

Evidence for the Formation of Ce^{139} by an $n\text{-}\gamma$ Reaction*[†]

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A radioactivity, found in old samples of pile-irradiated cerium, can be attributed to Ce^{139} (140d) formed by an $n\text{-}\gamma$ process. This activity had a half-life of more than 120 days, and absorption curves indicated it possessed a larger component of x-rays than the more abundant Ce^{141} (28d). The activity followed Ce in chemical separations from La, Ca, Ba, Pb, Fe, and Zn. The ratio of cross sections, $\sigma_{138}/\sigma_{140}$, was estimated to be 1.4.

I. INTRODUCTION

Ce^{139} , which decays by K -capture, has been prepared by deuteron bombardment^{1,2} of La, alpha-bombardment¹ of Ba and fission³ of Bi. Besides the La x-rays the radiation contains a 0.18 Mev gamma, partly converted, and possibly harder radiations of low intensity.^{1,2} Although it would be formed by the $n\text{-}\gamma$ reaction from Ce^{138} (abundance 0.25 percent), this process has not been previously observed because of the high intensity of radiation from Ce^{141} formed during the neutron irradiation of this element.⁴ Ce^{141} has been reported to decay with a 28-day half-life by β -emission with a maximum energy of 0.53 Mev and with a partially converted 0.146 Mev γ -ray accompanying 70 percent of the disintegrations.⁵⁻⁷

II. EXPERIMENTAL

It was desired to use for tracer experiments some old samples of cerium activity which had been prepared by neutron irradiation in the Oak Ridge pile for 30 days. As a routine procedure two portions of the activity were mounted for observance of their decay by means of a thin-wall, hydrogen-filled G-M counter tube. One sample was arranged with little absorber so that the major contribution came from beta-particles; the other stronger sample was separated from the counter tube by 270 mg/cm² of Al, sufficient to stop all the betas. Individual counting rates were always compared with that of a U_3O_8 standard sample. A difference in the disintegration periods of the two samples was soon noted and the decay curves followed from 240 to 530 days after irradiation. The results, shown in Fig. 1, indicated that although the β -component decayed exponentially with about a 26-day half-life, the gamma-

component contained in addition a long-lived fraction with a half-life greater than 120 days. Consequently the radiation was studied by absorption methods and a number of chemical separations performed in order to establish its identity. The first sample (A) had been formed by a one-month irradiation in the Oak Ridge pile of about one gram of carefully purified Ce metal which had been originally prepared by Dr. F. H. Spedding and co-workers. A similar long-lived activity was also found in a second sample of metal (B) which was irradiated for three months in the Oak Ridge pile. Both samples were employed in the various chemical tests.

Absorption curves for the radiation in Ta, appearing in Fig. 2, offered some information concerning the nature of the gamma-components. The upper curve shows the absorption of a sample of freshly irradiated ceric oxide. There was sufficient aluminum directly in front of the counter tube (225 mg/cm²) to stop completely all Ce^{141} betas. This curve was identical within experimental error to one obtained with sample (A) 203 days after irradiation. The half-thickness of the hard component (410 mg Ta/cm²) was consistent with either the 0.146 and 0.18 Mev gamma-rays of Ce^{141} and Ce^{139} . The original sample however contained a soft component

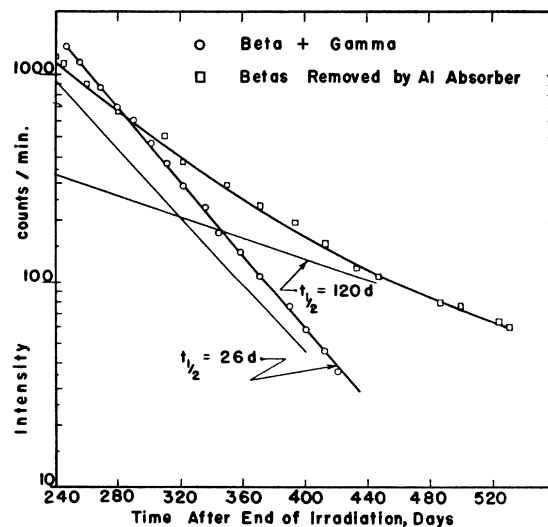


FIG. 1. Cerium decay curves.

* This work was performed in the Ames Laboratory of the AEC.

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¹ M. L. Pool and N. L. Krisberg, Phys. Rev. **73**, 1035 (1948).

² J. B. Chubuck and I. Perlman, Phys. Rev. **74**, 982 (1948).

³ R. H. Soekerman and I. Perlman, Phys. Rev. **73**, 1127 (1948).

⁴ G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

⁵ W. H. Burgus reported in "Plutonium Project Report," CC-680, p. 13 (May, 1943).

⁶ L. R. Shepard, Research **1**, 671 (1948).

⁷ Ter-Pogossion, Cook, Goddard and Robinson, Phys. Rev. **76**, 909 (1949).

TABLE I. Results of chemical separations.

Series	Sample	Days after irradiation	Element	Percent of activity carried (beta and gamma)
1	A	460	La	1.2
			Ce	99.
2	B	230	Pb	0.3
			Ca-Ba	0.2
			Ce	99.
3	B	290	Fe	5.
			Zn	1.4
			Ce	94.

with half-thickness of 39 mg Ta/cm² which might correspond to a photon of about 34 kev and in agreement with a light rare earth *K* x-ray. Such an x-ray would not appear in the spectrometer determination of photo-electrons from a uranium foil reported in reference 7. They might arise as a result of internal conversion of the gamma-rays following the Ce¹⁴¹ disintegration. The ratio of intensities for the hard to soft component was 1.6. In a similar absorption curve on the same material 363 days after irradiation the ratio of the hard to soft components had decreased to 1.0. Because of the low activity in the sample the line for the hard component was fitted to the data by the least squares method. These results would be consistent with the presence of Ce¹³⁹ which, decaying by electron capture, would emit the lanthanum *K* x-rays with high intensity.

There remained the necessity of associating the long-lived activity with Ce. A consideration of possible impurities in the metal indicated that the most probable chemical impurities would be other rare earths, alkaline earths from refractory liners, lead, iron, and zinc. A series of chemical separations were performed in which carriers from these groups of elements were added and

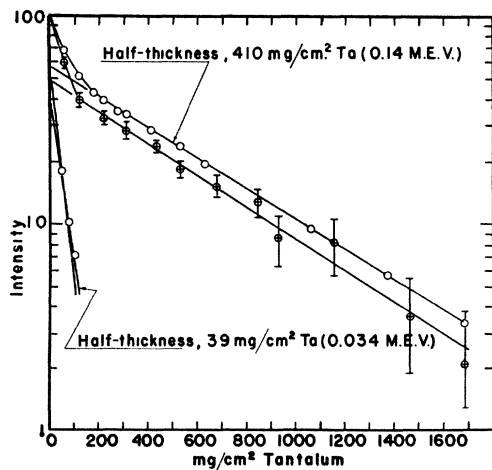


FIG. 2. Absorption curve of cerium photons in tantalum. O—203 days after end of irradiation (sample A). ⊕—363 days after end of irradiation (sample A). [270 mg/cm² of Al in front of counter to stop electrons.]

then separated from the radioactive cerium.^{7a} The results of these separations have been tabulated in Table I. In each case the major fraction of the activity followed the cerium. However, all of the separated elements, especially Fe, carried measurable activity. In each case the absorption curves of these activities appeared to be identical with the original Ce activity insofar as could be determined with the intensities at hand. In addition the decay rate of the photon component of radiation from cerium, which had undergone the chemical separations, was compared with that of the untreated cerium. To avoid effects due to changes in counter characteristics, both the separated and unseparated samples were always counted and their ratios computed. (Ratio: $R = (x-b)/(y-b)$ where x is the counting rate of the separated sample, y is the counting rate of the unseparated sample, and b is the background rate.) These ratios have been plotted in Fig. 3, each point

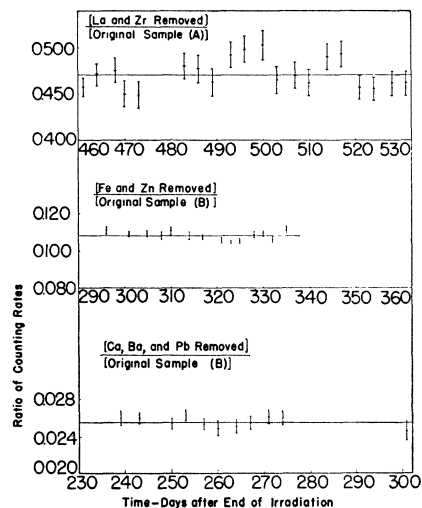


FIG. 3. Ratios of photon counting rates of separated and unseparated cerium samples.

with a width 2σ .

$$\sigma = [(\partial R/\partial x)^2 \sigma_x^2 + (\partial R/\partial y)^2 \sigma_y^2 + (\partial R/\partial b)^2 \sigma_b^2]^{\frac{1}{2}}$$

A chi-square determination of the data showed that the ratios did not deviate from a constant value by more

^{7a} In the first series the La carrier was separated from Ce by the method of Boldridge and Hume (AECD-2531-C). In this procedure the Ce was oxidized to the tetravalent state and precipitated as Ce(IO₃)₄. The La was subsequently precipitated as the oxalate. In the second series Ce was separated from Pb, Ca, and Ba carriers which should also have carried Sr and Ra. Lead was first precipitated as PbI₂. Cerium was precipitated with NH₃ in the presence of NH₄Cl and finally the alkaline earths were precipitated as sulfates. In the third series Fe and Zn carriers were separated from the cerium. Ferric iron was separated by 10 extractions into ether-HCl solution. After evaporation of the solvent, it was precipitated with ammonia, redissolved in HF, and finally precipitated again by ammonia after the removal of HF. Zn was separated by precipitation with H₂S from mixed citrate and formate buffers [H. A. Fales and G. M. Ware, J. Am. Chem. Soc. 41, 487 (1919).]

than would reasonably be expected from statistical fluctuations.

It was estimated from Fig. 1 that at 240 days after irradiation the long-lived activity contributed about one-fourth of the total gamma-count. Even if the chemical separations were only 50 percent efficient so that this contribution was reduced from one-fourth to one-eighth, it was calculated that the counting rate ratios should change by about 20 percent in two months. The absence of such variations in any of the three samples was considered as proof that the long-lived activity was not carried by any of the other elements and must therefore be associated with a cerium isotope.

The evidence for the presence of Ce^{139} consists in: (1) The existence of a half-life of greater than 120 days in the photon component of the irradiated cerium, (2) the greater contributions of soft radiation corresponding to x-rays following K capture in this activity, and (3) the impossibility of separating the activity chemically from cerium, by removal of the most likely impurities.

III. ESTIMATE OF CROSS SECTION RATIO

An estimate of the ratio of cross sections of (Ce^{138}/Ce^{140}) should now be possible from the fact mentioned above, that at 240 days after a 30-day irradiation the counting activity of the long-lived gammas was one-third of that for the short-lived activity which Shepard estimated to occur in 70 percent of the disintegrations of Ce^{141} . Then, estimating the counting efficiency for Ce^{139} radiation to be twice as great as that for Ce^{141} because of the larger fraction of x-rays of about 30 kev energy,⁸ the ratio of the disintegration rates 139/141 at this time would be 0.12. Using the half-life values of 140 and 28 days this ratio would have been 1.1×10^{-3} at the end of irradiation. For a 30-day bombardment, and using the isotopic abundances of 0.250 and 88.48 percent given by Ingham, Hayden, and Hess,⁹ the ratio of the cross sections was calculated to be: $\sigma(138)/\sigma(140) \approx 1.4$.

⁸ H. Maier-Liebnitz, *Zeits. f. Naturforsch.* **1**, 243 (1946).

⁹ Ingham, Hayden, and Hess, *Phys. Rev.* **72**, 967 (1947).

Variational Principles for Scattering Processes. I

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A systematic treatment is presented of the application of variational principles to the quantum theory of scattering.

Starting from the time-dependent theory, a pair of variational principles is provided for the approximate calculation of the unitary (collision) operator that describes the connection between the initial and final states of the system. An equivalent formulation of the theory is obtained by expressing the collision operator in terms of an Hermitian (reaction) operator; variational principles for the reaction operator follow. The time-independent theory, including variational principles for the operators now used to describe transitions, emerges from the time-dependent theory by restricting the discussion to stationary states. Specialization to the case of scattering by a central force field establishes the connection with the conventional phase shift analysis and results in a variational principle for the phase shift.

As an illustration, the results of Fermi and Breit on the scattering of slow neutrons by bound protons are deduced by variational methods.

I. INTRODUCTION

ALTHOUGH variational methods have long been applied to eigenvalue problems in many fields of physics, no systematic use had been made of variational procedures in connection with scattering processes until the period 1942-1946 when variational techniques, among others, were extensively employed in the solution of electromagnetic wave guide problems.¹ Variational formulations have also been devised for the treatment

of neutron diffusion,² acoustical and optical diffraction,³ and quantum-mechanical scattering problems.⁴ Indeed,

¹ J. Schwinger, unpublished; R. E. Marshak, *Phys. Rev.* **71**, 688 (1947).

² H. Levine and J. Schwinger, *Phys. Rev.* **74**, 958 (1948); **75**, 1423 (1949).

³ J. Schwinger, "Lectures on Nuclear Physics," Harvard University, 1947; J. Schwinger, *Phys. Rev.* **72**, 742 (1947); J. M. Blatt, *Phys. Rev.* **74**, 92 (1948); W. Kohn, *Phys. Rev.* **74**, 1763 (1948); J. M. Blatt and J. D. Jackson, *Phys. Rev.* **76**, 18 (1949). Variational principles for scattering problems have also been independently developed by L. Hulthén, see Mott and Massey, *The Theory of Atomic Collisions* (Oxford University Press, London, 1949), 2nd ed., p. 128, and I. Tamm, *J. Exp. Theor. Phys. USSR* **18**, 337 (1948); **19**, 74 (1949).

¹ "Notes on Lectures by Julian Schwinger: Discontinuities in Waveguides," prepared by David S. Saxon, MIT Radiation Laboratory Report, February 1945.