New Barium and Cesium Isotopes: 12.0d Ba^{131}, 10.2d Cs^{131}, and Long-Lived Ba^{133}*

SEYMOUR KATCOFF**
Argonne National Laboratory, University of Chicago, Chicago, Illinois
(Received August 11, 1947)

A new Ba-Cs chain of probable mass assignment of 131 has been formed by neutron activation of barium. The Ba^{131} isotope decays predominately by orbital electron capture with a half-life of 12.0 days, emitting gamma-radiations of about 0.26 Mev, 0.5 Mev, and roughly 1.2 Mev (the latter in low intensity). Detected also were 31-kev x-rays and 0.42-Mev and 0.24-Mev conversion electrons. The Cs^{131} decays exclusively by K-electron capture with a half-life of 10.2 days, emitting no detected gamma-rays. The Ba^{133} has a minimum half-life of 20 years and is very probably the lower state to which the 37.8 hour Ba^{133} decays. The former decays by K-capture and emits 31-kev x-rays, 0.36-Mev gamma-rays, 0.34-Mev conversion electrons, and possibly 4.3-kev x-rays.

I. INTRODUCTION

THE stable Ba and Cs isotopes and their percent abundances are as given in Table I. Neutron bombardment of barium is known to give 85m Ba^{139} and 37.8h Ba^{133}. It appeared probable that if Ba^{131} has a half-life of the order of several days that it might be detected in some barium carbonate which had received an intense irradiation at the center of the Clinton pile. (This is the same $BaCO_3$ sample that was used to determine the neutron-absorption cross section[1] of 85m Ba^{139}.) A considerable amount of activity actually was found when a radio-chemical analysis for Ba was performed 21 days after the end of the bombardment. Some of this purified Ba was then allowed to stand four days, and a radiochemical analysis for Cs was made. Some activity was found, and this was assigned to a radioactive Cs daughter of the new Ba isotope. The long-lived Ba^{133} was discovered after the Ba^{131} had decayed to a negligible activity.

12.0d Ba^{131}

In order to prove definitely that the first activity was really that of an isotope of Ba, some fractional precipitation experiments were performed. Three aliquots of the irradiated $BaCO_3$ were taken and labeled Ba b, Ba c, and Ba d. Then Ba b was scavenged six times with $La(OH)_3$ precipitations and twice with $Fe(OH)_3$ precipitations. It was then precipitated two times as $BaCl_2$ and once as $BaCrO_4$. Sample Ba c was treated likewise except that two $Ba(NO_3)_2$ and one more $BaCrO_4$ precipitations were added. Sample Ba d was first subjected to seven $La(OH)_3$ and two $Fe(OH)_3$ precipitations. It was precipitated three times as $Ba(NO_3)_2$, once as $BaCrO_4$, and four times as $BaCl_2$. Then it was subjected to slow fractional precipitation as $BaCO_3$. It was brought down in four batches and labeled Ba d_1, Ba d_2, Ba d_3, and Ba d_4. Each of these was then reprecipitated as $BaCrO_4$. The results are given in Table II.

The specific activity was essentially the same for all six samples when measured through 213-mg Al/cm² or through 1.8-g Pb/cm². The deviations from constant specific activity for the measurements made without added absorber can be completely accounted for by the differences in self-absorption of the very weak radiations corresponding to the differences in the weights of the samples. Therefore, it is established that this is an active isotope of Ba.

Its half-life was determined by following the decay of a sample covered with 1.8-g/cm² of lead. This amount of lead absorbs completely the radiations of the Cs daughter of this Ba isotope. The

TABLE I. Stable isotopes of Ba and Cs.

Ba^{130}	Ba^{132}		Ba^{134}	Ba^{135}	Ba^{136}	Ba^{137}	Ba^{138}
0.101	0.097		2.42	6.59	7.81	11.32	71.66
		Cs^{133}					
		100					

* This work was originally reported in Manhattan Project Reports CC-3148 (June 1, 1945) and CC-3456 (March 11, 1946); see also Plutonium Project Record IX B, 12.1.1 and IX B, 12.2 (1946).

** Now at Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

[1] S. Katcoff, Manhattan Project Report CC-2908 (April 7, 1945); Plutonium Project Record IX B, 7.59.4 (1946).

half-life was found to be 12.0 days for a decay time of nine half-lives. Absorption measurements in lead and in aluminum were made on freshly purified Ba samples. The lead-absorption curves (Figs. 1 and 2) show a 0.5-Mev γ-ray, a small amount of a harder γ-ray (roughly 1.2 Mev), and one or more softer γ-rays (roughly 0.26 Mev and below). The aluminum-absorption curves (Fig. 3) indicate 31.5-kev x-rays, 0.42-Mev conversion electrons, and 0.24-Mev conversion electrons. The latter two energies correspond fairly well with the 0.5-Mev γ-rays and 0.26-Mev γ-rays, respectively. From these measurements (especially from the large proportion of 31.5-kev x-rays) it seems that most of the disintegrations take place by K-electron capture. Some positron emission might also occur, especially since 0.5-Mev γ-rays are observed, part of which might be annihilation radiation.

10.2d Cs^{131}

The Cs daughter of this Ba decayed exponentially with a half-life of 10.2 days for a period of eight half-lives. A few periodic extractions were made of the Cs from the Ba. These indicated a 12-day half-life for the Ba and definitely established the fact that the 10.2d Cs is a daughter of this 12.0d Ba. The absorption of the Cs radiations in aluminum is shown in Fig. 4. This indicates only a 31.0-kev x-ray and some extremely soft radiation. Absorption in copper shows only a 30.8 kev x-ray. A lead absorber 1.8 g/cm² thick reduced the count of a Cs sample, which gave 30,000 c/m without absorber, to zero. Therefore, no γ-rays are emitted by this isotope, unless their energy is less than a few kev.[2] The very soft

TABLE II. Results of precipitations.

Sample No.	Weight of sample	Activity per milligram through no added absorber	through 213-mg Al/cm²	through 1.8-g Pb/cm²
Ba *b*	14.3 mg	545 c/m	160 c/m	215 c/m
Ba *c*	21.7	491	159	220
Ba d_1	14.1	556	162	212
Ba d_2	14.7	536	161	211
Ba d_3	8.4	602	161	224
Ba d_4	22.6	485	157	215

[2] In a recent note, Phys. Rev. **71**, 382 (1947), Fu-Chun Yu, Donald Gideon, and J. D. Kurbatov reported the emission of highly converted 0.145-Mev gamma-rays by Cs^{131}. These were not observed in the experiments reported here. The other data given in their note are in general agreement with the data given here.

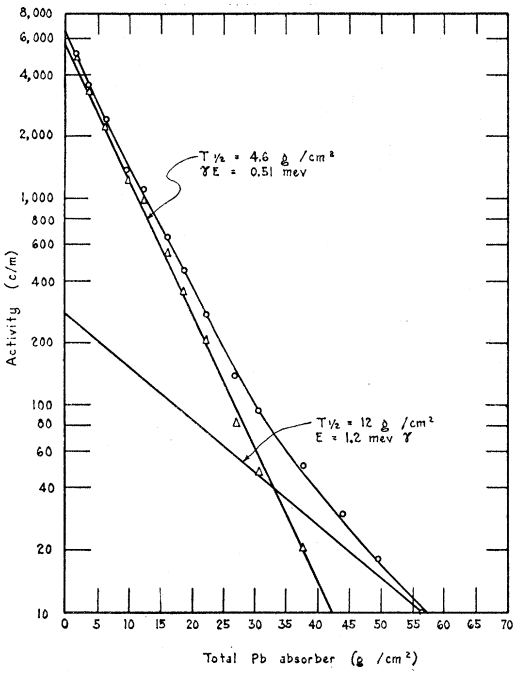

FIG. 1. Lead-absorption curve of Ba^{131}.

radiation shown by the first part of the absorption curve in aluminum is probably the L x-radiation. The other x-rays are very likely the K x-rays of Xe (29.6 kev) which follow the capture of the K electrons by this Cs isotope. The only isotope previously known which decays purely by orbital electron capture and does not emit γ-rays is 600d V^{49}.

II. MASS ASSIGNMENT TO CHAIN:

12.0d Ba→10.2d Cs

Mass numbers 129 and 131 are the only two possibilities which permit an active Ba isotope to be formed by neutron bombardment of ordinary barium and to have an active Cs daughter.[3] Mass number 129 can be eliminated by considering the neutron cross section of normal barium for the formation of this 12.0d Ba. This was found

[3] One other possibility is that the 10.2d Cs is an isomer of stable Cs^{133} which decays to the ground state by emission of 31-Kev γ-rays. This was made very improbable by the critical absorption measurements of B. Finkle, which indicated that the 10.2d Cs does decay to Xe. See B. Finkle, Phys. Rev. **72**, 1260 (1947).

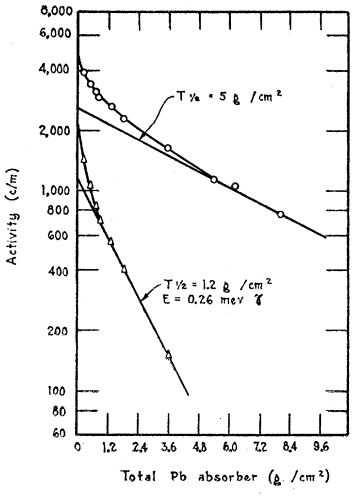

FIG. 2. Lead-absorption curve of Ba^{131}. Δ0.5-Mev and 1.2-Mev components subtracted from original points (cricles).

to be 0.006 barn. Ba^{129} would have to be formed by an $(n, 2n)$ reaction on stable Ba^{130}, whose isotopic abundance is 0.10 percent. The isotopic cross section would then be six barns. Such a high cross section for an $(n, 2n)$ reaction in a neutron flux, which is mostly near thermal, is extremely improbable. Therefore, it is fairly certain that the correct mass number of these isotopes is 131, and that Ba^{131} is formed from Ba^{130} by an (n, γ) process. The calculation of the cross section is based on the assumption that the decay of the 12.0d Ba takes place almost entirely by K-capture and that the γ-rays are internally converted to only a small extent. That is, one K x-ray was assumed for every disintegration. The counting efficiency of these x-rays was taken to be one percent (private communication from D. W. Engelkemeier).

III. LONG-LIVED Ba^{133}

Nine months after the neutron irradiation of the barium some residual activity was still present in a sample which had been extensively purified subsequent to the bombardment. After these nine months the sample was dismounted and purified further by making one $BaCO_3$ precipitation, two $BaCl_2$ precipitations, two $Fe(OH)_3$ scavenging precipitations, and one $BaCrO_4$ precipitation. The specific activity, as observed through 139 mg/cm² of aluminum absorber, was unchanged to within a possible experimental error of 10 percent. The observed activity must therefore be that of a long-lived isotope of barium.

The supernatant solution which remained from the $BaCO_3$ precipitation of this sample was radiochemically analyzed for cesium. Eight months had been allowed for a possible Cs daughter to grow in. Two hours after separation from the barium the cesium extract had an activity, though no added absorber, of about 2 c/m. Under similar conditions the parent barium sample had an activity of 2000 c/m; through 213 mg/cm² of aluminum absorber this activity was 360 c/m. Thus it seems that this long-lived Ba isotope has no active Cs daughter unless the latter is very short-lived, very long-lived, or has very soft radiations.

Over a period of 20 months the decay of this Ba activity could not be detected to within a possible error of 5–6 percent. This sets a lower limit of 20 years on the half-life. The radiations were investigated by means of absorption measurements. A lead-absorption curve (Fig. 5) indicates the presence of a 0.36-Mev gamma-ray. Aluminum-absorption curves (Fig. 6) show 0.34-

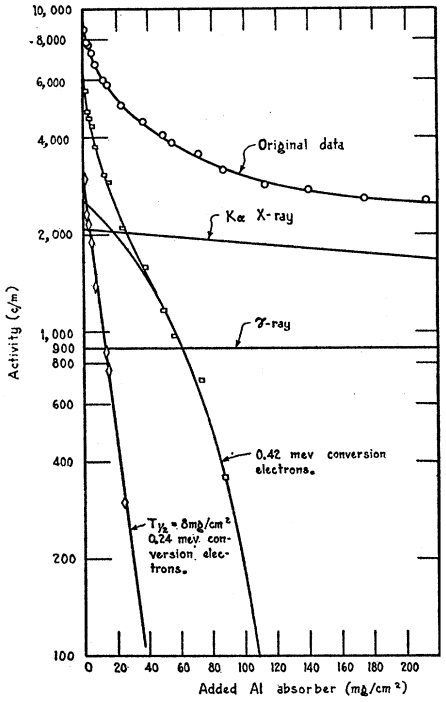

FIG. 3. Aluminum-absorption curves of Ba^{131}.

Mev conversion electrons, a 32-kev x-ray, and possibly a 4.6-kev x-ray. Critical absorption measurements were made with Sn, Sb, Te, and iodine absorbers in order to establish the origin of the harder x-rays. The absorbers were made by using solutions or suspensions of the elements in question in a fixed quantity of 6N HCl. These were placed in disk-shaped cells whose diameter was 7 cm and whose height was 2 cm. The bottom of each cell was made of mica. About 14 absorbers were used for each of the four elements. The x-rays showed half-thicknesses of 31 mg/cm^2 and 30 mg/cm^2 with Sn and Sb absorbers, respectively, whereas they showed half-thicknesses of 69 mg/cm^2 and 70 mg/cm^2 with Te and iodine absorbers, respectively. This clearly identifies the radiation as the K x-rays of Cs, the energy of which is 30.7 kev. Very few, if any, positrons are emitted because no 0.5-Mev annihilation γ-rays are observed. Therefore this long-lived Ba must decay by means of orbital electron capture. The soft 4.6-kev x-ray indicated by the aluminum-absorption curve (Fig. 6) is probably the L x-radiation of Cs, the energy of which is 4.3 kev.

The mass assignment can be made by a consideration of the cross section which was found for the formation of this isotope. By assuming that one 30.7-kev x-ray was formed for each dis-

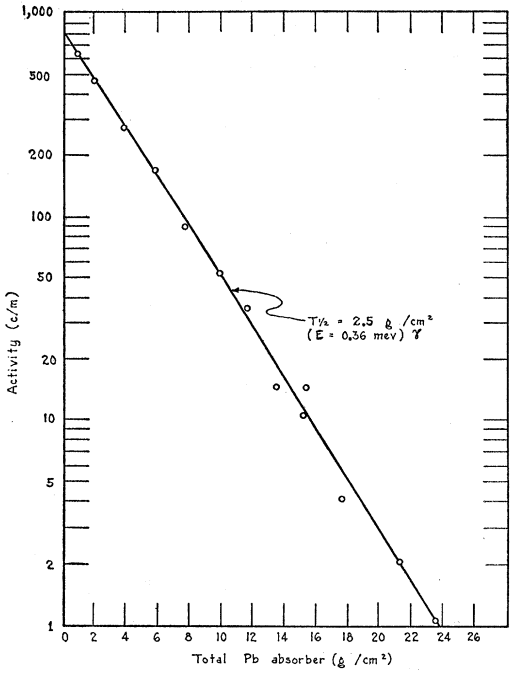

FIG. 5. Lead-absorption curve of >20y Ba^{133}.

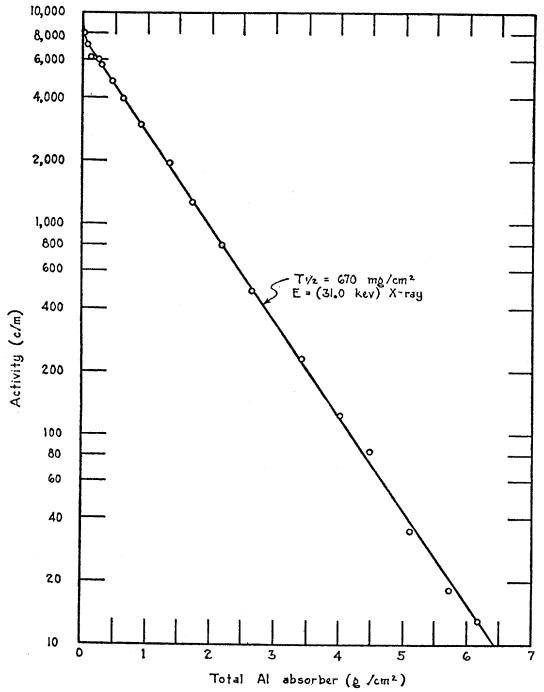

FIG. 4. Aluminum-absorption curve of Cs^{131}.

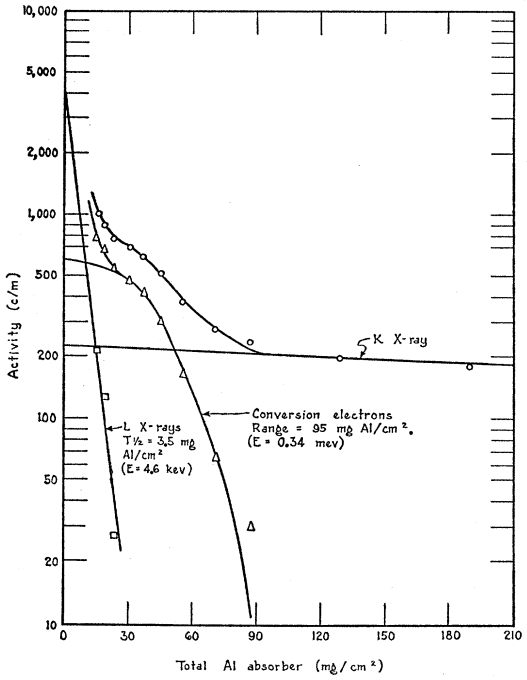

FIG. 6. Aluminum-absorption curve of >20y Ba^{133}.

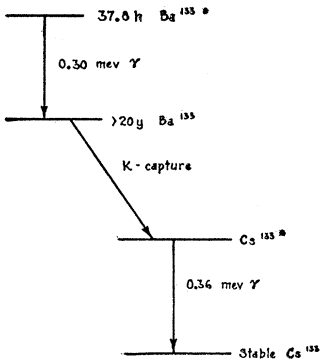

FIG. 7. Probable decay scheme of Ba^{133}.

integration and that the counting efficiency of these x-rays is one percent, this cross section for normal barium is at least 0.006 barn (corresponding to the minimum half-life of 20 years). If assignment is made to mass 129, then it could only have been formed from stable Ba^{130}, whose abundance is 0.10 percent, by an $(n, 2n)$ reaction. This would mean that the isotopic cross section for this process is at least 6 barns, which is extremely improbable since most of the pile neutrons are of low energy. Furthermore, Ba^{129} would decay to an active Cs^{129}, and no such activity could be detected. Assignment of the long-lived Ba to mass 131 is ruled out by the fact that it does not decay to the 10.2d Cs^{131}. The only possible remaining assignment is to mass 133. In this case the isotope could be formed from stable Ba^{132}, whose abundance is also 0.10 percent, by an (n, γ) reaction. The isotopic cross section for this process is then a minimum of about 6 barns. Thus an assignment to mass 133 is fairly certain. The 37.8h Ba which is also assigned to mass 133 is known to decay by isomeric transition. The lower state can now be identified as this >20y Ba^{133}. The decay scheme is probably as shown in Fig. 7.

This paper is based on work performed at the Metallurgical Laboratory of the University of Chicago under Contract W-7401-eng-37 for the Manhattan Project, and the information contained therein will appear in Division IV of the Manhattan Project Technical Series. Counting of the long-lived Ba^{133} is continuing at Los Alamos.

PHYSICAL REVIEW VOLUME 72, NUMBER 12 DECEMBER 15, 1947

Coincidence Measurements. Part I. Beta Spectra

M. L. WIEDENBECK AND K. Y. CHU
Department of Physics, University of Michigan, Ann Arbor, Michigan
(Received August 25, 1947)

The necessary and sufficient conditions for a simple β spectrum (not accompanied by internally converted γ radiation) are discussed. It is generally considered that the $\beta-\gamma$ coincidence rate per recorded β particle decreases with an increase of absorber thickness for a complex β spectrum. Although this is the most common condition, it is possible for the ratio to remain constant or even rise as the β absorber thickness is increased.

The ratio, $N_{\beta\gamma}/N_\beta$, is also calculated for simple spectra followed by non-delayed conversion electrons as well as for conversion in metastable levels of lifetime long compared with the resolving time of the coincidence circuit.

Data have been obtained for several nuclei including antimony^{124} (60 days), cesium^{134} (1.7 years), europium^{154} (5–8 years), gold^{198} (2.7 days), hafnium^{181} (55 days), iridium^{192,194} (60 days), mercury ^{203,205} (51.5 days), osmium^{193} (17 days), and potassium^{42} (12.4 hours).

1. INTRODUCTION

IN Part II of this set of articles we shall indicate a simple and accurate method for determining the total absolute conversion coefficients of nuclear levels. This method demands little knowledge of the γ radiation involved in the transition if the β spectrum is simple but a considerable amount of information must be available if the β spectrum is complex. Since little is known about the structure of most complex β emitters we have limited our investigation to nuclei which have simple β spectra.