### Alpha Decay of <sup>227</sup>U<sup>†</sup>

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The  $\alpha$  spectrum of the <sup>227</sup>U decay chain, previously reported in 1952 by Meinke *et al.*, was investigated by bombarding <sup>231</sup>Pa with protons from 35 to 54 MeV. The half-life of the parent nuclide <sup>227</sup>U was found to be  $1.1\pm0.1$  min, in agreement with the earlier value of  $1.3\pm0.3$  min. The  $\alpha$ -particle energies in the decay chain were determined more precisely than in the earlier study. In addition, some disagreement was found with the 1952 work. The nuclides and  $\alpha$ -particle energies observed in the present investigation are as follows: <sup>227</sup>U, 6.87\pm0.02 MeV; <sup>228</sup>Th, 7.33\pm0.02 MeV; <sup>219</sup>Ra, 7.70\pm0.02 MeV (relative intensity 0.7\pm0.1) and 7.99\pm0.02 MeV ( $0.3\pm0.1$ ); <sup>215</sup>Rn, 8.67\pm0.02 MeV; and <sup>211</sup>Po, 7.46\pm0.02 MeV.

### I. INTRODUCTION

**B**ECAUSE of our interest in the  $\alpha$  decay of neutrondeficient isotopes,<sup>1-3</sup> we have studied the  $\alpha$  decay of short-lived <sup>227</sup>U. This nuclide and its decay products were first characterized by Meinke *et al.*<sup>4</sup> in 1952; since that time, no additional work on <sup>227</sup>U has been reported.<sup>5</sup>

Meinke *et al.* prepared <sup>227</sup>U by irradiation of <sup>232</sup>Th with high-energy  $\alpha$  particles followed by rapid chemical separation of uranium. They determined the half-life of <sup>227</sup>U to be  $1.3\pm0.3$  min, while its daughters all had periods less than 1 sec. With the exception of the 7.43-MeV  $\alpha$  of <sup>211</sup>Po, which was known,<sup>6</sup> the  $\alpha$ -particle peaks observed in the spectrum were assigned to the members of the decay chain because of the approximate agreement of the experimental energies with values from  $\alpha$ -decay systematics that were then available.

As the authors pointed out, three major problems were encountered in their work. First, it was not a simple task, after the chemical processing, to prepare the thin samples so necessary for accurate  $\alpha$ -pulseheight analysis. Secondly, in their  $\alpha$  spectra there were, at best, approximately equal numbers of counts from 1-min <sup>227</sup>U and 9.3-min <sup>228</sup>U. Thirdly, the  $\alpha$ -particle energies from the <sup>228</sup>U decay chain are very similar<sup>5</sup> to those reported for <sup>227</sup>U. Precise determination of the <sup>227</sup>U energies under these conditions was accordingly very difficult.

In the present work, <sup>227</sup>U and <sup>228</sup>U were produced by irradiating <sup>231</sup>Pa with protons from the Oak Ridge Isochronous Cyclotron (ORIC). Instead of the 150MeV  $\alpha$  particles used for the <sup>232</sup>Th( $\alpha$ , 9*n*)<sup>227</sup>U reaction, proton energies of only 35–54 MeV were needed to observe the <sup>231</sup>Pa(p, 4*n*)<sup>228</sup>U and <sup>231</sup>Pa(p, 5*n*)<sup>227</sup>U reactions. By proper selection of the proton energy, the interferences due to the <sup>228</sup>U spectrum could be minimized. Also, at these relatively low bombarding energies, competition from other nuclear reactions was not very significant, so that  $\alpha$  spectra could be obtained without the need of prior chemical purifications.

### **II. EXPERIMENTAL PROCEDURE**

To facilitate the detection of the short-lived uranium radionuclides, the experiments were performed on-line at the ORIC. The target assembly has been described in a previous publication.<sup>2</sup> It provides for the collection of recoil nuclei ejected from a thin target followed by their rapid assay. Absorbers that decrease the beam energy, and beryllium catcher foils, are mounted on movable wheels that can be remotely driven. One can thus select and automatically position any appropriate combination of absorber and catcher for a given irradiation. After irradiation, the wheel can be automatically turned in  $\leq 10$  sec to place the beryllium catcher near a surface-barrier Si(Au) detector for assay of the  $\alpha$ -particle radioactivity.

Thin targets of <sup>231</sup>Pa were prepared by modifying a published procedure.<sup>7</sup> The purified oxide of <sup>231</sup>Pa was dissolved in 1N HF; the solution was adjusted to pH 6 with 1N NaOH, and then diluted with 1N NaF. An aliquot was placed in a Lucite plating cell and the <sup>231</sup>Pa electrodeposited onto a 1-mil-thick platinum foil. The current density used was ~550 mA/cm<sup>2</sup> at a voltage of ~15 V. Typical target thicknesses obtained were ~100  $\mu$ g/cm<sup>2</sup>.

During irradiation, the target was positioned so that first the platinum backing, cooled by helium gas, and then the protactinium deposit intercepted the beam. Evacuation of the space between target and catcher ensured that the recoil nuclei from the (p, xn) reactions would reach the catcher foil.

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<sup>&</sup>lt;sup>1</sup> R. L. Hahn, K. S. Toth, and T. H. Handley, Phys. Rev. **163**, 1291 (1967).

<sup>&</sup>lt;sup>2</sup> R. L. Hahn, M. F. Roche, and K. S. Toth, Nucl. Phys. A113, 206 (1968). <sup>3</sup> K. S. Toth, R. L. Hahn, and M. F. Roche, Phys. Rev. (to

<sup>&</sup>lt;sup>6</sup> K. S. Joth, K. L. Hann, and M. F. Koche, Phys. Rev. (to be published). <sup>4</sup> W. M. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev.

**<sup>5</sup>**, 429 (1952).

<sup>&</sup>lt;sup>6</sup>C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (Wiley-Interscience, Inc., New York, 1967), 6th ed. <sup>6</sup>R. F. Leininger, E. Segrè, and F. N. Spiess, Phys. Rev. 82,

A334 (1951).

<sup>&</sup>lt;sup>7</sup> C. Ferradini, J. Chim. Phys. 53, 714 (1956).

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FIG. 1.  $\alpha$  spectra observed from irradiations of <sup>281</sup>Pa with protons. (a) Spectrum taken during the first 2.5 min after bombardment; the proton energy was 44 MeV. (b) Spectrum also taken from the 44-MeV irradiation. The counting interval covered 9-20 min after bombardment. (c) Spectrum taken during the first 5 min after the 35-MeV irradiation.

 $\alpha$ -particle spectra were obtained with the Si(Au) detector coupled through a low-noise charge preamplifier, linear amplifier, and postamplifier to a 1600-channel analyzer. This pulse-height analyzer served as eight 200-channel analyzers to store spectra as a function of decay time. To obtain adequate counting statistics, multiple irradiations were done at each selected proton energy and the resulting spectra were summed in the memory of the analyzer.

Double differentiation in the linear amplifier was employed to minimize pileup between the  $\beta$ - $\gamma$  background and the  $\alpha$  pulses of interest. With these 0.75- $\mu$ sec-wide bipolar pulses, the full width at half maximum (FWHM) of standard  $\alpha$  peaks was ~50 keV. Under actual irradiation conditions, the FWHM increased to  $\sim$ 75 keV. A precision pulser in conjunction with a <sup>244</sup>Cm source mounted on the recoil-catcher wheel was used to set the energy scale of the spectrometer. A more accurate calibration was obtained with <sup>228</sup>U peaks observed in the spectra.

## **III. RESULTS AND DISCUSSION**

To survey the (p, xn) reactions on <sup>231</sup>Pa, irradiations were performed at proton energies of 35, 40, 44, 49, and 54 MeV. A typical  $\alpha$ -particle spectrum obtained from the 35-MeV bombardment is shown in Fig. 1(c). The major peaks are clearly identifiable as arising from <sup>228</sup>U and its daughters<sup>5,8</sup> and from <sup>229</sup>U.<sup>5</sup> Analysis of the decay data from this bombardment confirmed the presence of these nuclides.

Data from the multiple irradiations done at 44 MeV are plotted in Figs. 1(a) and 1(b). The spectrum obtained within the first 2.5 min after bombardment is shown in Fig. 1(a), while Fig. 1(b) shows the peaks observed within the interval from 9 to 20 min after irradiation. At the later counting times, as can be readily seen by comparing the spectra in Figs. 1(b) and 1(c), the peaks of the <sup>228</sup>U chain become prominent. These well-known  $\alpha$ -particle energies were used as internal standards to calibrate accurately the energy scale of the spectrometer. Members of other decay chains are also indicated in Fig. 1(b). Because of the complexity of the spectra and the 75-keV FWHM, we assign an uncertainty of  $\pm 0.02$  MeV to the energy values.

Figure 1(a) demonstrates the presence of a shortlived decay chain with energies different from those of <sup>228</sup>U and its descendants. Especially striking in the comparison of Figs. 1(a) and 1(b) are the shifts in peak position caused by the decay of the short-lived components. For example, after the 8.67- and 7.99-MeV peaks have decayed away, the 8.78- and 8.05-MeV peaks of <sup>228</sup>U are clearly seen in the spectrum. Least-squares analysis<sup>9</sup> of the decay data for the 6.87-, 7.33-, 7.46-, 7.70-, 7.99-, and 8.67-MeV  $\alpha$  particles indicated that

TABLE I.  $\alpha$ -particle energies (MeV) in the <sup>227</sup>U decay chain.

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Nuclide	Meinke et al.ª	This work
227 U	6.8±0.1	6.87±0.02
↓ 223Th	7.55±0.1	$7.33{\pm}0.02$
↓ <sup>219</sup> Ra	8.0±0.1	7.99±0.02 (0.3±0.1) <sup>b</sup> 7.70±0.02 (0.7±0.1) <sup>b</sup>
↓ <sup>215</sup> Rn	8.6±0.1	8.67±0.02
211Po	7.43±0.1°	$7.46 {\pm} 0.02$

<sup>a</sup> See Ref. 4.

<sup>&</sup>lt;sup>b</sup> Numbers in parentheses are relative emission probabilities, assuming that the two  $\alpha$  particles are emitted by the same nuclide.

<sup>&</sup>lt;sup>e</sup> The energy of the <sup>211</sup>Po  $\alpha$  particle was known prior to the work of Meinke et al. See Ref. 6.

<sup>&</sup>lt;sup>8</sup>C. P. Ruiz, University of California Radiation Laboratory Report No. UCRL-9511, 1961 (unpublished).
<sup>9</sup>J. B. Cumming, Natl. Acad. Sci. Natl. Res. Council, Nucl. Sci. Ser. NAS-NS 3107, 25 (1963).

they all decay with the same period. The average value found for the half-life was  $1.1\pm0.1$  min, in agreement with the previously reported<sup>4</sup>  $1.3\pm0.3$  min.

Table I lists the  $\alpha$ -particle energies found in this work and compares them with the results of Meinke *et al.*<sup>4</sup> Some differences with the earlier work are apparent. No evidence was found in the present study for a peak at 7.55 MeV. Instead, peaks were observed at 7.33 and 7.70 MeV. In addition, the 7.99-MeV peak [see Fig. 1(a)] was much smaller than any of the other peaks in the spectrum. Analysis of the decay data indicated that the sum of the intensities of the 7.99- and 7.70-MeV peaks was comparable to the intensities of the other short-lived peaks. Apparently, the 7.99and 7.70-MeV  $\alpha$  particles are emitted by the same nuclide, with relative emission probabilities of  $0.3\pm0.1$ and  $0.7\pm0.1$ , respectively.

Apart from the noted discrepancies, the present results, although more precise, agree in general with those reported earlier by Meinke *et al.*<sup>4</sup> This agreement allows us to assign the observed  $\alpha$  particles to the <sup>227</sup>U decay chain, as seen in Table I. [A comparison with the data taken at the various bombarding energies of  $^{281}$ Pa(p, xn) excitation functions for  $^{228}$ U and  $^{227}$ U, calculated by assuming compound-nucleus formation,<sup>10</sup> also supports this assignment.] Note that the 7.33-MeV  $\alpha$  particle is assigned to  $^{223}$ Th, to replace the previously reported<sup>4</sup> 7.55-MeV  $\alpha$  particle, and both the 7.99- and 7.70-MeV  $\alpha$  particles, as discussed above, are assigned to  $^{219}$ Ra. These assignments are consistent with the systematic trend generally observed for  $\alpha$  emitters with neutron number between 128 and 140: The  $\alpha$ -particle energy increases with each step down the decay chain.

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<sup>10</sup> I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116**, 683 (1960).

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# Distribution of Nuclear Charge in Low-Energy Fission of Transthorium Elements

### A. Notea\*

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The variation of the nuclear charge dispersion  $\sigma$  in the thermal neutron fission of <sup>235</sup>U was examined in three limited product mass ranges. By using measured or estimated  $\sigma$  values and fractional chain yields, the "experimental" most probable charge values  $Z_p(A)$  were derived. For chains outside the mass regions mentioned, the average dispersion  $\sigma = 0.57 \pm 0.07$  was used. From the derived experimental  $Z_p(A)$  values, a new empirical  $Z_p(A)$  function covering the whole fission-product mass region was drawn. This empirical function agrees with all published experimental data. The way of estimating product yields from this function is described. The effects of closed shells on the shape of the curve are discussed. The empirical function is preserved in shape for other types of low-energy fission, but is shifted by a constant  $\Delta\delta$ . A simple empirical relation for calculating  $\Delta\delta$  is given.

#### I. INTRODUCTION

THE division of nuclear charge between complementary fission fragments is characterized by the most probable charge  $Z_p(A')$  of a fragment with mass A'. From measured independent yields of individual fission products along a single mass chain A, it is possible to determine an "experimental"  $Z_p(A)$ . However since the latter corresponds to the situation after prompt neutron emission, it is not identical with  $Z_p(A')$ 

In order to determine  $Z_{\nu}(A')$  from the experimental function, it is necessary to know  $p_{\nu}(A', Z)$ , the probability for emission of  $\nu$  neutrons (where  $\nu = 0, 1, 2$ ,

etc.) from primary fragment (A', Z). But even  $\nu(A', Z)$ , the isobaric distribution of the average number of prompt neutrons emitted from the primary fragment A', has not yet been measured. Since only  $\nu(A')$ , the average value over charge number, is available, it is only possible to get a "representative"  $Z_p(A')$  for an average fragment mass number by adding  $\nu(A)$  to the product mass number for which experimental  $Z_p(A)$  is known.

$$Z_p(A')_{rep} = Z_p[A + \nu(A)]$$

where  $\nu(A)$  is calculated by the transformation  $A = A' - \nu(A')$ .

The accumulated information on fractional yields, especially in the thermal neutron fission of <sup>235</sup>U, indicates that the charge-distribution width at half-maxi-

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