Isotopes of Einsteinium and Fermium Produced by Neutron Irradiation of Plutonium

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(Received December 30, 1955)

Samples of einsteinium and fermium have been separated from neutron-irradiated plutonium and carefully purified. Four isotopes of einsteinium were found: 20.03-day E^{253}, emitting 6.636-Mev and 6.24-Mev alpha particles and 42- and 393-kev gamma rays, both magnetic dipole transitions, and with a spontaneous fission half-life of 7×10^5 years; 38.5-hr E^{254m}, a beta emitter with 1.04-Mev beta rays and 0.680-Mev gamma rays; ~320-day E^{254}, an alpha emitter with 6.42-Mev alpha particles; and 24-day E^{255}, a beta emitter. Two fermium isotopes were found: 3.24-hr Fm^{254} emitting 7.20-Mev alpha particles and with a spontaneous fission half-life of 246 days; and 21.5-hr Fm^{255} emitting 7.08-Mev alpha particles.

INTRODUCTION

EINSTEINIUM and fermium (atomic numbers 99 and 100) have recently been prepared by neutron irradiation[1,2] and by charged-particle bombardments.[3,4] To confirm and extend measurements of their nuclear properties, isotopes of these elements which can be prepared by neutron irradiation were extracted from a 348-mg sample of Pu which had been irradiated to an integrated flux of 1.46×10^{22} neutrons/cm^2 in the Materials Testing Reactor at the National Reactor Testing Station in Idaho.

The work formed part of a joint project of Atomic Energy of Canada, Limited and the Knolls Atomic Power Laboratory, and was a part of the technical cooperation program of the United States Atomic Energy Commission.

When einsteinium and fermium are prepared by neutron irradiation of Pu, a sequence of neutron capture reactions and β-decay processes occurs leading from Pu through Am, Cm, Bk, and Cf to E and Fm. A number of the members of this series undergo neutron fission so that the einsteinium and fermium produced are contaminated with large quantities of fission products, as well as α-emitting actinide elements. For example, in the present experiments there were about 20 curies of β activity and 740 mC of α activity, largely Cm^{242-4}, in the sample thirty hours after the end of the irradiation. The radiation field 3 feet from the sample was 19 roentgens per hour. It was first necessary therefore to purify E and Fm scrupulously before studying their nuclear properties.

* The Knolls Atomic Power Laboratory is operated by the General Electric Company for the U. S. Atomic Energy Commission.

[1] Thompson, Ghiorso, Harvey, and Choppin, Phys. Rev. **93**, 908 (1954).

[2] Studier, Fields, Diamond, Mech, Friedman, Sellers, Pyle, Stevens, Magnusson, and Huizenga, Phys. Rev. **93**, 1428 (1954).

[3] Ghiorso, Rossi, Harvey, and Thompson, Phys. Rev. **93**, 257 (1954).

[4] Atterling, Forsling, Holm, Melander, and Aström, Phys. Rev. **95**, 585 (1954).

EXPERIMENTAL METHODS

1. Chemical Purification

The chemical steps used to purify E and Fm from fission products, other actinide elements, and about 25 g of aluminium that clad the sample during irradiation, are given below. Steps (a) through (d) were done by remote control with 6 inches of steel shielding that reduced the radiation field at 3 feet to 80 mr per hr.

(a) The aluminium was dissolved in 175 ml of 5*M* NaOH, 3.5*M* $NaNO_3$ solution in a stainless-steel vessel fitted with a stainless-steel reflux condenser. Towards the end of the reaction the mixture was boiled to ensure complete solution. The solution was centrifuged and the insoluble residue washed with KOH solution.

(b) The insoluble residue was dissolved in 13*M* HCl-0.3*M* HNO_3. The lanthanide and actinide elements were then precipitated first as fluorides and, after dissolution in H_3BO_3-HNO_3 mixture, finally as hydroxides. The hydroxides were dissolved in 9.5*M* HCl-0.1*M* HNO_3.

(c) The plutonium was removed by passing the acid solution through an anion-exchange resin column.[5] Dowex *A*-1, 8% cross-linked, 100–200 mesh was used in a column 0.8 cm^2 in area and 5 cm long; the flow rate was 1 cm per min.

(d) The transplutonium elements were separated from fission-product lanthanide elements by cation exchange[5] using Dowex 50 12% cross-linked, in a column of the type shown in Fig. 1(A). The particle size was selected by grading the hydrogen form of the resin in an up-flow of water and the fraction with a settling rate between 0.75 and 1.2 cm per min was used. The elution was then done with conc. HCl at a flow rate of 0.3 cm/min.

(e) Individual actinide elements were separated from each other by cation exchange.[6] The *trans*-curium isotopes were separated from the bulk of the curium on a

[5] Diamond, Street, and Seaborg, J. Am. Chem. Soc. **76**, 1461 (1954).

[6] Thompson, Harvey, Choppin, and Seaborg, J. Am. Chem. Soc. **76**, 6229 (1954).

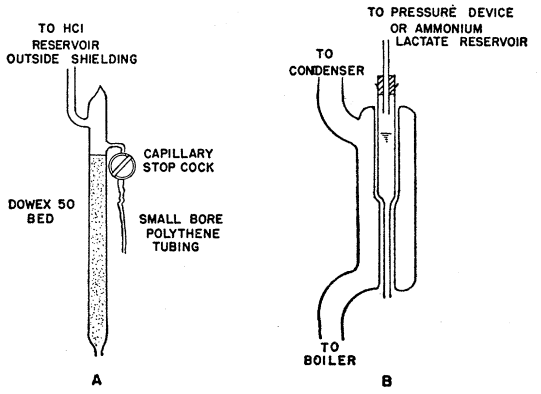

FIG. 1. (A) Cation exchange column (0.82 cm² cross-sectional area, 20 cm long) used for separation of actinide and lanthanide elements by remote control. Rinse and feed solutions are transferred to the top of the bed via the polythene tube and capillary stopcock by suction through the line to the HCl reservoir. Activity is eluted by closing the stopcock and applying pressure to the HCl reservoir. (B) Cation exchange column used for separation of individual actinide elements. Trichlorethylene vapor from the boiler maintains jacket temperature at 87°C. The interchangeable ion exchange column fits snuggly into the heating jacket and contains enough preheated eluant for a normal separation.

Dowex 50 column 0.78 cm² in area, 8.0 cm long at 87°C using ammonium lactate (0.4*M*, *p*H 4.58) as the eluant. The final separation of trans-curium actinides was done in a column of area 0.031 cm², 5 cm long [Fig. 1(B)] using white Dowex 50, 12% cross-linked, settling in water at a rate of 0.30–0.60 cm/min, with ammonium lactate (0.4*M*, *p*H 4.18) as eluant. A typical elution curve is shown in Fig. 2. To obtain pure samples of einsteinium and fermium, one or more of steps (b) to (e) were repeated where necessary.

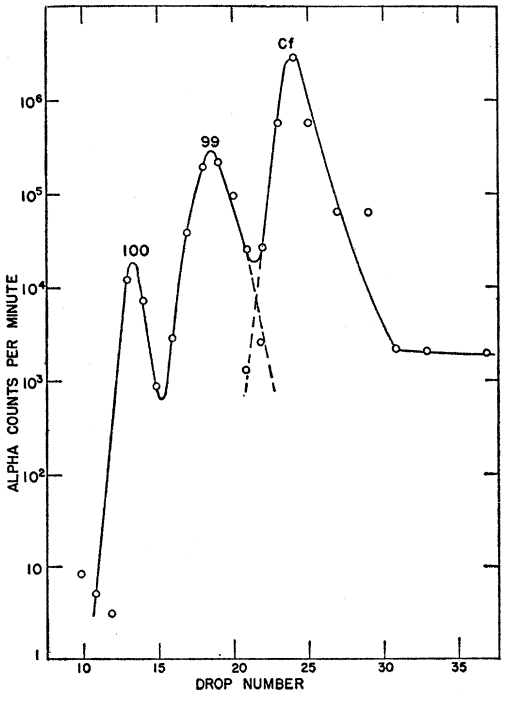

FIG. 2. Typical elution curve of transcurium elements from a Dowex 50 ion exchange column using 0.4*M* ammonium lactate at *p*H 4.15 as eluant. The drop size was 0.028 ml and the flow rate 0.5 cm/min. The points on the dotted lines between the Cf and E peaks were obtained by α-pulse analysis.

2. Counting Techniques

Alpha-particle spectra were measured by analysis of the pulses from a gridded ion chamber.[7] The signal from the chamber was fed first to a low-noise preamplifier and then to the main amplifier. The amplified pulses were "cut" by a biased diode and the "tops" further amplified before passing to a 30-channel pulse-height analyser.[8] Since the counting rates were often low, high stability was required of the equipment. The pulse size from a particular α emitter varied less than 0.2% in 24 hours.

Sources for the determination of α-particle energies were prepared by sublimation in vacuum from a tantalum filament onto a polished platinum disk. The resolution obtained with a Ra^{226} source, for example, prepared in this way was 0.63% full width at half-maximum (i.e., 30 kev at 4.78 Mev). Because of the small amount available, the determination of the α-particle energy of Fm^{255} was made with a source prepared by direct evaporation of a lactate solution of fermium including a little E^{253}, followed by ignition at about 700°C. The source so prepared was thicker than a sublimed source but a thickness correction was estimated from the 6.636-Mev α-particle peak of E^{253} and applied to the observed energy of Fm^{255}.

For each energy determination a pulse generator (stability 1 part in 10^4) was calibrated against Ra^{226}, Em^{222}, Po^{214} and Po^{218} α particles, and then the unknown was measured by bracketing the observed α-particle peak by pulse-generator peaks. The calibration and interpolation was done by least squares fitting. This procedure was repeated several times and the average taken. Source thickness and positive ion effect corrections were then applied to arrive at the final value of the energy.

When the decay of an alpha-emitting nuclide or its beta-emitting precursor was being observed by pulse analysis the sum of the counts over an α-particle peak, corrected for background, was used. A methane flow proportional counter, with about 2π geometry, was used for gross alpha-particle counting.

Routine β-particle counting was done with a methane flow proportional counter or an end-window G-M tube. A G-M tube, shielded by 4 inches of lead, with a steady background of 14 counts/min, was used when the β-disintegration rates were low.

The detecting equipment used for α, β, and γ scintillation spectrometry was as follows:

[7] Harvey, Jackson, and Eastwood, Can. J. Phys (to be published).

[8] Moody, Battell, Howell, and Taplin, Rev. Sci. Instr. 22, 551 (1951).

TABLE I. Nuclear properties of einsteinium and fermium.

Nuclide	Disintegrations per minute at end of the irradiation	Observed half-life	Period of observation (number of half-lives)	Method of measuring half-life	Primary mode of decay	Observed energy of particle (Mev)	Observed fission half-life
E^{253}	1.53×10^6	20.03±0.01 days	2	Gross α decay of E source	α	6.636±0.005 6.24 ±0.02	$(7\pm3)10^5$ yr
E^{254m}	2.45×10^5	38.5 ±1.0 hr	7	Gross β decay of E source	β	1.04 ±0.04	
		37 ±2 hr	6.5	Pulse analysis of equilibrium Fm^{254}			
		42 ±4 hr	2	Fission counting of equilibrium Fm^{254}			
E^{254}	180	320 days		α pulse analysis	α	6.42 ±0.02	
E^{255}	180	24 ±2 days	4.5	α pulse analysis of equilibrium Fm^{255}	β		
Fm^{254}	2.50×10^5	3.24±0.01 hr	7	Gross α-decay of Fm source	α	7.20 ±0.01	246 days
Fm^{255}	180	21.5 ±0.1 hr	10	Pulse analysis of α-decay of Fm source	α	7.08 ±0.01	

(a) a 3-inch by 3-inch cylindrical NaI(Tl) crystal with $\frac{1}{4}$ in.$\times1\frac{1}{2}$ in. re-entrant hole in the center of one of its circular faces and a Dumont type 6363 photomultiplier tube, as well as a 2 in.$\times$2 in. cylindrical NaI(Tl) crystal and Dumont type 6292 photomultiplier tube for x-ray and π-ray spectra.

(b) a 1-in.$\times\frac{3}{4}$-in.$\times\frac{1}{2}$-in. thick anthracene crystal with an RCA type 6199 photomultiplier tube for β spectra, and

(c) a $1\frac{1}{4}$-in. diameter, $\frac{1}{8}$-in. thick KI(Tl) crystal for α spectra.

To reduce the background, a shield of 2 inches of mercury and 4 inches of lead could be used if required.

The pulses from the detectors were analyzed and displayed in a manner similar to that used for α-spectrum analysis. For α-γ or β-γ coincidence studies, a suitable combination of detectors was used with standard coincidence counting techniques.

Gamma rays from Na^{22}, Zn^{65}, Cs^{137}, Ce^{144}, Au^{198}, and Am^{241} were used for the energy calibration of the NaI scintillation spectrometer, beta particles from P^{32} and Au^{198} for the β spectrometer and α particles from Am^{241} for the α spectrometer. The α-γ and β-γ coincidence circuits were set up with Am^{241} and Au^{198}, respectively. Sources of Na^{22}, Co^{60}, Cs^{137}, Ce^{141}, Au^{198}, and Am^{241} of known disintegration rate were used to determine the efficiency of the γ spectrometer.

The spontaneous fission counter was a simple ionization chamber filled with methane. The gas pressure was so adjusted that both the α particles and fission fragments had ranges greater than the spacing between the electrodes; then the larger amount of ionization produced by fission fragments at the beginning of their range gives a pulse several times larger than that produced by an alpha particle.

By using an amplifier with an integrating and clipping time constant of 10^{-7} sec, good discrimination between α particles and fission fragments could be achieved in the presence of 10^8 α disintegrations per minute. To

TABLE II. Previously reported values of einsteinium and fermium properties.

Nuclide	Half-life	Primary mode of decay	Energy of emitted particle Mev	Fission half-life
E^{253}	19.3±0.3 days[a] 20 days[b]	α	6.61±0.01[a] 6.63±0.02[b]	10^5 yr[a]
E^{254m}	37 ±1 hr[a] 36.3 hr[b]	β	1.1 ±0.1[b]	
E^{254}	2 yr[c]	α	6.44±0.01[c]	
E^{255}	30 days[b]	β		
Fm^{254}	3.3±0.2 hr[a]	α	7.17±0.01[a] 7.22±0.03[b]	220±40 days[a] 200 days[b]
Fm^{255}	15 hr	α	7.1[b] 7.1[d]	

[a] See reference 9.
[b] See reference 11.
[c] See reference 10.
[d] See reference 2.

TABLE III. Observed characteristics of einsteinium gamma rays.

Gamma-ray energy (kev)	Particle in coincidence with gamma ray	Approx. half-life	Abundance relative to: E^{253} α's	E^{254m} β's	E^{255}–Fm^{255}	E^{254}–Bk^{25}
21±2	α, β	20 days	6.5×10^{-3}		50	
42±3	α	20 days	3.8×10^{-4}		3	
115±10	α, β	20 days	3.6×10^{-4}		3	
393±5	α	20 days	5.5×10^{-4}		5	
660±15	β	38 hr		0.7		
975±50	β	56 days				0.63

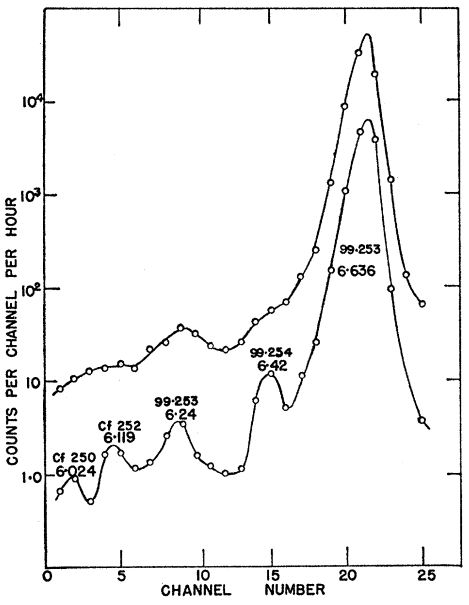

FIG. 3. Typical α spectra of an einsteinium source. The upper curve was obtained 21 days after the end of the irradiation and the lower one 64 days later. Each peak is labeled with the observed energy in Mev and the assignment.

determine the counting geometry, α particles from a Po^{210} source of known disintegration rate were counted in the chamber at increased pressure. It was found to be 48.2% for the α-emitting source, or 96.4% for a spontaneous fission source. A 24-channel pulse-height analyzer was used to record counts of short duration while a Brush Recording Oscillograph was used for long counts.

RESULTS

A summary of the results of this investigation is given in Table I, and for comparison, previously reported values are shown in Table II.[2,9–11]

The yields shown in the first column of Table I are extrapolated to the end of the irradiation by using the quoted half-lives. Where the half-life of a particular nuclide has been measured by different methods, all observed half-lives and the respective methods of measurement have been given, mainly to confirm genetic relationships. The first value given is the most reliable and except for E^{254} and E^{255} was obtained by analyzing the experimental data by the method of least squares. The errors are the standard deviations derived from the least squares fit. To give an indication of the radiochemical purity of the sources used, the number of half-lives over which the decay was observed is also given in the table. More detailed comments follow outlining the reasons for the assignments given in Table I.

Einsteinium.—Typical α spectra of an einsteinium source are shown in Fig. 3. In addition to the 6.636-Mev peak, a 6.24-Mev peak is apparent; and as the 6.636-Mev peak decays, the 6.42-Mev peak becomes better resolved.

Figure 4 shows a typical γ-ray spectrum of an einsteinium source shortly after the end of the irradiation. The γ-ray spectrum in coincidence with α and β particles has been investigated and the decay of the various rays has been followed; the results are summarized in Table III. The ratios of the disintegration rate of the various γ rays to the disintegration rate of isotopes of einsteinium of about the same half-life are also given. Since the activity of the einsteinium source was low, both the half-life and abundance values are only approximate.

Einsteinium-253.—The previously unreported 6.25-Mev α peak decays with a half-life of about 20 days, just as does the 6.636-Mev peak, and parallels the growth and decay of the 6.636-Mev peak of einsteinium in a Cf fraction containing the β emitter Cf^{253}; therefore it has been assigned to E^{253}.

Four photon peaks decay with a 20-day half-life and are in coincidence with α particles. These are probably associated with E^{253}. The alternative to this assignment, Fm^{255}, can be ruled out because the disintegration rates, as shown in Table III, are too high. Additional evidence concerning the assignment of the 393-kev peak to Bk^{249}, the daughter of E^{253} is as follows: (a) γ-α coincidence measurements show the 6.25-Mev α peak is in coincidence with γ rays of about 400-kev energy. (b) A bias curve of γ rays in coincidence with α particles shows a sharp break at about 400-kev γ energy. (c) The ratio of the 6.25-Mev to 6.636-Mev α particles was found to be $0.08\pm0.02\%$ by pulse analysis with the gridded ionization chamber and $0.09\pm0.02\%$ from the α-scintillation spectrum with and without coincidences with γ rays; the ratio of 393-kev γ-disintegration rate (including the fraction internally converted) to the α-disintegration rate was $0.09\pm0.02\%$. The 21- and 115-kev photon peaks are probably L and K x-rays of Bk arising from internal conversion of the 42- and 393-kev γ rays. The $L_{\alpha 1}$, $L_{\beta 1}$, $L_{\gamma 1}$, and K x-ray energies of Bk are calculated to be 15.3, 20.0, 23.4, and 112 kev, respectively.[12]

Contributions to the x-ray intensity from the 660- and 975-kev γ rays are small because the 660-kev peak had largely decayed when the abundances were measured, and the 975-kev peak is in low abundance and is unlikely to be highly converted. From the abundance of the 112- and 393-kev photons, the conversion ratio is calculated to be approximately 0.66. Assuming that the 393-kev peak is a single γ ray, this would indicate a magnetic dipole transition with some $E2$ admixture.[13] Conversion of the 393-kev γ ray therefore contributes little to the

[9] Fields, Studier, Méch, Diamond, Friedman, Magnusson, and Huizenga, Phys. Rev. **94**, 209 (1954).
[10] Harvey, Thompson, Choppin, and Ghiorso, Phys. Rev. **99**, 337 (1955).
[11] Choppin, Thompson, Ghiorso, and Harvey, Phys. Rev. **94**, 1080 (1954).
[12] S. Fine and C. F. Hendee, Nucleonics **13**, No. 3, 36 (1955).
[13] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **38**, 79 (1951).

L x-ray peak. Assuming that the fluorescence yield for Bk is approximately 50%, about that for U,[14] the K-conversion coefficient for the 42-kev γ ray is 34. The transition is probably magnetic dipole, although the observed coefficient is smaller than the value of 100 obtained by extrapolation from published tables.[15] Speculation about the decay scheme of E^{253} is of little value until more sensitive measurements of the γ and α groups have been made.

The fission half-life was determined by fission-counting an einsteinium source 47 days after the irradiation so that the contribution from Fm^{254} was negligible.

A portion of the einsteinium produced in the original irradiation was reirradiated in the *NRX* reactor at Chalk River, Canada after the E^{254m} had decayed. The yield of E^{254m} from the irradiation was determined by fission counting and also by alpha pulse analysis of the equilibrium Fm^{254}. The cross section for E^{254m} production from E^{253} was thus determined to be 450×10^{-24} cm^2 for *NRX*-reactor-spectrum neutrons, considerably larger than the value of 240×10^{-24} cm observed for irradiations in the MTR.[9]

Einsteinium-254 and 254m.—Evidence for the existence of two isomeric states of E^{254} has recently been found.[10] One state, thought to be the ground state, decays by the emission of a 6.44-Mev α particle with a half-life of about 2 years. The other, probably an excited state, decays mainly by β^- emission, but also by electron capture. In the present investigation both states were observed; the 6.42-Mev α peak in the einsteinium spectrum is evident in Fig. 3. Alpha particles of this energy are absent from the spectrum of a decaying Cf source, which of course contains einsteinium of mass 253 only, thus tending to confirm the mass assignment.

The recoil nuclei from the α decay of an einsteinium source were collected by placing a platinum plate close to and parallel with the einsteinium source *in vacuo*. The source so prepared decayed by β emission with the 3-hour half-life of Bk^{250},[11] the daughter of E^{254}. Furthermore, a beta emitter with radiations similar to those of Bk^{250} was found in a highly purified einsteinium source shortly after separation and was observed to decay with the half-life of E^{254}.

The maximum β-particle energy of E^{254m} was determined by Feather analysis of an Al absorption curve obtained with a standard end-window G-M tube. To minimize interference from the radiations produced by the spontaneous fissions of Fm^{254}, a freshly prepared source was used and counting was done as rapidly as possible.

The 660-kev peak in the γ spectrum of einsteinium (Fig. 4) decays with a half-life of about 40 hours, the half-life of E^{254m}, and it has therefore been assigned to this nuclide. The ratio of the photon disintegration rate to the β disintegration rate of E^{254m} was not measured accurately but is between 0.6 and 0.8. The absence of a component of about 360-kev maximum energy in the β-absorption curve supports the high figure for the ratio and indicates that a large fraction of the E^{254m} β particles are followed by a γ ray of 660 kev.

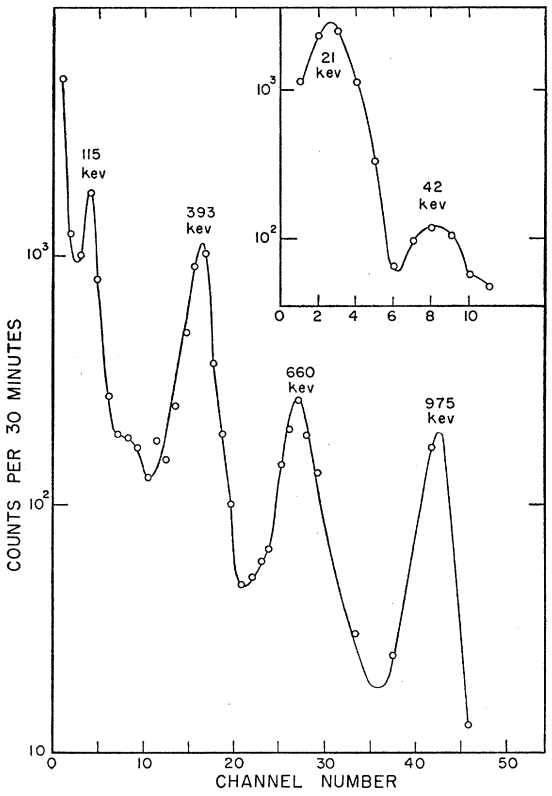

FIG. 4. A typical γ spectrum of einsteinium observed with a NaI(Tl) scintillation spectrometer fourteen days after the end of the irradiation. The low-energy portion of the spectrum has been expanded and is shown in the insert.

Fermium-255.—The α-particle peak in the spectrum of this nuclide at 7.08 Mev is significantly wider than would be expected for a single α group even in a source prepared by evaporation, and could be interpreted as representing at least two groups separated by about 35 kev.

ACKNOWLEDGMENTS

The authors wish to acknowledge the special help of Mr. R. F. Van Wye and A. Matheson of the Schenectady Operations Office, U. S. Atomic Energy Commission, in setting up and carrying out the joint Atomic Energy of Canada Limited-Knolls Atomic Power Laboratory Program as part of the technical cooperation program of the U. S. Atomic Energy Commission. The authors also wish to thank Mr. R. D. Werner of Atomic Energy of Canada Limited for his help with the chemical separations and counting, and the *MTR* and *NRX* reactor operating staffs for irradiations.

[14] B. B. Kinsey, Can. J. Research **A26**, 404 (1948).

[15] M. E. Rose in *Beta- and Gamma-Ray Spectroscopy* edited by K. Siegbahn (North Holland Publishing Company, Amsterdam, 1955).