

Short-Lived Cerium Isotopes from Uranium Fission†

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It was demonstrated that the previously reported 1.8-hr Ce^{146} and 4.5-hr Pr^{146} do not occur among the uranium fission products. Ce^{146} and Pr^{146} were briefly reinvestigated: the former decays with a half-life of 13.9 ± 0.6 minutes and emits β -rays of 0.9-Mev maximum energy followed by soft γ -rays; the latter decays with a half-life of 24.4 ± 0.5 minutes and its maximum β -ray energy is 3.8 Mev. In addition, it was shown that any undiscovered short-lived Ce fission products must decay with half-lives shorter than 6 minutes.

A STUDY was made of short-lived cerium isotopes, produced by fission of uranium to confirm, or disprove, the existence of 1.8-hr Ce^{145} and 4.5-hr Pr^{145} .¹⁻³

The chemical procedure used was a modification of the method of Boldridge and Hume,⁴ which is based on the fact that ceric iodate is insoluble in 6 *N* nitric acid while all trivalent rare earth iodates remain in solution. The following changes were found necessary in order to obtain sufficient radiochemical purity: (1) Two rare earth fluoride precipitations were made instead of one, and rhodium was used as hold-back carrier; (2) the cerium was passed through six precipitation cycles as $Ce(IO_3)_4$ instead of three, and barium was used as hold-back carrier twice; (3) three zirconium iodate precipitations were made instead of one. When the praseodymium daughters were isolated from cerium, ceric iodate was precipitated at least six times.

Seven irradiations of uranyl nitrate followed by separation of cerium yielded decay curves showing the 14.6-min Ce^{146} -24.6-min Pr^{146} chain and 33-hour Ce^{143} . In no case was any 1.8-hr Ce observed or any other half-time between 24.6-min and 33-hrs. Also, on four irradiations a Pr fraction was isolated from pure cerium which had decayed for times between 30 minutes and 2.5 hours. The Pr decay curves showed the 24.6-min Pr and the 13.8-day Pr^{143} , but no 4.5-hr Pr or any other intermediate half-time. These experiments show conclusively that the previously reported Ce^{145} - Pr^{145} chain does not occur among the uranium fission products. If it did occur with a fission yield greater than 0.1 percent, it would have been easily observed in these decay curves; the yield expected for mass 145 is about 4 percent. The previously reported Ce^{145} - Pr^{145} chain was probably due to contamination, perhaps by 2.0-hr Nd^{149} and 3.7-hr La^{141} . Only one or two $Ce(IO_3)_4$ precipitations were

done formerly, while in the present work far greater purity was achieved. The report of 0.8-sec Xe^{145} ³ is also based on insufficiently purified cerium and is therefore probably in error.

The Ce^{146} - Pr^{146} chain, previously reported by Götte,⁵ and Schuman,⁶ was observed and investigated further. Pure cerium and praseodymium fission products were quickly isolated and their activities followed with an end-window Geiger tube through a series of aluminum absorbers. The half-life of Pr^{146} was found to be 24.4 ± 0.5 minutes as determined from twelve decay curves, each followed for four to nine half-lives. The maximum beta-energy was found to be 3.8 Mev from the absorption curves which gave a Feather plot range of 1900 mg/cm². Schuman⁶ reported 24.6 ± 0.3 minutes for the half-life, about 3 Mev for the maximum β -ray energy, and 1.4 Mev for the γ -ray energy.

A growth of activity was observed in the cerium fractions when measured through absorbers thicker than 50 mg/cm². This resulted from the fact that the beta-rays of the Ce^{146} are soft and easily absorbed relative to the energetic Pr^{146} betas. The four growth-decay curves obtained with enough absorber to completely remove the Ce^{146} beta-rays were analyzed to determine the half-life of Ce^{146} , as follows. The observed activity at various times was subtracted from the asymptote (corresponding to the 24.4-min half-life of the daughter) which the curves approach after about three hours. The resulting difference curves were exponential and corresponded to the decay of the Ce^{146} through a factor of about 20 with a half-life of 13.9 ± 0.6 minutes. Schuman's value⁶ was 14.6 ± 0.8 minutes determined by counting a series of cerium aliquots that were periodically cleaned of Pr daughter activity. The maximum β -ray energy of the Ce^{146} was found by analyzing absorption curves obtained for the Ce^{146} - Pr^{146} mixture. The observed half-thickness value in aluminum for the Ce^{146} β -rays was 31 mg/cm², which corresponds⁷ to about 0.9-Mev maximum energy. It was also found that 13.9-min Ce emits soft γ -rays, probably between

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¹ N. E. Ballou, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 173, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

² S. Katcoff, Paper No. 174 of reference 1.

³ Dillard, Adams, Finston, and Turkevich, Paper No. 68 of reference 1.

⁴ W. F. Boldridge and D. N. Hume, Paper No. 294 of reference 1.

⁵ H. Götte, *Z. Naturforsch.* **1**, 377 (1946).

⁶ R. P. Schuman, Metallurgical Laboratory Reports CN-2799, March, 1945 and CN-2929, April, 1945 (unpublished).

⁷ D. W. Engelkemeir, Figs. I-7 of "Introduction to Part I," *Radiochemical Studies: The Fission Products*, p. 18, reference 1.

0.2 and 0.3 Mev. This is based on an analysis of two runs in which the cerium fractions were measured through three lead absorbers (2, 3, and 4 g/cm²). A more thorough investigation of these radiations is planned by means of a scintillation spectrometer.

In the above cerium runs counting was begun as soon as 28 minutes after the irradiation and 3.5 minutes after the final separation from Pr. Since no component

shorter than 13.9-min was seen, it is concluded that any other short-lived Ce fission products must have half-times shorter than 6 minutes.

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Gamma-Radiation from Interaction of 14-Mev Neutrons with Iron

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Experiments have been performed to observe gamma-rays produced by the interaction of 14-Mev neutrons with iron. Although the gamma-spectrum appears to be continuous below 3.0 Mev, there may be discrete gamma-rays at 3.3, 4.4, 5.8, 7.1, and 8.75 Mev. The total cross section for gamma-ray production is 4.6 ± 0.5 barns.

I. INTRODUCTION

SOME observations^{1,2} of the gamma-radiation produced by interaction of fast neutrons with matter have been carried out primarily by absorption techniques. To obtain better resolution, we have made measurements with a spectrometer consisting of a single crystal of NaI(Tl) in conjunction with an eighteen-channel pulse-height analyzer. The spectra examined so far appear to consist of a low energy

continuum overlaid by a series of discrete lines or bands at higher energies.

II. DESCRIPTION OF THE EXPERIMENT

A 250-kilovolt Cockcroft-Walton accelerator³ employing the $T(d,n)He^4$ reaction was used as a source⁴ of 14-Mev neutrons, which were allowed to fall upon an iron converter arranged as shown in Fig. 1.

The converter and the accelerator monitor were so placed that whenever an alpha-particle entered the monitor a corresponding neutron entered the converter. The iron converter, approximately 0.75 inelastic neutron mean free paths long, was just wide enough to cover the "coincidence neutron beam." On the average a gamma-ray traveled about 0.25 mean free paths before leaving the converter and reaching the detector.

A block diagram of the electronic arrangement is shown in Fig. 2. A single crystal of NaI(Tl) and a 5819 photomultiplier in conjunction with an eighteen-channel pulse-height analyzer detected the gamma-rays produced in the converter. The multichannel analyzer was gated only by coincidences between the alpha-monitor and the spectrometer, thereby eliminating background counts from direct neutrons on the crystal as well as radiation scattered off the walls of the room. The amplifier discriminator in the alpha-channel was set to eliminate counts from the $D(d,n)He^3$ reaction, from electrons and low level noise. Scalers were placed at all points in the system that gave useful information.

Figure 3 shows typical calibration curves with the 4.45-Mev line of Po-Be and the two gamma-lines of

³ Bergstrahl, Dunning, Durand, Ellison, Howerton, and Slavin (to be published).

⁴ Graves, Rodrigues, Goldblatt, and Meyer, *Rev. Sci. Instr.* **20**, 579 (1949).

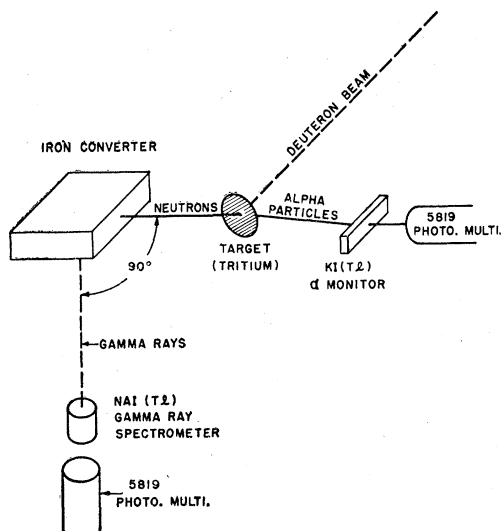


FIG. 1. Schematic representation of the experimental arrangement showing accelerator target, alpha-monitor, iron converter, and crystal detector.

¹ D. F. Lea, *Proc. Roy. Soc. (London)* **150**, 637 (1935).

² J. H. Coon, private communication.