Radioactivity of the Cerium-137 Isomers

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A cerium isotope with an 8.7-hr half-life has been assigned to Ce^{137} and shown to decay by electron capture with about 3% of the captures to a 445-kev level in La^{137}. The 34.5-hr decay period previously characterized as electron capture of Ce^{137} is an isomeric transition from a 255-kev level in Ce^{137}. Spin assignments consistent with the experimental data indicate that the transition between ground states of La^{137} and Ba^{137} should be allowed. However, the K or L electron capture half-life of La^{137} was found to be greater than 10^5 years.

I. INTRODUCTION

A RADIOACTIVE decay period of 36 hr was assigned to Ce^{137} by Chubbock and Perlman[1] who reported decay by electron capture through a 280 kev level in La^{137}. Stover[2] confirmed the work of Chubbock and Perlman but found the gamma energy to be 240 kev. In a search for an isomeric transition in Ce^{137} Hill[3] confirmed the mass assignment of the 36 hr period by the use of enriched isotopes and found the K/L conversion ratio of a 257-kev gamma ray to be about 4. Keller and Cork,[4] who also used enriched isotopes, confirmed the mass assignment and gave an energy of 253.4 kev for the gamma ray. They found a K/L conversion ratio of about 10. In the present work the K conversion coefficient as well as the K/L conversion ratio has been measured. This has led to a new assignment of the 36 hr decay period and to the discovery of a new radioactive species which has been assigned to Ce^{137}.

II. ISOMERIC TRANSITION IN CERIUM-137

A mixture of Ce^{137}, Ce^{139}, and Ce^{141} was produced by bombardment of a BaF_2 target with 48-Mev alpha particles in the Berkeley 60-in. cyclotron. The cerium fraction from the target was identified and purified with a cation exchange column.[5] The conversion electron spectrum of a source prepared from this material was measured with a thin magnetic lens spectrometer.

The intensity of the K conversion peak of the 255-kev gamma was measured relative to the K conversion peak of the 166-kev gamma of Ce^{139}. A NaI scintillation spectrometer was used to measure the relative intensities of the unconverted 255 kev and 166-kev gammas. From these intensity ratios and the published[6] K conversion coefficient (0.2 ± 0.05) of the 166-kev gamma, a K conversion coefficient of 5.5 ± 1.5 was found for the 255-kev gamma ray. This conversion coefficient and the K/L conversion ratio of 2.3 ± 0.2 indicate that the 255-kev gamma is emitted in an $M4$ transition.[7,8]

An $M4$ isomeric transition in Ce^{137} is expected from the systematics of energy level spacings.[9] X-ray gamma-ray coincidence measurements show that the 255-kev gamma is not coincident with capture x-rays and thus support the conclusion that it is emitted from an isomeric level. This conclusion is confirmed by the fact that the x-rays associated with the 34.5 hr period are cerium rather than lanthanum x-rays. This was shown by using a xenon filled proportional counter, linear amplifier and pulse analyzer. Both the $K\alpha_1$, and the $K\beta$ x-rays of lanthanum are critically absorbed. Escape of the fluorescent $K\alpha$ and $K\beta$ x-rays of xenon results in pulse height peaks at about 5 kev and 9 kev when either cerium or lanthanum x-rays are counted. However, the ratio of the intensity of the peak at 5 kev to the peak at 9 kev should be about 10 times greater for cerium than for lanthanum K x-rays. A comparison of the pulse-height spectra of Ce^{139} and Ce^{137} shows that the K x-rays emitted with a 34.5 hr half-life are those of Ce. The conversion electrons and unconverted gamma of the 255-kev transition also decay with a 34.5 ± 0.5 hr half-life.

Cerium samples enriched in Ce^{136} have been used to confirm the mass assignment of the 34.5-hr decay period. One sample was enriched to 30% Ce^{136} and another to 6% Ce^{136}. Both were bombarded for the same period at the same neutron flux in the Oak Ridge National Laboratory graphite reactor. The samples were measured with a scintillation spectrometer and the 255-kev gamma ray activities per microgram of each of the cerium isotopes in the samples are shown in Table I. These data show that the yields of the 34.5-hr isomer per microgram of Ce^{136} are the same in the two samples and therefore the isomeric transition is in Ce^{137}.

III. ELECTRON CAPTURE DECAY OF Ce^{137}

When a mixture of stable cerium isotopes is bombarded with slow neutrons, a 445-kev gamma ray and x-rays which decay with an 8.7-hr half-life are produced. Chemical separation of cerium from rare earth impuri-

[1] J. B. Chubbock and I. Perlman, Phys. Rev. **74**, 982 (1948).
[2] B. J. Stover, Phys. Rev. **81**, 8 (1951).
[3] R. D. Hill, Phys. Rev. **82**, 449 (1951).
[4] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951).
[5] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. **69**, 2800 (1947).
[6] C. H. Pruett and R. G. Wilkinson, Phys. Rev. **96**, 1340 (1954).
[7] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79, (1951).
[8] M. E. Rose (privately distributed L conversion coefficients).
[9] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

TABLE I. Neutron capture yields referred to isotopic assays.

	255 kev counts/min μg				455 kev counts/min μg			
	Ce^{136}	Ce^{138}	Ce^{140}	Ce^{142}	Ce^{136}	Ce^{138}	Ce^{140}	Ce^{142}
Isotopic enrichment No. 1	9.8	430	4.5	71	30.8	1,340	14.2	220
Isotopic enrichment No. 2	10.1	6	0.8	21	31.0	18.3	2.4	64

ties shows that the activity is associated with a cerium isotope. The data in Table I show that the amount of the 445-kev gamma emitter formed is proportional to the amount of Ce^{136} bombarded and therefore, the transition must occur in the decay of Ce^{137}. The 445-kev gamma is in prompt coincidence with K x-rays which are not critically absorbed in the xenon proportional counter. These facts show that Ce^{137} decays by electron capture with at least some of the captures to a 445-kev level in lanthanum.

Decay curves of the x-rays and the 445-kev gamma ray show that the 8.7-hr electron capture process is the daughter of the 34.5-hr isomeric transition. When the parent and daughter are in equilibrium, the relative number of 255-kev and 445-kev transitions indicates that 3% of the electron captures are to the 445-kev level in La^{137}. An approximate K conversion coefficient of 0.02 for the 445-kev transition was computed from intensity ratios and indicates an $M1$ or $E2$ transition.

Approximate cross sections of Ce^{136} for pile neutrons have been computed from the yields. These give 20 barns for capture to the ground state and 2 barns for capture to the isomeric state of Ce^{137}. These cross sections are known within a factor of two and are in reasonable agreement with 25 barns±100% given by Pomerance[10] for the thermal neutron absorption cross section of cerium-136.

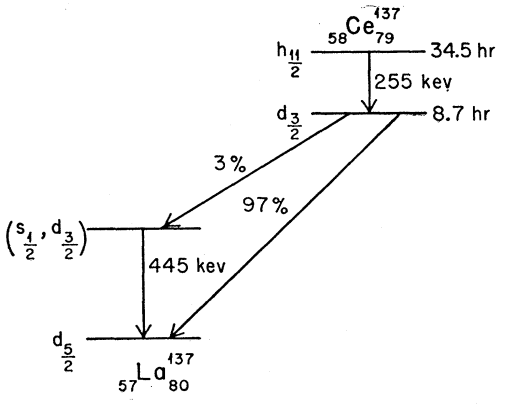

FIG. 1. Cerium-137 decay scheme.

[10] H. S. Pomerance, Phys. Rev. **88**, 442 (1952).

IV. DISCUSSION

On the basis of the single-particle model[11] an isomeric transition between $h_{11/2}$ and $d_{\frac{3}{2}}$ levels is probable in Ce^{137}. In the 79-neutron isotopes of Te, Xe, and Ba the single-particle predictions have been verified and isomeric transitions from $h_{11/2}$ levels to $d_{\frac{3}{2}}$ levels are known. In the case of Ce^{137} the K conversion coefficient and the K/L conversion ratio indicate an $M4$ transition for the decay of the 255-kev isomeric state. Therefore, the isomeric state is shown as an $h_{11/2}$ level in the decay scheme proposed for Ce^{137} in Fig. 1. Although it seems quite clear that the isomeric transition is to a $d_{\frac{3}{2}}$ level, it is not as certain that the ground state of Ce^{137} is a $d_{\frac{3}{2}}$ level. The ground states of many of the odd-neutron isotopes near the end of the 82-neutron shell are $s_{\frac{1}{2}}$ levels. The systematics of energy level spacing[9] do not lead to a definite prediction concerning the ground state of Ce^{137}. However, no experimental evidence for a $d_{\frac{3}{2}}-s_{\frac{1}{2}}$ transition following decay of the isomer was found. The fact that electron capture to the 445-kev and to the ground state of La^{137} is allowed can also be used as an argument against an $s_{\frac{1}{2}}$ ground state for Ce^{137}.

Of the single-particle levels available for assignment to the ground state of La^{137} only $d_{\frac{3}{2}}$ fits the experimental observations. Since the spin of Ba^{137} is known to be $d_{\frac{3}{2}}$, a transition between the ground states of La^{137} and Ba^{137} should be allowed. However, no evidence of K or L x-rays which could be attributed to electron capture decay of La^{137} was found. Because of the high x-ray counting efficiency of the detectors used in this work the lower limit for the K or L electron capture half-life for La^{137} must be increased from 400 years[1] to 10^5 years. The long half-life of La^{137} may mean that the energy available for the transition is very low. On the other hand, this may be another case where the probability of a transition is reduced because it occurs between a state with a mixed configuration and a pure single particle state.[12]

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[11] M. G. Mayer, Phys. Rev. **78**, 16 (1950).

[12] A. de-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).