Electron Spectra of Pu^{239}, Pu^{240} and Pu^{241}

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Two plutonium samples, containing different amounts of Pu^{239}, Pu^{240}, and Pu^{241}, have been analyzed on a 180° magnetic β-spectrometer. Using thin celluloid films and radioactive sources, spectra were taken in an energy range beginning at ≈ 1 kev.

From the conversion lines it was possible to determine the following energies for γ-transitions in U^{235} and U^{236}: for U^{235} – 3 (?), 12.5, 38.3, 50.8, 117 (?) kev and for U^{236} – 44.6 kev.

The Pu^{241} β-spectrum has the allowed shape with end point $E_0 = 20.8$ kev.

INTRODUCTION

IMPORTANT data about the energy levels of radioactive nuclei can be obtained both from their electron spectra and their α and γ spectra. Thus, by observing the spectra of conversion electrons from α-active isotopes of plutonium, one can obtain information about γ-transitions in the uranium isotopes. However, up to the present time, no investigations of the electron spectra from Pu^{239}, Pu^{240}, or Pu^{241} have been published.

We summarize briefly the published work on the decay of Pu^{239}, Pu^{240}, and Pu^{241}. The fact that Pu^{239} is α-active was first apparent in the years 1944–1946. The results of Ghiorso, Sullivan et al,[1] and Rosenblum, Valadares and Goldschmidt [2] indicate that the α-spectrum is complicated. According to their results, U^{235}, the daughter nucleus of Pu^{239}, should emit γ-rays of energies about 0.05, 0.2 (weak), 0.3 (weak) and 0.42 (weak) mev. According to Skaggs et al[3], the half life of Pu^{239} is about 24,400 years.

The only information available about Pu^{240} was that it was α-active, with a half life of about 6000 years.[4]

Seaborg, James and Morgan[5] showed tha Pu^{241} is mostly β-active (the α-activity amounting to only 0.002% of the total) with a half life of about 10 years. The electron energy is 10 to 20 kev.

It was with the above scarcity of information about the radiations from these isotopes in mind, that the present work was undertaken.* The results of several other investigations, carried out concurrently with the one described here, and also in the last year, agree with ours.

Albouy and Teillac[6], and Dunlavey and Seaborg[7] used electron sensitive emulsions to detect the conversion electrons, and found transitions in U^{235} (obtained by the α-decay of Pu^{239}) at energies about 35, 50, 115 and 200 kev. Using a proportional counter, West and Dawson[8] found α-rays of energies 38.5, 51.8, 101 (?) and 115 (very weak) kev for U^{235} and another at 45.1 kev belonging, apparently, to U^{236} (from the α-decay of Pu^{240}). The α-spectra of Pu^{239} and Pu^{240} have been examined with a magnetic α-spectrometer by Asaro and Perlman[9] and also by Gol'din, Tretiakov and Novikov.[10]. Their results confirm the fact that these spectra are very complicated, and point to the possibility that U^{235} might have γ-rays at about 13, 38, and 51 kev, while U^{236} might have a γ-ray at about 45 kev.

The only work besides ours, on the electron spectra of Pu^{239}, Pu^{240}, and Pu^{241} was carried out by Freedman, Wagner and Engelkemeir,[11] on a magnetic lens β-spectrometer. These authors also investigated the γ-rays, using a proportional counter and a scintillation spectrometer. γ-transitions were found at 39, 53, 100 and 120 kev for U^{235}, 46.9 kev for U^{236}, and 100 and 145 kev for U^{237} (from the decay of Pu^{241}). The β-spectrum of Pu^{241} taken by these authors is strongly distorted. According to their data, the upper limit of the β-spectrum is at 20.5 kev. The electron spectrum is not shown in their paper.

There are great experimental difficulties in the study of the soft β-radiations from the decay of plutonium, which might explain why so little work has been done on them.

1. EXPERIMENTAL TECHNIQUE

a) β-SPECTROMETER

The electron spectra reported in this paper were taken on a conventional, magnetic β-spectrometer with 180° focussing. The middle electron orbit had a radius of curvature of 15 cm. The spectrometer resolution, as measured by the relative half width of the Hg^{198} conversion line ($E_\gamma = 411.2$ kev Au^{198}), was about 2% in one experiment and about 1% in the other. The same conversion line was used to calibrate the spectrometer. The relative solid angle was about 0.06% of 4π. A mercury diffusion pump, with a liquid air trap, gave a vacuum of about

*The work was done in 1951.

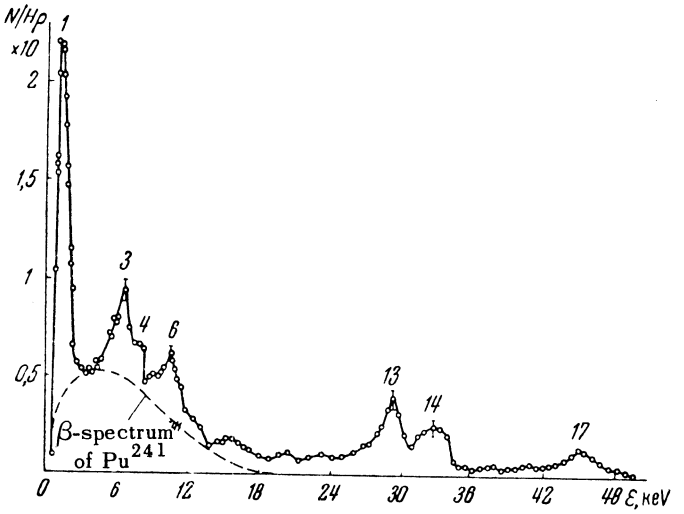

FIG. 1. Electron spectrum of sample No. 1 (source on film)

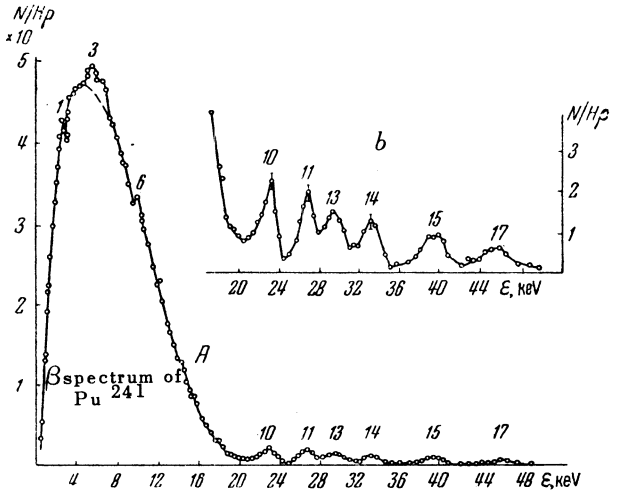

FIG. 2. Electron spectrum of sample No. 2 (source on film)

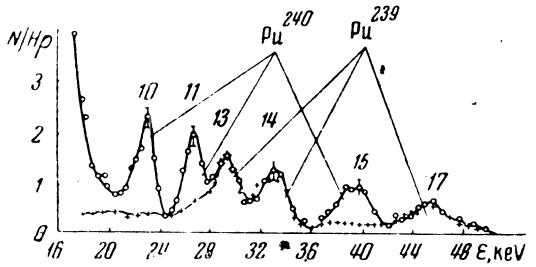

FIG. 3. Superposition of spectra from samples No. 1 and 2

10^{-5} mm Hg in the spectrometer. A constant current electromagnet, made of "Armco" soft iron, generated the magnetic field. The electromagnet current was supplied by high capacity storage batteries ($\sim$3000 amp-hr). The field was measured to 1% by a search coil and ballistic galvanometer.

A Geiger-Muller counter, with a slit to admit electrons, was used as a detector. The celluloid window was $5\,\mu\ g/cm^2$ thick, and was supported by a wolfram wire net (wire thickness 0.04 mm, separation 0.3 mm) giving about 80% transmission. The counter was filled with a gaseous mixture of argon (90%) and ethyl alcohol (10%) at a pressure of 20 to 40 mm. Hg. The counter pulses were recorded by a conventional electronic circuit.

b) PREPARATION OF SOURCES

Plutonium is usually prepared by irradiating uranium with slow neutrons. Pu^{239} is formed by the reaction

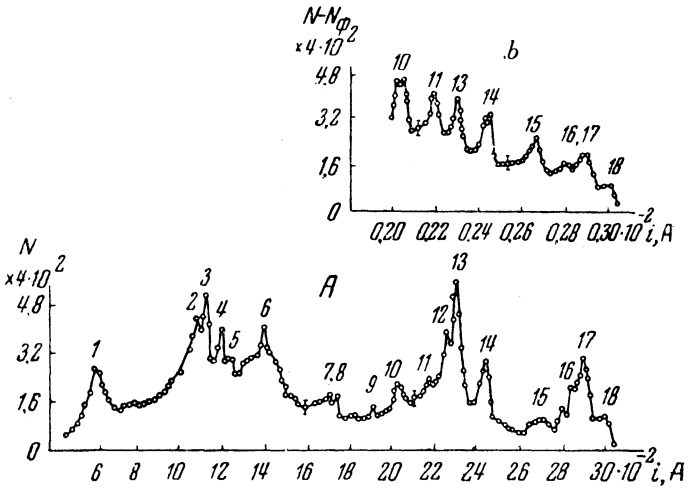

FIG. 4. Electron spectra: A – sample No. 1, B – sample No. 2; sources on aluminum foil. Spectrometer resolution ~ 1%.

$$U^{238}(n, \gamma)\, U^{239} \xrightarrow[23.5\ \text{min}]{\beta} Np^{239}$$

$$\xrightarrow[2.3\ \text{days}]{\beta} Pu^{239}\ (T \sim 24\,400\ \text{yr}).$$

Further neutron irradiation of Pu^{239} leads to the formation of Pu^{240} and Pu^{241}:

$$Pu^{239}(n, \gamma)\, Pu^{240}\ (T \sim 6000\ \text{yr});$$

$$Pu^{240}(n, \gamma)\, Pu^{241}\ (T \sim 13\ \text{yr}).$$

Two plutonium samples, No. 1 and No. 2, were used in the present experiment. They differed in the relative amounts of Pu^{239}, Pu^{240}, and Pu^{241} contained, sample No. 2 being richer in the Pu^{240} and Pu^{241} isotopes than sample No. 1. The plutonium samples were carefully purefied chemically to rid them of products from the fission of uranium and plutonium. The radioactive source (in the form of a strip 1 to 2 × 30 mm^2) was precipitated out from an aqueous solution of plutonium nitrate on a celluloid film 10 $\mu g/cm^2$ thick. To avoid charging, a layer of aluminum (about 1 $\mu g/cm^2$ thick) was evaporated in vacuum on the radioactive source. The surface density of the sources was measured by their α-activity and was ~ 3 to 10 $\mu g/cm^2$. Each run was long because the sources were weak.

2. RESULTS

Figures 1 and 2 show the electron spectra from samples No. 1 (surface density ~6 g/cm^2) and No. 2 (surface density ~ 3 g/cm^2) taken on the spectrometer with 2% resolution. Part of the spectrum from No. 2, starting from 18 kev, is shown in Fig. 2 on an enlarged scale (curve B). In the energy interval from 1 to 50 kev, the figures show a β-spectrum and several conversion lines.*

In order to interpret lines 10, 11, 13, 14, 15, 17, the electron spectra were superposed in the energy region from 18 to 50 kev (Fig. 3).

From a knowledge of the concentrations of the various plutonium isotopes in the samples used, together with the intensities and positions of the lines in the β-spectra, it was possible to establish the following:

a) lines No. 1, 3, 4, 6, 13, 14, 17 are conversion lines from U^{235}.

b) lines 10, 11, 15 are conversion lines of U^{236}.

c) the β-spectrum shown in Figs. 1 and 2 is that of Pu^{241}.

It was difficult to determine the γ-transition energies accurately from the conversion lines in Figs. 1 and 2, because these lines were poorly resolved. Because of this, we took the spectra of the same samples (Nos. 1 and 2) again, this time with better resolution (~ 1%). Figure 4 shows these spectra (curve A for sample No. 1, curve B for sample No. 2), using sources with surface densities ~ 10 g/cm^2. An aluminum foil was used as source backing.

In addition to the earlier, intense lines, several new, weak lines appear in the spectra of Fig. 4. The results of measurements on the conversion lines of the Pu^{239} and Pu^{240} isotopes are shown in the table. In the spectra of all the figures,

*The electron spectrum of Pu^{240} does not appear in Fig. 1 because little of this isotope was present.

TABLE

Interpretation of lines in the electron spectra of plutonium samples Nos. 1 and 2.

Line number from Fig. 4		Electron kinetic energy (in kev)	Conversion Shell	Transition energy in kev		Plutonium isotope	Notes
Line	Curve						
1	*A* Not Shown	~2 ~95(?)	*K* *L*	117 117 117(?)		Pu^{239}	Very weak radiation
1	*A*	~2	*N,O*	~3(?)		Pu^{239}	
2	*A*	6.7	M_I	12.3			
3	*A*	7,2	M_{II}	12.4			
4	*A*	8,3	M_{III}	12.6	12.5	Pu^{239}	
5	*A*	8,8	$M_{IV,V}$	12.4			
6	*A*	11,1	N_I	12.5			
7	*A*	16.7	L_I	38.5			
8	*A*	17.4	L_{II}	33,3	38.3	Pu^{239}	Very weak radiation
9	*A*	20.9	L_{III}	38.1			
10	*A*	23	L_I	44.8			
10	*B*	23.8	L_{II}	44.7			
11	*B*	27.3	L_{III}	44.5		Pu^{240}	
11	*A*	27.1	L_{III}	44.3	44.6		
15	*B*	40.4	$M_{I\div V}$	~45			
15	*A*	40.4	$M_{I\div V}$	~45			
16	*B*	44.2	*N,O*	~45			
12	*A*	28.8	L_I	50.6			
13	*A*	29.8	L_{II}	50.7			
13	*B*	29.9	L_{III}	50.8			
14	*A*	33.6	L_{III}	50.8			
14	*B*	33.9	L_{III}	51	50.8	Pu^{239}	
17	*B*	46.5	$M_{I\div V}$	~51			
17	*A*	49.5	$M_{I\div V}$	~51			
18	*A*	50.7	*O*	~51			
18	*A*	50.9	*O*	~51			

1–4, identical conversion lines are labeled by the same indices,

In the energy region 0.05 to 2 mev, none of the electron lines from Pu^{239}, Pu^{240} or Pu^{241}, nor any of the β-spectra, were more than a factor 2 above the background.

The most intense line, 1 (see Fig. 1 at electron energy about 2 kev), is difficult to interpret because it is somewhat different from the nearest conversion lines. If we take line 1 to be a K-conversion line, then the corresponding γ-ray energy, as calculated from the energy of the conversion electrons, is about 117 kev. The presence of a weak L-conversion line (electron energy ~ 95 kev, intensity

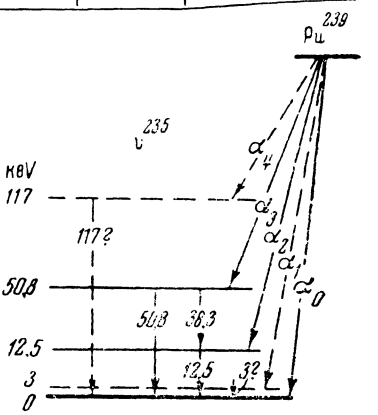

FIG. 5. Decay scheme of Pu^{239}

$\sim$ 1.5 times the background) lends some support to this view. Approximate calculations indicate that the intensity of the K-conversion line should be considerably less than the observed intensity of line 1. Hence, we cannot take line 1 as due only to K-conversion electrons. Similarly, we are not justified in considering line 1 to be due only to Auger-electrons. Similar Auger-lines should appear at energies > 3 kev, but are not observed on the experimental spectra.

Hence, it is difficult to explain the large intensity* of line 1, except as due to K-conversion and Auger-electrons combined.

We cannot exclude the possibility that line 1 is due to conversion electrons from the N and O shells. In this case, the γ-ray energy would have to be about 3 kev. If such a ray is actually present in the γ-decay of Pu^{239}, we would have to assume that one of the excited levels in U^{235} lies $\sim$ 3 kev above the ground state. Another effect that might contribute significantly is the ionization of plutonium when it decays by α-emission. These conjectures need confirmation.

The data in the table show that the observed conversion lines are associated with the following γ-transitions in U^{235} (from the α-decay of Pu^{239}): 3(?), 12.5, 38.3, 50.8, 117 (?) kev and in U^{236} (from the α-decay of Pu^{240}) – 44.6 kev. The 12.5 kev γ-ray in U^{235}, which certainly exists, is reported here for the first time. The 3 and 117 kev transitions need to be checked. Figure 5 shows the proposed α-decay scheme of Pu^{239}. A Kurie plot of the Pu^{241} β-spectrum shows that this spectrum has the allowed shape.** The experimental points in this graph lie close to a straight line in the energy interval 3 to 19 kev. The upper limit of the β-spectrum is at 20.8 $\pm$ 0.2 kev.

In conclusion it is my pleasant duty to express my gratitutde to G. N. Iakovlev and G. A. Chistiakova for preparing the chemically pure plutonium samples.

*The observed line intensity is considerably lessened by electron absorption in the source itself and in the window of the Geiger-Muller counter.

**A more detailed discussion of the Pu^{241} β-decay will be given in another paper.[12]

Note added in proof: The level sequence 0, 50.8 and 117 kev can be described by the well known theoretical formula of Bohr and Mottelson[13] for the rotational energy levels of nuclei. Assuming that the ground state of U^{235} is the first rotational level, with spin 7/2 $\pm$,[14] then the spins of the first and second levels will be 9/2 $\pm$ and 11/2 $\pm$. In our case, the ratio $E_2 : E_1$ for these levels is 117: 50.8 = 2.3, which agrees with the value 2.22 given by the Bohr–Mottelson formula. Better agreement is obtained by taking the spin of the first rotational level to be 5/2 $\pm$.

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