

by a spin zero nucleus.^{12, 13}

$$(\sigma_{\max})^{\frac{1}{2}} - (\sigma_{\min})^{\frac{1}{2}} = (J + \frac{1}{2}) \lambda P_L(\cos\theta).$$

Table II shows that the 465-kev level is best assigned $J = \frac{1}{2}$. Some loss in scintillation counter efficiency would account for the smaller experimental intensity. Since only the assumption of $L=0$ gives a level "width without barrier" of reasonable value,¹⁴ the level is probably ${}^2S_{\frac{1}{2}}$. Further, the gamma-radiation intensity from the corresponding level in C^{13} to its ground state, presumed to be ${}^2P_{\frac{1}{2}}$, appears to be electric dipole radiation.⁵ The data for the 1.7-Mev level¹⁵ show an intensity midway between $J=3/2$ and $J=5/2$. However, the small concomitant maximum at 1.61 Mev and minimum at 1.83 Mev may be the result of a second level in this neighborhood, and the single level intensity analysis may not be valid. §

¹² C. L. Critchfield and D. C. Dodder, Phys. Rev. **76**, 602 (1949).

¹³ This extension of the formulas in reference 12 is due to R. A. Laubenstein.

¹⁴ Fowler, Lauritsen, and Lauritsen, Revs. Modern Phys. **20**, 236 (1948).

¹⁵ From comparison of the calculated level shift between the levels in N^{13} (3.5 Mev) and C^{13} (3.7 Mev) with the experimental value, R. G. Thomas concludes that a p wave should be assigned here (private communication).

§ Note added in proof.—Preliminary results of an analysis by H. L. Jackson and A. I. Galonsky (to be published) show that a

It is apparent that a more reliable absolute cross section would be of value in the assignment of level configurations. The cross-section measurement could be improved by extending the present data below 300 kev into a region of pure Rutherford scattering. Another possibility is the use of a methane gas target and a proportional counter for both absolute cross section and angular distribution measurements. Angular distributions, taken at the two resonances, would help further in assignment of angular momenta.¹⁶

We wish to thank Professor H. T. Richards for suggesting this problem to us and for many helpful discussions. We are also indebted to R. A. Laubenstein, H. L. Jackson, and M. J. W. Laubenstein for aid in taking data.

P (1.68-Mev) level and a $D_{\frac{3}{2}}$ (1.73-Mev) level of comparable widths account for the shape near resonance. For the corresponding level of the mirror nucleus C^{13} , J. Rotblat (Proc. of the Harwell Nuclear Physics Conference, 1950) reports finding a doublet in place of the 3.7-Mev level, from the $C^{12}(d,p)C^{13}$ reaction. He gives the values 3.77 and 3.90 Mev for the two components. See also a forthcoming paper by R. G. Thomas on this subject.

¹⁶ W. D. Whitehead, Phys. Rev. **79**, 1022 (1950), has measured the angular distribution of elastically scattered protons from carbon for bombarding energies between 2 and 2.75 Mev. The data indicate the scattering of both s and p waves.

Radiations of UY

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(Received January 10, 1951)

The radiations of UY have been examined by absorption curve techniques. Among the radiations are: (1) 0.2-Mev beta, (2) L x-rays, (3) 35-kev gamma-ray, (4) soft electronic component, probably conversion electrons about 50 kev, (5) soft quantum component, probably M x-rays, and (6) two harder gamma-rays, 65 to 75 kev and about 100 kev. On abundance and coincidence considerations, a tentative decay scheme is suggested which includes a 0.2-Mev beta followed by 35- and 65-kev gamma-rays in cascade with a 100-kev cross-over transition. The half-life was found to be 25.64 ± 0.1 hours.

I. INTRODUCTION

IN the course of a cross-section measurement¹ on the reaction $Th^{230}(n,\gamma)Th^{231}$ by direct determination of the Th^{231} (or UY) disintegration rate, it was found necessary to have some information on the UY decay scheme. Such information was necessary in order to evaluate the fraction of the counts measured which were due to disintegration betas. Some preliminary work has been done on the decay scheme, by the use of absorption methods. The poor resolution of these methods enabled the determination of only the gross features of a decay scheme, which were, we believe, sufficient for the purposes of evaluating disintegration rates (with the aid of suitable corrections). Our results indicate that further work is necessary to determine the correct scheme.

As indicated above, UY can be made through the

¹ A. H. Jaffey and E. K. Hyde, Argonne National Laboratory Report ANL-4249, Febuary, 1949, unpublished.

absorption of neutrons by ionium, or it can be isolated from uranium samples, since it is the daughter of U^{235} decay ($U^{235} \xrightarrow{\alpha} Th^{231}$). Prior to World War II, the usual source of UY had been natural uranium. By separating the thorium fraction from a large amount of uranium, it is possible to get a reasonable amount of UY.

Unfortunately, UY so separated always has large amounts of $UX_1(Th^{234})$ and UX_2 present, even when short periods (one to two days) of growth are used in order to increase the relative concentration of the short-lived (25.6 hr) UY over that of the 24.1-day UX_1 . Most measurements made on UY separated from natural uranium were made by difference. The UX_2 radiation was measured after the decay of the UY; and after correcting for the UX_1 decay, these measurements were subtracted from the measurement on the original $UX_2 + UY$ radiations.

Because of the difficulties in such measurements,

most of the work²⁻⁵ prior to the war gave incomplete results. A soft component was found, although the observed aluminum half-thicknesses varied considerably [3.2 mg/cm² (Erchova); 6.2 (Antonoff); 6.2 (Kirsch); 7.6 (Nishina)]; Erchova also found a harder component with a 35-mg/cm² half-thickness, while Antonoff found a very soft component (0.75 mg/cm² half-thickness) which he ascribed to alpha-radiation.

With the availability of sources of concentrated U²³⁵ from the Oak Ridge plants, it has become possible to isolate UY samples with much smaller amounts of UX contamination. Such samples have been studied⁶⁻⁸ at this laboratory and by Knight and Macklin at Oak Ridge. Earlier measurements reported from this laboratory⁷ on a UY preparation separated from uranium containing 65 percent U²³⁵, showed the presence of a number of radiations, including a soft beta of 5.6-mg/cm² aluminum half-thickness, *L* x-rays with half-thicknesses ranging from 40 to 120 mg/cm² aluminum, and a very soft radiation with about 1.4-mg/cm² half-thickness. The abundance of the very soft radiation was almost twice that of the beta-particle component. In addition to the *L* x-rays, a harder quantum was found, for which the data were too poor to determine the energy, although it was suspected that it might be a *K* x-ray. The quantum radiations were proved to be such by measurement through sufficient beryllium absorber to cut out all beta-particles.

Knight and Macklin⁶ found the soft beta-particle, determining a value of 7.0 mg/cm² for the aluminum half-thickness. In addition, they found two more components, with half-thicknesses of 820 mg/cm² and 104 mg/cm², which they ascribed to a 34-kev gamma-ray and to a 16-kev *L* x-ray. They suggested a decay scheme involving a beta-decay (7-mg/cm² component) followed by a 34-kev gamma, which was highly converted, thus giving rise to *L* x-rays. They ascribed the very soft component found in this laboratory to a mixture of conversion electrons and *M* x-rays.

More recent investigation in this laboratory has shown that this decay scheme is probably incomplete. The work described in this paper summarizes both the earlier experiments and the more recent measurements. Since both our results and those of Knight and Macklin suggest a single beta-particle followed by a number of other radiations (very soft conversion electrons, x-rays, gammas), it is hoped that the determination of the true disintegration rate from the counting rate is not affected seriously by the details of the disintegration scheme.

² G. N. Antonoff, *Phil. Mag.* **22**, 419 (1911).

³ G. Kirsch, *Sitzberichte Akad. Wiss. Wien, Math.-naturw. Klasse. Abt. IIa* **129**, 309-34 (1920).

⁴ Z. V. Erchova, *J. phys. radium* **8**, 501 (1937).

⁵ Nishina, Yasaki, Kimura, and Ikawa, *Nature* **142**, 874 (1938).

⁶ G. B. Knight and R. L. Macklin, *Phys. Rev.* **75**, 34 (1949).

⁷ A. H. Jaffey and E. K. Hyde, *Metallurgical Laboratory Report CN-3001*, pp. 21-4, May, 1945, unpublished.

⁸ Jaffey, Lerner, and Warshaw, *Argonne National Laboratory Report ANL-4112*, January 15, 1948, unpublished.

TABLE I. Radiations of UY.

Probable type of radiation	Aluminum half-thickness	Energy	Relative cpm at zero absorber	Approx. relative abundance
Mixture <i>M</i> x-rays and conversion e's	~1 mg/cm ²	2.5 to 5 kev } ~50 kev }	2	~2
Beta-rays	6.1 mg/cm ²	0.20 Mev	1.0	1.0
<i>L</i> x-ray	47 mg/cm ² 108	12.3 kev } 16.3 kev }	0.03	1
Gamma-ray	800 mg/cm ²	35 kev	0.003	1
Gamma-ray Gamma-ray		65 to 75 kev 100 kev or greater	small small	

II. CHEMICAL SEPARATION PROCEDURE

The enriched uranium sample used contained about 95 percent U²³⁵ by weight, which meant that the U²³⁵/U²³⁸ activity ratio was approximately 100. Since, at equilibrium, the UY/UX₂ activity ratio would be the same, a thorium sample directly separated from the uranium could have been used. However, to insure that the UX₂ activity would not interfere seriously even after the sample had decayed somewhat, the thorium fraction was completely separated, the uranium was purified, and the daughters allowed to grow in for one or two days. The thorium daughters were then separated from the uranium, the resulting UY/UX₂ activity ratio being increased by a factor of 12 to 15 over the equilibrium value.

In the first purification of the uranium, the thorium fraction was removed by precipitation with a LaF₃ carrier, after which the uranium was separated from fluoride ion by precipitation as the hydroxide, and further purified by a hexone (methyl isobutyl ketone) extraction from 10 *M* ammonium nitrate. Following the growth period, the thorium daughters were separated by precipitation with LaF₃ carrier. To complete the separation from uranium, two more purification cycles were used, involving precipitations with zirconium iodate and LaF₃ carriers. On solution of the LaF₃, the activity was separated from the lanthanum salt by extracting the thorium activity with TTA (thenoyl tri-fluor-acetone) in benzene solution. The nitric acid extract from the TTA contained essentially carrier-free UY, and the samples were prepared from this solution.

III. MEASUREMENTS

The results are summarized in Table I. Our best value for the aluminum half-thickness of the soft beta-component is about 6.1 mg/cm². According to the data summarized by Libby⁹ this corresponds to an energy of 0.20 Mev. Our results also indicate that the harder radiations are due to *L* x-rays and several gamma-rays.

⁹ W. F. Libby, *Anal. Chem.* **19**, 2 (1947).

An aluminum absorption curve gave two components with half-thicknesses 47 mg/cm² and 108 mg/cm², which correspond to 12.3 keV and 16.3 keV, respectively. These energies are in the region of the Pa *L* x-rays, which could be excited if a gamma following the beta-decay were internally converted. Although the *L* x-rays of Pa range from 13 to 20 keV, an aluminum absorption curve of the mixture of components can be (somewhat arbitrarily) "resolved" into two components, as shown in the case of Np²³⁸ decay by Jaffey and Magnusson.¹⁰ Thus, the identification of these two components with *L* x-rays arising in UY decay seems reasonable. In addition to the *L* x-rays, the aluminum absorption curve showed a component with a half-thickness of about 800 mg/cm² (34 keV). All these components were proven to be quantum radiations by measuring them in a magnetic field strong enough to bend away all electrons, and also by measuring them through a beryllium absorber sufficiently thick to absorb all emitted electrons (1900 mg/cm², 234 mg/cm²).

Lead absorption curves showed components with half-thicknesses 4.5 mg/cm² and 40 mg/cm², which were shown to be quantum radiations by bending away electrons with a magnetic field. Since the first value lies in the midst of the lead *L*-absorption edges, the 4.5-mg/cm² component could correspond to 9.6, 13.4, or 16.5 keV. Evidently, this is the *L* x-ray observed in the aluminum curve. The harder component corresponds to an energy of 38 keV, which checks fairly well with the results of the aluminum absorption curve. Silver and copper absorption curves similarly showed *L* x-rays and a gamma of either 34 or 38 keV, respectively.

In addition to these components, harder quantum radiations of lower intensity were apparent in the absorption curves taken with aluminum, lead, silver, and copper. Agreement on energy values was quite poor, but it seemed fairly certain that there was a gamma with an energy lying between 65 and 75 keV and one with an energy at or above 100 keV which could possibly be ascribed to a *K* x-ray.

A soft component was observed in a low absorption counter¹¹ with a half-thickness (aluminum) of 0.9 to 1.0 mg/cm², and an abundance about twice that of the 6.1-mg/cm² component. The same radiation was also observed in absorption curves taken with ordinary mica window G-M tubes with fairly thin windows. When a beryllium absorber or a magnetic field was used to remove electrons, the half-thickness increased to about 1.25 mg/cm². It is possible, then, that the 0.9-mg/cm² component represents a mixture of soft conversion electrons and *M* x-rays.

¹⁰ A. H. Jaffey and L. B. Magnusson, "Radiations of Np²³⁸ and the half-life of Pu²³⁸," *The Transuranium Elements* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 14.2, National Nuclear Energy Series, Vol. 14B, Division IV.

¹¹ Elliott, Sullivan, Sleight, Gladrow, Raynor, and Freedman, "Low Absorber Counters," *Radiochemistry: The Fission Products* (McGraw-Hill Book Company, Inc., New York, to be published in 1951), Paper No. 2.8, National Nuclear Energy Series, Vol. 9B, Division IV.

For x-rays, a 1.25-mg/cm² half-thickness may be due to a 1.3-keV or a 3.4-keV x-ray (multivalued because of *K*-absorption edge of aluminum). The *M* x-rays from the Pa²³¹ daughter of UY range from 2.4 to 4.9 keV, which is consistent with these results, considering the experimental error in the half-thickness measurements. If the soft conversion electrons were 0.5 to 0.7 mg/cm² in half-thickness, their energy would lie⁹ between 45 and 55 keV. If these electrons were due to conversion in the *L*-shell, the converted gamma would have an energy of approximately 65 to 75 keV (20 keV taken as average Pa binding energy in the *L* shell), which apparently checks with the observation of a low intensity gamma-ray of about this energy. If the soft electrons had been due to conversion of the 35-keV gamma, as suggested by Knight and Macklin,⁶ their energy would have been about 15 keV which corresponds⁹ to an aluminum half-thickness of about 0.15 mg/cm². No such component was observed in the low absorption counter (minimum absorption 0.3 mg/cm²).

It is further unlikely that the large number of conversion electrons observed were due to conversion of the 35-keV gamma, since, after correcting for counting efficiencies, the abundances of the *L* x-rays and the gamma seemed to be equal (Table I). The ratio (at zero absorber) of the *L* x-ray counts to the counts due to the 35-keV gamma-ray was found to be approximately 10. Taking the average energy of the *L* x-ray to be 16 keV, the half-thickness in argon of this radiation would be 42 mg/cm², while the corresponding half-thickness for a 35-keV gamma is 380 mg/cm². For low energy quanta, most of the counts in a G-M tube are those arising from absorption of the quanta in the gas of the tube, so that at low efficiencies, the counting efficiency for a given type of such radiation is inversely proportional to the half-thickness for absorption in the gas. Neglecting the small effect due to the alcohol, the absorption of a 16-keV x-ray in an ordinary argon-alcohol G-M tube relative to the absorption of a 35-keV gamma would be 380/42 = 9.0. The agreement between this value and the ratio of observed counts suggests that the number of *L* x-rays and 35-keV gammas are approximately equal, with a relatively small percentage (if any) of conversion of the gamma.

It also seems likely that the number of beta-particles is about equal to the number of *L* x-rays emitted (Table I). Using an argon-alcohol G-M tube, the ratio of the 6.1-mg/cm² (beta) component counts (at zero absorber) to the x-ray counts was found to be about 30. If it is assumed that the number of *L* x-rays is approximately equal to the number of betas, the efficiency for counting the *L* x-rays of Pa is found to be about 3 percent. This value is not unreasonable, since Jaffey and Magnusson¹⁰ found a value of 2½ percent^{11a} for the *L* x-rays of Pu from the decay of Np²³⁸.

^{11a} Since the *L* x-rays of Pa are lower in energy than those of Pu, the absorption coefficient in argon of the former would be greater, and hence the counting efficiency higher.

As mentioned above, the very soft component may be a mixture of soft conversion electrons and M x-rays. The absorption coefficients of the M x-rays in argon are quite large, so that their counting efficiency is high. It is fairly reasonable to assume, then, that somewhat less than half of the soft component counts is due to M x-rays and the remainder due to conversion electrons.

Some coincidence-measurements were made, but proved relatively little, because of the very low counting efficiencies for the soft gammas involved. In these measurements, two G-M tubes were used with a permanently fixed absorber over one and coincidences measured as a function of the aluminum absorber thickness in front of the other tube. In one measurement, 475 mg/cm² Be was used as the permanently placed absorber; in another, 1000 mg/cm² Al was used. The former cut out electrons, but allowed the L x-rays and gammas through; the latter cut out L x-rays as well as electrons. The results showed that the 6.1-mg/cm² component and the L x-rays were in coincidence with radiation that could penetrate 475-mg/cm² Be or 1000 mg/cm² Al; i.e., the beta-particle and L x-rays were in coincidence with quantum radiation, some of which were gamma-rays.

The half-life of UY was measured by means of a thin-walled glass G-M tube; this type of tube has been found to have more reproducible characteristics than the mica window tubes available. The tube was frequently checked with a standard and most of the counts were within the expected statistical deviation of 0.3 percent. Because this glass tube had a minimum absorption of about 30 mg/cm², it attenuated the UY beta-radiation by almost 5 half-thicknesses (a factor of about 30). As a result, the hard radiation of UX₂ was found to give a long-lived tail in the decay curve. Although the UX₂ concentration was quite a small fraction of the total activity, the severe attenuation of the UY betas made the UX₂ radiation (which was only partially attenuated) a sizeable part of the total counts measured.

A better decay curve was determined by using 1230 mg/cm² of Be to cut out all betas. Under these conditions only the quanta emitted in the UY decay were measured, although a nondecaying residuum of several counts was found, after the UY decayed out. This was ascribed to incomplete separation of U²³⁵ and U²³⁴, both of which emit quantum radiation.¹²⁻¹⁵ The resolution losses were determined by using the paired-source technique,¹⁶ and were found to be 0.42 percent per

¹² R. L. Macklin and G. B. Knight, MDDC-976, December, 1946 (unpublished).

¹³ R. L. Macklin, Carbon and Carbide Report No. A-3640, April, 1946 (unpublished).

¹⁴ R. L. Macklin, Carbon and Carbide Report No. K-97, December, 1947 (unpublished).

¹⁵ B. Scott, Metallurgical Laboratory Report CC-3715, Nov., 1946 (unpublished).

¹⁶ T. P. Kohman, "A general method for determining coincidence corrections of counting instruments," Paper No. 22.50, *The Transuranium Elements*; Metallurgical Laboratory Report CP-3275, June, 1945 (unpublished).

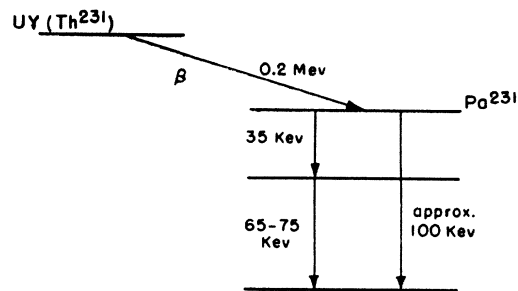


FIG. 1. Tentative decay scheme for UY disintegration. The results are consistent with the assumptions that the 35-kev gamma is relatively unconverted, while the 65- to 75-kev gamma is highly converted. Associated with the conversion are L and M x-rays and conversion electrons.

1000 cpm. Two separate preparations of UY were made from the U²³⁵ sample. Sample I (initial counting rate about 700 cpm) was counted down to background (about six half-lives). Sample II (initial counting rate about 3600 cpm) was also counted down to background (about eight half-lives). The half-lives were calculated using a least-squares method, Sample I giving a value 25.56 ± 0.06 hours and Sample II giving 25.73 ± 0.06 hours. (The deviations represent the standard deviation calculated from the dispersion of the data.) The results probably justify a half-life value of 25.64 ± 0.1 hours.

SUMMARY

The results described indicate that the following components are probably present with approximately equal intensity (within perhaps 25 percent): 0.20-Mev beta, L x-rays, M x-rays, 50-kev conversion electrons, 35-kev gamma. In addition there seems to be evidence for the existence of a highly converted gamma in the range 65 to 75 kev and of a low intensity gamma with an energy of 100 kev or greater. Despite the scanty evidence, it may be possible to combine these data into a consistent decay scheme.* A tentative scheme is suggested in Fig. 1. The order of the two softer gamma-rays is, of course, uncertain.

At zero absorber (Table I), using an argon-alcohol mica window G-M tube (9 cm argon, 1 cm alcohol) the very soft component (presumably M x-rays and conversion electrons) had almost twice as many counts as the beta-ray component; the counting rate of the L x-rays was about 1/30 that of the beta-rays; the 35-kev counting rate was about 1/10 that of the L x-rays; while the counting rates of the other gammas were somewhat lower yet.

The half-life of UY was measured as 25.64 ± 0.1 hours.

* *Note added in proof.*—Further work in this laboratory with a beta-spectrometer and a NaI scintillation gamma-spectrometer indicates that the decay scheme is much more complex than was found by absorption techniques. The single beta-ray described in this paper has been resolved into three components, the hardest beta having an energy of 302 kev, and a total of nine gamma-rays have been found. A disintegration scheme has been devised from which it appears that the total disintegration energy is 324 kev. It is planned to publish these results in the near future.