Radioactive Isotopes of Barium from Cesium

J. M. CORK AND GAIL P. SMITH
Department of Physics, University of Michigan, Ann Arbor, Michigan
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Radioactive isotopes of barium of half-lives 40.0 hours and 340 hours are made by bombarding cesium with deuterons of 9.5 Mev energy. The former is shown by absorption measurements and by the magnetic beta-spectrometer to consist of a partially converted gamma-ray of energy 276.4 kev. The latter activity is associated with K-electron capture in Ba^{133} and consists solely of a 17-kev gamma-ray and the K x-radiation of cesium of energy about 30 kev. The 40.0-hour activity is probably due also to a highly excited state of Ba^{133}, although the existence of an excited state of the stable Ba^{134} would equally well satisfy the data. The probability of total conversion for the 40.0-hour gamma-ray is about 71 percent, and the ratio of the K and L components is found to be 3.18. From the calculations of Hebb and Nelson this indicates a change in angular momentum of four units.

CESIUM bombarded with deuterons of 9.5-Mev energy, gives rise to strong radioactivity in barium. Since cesium has but a single isotope of mass 133 only three possibilities exist for the masses of the barium atoms formed. The most likely process is the (D, N) reaction resulting in barium (134). The less probable $(D, 2\ N)$ transformation leads to barium (133) while the least probable (D, γ) interaction would result in barium (135). Since both barium isotopes of mass 134 and 135 exist in nature any radioactivity due to them must be associated with the existence of excited states of the stable element.

Kalbfell and Cooley[1] bombarded barium with neutrons and reported a radioactivity of 30 hours half-life consisting of a characteristic gamma-ray of energy about 250 kev but too weak to be observed in their spectrometer. This activity whose half-life is shown to be 40.0 ± 0.5 hr. is made very strong by the bombardment of cesium with deuterons and a subsequent separation of barium. In addition to absorption measurements by an ionization chamber and string electrometer, it has been studied in a magnetic spectrometer[2] of high resolution. A preliminary report[3] of the observed results has already been given. In addition to the activity of half-life 40.0 hr. another radioactivity of half-life 340 ± 10 hr. is found in the barium precipitate.

[1] D. C. Kalbfell and R. A. Cooley, Phys. Rev. **58**, 91 (1940).
[2] For description see Phys. Rev. **57**, 6 (1940).
[3] H. W. Collar, J. M. Cork, and G. P. Smith, Phys. Rev. **59**, 937A (1941).

The radioactivity of shorter half-life is shown to be due to a gamma-ray of energy 276.4 kev. This gamma-ray on leaving the nucleus has a large probability of ejecting electrons from the outer atomic levels of the same barium atom. There thus appear electrons of discrete energy values together with the unconverted gamma-radiation. This electron distribution as observed in the beta-spectrometer for two slit widths is shown in Fig. 1. The intensities have been normalized to the same ordinate, and in order not to overlap, the zero ordinate for the curve with the 3-mm slit is shifted upward. The small upper figure is an enlarged view of the M component. These observations have been corrected for background, decay of the source, efficiency of the counter, absorption in the window of the counter, and losses due to counting rate. The peaks correspond to conversion of K, L, and M orbital electrons by the gamma-rays from the active nuclei. In terms of energies these values are 239.0, 270.5, and 275.0 kev, respectively.

These data when compared with the known binding energies for electrons from x-ray spectroscopy, enable a choice to be made regarding the atomic number of the element which emits the gamma-ray. Thus if the radioactive barium should emit a charged particle as a positron and immediately a gamma-ray then the conversion and the K–L difference would be characteristic of cesium; whereas if the half-life is attributed to the gamma-emission the conversion and the K–L energy difference would be characteristic of barium. This latter condition is found to be the case since the observed K–L difference is 31.5 kev and the corresponding differences in K–L binding energies are 30.2 for cesium and 31.4 for barium. Moreover, no general electron or positron radiation of half-life 40.0 hr. was observed. From this evidence the active body could be either Ba^{133} or an excited state of Ba^{134}.

A gamma-radiation can be characterized by such a long half-life only when a large change in spin accompanies the emission. By observing quantitatively the conversion coefficients an estimate can be made of this spin change. An approximate value of the total conversion factor can be obtained by means of an ionization chamber if the relative sensitivity of the chamber to electrons and to photons is known.

To do this it is possible to make use of an absolute calibration[4] already made in the study of the radioactivity of indium. Here the partially converted 4.5-hr. indium activity is in equilibrium with its parent 56-hr. cadium electron activity so that the areas corresponding to the number of electrons could be integrated and the ratio determined. To the same accuracy that the total conversion in the 4.5-hr. indium was found to be 49 ± 10 percent, the total conversion coefficient for the 40.0-hr. barium activity is found to be 71 percent.

The ratio of the conversion coefficients α_K, α_L, and α_M for the various shells can be determined with considerable accuracy by integration of the separate peaks. From the data for a 3-mm slit system the ratio $\alpha_K/(\alpha_L+\alpha_M)$ was found to be 2.76 and for the 1-mm slit system, 2.80. From the shape of the lines obtained with best resolution an estimate can be made of the M conversion alone. Such an estimate is $\alpha_L/\alpha_M=7$, from which it follows that $\alpha_K/\alpha_L=3.18$. Hebb and Nelson[5] have calculated the probability of conversion of a gamma-ray in the L shell and have indicated the dependence of the K, L ratio of conversion upon the charge of the nucleus, the angular momentum and the energy of the gamma-ray. For a gamma-ray of energy 276.4 kev in barium the ratio $\alpha_K/\alpha_L=3.18$, indicates very closely a change in angular momentum of four units.

The 40.0-hr. activity was also made by neutron excitation. It was found that the radioactivity induced by 9.5-Mev deuterons on Be in a sample placed at right angles to the direction of the beam was almost as strong as in the sample placed directly behind the Be target. Samples in paraffin however were not noticeably activated. It thus appears that the threshold excitation is not exceedingly high. If the activity were due to an excited state of Ba^{134} there might exist the possibility of activation by exposing barium to photon radiation of sufficiently high energy. A quantity of barium was given an enormous exposure[6] to 500-kv x-rays. No trace of radioactivity was observed.

From the data obtained it is evident that this activity could be ascribed equally well to either Ba^{133} or Ba^{134}. However it is known[7] that the Rochester group have found this same activity by bombarding cesium with protons. In their bombardment it is possible to produce barium by the (P, N) and (P, γ) reactions. Although the latter process is known to occur, the former is much more probable and favors the assignment to Ba^{133}.

The activity in barium whose half-life is 340 hr. is shown to consist of two low energy gamma-

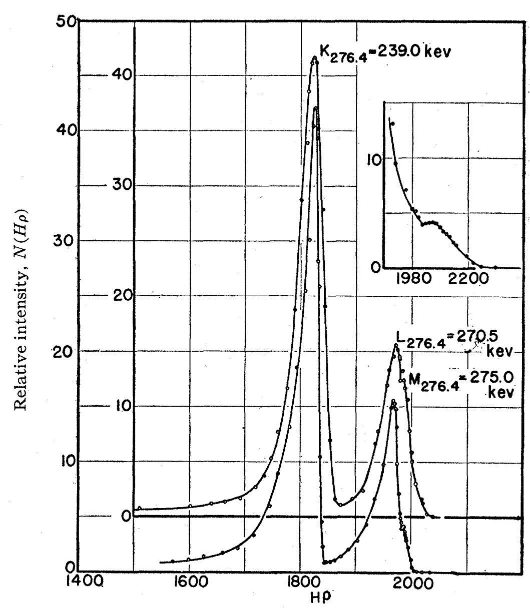

FIG. 1. Distribution of electrons due to internal conversion in 40-hr. barium activity. Upper curve, 3-mm slit; lower curve, 1-mm slit. Intensities are normalized and curves shifted to avoid overlap.

[4] J. L. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940).
[5] M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).

[6] This exposure was very kindly made by Dr. K. Corrigan with the high voltage x-ray tube of Harper Hospital.
[7] Private communication.

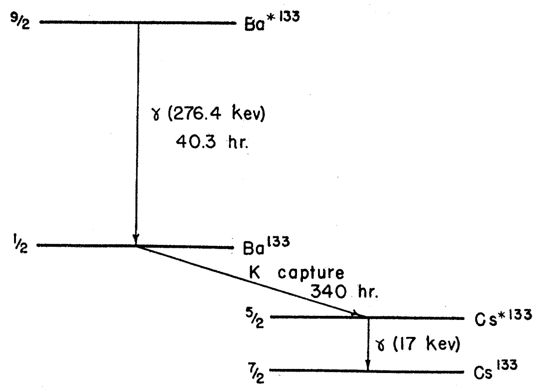

FIG. 2. Energy level scheme to account for observed radioactivities in barium.

rays. By absorption in aluminum the energies of the gamma-radiations appear to be 17 kev and 30 kev. Isotopes of barium could decay to stable cesium only by the emission of positrons, or an equivalent process, namely the capture by the nucleus of the atom of one of its own K electrons. No positron emission appeared to accompany the gamma-radiation, hence the radioactive decay was undoubtedly associated with the K-capture process. This would suggest that the 30-kev radiation is the characteristic K radiation of the atom after the capture of the K electron and hence in this case would be the K radiation of cesium. To establish this fact, absorbing films having about 21 mg/cm^2 were prepared of the following elements; iodine, antimony, tellurium, and tin. If the x-rays are characteristic of cesium they should be weakly absorbed by iodine and tellurium and strongly absorbed in antimony and tin. If originating in barium the x-rays should be weakly absorbed in iodine and strongly absorbed in tellurium, antimony and tin. The percentage of the incident beam absorbed by the foils is shown in Table I. It is thus quite certain that the radiation is characteristic of cesium following K-electron capture by the barium nucleus. The existence of the 17-kev gamma-ray indicates that the nucleus after K capture is left in an excited state. This probably has a short half-life and is in equilibrium with the component of long life. It thus appears reasonable to construct an energy level scheme as shown in Fig. 2. The values assigned to the spins are not unique but are sufficient to allow transitions leading to the observed results.

In studying the products formed by the fission of thorium and uranium an activity in barium of half-life 300 hr. has been reported.[8] This activity, however, has been identified as linked in a chain reaction with a cesium activity of 33-min. half-life, which in turn is derived from a xenon activity of very short half-life. It has therefore been assigned to an isotope of barium of mass 139 or greater and hence is not the activity observed in this investigation.

TABLE I. *Absorption of x-rays.*

Absorbers	Percent Absorbed
Iodine	8 ± 4
Tellurium	7 ± 4
Antimony	26 ± 4
Tin	32 ± 5

The chemical separations have been made by Mr. T. Berlin. For discussions of the theory we are especially indebted to Professor G. E. Uhlenbeck. This investigation was made possible by a grant from the Horace H. Rackham Fund.

[8] Heyn, Aten and Bakker, Nature **143**, 516 (1939); Hahn and Strassmann, Naturwiss. **27**, 529 (1939).