per neutron incident upon the scatterer. The neutron beam was monitored by means of BF_3 long counters placed at 90° with respect to the proton beam. Preliminary measurements with BF_3 counters gave a calibration between the neutron flux at the scatterer and the flux incident upon the monitor counters. Experiments are in progress to determine the absolute value of σ_i.

The NaI spectrometer produces three (possibly four) relatively intense gamma rays when subjected to neutron bombardment, Fig. 3. These gamma rays are tentatively assigned to inelastic processes in the NaI crystal. The gamma energies are: 200 kev, ~410 kev (possibly two gamma rays superposed), and 630 kev. The gamma at 135 kev is attributed to the electric excitation of the Ta backing of the target.[4] A sample gamma spectrum for Fe is also shown in Fig. 3.

[4] C. L. McClelland and C. Goodman, Phys. Rev. **91**, 760 (1953).

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A New 3.0-min Ce Fission Product and its 5.95-hr Pr Daughter*

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Rapid chemical separations of Ce from neutron-irradiated U led to identification of a 3.0-min Ce which decays to 5.95-hr Pr. The former emits β rays, whose maximum energy is approximately 2.0 Mev, and γ rays whose spectrum was not investigated. The Pr decays by emission of a single β ray whose maximum energy is 1.7 Mev; no γ rays were found. This chain has tentatively been assigned to mass 145.

I. INTRODUCTION

IN 1942–43 Ballou reported[1] a 1.8-hr Ce—4.5-hr Pr fission product chain. Recently Caretto and Katcoff showed[2] that there was no Ce fission product with a half-life between 13.9 min and 33 hr, and that previous reports of a 1.8-hr Ce probably resulted from insufficient purification of the cerium. Furthermore, it was shown that no Pr activity with a half-life between 24.4 min and 13.8 days can be isolated from pure fission product Ce which had decayed for 0.5 to 2.5 hours. Since the chemical method for Ce required a minimum of 28 minutes, it was concluded then that any undiscovered short-lived Ce fission products must decay with half-lives shorter than 6 minutes.

A new and shorter radiochemical procedure developed by Glendenin[3] was used by us in a recent scintillation spectrometer investigation[4] of the radiations from 13.9-min Ce^{146} and 24.4-min Pr^{146}. It was noticed that a 6-hr half-life appeared in Pr separated from fission Ce which had been purified in 11 to 27 minutes after irradiation. In one run where the Ce was allowed to decay for 43 minutes before its purification was completed, virtually no 6-hr component was seen. From these observations it was postulated that a Ce fission product with about a 3-min half-life was decaying to a 6-hr Pr daughter. Subsequent experiments, described below, verified this supposition.

In Ballou's early work,[1] he made separations of Ce and Y from rare earth fission products, and then he showed by a partial separation ($NaNO_3$ fusion method[5]) of La from the remaining rare earths that a 4.5-hr component was probably not associated with La and that it probably was a Pr, Nd, or Pm activity. At the present time it appears that his 4.5-hr component resulted from the 6-hr Pr plus various impurities. He did not prove that the 6-hr activity was a daughter of Ce, and therefore a Pr isotope, because his procedure for Ce was too long to permit isolation of 3-min Ce.

II. CHEMICAL PROCEDURE

Samples of uranyl nitrate (10 to 100 mg) were irradiated in the Brookhaven pile at a flux of 4×10^{12} neutrons/cm² sec for one to three minutes. The chemical separation of Ce was begun almost immediately. It is based on solvent extraction of tetravalent Ce from $10M$ HNO_3 with methyl isobutyl ketone. The trivalent rare earths as well as most of the other fission products remain in the aqueous phase. $NaBrO_3$ was used as the oxidizing agent for the Ce. The organic layer containing the Ce^{+4} was washed with $10M$ HNO_3 and then the Ce was back-extracted into an aqueous phase after reduction to Ce^{+3} with H_2O_2. This cycle was then repeated two or three more times. The Ce samples were

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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] A. A. Caretto, Jr. and S. Katcoff, Phys. Rev. **89**, 1267 (1953).
[3] L. E. Glendenin (private communication). A similar solvent extraction method was used in this laboratory by J. W. Gryder and R. W. Dodson to separate tetravalent from trivalent Ce: J. Am. Chem. Soc. **73**, 2890 (1951).
[4] Bernstein, Markowitz, and Katcoff (to be published).
[5] N. E. Ballou and J. A. Marinsky, Paper No. 300 of reference 1.

mounted as ceric iodate, usually about 9 minutes after the end of irradiation.

The Pr samples were extracted from Ce preparations which had been purified in 7 to 18 minutes and which were then allowed to decay for 6 to 75 minutes. La carrier was used for the Pr activity which was purified, usually by making 3 ceric iodate precipitations (to remove Ce activity), 2 LaF_3 precipitations, and one lanthanum oxalate precipitation. The latter was either mounted directly, or it was ignited to the oxide for weighing and mounting. Table I shows the results of an experiment in which a purified Ce preparation was divided into 4 equal parts. Pr activity was separated from each, but with variable amounts of purification as shown in the table. In each case a final lanthanum oxalate precipitate was ignited to the oxide, weighed, mounted, and the decay of its activity followed. It is seen that the specific activity of the 6-hr period is the same for all samples within experimental error. This demonstrated that the 6-hr activity must be a rare earth, probably Pr.

III. HALF-LIFE AND RADIATIONS OF Ce^{145}

In order to prove that the 6-hr activity is the Pr daughter of a short-lived Ce parent, three experiments were performed. In each one a series of equal Ce samples was thoroughly purified of any daughter activities at successive intervals of 2 to 3 minutes. Then the samples were allowed to decay for at least 20 minutes, a measured amount of La carrier was added to each, and Pr was extracted by the method described above. In each Pr sample the amount of the 6-hr activity (corrected for chemical yield) should be directly proportional to the activity of its parent in the Ce sample from which it was derived, at the beginning of the growth interval. The results of one experiment are shown in Fig. 1, circled points. The observed decrease in 6-hr Pr activity with each successive sample shows that it is the daughter of a 3.0-min Ce. The other two runs gave the same result but the experimental points were a bit more scattered.

Direct measurement of the decay of the 3-min Ce was complicated by the presence of 13.9-min Ce^{146} and its 24.4-min Pr daughter. The best results were obtained when the Ce was purified within 7 minutes after bombardment and its radiations measured, starting $2\frac{1}{2}$ minutes later, through 243 mg/cm² of Al; this absorber was sufficient to remove the beta radiation of 13.9-min Ce^{146}. An end-window proportional counter was used as detector and the decay was followed for about 42 minutes (Fig. 1, square points). The activity measured during the last 22 minutes was due almost entirely to the 24.4-min Pr^{146}. The contribution of the latter to the activity measured during the first 20 minutes was determined by preparing another Ce sample identical with the preceding except that its isolation was delayed 30 minutes following bombardment to allow the 3-min Ce to decay away. Then all other conditions were duplicated and the activity of the sample was measured for 45 minutes. The shapes of the decay curves were identical for the last 22 minutes, where the curves were normalized. Subtraction of the second curve from the first for the first 20 minutes resulted in the triangles shown in Fig. 1. From this curve, from numerous other curves of this type obtained through various absorbers, and from the successive daughter extraction experiments, we conclude that the Ce^{145} half-life is 3.0 ± 0.1 minutes, where 0.1 min is the estimated maximum error.

Decay of the Ce was measured through a series of 15 Al absorbers up to 1450 mg/cm². An absorption curve for the 3-min Ce radiation was constructed and analyzed by the Feather method. The maximum range of the beta rays was about 1000 mg/cm² corresponding to a maximum energy of approximately 2.0 Mev. Gamma radiation contributed about 2 percent to the total counting rate, in the brass-wall proportional counter, but its spectrum was not investigated.

TABLE I. Effect of variable amounts of purification on the 6-hr period separated from fission Ce.

Sample	Precipitations	Wt. of La_2O_3 (mg)	Activity per mg
1	2 ceric iodate, 1 LaF_3	13.3	207
2	3 ceric iodate, 2 LaF_3	10.8	211
3	3 ceric iodate, 3 LaF_3	11.6	197
4	4 ceric iodate, 4 LaF_3	10.9	198

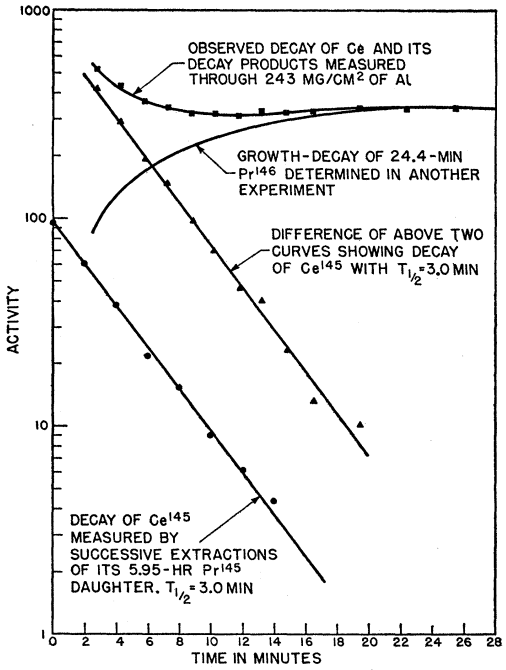

FIG. 1. Decay of Ce^{145} measured directly and by successive daughter extractions.

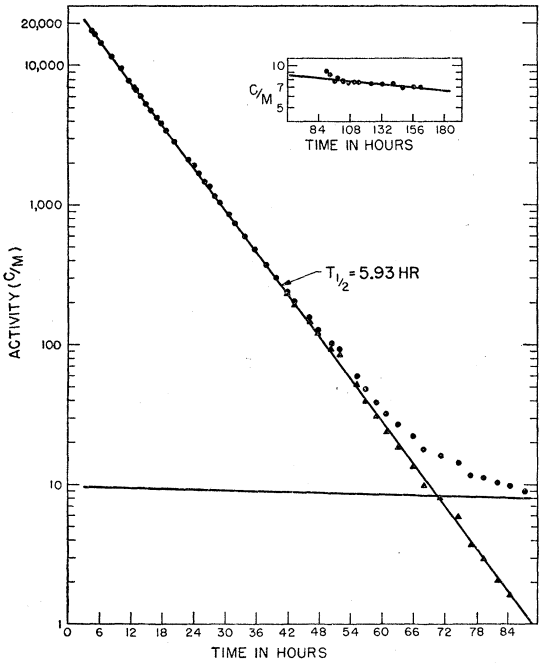

FIG. 2. Decay of Pr^{145}.

IV. HALF-LIFE AND RADIATIONS OF Pr^{145}

Two samples of Pr^{145} were prepared for a half-life determination. One was measured with an end-window G-M tube and yielded a value of 6.0 hours. The 13.8-day Pr^{143} contributed 0.14 percent to the initial counting rate. The second sample was more active than the first and the longer-lived component contributed only 0.05 percent to the initial activity because the growth interval of Pr from Ce was reduced to only 6 minutes. (The parent of 13.8-day Pr^{143} is 33-hr Ce.) The decay of this sample was followed with an end-window proportional counter and the Pr^{145} activity was observed to decay exponentially through 14 half-lives (Fig. 2) with a half-time of 5.93 hours. A weighted average gives a value of 5.95 hours with an estimated maximum error of 0.10 hr. Several other decay curves, which were not determined as carefully as these two, also yielded values very close to 6.0 hours.

Aluminum absorption curves of the 5.95-hr Pr radiation indicated a range of 800 mg/cm^2 for the beta rays, both by visual extrapolation and by the Feather method of analysis. This range corresponds to a maximum energy of 1.7 Mev. The counting rate beyond 800 mg/cm^2 remained nearly constant at about 0.02 percent of the rate measured without absorber. Presumably this residual radiation was electromagnetic in character.

Its exact nature was investigated with a scintillation spectrometer consisting of either a 3 cm$\times$2 cm or a 0.2 cm$\times$2 cm NaI(Tl) crystal, a Dumont K-1186 photomultiplier tube, and a gray wedge pulse-height analyzer.[6] The sample, mounted on an Al card, was surrounded with Be absorber sufficient to stop the beta particles. Most of the radiation was shown to consist of bremsstrahlung produced by the 1.7-Mev β rays in the Al mounting and the Be absorbers. In addition, two weak low-energy lines were observed at 36$\pm$1 kev and 70$\pm$3 kev. In order to verify that the 36-kev line was not produced by La fluorescence in the source[7] from the beta radiation, a similar source was prepared in which the 5.95-hr Pr activity was replaced by twice this activity of P^{32}, a pure beta emitter with approximately the same β-ray energy. A similar bremsstrahlung spectrum was obtained and a very weak line was seen at 33$\pm$1 kev characteristic of La x-rays. Therefore, the 36-kev line probably represents Pr or Nd x-rays (35.6 and 36.9 kev, respectively) arising from internal conversion of the 70-kev γ ray. In order to determine whether these x-rays were associated with 5.95-hr Pr, their decay was followed for 10 hours with a thin NaI(Tl) crystal and a single-channel analyzer. The 70-kev γ rays were too low in intensity for this measurement. The half-life observed for the 36-kev line was 3.3$\pm$1.0 hr and, therefore, it is not associated with the 5.95-hr Pr. Presumably the same is true for the 70-kev γ rays. The origin of these radiations will be investigated.

V. DISCUSSION

A tentative mass assignment of 145 was made for the 3.0-min Ce—5.95-hr Pr chain on the basis of a comparison of the observed energies of decay with those calculated from the semi-empirical mass formula where the constants used are those recommended by Coryell.[8] The observed value for 5.95-hr Pr is 1.7 Mev, and the calculated values are 1.8 Mev for mass 145, 3.6 Mev for mass 146, and 2.7 Mev for mass 147. The last mass number is ruled out because the 5.95-hr Pr does not decay to the well-known 11.3-day Nd^{147}. For example, in Fig. 2 the long-lived component is lower by a factor of 100 than that required by an 11.3-day daughter of the 5.95-hr Pr. Mass assignments above 147 would increase the discrepancy between the observed and calculated disintegration energies. For Ce^{145} the calculated value is 2.4 Mev, close to the observed 2.0 Mev maximum β-ray energy which may represent a transition to the ground state or to a low excited state.

One of the authors (S.S.M.) is indebted to Dr. R. W. Dodson and the Staff of the Chemistry Department for the opportunity to carry out this research at the Brookhaven National Laboratory during the past summer.

[6] Bernstein, Chase, and Schardt, Rev. Sci. Instr. 24, 437 (1953).
[7] La was used as carrier and it was mounted as lanthanum oxalate.
[8] C. D. Coryell, Ann. Rev. Nuclear Sci. 2, 305 (1953).