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Ranges in Air and Mass Identification of Plutonium Fission Fragments

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Determinations were made of the mean and extrapolated ranges in air of plutonium fission fragments for twenty individual masses between 83 and 157. Collimated fission fragments passing through air at 120 or 140 mm pressure were deposited, after being stopped by the air, on a series of fourteen thin lacquer films. These were analyzed radio-chemically for individual fission products. The corrected activities were plotted against distance traversed by the fragments, yielding differential range curves whose widths at half-maximum were 11.7±1.3 percent, independent of fragment mass. The activities found beyond each distance were plotted against distance giving integral range curves. Mean and extrapolated ranges were derived from these. In the light group the extrapolated ranges decrease from 2.90 cm (15°C and 760 mm) for mass 83, to 2.25 cm for mass 117; in the heavy group they decrease from 2.25 cm for mass 127, to 1.95 cm for mass 157. From the range-mass curve drawn for well-known masses, definite assignments of 92, 93, and 132 were given to 3.5-hr. Y, 10-hr. Y, and 77-hr. Te, respectively. Highly probable assignments of 94 and 134 were given to 20-min. Y and 54-min. I, respectively.

I. INTRODUCTION

SINCE the discovery of the process of nuclear fission, several measurements have been made of the range of the fission recoil fragments. Corson and Thornton[1] observed in a cloud chamber that the maximum range was about 3 cm standard air. Booth, Dunning, and Slack[2] used a shallow ionization chamber to measure the number of fission fragments as a function of air pressure in the system. Two groups appeared with maximum ranges of 1.5 cm and 2.2 cm. E. McMillan[3] irradiated a plate of U_3O_8 covered with a series of aluminum foils. By plotting the gross recoil radioactivity in each foil against thickness of aluminum, a maximum range of 2.2 cm air equivalent was obtained.

Segrè and Wiegand,[4] using two methods, measured the relative stopping power for fission fragments of collodion, aluminum, copper, silver, and gold. In one method, the gross radioactivity that penetrated various thicknesses of foil was collected on a celluloid plate and counted. The maximum range in aluminum was found to be 3.7 mg/cm² or about 2.5 cm normal air equivalent. In the other method, fission pulses which penetrated various thicknesses of foil were counted in a shallow ionization chamber. It was found that the relative stopping powers for fission fragments of the above materials are roughly the same as for 4.6-Mev alpha-particles.

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[1] D. R. Corson and R. L. Thornton, Phys. Rev. **55**, 509 (1939).

[2] E. T. Booth, J. R. Dunning, and F. Slack, Phys. Rev. **55**, 982 (1939).

[3] E. McMillan, Phys. Rev. **55**, 510 (1939).

[4] E. Segrè and C. Wiegand, Phys. Rev. **70**, 808 (1946).

Bøggild, Brostrøm, and Lauritsen,[5] and Bøggild, Arrøe, and Sigurgeirsson[6] have made extensive cloud-chamber studies of the stopping of fission fragments from uranium in xenon, argon, helium, hydrogen, and deuterium. The heavy fragment tracks usually show some curvature caused by many collisions with gas atoms that are not close enough to give observable recoil tracks. Numerous collisions, however, do give rise to definite branches, and a tuft of several short branches frequently appears at the end of the range of a fission fragment. By a study of the distribution of branches along the tracks and also by direct measurements of the angles and lengths of individual large branches, velocity-range curves were constructed for the light and heavy groups of fission fragments. These curves show that first the fragments are slowed down mostly by electronic interactions but that during the last portion of the path most of the stopping results from nuclear collisions. The greatest variation in range reduced to normal air was from a mean total range for both fragments of 5.2 cm in helium to 3.9 cm in hydrogen.

Experiments to determine the range of fission fragments of a single mass were performed by Freedman, Metcalf, and Sugarman.[7] Their apparatus consisted of a small ground quartz joint containing a thin enriched uranium source at one end, and a Cellophane disk at the other end for collecting the fission fragments. An aluminum foil, 1.4 mg/cm^2 thick, was placed just in front of the Cellophane to prevent diffusion of stopped fission fragments to the Cellophane. The chamber was given a series of irradiations for various pressures of air. After each bombardment the Cellophane collecting disk was analyzed radio-chemically for 85-min. Ba^{139} and 9.7-hr. Sr^{91}. Then by plotting activity against air pressure, absorption curves were obtained for fission recoils of the given mass numbers. The maximum ranges were 2.58 cm and 2.05 cm of normal air for masses 91 and 139, respectively. Since the absorption curves that were obtained were not horizontal, as expected, over the first portion of the range, the experiments were repeated by Finkle, Hoagland, Katcoff, and Sugarman.[8] A very thin zapon film ($\sim$15 μg/cm^2) was substituted for the aluminum foil and measurements were made for plutonium as well as for U^{235}. This time the absorption curves were nearly horizontal over the initial portion and resembled corresponding curves for alpha-particles. The maximum ranges were 2.58 cm and 1.85 cm normal air for masses 91 and 139, respectively, from U^{235}; 2.69 cm and 1.95 cm, respectively, for these masses from plutonium.

In continuing a series of experiments initiated by F. Joliot,[9] Suzor[10] irradiated with thermal neutrons a stack of aluminum foils placed over a thin layer of uranium oxide. Half of the fission recoils of a given range leaving any point in the source would thus be deposited in the aluminum on a hemisphere whose radius is equal to the range. It follows from geometrical considerations that the activity of a fission product deposited in any foil is proportional to the thickness of that foil up to a distance from the uranium equal to the range. Three isotopes were separated radiochemically from each of the foils and the activity per unit thickness plotted against distance from the uranium. The curves were horizontal over the initial portion and then dropped gradually to zero. The average ranges for 17-hr. Zr^{97}, 67-hr. Mo^{99}, and 77-hr. Te^{132} were found to be 3.78, 3.78, and 3.11 mg/cm^2, respectively. The corresponding maximum ranges were 4.50, 4.40, and 3.85 mg/cm^2.

A similar series of experiments was performed by Finkle, Hoagland, Katcoff, and Sugarman.[11] In this case eight fission products from uranium were investigated and the total activity of each isotope which passed through each foil was plotted against the thickness of aluminum. Straight lines were obtained over most of the range, and these were extrapolated to zero activity to give values of 3.74, 3.64, 3.57, 3.34, 3.16, 2.75, 2.69, and 2.54 mg/cm^2 for masses 89,

[5] J. K. Bøggild, K. J. Brostrøm, and T. Lauritsen, Kgl. Danske Vid. Sels. Math-Fys. Medd. (Math-Phys. Comm., Acad. Sci. Copenhagen) **18**, 4 (1940).

[6] J. K. Bøggild, O. H. Arrøe, and T. Sigurgeirsson, Phys. Rev. **71**, 281 (1947).

[7] M. S. Freedman, R. P. Metcalf, and N. Sugarman, Manhattan Project Report CC-1559 (April 8, 1944); Plutonium Project Record IXB, 6.6.1 (1946).

[8] B. J. Finkle, E. Hoagland, S. Katcoff, and N. Sugarman, Manhattan Project Report CK-1806 (June 30, 1944); Plutonium Project Record IXB, 6.6.2 (1946).

[9] F. Joliot, Comptes Rendus **218**, 488 (1944).

[10] F. Suzor, Comptes Rendus **224**, 1155 (1947); J. de phys. et rad. **8**, 39 (1947).

[11] B. J. Finkle, E. Hoagland, S. Katcoff, and N. Sugarman, Manhattan Project Report CC-2076 (August 25, 1944); Plutonium Project Record IXB, 6.6.3 (1946).

95, 103, 129, 131, 140, 141, and 144, respectively. These results are not accurate because it was discovered later that the method used to determine the thickness of the aluminum foils was unreliable.

N. Sugarman[12] measured the approximate ranges of the delayed neutron-emitting isotopes from uranium fission by means of a slightly modified aluminum foil technique. From fission yield considerations and from a comparison of the ranges of the 4.51-second and 1.52-second isotopes with the ranges of the better-known 55.6-second and 22.0-second isotopes, the former were assigned possible mass numbers of 87 to 90 and 129 to 135, respectively.

During the investigation reported here the ranges in air of plutonium fission fragments were measured for twenty mass numbers between 83 and 157. Both differential and integral range curves (Figs. 2 and 3) were obtained directly from the data for each mass. Then from a range *vs.* mass curve (Fig. 4) it was possible to make definite assignments of mass to three decay chains whose assignments were doubtful previously, and to give strong support for mass assignments to two other chains. Some conclusions were made as to the dependence of range on the mass and energy of the fragments, and data were obtained on the distribution in range of fragments of uniform mass.

II. EXPERIMENTAL METHOD AND RESULTS

The apparatus consisted of a beryllium tube about eight inches long and one inch in diameter. At one end was a thin foil of plutonium ($\sim$0.1 mg/cm^2) perpendicular to the axis of the tube (Fig. 1). Starting at about four inches from this foil there were mounted on beryllium spacer rings at intervals of one-eighth inch a series of about fourteen zapon lacquer films (8 μg/cm^2 thick). The spacer rings were measured accurately with micrometer calipers. The apparatus was usually filled with dry air to a pressure of 120 or 140 mm mercury at a known temperature. It was then irradiated with neutrons in the Los Alamos homogeneous pile for times varying from 30 minutes to 14 hours. The zapon films were so situated that nearly all fission fragments in the mass range being studied (which did not strike the walls of the tube) were stopped in the region of the films. Practically all of the stopping was by the air, the zapon contributing at most only about five percent. It was assumed as a first approximation that the stopped fission fragments would deposit on the nearest zapon film, i.e., fragments stopped within $\frac{1}{16}$ inch on either side of a film belong to that film.

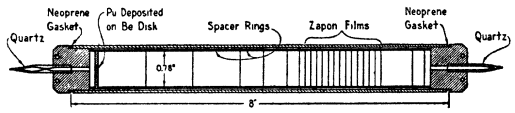

FIG. 1. Sectional view of fission recoil range apparatus. All parts are of Be except where otherwise indicated. The chamber is attached to a vacuum line by means of the quartz tube on the right for filling to the proper pressure. After an irradiation the tip of the left-hand quartz tube is broken so that air leaks through the capillary to bring the pressure to atmospheric without rupturing the zapon films. The spacer rings contain small notches to permit rapid equalization of pressure.

After irradiation, each film, except the first and last, was dissolved in acetone or concentrated HNO_3 and analyzed radio-chemically for a few fission products. The procedures were modified from those reported in Chapter 8, Volume IXB, of the Plutonium Project Record. Some modification was usually needed because two or more elements were isolated from all of each solution. Ordinarily, a solution is divided into aliquots, one for each of the elements analyzed for. In this work the activities were too low for such a procedure. Radio-chemical exchange among the various chemical states of each element was always effected before any precipitations were made. In the case of the yttrium analyses no chemical separations from the rare earths were necessary because of the great difference in fragment range. Element 61 was not isolated from any of the other rare earths except cerium and samarium. La^{140} activity ($T_{\frac{1}{2}}=40$ hr.) was kept down to about two percent of the 47 hr. 61^{149} activity by removing the 12.8-day Ba^{140} parent immediately after 2- to 3-hour irradiations. The 47-hr. Sm^{153} was partially extracted from the element 61 by means of sodium amalgam. The Sm^{153} fission yield is only one-third of the 61^{149} yield; furthermore, a small 47-hr. Sm^{153} contamination would hardly affect the range of mass 149 because the range of the former is shorter than that of the latter. The effect of all other contaminating rare earth

[12] N. Sugarman, J. Chem. Phys. **15**, 544 (1947).

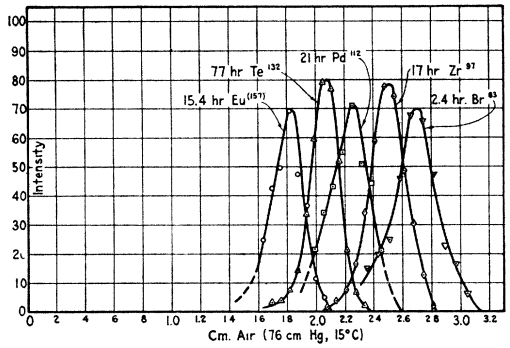

FIG. 2. Five typical differential range curves. The ordinates for each curve are entirely arbitrary.

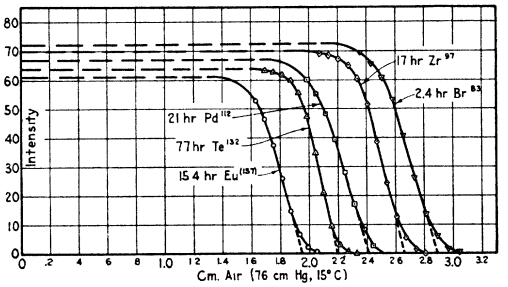

FIG. 3. Five typical integral range curves. The ordinates for each curve are entirely arbitrary.

isotopes was removed by a resolution of the decay curves.

The activity of each sample was followed with a G-M tube for one or two half-lives. In some cases it was necessary to analyze the decay curves into two components. The duration of the bombardment and the time allowed for decay before analysis were chosen so as to emphasize the component under investigation. The activity at a specified time of a given isotope from each film was corrected to 100 percent chemical yield and to the same solid angle, and then plotted against distance of air traversed by the fragments. Figure 2 shows five representative differential range curves (these are really smoothed-out histograms). The vertical scale for each of the curves is entirely arbitrary. The distance, plotted as abscissae, was corrected to air at 15°C and 760 mm mercury pressure (normal temperature and pressure). The zapon thickness was taken into account by relating its stopping power for fission fragments to that of aluminum[13] and then relating the stopping power of aluminum for alpha-particles to that of air.[14] One-half the thickness of the plutonium foil was also included in the distance, taking one mg/cm^2 of the foil equivalent to 0.36 mg/cm^2 of air.[4] By plotting the total activity of a given isotope *stopped* beyond each film against the distance of the film from the source, integral range curves were obtained. Figure 3 shows five such curves corresponding to the differential curves of Fig. 2; again the vertical scale for each mass is entirely arbitrary. The dashed horizontal portions were drawn in by extrapolation. The steepest slopes of all the curves were extended to zero activity to give *extrapolated ranges*.

In Table I the individual runs are separated into blocks enclosed by lines. The upper figures within each block (roman type) give the measured values of extrapolated ranges, while the lower figures (italics) give normalized extrapolated ranges. Nearly all of the runs overlapped each other: in each successive run the range of at least one mass that was measured in an earlier run was redetermined. Relative values of the range as a function of mass number could be obtained accurately only in this way. As a first approximation, each range value was normalized in as direct a manner as possible to the average value (2.504 cm) of the extrapolated range of mass 109. For example, each measured range of mass 157 was corrected by normalizing to the average normalized value of mass 143; each normalized value of mass 143 was obtained from its corresponding measured value by normalizing to the average normalized value of mass 132; the latter was obtained from two runs in which the ranges of both mass 132 and mass 109 were measured simultaneously. In general, there were small differences between the average measured values and the corresponding average normalized values. The algebraic sum of these differences, weighted according to the number of runs for each mass, showed that the average normalized value was 0.17 percent smaller than the corresponding measured value. This resulted from the arbitrary choice of the standard for normalization. Therefore this standard was increased by 0.17 percent (from 2.504 cm to 2.508 cm for mass

[13] E. Segrè and C. Wiegand, see reference 4. Zapon was taken to be the same as collodion since the atomic compositions are nearly identical.

[14] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. 9, 272 (1937).

109) and the measured ranges were then renormalized to this new and final value. In Table I the figures in italics give these final normalized extrapolated ranges and the average value for each mass is plotted in Fig. 4 as a function of mass number.

Table II gives both the average values of the normalized extrapolated ranges and the average values of the normalized mean ranges. The latter were calculated from the measured data in the same manner as were the former. However, the mean ranges could not be measured as precisely as the extrapolated ranges. The last column of Table II gives the average values of the width at half-maximum of the differential range curves for the various mass numbers. The indicated limits of error are merely the standard deviations from the mean of the individual determinations. Parentheses indicate that only two individual measurements were made. The widths are all the same within experimental error and the over-all average is 11.7 ± 1.3 percent of the mean range.

Radio-chemical analyses were not performed directly for some of the isotopes listed in Table I. Instead, their radioactive daughters were first extracted quantitatively from the gross fission product activity, discarded, and then allowed to grow in again from the parents. This method was used for 60-min. Te^{133}, whose daughter is 22-hr. I^{133}, because a direct measurement is made very difficult by other tellurium isotopes and their decay products. A direct isolation of the iodine is also troublesome because 6.7-hr. I^{135} and its 9.2-hr. Xe daughter interefere seriously. Since the tellurium parent of 6.7-hr. I^{135} is very short-lived (<2 min.), an initial iodine separation removes the 6.7-hr. I permanently, but more 22-hr. I^{133} can be formed from its 60-min. Te parent. This regenerated 22-hr. I was then isolated together with the 54-min. $I^{(134)}$ that was formed from its 43-min. $Te^{(134)}$ parent. The decay curves were easily resolved into 54-minute and 22-hour components. By suitably adjusting the times of bombardment, decay, and growth, and by counting the beta-rays from the 22-hr. I through 71 mg/cm² of aluminum absorber, all other components were made negligibly small. The measurements with 21-hr. Pd^{112} were made without the interference of 13.4-hr. Pd^{109} by utilizing the 3.2-hr. Ag daughter of 21-hr. Pd^{112} in a manner similar to that described for the iodine daughters. The range for mass 92 was measured once by

TABLE I. Extrapolated ranges in air of plutonium fission fragments. The values are given in centimeters at 760 mm Hg and 15°C. The individual runs are indicated by the enclosing lines. The upper values in each block are the actually measured ones; the lower italicized values were obtained by normalizing to a value of 2.508 cm for mass 109.

ISOTOPE	Br^{83} 24h	Sr^{91} 9.7h	Y^{92} 3.5h	Y^{93} 10h	$Y^{(94)}$ 20m	Zr^{97} 17h	Mo^{99} 67h	Rh^{105} 36.5h	Pd^{109} 13.4h	Pd^{112} 21 h	In^{117} 1.95h	Sb^{127} 93h	Sb^{129} 4.2h	Te^{132} 77h	Te^{133} 60m	$Te^{(134)}$ 43m	Ba^{140} 12.8d	Ce^{143} 33 h	61^{149} 47 h	$Eu^{(157)}$ 15.4 h
	2.976 *2.967*	2.746				2.736			2.512			2.256 *2.242*	2.250 *2.236*	2.212				2.002	1.958 *1.995*	
	2.860 *2.893*	2.707			2.758			2.580 *2.586*	2.502	2.421 *2.427*		2.269 *2.254*	2.266 *2.251*	2.213				1.947	1.879 *1.969*	
	2.848 *2.815*	2.770		2.754	2.749 *2.692*			2.598 *2.589*	2.517		2.195 *2.232*		2.206	2.192	2.184 *2.190*	2.182 *2.188*		2.011	1.940 *1.968*	
	2.910 *2.921*	2.728		2.738 *2.702*		2.683 *2.648*	2.657 *2.623*	2.621	2.540	2.436 *2.405*	2.204 *2.272*		2.176	2.178	2.157 *2.177*	2.163 *2.183*		2.017 *2.036*		
	2.840 *2.878*	2.702		2.720 *2.701*		2.665 *2.647*	2.661 *2.643*	2.605	2.506		2.266 *2.268*			2.223	2.198 *2.173*	2.193 *2.168*		2.068 *2.045*		
		2.802 *2.743*	2.783 *2.724*	2.755		2.747 *2.689*			2.478		2.188 *2.214*						2.029 *2.078*	1.992		1.904 *1.950*
		2.762 *2.732*	2.740 *2.710*	2.727			2.669 *2.640*		2.485		2.222 *2.243*						2.026 *2.088*	1.979		1.885 *1.943*
				2.794	(2.815) *(2.717)*				2.488		2.138 *(2.155)*						(1.921) *2.083*	(1.881)		(1.802) *1.954*
				2.774	2.761 *2.684*				2.451					2.141 *2.191*			2.112 *2.071*	2.080		
				2.797	2.783 *2.684*				2.481					2.181 *2.205*				2.038		
				2.742 *2.688*					2.558											
				2.720 *2.698*					2.528											
MEASURED AVERAGE	2.887	2.745	2.762	2.752	2.763	2.708	2.662	2.601	2.504	2.429	2.202	2.263	2.225	2.191	2.180	2.179	2.056	2.015	1.926	1.894
NORMALIZED AVERAGE	*2.895*	*2.738*	*2.717*	*2.697*	*2.687*	*2.661*	*2.635*	*2.587*	*2.508*	*2.416*	*2.246*	*2.248*	*2.243*	*2.198*	*2.180*	*2.180*	*2.080*	*2.040*	*1.977*	*1.949*

TABLE II. Extrapolated and mean ranges of plutonium fission fragments in normal air, and the straggling as indicated by the widths at half-height of the differential range curves.

Mass number	Isotope	Normalized extrapolated range (cm)	Normalized mean range (cm)	Average width at half-maximum (percent)
83	2.4-hr. Br	2.895	2.65	13.4±1.5
91	9.7-hr. Sr	2.738	2.55	11.4±0.7
92	3.5-hr. Y	2.717	2.55	10.5±(0.6)
93	10-hr. Y	2.697	2.53	10.1±0.7
(94)	20-min. Y	2.687	2.52	10.5±0.7
97	17-hr. Zr	2.661	2.50	10.7±1.1
99	67-hr. Mo	2.635	2.48	10.8±0.5
105	36.5-hr. Rh	2.587	2.42	11.4±0.6
109	13.4-hr. Pd	2.508	2.35	10.7±0.9
112	21-hr. Pd	2.416	2.24	13.4±(0.2)
117	1.95-hr. In	2.246	2.08	10.1±1.7
127	93-hr. Sb	2.248	2.09	11.9±(1.3)
129	4.2-hr. Sb	2.243	2.09	12.5±0.5
132	77-hr. Te	2.198	2.05	11.5±0.6
133	60-min. Te	2.180	2.04	11.8±0.8
(134)	43-min. Te	2.180	2.04	11.4±1.3
140	12.8-day Ba	2.080	1.92	12.6±1.3
143	33-hr. Ce	2.040	1.89	11.8±0.6
149	47-hr. 61	1.977	1.82	13.1±1.2
(157)	15.4-hr. Eu	1.949	1.79	15.1±1.3

isolating 3.5-hr. Y^{92} directly and once by isolating that 3.5-hr. Y which grew from its 2.7-hr. Sr^{92} parent during an interval after the irradiation. The two values are in close agreement, within 0.5 percent.

When attempts were made to measure the range of the fission fragments of mass 139 by analyzing for 85-min. Ba^{139}, differential range curves were obtained that were considerably distorted. This is attributed to appreciable migration in the apparatus of the 41-sec. Xe^{139} grandparent. The differential curves obtained from 12.8-day Ba^{140} were normal, although its xenon ancestor has a half-life of sixteen seconds. Therefore, it appears that this method of range determination does not apply to those mass numbers which contain in the early part of their decay chains a gaseous isotope with a half-life greater than 20–30 seconds. The differential range curves obtained from 2.4-hr. Br^{83} looked normal, although bromine may exist in volatile chemical forms. However, bromine in such volatile states might react rapidly with the beryllium and zapon, thus preventing the undesirable migration.

By measuring the range of fission fragments of uncertain mass an assignment can be made in many cases by reference to the range-mass curve of Fig. 4. This was done for five decay chains. In three separate experiments the range of fragments that result in 77-hr. Te was found to be greater than the range of those whose mass is known to be 133. This places an upper limit of 132 on the mass of 77-hr. Te, its 5-min. Sb parent, and its 2.4-hr. I daughter. Masses lower than 132 are ruled out because they are assigned to other well-known isotopes. These facts combined with some secondary considerations (e.g., fission yield) establish the mass assignment at 132. The same three experiments showed that fragments resulting in 43-min. Te have, within experimental error, very nearly the same range as those of mass 133. For the same reasons that were presented above, the mass of 43-min. Te and its 54-min. I daughter must be greater than 133. Since mass 135 is definitely assigned to other isotopes, 134 is very probably the correct mass number. Assignment to a greater mass would require serious distortion of the range-mass curve.

The mass numbers of 3.5-hr. Y, 10-hr. Y, and 20-min. Y have all been uncertain. The present range studies (Fig. 4 and Tables I and II) demonstrate that all three masses are in the interval 92–96 inclusive, and that the mass increases in the order just given. This immediately locates 3.5-hr. Y at mass 92 because assignment to 94 would put 10-hr. Y at 95 which has been previously excluded;[15] assignment to 93 is impossible because 3.5-hr. Y has been prepared by a (n, p) reaction,[15] and zirconium does not have a stable isotope there. The 10-hr. Y almost certainly belongs at mass 93. An assignment to mass 94 is highly improbable because 10-hr. Y could not be prepared from zirconium by a (n, p) reaction under conditions which did produce[16] 3.5-hr. Y and 20-min. Y; moveover, 20-min. Y would then have to be assigned to mass 96 which is also unlikely since this would require considerable distortion of the mass *vs.* range curve. Assignment of the 20-min. Y to 95 is impossible because then it could not have been produced by (n, p) from natural zirconium which is not stable there. Thus assignment of 20-min. Y to a mass of 94 is very probably correct.

[15] "Nuclei formed in fission," J. Am. Chem. Soc. **68**, 2411 (1946); Rev. Mod. Phys. **18**, 441 (1946).
[16] W. Seelmann-Eggebert, Naturwiss. **31**, 510 (1943).

The foregoing discussion of mass determination assumes that there are no radical changes in slope of the mass-range curve in the regions considered. In Fig. 4 the crossed points indicate mass numbers whose assignments have been facilitated by these range measurements.

III. DISCUSSION OF RESULTS

The uncertainty in the absolute values of the normalized extrapolated ranges which are listed in Table II is about 1.5 percent. Much of this may be attributed to variations in the thickness of the zapon films. These were not measured individually; instead, several large films of known area were weighed. The mean thickness value obtained in this way was applied to all films which were made in as reproducible a fashion as possible from the same batch of zapon lacquer. If any air leaked into the apparatus before or during an irradiation, the measured ranges would be low. However, a leak was suspected in only one or two of the runs. Small systematic errors may have been introduced by inaccurate reduction of range in zapon and in the plutonium foil to range in air. A few duplicate runs were made with a different plutonium foil, and the results are indistinguishable from those obtained with the main foil. All of the preceding errors can have only a small effect on the ratio of the ranges of different fission fragment masses that are measured in a single run.

The scattering of points in the differential curves may be produced by several factors. Although the zapon films were stretched tightly across the spacer rings when inserted into the tube, they may have buckled occasionally during the irradiations. The films have been observed to expand under certain conditions. The radiochemical analyses were subject to the usual errors. No corrections were applied for the differences in self-absorption of the beta-rays from samples of different weight because the radiation was generally hard.

The graphical method used to obtain the extrapolated fission fragment ranges is subject to some inaccuracy; individual authors were able to arrive independently at virtually the same results. However, there is an uncertainty in the absolute range values of about one percent caused by the arbitrary assumption that the stopped fragments deposit on the nearest zapon film.

Many of the above errors become more important in the measurements of the straggling. The observed spread in range of fragments of a single mass arises from three fundamental sources: (1) the variation in initial charge and kinetic energy of the fragments; (2) the statistical variation in the number of collisions, electronic and nuclear, that are encountered by the fragments; and (3) limitations introduced by the experimental arrangement. The latter were small and consisted of approximately a 1.4 percent total spread in range caused by the thickness of the plutonium source and roughly a 1.5 percent total spread caused by the difference between a perpendicular path from the source to zapon and an extreme diagonal path. These experimental factors were slightly more important for the heavy group than for the light group; therefore this may account for the slightly greater spread that seems to occur in the heavy group. None of the differential curves (Fig. 2) is Gaussian in form; all are steeper on the side distant from the source. Unfortunately, the work performed so far does not reveal whether this form of the range distribution is inherent in the fission process or in the experimental arrangement. If the plutonium fission source were thick over a small fraction of its area or if a small portion of it were covered with foreign matter, then an originally Gaussian distribution would be distorted to the observed shape. Small angle scattering of fission fragments from the walls of the tube might also lead to the same results.

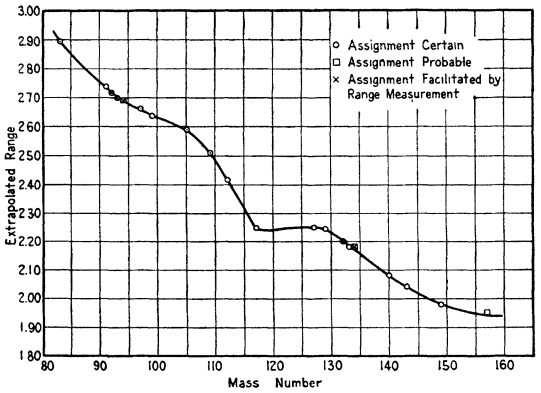

FIG. 4. Extrapolated range (76 cm pressure, 15°C) of plutonium fission fragments in air as a function of mass.

According to a relation derived by Bohr,[17] the range R of a fission fragment should be approximately proportional to the square root of its energy E, and inversely proportional to the sixth root of its mass M. This assumes the charge Z proportional to M. Deutsch and Ramsey[18] have investigated the energy release of plutonium fission fragments in a double ionization chamber. By combining values for the maximum energy derived from their paper with the range values found here, calculations were made of $RM^{1/6}/E^{1/2}$ for both the light and heavy fragments for mass ratios between 1.3 and 1.8. The ionization chamber data are most reliable in this region. The value of $RM^{1/6}/E^{1/2}$ was roughly constant, in approximate agreement with Bohr's calculations; it decreased monotonically from 0.584 to 0.548 for both the light and heavy fragment as the mass ratio was varied from 1.8 to 1.3. Below a mass ratio of 1.2 there is a sharp change in slope of the range *vs.* mass curve for both the light and heavy fragments. This seems to indicate that the kinetic energy release in the fission of plutonium reaches a maximum at a mass ratio of about 1.2 instead of at symmetric fission as calculated by Jentschke.[19] The data of Deutsch and Ramsey on energy release do not extend below a mass ratio of 1.2. However, Jentschke[19] does give data down to a ratio of about 1.1 for U^{235} and U^{238} fission. There is also some indication from his curves that the kinetic energy decreases as symmetric fission is approached. It would be interesting to refine and extend the ionization experiments so that this point could be settled.

Comparison of the ranges reported here with those reported in previous work is difficult. One reason is that the various experiments were not all performed in the same manner and the data are plotted in different ways. Most previous work was on the fission of U^{235} instead of Pu^{239}, and many of the experiments measured the ranges in aluminum instead of in air. The only directly comparable experiment was by Finkle, Hoagland, Katcoff, and Sugarman.[8] The *extrapolated* ranges given there for masses 91 and 139 are 2.66 cm and 1.92 cm (at normal temperature and pressure) instead of 2.74 cm and 2.09 cm as reported here. The cause of this discrepancy is probably that the earlier plutonium foil was far from uniform in thickness, as was pointed out in the original paper. There was only one previous investigation[11] in which the ranges of a sufficient number of fragment masses were measured to obtain a range-mass curve. This was for U^{235} fission fragments in aluminum. Only a limited region was covered, but the shape of the curve was considerably different from the corresponding curve plotted in Fig. 4 for Pu^{239} fission fragments in air. This may be due to the difference between the stopping by aluminum and by air. The older work was based on only a single experiment which should be repeated and extended to include more fragment masses and also plutonium fission.

The apparatus described here (Fig. 1) can be applied to the study of the shorter-lived fission products. Complete or partial separations of certain fission products from certain others can be effected during the bombardment because of the differences in range (see Fig. 2). This makes feasible simplified chemical procedures, so that more rapid isolations could be carried out.

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