The Disintegration of Ce^{144} and Pr^{144}†

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The radiations from the radioactive chain $Ce^{144}\rightarrow Pr^{144}\rightarrow Nd^{144}$ have been investigated, using chemically purified sources. Ce^{144} (295-day half-life) decays to the ground state of Pr^{144} through a 70 percent abundant beta-group having a maximum energy of 304±2 kev. Conversion electrons have been observed which indicate that within the praseodymium nucleus there are transitions having energies 33.7, 53.5, 80.7, 100.3, and 134.2 kev. The predominant number of the Pr^{144} beta-transitions go directly to the ground state of Nd^{144} through a 2.97±0.01 Mev beta-group. Conversion electrons from a weak 60.3-kev transition in neodymium have been observed. Very weak gamma-radiation having energies 0.696, 1.5, and 2.185 Mev have been observed. The beta-spectrum indicates that less than 2 percent of the beta-transitions go to excited states of neodymium at 0.696 and 2.185 Mev. Using the evidence available a disintegration scheme has been proposed.

I. INTRODUCTION

RADIOACTIVE cerium, having a half-life[1] of approximately 300 days and a mass[2] number 144, has been observed to decay through beta-emission to Pr^{144}, which in turn decays by beta-emission[3] to stable Nd^{144}, the half-life of this isotope being reported as 17.5 minutes.

The most recent works[4–7] involving these isotopes indicate gamma-ray transitions of low intensity to be associated with the decay of both. There still seems to be no general agreement[4–8] as to the relative intensities of the beta- and gamma-radiations associated with this radioactive chain.

It seems, however, to be generally conceded that the great majority of beta-transitions are of the ground-state to ground-state type for both isotopes. Since both Ce^{144} and Nd^{144} are even-even nuclei, it would seem plausible that both should have a spin zero. Therefore, it would appear that the comparative half-lives of the ground-state to ground-state transitions of both these beta-transitions might be expected to be approximately equal one to the other. This is not the case at present.[9]

This unsettled state of knowledge concerning the radioactive chain $Ce^{144}\rightarrow Pr^{144}\rightarrow Nd^{144}$ seems sufficient reason to investigate its radiations.

II. APPARATUS

The chief piece of apparatus used in the investigation of the beta-spectra has been the semi-circular uniform field magnetic spectrometer having a radius of curvature of 14 cm.[10] A new baffle system has been built into the spectrometer, this system incorporating the results of the information found earlier[11] regarding spectrometer baffle systems. To further reduce scattering at the source a fork type source holder has replaced the former ring type holder.

The source was deposited onto an aluminum ribbon having a surface density of 1.5 mg/cm² and was defined by a prior deposition of insulin in the customary manner.[12] The surface density of the source was approximately 0.5 mg/cm².

III. RESULTS

The lower energy portion (below 300 kev) of the electron spectrum is shown in Fig. 1. The underlying beta-spectrum is primarily that of Ce^{144}. In addition, there are several prominent internal conversion lines which are summarized in Table I. The energies and K/L conversion ratios are used in the discussion to prepare a tentative disintegration scheme.

In studying the beta-spectrum four sets of data were obtained from a single source; each set was completed within a period of less than three days, such that any uncertainty in half-life of Ce^{144} contributed negligible error. The four sets were distributed over a period of 70 days. Incidentally, our data are consistent with a half-life of 295 days. An F-K analysis of that set of data showing the largest positive deviation from a straight line is given by Fig. 2. It is not difficult to draw a straight line which apparently fits the experimental data between 500 kev and the end-point energy at 2.97 Mev. However, Alburger *et al.*[5] have reported beta-groups at 2.3 and 0.86 Mev, and Langer and Moffat[6] at 0.78 Mev. To obtain an unbiased result, a least-square fit was made of our F-K analysis data between 2.3 Mev and the end-point energy. When the line so obtained is extended toward lower energies, three of the four aforementioned observations show a

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[5] Alburger, der Mateosian, Goldhaber, and Katcoff, Phys. Rev. **82**, 332 (1951), and private communication at New York Meeting, 1952.
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[7] F. T. Porter and C. S. Cook, Phys. Rev. **85**, 733 (1952).
[8] Chen, John, and Kurbatov, Phys. Rev. **85**, 487 (1952).
[9] A. M. Feingold, Revs. Modern Phys. **23**, 10 (1951).
[10] Ter-Pogossian, Cook, Porter, Morganstern, and Hudis, Phys. Rev. **80**, 360 (1950).
[11] G. E. Owen and C. S. Cook, Rev. Sci. Instr. **20**, 768 (1949).
[12] L. M. Langer, Rev. Sci. Instr. **20**, 216 (1949).

slight excess of beta-particles below 2.3 Mev. However, one set of experimental results actually falls below the least-squares line in the energy region below 2.3 Mev. At least 10,000 real counts were taken for each experimental point so that the statistical error will always be less than one percent.

If it is assumed that the two groups reported by Alburger *et al.* are actually present in the decay of Pr^{144}, our data indicate that the maximum possible abundance of the 2.3-Mev group relative to the 2.97-Mev group is 1.0 percent and the maximum possible relative abundance of the 0.78-Mev group is also 1.0 percent. These are considerably lower than the values of approximately 5 percent for each quoted by Alburger,[5] especially when it is considered that the values we quote are the maximum values our data will allow. Without the prior determinations by Alburger we would probably consider them as nonexistent.

In view of the fact that the energies of the proposed weak beta-groups are quite close to those of the decay chain $Sr^{90}\rightarrow Y^{90}\rightarrow Zr^{90}$, a special chemical search was made for us by Dr. Wahl and Mr. Mills of the radiochemistry laboratory. Their results indicate less than one part in 10^4 of radioactive strontium to radioactive cerium in the final source material.

The 300-kev beta-group, associated with the decay of Ce^{144}, is found to be 70 percent as abundant as the 2.97 Mev group. Its F-K plot is linear down to an energy of about 170 kev. However, a measure of the relative abundance of the 170-kev group and any other lower energy groups is impossible because of masking by the extensive number of internal conversion electrons at the lower energies.

A photoelectron spectrum from a uranium radiator clearly shows the L and M lines of the 134.2-kev gamma-ray and the K-lines of the 0.696 and 2.185-Mev gamma-

TABLE I. Internal conversion electrons in the beta-spectrum of $Ce^{144}\rightarrow Pr^{144}\rightarrow Nd^{144}$.

Electron energy[a] kev	Transition energy[a] kev	Transition in Pr or Nd?	Relative abundance[b]	K/L ratio
26.9	L_I	33.7±0.3	(Pr?)[c]	see d	
32.0	M_I				
46.6	L_I	53.5±1	(Pr?)[c]		
see e	M_I				
16.7	K				
53.1	L_I	60.3±0.3	Nd	see f	see f
58.8	M_I				
38.7	K				
74.1	L_I	80.7±0.5	Pr	1.9%[g]	3.5[g]
79.2	M_I				
58.0	K				
93.8	L_I	100.3±0.5	(Pr?)[c]	see h	see h
98.6	M_I				
92.2	K				
127.3	L_I	134.2±0.5	Pr	6.1%	8.8
133.0	M_I				
29.2±0.5	see i		Pr		
34.0±0.5	see j		Pr		

[a] Binding energies of the praseodymium and neodymium shells were taken from "A Table of Critical X-ray Absorption Edges" by Lewis Slack of the Naval Research Laboratory (private communication).
[b] The relative abundance referred to in this case is the ratio of intensity of all internal conversion electrons of the appropriate transition to the intensity of the 2.97-Mev beta-ray group.
[c] Presumably in praseodymium—appropriate electron lines not sufficiently distinct to distinguish between Pr or Nd.
[d] Greater than 0.25 percent. The lines are too near the spectrometer window cut-off energy to allow an accurate measurement.
[e] The M line of this transition is masked by the L of the 60.3-kev transition. The slightly greater width at half-maximum of the L-conversion line of the 60.3-kev transition, however, indicates its presence.
[f] The K-conversion line is too near the cut-off energy of the spectrometer counter window to give an accurate estimate of either of these.
[g] Both values probably slightly low because of effect of diminution in intensity by counter window.
[h] L line masked by K line of 134.2-kev transition. Approximate values are 0.25 percent and 3, respectively.
[i] K-2L Auger electron line.
[j] K-L-M Auger electron line.

FIG. 1. The electron spectrum below 300 kev. The arrows indicate the expected position of the internal conversion electrons from the indicated shells for the indicated transition energies. In all cases the L_I and M_I energies have been used. All energies are in kev.

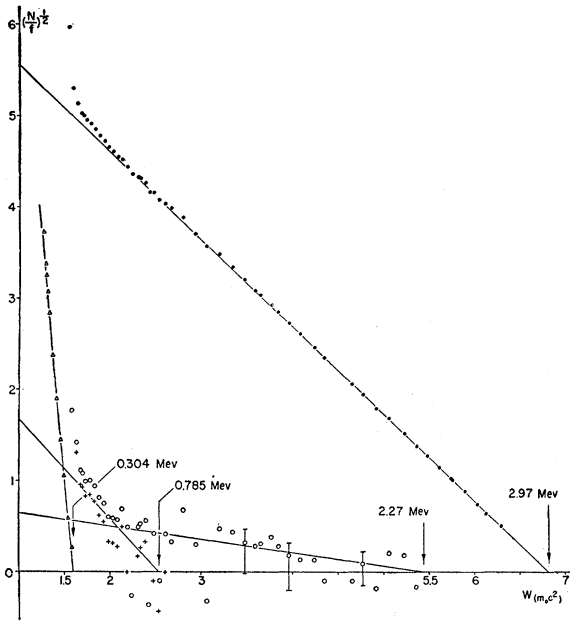

FIG. 2. F-K analysis of that set of data showing the largest excess of electrons in the region between 300 kev and 2.3 Mev. The initial straight line was obtained by a least squares fit on the experimental points between 2.3 Mev and the end-point energy (2.97 Mev). The solid circles are the experimental points; the open circles are the first extrapolation (2.3-Mev group); the crosses are the second extrapolation (0.780-Mev group); the triangles are the third extrapolation (0.304-Mev group). In order that it can be shown on the same diagram, the ordinate for the abundant 0.304-Mev group in Ce^{144} has been reduced by a factor of $\frac{1}{5}$. The statistical errors for the closed circles and the triangles are of the order of magnitude of the size of the symbols. Samples of the statistical errors for the open circles and crosses are as indicated. The end-point energies for the 0.780- and 2.3-Mev groups were determined in accordance with the available gamma-ray information.

rays. The reported 1.48-Mev radiation's photoelectron lines were not distinctly visible. To obtain these results required a source of several millicuries and to improve significantly the results would have required considerably stronger sources. Since the chemical procedure deemed adequate resulted in a yield of about 20 percent, the amount of initial radioactive material required (greater than 100 mC) is more than we are prepared to handle at present. However, a scintillation spectrometer has confirmed the existence of the 0.7, 1.5, and 2.2-Mev gamma-rays in all our sources.

IV. DISCUSSION

The large variety of results which have been reported in the literature for the decay chain $Ce^{144} \rightarrow Pr^{144} \rightarrow Nd^{144}$ makes it appear that one of the most important items which must be considered is that of possible impurities within the source. A first source was prepared and underwent a rather extensive chemical separation. The results obtained from it have been reported[7] briefly at the 1951 Chicago meeting. Because of discrepancies in results between the various groups who had at that time being examining this chain, a new source was prepared which underwent several more steps in the chemistry than did the first source. All beta-spectral results reported here were obtained from the second source. The photoelectron spectrum, however, was from the first source.

The procedure for the second chemistry, as performed for us by Dr. A. C. Wahl and Mr. R. Mills of the radiochemistry laboratory, follows:

To the original solution of $Ce(Cl)_3$, 3 mg of Ce(III), 10 mg of La(III), and 20 mg of Sr(II) were added. The hydroxides of Ce and La were precipitated, the Sr acting as a holdback carrier. The hydroxides were dissolved in HBr and 10 mg Te(VI) and 20 mg Sr(II) carriers were added. The fluorides were precipitated with Sr and Te remaining in solution. The fluorides were dissolved in $HClO_4$ and 5 mg Ru(III) added. RuO_4 was fumed off. The solution was diluted, Sr holdback carrier added and the hydroxides again precipitated.

The foregoing was repeated except that no additional Ce or La carrier was added.

The hydroxides were dissolved in HNO_3 and 5 mg Zr(IV) added. Zirconium iodate was precipitated with HIO_3 from an approximately 4 *M* acid solution. In addition to $Zr(IO_3)_4$, $Th(IO_3)_4$ should be carried with this precipitation. This precipitation was repeated with an additional 5 mg of Zr(IV).

The hydroxides were precipitated, dissolved in HNO_3 and the Ce(III) oxidized to Ce(IV) with BrO_3^-. $Ce(IO_3)_4$ was precipitated with HIO_3 in approximately 4.5 *M* acid solution, leaving La and the rare earths in in solution. The precipitate was redissolved in HCl, reduced to Ce(III) with H_2O_2, more La carrier added, Ce(III) reoxidized with BrO_3^-, and again $Ce(IO_3)_4$ precipitated. The sequence of the previous sentence was carried out four times.

The final $Ce(IO_3)_4$ precipitate was redissolved, reduced to Ce(III) with HSO_3^- and reprecipitated as the hydroxide, which was then dissolved in a few drops of dilute HNO_3. The spectrometer source was prepared from this solution.

The beta-groups reported here have all been reported at some prior time by some other groups of investigators. However, our maximum possible abundance for the 0.8-Mev and 2.3-Mev groups are distinctly less than the equivalent percentages reported by Alburger *et al.*[5] In fact, had the 0.7-, 1.5-, and 2.2-Mev gamma-radiation not been observed by means of a scintillation spectrometer in this laboratory, we would have had considerable doubt as to the real existance of such beta-groups. However, it now appears that they do exist, but with an abundance in each case of the order of one percent or less of the 2.97-Mev group. With this interpretation of the F-K plot we no longer observe the 450-kev beta-group, reported at the Chicago meeting.[7] Also, it is not possible from our data to obtain the intermediate groups of the intensity or have the energies reported by Cheng, John, and Kurbatov.[8]

With regard to gamma-radiation, we can find no indication, whatsoever, of the 231-kev group reported by the Ohio State group.[8] A careful and thorough study of the region in which the internal conversion lines of this group should appear reveals nothing but the continuous beta-spectrum, as illustrated in Fig. 1.

We are also unable to find any conclusive evidence for a 95-kev group reported by Keller and Cork.[4] The K-conversion electrons from a 95-kev transition in praseodymium would occur at an energy differing by only 0.1 kev from the L-conversion line of the 60.3-kev transition. However, L-conversion electrons from this 95-kev transition would have an energy differing by 4 kev from the K-conversion electrons of the 134.2-kev transition. We detect no fine structure in the 134.2-kev line. Also, we find no evidence for a 46.8-kev transition[4] and assign the electron line at 34.0 kev to the K–L–M Auger electrons rather than to the L of a 41.3-kev transition.

The electron line whose energy is slightly greater than 58 kev may logically be assigned as a combination of the K of the 100.3-kev transition and the M of the 60.3-kev transition, since its intensity is larger than would be expected if it were only the M of the 60.3-kev transition.

Making use of the information available the disintegration scheme of Fig. 3 seems plausible. The arrangement and approximate abundances of the chief beta-groups has been explained earlier. The assignment of the gamma-rays, however, requires some explanation.

The 134.2-kev gamma-ray transition appears comparatively strong both in the photoelectron and in the internal conversion spectra. Its experimentally determined K/L internal conversion intensity ratio is 8.8. Using the empirically determined values of Goldhaber and Sunyar,[13] this ratio would lead to the supposition that the 134.2-kev radiation is magnetic dipole. Goldhaber and Sunyar give 7.8 for the $M1$ type of transition. Following the example of Goldhaber and Sunyar an extrapolation to lower energies of Rose's values for the K-conversion coefficient for Pr ($Z=59$) gives $\sim$0.5 at 134.2 kev for an $M1$ transition. Considering the definite appearance of the 134.2-kev group in the photon spectrum in some strength makes this value reasonable. For no other type transition does Goldhaber's empirically determined K/L ratio and Rose's theoretically determined K-conversion coefficient give values which fit so well with our experimental observations. It, therefore, seems consistent with known facts to consider the 134.2-kev transition to be of the magnetic dipole type. With the K-conversion peak being 5.8 percent as abundant as the Pr^{144} 2.97-Mev beta-group, these values would give the 134.2-kev transition an abundance of approximately 18 percent, with an error of several percent.

On the other hand, the 80.7-kev transition appears to be quite highly converted. Its measured K/L ratio is 3.5, which, if anything, is slightly small because of the approach of its K line toward the low energy cutoff of the spectrometer counter window. The best fit for this transition is an $M2$ type whose extrapolated K conversion coefficient would be expected to be about 10 and whose empirical K/L internal conversion ratio is 5.0. The $E1$ type whose K/L ratio is close to the experimental value has too low a K-conversion coefficient.

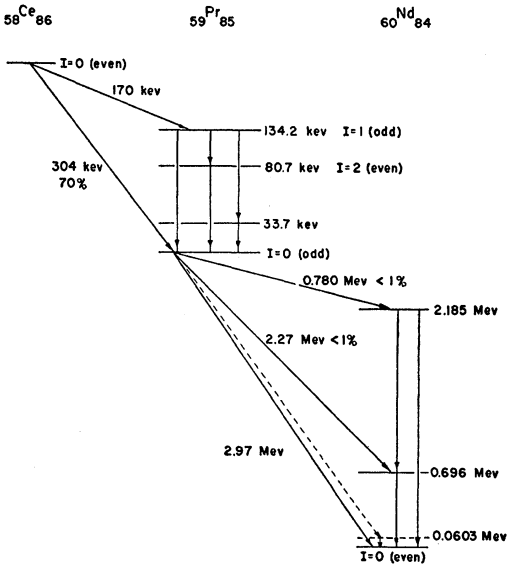

FIG. 3. Proposed decay scheme. The order of emission of the 33.7- and 100.3-kev gamma-rays in praseodymium is arbitrary. The weak 60.3-kev transition is assigned to neodymium, but its position in the disintegration scheme is very tentative.

The K/L-conversion coefficient ratios of the other gamma-transitions have not been determined sufficiently well at this date to establish their transition type.

This information is, however, sufficient to assign spins and parities to two of the Pr^{144} excited states. The assignment of 0 (even) to the ground state of Ce^{144} and Nd^{144} is based upon the fact that they are even-even nuclei. The assignment of 0 (odd) to the ground state of Pr^{144} is made from considerations of Nordheim's empirical rules for the beta-disintegration of even A nuclei.[14] With this assignment, the magnetic dipole character of the 134.2-kev transition and the magnetic quadrupole character of the 80.7-kev transition makes assignment of 2 (even) and 1 (odd) to the 80.7- and 134.2-levels, respectively, appear rather obvious. This makes the Ce^{144} beta-transition to the 134.2-kev level first-forbidden, as well as the Ce^{144} transition to the Pr^{144} ground state, such that they may have abundances of the same order of magnitude. A transition from Ce^{144} to the 80.7-kev level must, however, be second-forbidden and would not be expected to be observed. Nothing can be said at present about the 33.7-kev level (or the

[13] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[14] L. W. Nordheim, Phys. Rev. **78**, 294 (1950); Revs. Modern Phys. **23**, 332 (1951).

100.3-kev level if their order of decay is reversed) because of the uncertain state of knowledge concerning the K/L internal conversion intensity ratio for the 100.3-kev transition or the conversion coefficients of either transition.

The fact that the 60.3-kev transition belongs to Nd^{144}, as proposed by Keller and Cork,[4] is verified by our results. The observed intensity, however, appears insufficient for it to follow a large fraction of the Pr^{144} betas. It is, however, possible for it to represent a transition to the ground state of Nd^{144} from a level 60 kev above the ground state level without changing the shape of our spectra. A calculation of the F-K plots resulting from the addition of two spectra whose end-point energies are 2.97 and 2.91 Mev, respectively, shows no distinct change from a straight line except in the region in the immediate vicinity of the end-point energy. This is the result regardless of the relative abundance of the two groups. Our investigations of the beta-spectra, therefore, are not inconsistent with such a scheme.

The present investigations indicate that the difference in ft values for ground-state to ground-state transitions is real. For the disintegration scheme proposed here the values are such that $\log ft=7.43$ for the Ce^{144} ground-state transition (assuming 70 percent abundance), $\log ft\geqslant 7.03$ for Ce^{144} decay to the 134.2-kev excited state of Pr^{144} (assuming $\leqslant 30$ percent abundance), and $\log ft=6.53$ for the Pr^{144} decay (assuming 98 percent ground-state to ground-state transition). The 60-kev transition does not appear to have sufficient total intensity to alter the latter value appreciably.

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The Electrical Conductivity in the Solar Atmosphere

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The electrical conductivity in the solar atmosphere is computed from the physical properties of the solar material. It is shown that as a consequence of the high abundance of hydrogen the mean free paths of the free electrons are determined by collisions with neutral hydrogen atoms and not by collisions with the much less abundant ions. Furthermore, for magnetic fields less than about 750 gauss the curvature of the trajectories of the free electrons is small enough for the ordinary Ohm's law to be a good approximation.

THE conditions under which convection can begin in a thermally unstable fluid that is a good electrical conductor have recently been studied by Chandrasekhar.[1] He finds that the presence of a magnetic field has a large effect in inhibiting the onset of convection and that the higher the electrical conductivity of the fluid is, the larger this effect is. One of the proposed applications of this theory is to the sun and to sunspots, and in this connection values of the electrical conductivity of the solar atmosphere are needed. The present paper contains calculations of these values.

It is well known that the electrons in a gas consisting of a mixture of electrons, ions, and neutral atoms, are responsible, on account of their small masses and consequently large velocities, for carrying practically all the current. Thus in finding the electrical conductivity of such a gas one is essentially finding the mean free paths of the electrons.Values of the conductivity of the solar atmosphere have been computed in the past by using the formulas derived for the case of an ionized gas, in which the mean free path of the electrons is set by the Coulomb scattering cross section of the positive ions. Now in the solar atmosphere the number of positive ions is relatively small, but the number of neutral hydrogen atoms is very large, so that in spite of the very large Coulomb cross sections of the ions, the effect of scattering by neutral hydrogen atoms may be expected to be of some importance. The elastic scattering cross section of a hydrogen atom is rather large at the low velocities of importance in the solar atmosphere and in fact the calculations summarized below show that the neutral hydrogen atoms reduce the mean free path of the electrons and hence the electrical conductivity by a factor of at least ten from the case in which only Coulomb scattering is taken into account.

To compute the electrical conductivity for electrons scattered by neutral hydrogen atoms, we shall use the formalism developed by Chapman and Cowling.[2] Collecting the formulas from their book (the notation is somewhat simplified here), we can reduce the calculation of the conductivity σ in the first approximation to the

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[1] S. Chandresekhar, Phil. Mag. (to be published).

[2] S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-uniform Gases* (Cambridge University Press, Cambridge, 1939). The expression for the conductivity is given in Eq. 18.11, 5 of this book and all the other equations may be found by tracing the definitions of the various symbols in this equation.