beryllium is soluble in sodium hydroxide solution. Finally, beryllium as the basic acetate was extracted several times into chloroform in the manner described by McMillan.[14]

(B) Identification of Be^7

Since the radiation from Be^7 consists solely of a gamma-ray of 0.48 Mev, the absence of electrons as well as decay with proper half-life and the distinctive chemistry were used as criteria for the purity of the Be^7. In all cases except that of the gold target it was possible to show that the gamma-rays had the proper half-thickness in lead. The activity isolated from the gold target had but six counts per minute, and positive identification could not be made, especially since there were some shorter lived impurities which had to be resolved.

(C) Cross Sections

The means of monitoring the beam were different for the different targets. For the beryllium target, aluminum foils were used and the Na^{24} measured as already mentioned. The amount of Na^{24} also served to monitor the beam for the aluminum target. For the carbon target, the yield of C^{11} was used according to the excitation curve of Peterson,[15] which shows a cross section of 0.040–0.050 barn in the energy range 350–200 Mev for the reaction $C^{12}(p,pn)C^{11}$. The annihilation radiation of the C^{11} positrons was also used to calibrate the efficiency of the counter for the Be^7 gamma-rays making the assumption that the counting efficiencies for the 0.48-Mev and 0.51-Mev gamma-rays would be the same.

The cross section for the copper target was determined with the yield of Cu^{64} used as a monitor, which has been shown to be 0.025 barn for 340-Mev protons.[16] For the silver target, the yield of 8.2-day Ag^{106} was determined in a separate irradiation with an aluminum monitor, after which it served as a monitor. The gold

[13] P. C. Stevenson and R. L. Folger (private communication).

[14] E. M. McMillan, Phys. Rev. 72, 591 (1947).

[15] Aamodt, Peterson, and Phillips (University of California Radiation Laboratory Unclassified Report UCRL-526, November, 1949) (unpublished).

[16] R. Batzel and G. T. Seaborg (private communication).

targets were handled similarly, yields being based on an arbitrary point on the decay curve of the gold fraction two days after irradiation. The reason for the secondary monitoring for the copper, silver, and gold targets was to eliminate introduction of Be^7 into the target from the aluminum monitor by recoil.

As mentioned, the counting efficiency of the Be^7 gamma-ray was determined by calibrating the counter against annihilation radiation of C^{11}. For the argon-chlorine Geiger tubes used in these studies, the efficiency was found to be 0.65 percent. Finally, to calculate the cross sections for production of Be^7, the yield of 0.48-Mev gamma-rays was taken to be 1 in 10 disintegrations.[11,12] The errors in cross section include those from counting statistics, counter efficiency calibration, chemical yields, and uncertainties in monitor effectiveness. We estimate that the combined uncertainties are some 30 to 40 percent for the targets other than gold, while the yield obtained from the gold is probably reliable only within a factor of two or three.

The yields of Be^7 from 335-Mev protons are plotted in Fig. 1 against the mass number of the target. The general decrease in yield with increase in atomic number or mass number is apparent and amounts to a factor of 1000 between carbon and gold. It is highly probable that Be^7 results from a different process in the different targets. From carbon, one may visualize Be^7 as the residue from spallation reactions of the type, $C^{12}(p,p\alpha n)Be^7$; but, from the heavier nuclei, the Be^7 must be considered as the ejected fragment.

On the basis that the turning point in principal mode of formation might be reached at aluminum between formation of Be^7 as an ejected fragment or as a spallation residue, several spallation products of aluminum were examined. The yields are shown in Table I for 335-Mev protons on aluminum. Since the yields decrease in the expected manner and that of Be^7 is slightly below C^{11}, it is not unlikely that at least part of the Be^7 arises as a spallation residue.

Returning to Fig. 1, it is noted that the yield of Be^7 from a beryllium target (Be^9) is lower than that from a carbon target (C^{12}). Without further study it is not possible to give an explanation. One factor which could contribute to a low yield of Be^7 from Be^9 is the following. It may be assumed that the most important form of excitation in the reaction is the creation of an excited state of Be^9, since the incoming proton will not stay in the nucleus. After this, Be^7 would be reached by evaporation of two neutrons. The first step would result in an excited state of Be^8 from which dissociation into two alpha-particles would compete with further neutron evaporation.

III. ABNORMAL CHARGE INCREASE

In irradiations of tin with ~350-Mev alpha-particles and protons, it has been observed that iodine activities are produced, which means that an increase of three in atomic number has taken place. The iodine activities were identified[17] as the new neutron-deficient isotopes I^{120}, I^{121}, I^{123}, as well as previously known I^{124} and I^{126}; therefore, none could have come from β^- decay processes. It was also possible to rule out fairly conclusively impurities in the tin as the source of the iodine. The most likely explanation is that the iodine nuclei result from second-order reactions in which lithium nuclei are postulated to be ejected from tin, and these in turn transmute other tin nuclei into iodine. The implications of the yields obtained will be discussed.

TABLE I. Yields of products from irradiation of aluminum with 335-Mev protons.

Nucleus	Yield (barn)
Na^{24}	1.0×10^{-2}
Na^{22}	1.2×10^{-2}
F^{18}	5.5×10^{-3}
C^{11}	1.9×10^{-3}
Be^7	1.4×10^{-3}

(A) Methods

The tin targets were irradiated in the circulating beam of the cyclotron for periods of one to eight hours. After irradiation, the targets were placed in a distilling flask with 10 mg of I^- and 30 cc of H_2SO_4. Upon heating, the tin was dissolved and the iodine was distilled over, probably as hydrogen iodide, and was trapped either in ice-cold solution of sulfur dioxide or in sodium hydroxide solution. The solution in either case was acidified with sulfuric acid and the iodide oxidized to iodine with excess nitrite and distilled as iodine into sodium hydroxide solution. Upon acidification the iodine was extracted into carbon tetrachloride. After removal of the iodine from the carbon tetrachloride with sulfur dioxide solution or sodium hydroxide, the extraction cycle was repeated three times. The iodine was finally isolated as silver iodide, which was weighed to determine the chemical yield.

The iodine activities were resolved by following the decay curves and using the counting efficiencies previously estimated,[17] the yields were calculated. From the longer irradiations, it was possible to identify with a spectrometer the conversion electron line of I^{123}, the positron spectrum of I^{124}, and the β^- spectrum of I^{126}.

TABLE II. Cross sections for formation of iodine activities from high energy particles on tin.

Isotope	350-Mev protons (barn)	350-Mev alphas (barn)
30-min I^{120}	0.5×10^{-5}	0.9×10^{-5}
1.8-hr I^{121}	0.1×10^{-5}	1.1×10^{-5}
13-hr I^{123}	0.7×10^{-5}	1.3×10^{-5}
4.5-day I^{124}	0.4×10^{-5}	0.4×10^{-5}
13-day I^{126}	0.6×10^{-5}	0.1×10^{-5}
Total	2.3×10^{-5}	3.8×10^{-5}

[17] L. Marquez and I. Perlman, Phys. Rev. **78**, 189 (1950).

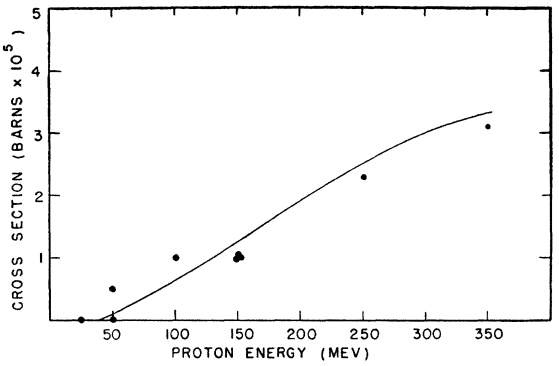

FIG. 2. Excitation function for protons (total iodine cross section).

(B) Yields of Iodine from Tin

Table II shows cross sections for the several iodine activities resulting from high energy protons and alpha-particles on tin. The difficulties of accurate resolution of the different species were considerable and no great confidence can be had in any particular value. Probably comparison of the sums of the several cross sections is more meaningful. It is seen that the total yield with alpha-particles is somewhat greater than with protons but not significantly so. It may be mentioned that 4-min I^{122} and 56-day I^{125} probably were formed also, but the one was not detected because of its short half-life and the other because of its long half-life and low counting efficiency.

In order to see how the yields of iodine activities varied with particle energy, excitation functions were determined for both protons and alpha-particles and are shown in Figs. 2 and 3. The values for energies below 100 Mev became quite uncertain because of low activities, and at about 25 Mev no iodine could be detected. The threshold of detection appeared to be at about 50 to 60 Mev.

The possible role of impurities in the tin as the source of the iodine activities can best be discussed at this point. Spectroscopic examination of the tin showed only traces of the elements lead, copper, iron, arsenic, antimony, bismuth, and silver, each of which was estimated to be present to the extent of less than 5 ppm. Since the formation of iodine with 350-Mev alpha-particles was found to have a cross section of $\sim 10^{-5}$ barn, antimony is ruled out as a source of the iodine. The actual cross sections expected from antimony are at least 100 times lower than would be required. Furthermore, it would not be possible to explain the similar yields for protons and alpha-particles. Tellurium cannot be ruled out quite so conclusively from the chemical analysis, because the limits of detection are poor. However, if tellurium were present, the trends of the excitation curves are the opposite of what would be expected. Also, I^{130} and I^{131} would be formed, and when a low resolution beta-ray spectrometer was used, their spectra were not encountered. Elements of higher atomic number than iodine were ruled out by an analysis for barium radioactivities. Barium activity would be encountered in higher yield than iodine, yet an upper limit for its formation cross section could be set at 10^{-6} barn and it is probably much lower. The iodine activities could not arise from thorium or uranium through fission, since the wrong iodine isotopes are found; and, in any case, very low limits could be set for the presence of these elements from the inability to detect alpha-radioactivity which would be found in high yield from spallation products. Iodine itself as an impurity in the tin was eliminated from consideration by chemical analysis which showed that less than 10 ppm were present. From the yields of comparable reactions to form light antimony isotopes from an antimony target,[18] the iodine would have had to be present to an extent at least ten times greater than the upper limit measured.

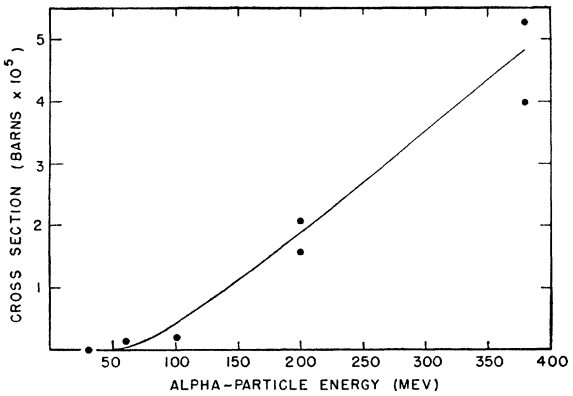

FIG. 3. Excitation function for alphas (total iodine cross section).

IV. DISCUSSION

The explanation for the observed reaction products by means of second-order reactions involving lithium nuclei demands examination with regard to expected and observed yields. The mechanism is in qualitative agreement with the features observed; that is, it accounts for the particular iodine isotopes found, the increase in yield with increase in projectile energy, and the fact that the activities and their yields are nearly the same whether alpha-particles or protons are used. That lithium nuclei can be ejected in high energy nuclear reactions is also inferred from the proof that specific nuclei Li^8 and Be^7 are formed. The objective is then to piece together conditions for each stem of the two-step process which could give the observed over-all results.

There are three important parameters that enter into the determination of the over-all yield, for none of which do we have values: (1) the cross section (σ_{Li}) for the production of lithium nuclei, (2) the energy distribution of the lithium nuclei, and (3) the cross section for the formation of iodine isotopes from lithium nuclei

[18] M. Lindner and I. Perlman, Phys. Rev. 78, 499 (1950).

on tin. The energy distribution for the lithium nuclei (item 2) is of great importance, not only in its effect on the cross section for producing iodine from tin, but also as it determines the ranges of the lithium nuclei in the tin.

The first simplifying assumption made is that regarding the cross section of the lithium reactions with tin to give iodine. It is assumed that all lithium nuclei which enter tin nuclei result in iodine isotopes; that is, all three protons which enter remain. This assumption is probably not seriously in error, because it is known that for alpha-particles in the proper energy range, reactions of the type (α,xn) are most prominent in medium-heavy and heavy nuclei. Recently, it has been shown that (C^{12},xn) reactions occur[19,20] with appreciable cross section, which means that six protons entering a nucleus can all remain. We shall then assume that the cross section for the formation of iodine isotopes is nearly the same as the penetration cross section for lithium into tin given by the formula:

$$\sigma=\pi R^2(1-B/E),$$

where B is the potential barrier height (taken to be 30 Mev), E is the energy of the lithium ion ($E>B$), and πR^2 is the geometric cross section of tin (1.7×10^{-24} cm^2). The total cross section for the production of iodine isotopes (σ_I) is estimated from the measured values for full energy protons or alpha-particles to be 4×10^{-29} cm^2 (Table II).

The cross section for formation of lithium nuclei in the primary reaction can then be calculated for various assumed values of monoenergetic lithium ions of energy ϵ:

$$\sigma_{Li}=\sigma_I\Big/\left[\pi R^2\int_{E=B}^{E=\epsilon} n(1-B/E)dx\right],$$

in which n is the number of tin nuclei per cm^3 and x is the distance traveled by the lithium ions. The relation between E and x was obtained from the theory of the interaction of heavy ions with matter, and the integrations were performed numerically. Some typical results

[19] Miller, Hamilton, Putnam, Haymond, and Rossi, Phys. Rev. **80**, 486 (1950).

[20] Ghiorso, Thompson, Street, and Seaborg, Phys. Rev. **81**, 154 (1951).

TABLE III. Relation between calculated cross section and energy of lithium nuclei.

ϵ(Mev)	36	40	50	80	120	200
σ_{Li}(barn)	3.0	1.2	0.3	5×10^{-2}	2×10^{-2}	5×10^{-3}

are shown in Table III, from which it is seen that the cross sections are surprisingly high, and only those seem to be reasonable which assume that the lithium ions are ejected with high energy, say, 80 Mev or higher. Even if it is assumed that the effective energy of the lithium ions is 80 Mev, the resulting cross section is 0.05 barn, which is 500 times greater than that observed for the formation of Be^7 from silver. The differences in yields for these two cases would have to be reconciled by the arguments that in the one case the sum of a number of lithium isotopes is compared with the single isotope, Be^7, and the greater charge of beryllium as compared with lithium should make its ejection more difficult.

If the deduction that the lithium nuclei are ejected with high kinetic energy should be borne out by some form of direct measurement, it would seem to be impossible to explain this phenomenon through a compound nucleus model. Instead, one must assume that such fragments are ejected before the excitation energy is distributed in order that they may carry away a large fraction of the available energy. Indeed, it is probable that when energy of the order of 200 Mev is evenly distributed throughout a medium heavy nucleus, a fragment containing three protons would compete very poorly in evaporation probability with particles such as protons, and especially neutrons. Qualitatively, these arguments are restatements of the picture given by Serber[21] for high energy nuclear reactions. The cosmic-ray work already cited in which charged particles of high energy are expelled from nuclei may be concerned with the same process, although neither the nature of the exciting particle nor cross sections are known which would be necessary to make comparisons.

We wish to thank Mr. James T. Vale and the other members of the 184-inch cyclotron operating group for making the irradiations.

[21] R. Serber, Phys. Rev. **72**, 1114 (1947).