The Decay Scheme and Angular Correlation of Pr^{144}†

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The decay scheme and angular correlation of Pr^{144} have been studied with lens spectrometer and scintillation counter techniques. The beta-ray spectrum shows three groups with end points at 2.965 ± 0.015, 2.3 ± 0.1, and 0.86 ± 0.1 Mev, the first having a relative intensity of 90 percent, the others each about 5 percent. Gamma-rays were observed by photoelectric conversion and have energies of 0.695 ± 0.005, 1.48 ± 0.01, and 2.185 ± 0.015 Mev and relative intensities of 1:0.4:1.1, respectively. The data indicate that beta-emission takes place to states in Nd^{144} at zero, 0.695, and 2.185 Mev. The absorption of beta-rays in coincidence with gamma-rays shows two components with ranges of 0.34 and 1.0 g/cm² aluminum. The corresponding energies and intensities of these beta-groups confirm the above decay scheme. Gamma-gamma coincidences are also present and their angular correlation has the form $1-0.33\cos^2\theta$, characteristic of either a 1–2–0 or a 1–1–0 cascade. It is suggested that the first excited state of Nd^{144} has spin 2 and even parity and the second level spin 1. Some of the features of Ce^{144} decay are described and the usefulness of this fission-product isotope as a source of homogeneous photoneutrons is discussed.

INTRODUCTION

AMONG long-lived fission-product isotopes examined[1,2] in this laboratory, Ce^{144} (half-life 282 days[3]) in equilibrium with 17-min Pr^{144} was found to give a moderate photoneutron yield from Be and a very weak yield from heavy water. The intensity of gamma-rays above the Be threshold was estimated to be about 2 percent per beta-ray. Because of the convenient half-life and high fission yield of Ce^{144} ($\sim$5 percent[4]) this isotope could well be useful for photoneutron sources.

The present investigation was undertaken initially to measure the gamma-ray spectrum responsible for photoneutron production. Subsequent studies were made to determine the decay scheme and to assign spins by means of angular correlation experiments.

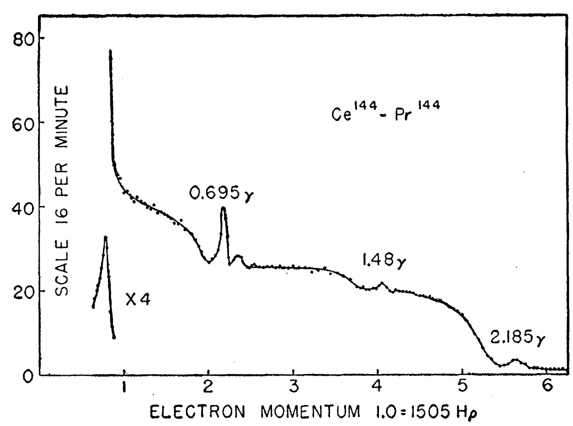

FIG. 1. External conversion electron spectrum due to $Ce^{144}-Pr^{144}$ gamma-rays taken with a 22 mg/cm² Pb converter.

Present information[5-7] indicates that the parent nuclide Ce^{144} decays by emission of beta-rays of about 300 kev maximum energy. A number of soft gamma-rays also occur[7-9] as shown by low energy internal conversion electrons having *K-L* energy separations characteristic of praseodymium. The strongest of these correspond to gamma-rays of 134 kev.

The daughter nucleus Pr^{144} decays [5-7,10,11] mostly to the ground state of Nd^{144} by emission of beta-rays with an end point of 3 Mev. Probable complexity of the beta-spectrum was first indicated by measurement[2] of hard gamma-rays which were subsequently shown by Mandeville and Shapiro[12] to be associated with 17-min Pr^{144}. The latter workers found several gamma-components by absorption techniques and observed coincidences between gamma-rays. In a recent investigation[7] of Ce^{144} and Pr^{144}, Lin-sheng, John, and Kurbatov have found beta-ray groups with end points at 3.00 ± 0.06 Mev, 1.30 ± 0.02 Mev, 605 ± 8 kev, 446 ± 8 kev, and 307 ± 6 kev. Except for the 307-kev group the results on other inner beta-ray groups do not agree with those of the present authors[11] which are reported here in more detail.

EXPERIMENTAL METHODS AND RESULTS

A lens spectrometer of the conventional type was used for investigations of the beta- and gamma-ray

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‡ Part of this work was done in partial fulfilment of the requirements for the Ph.D. degree in physics at Syracuse University.
[1] M. Goldhaber and E. der Mateosian, Brookhaven National Laboratory Report No. 51 (S-5) (1950).
[2] Alburger, der Mateosian, Goldhaber, and Katcoff, Phys. Rev. **82**, 332 (1951).
[3] R. P. Schuman and A. Camilli, Phys. Rev. **84**, 158 (1951).
[4] Appendix B, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1950), National Nuclear Energy Series, Plutonium Project Record, Vol. **9**, Div. IV.
[5] V. A. Nedzel, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1950), Paper No. 187. National Nuclear Energy Series, Plutonium Project Record, Vol. **9**, Div. IV.
[6] Peacock, Jones, and Overman, Plutonium Project Report Mon N-432, 56 (1947).
[7] Lin-sheng, John, and Kurbatov, Phys. Rev. **85**, 487 (1952).
[8] Emmerich, John, and Kurbatov, Phys. Rev. **82**, 968 (1951).
[9] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951).
[10] F. T. Porter and C. S. Cook, Phys. Rev. **85**, 733 (1952).
[11] D. E. Alburger and J. J. Kraushaar, Phys. Rev. **86**, 633(A) (1952).
[12] C. E. Mandeville and E. Shapiro, Proc. Nat. Inst. Sci. India **17**, 45 (1951).

spectra. This is equipped with the usual resolution and antiscattering baffles and in addition has a set of spiral baffles which further reduces the number of scattered beta-rays reaching the detector.

A 25-millicurie cerium-144 source was obtained from Oak Ridge in the summer of 1949. For the gamma-ray measurements the entire amount was deposited without purification in a stainless steel capsule with walls thick enough to absorb the beta-rays. Photoelectric converters 6 mm in diameter were attached to the capsule and measurements on the electron spectrum were carried out at 3 percent resolution. The first observations were made in December, 1949, and a second set of data taken in December, 1950, gave the same results only with an intensity reduced by an amount consistent with the Ce^{144} half-life. The spectrometer yield *versus* electron momentum taken with a lead converter 22 mg/cm^2 thick is shown in Fig. 1. Photoelectric peaks corresponding to gamma-rays of 0.695 ± 0.005 Mev, 1.48$\pm$0.01 Mev, and 2.185$\pm$0.015 Mev are indicated. Calibration was made with Co^{60} gamma-rays. Because of the proximity of the highest energy gamma-ray to the deuterium (γ,n) threshold (2.23 Mev) a check on the energy of this line was made by a comparison measurement on the 2.20-Mev gamma-ray of radium[13] taken under identical geometrical conditions. The Pr^{144} peak was slightly lower in energy than the radium photoelectric peak.

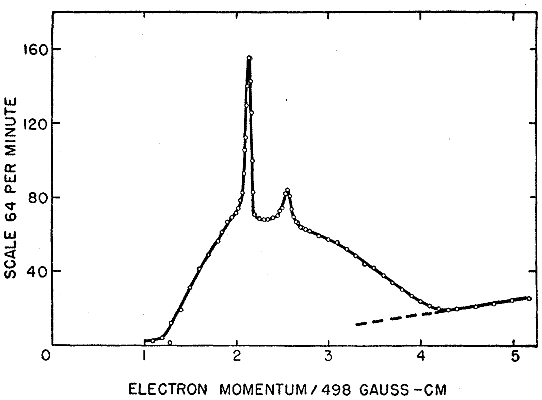

FIG. 2. Electron spectrum of Ce^{144} showing beta-rays of about 300-kev maximum energy and conversion lines of a 135-kev gamma-ray. The rise at the right is due to Pr^{144} beta-rays.

The high intensity low energy line in Fig. 1 is thought to consist of L-shell electrons. The energy as determined from the extrapolated high energy edge corresponds to a gamma-ray of about 135 kev.

Relative intensities of the gamma-rays were obtained from the K-conversion peaks by measuring the areas under these lines on a plot of $N/H\rho$ *versus* $H\rho$ and correcting for photoelectric conversion efficiency according to the empirical formula of Gray.[14] The 0.695-, 1.48-, and 2.185-Mev lines have intensities ratios of 1:0.4:1.1, respectively, the probable errors being about 20 percent.

[13] Way, Fano, Scott, and Thew, National Bureau of Standards Circular No. 499 (1950).

[14] L. H. Gray, Proc. Cambridge Phil. Soc. **27**, 103 (1931).

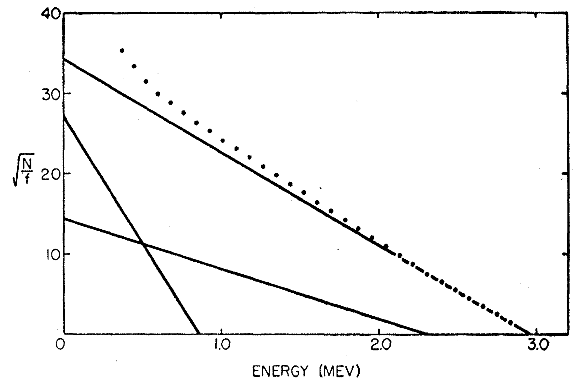

FIG. 3. Kurie plot analysis of the Pr^{144} beta-ray spectrum.

The beta-ray spectrum was first observed using a source of unpurified cerium activity deposited on 0.09 mg/cm^2 Nylon foil. The resulting spectrum consisted of a strong low energy beta-ray group, a pair of prominent K- and L-conversion electron lines and high energy beta-rays. The low energy portion of the spectrum is given in Fig. 2. This shows the presence of beta-rays with an end point of approximately 300 kev and conversion lines corresponding to a gamma-ray of 135 kev. Because of the counter window (1.7 mg/cm^2) and source thickness, the other low energy lines known[7–9] to be present are obscured. Very weak conversion lines of the 230-kev gamma-ray were not evident at the resolution setting used.

At first sight the high energy beta-ray distribution appeared to be a single group. However, the Kurie plot analysis in Fig. 3 shows, in addition to beta-rays with an end point of 2.965$\pm$0.015 Mev, two inner groups with end points of 2.3$\pm$0.1 Mev and 0.86$\pm$0.1 Mev. In these computations corrections were made for counter dead-time, and accurate relativistic Fermi functions were taken from a table kindly supplied by I. Feister of the Bureau of Standards. From the areas under the plots of $N/H\rho$ *versus* $H\rho$ the 2.965-Mev group was shown to contain about 90 percent of the intensity while the two inner groups were each roughly 5 percent. The intensities of the two weak components are accurate to only about 30 percent since subtraction of the main group depends upon an extrapolation from a relatively short region near the Kurie plot end point. In this subtraction the 2.965-Mev group, which is straight above 2.3 Mev, is assumed to have the allowed shape. It then follows from the fit of the 2.3-Mev group to a straight line Kurie plot that the beta-ray distribution in this transition has an experimentally allowed shape.

In the experiments on the high energy beta-rays of Pr^{144} the K-conversion line of the 135-kev Ce^{144} gamma-ray served as a convenient guide to the quality of sources in regard to source thickness effects. Although the actual thickness of the sources was not determined, the experimental width of the conversion line was in most cases equal to the spectrometer resolution setting. It was felt that if the energy spread of a line at 90 kev were small then the effect of the source thickness on the high energy spectrum would be negligible.

The beta-ray measurements just described were made more than two years after the source was received which should exclude difficulties due to short-lived impurities. However, the presence of long-lived fission-product impurities was considered possible, in particular Sr^{90} whose daughter Y^{90}, has a beta-ray with an end point of 2.3 Mev. To check the results a portion of the cerium activity was purified and remeasured. Chemical puirfication was kindly carried out by Dr. Seymour Katcoff of this Laboratory. The procedure was as follows: 5-ml nitric acid and 20-mg $Sr(NO_3)_2$ were added. The cerium was oxidized to the tetravalent state by adding 150 mg of sodium bromate (0.4 ml of saturated solution). In this form it was extracted into ether. The 17-min daughter Pr^{144} was observed to remain behind in the aqueous phase. The ether was then shaken with dilute sulphurous acid solution to reduce the cerium to the trivalent state, thereby returning it to an aqueous phase. The above cycle was repeated but without the addition of Sr^{++}. The aqueous phase containing the Ce^{144} was evaporated to dryness. Heating with an open flame was necessary to drive off a small amount of H_2SO_4. A few drops of HCl were added and boiled off and the activity was taken up in a small amount of water. For beta-ray measurements the activity was evaporated on a Zapon film ($\sim$20 μg/cm^2) stretched on a Lucite cup as in previous experiments. The spectrum showed the same features as with the unpurified source.

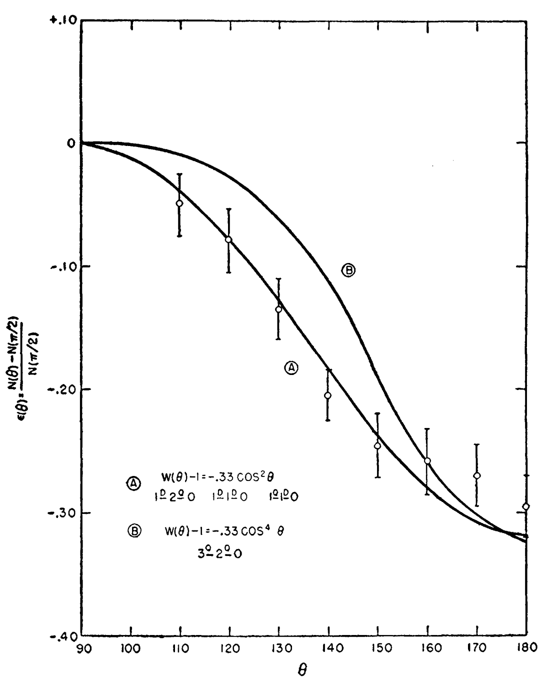

FIG. 4. Angular correlation of Pr^{144} gamma-rays. Resolving time=0.12 μsec; angular resolution=12°. The best fit is obtained with the function $1-0.33\cos^2\theta$.

As a check on the decay scheme suggested by the lens spectrometer measurements the absorption of beta-rays in coincidence with gamma-rays was examined by means of two scintillation detectors. Anthracene was used for the beta-counter and NaI(Tl) for the gamma-counter. The resulting coincidence yield as a function of aluminum absorber thickness in front of the beta-counter was corrected for random coincidences and for gamma-gamma coincidences. The corrected absorption curve shows clearly that two components are present with ranges of about 0.34 and 1.0 g/cm^2 Al and the corresponding beta-ray maximum energies, as obtained from the Feather relation, are 0.9 and 2.1 Mev, respectively. Extrapolation of the individual curves to zero absorber thickness indicates that about twice as many beta-gamma coincidences are associated with the 0.9-Mev beta as with the 2.1-Mev beta. Rough confirmation of the lens spectrometer data is thus obtained.

Angular correlation experiments on the gamma-rays of Pr^{144} were carried out with two sodium-iodide-5819 scintillation detectors connected in coincidence. The arrangement was similar to that used[15] in measurements of the angular correlation of Rh^{106} gamma-rays. Co^{60} gamma-rays which have a well-established angular correlation were used to check the apparatus. Measurements on Pr^{144} were made with sufficiently high discriminator settings so that the low energy lines in the decay of Ce^{144} were not observed. Scattering from one counter to the other was effectively prevented by lead shielding.

Figure 4 shows the experimentally observed angular correlation as well as the computed curves for $1-0.33\cos^2\theta$ and $1-0.33\cos^4\theta$ distributions. The computed curves have been corrected for the angular resolution of the apparatus. The presence of annihilation radiation at first caused the points near 180° to be high, but this effect was later diminished by increasing the bias on one of the counters to a point where it no longer had an appreciable efficiency for 0.5-Mev gamma-radiation.

DISCUSSION

The measurements on Pr^{144} described above are consistent with the decay scheme given in Fig. 5. The 0.695- and 1.48-Mev gamma-ray transitions are evidently in cascade while the 2.185-Mev line is a cross-over. The fact that the 0.695-Mev gamma-ray is low is indicated by its greater intensity and by the observation of beta-ray transitions feeding this state.

In the angular correlation of the gamma-rays, if pure dipole, quadrupole, and octapole radiation are considered, and if the ground state of Nd^{144} is assumed

[15] J. J. Kraushaar, Phys. Rev. 85, 727 (1952).

to have spin 0, then the two computed curves are the only ones which bear a similarity to the experimental results. It is apparent that the $1-0.33\cos^2\theta$ curve, characteristic of a 1–2–0 or a 1–1–0 cascade, best fits the data. As a further argument against the $1-0.33\cos^4\theta$ correlation, which would require a spin of 3 for the second excited state, one would not expect the cross-over to occur with the observed intensity. Of the mixture possibilities only 1–2–0 (*D-Q*) with up to 6 percent admixture of quadrupole radiation in the first transition and $\delta=0°$ satisfactorily fits all the data.

Between the two possibilities allowed by the observed correlation (1–1–0 or 1–2–0) the choice of spins for Nd^{144} given in Fig. 5 is based on the general rule[16] that the first excited states of most even-even nuclei have spin two and even parity. The parity of the 2.185-Mev level cannot be assigned at present.

The log *ft* value of 6.6 for the ground-state beta-ray would assign this transition as probably first forbidden according to the classification[17] of Nordheim. For the 2.3-Mev beta the log *ft* is perhaps a bit higher, but it too could be classed as first forbidden. Pr^{144} could then have spin 1 or 0 and the parity of this nucleus, according to the shell model, might be expected[18] to be odd since the 59th proton is probably even and the 85th proton odd.

According to this scheme the 2.185-Mev line accounts for about 40 percent of all Pr^{144} gamma-rays and occurs in about 3 percent of the beta-decays. The latter figure is in agreement with estimates from the photoneutron data previously mentioned. A simple calculation shows that a source of $(Ce^{144}-Pr^{144})$ plus Be should emit monoenergetic photoneutrons of about 460 kev.

In the photoneutron measurements[1] of Goldhaber and der Mateosian the ratio of yields from Be and D_2O was 650 using a Ce^{144} source, whereas the corresponding ratio obtained with a radium source in the same geometries was 20. However, the observation of the weak yield from heavy water is not explained by the decay scheme given in Fig. 5. After assuming that an impurity is not responsible, there are two other possibilities. A level might exist in Nd^{144} having an energy above the $D(\gamma n)$ threshold, and this could be weakly excited by beta-emission of Pr^{144}. If this is the case the direct gamma-ray transition to the ground state will be very difficult to measure by conventional methods because of the low intensity. The remaining alternative is the existence of bremsstrahlung due to the 3-Mev beta-rays.

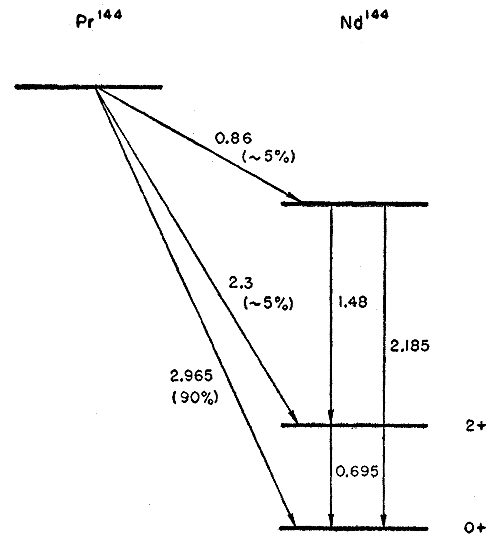

FIG. 5. Proposed decay scheme of Pr^{144}.

Since the relative number of 3-Mev betas and 2.2-Mev gammas is about 30 to 1 on the basis of the proposed decay scheme and since the photoneutron work[1] indicates a ratio of approximately 650 in the number of gammas below 2.23 Mev to those above this energy (the photoneutron measurements do not give directly the relative numbers of gamma-rays), then it may be estimated that the number of gammas above 2.23 Mev per beta-disintegration is about 5×10^{-5}. Inasmuch as no allowance has been made for the relative (γ,n) cross sections in Be and D_2O above 2.3 Mev, the photoneutron yield observed from D_2O might be a reasonable consequence of bremsstrahlung.

The authors are indebted to Dr. Seymour Katcoff for chemical purification of the beta-source and to Dr. M. Goldhaber for suggestions and discussions of this problem.

[16] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[17] L. W. Nordheim, Phys. Rev. **78**, 294 (1950).
[18] M. Goldhaber, private communication.