The Alpha-Spectra of Pu^{239} and Pu^{240}†

FRANK ASARO AND I. PERLMAN
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received July 30, 1952)

Utilizing a magnetic alpha-particle spectrograph the complex alpha-spectrum of Pu^{239} was found to consist of three groups: α_0 (69 percent), α_{13} (20 percent), and α_{51} (11 percent). The spectrum of Pu^{240} consisted of two groups, α_0 (76 percent) and α_{44} (24 percent). The nomenclature α_x refers to that alpha-group which leaves the product nucleus with an energy of x kev above the ground state. The alpha-particle energy of α_0 of Pu^{239} was measured as 5.150±0.002 Mev and that of Pu^{240} as 5.162±0.004 Mev. These values are correlated with the gamma-rays and conversion electrons observed by other workers to form a partial decay scheme.

INTRODUCTION

SOON after the availability of Pu^{239} in supra-tracer quantities the alpha-activity was found to be accompanied by gamma-radiation in low abundance.[1,2] These and later measurements will be discussed below where the decay scheme is considered. The complex alpha-spectrum presumed from these data was first observed directly by Rosenblum, Valadares, and Goldschmidt[3] who employed a large permanent magnet spectrograph. They found the highest energy group in highest abundance and a prominent group at 50-kev lower energy. Some evidence was also found for a very weak group 200 kev below the main group. The present study amplifies these results and attempts to explain the gamma-ray spectrum which has been reported by several sources. Samples of plutonium containing measurable Pu^{240} activity have been used to determine its alpha-particle spectrum.

EXPERIMENTAL RESULTS

Pu^{239} Alpha-Spectrum

The magnetic alpha-particle spectrograph used for these measurements has been described in other reports.[4,5] The source in each case was prepared by vacuum sublimation from a tungsten filament to a platinum plate of the dried residue from a solution of plutonium (IV) in hydrochloric acid. The platinum plate was masked to give a $\frac{1}{8}\times 1$ inch band and in the case of Pu^{239} approximately 2 micrograms was deposited. Samples so prepared have the extreme uniformity demanded for alpha-particle spectroscopy.

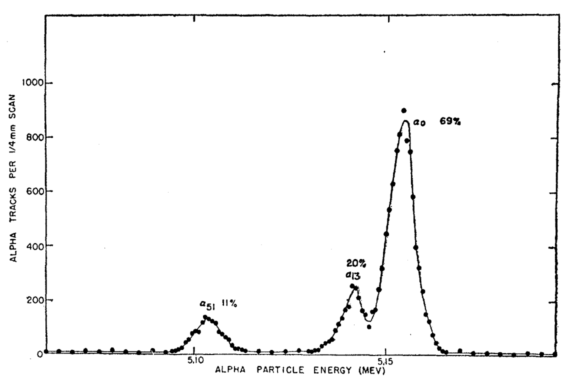

FIG. 1. Pu^{239} alpha-spectrum—peak region.

Figure 1 shows the spectrum of isotopically pure Pu^{239}. The source consisted of 2.5 micrograms of Pu^{239} and was exposed for 110 hours. The designations for the alpha-groups follows the same convention previously used in which the highest energy group, presumed to be the ground-state transition, is termed α_0 and other groups are shown with subscripts indicating the corresponding energy levels in kilovolts above the ground state. Although no higher energy group than that designated α_0 has been detected, there is some evidence to be discussed later which indicates that α_0 is not the ground-state transition. If this proves to be so, these designations will have to be revised.

All of the three peaks observed have the same width at half-maximum (~8 kev), and the abundances as indicated were found both by comparing peak heights and by integrating the total alpha-tracks.

In another exposure for 90 hours the slit system was changed to give higher transmission in order to look for rare alpha-groups. The peak widths went up to 21 kev and α_0 and α_{13} were no longer clearly resolved. The abundance of α_{51} was found to be 11.7 percent which agrees with the other measurement cited. A careful search was made for other alpha-groups and the data are recorded in Fig. 2. The energy range covered was 4.82–5.57 Mev which extends from 330 kev below the main group to 430 kev above it. No alpha-group was found and the limits can be set as follows: from 25 kev above α_0 to 430 kev there is no group in greater than 0.15 percent abundance; from 70 kev below α_0 (20 kev below α_{51}) to 330 kev there is no group in greater than 0.3 percent abundance. The plate from which Fig. 1 was derived was also counted over an extended range and the results were substantially the

† This work was performed under the auspices of the AEC.
[1] A. Ghiorso, Metallurgical Laboratory Report CK-1511 (April, 1944) (unpublished).
[2] Sullivan, Kohman, and Swartout, Hanford Report HEW-3-1635 (February, 1945) (unpublished).
[3] Rosenblum, Valadares, and Goldschmidt, Compt. rend. **230**, 638 (1950).
[4] F. L. Reynolds, Revs. Sci. Instr. **22**, 749 (1951).
[5] Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952).

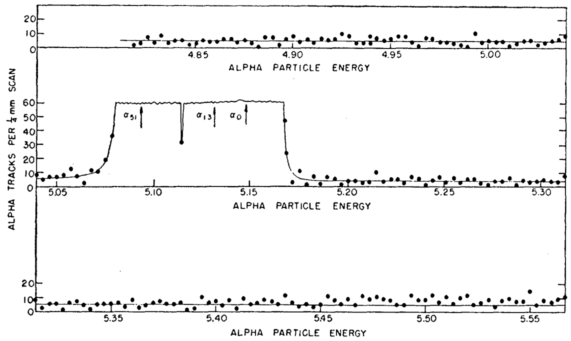

FIG. 2. Pu^{239} alpha-spectrum—high and low energy region.

same although the limits of detection were not so low because of the fewer total tracks recorded.

The energy for Pu^{239} was determined by comparing α_0 with Po^{210} as the standard for which the particle energy was taken as 5.298 Mev.[6] The energy for α_0 of Pu^{239} was found to be 5.150±0.002 Mev as compared with the other spectrograph value,[3] 5.147, obtained using ThC as a standard.

These spectrograph values are in good agreement with reported values obtained using ionization chambers and range methods. Cranshaw and Harvey[7] using an ionization chamber with a pulse-height analyzer reported 5.159 (±0.005) Mev and Jesse and Forstat[8] obtained 5.140 (±0.005) Mev by measuring total ionization current. An air range determination by Chamberlain *et al.*[9] gave 5.15 Mev.

The energies of the two shorter range groups were obtained from their displacements from the main group using the dispersion of the instrument as described previously.[5] The differences in energy from the main group, α_0, were 12.8±0.7 kev and 49.7±0.7 kev. Rosenblum, Valadares, and Goldschmidt[3] resolved α_{51} and found its energy to be 50 kev lower than the main group and our result is in excellent agreement for this group. Adding a correction of 1.7 percent to these alpha-particle energy differences to obtain the spacing between energy levels and rounding off to the nearest kilovolt, the level differences become 13 and 51 kev, hence α_{13} and α_{51}. These energies would correspond to gamma-ray energies between the appropriate levels.

Pu^{240} Alpha-Spectrum

This isotope of plutonium is an alpha-emitter of 6600-year half-life[10] and is best prepared by neutron capture by Pu^{239}.[11] Its alpha-energy is known to be very close to that for Pu^{239} and had not previously been resolved for sure.

A sample containing 5×10^5 disintegrations per minute of mixed Pu^{239} and Pu^{240} activities was exposed for 46 hours, and the spectrum shown in Fig. 3 was registered. The peaks were readily assigned to their respective isotopes because (1) mass spectrographic analysis indicated that Pu^{240} was in an abundance such that its alpha-activity should predominate,[12] and (2) the energy differences and relative abundances of the three peaks assigned to Pu^{239} were in close agreement with those for pure Pu^{239} (see Fig. 1).

The energy of the main group of Pu^{240} is 12±2 kev greater than that for Pu^{239}. Using our value, 5.150 Mev, for the energy of the α_0 group of Pu^{239}, the energy for Pu^{240} α_0 group is 5.162. Since we are fairly certain that the most abundant group of an even-even alpha-emitter represents the ground-state transition we may then calculate the decay energy for Pu^{240} as 5.250 Mev.

The second alpha-group of Pu^{240} is 43.5±2 kev lower energy than the main group; hence it is designated α_{44} signifying that it leads to a state 44 kev above the ground state. Its abundance is 24 percent; therefore the partial alpha half-lives for the two groups, based on the 6600-year measured half-life, are 8700 years and 27,000 years. The half-life *versus* energy relationships for these groups are close to expectations of alpha-decay as can be seen from Fig. 4. The curve is the calculated half-life *versus* energy relationship for plutonium isotopes in which the rigorous one-body decay theory was applied.[13,14] The nuclear radii were taken to follow the function, $r=1.51A^{\frac{1}{3}}\times10^{-13}$ cm, and the effective alpha-energies were obtained by adding to the decay energies 39 kev to account for the change in orbital electron binding as suggested by Ambrosino and Piatier.[15] A similar curve for curium and its method of derivation are shown in another publication.[5]

Figure 4 shows that both Pu^{240} groups are in close agreement with the theory, a situation which we now accept as normal for an even-even nuclide. Similarly

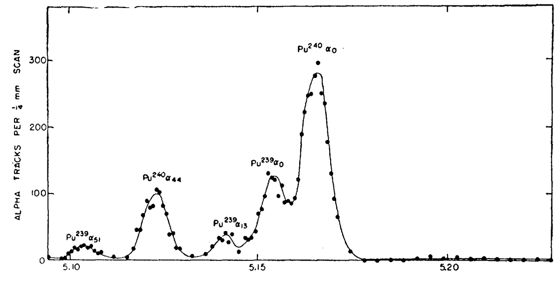

FIG. 3. $Pu^{239,240}$ alpha-spectrum.

[6] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 (1938).
[7] T. E. Cranshaw and J. A. Harvey, Can. J. Research **A26**, 243 (1948).
[8] W. P. Jesse and H. Forstat, Phys. Rev. **73**, 926 (1948).
[9] Chamberlain, Gofman, Segrè, and Wahl, Phys. Rev. **71**, 529 (1947).
[10] Inghram, Hess, Fields, and Pyle, Argonne National Laboratory Report ANL-4653 (June, 1951) (unpublished).
[11] Ghiorso, James, Morgan, and Seaborg, Phys. Rev. **78**, 472 (1950).

[12] We are indebted to Dr. S. G. Thompson for the plutonium used in this measurement and to Mr. F. L. Reynolds for the mass analysis.
[13] M. A. Preston, Phys. Rev. **71**, 865 (1947).
[14] I. Kaplan, Phys. Rev. **81**, 962 (1951).
[15] G. Ambrosino and H. Piatier, Compt. rend. **232**, 400 (1951).

the groups for Pu^{239} follow the now familiar pattern of hindered decay expected of nuclides with odd nucleons.

DISCUSSION OF DECAY SCHEME

It will be pointed out that the Pu^{239} alpha-spectrum as observed here cannot explain all features of the gamma-ray and conversion electron spectra which have been reported. First we shall examine the area of agreement which is indicated in Fig. 5.

A gamma-ray of 50 kev was reported early by Ghiorso,[1] and this has been abundantly confirmed. West and Dawson[16] using a proportional counter spectrometer found a 52.3 ± 0.4 kev photon in an abundance of 7×10^{-5} per alpha-particle, Martin[17] in this laboratory has measured this gamma-ray as 53 ± 2 kev with a scintillation spectrometer, while two laboratories[18,19] have found conversion electrons corresponding to a 50-kev gamma-ray in coincidence with alpha-tracks in Pu^{239} impregnated nuclear emulsions. Albouy and Teillac[18] found the electrons to be present to the extent of 16 per 100 total alpha-particles, while Dunlavey and Seaborg[19] found 12.5 electrons per 100 alpha-particles. It seems almost certain that this gamma-ray represents the transition between the levels reached by α_{51} and α_0 because of the agreement in energy and moderate agreement in abundance. Rosenblum and co-workers[3] reported a much higher abundance than ours for α_{51}, but their resolution would have been more difficult.

West and Dawson[16] also found a gamma-ray of 37.2 ± 0.5 kev in low abundance which corresponds exactly to a transition between α_{51} and α_{13}. They also found in low abundance gamma-rays of 59.2 and 32.0 kev. The close agreement in energy of one of these photons with that for the most prominent gamma-ray of Am^{241} (59.78 kev,[20] 59.7 kev[21]) makes it tempting to

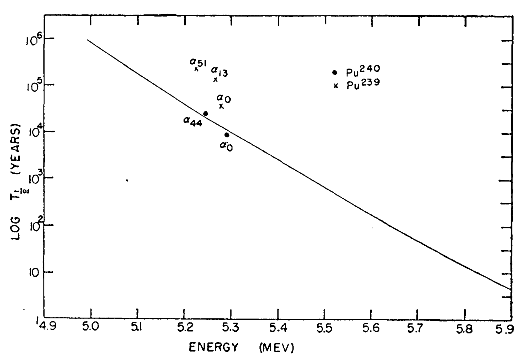

FIG. 4. Theoretical alpha-decay curve for plutonium.

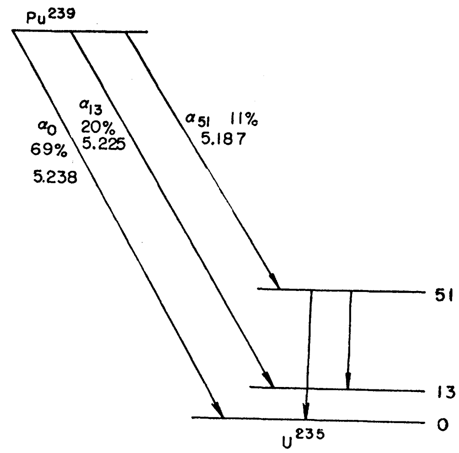

FIG. 5. Pu^{239} partial decay scheme.

consider that it is from Am^{241} which grew from Pu^{241}.[22] A similar but more extensive series of conversion electron lines has been found by Wagner, Freedman, and Engelkemeir[23] in a sample not isotopically pure Pu^{239}. The latter paper reports four weak lines in pure Pu^{239} and these could be interpreted as the L_{I} or L_{II}, L_{III}, M, and N lines of the 51-kev transitions or two of them could arise from the 38-kev transition.

The preceding discussion shows that there need be no sharp disagreement between our alpha-spectrum and the gamma-ray transitions so far considered. However, there is harder gamma-radiation which is more difficult to explain. There have been reported gamma-rays of 300 kev[1] and 420 kev[2] in very low abundance, and these could well correspond to low energy alpha-groups in quantity below our level of detection. Another gamma-ray can be deduced from the work by Albouy and Teillac[18] and by Dunlavey and Seaborg[19] who both measured electron tracks stemming from alpha-tracks in nuclear emulsions. Albouy and Teillac reported theirs as the "K conversion electrons from a 200-kev gamma-ray" which would mean that the electron line was 100 kev and the abundance as 0.1–1 percent of the alpha-particles. It is doubtful that these electrons are K shell converted because the corresponding K x-rays have not been seen in requisite abundance. Dunlavey and Seaborg found similar electrons in the energy range 100 ± 20 kev in 0.5 percent abundance. If the electrons are the L conversion line of an $\sim$120-kev gamma-ray and the abundance is correct, the corresponding alpha-group should have been seen. According to the data of Fig. 3 an alpha-group of lower energy than α_0 by 70 kev or more would be detected if present in 0.3 percent

[16] D. West and J. K. Dawson, Proc. Phys. Soc. (London) **64**, 586 (1951).
[17] D. Martin (unpublished data).
[18] G. Albouy and J. Teillac, Compt. rend. **232**, 326 (1951).
[19] D. Dunlavey and G. T. Seaborg, Phys. Rev. **87**, 165 (1952).
[20] C. I. Browne and I. Perlman, Phys. Rev. **85**, 758 (1952).
[21] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952).
[22] Dr. West has kindly informed us that another article is in publication showing that the 59-kev gamma-ray is from Am^{241} and that the 32-kev peak may have some other origin.
[23] Wagner, Freedman, and Engelkemeir, AEC Report AECD-3304 (November, 1951) (unpublished).

abundance or greater. The discrepancy between the abundance of the $\sim$120-kev transition and the absence of a corresponding alpha-group is too close to the limits of error in measurement to be considered real at this stage. If it should prove to be real, this may be taken as evidence that the ground-state transition for the alpha-decay of Pu^{239} has not yet been seen.

We wish to acknowledge the assistance of Miss Beverly Turner and Mr. James Vanderveen in counting the alpha-tracks.

PHYSICAL REVIEW VOLUME 88, NUMBER 4 NOVEMBER 15, 1952

Radioisotopes of Osmium*

J. B. SWAN† AND R. D. HILL
Physics Department, University of Illinois, Urbana, Illinois
(Received July 30, 1952)

Osmium activities of 15-day and 32-hour half-lives have been identified with Os^{191} and Os^{193}, respectively. A 14-hour activity, decaying by a 74.2-kev gamma-transition into the 15-day activity, has been assigned to an isomeric state in Os^{191}. The decay scheme of Os^{191} has been investigated and a 7/2+ assignment has been made to the 15-day ground state and an $i_{13/2}$ assignment to the 14-hour isomeric state. The 7/2+ state is attributed to a coupling of an odd number of $i_{13/2}$ neutrons.

INTRODUCTION

OSMIUM has seven stable isotopes, with mass numbers from 184 to 192, except for vacancies at 185 and 191. Three radioisotopes have so far been observed, and these have been allocated to mass numbers 185, 191 and 193.[1–13] We report in the present paper a new activity and a revised assignment of two of the already known activities.

Os^{191} AND Os^{191m}: EXPERIMENTAL OBSERVATIONS

Ordinary osmium, in powdered metallic form, was irradiated by neutrons in the heavy water pile at Argonne. The internal conversion electron spectrum, measured with 180° magnetic spectrographs, showed three groups of conversion lines. Accurate energy determinations, which are given in Table I, indicate that the gamma-transitions of 41.7 and 129.1 kev take place in iridium and that the 74.2-kev gamma-transition occurs in osmium.

Lifetime tests on the individual conversion lines showed that there was an initial rise in the intensity of the 41.7- and 129.1-kev lines, which changed over after a period of several days into a constant decay of 15-day half-life. On the other hand, the 74.2-kev transition lines decayed throughout with a half-life of 14±2 hours. A set of decay curves for a particular source, which was bombarded one day in a pile, is shown in Fig. 1. It is clear that the growth of the 41.7- and 129.1-kev transition intensities is consistent with a partial production of the 15-day activity from a parent of 14-hour half-life.

Osmium was also bombarded by gamma-rays from a 22-Mev betatron, using an internal target arrangement.[14] A conversion spectrum the same as, although weaker than, that from a pile irradiated source was obtained. The 15-day activity is therefore to be identified with mass number 191, instead of 193, and the 14-hour activity is thus an isomer of Os^{191}. The present assignment of the 15-day activity is in accord with beta-disintegration energy systematics, as pointed out by Way.[15,‡]

TABLE I. Energies in kev of electron conversion lines from Os^{191} and Os^{191m} activities.

Gamma-transition	Sub-shell origin of conversion electrons K	L_I	L_{II}	L_{III}	M_I	M_{II}	M_{III}	N
41.70 kev		···	28.85	30.48	···	38.80	39.19	41.15
74.17 kev		61.24	61.78	63.36	71.09	···	71.72	73.45
129.1 kev	53.15	115.49	116.14	117.97		126.29		128.76

* Assisted by the joint ONR and AEC program.
† Fulbright and University of Illinois Postdoctoral Fellow. On leave from University of Western Australia, Perth, Australia.
[1] Kurtchatow, Latyschew, Nemenow, and Selinow, Physik Z. Sowjetunion **8**, 589 (1935).
[2] E. Zingg, Helv. Phys. Acta **13**, 219 (1940).
[3] G. T. Seaborg and G. Friedlander, Phys. Rev. **59**, 400 (1941).
[4] L. J. Goodman and M. L. Pool, Phys. Rev. **70**, 112 (1946); **71**, 288 (1946).
[5] Cork, Schreffler, and Fowler, Phys. Rev. **72**, 1209 (1947).
[6] L. I. Katzin and M. Pobereskin, Phys. Rev. **74**, 264 (1948).
[7] D. Saxon, Phys. Rev. **74**, 1264 (1948); Atomic Energy Commission Report, AECD, 1860 (1948) (unpublished).
[8] Mandeville, Sherb, and Keighton, Phys. Rev. **74**, 888 (1948).
[9] Bunker, Canada, and Mitchell, Phys. Rev. **80**, 126 (1950); **79**, 610 (1950).
[10] F. K. McGowan, Oak Ridge National Laboratory Report, ORNL-952 (1951) (unpublished); Phys. Rev. **79**, 404 (1950).
[11] T. C. Chu, Phys. Rev. **79**, 582 (1950).
[12] E. Kondaiah, Ark. Fys. **3**, 47 (1951).
[13] *Nuclear Data*, National Bureau of Standards, Circular No. 499 (1950), p. 222.
[14] R. A. Becker, Rev. Sci. Instr. **22**, 773 (1951).
[15] K. Way, private communication and reference 13.
‡ *Note added in proof:*—A revision of the original Os^{191}, Os^{193} assignment by Seaborg and Friedlander (see reference 3) was later made by G. Friedlander (quoted in *Table of Isotopes*, Seaborg and Perlman, Revs. Modern Phys. **20**, 585 (1948)). According to J. M. Hollander (private communication) this revision was also made on the basis of γ-irradiations.