value, based on the intensity of the *F*-line relative to that of the parent beta spectrum, is 0.71.[16]

With the lens spectrometer and a thin carbon target, backed with a 20.2-mg/cm^2 thorium converter, a search was made for the gamma lines representing transitions to the 3.1-Mev level. The search was unsuccessful, and their intensities are estimated at less than three percent of those of the ground state transitions.

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Electron Capture and Alpha Decay of Np^{235}†

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The electron capture and alpha decay of Np^{235} have been investigated. An L/K capture ratio of 30 ± 2 is found from absolute L and K x-ray measurements. No gamma rays are observed in the electron capture decay of Np^{235}. An alpha branching ratio of $(3.5\pm0.4)\times10^{-5}$ was measured for this nuclide. The alpha particles have coincident 26- and 85-kev gamma rays with an intensity of 0.054 ± 0.005 gamma per alpha disintegration for the 85-kev gamma ray. The intensity of the 26-kev gamma ray is comparable, although somewhat more uncertain.

I. INTRODUCTION

THE nuclide Np^{235} was originally discovered as a product of deuteron irradiations of U^{235}.[1] In later work by James *et al.*,[2] a half-life of 410 days was measured for this nuclide, exhibiting predominantly electron capture decay with an L/K capture ratio of >9. A slight alpha branching (5×10^{-5}) was also reported with an alpha-particle energy of 5.06 Mev. The present report concerns a more detailed study of the decay of this nuclide using a scintillation crystal spectrometer and a proportional counter in connection with a multichannel pulse-height analyzer. Both gamma-gamma and alpha-gamma coincidence techniques were used in a study of the decay scheme.

II. EXPERIMENTAL METHODS AND RESULTS

The Np^{235} used in the present study was produced by the $(d,2n)$ reaction in a cyclotron bombardment of high-purity U^{235} (99.9+%) with 18-Mev deuterons. The target received a total of 2489 microampere hours of intermittent irradiation over a period of three months. After dissolution of the uranium foil in hydrochloric acid, the neptunium fraction was separated and purified by repeated hydroxide and fluoride precipitations with lanthanum carrier added, anion exchange column elutions using Dowex-1 resin[3] in a hydrochloric acid medium, and solvent extraction of the neptunium using thenoyltrifluoroacetone (TTA) dissolved in benzene.[4] The final purification was performed after waiting a sufficient length of time to insure the complete decay of shorter-lived neptunium isotopes (Np^{234}, Np^{236}) present in the original sample. An aliquot of the final preparation was followed for decay in a 4π counter as a check on radiochemical purity.

The relative intensities of the K and L x-rays resulting from the electron-capture decay of Np^{235} were measured using both a sodium iodide (Tl-activated) crystal and a xenon-filled proportional counter as detectors. The initial pulse, either from the phototube attached to the crystal or directly from the proportional counter was introduced into a 50-channel differential pulse-height analyzer, after being amplified in a preamplifier and linear amplifier. The 60-kev gamma ray of Am^{241} was used to calibrate sample geometry for the sodium iodide crystal. Knowing the dimensions of the proportional counter and the pressure of the xenon gas within the counter, it was possible to calculate the sample geometry and counting efficiencies for the L x-rays of the Np^{235} sample. The counting efficiencies of the L_α, L_β, and L_γ x-rays of uranium were 53.7%, 34.0%, and 23.5%, respectively. The calculated geometry and counting efficiencies were experimentally checked using the L x-rays of an Am^{241} standard.[5] The K and L x-ray spectra of Np^{235} are shown in Fig. 1. From the absence of

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] James, Florin, Hopkins, and Ghiorso, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV.

[2] James, Ghiorso, and Orth, Phys. Rev. **85**, 369 (1952).

[3] Dow Chemical Company, Midland, Michigan.

[4] E. K. Hyde, in *The Actinide Elements*, edited by G. T. Seaborg and J. J. Katz (McGraw-Hill Book Company, Inc., New York, 1954), Chap. 15, National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV.

[5] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952) and Phys. Rev. **87**, 1144 (1952).

detectable gamma rays, it is concluded that the majority (99+%) of the electron capture decay of Np^{235} does not populate an excited level of U^{235} but rather proceeds directly to the ground state. L x-ray—K x-ray coincidences were measured using the proportional counter and sodium iodide crystal detectors in conjunction with a coincidence circuit of 2×10^{-6}-sec resolving time. A single-channel pulse-height analyzer was used to discriminate against radiation other than the K x-ray gates, while a 50-channel differential pulse-height analyzer was used to analyze the energy of the coincident L x-ray radiation. The relative K and L x-ray intensities for Np^{235} are listed in Table I, these intensities having been corrected for sample geometry and counting efficiency in the detector. After subtracting the L x-rays arising from the filling of a vacancy in the K shell (i.e., those L x-rays in coincidence with the K x-rays), an L/K capture ratio of 30±2 was calculated using a fluorescence yield of 98% for the K shell[6] and an over-all fluorescence yield of 42% for the L shells.[7]

The energy available for the electron capture of Np^{235} has been estimated with the aid of closed-decay cycles[8] to be 0.170 Mev. A search was made for the continuous gamma spectrum of Np^{235} in both the singles spectrum and in coincidence with K x-rays. However, the low decay energy and expected low intensity[9] caused the continuous gamma spectrum to be obscured by the other radiations of the sample.

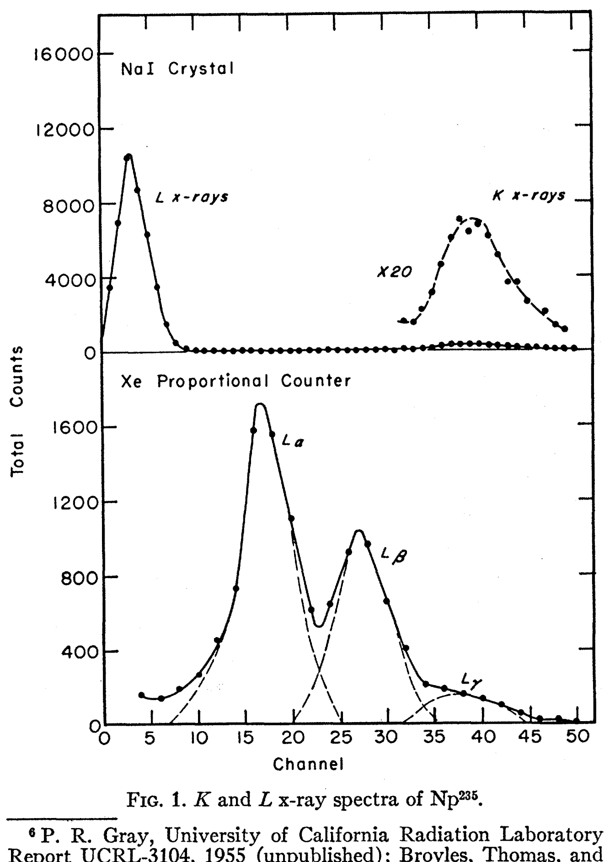

FIG. 1. K and L x-ray spectra of Np^{235}.

TABLE I. Relative K and L x-ray intensities for the electron-capture decay of Np^{235}.

X-ray	Energy (kev)	Relative intensities	Coincident with K x-rays
K	98	762±12	...
L_α	13.4–13.7	4600±70	106±6
L_β	16.4–17.5	4240±70	89±6
L_γ	20.1–20.9	1080±50	12±3

Alpha-particle pulse analysis revealed that the sample contained, in addition to the 5.06-Mev alpha particles of Np^{235}, 4% Np^{237} alpha activity which was probably formed during the cyclotron bombardment by the $(d,3n)$ reaction on the U^{238} impurity in the target. The alpha branching ratio of Np^{235} was redetermined by comparison of the alpha counting rate in an argon counter of known efficiency and the electron capture disintegration rate as measured by the absolute K and L x-ray intensities. A branching ratio of $(3.5\pm0.4)\times10^{-5}$ was found as compared with a value of 5×10^{-5} measured previously.[2]

The gamma spectrum in coincidence with the alpha particles of the sample was measured using a zinc sulfide screen to detect the alpha particles, a sodium iodide crystal detector for the gamma rays, and a fast-slow coincidence apparatus of 7×10^{-8}-sec resolving time. A 50-channel differential pulse-height analyzer was used to analyze the energy of the coincident radiation. Gamma rays of 26 and 85 kev and L x-rays were observed to be in coincidence with the alpha particles of the sample.[10] The over-all half-life of the coincident radiation was measured to be $(3.7\pm0.4)\times10^{-8}$ sec (Fig. 2). This represents a composite half-life due to a contribution from the 30- and 87-kev gamma rays associated with the alpha decay of Np^{237}.[11,12] Correcting for the 87-kev gamma ray in Np^{237}, the intensity of the 85-kev gamma ray is 0.054±0.005 gamma per alpha disintegration. The intensity of the 26-kev gamma ray is comparable, although somewhat more uncertain.

III. CONCLUSIONS

The L/K capture ratio, which is relatively easy to determine in Np^{235} decay since there are no gamma rays involved, can be used to calculate the electron-

[6] P. R. Gray, University of California Radiation Laboratory Report UCRL-3104, 1955 (unpublished); Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).

[7] B. B. Kinsey, Can. J. Research **26A**, 404 (1948); B. L. Robinson and R. W. Fink, Revs. Modern Phys. **27**, 424 (1955).

[8] Glass, Thompson, and Seaborg, J. Inorg. and Nuclear Chem. **1**, 3 (1955).

[9] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).

[10] These same gamma rays have been observed in the beta decay of Th^{231}. Their half-life is reported to be $(4.1\pm0.4)\times10^{-8}$ sec by D. Strominger and J. O. Rasmussen, Phys. Rev. **100**, 844 (1955).

[11] D. W. Englekemeir and L. B. Magnusson, Phys. Rev. **94**, 1395 (1954).

[12] F. S. Stephens, Jr., University of California Radiation Laboratory Report UCRL-2970, 1955 (unpublished).

capture decay energy.[13] At very low decay energy (just above the K edge of the element) the L/K capture ratio is very sensitive to a change in decay energy. For the experimentally observed L/K ratio of 30 ± 2, the Np^{235} decay energy is calculated to be 0.130 Mev. This decay energy is to be compared with the closed-decay-cycle estimate of 0.170 Mev,[8] corresponding to an L/K capture ratio of 1.5 which is far outside of the experimental error on the measured ratio. The closed-decay-cycle estimate of the decay energy is considered to be 40 kev too high. An uncertainty of this magnitude is not unexpected in the closed-cycle estimates.

The value of log ft for the electron capture decay of Np^{235} is 6.5. This most probably indicates first-forbidden decay, $\Delta I=0$, 1, yes.[14] The spins of U^{235} and Np^{237} have been measured to be 7/2[15] and 5/2,[16] respectively. From the shell model the nuclide Np^{235} might also be expected to have spin 5/2 and to have opposite parity (i.e., odd parity) to that of U^{235}. Thus, the assignment of $\Delta I=0$, 1, yes for the decay of Np^{235} is consistent with the present data on spins in this region.

From a correlation of data available on relative abundances of L x-rays in the region of $Z=92$,[17] it is found that L_α x-rays arise predominantly from the filling of L_{III} shell vacancies, while L_β and L_γ x-rays are produced from the filling of a combination of L-shell vacancies. As first noted by James *et al.*,[2] the relative L x-ray intensities associated with the L capture of Np^{235} indicate a large fraction of L_{III}-shell vacancies. Now for allowed or first-forbidden, $\Delta I=0$, 1 electron capture transitions, capture from the L_I shell is predominant with approximately 10% capture from the L_{II} shell and a negligible contribution from the L_{III} shell.[13] Thus, any L_{III} shell vacancies observed must arise from Coster-Kronig type transitions.[18] Although the uncertainties of the individual L-shell fluorescence yields for uranium preclude a detailed analysis of the Np^{235} data, it can be said that a large fraction of the vacancies in the L_I shell due to capture are filled by L_{III}-shell electrons in Coster-Kronig type transitions. This is in agreement with the results of Kinsey[7] and Ross *et al.*[19]

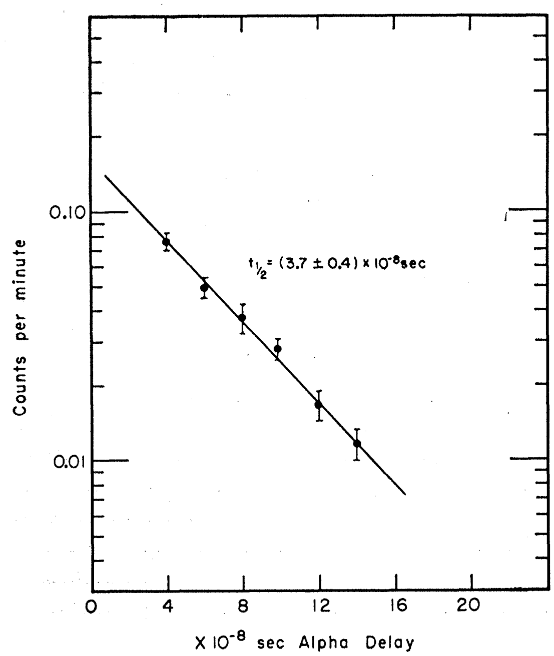

FIG. 2. Half-life of 26- and 85-kev gamma rays in coincidence with alpha particles of Np^{235}.

The partial alpha half-life for Np^{235}, calculated from the experimentally measured alpha branching ratio, is $(3.2\pm0.3)\times10^4$ years. Although there are not sufficient data to comment on a scheme for the alpha decay of Np^{235}, the nuclide is similar to a number of odd-mass, odd-proton alpha emitters in this region for which a large number of $E1$ gamma transitions have been observed, many of these having measurable half-lives which are much longer than would be expected from the predictions of the Weisskopf formula.[12]

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[13] R. E. Marshak, Phys. Rev. **61**, 431 (1942); R. W. Hoff and J. O. Rasmussen, Phys. Rev. **101**, 280 (1956); H. Brysk and M. E. Rose, Oak Ridge National Laboratory Report ORNL-1830 (unpublished).

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[16] J. E. Mack, Revs. Modern Phys. **22**, 64 (1950).

[17] Jaffe, Passell, Browne, and Perlman, Phys. Rev. **97**, 142 (1955); Paul Day, Phys. Rev. **97**, 689 (1955); H. Jaffe, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-2537 (unpublished); C. I. Browne, Jr., Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1764 (unpublished).

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