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Artificially Radioactive Element 85

D. R. CORSON, K. R. MACKENZIE AND E. SEGRÈ
Radiation Laboratory, Department of Physics, University of California, Berkeley, California

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Bismuth bombarded with 32-Mev alpha-particles becomes radioactive. Two ranges of alpha-particles are emitted, one of 6.55 cm and one of 4.52 cm. These two alpha-particles are not genetically related. There are also x-rays which show the absorption characteristics of polonium x-rays. All these radiations separate together chemically as element 85, and all show the same half-life of 7.5 hours. The probable explanation of these effects is the following: Bi^{209}, by an (α, $2n$) reaction, goes to 85^{211}, which decays either by K-electron capture to actinium C′(Po^{211}) or by alpha-particle emission (range 4.5 cm) to Bi^{207}. The 6.5-cm alpha-particles are those of actinium C′. According to this scheme the second branch from 85^{211} leads to Bi^{207} which should decay to Pb^{207}. As yet we have been unable to find this activity. We discuss the chemical properties of element 85 and show that in general its behavior is that of a metal.

INTRODUCTION

THE sixty-inch cyclotron[1] of the Crocker Radiation Laboratory produces a beam of several microamperes of 32-Mev alpha-particles. We have used this beam to bombard a number of heavy elements. Lead and bismuth yield radioactive products which emit alpha-particles.[2, 3] In the present paper we report the results of our experiments on bismuth. We chose to investigate this activity, first because it could be due to element 85, and because bismuth has a single stable isotope (209), thereby simplifying the identification of the radioactive products.

The bombarded bismuth shows a number of different activities: (1) an alpha-particle group of 6.55 cm range, (2) an alpha-particle group of 4.52 cm range, (3) a gamma-ray with an energy of about 0.5 Mev, (4) an x-ray or soft gamma-ray with an energy of about 80 kev, (5) a soft x-ray, and (6) a few low energy electrons. All these radiations exhibit the same half-life of 7.5 hours.[4]

GENERAL DISCUSSION

The fact that all the activities show the same half-life suggests either (1) that they are genetically related, with the short periods in equilibrium with a long one (7.5 hours), or (2) that there are branching disintegrations, or (3) that there is a combination of chains and branchings. We first investigated the possibility of a genetic relationship between the two alpha-particle groups. In the first place, there is no growth of either group—after a 30-second bombardment both groups are present in their full intensity. According to the Geiger-Nuttall rule a 6.5-cm alpha-particle must have a half-life of the order of 10^{-3} sec., so that the most reasonable assumption is that the short range alpha-particle precedes the long range one. Hence we attempted to count coincidences, with various resolving times, between the long range and short range alpha-particles. No coincidences were found. For the same purpose a photographic emulsion (Ilford, R 1) was impregnated with a solution containing the activities. Upon development the alpha-particle tracks occurred singly in the emulsion. Any sort of genetic relationship between the two alpha-particles, even with an intermediate beta-process, would lead to two tracks originating at the same point. As a third test, we arranged an evacuated, right-angled tube in such a manner that nuclei recoiling from one alpha-disintegration were collected in front of a counter. Only alpha-particles from a second disintegration could be detected by this counter. No counts were recorded.

One might think that the two alpha-processes are alternative ways of disintegration of the

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[1] E. O. Lawrence *et al.* Phys. Rev. **56**, 124 (1939).

[2] D. R. Corson and K. R. MacKenzie, Phys. Rev. **57**, 250 (1940).

[3] D. R. Corson, K. R. MacKenzie and E. Segrè, Phys. Rev. **57**, 459 (1940).

[4] In addition to these activities there are energetic positrons whose maximum energy of about three Mev and 9.4-hour half-life suggest that they are due to Ga^{66} contamination produced by the bombardment of Cu^{63} with alpha-particles. This is confirmed by chemical evidence.

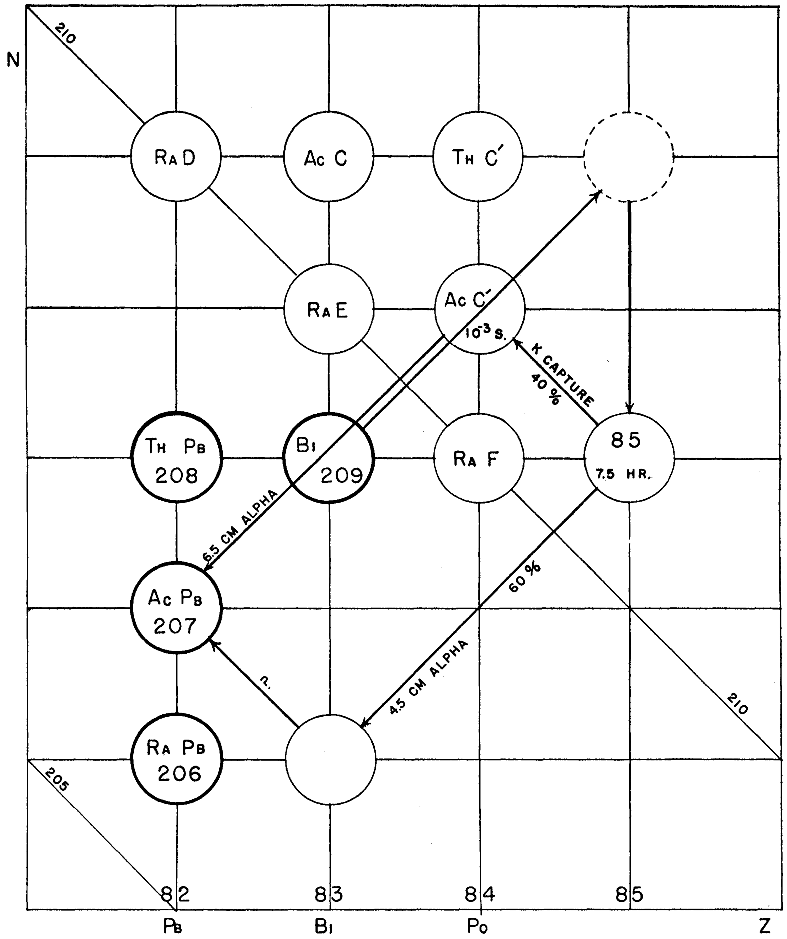

FIG. 1. Observed transmutations in a proton-neutron diagram. Bi^{209}, by an $(\alpha, 2n)$ reaction, goes to 85^{211}. This in turn disintegrates by alpha-particle emission (4.5 cm range) to Bi^{207} or by K-electron capture to Po^{211} (actinium C′). Actinium C′ emits alpha-particles (6.5 cm range) to go to Pb^{207}. The disintegration of Bi^{207} into Pb^{207} has not yet been observed. Stable nuclei are indicated by heavy circles.

same isotope. This would be in conflict with the Geiger-Nuttall rule because, as stated above, the nucleus in question should have a half-life of the order of 10^{-3} sec. instead of the observed 7.5 hours. Moreover, careful measurements show that 60 percent of the alpha-particles are in the long range group and 40 percent in the short. This is additional evidence against the direct genetic relationship between the alpha-particles, and also against the branching from the same nucleus, because in such a case the Geiger-Nuttall rule requires a branching ratio given by $\lambda_{4.5}/\lambda_{6.5} = 10^{-3}$ sec./7.5 hr. $= 3.7\times 10^{-7}$, instead of the experimental value of 0.67.

On the other hand, the fact that both groups have the same half-life strongly suggests that both groups originate from the same nucleus through some intermediate process, e.g., the nucleus with the 7.5-hour half-life could disintegrate either by an alpha (4.5 cm) or by a beta-process, the long range alpha-particle following immediately the beta-process. The ratio of short range to long range alpha-particles would then give the branching ratio of the alpha-beta process.

Since the radioactive nucleus must lie below the "stability" line, the beta-process would be expected to be a positron emission or a K-electron capture. However, for such a heavy nucleus and for a disintegration energy compatible with the observed half-life, the probability of K-electron capture should be much larger than the probability of positron emission.[5] In fact, no positrons with a 7.5-hour period are present whereas 7.5-hour x-rays are found. This radiation consists in large part, at least, of polonium x-rays. This

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[5] C. Møller, Phys. Rev. **51**, 84 (1937).

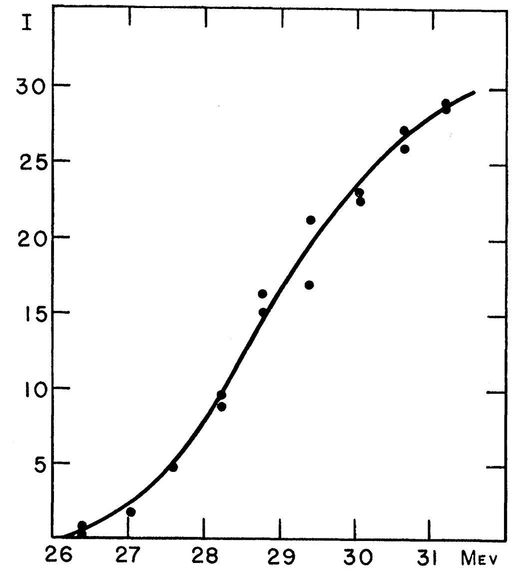

FIG. 2. Excitation curve.

was shown by critical absorption measurements. Since in a disintegration any x-rays follow the nuclear process, the product of the disintegration responsible for the x-rays must be polonium. We have shown that the parent substance is neither polonium nor bismuth by separating the unknown activity from these substances by evaporation from a bismuth target which had been bombarded with both alpha-particles and deuterons. The alpha-particles produce the activity in question, while the deuterons produce radium F (polonium). By heating filings from this target to the melting point in a vacuum the unknown activity was collected in an invisible layer on a cold glass plate, while the polonium remained behind. From this evidence we conclude that the disintegrating substance is element 85 going to polonium by a *K*-electron capture.

The high energy alpha-particle has a range (6.55 ± 0.04 cm), which is very close to the known range[6] (6.57 cm) of actinium C′, an isotope (211) of polonium. This strongly suggests that the primary product of the disintegration is 85^{211} formed by an (α, $2n$) reaction. 85^{211} then decays with a half-life of 7.5 hours by alpha-particle (4.5 cm) emission to Bi^{207}, or by *K*-electron capture to Po^{211} (actinium C′). Actinium C′ immediately disintegrates by the emission of an alpha-particle (6.5 cm), going to stable Pb^{207}. According to this scheme (see Fig. 1), there must be one *K* x-ray for each long range alpha-particle. In a thin-walled Geiger counter, about 500 times as many long-range alpha-particles as *K* x-rays were counted. Considering the efficiency of the counter for x-rays, this is consistent with the hypothesis of equal numbers of alpha-particles and x-rays.

The Bi^{207} formed by the first branch should be unstable and decay to Pb^{207}, but we have not yet found this activity. A short period for it seems to be ruled out because (1) Pb^{207} would be in equilibrium with 85^{211} and the x-rays or positrons would be in evidence, (2) we have separated element 85 from bismuth in ten seconds, and have found no activity in the bismuth, and (3) there is no growth of activity in element 85 after the separation. On the other hand, a period of a month or less would have shown up as a tail on our decay curves, but we have followed the x-rays over an intensity ratio of 500 with no clear evidence for any period other than the 7.5-hour one. This difficulty is made even more outstanding by considering the energy balance in the branching process. Bi^{207} must have an energy excess of at least one Mev over Pb^{207}, which should lead to an observable half-life, unless the transition is a forbidden one.

The question arises why only the product of the (α, $2n$) reaction should appear. The competing reactions (α, p), (α, $2p$), (α, np) lead to thorium C′, actinium C and actinium C′, respectively, none of which would be observable in our experiments. An (α, n) reaction leads to 85^{212}, but it can be

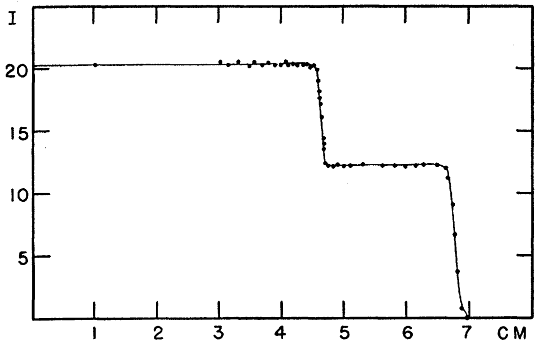

FIG. 3. Range curve of alpha-particles from a thin 85 source (measured at 25°C and 757 mm pressure).

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[6] Lord Rutherford, C. E. Wynn-Williams and W. B. Lewis, Proc. Roy. Soc. **133**, 351 (1931).

shown that this would be very improbable compared to the (α, $2n$) reaction.[7]

We have made an excitation curve by bombarding a stack of aluminum foils on which thin layers of bismuth had been evaporated. The foils had a nominal thickness of 0.0005 inch. The measured air equivalent thickness for alpha-particles of 4.5 Mev was 2.30 cm. For energies of 25 to 30 Mev we have taken the air equivalent to be 2.40 cm.[8] The thickness of the bismuth was determined by evaporating a similar film on Cellophane and determining the reduction in intensity of a light beam in passing through it. The air equivalent measured in this way was of the order of 0.1 mm, and has been neglected. The bismuth layers were assumed to be of uniform thickness since they were adjacent eighth-inch squares cut from a single sheet on which the bismuth had been evaporated. We have converted range to energy, using the data of Livingston and Bethe.[8]

The excitation curve determined in this way is shown in Fig. 2. There are two uncertainties in the energy scale, one caused by the inhomogeneity[9] of the cyclotron beam and the other by the fact that the average energy is determined only by the resonance conditions in the cyclotron. Dr. R. R. Wilson estimates that the inhomogeneity may amount to ten percent of the maximum energy. The uncertainty in the energy calculated from the resonance conditions exists because the ion source may not be at the geometrical center of the cyclotron.

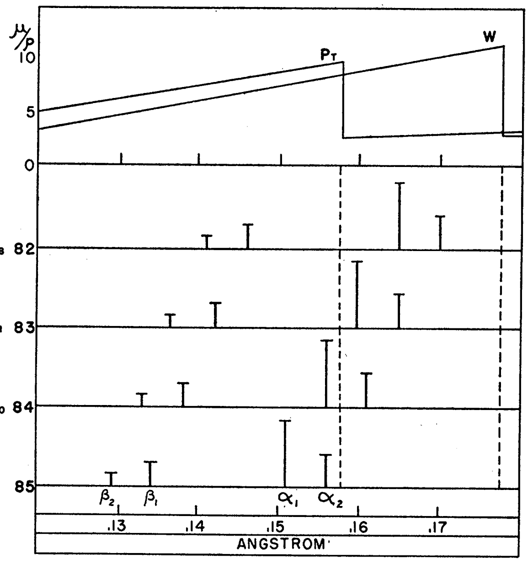

FIG. 4. The lower part of the diagram shows the K x-ray lines of lead, bismuth, polonium and 85. The length of the lines are proportional to their intensity. Dotted lines indicate the position of the absorption edges of platinum and tungsten. The upper part of the diagram shows the mass absorption coefficient (cm^2/g) of platinum and tungsten as a function of wave-length.

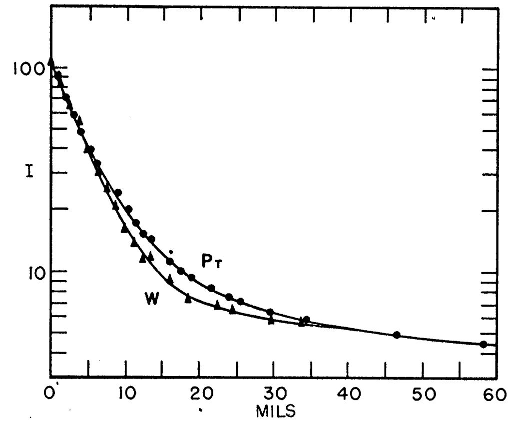

FIG. 5. Absorption curves of x-rays and gamma-rays in platinum and tungsten. Thickness of absorber measured in thousandths of an inch.

DETAILS OF MEASUREMENTS

All the sources used in the alpha-ray and x-ray measurements were prepared in a manner similar to that described by Rona and Schmidt for polonium.[10] The bombarded bismuth was placed in a glass tube with a constriction at one end and through which a stream of helium flowed. The walls of the tube were heated uniformly to the melting point of bismuth. The radioactive substance evolved from the bismuth was collected on a water-cooled plate placed in front of the constricted opening. In this way we obtained a source of about 20 microcuries (determined from the number of alpha-particles per second) on an area one cm in diameter. The bismuth residue contained the positron contamination previously mentioned.

The alpha-particles were counted with a shallow (one-mm) ionization chamber in con-

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[7] V. F. Weisskopf and D. H. Ewing, Phys. Rev. **57**, 472 (1940).

[8] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 245 (1937).

[9] R. R. Wilson, unpublished.

[10] E. Rona and E. A. W. Schmidt, Zeits. f. Physik **48**, 784 (1928).

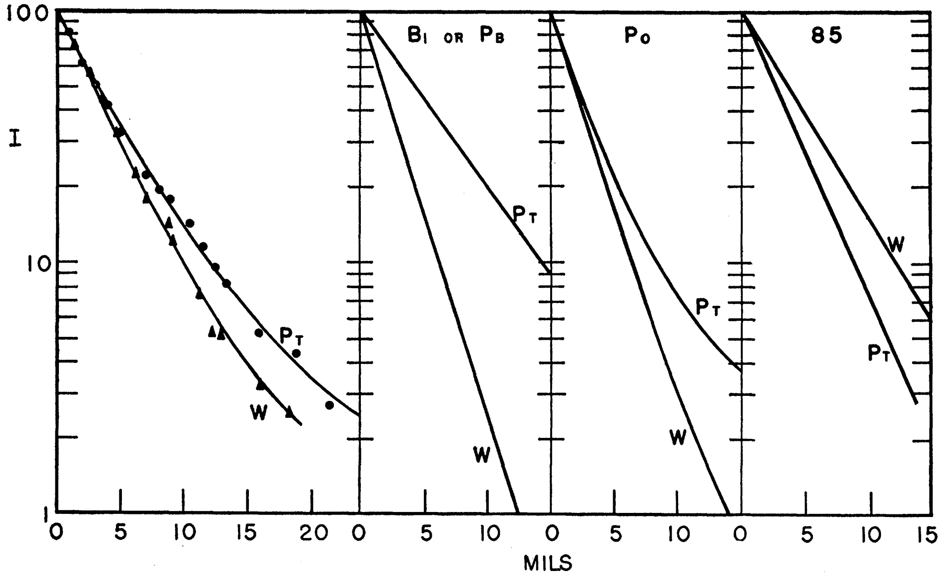

FIG. 6. Absorption curves of x-rays after subtracting gamma-ray. Also theoretical absorption curves of lead, bismuth, polonium and 85 x-rays in platinum and tungsten computed from the data of Fig. 4.

junction with a linear amplifier connected to a scale-of-eight recording circuit. To determine the range curve, the source and detector, together with collimation screens, were placed in a chamber in which the pressure could be varied. Then with a fixed source-to-detector distance, the counting rate was determined as a function of air pressure. As the pressure was changed, the depth of the ionization chamber was also changed to maintain a constant effective depth. To cover the complete range it was necessary to use three different source-to-detector distances. Thus, the final curve is the result of these three sets of data. Figure 3 shows the composite range curve, for 757 mm of mercury pressure and 25°C, with the abscissae being distances from the source to the front of the ionization chamber. The mean ranges so measured are 6.76 cm and 4.65 cm. When reduced to 760 mm of mercury and 15°C these become 6.50 cm and 4.47 cm. A further correction must be added to these values for the effective depth of the ionization chamber.[11] To determine the approximate magnitude of this correction we placed the ionization chamber at a distance from the source equal to the mean range of the high energy group and then decreased the depth of the chamber until the counting rate began to decrease. At this point the chamber depth was 0.5 mm, which we take as the effective depth to be added to the range values. This gives final values of 6.55 ± 0.04 cm and 4.52 ± 0.03 cm. The corresponding energies are 7.46 and 5.94 Mev, respectively.

The x-ray wave-lengths could be measured only by absorption methods because of the low intensities available. It is difficult to identify characteristic K radiations of heavy elements by critical absorption methods because of the relatively large differences in the wave-lengths of the various K lines of a given element. This is evident from Fig. 4, in which the wave-lengths of these lines are shown for the elements near bismuth. The x-ray wave-lengths for polonium and element 85 are determined by interpolation. We assume an intensity distribution of 50 : 25 : 17 : 8 for $K\alpha_1 : K\alpha_2 : K\beta_1 : K\beta_2$, corresponding to the measured values for gold.[12] The absorption edges of platinum and tungsten are indicated by the dotted vertical lines. All wave-lengths to the left of one of these lines are critically absorbed. The absorption coefficient of platinum jumps from about 2.5 cm^2/g to about 9.5 cm^2/g in crossing the critical wave-length.

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[11] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 (1938).

[12] F. K. Richtmyer, Rev. Mod. Phys. **9**, 391 (1937).

Thus, for the *K* x-rays of element 85, all four components will be highly absorbed in platinum, while in the case of polonium x-rays only three components, representing 75 percent of the intensity will be highly absorbed in platinum. In the case of bismuth and lead x-rays only two components, representing 25 percent of the intensity, will be critically absorbed in platinum. Our method has been to calculate theoretical absorption curves for the *K* x-rays of lead, bismuth, polonium and 85, in platinum and tungsten, and then to compare these curves with the experimental ones. Figure 5 shows the experimental curves, and Fig. 6 shows the same curves with the gamma-ray subtracted, together with the various theoretical curves. The shapes of the experimental curves are in good agreement with the theoretical curves for polonium *K* x-rays. The absolute value of the absorption coefficient does not agree very well, but this depends to a large degree on the geometry of the experimental arrangement. The absorption coefficient approaches the theoretical value as the geometry is improved. The final measurements were made with the best geometry consistent with the available x-ray intensity. The arrangement is shown in Fig. 7. By replacing the aluminum filter with paraffin one can observe some soft x-rays whose absorption coefficient in aluminum is consistent with that of *L* x-rays of polonium.

Chemical Properties

Element 85 is the upper homolog of iodine, and it could be expected to have the properties of a halogen. On the other hand, in the last rows of the periodic system resemblance between elements with atomic number differing by one unit is quite marked and often more evident than the resemblance with the lower homologs. The neighbors of 85 are bismuth and polonium to the left, iodine as its lower homolog, and radon to the right. Element 85 seems to have a closer resemblance to polonium than to iodine and generally speaking has quite marked metallic properties.

For the following experiments we filed the surface of the bismuth target and dissolved the filings in concentrated nitric acid, diluting the solution afterwards to 0.25 normal. The analytic behavior of element 85 with common reagents is as follows: (1) Hydrochloric acid does not precipitate it using lead or thallium as a carrier. This reaction is suitable for separating 85 from lead or thallium. (2) Hydrogen sulfide: In acid solution up to six normal hydrochloric acid, 85 is precipitated quantitatively using various carriers (bismuth, mercury, silver and antimony). A fractional precipitation with bismuth as a carrier obtained by decreasing the acidity from six normal hydrochloric acid shows a concentration in the first fractions. 85 does not precipitate quantitatively in ammonium sulfide solution using silver or mercury as a carrier. On the other hand the extraction from a mixed precipitate of mercury and antimony sulfides, by ammonium sulfide, even with prolonged digestion, is incomplete. (3) Ammonia or fixed alkali precipitate 85 with various carriers (mercury, iron, etc.). Probably these precipitations are due to absorption since often they are not quantitative. (4) Fractional hydrolysis of bismuth nitrate by dilution with water enriches 85 in the first fractions. (5) Reducing agents such as sulfur dioxide, zinc, or stannous chloride in hydrochloric or sulfuric acid solution precipitate 85 quantitatively. Precipitation with sulfur dioxide in three normal hydrochloric acid with tellurium as a carrier affords a method for separating 85 quantitatively from polonium, which stays in

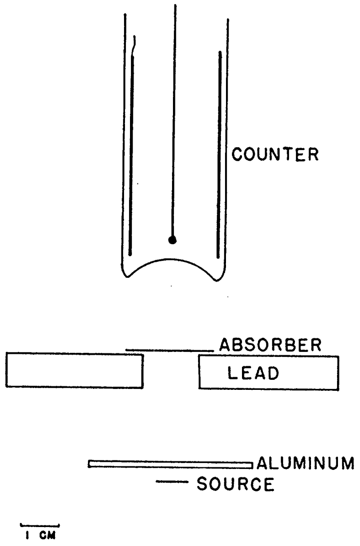

Fig. 7. Experimental arrangement for measuring absorption coefficient of x-rays.

solution.[13] Sodium stannite does not precipitate 85 and this reaction affords a good method for separating it from bismuth, tellurium, selenium, mercury, etc. (6) Silver nitrate does not precipitate 85 from a slightly nitric acid solution using potassium iodide as a carrier. Sometimes it is possible to collect a small fraction of the activity in this precipitation, but a further purification proposed by Buch-Andersen[14] in which the silver iodide is dissolved with sulfuric acid in the presence of zinc powder, the solution filtered and silver iodide reprecipitated from the filtrate leads to a completely inactive iodide, whereas the activity is found in the zinc. This behavior is interesting because it is definitely very different from the behavior of other halogens. It shows also that the methods used by Buch-Andersen in the search for element 85 in the natural radioactive families would not have led to an enrichment of 85 in the fractions studied by him. Hence, his negative experiments are not conclusive and 85 might exist in nature. In this connection mention should be made of some observations by Minder[15] which he interprets as proof for the existence of an isotope of 85 of mass 218 produced by beta-decay of radium A. Hulubei and Cauchois[16] have also reported some experiments which they interpret as evidence for the existence of element 85 in the decay products of radon. (7) Some experiments were made in order to collect element 85 by distillation from a slightly nitric acid solution. Potassium iodide was added as a carrier, the distilled iodine was collected in a sulfite solution, and the 85 precipitated with hydrogen sulfide. Under our operating conditions the recovery of iodine is practically complete, but that of 85 was very poor and irregular. Under the same conditions polonium does not distill at all. By adding concentrated sulfuric acid as well as potassium iodide to the solution, the yield of the distillation was increased to about 50 percent. In these experiments the presence of the carrier seems to be essential and may possibly indicate a chemical action of the carrier on 85. (8) Extraction with carbon tetrachloride with iodine as a carrier also gave a poor, but definite yield. (9) 85 is deposited on a copper plate from a 0.25 normal nitric acid solution containing bismuth and mercury. By heating cautiously the mercury which is also deposited on the plate disappears and the 85 stays behind. Some 85 can also be collected on bismuth by immersing a bismuth plate in a 0.25 normal nitric acid solution containing 85 and bismuth nitrate. (10) There are some indications for the formation of a hydride by the method of Paneth.[17] (11) The very characteristic volatility and its application to the separation from bismuth has been mentioned before. (12) Drs. J. G. Hamilton and M. H. Soley[18] have shown that 85 is concentrated in the thyroids of normal and thyrotoxic guinea pigs and excreted in a manner similar to iodine, thus establishing the similarity of physiological properties of these two halogens.

The chemical properties of 85 as outlined above are characteristic enough to rule out the rather remote chance that the activity be due to a fission product. As a matter of fact, by one or more of the reactions listed above it is possible to separate it from any other element. For the cases in which there could have been any suspicion, a direct separation has been effectively carried out; e.g., we have separated 85 from polonium, bismuth, thallium, mercury, selenium, tellurium, iodine, copper and silver.

In the conventional systematic analysis, 85 would be found in the hydrogen sulfide group. The separation from the naturally radioactive elements should not offer very serious difficulties. It must be borne in mind that the above results have been obtained with unweighable amounts of substance, and in applying them to macroscopic amounts the rules of radiochemistry must be taken into account.

In conclusion, we wish to thank Professor E. O. Lawrence for his continued interest in this work. We are indebted to the Rockefeller Foundation and to the Research Corporation for financial support.

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[13] M. Haissinsky, J. de Chemie Phys. **34**, 94 (1937).

[14] E. Buch-Andersen, K. Danske Vidensk. Selskab **16**, 5 (1938).

[15] W. Minder, Helv. Phys. Acta **13**, 144 (1940).

[16] H. Hulubei and Y. Cauchois, Comptes rendus **209**, 39 (1939).

[17] F. Paneth, *Radioelements as Indicators* (McGraw-Hill, 1928), p. 80.

[18] J. G. Hamilton and M. H. Soley, Proc. Nat. Acad. Sci. **26**, 483 (1940).