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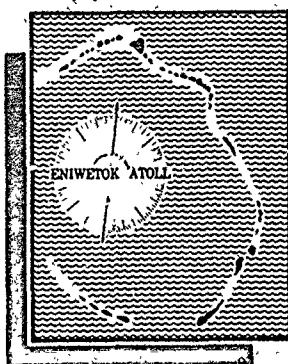
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November 1952

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HEAVY NUCLIDES IN BOMB DEBRIS

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Report to the Scientific Director

HEAVY NUCLIDES IN BOMB DEBRIS

Edited by

Charles I. Browne

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Sponsoring Agency to:

Los Alamos Scientific Laboratory
University of California
Los Alamos, New Mexico
March 1953

March 1953

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ABSTRACT

This report presents the results of the work of personnel of the Argonne National Laboratory, the Los Alamos Scientific Laboratory, and the University of California Radiation Laboratory on the heavy nuclides formed in the detonation of the Mike bomb of Operation Ivy. Many problems in the identification of the atomic number and mass number of the activities found in the debris have not yet been solved and, indeed, may well require another year of work on the debris before firm assignments are obtained. This report offers an interim statement of the conclusions which are available as a result of the primary work on the debris.

It is clear that isotopes of uranium far heavier than those known from other bombardment sources were formed in the Mike detonation. The beta decay of these nuclides led to the formation, in turn, of new heavy isotopes of plutonium, americium, curium, berkelium, and californium. It seems quite probable that isotopes of at least two new elements, those of atomic numbers 99 and 100, were formed. From the yields of these nuclides it is possible to construct a mass-yield curve of considerable interest in diagnostic information.

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ACKNOWLEDGMENTS

The data contained in this report have been obtained from samples of debris obtained in two ways. The first samples were those obtained from the sampling of the bomb cloud itself; the later samples were derived from coral from islands neighboring the detonation point. It is apparent that the results of this report, even though they encompass a great deal of work in rather complex chemistry and interpretation, are subsidiary to the very large effort and skill required to manufacture the Mike weapon itself and to gather the samples of the bomb cloud. For this reason the primary acknowledgment is to the personnel of the Los Alamos Scientific Laboratory (LASL) for the design and construction of the thermonuclear weapon, which gave rise to the extreme neutron flux required to produce the very heavy nuclides with which this report deals. In the initial sampling we wish to acknowledge the skill and application of H. F. Plank in the derivation of the methods upon which the sampling system rests; the courage and ability of the pilots and personnel of Task Unit 132.4.2, USAF, in the actual gathering of the cloud samples; and the effort of the personnel of Task Force 132, upon whom the burden of preparation for the detonation rested. We wish, especially, to acknowledge, in memoriam, our indebtedness to 1st Lt Jimmy Robinson, USAF, who lost his life in the sample-gathering operation.

In the collection of the coral samples, we are indebted to H. S. Allen and Victor R. Benson of LASL for their cooperation in the gathering of the samples.

In addition, the Argonne group wishes to acknowledge helpful discussions with John Huizenga, W. M. Manning, and A. Turkevich and their indebtedness to Dr. D. F. Peppard and his group (George Mason, John Maier, and Richard Wallace), who isolated a heavy-element fraction from the coral. The LASL group wishes to acknowledge their gratitude to R. A. Penneman and personnel of his group for their cooperation and the use of their pulse analyzer. The University of California Radiation Laboratory (UCRL) group wishes to acknowledge the cooperation of K. Street, W. W. T. Crane, R. W. Spence, C. I. Browne, L. Zumwalt, L. B. Werner, N. E. Ballou, and I. J. Russell in providing samples; N. B. Garden, Rosemary Barrett, and Richard Glass for active assistance; and the entire office staff of the UCRL chemistry group for their invaluable aid.

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CHAPTER 1

BACKGROUND

Early indication that debris from the Mike detonation might contain very heavy nuclides in appreciable quantity was obtained from radiochemical analyses and mass spectrographic data, which indicated a very high ratio of Pu^{240} and Pu^{241} to Pu^{239} . Since all these nuclides could be formed only by successive capture in U^{238} , the implication was clear that the successive capture processes might have continued a sufficiently large number of times to form detectable quantities of isotopes of very high mass number. The next concrete evidence of heavy-element production was the discovery of Pu^{244} by mass spectrometer analyses at Argonne National Laboratory (ANL). Further indication of new heavy nuclides was obtained at Los Alamos Scientific Laboratory (LASL) and at ANL when it was found that the plutonium fraction could not be purified of beta activity by many chemical purification steps. Since the plutonium isotopes through mass 243 were known and had such characteristics as to preclude their being responsible for the activity, the presence of the beta activity required the conclusion that isotopes of mass at least 244 were present in quite high abundance. With this information all three participating laboratories embarked upon investigations of the heavy-element fractions. Such investigations required employment of the techniques developed largely at the University of California Radiation Laboratory (UCRL) in connection with the work on the elements from americium to californium. With these techniques and the actinide concept developed by G. T. Seaborg and coworkers, it also seemed certain that the chemical properties of elements immediately beyond californium could be predicted by the actinide concept; in this case elements 99 and above would possess the following properties: (1) They would exist in aqueous solution in their most stable state as tripositive ions with rare-earth-like properties and (2) they would continue the sequence of other tripositive actinide elements with respect to ionic radius so that this property could be applied in identifying these elements by elution from ion-exchange resins.

The objectives of this investigation were not solely concerned with the extension of previous work on the heaviest elements. Another objective of possibly more immediate practical interest was the study of yields of various isotopes as a function of mass number. The variation in such yields would depend on the fast-neutron cross sections of the series of uranium isotopes and on the neutron flux. The yields of the different primary uranium products could be deduced from the amounts of the various products of the beta-decay chains.

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CHAPTER 2

CHEMICAL METHODS

2.1 DISSOLVING OF THE FILTER PAPERS

The initial samples were obtained on intermittent-particle-collector (IPC) filter papers, which were dissolved in different ways by the various laboratories. Most of the activity collected in this manner was dissolved at Los Alamos using the following procedure: The papers were digested with fuming nitric acid until the solution was essentially clear; concentrated perchloric acid was added to this solution, and evaporation was carried on until the exothermic reaction of perchloric acid with the organic matter was completed. Dilute hydrochloric acid was added to this solution, the resulting solution was centrifuged, and the supernatant was poured off. The precipitate was treated with perchloric and hydrofluoric acids, heated until the hydrofluoric acid had been driven off, taken up in dilute hydrochloric acid, centrifuged, and the supernatant was added to that obtained above. Any remaining precipitate was fused with sodium carbonate, then dissolved in dilute hydrochloric acid, centrifuged, and the supernatant was once again added to the stock solution. Any precipitate still remaining was treated again with perchloric and hydrofluoric acids, heated to fuming, and taken up with dilute hydrochloric acid. Further treatment was usually not necessary. The solution thus obtained was diluted further with 2N hydrochloric acid to a standard volume. The final solution, often referred to as Los Alamos "A" solution, contained the activities in essentially 2N hydrochloric acid.

In the ANL procedure usually one-half a filter was cut into 1-in. squares, placed inside a 500-ml polythene bottle, and shaken vigorously with 30-ml solid glass beads and 200-ml acetone according to the method developed by the Berkeley Division of Tracerlab, Inc. The acetone was rapidly decanted through an organdy screen by squeezing the bottle. The active particles which were shaken loose from the filter paper were carried along with the acetone, whereas the filter paper remained behind. This procedure was repeated until the desired removal of activity from the filter paper was accomplished. Monitoring both the pulped filter paper and the acetone filtrate gave an approximate idea of the progress of the removal of the active particles. Usually 60 to 80 per cent of the activity was removed from the filter paper in three cycles.

The acetone, containing the suspended particles, was evaporated to dryness, leaving the particles and a gummy residue, probably the Kronosol with which the IPC paper had been impregnated. Some small quantity of pulped filter paper was also present.

The Kronosol and filter-paper fibers were destroyed, and the bomb debris was brought into solution by cautious treatment with HNO_3 , to which a few drops of HClO_4 had been added. As the reaction subsided and the liquid evaporated with cautious heating, more HNO_3 and HClO_4 were added. This procedure was repeated until a clear colorless solution was obtained.

In the UCRL procedure the organic matter was removed by wet ashing. At the beginning of this work fuming nitric acid was used to remove most of the organic matter. In some cases

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fuming perchloric acid was used to remove residual organic matter. Very extensive fuming with HNO_3 was required to reach the point that it was considered safe to add HClO_4 . In later experiments a large number of filter papers was wet ashed using fuming sulfuric acid and small frequent additions of nitric acid. Organic matter was removed much more rapidly by this method than by any of the other procedures tried; the only disadvantage was that insoluble sulfates of certain elements, such as calcium, were produced. Since the amounts of these elements present in the filters were not large, the difficulties were not serious, and the sulfuric acid could be fumed almost to dryness. Then 6M nitric acid was added, and the mixture was transferred to platinum dishes. Small amounts of HF were added frequently to provide average concentrations of about 0.05M to 0.10M HF during a period of near boiling of the solution. This HF treatment appeared to be necessary to solubilize refractory oxides and other difficultly soluble compounds of the heavy elements. The final fuming operations included treatment with stronger HF to remove as much silica as possible in the conventional manner. Finally the HF was removed by bringing the mixture to strong fumes of H_2SO_4 . When H_2SO_4 had been fumed to near dryness, the dish was cooled, and the contents were transferred with ~6M HCl to a large glass beaker; part of the insoluble residue was dissolved by heating.

2.2 DISSOLVING OF THE CORAL

When initial investigation of the contents of the filter-paper samples had shown that the debris was a rich source of new heavy nuclides, samples of the surface coral from Edna (San Ildefonso), an island neighboring to the island on which the detonation took place, were gathered to determine their value as a source of these nuclides. Some 600 lb of the most active coral was brought to the United States and distributed among the three laboratories.

At ANL the procedure for treating the coral involved dissolving in nitric acid, saturating this solution with aluminum nitrate, and extracting with tributyl phosphate to remove the actinide elements from the aqueous phase. The tributyl phosphate was then stripped with water to return the actinides to a water solution, from which further chemistry could be carried on.

At Los Alamos the coral was dissolved in nitric acid, leaving a not inconsiderable precipitate of organic matter and iron in an undetermined form. The supernatant was decanted, precipitated with ammonium hydroxide, washed, dissolved in hydrochloric acid, and decanted once more. This procedure of precipitating, washing, dissolving, and decanting was repeated until the supernatant was clear and in sample centrifuging gave no indication of a precipitate. The precipitate obtained in the early stages was treated with hydrofluoric and perchloric acids, heated to fuming, and taken up with hydrochloric acid.

At UCRL samples were obtained of fall-out collected in rain gauges located on barges stationed at various points throughout the Atoll at the time of explosion. The same chemical treatment was used with these samples as with the coral; the solid material (primarily calcium and magnesium carbonates but containing also iron, aluminum, phosphate, and organic and silicious material) was dissolved in HCl. The addition of HCl resulted in the evolution of CO_2 , and in dissolving the soil samples there was considerable foaming, presumably due in part to the presence of organic matter. Difficulties with foaming were minimized by spraying the foam with HCl and with surface agents such as aerosol and alcohol. Precipitation of the insoluble hydroxides with NH_4OH or NH_3 gas was generally the first step in the chemical separations, but the subsequent steps were essentially as described where the starting material was filter paper.

Since there was considerable iron present in the material, $\text{Fe}(\text{OH})_3$ was used as a carrier for the elements above uranium. Consequently the calcium and magnesium present were partially separated by making the solutions alkaline (pH 8 to 9) with NH_4OH to precipitate $\text{Fe}(\text{OH})_3$ and $\text{Al}(\text{OH})_3$. The presence of phosphate resulted in the familiar complications in the ammonium hydroxide precipitation. The separation of calcium and magnesium from the ammonia precipitate when phosphate is present is improved by the addition of excess $\text{Fe}(\text{III})$, which

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carries down the phosphate and prevents the precipitation of calcium phosphate. Aluminum was partially dissolved as aluminate by treatment of the ammonia precipitate with KOH and in some cases by precipitation of $\text{Fe}(\text{OH})_3$ from an acid solution by adding KOH.

2.3 URANIUM AND PLUTONIUM PURIFICATION PROCEDURES

The plutonium fraction was investigated at ANL and LASL, and the uranium fraction was investigated at ANL. For mass spectrographic purposes at ANL the chemistry involved evaporating the solution (from filter papers) to dryness and dissolving in a few milliliters of 2N nitric acid. In some cases calcium and magnesium salts were present to such an extent as to interfere with subsequent operations, and auxiliary methods for their removal were employed. Otherwise this solution, containing uranium, transuranium elements, and all the nonvolatile long-lived fission products, was oxidized with a few milligrams of KBrO_3 at 90°C for 1 hr. It was then cooled, saturated with NH_4NO_3 , and extracted four times with preoxidized hexone. Each extract was scrubbed twice with an equal volume of saturated NH_4NO_3 -2N HNO_3 solution and then stripped with water. The water strip contained most of the plutonium and uranium, with possibly a small amount of fission-product zirconium and ruthenium. The transplutonium elements remained in the original HNO_3 solution. The small amount of NH_4NO_3 carried along during the extraction was destroyed by heating with aqua regia.

The plutonium was purified by extraction into a TTA-benzene solution from 0.5N HNO_3 , to which a drop of H_2O_2 had been added to ensure the plutonium being in the IV state. After the plutonium was stripped into 8N HNO_3 , the acid was evaporated, and the organic material was destroyed with HClO_4 . The plutonium was now ready for mass spectrometric analysis.

The uranium was purified by a conventional ether extraction from 2N HNO_3 solution saturated with NH_4NO_3 . On evaporation of the strips from this extraction, destruction with aqua regia of the small quantity of NH_4NO_3 that was carried along, and treatment with HClO_4 , the uranium was ready for mass spectrometric analysis.

No particular effort was made to separate neptunium from either the uranium or plutonium since the samples were too old to contain significant amounts of Np^{239} by the time they were purified.

For purification in the investigation of the activities in the plutonium, the ANL experimenters extracted the plutonium into ether from an oxidized solution, returned the plutonium to the aqueous phase, precipitated lanthanum fluoride, extracted the plutonium into TTA-benzene solutions, returned it to the aqueous phase, and passed it through an anion-exchange column. These operations were repeated until the ratio of alpha to beta activity remained constant within the experimental error.

At Los Alamos the plutonium was purified by initial extraction of the plutonium in the IV state into a cupferron-chloroform solution, destruction of the organic phase, the taking up into nitric acid, and the carrying out of a series of lanthanum fluoride precipitations from oxidized and reduced solutions. In the case of the reduced solutions, in which the plutonium is carried on the lanthanum fluoride, the precipitate was treated in the usual manner with nitric and boric acids; the lanthanum precipitated as the hydroxide with ammonium hydroxide and was dissolved in dilute nitric acid to initiate another cycle. These cycles were repeated until the alpha to beta activity ratio was constant.

2.4 TRANSPLUTONIUM ELEMENT PURIFICATION PROCEDURES

The methods employed in the purification of the transplutonium elements involved, in all cases, the use of ion-exchange resin columns. At ANL the aqueous phase from the ether extraction of uranium and plutonium was heated with aqua regia to destroy the ammonium salts and then run through an anion Dowex A-1 resin column to remove iron carrier, fission products, and the last traces of plutonium. The heavy-element fraction was then adsorbed on a

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cation column of Dowex 50 resin and eluted with concentrated hydrochloric acid. After this column run, the solution was converted to dilute nitric acid, salted with ammonium nitrate, and extracted with tributyl phosphate. The heavy elements were stripped from the tributyl phosphate with 1N nitric acid. A second Dowex 50 resin column run was then made, with hydrochloric acid again as the elutriant. At this point the alpha to beta particle ratio was about 1 at the peak of the elution curve. Separation of the individual elements was accomplished by running them through a Dowex 50 cation column at 87°C using an ammonium citrate-citric acid (0.25M citrate) mixture at a pH of 3.5 as the elutriant. After the first column volume had passed through, each drop was collected, evaporated on a separate plate, and counted for alphas, betas, and spontaneous fissions. All the plates collected before the americium peak were pulse analyzed.

The only deviation in the procedure for the coral and filter-paper samples was that in the case of the coral the eluting citrate solution was at pH 3.4 rather than 3.5.

At Los Alamos the procedure used for the filter-paper samples involved successive precipitations with fluoride, ammonium hydroxide, and sodium hydroxide to remove calcium, magnesium, and aluminum. Transplutonium elements were finally separated by a fluoride precipitation from an oxidized solution and were separated from rare earths by runs through a Dowex 50 four per cent cross-linked cation-exchange column, with 13M HCl as the eluting agent. No citrate separations were attempted. In the coral samples essentially the same chemistry was followed, except that the calcium was finally removed by passing the solution through a large column of Dowex 50 four per cent cross-link, with 13M HCl as eluting agent, and the iron was removed by passing the solution through a Dowex A-1 anion column, with 10M HCl as the eluting agent. The transplutonium elements were again separated by the use of small cation columns.

At UCRL lanthanum fluoride was precipitated to carry the desired elements and to separate a major part of the Fe(III). In view of the necessity of using a large amount of lanthanum to carry the actinides, the procedure was altered in some cases to that of passing the mixture in 8M to 10M HCl through Dowex A-1 anion resin to remove Fe(III) and anionic impurities before the fluoride precipitation step. This procedure was in some cases further varied with respect to the adsorption of plutonium. In some cases plutonium was reduced to the tripositive oxidation state and allowed to accompany Am(III) and the heavier elements. In other cases it was allowed to adsorb on the resin in the tetrapositive state and was removed separately. Whenever Pu(III) accompanied the heavier elements in the first Dowex A-1 column separations, it was separated as Pu(IV) in subsequent Dowex A-1 steps so as to minimize interference with the detection of much heavier elements in the final separation of the individual actinides on a standard hot citrate column. Usually two or three Dowex A-1 column separations with 8M to 10M HCl were employed in order to ensure adequate decontamination and purification from anionic constituents. Precipitations of lanthanum fluoride were generally accomplished in 1M to 3M HCl with 3M to 5M HF. Lanthanum hydroxide carrier was usually dissolved in HCl after conversion of the fluoride to lanthanum hydroxide with 2M to 4M KOH in at least two treatments at elevated temperatures. Then the reprecipitation of lanthanum hydroxide with ammonia and treatments with KOH were repeated to reduce the volume further. In a few cases a useful separation prior to the resin columns was that of leaching the complex mixture of hydroxides at least twice with hot 45 per cent K_2CO_3 . Usually a high proportion of the actinides passed into the carbonate solution, leaving an insoluble carbonate residue which could be dissolved in HCl. In this case the hydroxides were precipitated with ammonia, and the K_2CO_3 leaching process could be repeated on the hydroxides to increase the yield when necessary. The combined K_2CO_3 solutions were then diluted with ~4M KOH to 10 to 20 per cent K_2CO_3 and heated to precipitate $La(OH)_3$, which was washed, dissolved in acid, and reprecipitated with ammonia. In this procedure it is assumed that the heavy actinides precipitate as do $La(OH)_3$ and $Am(OH)_3$.

The $La(OH)_3$ obtained at this point was usually dissolved in HCl, reprecipitated with ammonia, and the washed hydroxides were saturated with HCl gas at room temperature. If NH_4Cl

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was present, it was destroyed with aqua regia, and the solution was saturated finally with HCl gas. The 13M HCl solution was usually transferred to a short (4 to 5 cm length) Dowex 50 four per cent cross-linked column which had been thoroughly washed with 13M HCl. The cross-sectional area of the resin column to be used was determined by the amount of rare-earth carrier judged to be present (maximum ~50 mg lanthanum per square centimeter). The rate of elution was adjusted to less than 1 ml/min/cm². It was found experimentally that three separate column separations, with small column diameters for the last two separations, were necessary in order to achieve maximum decontamination by this method. The separate fractions eluted from the HCl column were examined by taking small aliquots, transferring them to platinum disks, drying them under a heat lamp, and igniting them to a dull-red heat. The fractions containing the elements of interest were combined and subjected to further separations.

For the final separation of the individual elements above plutonium, the combined HCl fractions containing the elements of interest were evaporated to near dryness, using a water bath and an air jet, water was added, and the actinides were adsorbed on Dowex 50 spherical fines. This resin was transferred to a standard hot citrate column. The pH of the citrate eluting agent was adjusted to 3.35 ± 0.05 so as to elute the heaviest fractions more slowly than with pH 3.5 citrate and provide a larger separation between the individual elements.

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CHAPTER 3

INSTRUMENTATION

With the exception of the mass spectrometer at ANL, the instruments employed in the experiments described here have become so conventional that there is no need to describe them in detail. In all cases except that of the plutonium beta activities, the instrument of primary reliance was the pulse analyzer and chamber used to measure the alpha-particle energies. The details of the chamber and analyzer varied among the participating laboratories but not in so gross a fashion as to warrant description of the individual systems.

In the measurement of beta activities and half-lives, the variation among the laboratories was sharper. ANL used a conventional Geiger counter, LASL used a methane flow counter of Los Alamos design operated in the proportional region, and UCRL used a windowless methane flow counter of the Nucleometer type.

In the measurement of the spontaneous-fission events, the UCRL instruments are described as consisting simply of revamped alpha counters, each equipped with a chart recorder to show when each fission was observed. The gain of each amplifier was reduced by a factor of approximately 20 so that the counter was insensitive to events of energy less than about 20 Mev; otherwise the conventional argon-filled flow-type ionization chambers normally used for counting total alpha-particle activity were used. For the Pu^{242} spontaneous-fission determination a very fast fission counter was used. The rise time of the chamber was made very short by the use of argon plus 4 per cent CO_2 , and the pulse amplifier had a wide frequency-response range. The high resolution nature of the device was necessary to prevent alpha pile-ups caused by the large amount of Pu^{238} alpha activity present in the Pu^{242} sample.

The instruments for the scintillation spectrometer work at ANL and LASL were quite similar. Both employed a sodium iodide (Tl activated) crystal as the scintillator, with an RCA 5819 photomultiplier tube at ANL and a DuMont K1187 tube at LASL as the detector. The spectra were taken with a single-channel analyzer at ANL and with a 100-channel analyzer at LASL. The coincidence work done at ANL employed two sodium iodide crystals for the gamma-gamma coincidences and an anthracene crystal in coincidence with a sodium iodide crystal for the beta-gamma coincidences. The beta spectra were determined with an anthracene crystal in a scintillation counting assembly.

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CHAPTER 4

MASS SPECTROMETRIC MEASUREMENTS

All mass spectrometric measurements were made at ANL on filter-paper samples from various sources. Samples of the Mike shot, both close in and distant, as well as samples of the Los Alamos A solution were received from B-29 sampling aircraft. Tables 4.1 and 4.2 present the results on the isotopic composition of the Mike debris. For comparison and interest the isotopic composition of King debris is also presented (see Table 4.3).

It should be emphasized that the uranium isotopic ratios have not been corrected for the uranium present which came from coral. At the present time the contribution from coral uranium is not known. It is at least 25 per cent of bomb uranium and may constitute over half of all the uranium in the samples.

For comparison between samples an isotopic analysis was run on a sample of Los Alamos A solution from the King detonation (Table 4.4). The only chemical operation performed was purification by an anion column. Comparison of these data with those of Table 4.3 indicates a possibility of contamination with natural uranium. An additional check on the mass spectrometric values was obtained by fission to alpha ratio measurements on the Mike plutonium. These measurements led to a value of 24.9 per cent Pu^{240} corrected for Pu^{241} content.

The mass ratio of Am^{243} to Am^{241} was determined on samples from the hot citrate column separations to be 7.94 ± 0.22 . The figure in Table 4.1 for the abundance of mass 243 was derived from this datum and the calculation of the fraction of Pu^{241} which had decayed into Am^{241} at the time of separation.

Mass spectrometric analysis of the curium fraction from the hot citrate columns led to a Cm^{246} to Cm^{245} mass ratio of 0.37 ± 0.03 . The absolute percentage of these nuclides with respect to plutonium was calculated from the absolute disintegration rate of Pu^{246} (see Chap. 5).

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Table 4.1—ANL MASS SPECTROMETER MEASUREMENTS OF ISOTOPIC COMPOSITION OF MIKE (CLOSE IN)*

Mass	Pu abundance, %	Mass	U abundance, %
239	70.3 ± 0.3	238	99.06 ± 0.012
240	25.5 ± 0.3	236	0.166 ± 0.004
241	2.74 ± 0.04	235	0.765 ± 0.010
242	1.34 ± 0.02	234	0.0091 ± 0.0005
243	(0.137)		
244	0.083 ± 0.005		
245	(0.0086)		
246	0.0024–0.0032		

*The values within the parentheses are not from mass spectrometric data on the elements indicated but are calculated from mass spectrometric data on other elements.

Table 4.2—ANL MASS SPECTROMETER MEASUREMENTS OF ISOTOPIC COMPOSITION OF MIKE (DISTANT)

Mass	Pu abundance, %	Mass	U abundance, %
239	70.8 ± 0.5	238	99.1 ± 0.2
240	25.1 ± 0.5	236	0.06 ± 0.02
241	2.81 ± 0.05	235	0.81 ± 0.05
242	1.32 ± 0.02	234	<0.04
244	0.09 ± 0.02		

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Table 4.3—ANL MASS SPECTROMETER MEASUREMENTS OF ISOTOPIC COMPOSITION OF KING (CLOSE IN)*

Mass	Pu abundance, %	Mass	U abundance, %
239	95.02 ± 0.05	238	74.9 ± 0.2
240	4.92 ± 0.05	237	(0.35)
241	0.056 ± 0.006	236	2.32 ± 0.02
242	<0.003	235	22.4 ± 0.2
		234	0.36 ± 0.01

*The value within the parentheses is not from mass spectrometric data on the element indicated but is calculated from mass spectrometric data on other elements.

Table 4.4—ANL MASS SPECTROMETER MEASUREMENTS OF ISOTOPIC COMPOSITION OF KING URANIUM FROM LOS ALAMOS A SOLUTION

Mass	U abundance, %
238	65.6 ± 0.3
236	3.21 ± 0.06
235	30.8 ± 0.3
234	0.49 ± 0.04

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CHAPTER 5

PLUTONIUM ACTIVITIES

5.1 BETA ACTIVITIES

The presence of hard-beta activity in a carefully purified plutonium fraction was observed by personnel of ANL and LASL. The suspicion that the beta activity was due, at least in part, to a plutonium daughter was confirmed in both laboratories by milking experiments. At ANL the activity was milked from plutonium in a TTA-benzene solution by washing with 0.5M HCl and precipitated on lanthanum fluoride. At LASL it was milked from plutonium by oxidizing the plutonium with potassium permanganate, precipitating lanthanum fluoride, quickly reducing the supernatant, precipitating lanthanum fluoride, and observing the growth in the plutonium fraction and activity decay in the lanthanum fluoride precipitate from the oxidized solution. Similar experiments at ANL confirmed the fact that there existed a short-lived activity in equilibrium with a longer lived beta activity in the plutonium fraction.

The half-life of the daughter activity was measured at LASL by milking the daughter from the plutonium parent. This was done by putting the plutonium on a Dowex A-2 anion column from a solution 10M in hydrochloric acid and approximately 0.1M in nitric acid and eluting the daughter from the column with solution of the same composition. Under these conditions, in a typical experiment with approximately 3×10^5 c/m (alpha) on the column, only 6 or 7 c/m of plutonium would be eluted or carried off by passing a milliliter of elutriant through a column 4 mm in diameter and 5 cm long. The daughter activity could be followed into background without observation of an appreciable tailing of the activity. As an average value of several experiments in each of which the activity was followed over some nine half-lives, the LASL chemists report a value of 25.0 ± 0.2 min for the half-life.

At ANL the half-life of the daughter was measured to be 25 min by following a sample of some 6×10^4 c/m through approximately 13 half-lives. No deviation from straight-line decay was observed over that interval.

The decay of the beta activity in the plutonium fraction was followed for some 60 days at ANL and led to a half-life value of 11.2 ± 0.2 days for the parent plutonium. In similar experiments at LASL a half-life of 10.5 ± 0.5 days was obtained; this value is considered less reliable than the ANL datum since the activity was not followed for nearly so long a period.

A further experiment performed at LASL was the repeated milking and 4π counting of the daughter activity from a plutonium "cow" which was held on an anion column over a period of 24 days. In each milking the activity was eluted from the column, and the sample for counting was eluted again 3 hr later, since experimental difficulties made it doubtful that the first elution would be quantitative. The eluted solutions were counted for alpha activity to ensure that the plutonium loss from the column was negligible. This technique led to a half-life for the parent of approximately 10 days but had so few points and was over so short a period that no great accuracy can be claimed.

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In experiments to prove the atomic number of the daughter activity, the ANL chemists concluded that the activity was americium on the basis that it could be milked from plutonium in TTA-benzene by washing with 0.5M HCl; that it could be precipitated on lanthanum fluoride; that it behaved like americium on an anion column; and that it could be oxidized under the conditions for the oxidation of americium and remained in the supernatant solution with Am^{241} on precipitation of lanthanum fluoride. In the last experiment the americium which carried with the lanthanum fluoride through incomplete oxidation showed the same alpha to beta activity ratio as that in the supernatant. The ANL chemists noted that this chemical behavior does not definitely eliminate the possibility that the activity could be due to a beta emitter of atomic number higher than americium but pointed out that systematic considerations make a decay chain involving more than two beta-decay steps unlikely in this region.

The LASL chemists also concluded that the daughter activity was that of an americium isotope. The experiments in which the activity was milked from the plutonium on an anion column and by precipitations of lanthanum fluoride from a solution in which the plutonium was oxidized to the VI state showed that the emitter followed the chemistry of americium or curium. In another set of experiments the daughter activity was added to a solution containing tracer quantities of Am^{241} and Cm^{242} . This solution was oxidized with ammonium persulfate, lanthanum fluoride—precipitated, reduced, and lanthanum fluoride—precipitated once more. Less than 10 per cent of the beta activity was present in the lanthanum fluoride precipitate from the oxidized solution. Subsequent pulse analyses showed that about 5 per cent of the Am^{241} was present in the lanthanum fluoride precipitate from the oxidized solution, and approximately 5 per cent of the curium was present in the precipitate from the reduced solution. On this basis the beta activity could not be curium and must have been either americium or an activity of atomic number 97 or higher. The latter possibility was eliminated on the basis that it was very doubtful that two beta activities would escape undetected in either the equilibrium mixture or the daughter activity.

Aluminum absorption measurements were made on the equilibrium mixture and the daughter activity at LASL and ANL. In experiments at both laboratories the daughter activity exhibited only one beta activity, with an energy of approximately 1.2 Mev. The equilibrium mixture showed two beta activities, one with the 1.2-Mev energy observed in the daughter activity and another which was measured to be 0.2 to 0.4 Mev at Los Alamos and 0.15 Mev at Argonne. In experiments at Argonne the 0.15-Mev beta activity was found in freshly purified plutonium, and it exhibited no growth or decay over a period of 40 min.

The assignment of a mass number to this decay chain was based largely on isotopic-abundance data from the mass spectrometer at Argonne. From the characteristics of known plutonium isotopes, it was clear that the mass number must be greater than 243. The 25-min half-life of the americium activity suggested Am^{244} , which has been previously reported¹ to have a 25-min half-life. However, pulse analyses of milkings over a long decay period enabled LASL personnel to set an upper limit of 10^{-2} , and the ANL chemists, a limit of 10^{-3} , to the ratio of Cm^{244} in the milkings to that expected on the basis of its reported characteristics.² At ANL, in order to estimate the yield of the plutonium beta activity, the β^- -disintegration rate of a sample containing a known weight of plutonium was measured by beta-gamma coincidence techniques. The gamma spectrometer was set to accept only gammas with energies greater than 400 kev, and the beta spectrometer was set to accept only betas with energies greater than 200 kev. Thus each counter was sensitive only to the radiations of the 25-min americium in equilibrium with the 11.2-day plutonium. The bias setting on the beta counter corresponds to the maximum of the curve in Fig. 5.1 in which $N_{\beta\gamma}/N_{\beta}$ is plotted as a function of the integral bias (or β^- energy). The disintegration rate was calculated from the relation

$$N_0 = \frac{N_{\beta} \times N_{\gamma}}{N_{\beta\gamma}}$$

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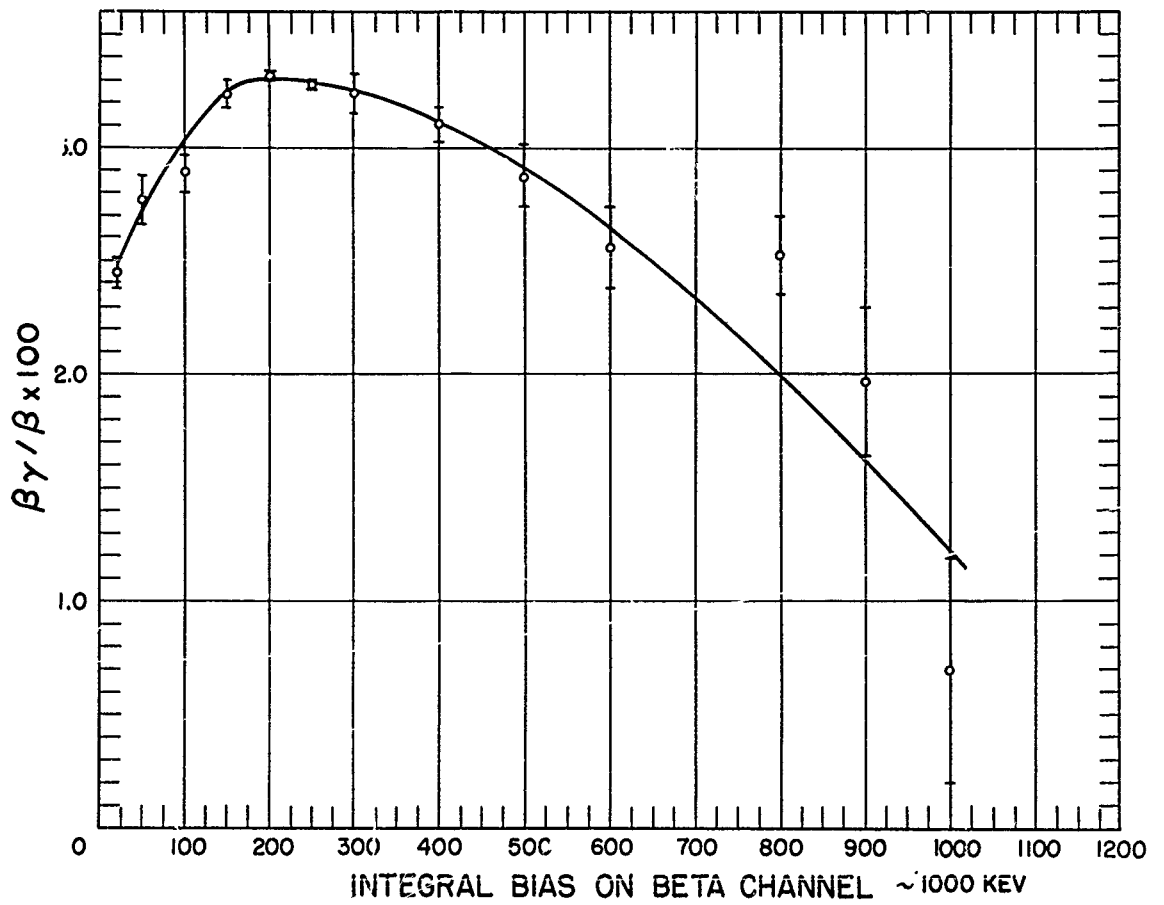


Fig. 5.1—Beta-gamma coincidences per beta in equilibrium Pu²⁴⁶-Am²⁴⁶ sample.

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which is valid for a simple decay scheme and very nearly true for any decay scheme, provided that $N_{\beta\gamma}/N_{\beta}$ is extrapolated to zero energy and that the number of conversion electrons detected relative to the betas is negligible. In an experiment employing this technique, the beta-disintegration rate of the 25-min americium activity in an equilibrium sample giving 1.36×10^4 c/m (alpha) in 2π geometry was measured to be 2494 dis/m at midnight, Jan. 21, 1953, with a statistical uncertainty of ± 1 per cent and an absolute error of possibly 10 to 20 per cent. As of the detonation time on Nov. 1, 1952, the disintegration rate of the plutonium, on the basis of an 11.2-day half-life, would have been 3.98×10^5 dis/m (beta), which corresponds to 3.79×10^{-6} μg . The weight of plutonium represented by 1.36×10^4 c/m (alpha) is 0.118 μg . On this basis ANL chemists derived an isotopic percentage of 0.0032 for the plutonium activity at the time of formation.

The procedure employed at LASL to determine the yield of this chain involved 4π counting of the 25-min activity milked from a known amount of plutonium. On Dec. 17, 1952, 1.84×10^5 dis/m of this activity was found to be in equilibrium with 1.57×10^5 c/m of plutonium. Calculations based on the isotopic composition of the plutonium, as determined by ANL, and the Los Alamos half-life value for the plutonium emitter lead to a value of 0.0024 per cent for the yield of this chain at formation. If the ANL half-life of 11.2 days is used, the yield figure becomes 0.0020 per cent. The absolute error, aside from half-life uncertainties, is estimated at 5 to 10 per cent.

The highest mass number measurable on the ANL spectrometer, 244, was determined to be present in 8.3×10^{-2} per cent. If the convenient assumption of equal capture cross section for successive mass numbers is employed, the yields of the successive mass numbers would be calculated as 245, 1.4×10^{-2} ; 246, 2.0×10^{-3} ; 247, 2.5×10^{-4} ; and 248, 2.8×10^{-5} per cent. From the trend of the known plutonium beta half-lives (Pu^{243} has a half-life of 5 hr), it seems quite likely that any heavy plutonium isotope of 10- to 11-day half-life must have an even mass number. Since it also seems unlikely that the equal cross-section assumption is in error by a factor of 100 over a few mass numbers, the mass number of the chain under investigation is very probably 246. The same conclusion is reached by extrapolation of the mass-yield curve described in Chap. 6.

An insufficient quantity of the 25-min activity was available to allow the conclusive experiment of detection of the alpha particles of the probable Cm^{246} daughter to be performed. On the basis of the knowledge of the characteristics of Cm^{246} (half-life of at least 2000 years), the activity available in milkings was well below the limits of detection of Cm^{246} .

5.2 SCINTILLATION SPECTROMETER STUDIES

Analysis of the gamma radiation from the plutonium activities was undertaken at both ANL and LASL, using the usual techniques of analysis of pulses from a scintillation counter. At ANL the observed peak positions were translated into energy by making corrections for channel width and zero level of the analyzer and for the nonlinear response of sodium iodide. The nonlinearity correction was obtained by measuring the pulse heights of 14 x-ray and gamma-ray lines of known energy, covering the energy range from 22.5 to 1332 kev. With 662 kev as the standard, the correction varied from -21 to +3 per cent over the range since low-energy gamma rays produced a greater pulse height per energy unit than those of high energy. The sample of the equilibrium mixture, which contained about 5 μg of plutonium, consisted of a 1-ml volume of solution in a 2-ml volumetric flask inserted in a hole drilled in a $\frac{1}{2}$ -in. plastic holder. A 245 mg/cm² aluminum absorber was placed over the counter, which had a 175 T aluminum window weighing 95 mg/cm². In several determinations the average values of the energies of the observed gamma rays were 18.5, 43, 103, 175, 220, 795, and 1069 kev. A representative spectrum is shown in Fig. 5.2.

Figure 5.3 presents the gamma-ray spectrum of Am^{246} separated from Pu^{246} at ANL. Gamma rays are identified which have energies of 18.5, 103, 795, and 1069 kev, with the inten-

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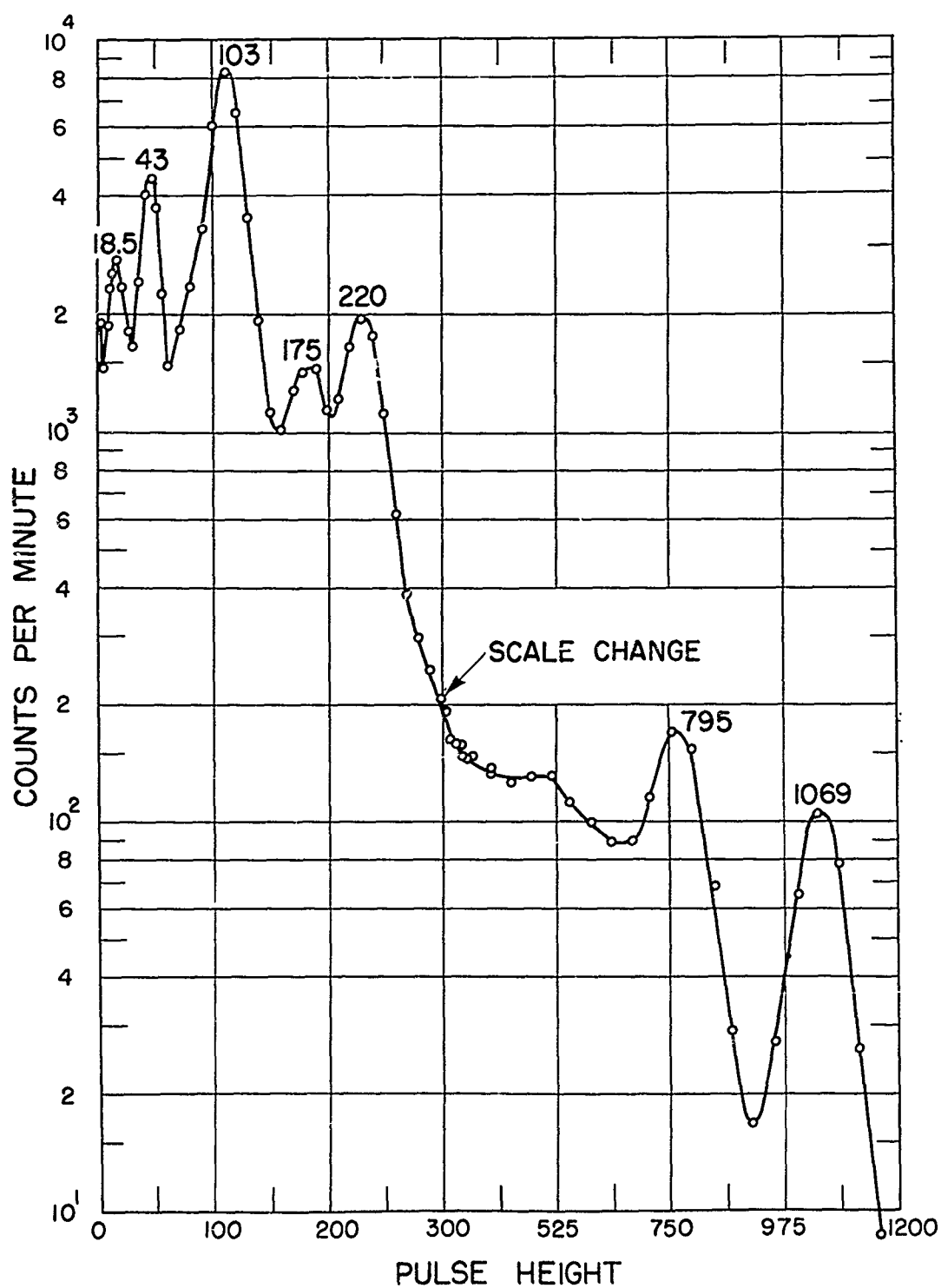


Fig. 5.2—ANL gamma spectrum of Pu^{246} - Am^{246} equilibrium mixture.

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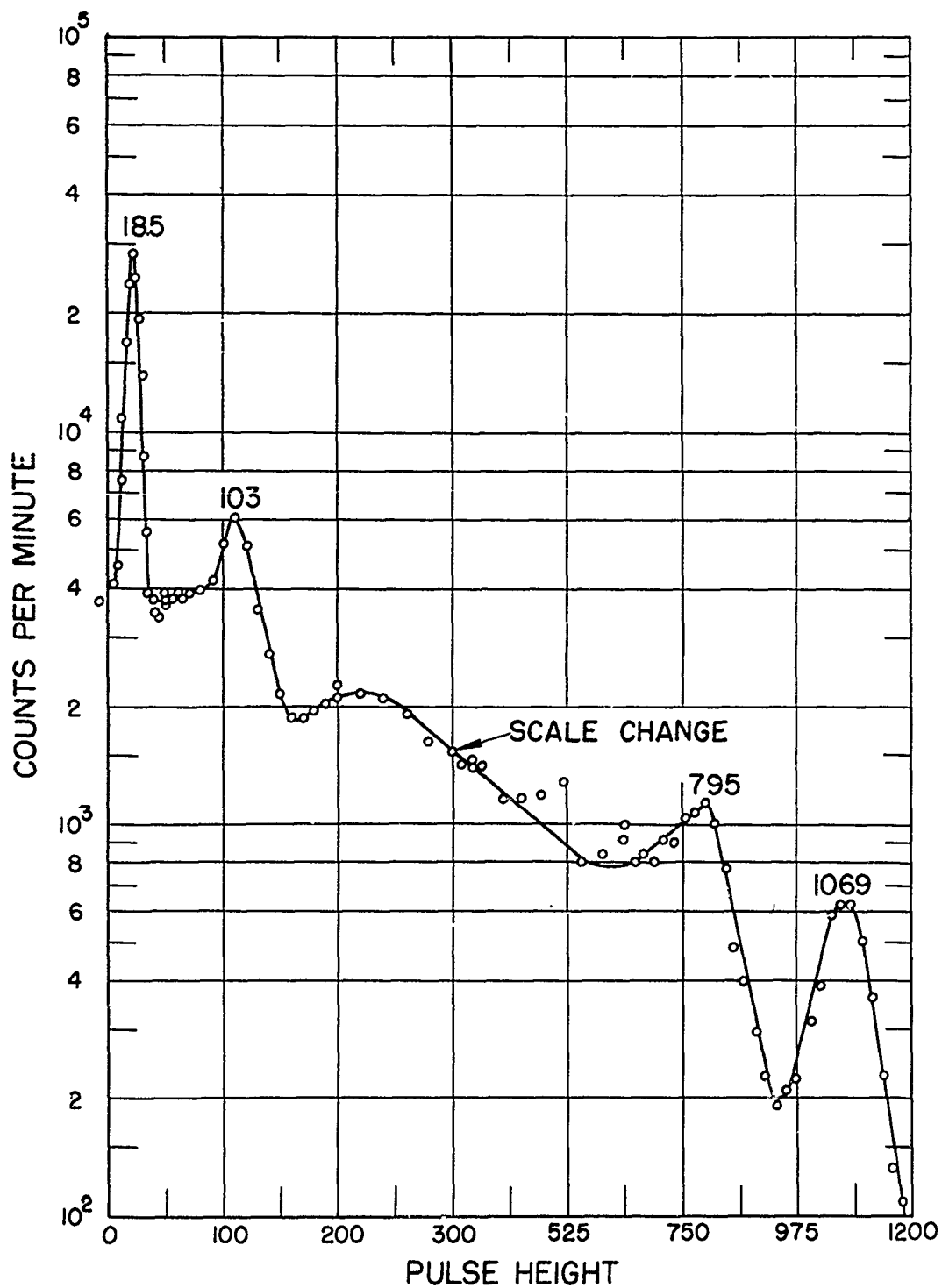


Fig. 5.3—ANL gamma spectrum of Am^{246} .

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sity of the 103 line greatly diminished from that obtained with the equilibrium sample. By difference, gamma rays with energies of 43, 103, 175, and 220 kev may be assigned to the plutonium fraction. In the determination of the americium spectrum, the 245 mg/cm² aluminum absorber was not used, and consequently the absorption of the 18.5-kev line was greatly diminished.

The contribution of the plutonium isotopes with masses less than 244 to any of the peaks observed above was no more than 10 per cent in the case of the 18.5-kev peak and less than 1 per cent for any of the others. It was also observed that the shape of the spectrum did not change appreciably over a period of one month.

At Los Alamos only the lower energy spectrum of the equilibrium mixture was studied closely. A linear relation between channel and energy was obtained in the region below 200 kev by calibrations using the gamma rays and x rays emitted by Lu¹⁷⁷, Am²⁴¹, and Hg²⁰³. The sample used consisted of some 90,000 c/m of plutonium on a platinum plate covered with 0.0005 in. of nylon; the only absorbers present, in addition to several centimeters of air, were 0.005 in. of Al and $\frac{1}{16}$ to $\frac{1}{32}$ in. of MgO. Repeated observations of this sample led to average values for the gamma-ray energies of 224 ± 10 , 111 ± 3 , 72 ± 2 , and 18 ± 0.5 kev. Figure 5.4 presents a representative low-energy spectrum.

At ANL two sodium iodide scintillation spectrometers were used in coincidence to measure the spectrum of gammas in coincidence with the 1069-kev gamma. Spectrometer B was set to select only the photopeak pulses of the 1069-kev gamma, whereas the pulse-height channel of spectrometer A was varied. A peak at approximately 100 kev, followed by a low shoulder at approximately 200 kev, was observed (Fig. 5.5). The relative intensity of the 100-kev radiation may be obtained from the ratio of the integrated coincidence rate over the 100-kev peak divided by the counting rate of the 1069-kev gamma in spectrometer A. The value of this ratio is found to be 0.013, and, since the ratio should equal the efficiency of counter A for a 100-kev photon (about 0.30, if every 1069-kev gamma were in coincidence with a 100-kev photon), it may be concluded that approximately 4 per cent of the 1069-kev gamma rays are accompanied by a 100-kev photon.

The Kurie plot (Fig. 5.6) of the beta spectrum taken at ANL from an equilibrium mixture of plutonium and americium shows a curvature which is characteristic of a complex spectrum. The spectrum was taken with an anthracene scintillation spectrometer, and only those beta particles were counted which were in coincidence with the 1069-kev gamma. This counting was accomplished by placing the sodium iodide spectrometer behind the sample and selecting only those pulses in the photopeak of the 1069 gamma. The coincidence circuit had a resolving time (2 τ) of 0.14 μ sec. The Kurie plot of the coincidence spectrum shows a straight-line portion which extrapolates to a maximum energy of 1213 kev. Correction for window absorption increases this energy to 1222 kev. The upswing at about half the maximum energy is characteristic of the instrument and is due to the scattering of electrons out of the crystal before they have expended all their energy. This upswing makes it impossible to obtain the energy of any possible lower energy components and also masks the spectrum of the plutonium. The large tail on the total β^- spectrum above 1222 kev indicates that betas with an energy greater than 1222 kev are present. However, the low intensity did not permit a determination of their energy. Further evidence for the presence of a higher energy component is shown in Fig. 5.6, in which the beta-gamma coincidence rate per beta is plotted as a function of beta energy. The gamma detector was set to detect all gammas with energies greater than 400 kev, and the beta detector was set to count all betas with energies greater than the abscissa value plotted. For a simple beta spectrum followed by a gamma, the value of $\beta\gamma/\beta$ will remain constant. The decrease of $\beta\gamma/\beta$ will remain constant. The decrease of $\beta\gamma/\beta$ at the higher beta energies indicates that betas are present having a higher energy than those in coincidence with the 795- and 1069-kev gammas. The decrease of $\beta\gamma/\beta$ at lower beta energies is probably due to the low-energy betas of plutonium.

The radiation of energy of about 100 kev and that of 18 kev were interpreted at ANL and

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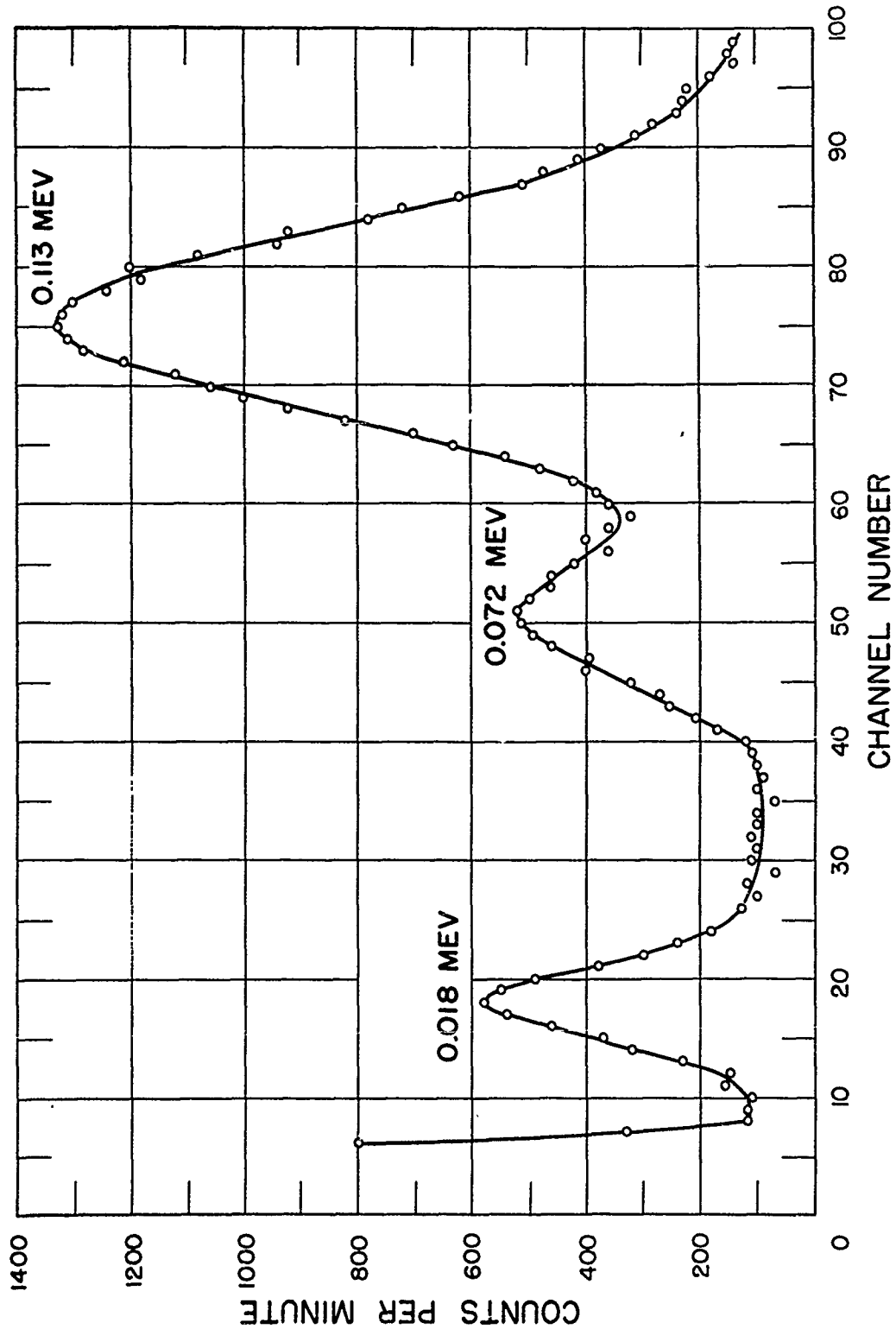


Fig. 5.4—LASL gamma spectrum of $\text{Pu}^{246}\text{-Am}^{246}$ equilibrium mixture.

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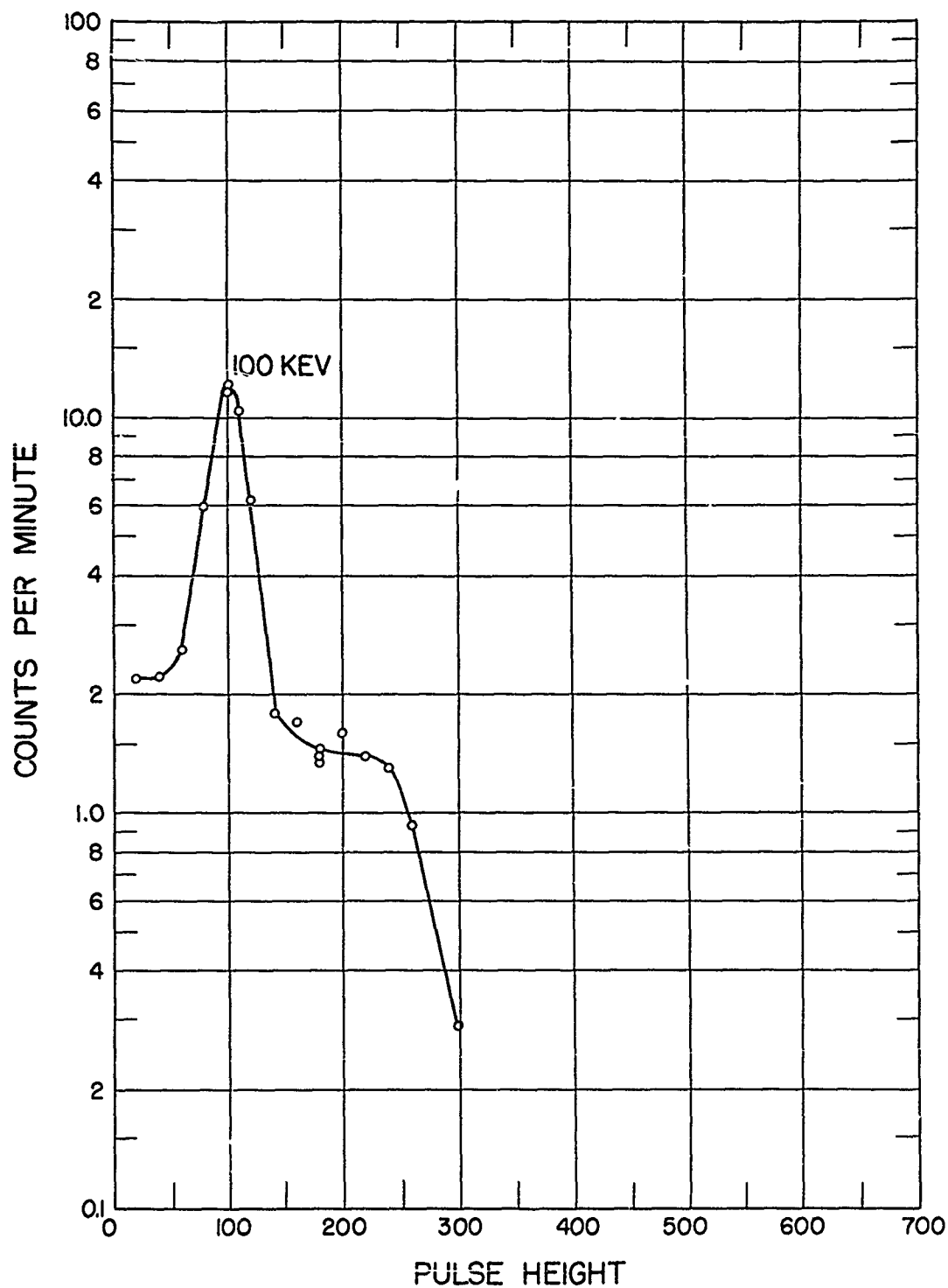


Fig. 5.5—ANL gamma-gamma coincidence spectrum of Pu^{246} - Am^{246} equilibrium mixture.

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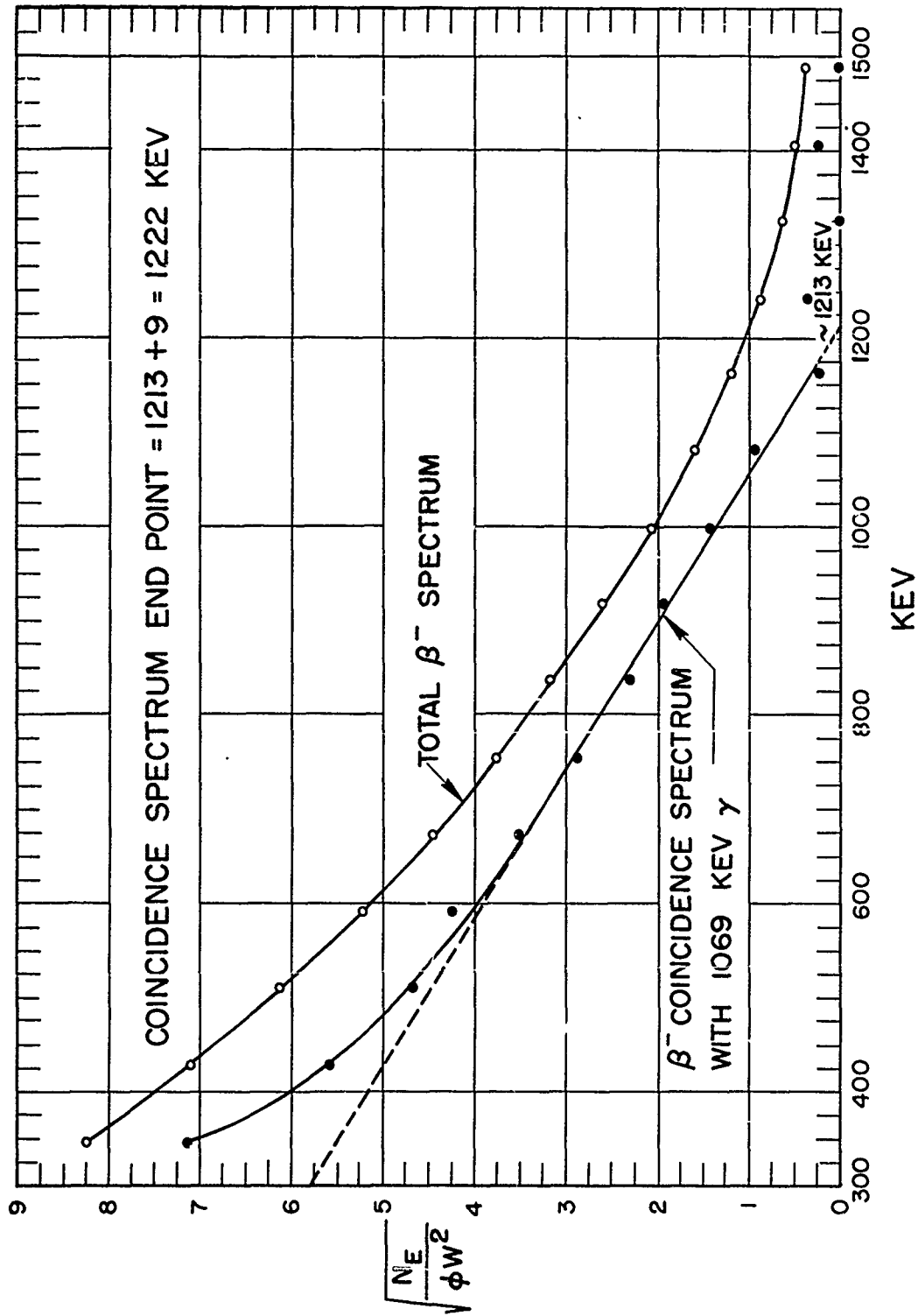


Fig. 5.6—ANL beta spectrum of $\text{Pu}^{246}\text{-Am}^{246}$ equilibrium mixture.

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LASL as probable K and L x rays since these energies roughly correspond to the expected values for the x rays of americium and curium. The 100-kev photon in coincidence with the 1069-kev radiation could be either a curium K x ray or a gamma ray. The total disintegration energy of Am^{246} was interpreted by ANL chemists to be the sum of the energy of the 1222-kev beta ray and that of the coincident 1069-kev gamma ray, or 2.29 Mev, in fair agreement with the 1.94 Mev estimated by Seaborg³ from considerations of closed decay cycles.

5.3 ALPHA ACTIVITIES

The plutonium fraction was examined carefully at UCRL for Pu^{238} (5.48 Mev) alpha activity. It was found that 0.05 per cent or less of the total plutonium alpha activity could be due to this isotope. When corrected for the change in specific alpha activity of the plutonium as determined by mass analysis (approximately 40 per cent of the alpha activity should be due to Pu^{240}), the atom ratio of Pu^{238} to Pu^{239} is calculated to be equal to, or less than, 4×10^{-6} .

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CHAPTER 6

MASS-YIELD CURVE

The mass spectrographic data obtained at ANL were used by the ANL and UCRL groups to construct a mass-yield curve similar to that shown in Fig. 6.1, in which the logarithm of the yield relative to mass 239 is plotted as a function of the mass number of the nuclide. The data through mass 244 suggest strongly that the yield function can be described by two straight, essentially parallel lines on this graph, with one line for the odd masses and the other for the even. Such a description and the extrapolation of these lines were used extensively by both groups in the tentative assignment of mass numbers to the observed activities.

Such a mass-yield function is in variance with the function anticipated in which (on the basis of an assumption of roughly equal capture cross sections for succeeding masses) the yield would fall with rising mass number in a function described by the reciprocal of a factorial increasing by 1 with each successive mass. Thus the yield of mass 240 relative to 239 would be $1/2!$, that of 241 relative to 239 would be $1/3!$, etc. A mass-yield relation of this type is presented for contrast in Fig. 6.1 and is concave toward the origin, with much lower yields at high mass numbers than those predicted by the straight-line extrapolation.

The linear relation, if it is indeed true for high mass numbers, has been explained tentatively as produced by the sum of several factorial curves; this arises from the fact that U^{238} located in different portions of the bomb would be subjected to neutron fluxes differing widely in magnitude.

Insufficient data on mass assignments have been obtained to make a clear distinction as to which type of function is being followed at high mass numbers. As Fig. 6.1 shows, at mass 246, if this assignment can be regarded as firm, the yield predictions of the two functions are so close that no conclusive decision can be made. If cross-sectional variations are admitted, the data through 246 could be explained by the factorial type of function. Until a mass number at least as high as 248 is conclusively assigned, on the basis of other evidence, the mass-yield relation so often mentioned in the following chapters must be regarded only as tentative collateral evidence.

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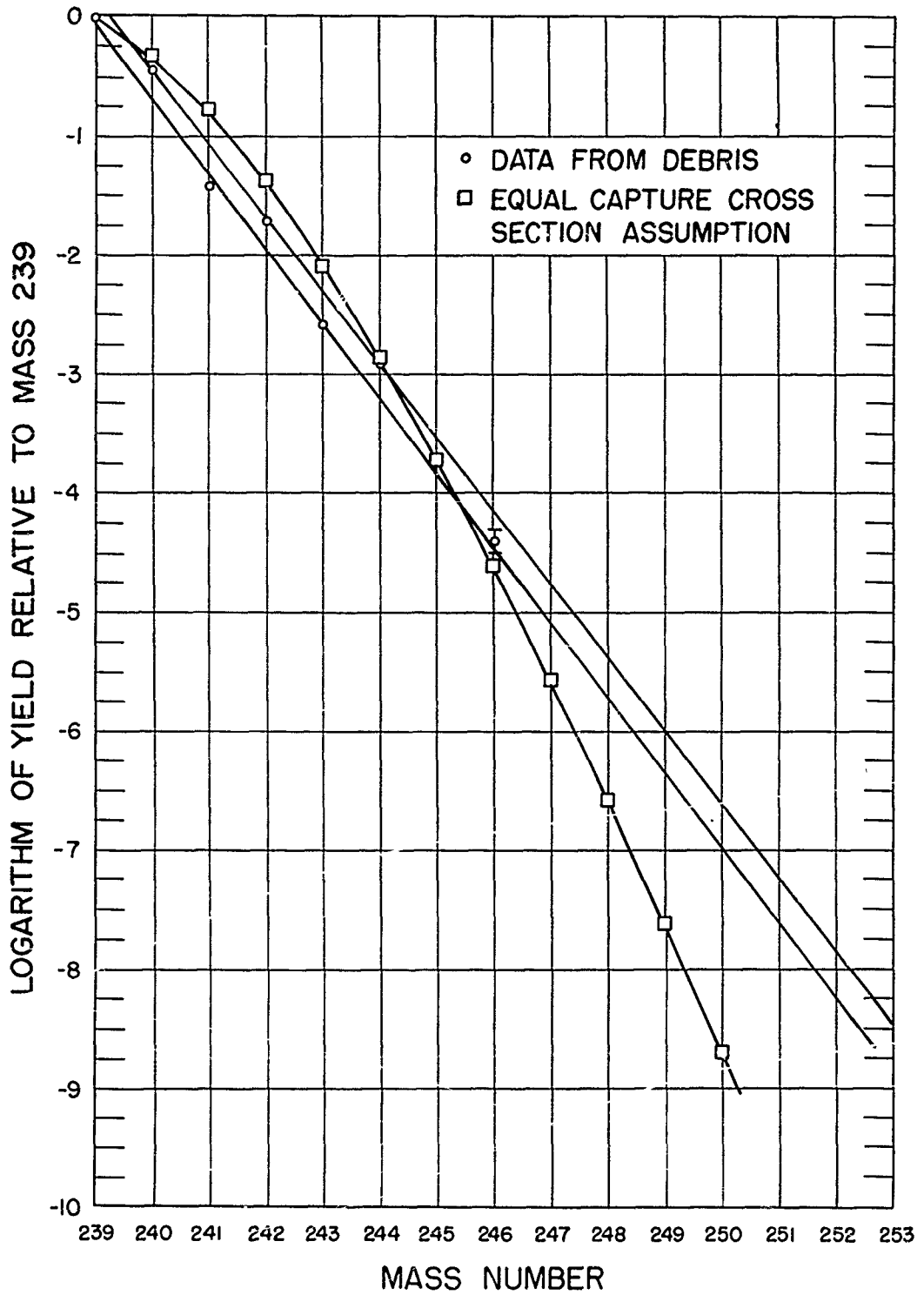


Fig. 6.1—Mass-yield curve.

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CHAPTER 7

AMERICIUM AND CURIUM ACTIVITIES

7.1 AMERICIUM ACTIVITIES

An alpha-particle pulse analysis at ANL determined the ratio of Am^{243} to Am^{241} activities as 0.60 ± 0.06 , which, coupled with the datum on the mass ratios, led to a value of 6380 ± 700 years for the half-life of Am^{243} . This value is in poor agreement with the 8800 ± 500 years obtained at ANL by analysis of the americium fraction from irradiated plutonium. A similar experiment on the Mike americium fraction at UCRL gave results which, upon assumption of an 8600-year half-life for Am^{243} , led to a ratio of 0.1 for masses of U^{242} to U^{241} produced at detonation time. Figures 7.1 and 7.2 present the pulse analyses obtained at ANL and UCRL, respectively.

At ANL a measurement of the ratio of alpha to beta particle activity before the first citrate column run permitted limits of less than three days or greater than 10 years to be set on the Am^{245} half-life. On the basis that the mass spectrometer showed no americium isotopes of mass 245, the longer half-life possibility was considered to be excluded. A similar conclusion was reached at UCRL, where an americium fraction was examined in a windowless methane proportional counter for beta activity. The absence of beta particles within the limits of statistical fluctuation, together with yield estimates based on the mass-yield curve discussed in Chap. 6, permitted limits of less than two days or greater than 3000 years to be set to the beta half-life of Am^{245} .

7.2 CURIUM ACTIVITIES

Pulse analyses of the curium fraction were interpreted at ANL as revealing two peaks, one at 5.4 Mev and one at 6.1 Mev. The 6.1-Mev peak was taken as that of Cm^{242} , produced by neutron capture in the Am^{241} present in the plutonium of the initiator bomb. The 5.4-Mev peak was interpreted as that of Cm^{245} , Cm^{246} , or both. The ANL chemists considered it not unlikely that the alpha-particle energies of Cm^{245} and Cm^{246} are close together and appeared unresolved in the 5.4-Mev peak, which showed little or no fine structure. Calculations of the extreme possibilities showed that, if the activity were due entirely to Cm^{245} , its half-life is about 5000 years, and the Cm^{246} half-life is greater than 10^4 years. On the other hand, if the activity were due entirely to Cm^{246} , its half-life is about 2000 years, and the half-life of Cm^{245} is greater than 5×10^4 years. Figure 7.3 presents the alpha-particle spectrum of the ANL curium fraction.

Alpha-particle pulse analysis experiments at UCRL were considered to exhibit three peaks, which were attributed to Cm^{242} , Cm^{246} , and, probably, Cm^{245} . The 5.48-Mev Cm^{246} peak was shown to be free of Am^{241} contamination by a comparison of alpha pulse analyses of several plates on the leading edge of the curium elution peak. The 5.36-Mev alpha peak was at-

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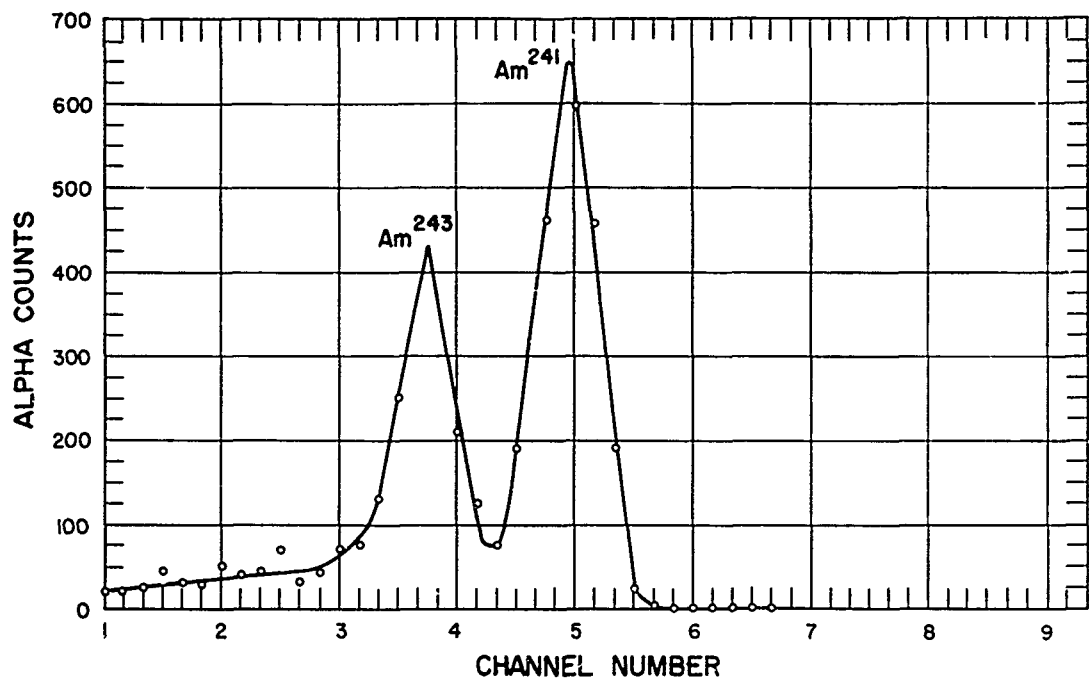


Fig. 7.1—ANL americium alpha spectrum, 10 min.

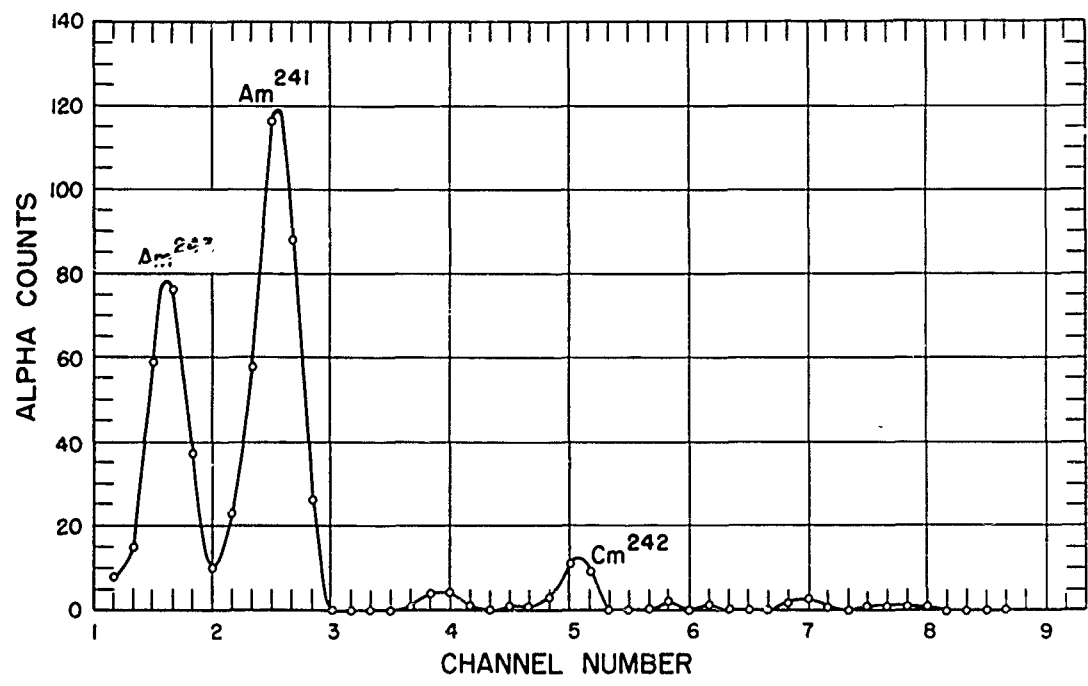


Fig. 7.2—UCRL americium alpha spectrum, 103 min.

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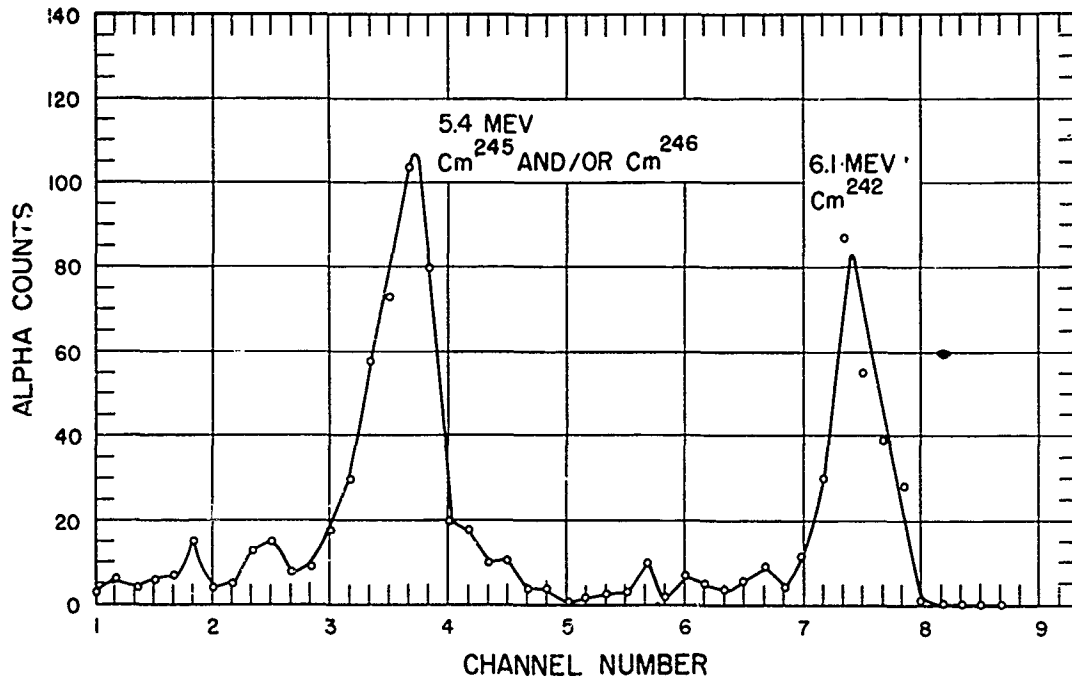


Fig. 7.3—ANL curium alpha spectrum, 236 min.

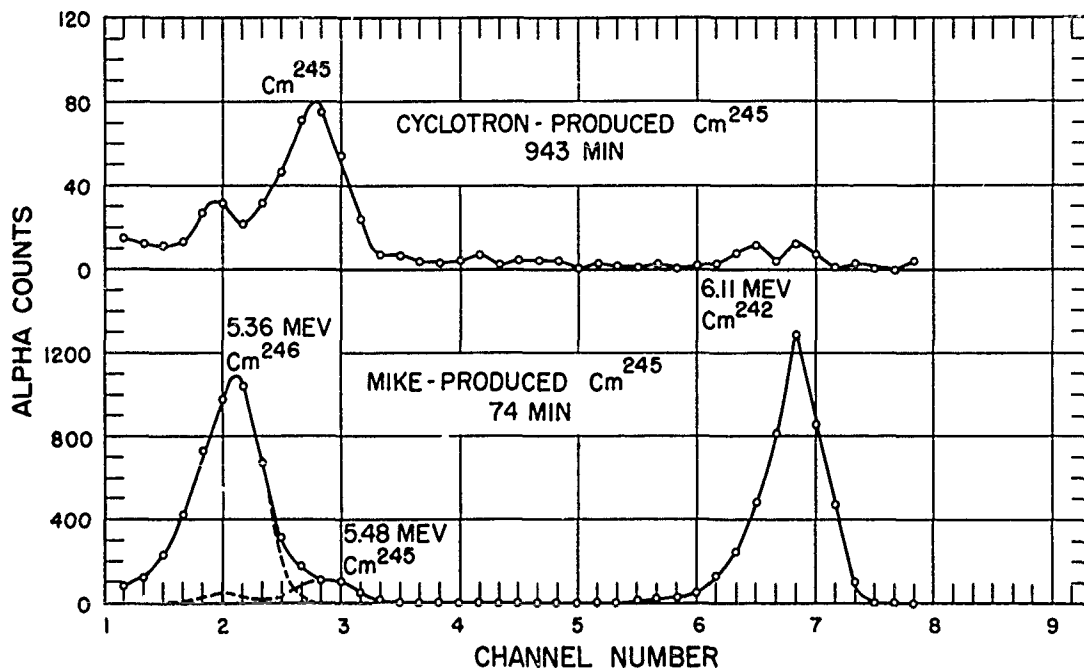


Fig. 7.4—UCRL curium alpha spectrum.

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tributed to Cm^{246} on the basis of its approximate yield and the alpha energy expected of this nuclide. The ratio of the 5.48-Mev alpha activity to that of 5.36 was 0.12; from mass spectroscopic data the atom ratio of Cm^{245} to Cm^{246} at the time of measurement was taken as approximately 2.5. On the basis of these data and an estimated 6000-year half-life for Cm^{246} , the half-life of Cm^{245} was calculated as 1.3×10^5 years. This value is quite at variance with the half-life for this isotope as determined by growth data from its cyclotron-produced Bk^{245} parent.⁴ Figure 7.4 shows an alpha pulse analysis of this Cm^{245} activity for comparison with a Mike sample presumably containing the same isotope. The Bk^{245} & Cm^{245} data indicate a half-life of 3000 to 4000 years for Cm^{245} . The reason for this gross discrepancy is not clear at this time.

The spontaneous fissions in the curium fraction were studied at ANL and UCRL. On the assumption that all the alpha activity and the fissions were due to Cm^{246} , the value reached at ANL for the ratio of fission to alpha-particle activity was 1 to 5000. At UCRL the value for this ratio, considering only the alpha activity attributed to Cm^{246} , was 1 to 4800, from which the spontaneous-fission half-life of Cm^{246} was calculated as 3×10^7 years, if all fission activity were due to Cm^{246} .

On the basis of the atom ratio of Pu^{244} to Pu^{241} and the fact that no Cm^{244} could be detected, the UCRL chemists calculated that the beta half-life of Pu^{244} must be at least 1000 years.

In another experiment at UCRL an isolated curium fraction was allowed to stand for a period of time and again was chemically separated on a citrate column. No significant berkelium or californium activities were detected, which leads to the conclusion that there were no significant quantities of curium beta-particle emitters with half-lives in the rough range of one month to 10^3 years.

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CHAPTER 8

TRANSCURIUM ACTIVITIES

8.1 BERKELIUM ACTIVITIES

No alpha-particle activities were observed which could be definitely attributed to a berkelium isotope. The UCRL group was able to establish the presence of a berkelium beta-particle emitter with a half-life greater than one month by observing the growth of 5.8-Mev alpha-particle activity in a separated berkelium fraction. Since an alpha particle of this energy was observed in the californium fraction, the growth of such an activity in the berkelium was presumed to be due to a californium daughter. The quantity of alpha-particle activity grown in the two-week observation time immediately prior to the writing of this report was small (about 0.2 c/m) but was considered quite definite. The UCRL group tentatively assigned a mass number of 250 or 251 to this nuclide. In previous experiments it was concluded that the half-life of the berkelium parent of the californium 6.1-Mev alpha-particle emitter (see Sec. 8.2) was shorter than a few days.

At ANL the absence of alpha activity in the berkelium fraction led to the conclusion that Bk^{247} was probably not present since considerations of yield and expected half-life indicated that its presence would result in abundant alpha-particle activity. From this it was concluded that Cm^{247} is either beta stable or a very long-lived beta emitter. It was considered that Bk^{249} , if present, might be so long-lived as to be undetectable.

8.2 CALIFORNIUM ACTIVITIES

The alpha-particle activity in the ANL elution from a hot citrate column run on a sample of Los Alamos A solution is presented in Figs. 8.1 and 8.2. The activity in the first two peaks consisted of alpha particles of the energy of Pu^{239} radiation and was interpreted as caused by this nuclide. The activity of the third elution peak was made up of alpha particles having energies of 6.56, 6.3, and 6.1 Mev. Further examination of the samples of this peak revealed a large amount of spontaneous-fission activity which peaked later than the alpha-particle activity in the run of Fig. 8.1. Pulse analyses of the individual drops of this peak are shown in Fig. 8.2, which illustrates the location of the activity peaks of the 6.56-Mev alpha particles, the 6.1-Mev alpha particles, and the spontaneous-fission activity. Figures 8.3 to 8.5 illustrate the relative amounts of activity of the alpha particles on drops 19, 20, and 22, respectively. Pulse analysis of a similar elution peak from a coral sample (elution 2) is presented in Fig. 8.6. Figure 8.7 shows the americium-curtium separation in this run, and Figs. 8.8 to 8.14 show the pulse analyses of drops 67, 68, 69, 70, 73, 75, and 77 of this elution, representing the 6.1-Mev alpha-particle spontaneous-fission peak. The ANL group found that the 6.1-Mev alpha activity was in constant ratio to the spontaneous-fission activity over the elution peak and that

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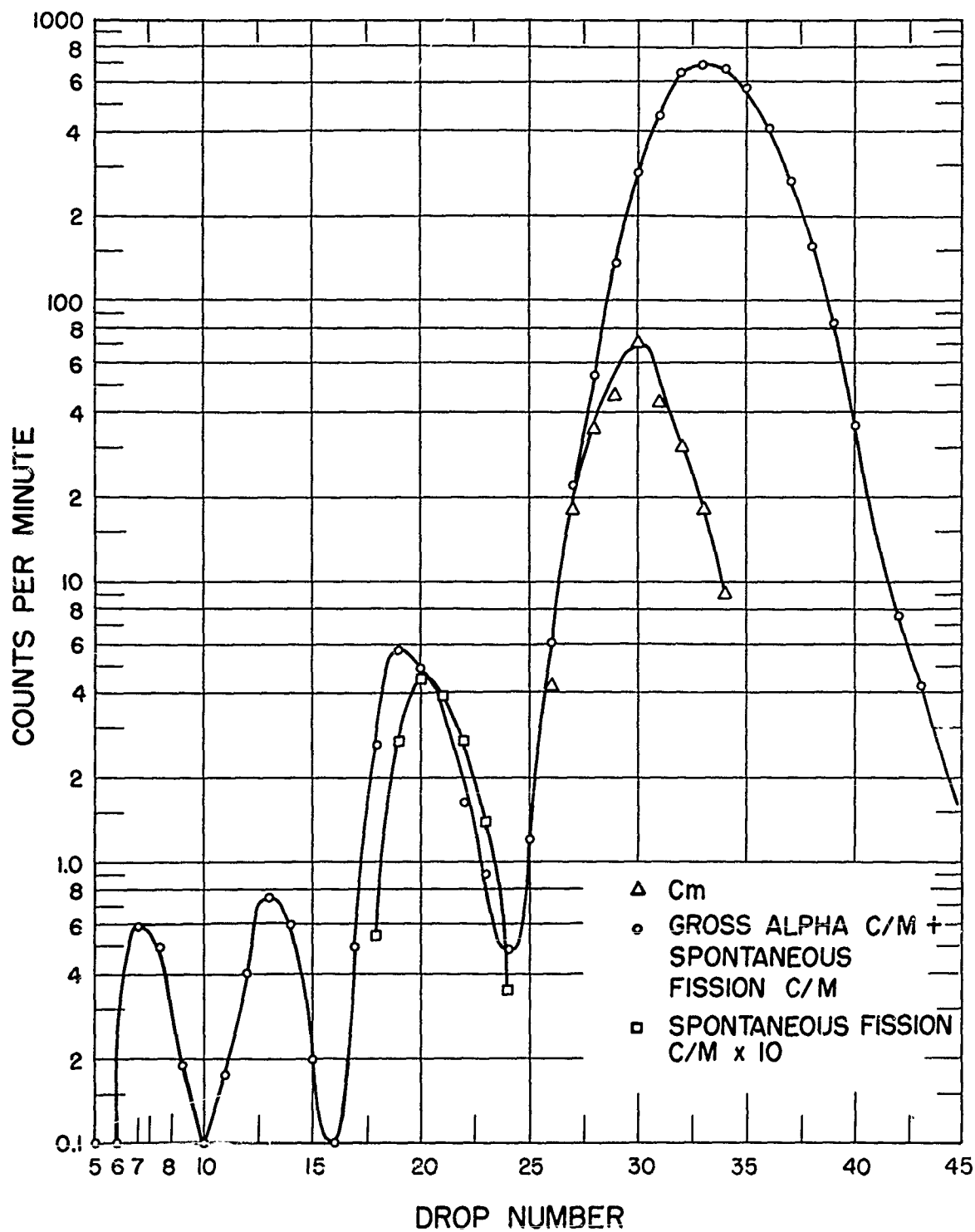


Fig. 8.1—ANL elution curve No. 1; gross alpha activity.

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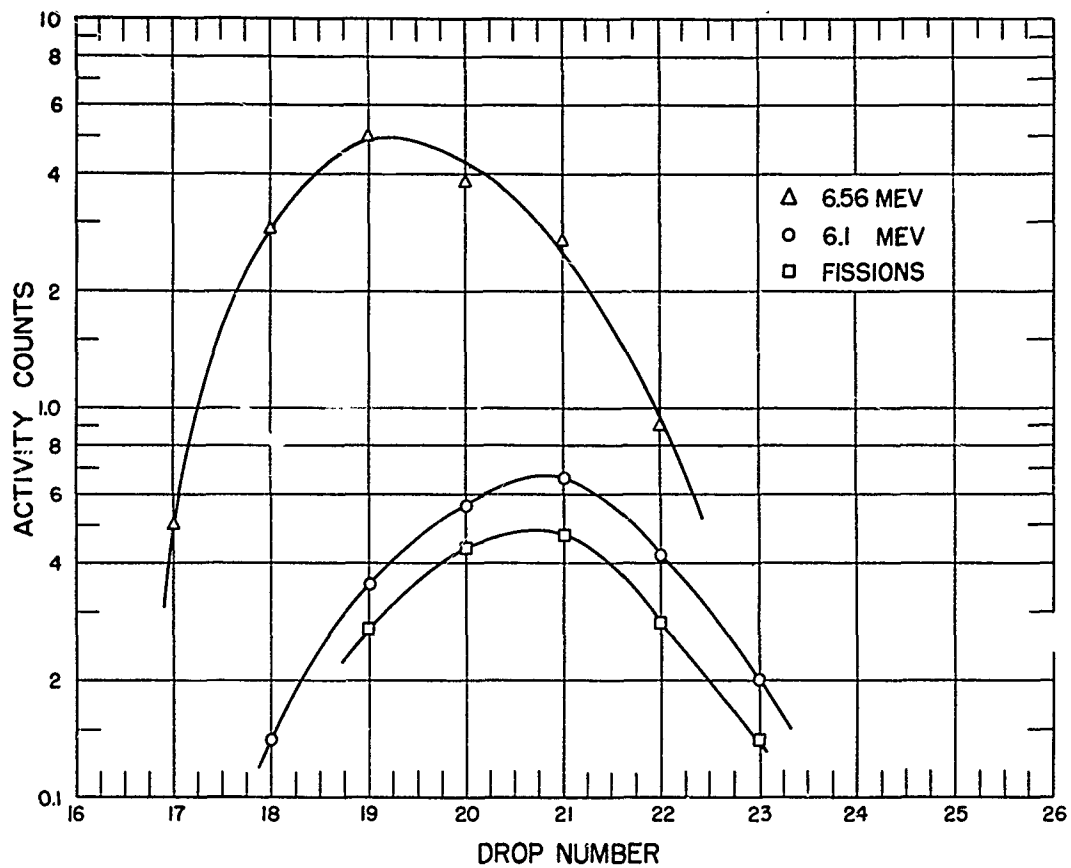


Fig. 8.2—ANL elution curve No. 1; pulse analysis and fissions.

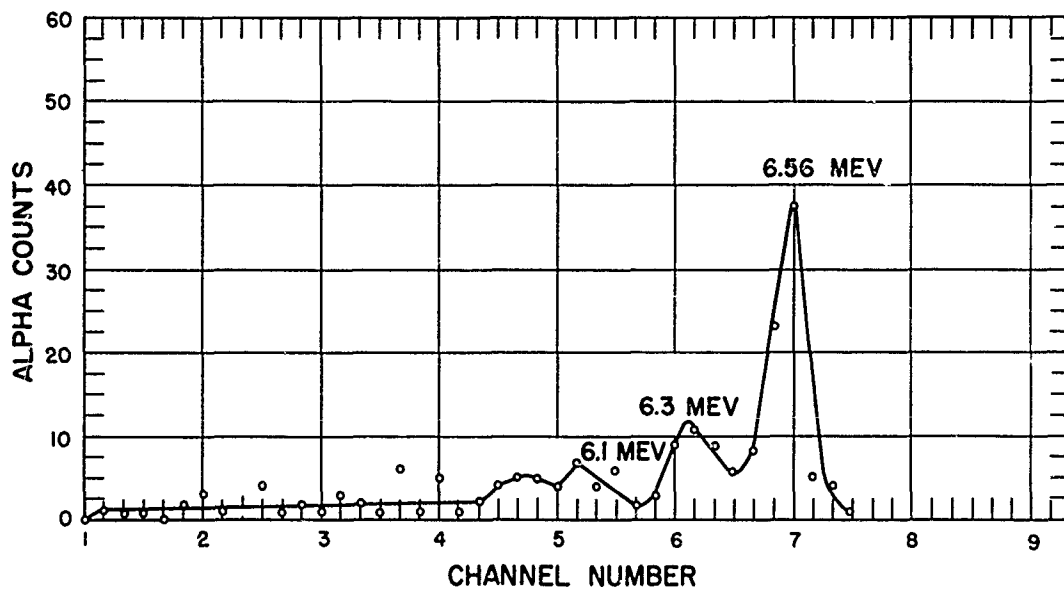


Fig. 8.3—ANL elution curve No. 1; pulse analysis of drop 19, 36 min.

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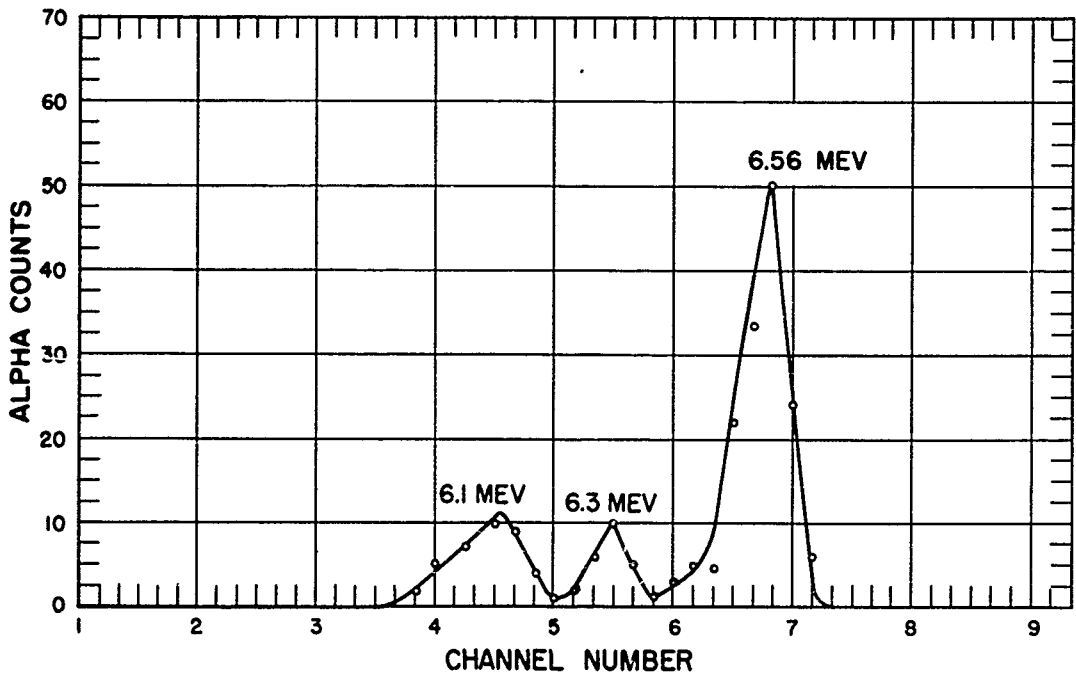


Fig. 8.4—ANL elution curve No. 1; pulse analysis of drop 20, 88 min.

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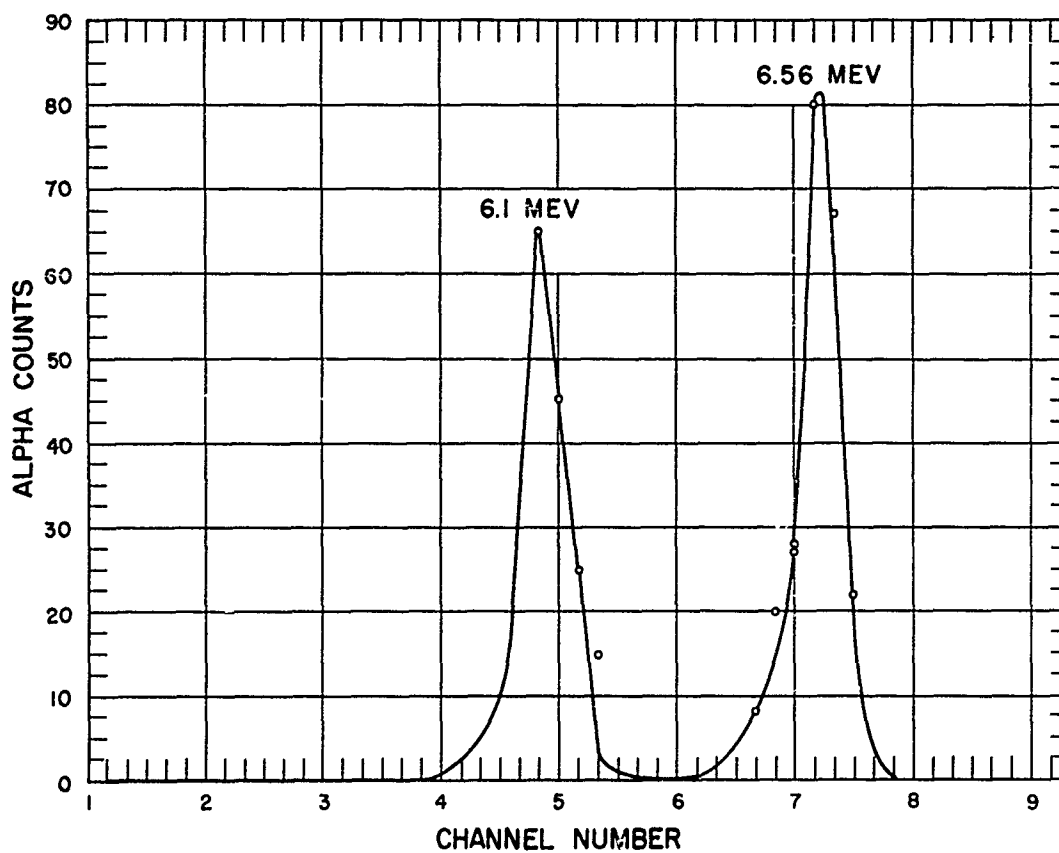


Fig. 8.5—ANL elution curve No. 1; pulse analysis of drop 22, 594 min.

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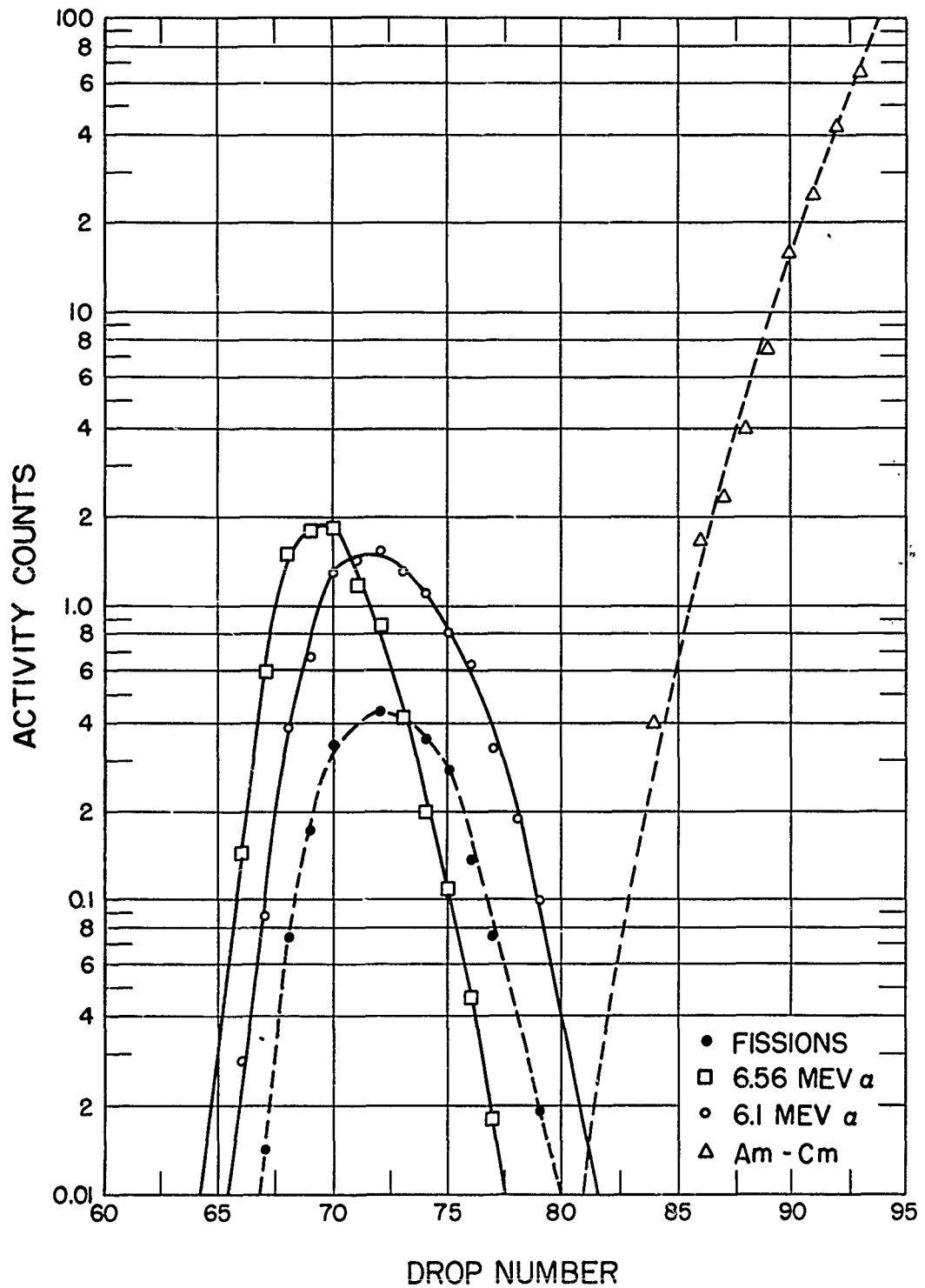


Fig. 8.6—ANL elution curve No. 2; pulse analysis and fissions.

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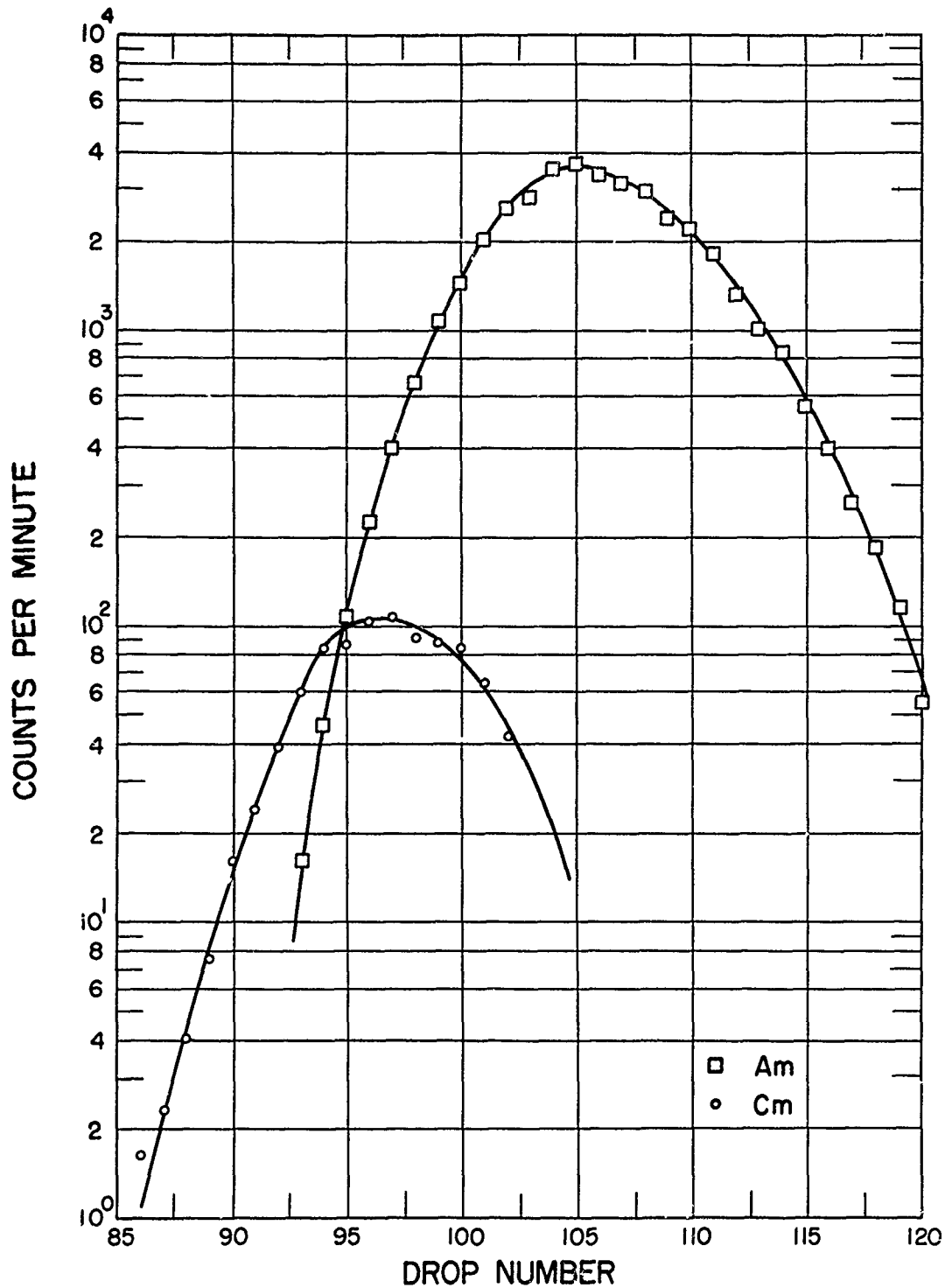


Fig. 8.7—ANL elution curve No. 2; americium-curium separation.

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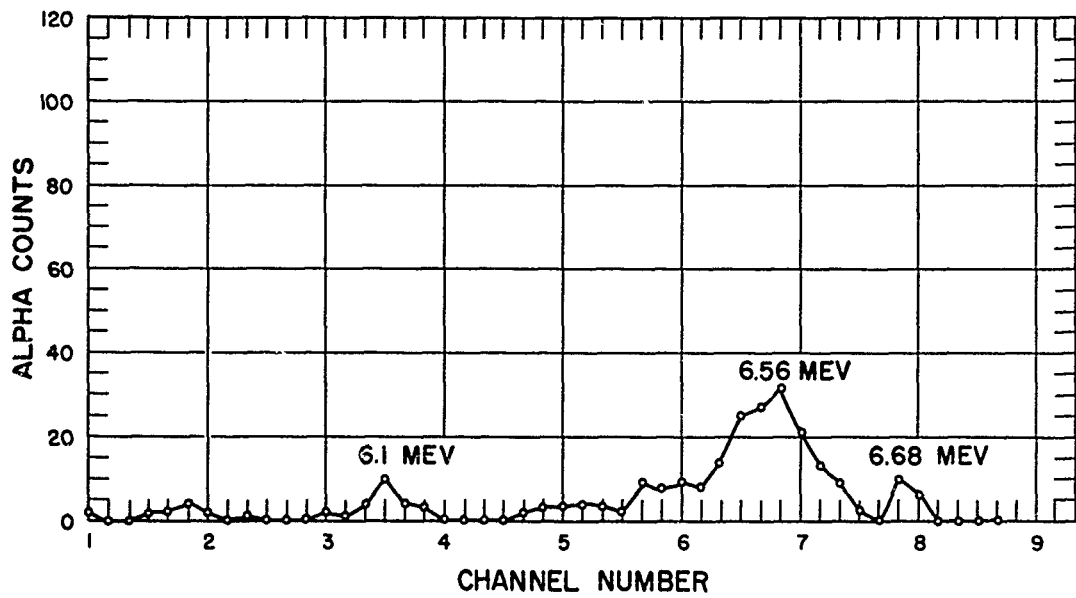


Fig. 8.8—ANL elution curve No. 2; pulse analysis of drop 67, 566 min.

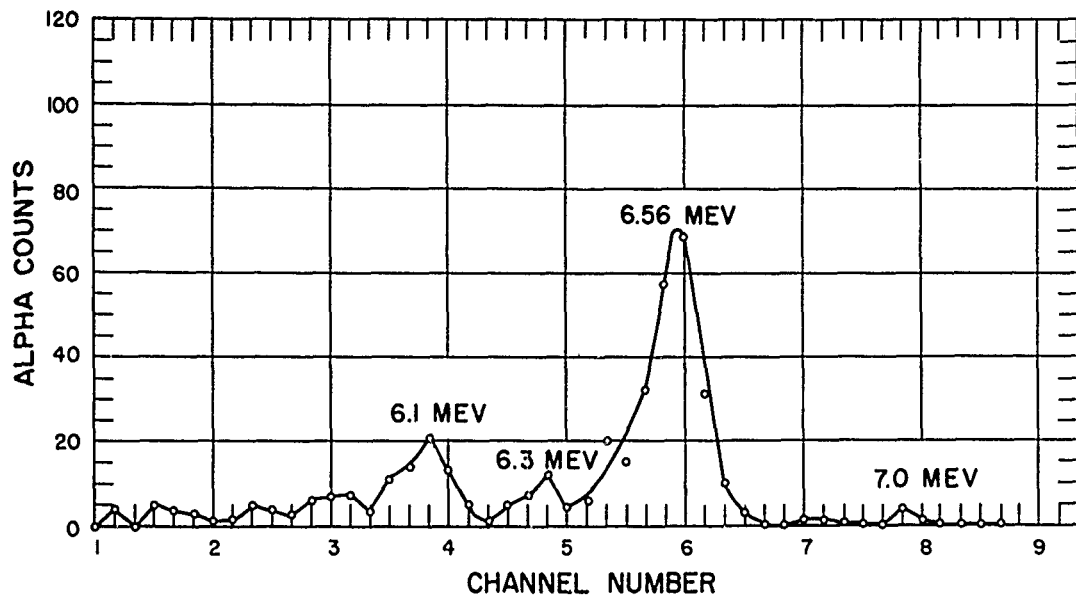


Fig. 8.9—ANL elution curve No. 2; pulse analysis of drop 68, 318 min.

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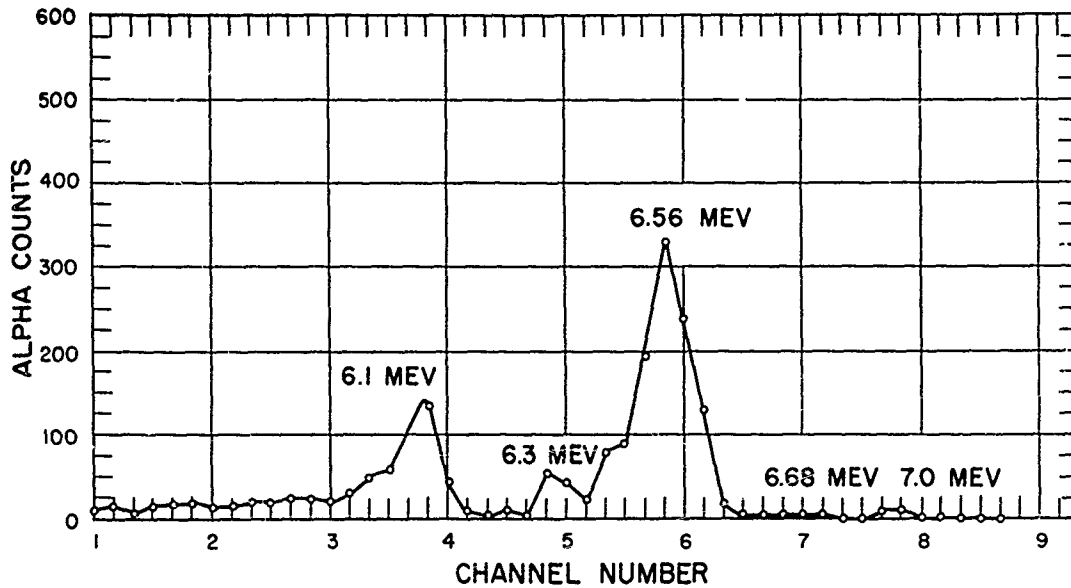


Fig. 8.10—ANL elution curve No. 2; pulse analysis of drop 69, 1040 min.

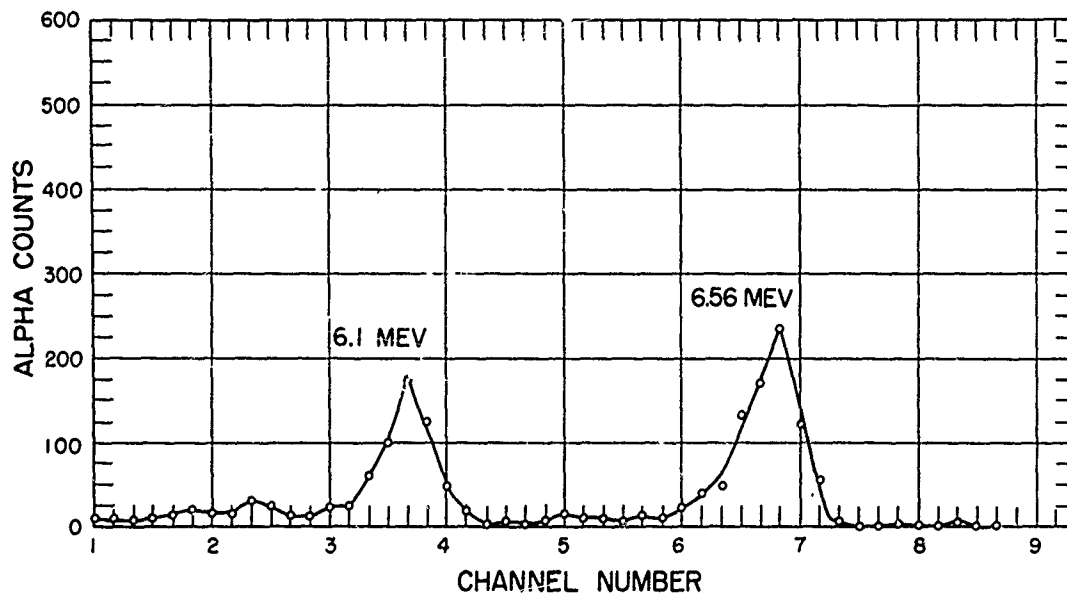


Fig. 8.11—ANL elution curve No. 2; pulse analysis of drop 70, 912 min.

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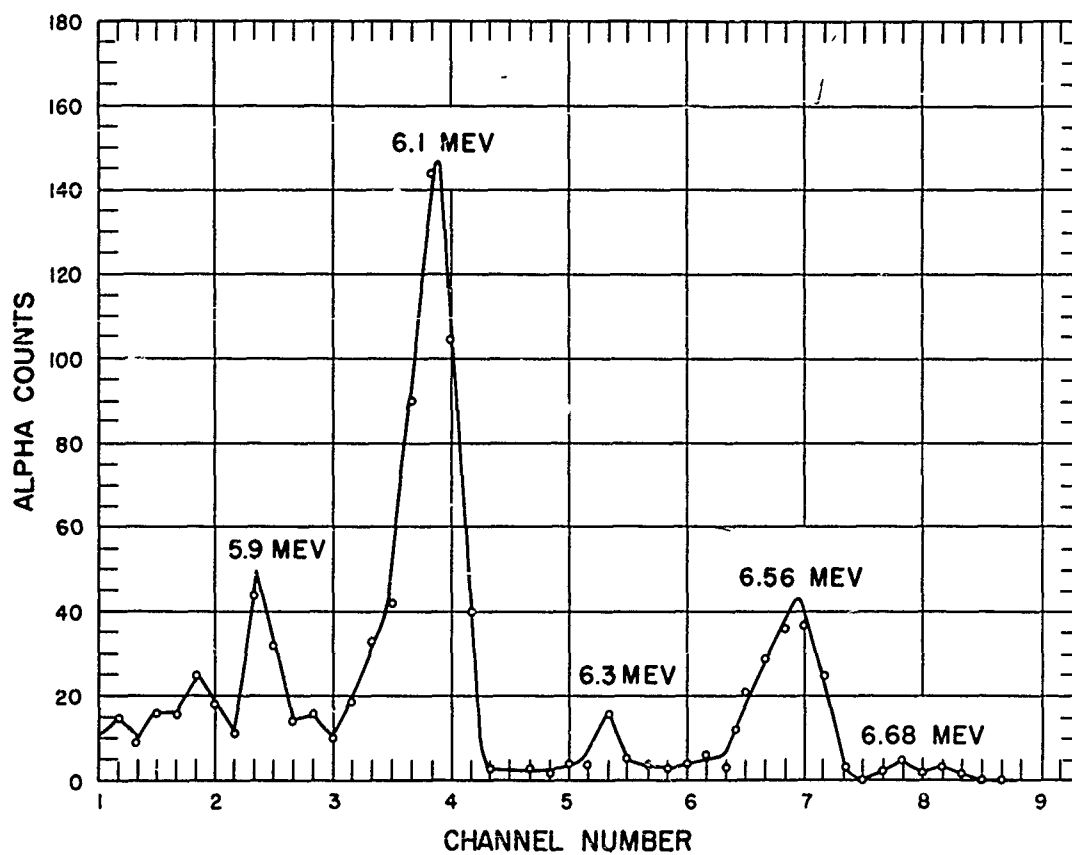


Fig. 8.12—ANL elution curve No. 2; pulse analysis of drop 73, 680 min.

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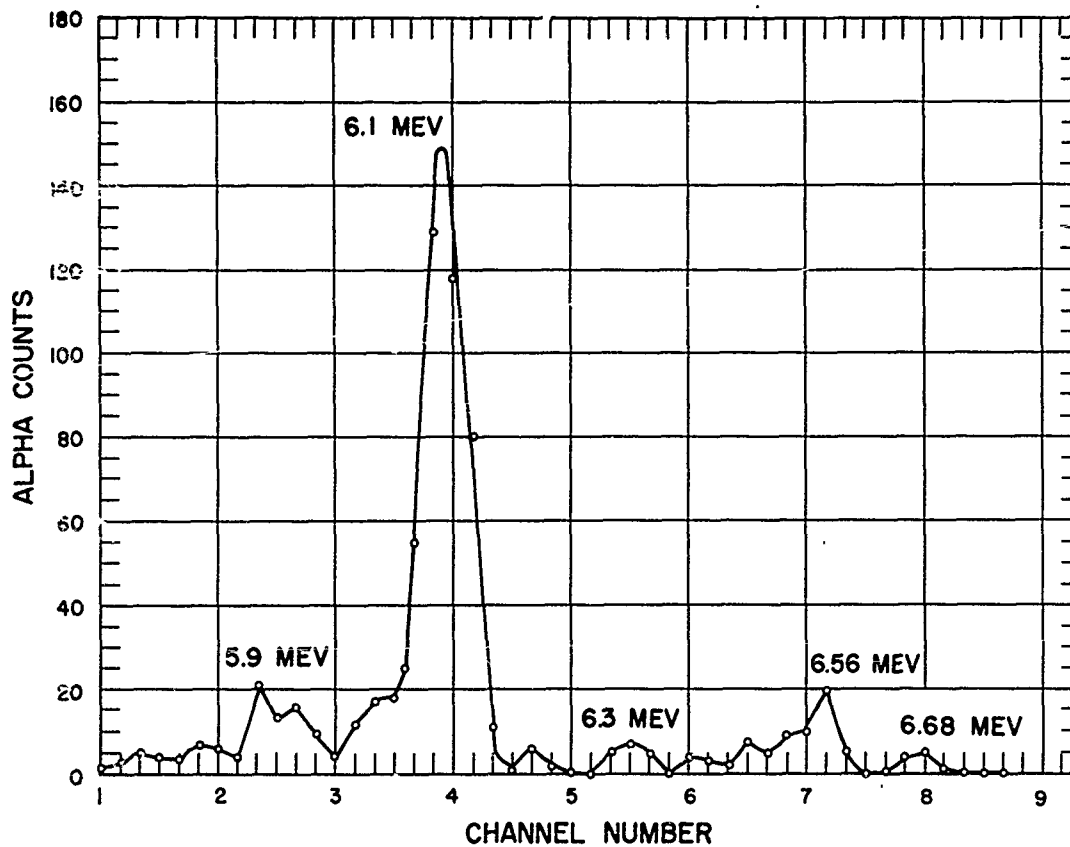


Fig. 8.13—ANL elution curve No. 2; pulse analysis of drop 75, 820 min.

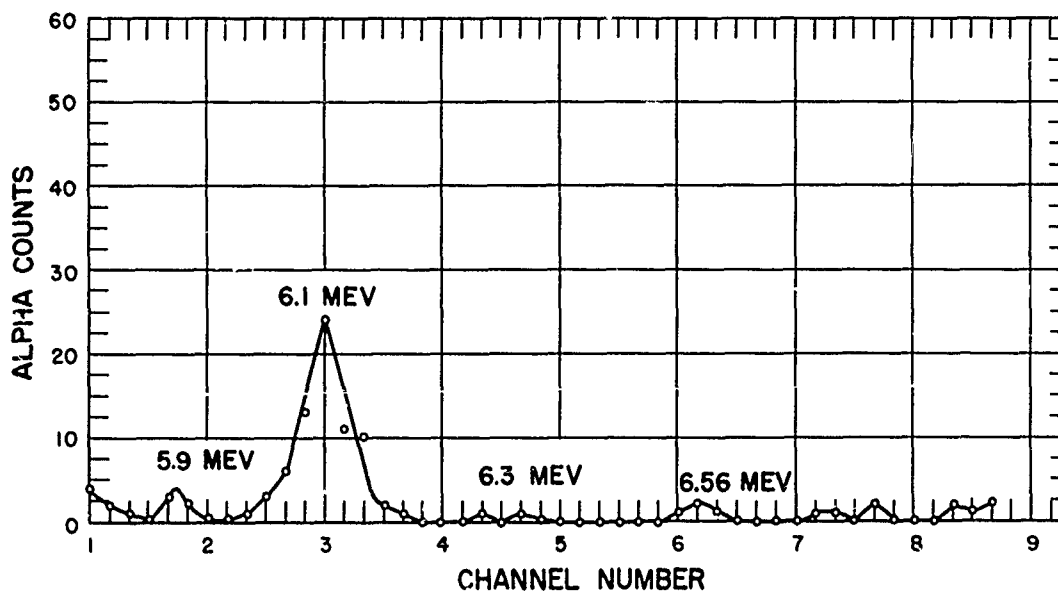


Fig. 9.14—ANL elution curve No. 2; pulse analysis of drop 77, 448 min.

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both eluted after the 6.56-Mev alpha-particle activity. The constancy of the ratio of the 6.1-Mev alpha activity to the spontaneous fissions led to the interpretation that they were due to isotopes of the same element. It was concluded from the position of the elution peak and from spontaneous-fission systematics (see Chap. 9) that the element was probably californium. On following the samples of the first elution for decay, ANL chemists found that the spontaneous-fission activity decayed with a 68-day half-life in all drop samples, whereas no decay in the 6.1-Mev alpha activity was detectable. Figures 8.15 to 8.17 illustrate the spontaneous-fission decay in the gross sample and in the individual drops.

On the basis of the decay of the spontaneous fissions and the constancy of the 6.1-Mev alpha activity, the ANL group concluded that there were two californium isotopes present with very low alpha activity associated with the spontaneously fissioning nuclide. A mass number of 254 was tentatively assigned to the fissioning nuclide from spontaneous-fission systematics and the estimated alpha half-life.

In later experiments an alpha-particle activity of 5.9 Mev was detected which was interpreted as associated either with the californium or with the berkelium. Also, the possibility was suggested that the 6.3-Mev alpha particle was associated with the decay of a nuclide of higher atomic number than californium.

The results of a pulse analysis of an elution obtained at UCRL with a coral sample are illustrated in Fig. 8.18, and the alpha spectrum of the californium fraction is given in Fig. 8.19. Two peaks, of 6.1 and 5.8 Mev, were interpreted as associated with californium isotopes; the peak at 5.48 Mev was interpreted as associated with Am^{241} breaking through the column. In later experiments the yield of the 5.8-Mev alpha particles was found to increase relative to the 6.1-Mev peak; this is consistent with the evidence found in the berkelium fraction for the existence of a berkelium beta-particle emitter of intermediate half-life. From the amount of berkelium beta activity, the time of growth, and the amount of 5.8-Mev alpha activity which grew in the californium fraction, the half-life of this californium isotope (tentatively designated as Cf^{250} or Cf^{251}) was calculated to be approximately 400 years.

The UCRL group found by observing the decay of the 6.1-Mev alpha peak over a period of three months that its half-life was greater than five months. They reasoned from the relation between the energy and half-life of alpha-particle emitters that the minimum possible half-life value for a californium isotope emitting an alpha particle of this energy should be about 10 years. If the mass number is even and the half-life is, then, 10 years, reference to the mass-yield curve discussed in Chap. 6 leads to the conclusion that the amount of activity observed requires a mass assignment of 252. In order for this hypothesis to be consistent, the UCRL group considered the possibility of a minor closed shell of 152 neutrons. An alternative assumption made was that the 6.1-Mev alpha activity was due to Cf^{249} , and on this basis the observed yield indicated an alpha half-life of roughly 500 years.

Early experiments at UCRL revealed a high rate of spontaneous fission in the californium fraction, which was observed to decay with a half-life of approximately two months, as illustrated in Fig. 8.20. No alpha decay of this half-life was found. From this half-life and the amount of activity, it was concluded from the mass-yield curve that the mass number should be 256, but it was stressed that the two-month half-life was only a gross value from three months of observation and that the contribution from Cf^{252} and Cf^{254} could not be determined. It was believed, however, that these two isotopes could not contribute more than one-half the observed fissions. The Cf^{256} was expected to be beta unstable and to decay through elements 99 to 100^{256} . A search for alpha particles of energy greater than 6.6 Mev was without success; therefore it was concluded that, unless 100^{256} has an alpha-particle energy in the vicinity of 6.6 Mev (in which case it would be masked by the 6.6-Mev alpha particles of element 99 which were found to grow into the californium fraction), the beta half-life of Cf^{256} must be at least 20 times its spontaneous-fission half-life.

The UCRL chemists observed that a 6.6-Mev alpha activity grew into the californium fraction. Since an alpha particle of this energy was observed in the element 99 fraction, it was suspected that a beta-emitting californium isotope was present. The californium fraction was

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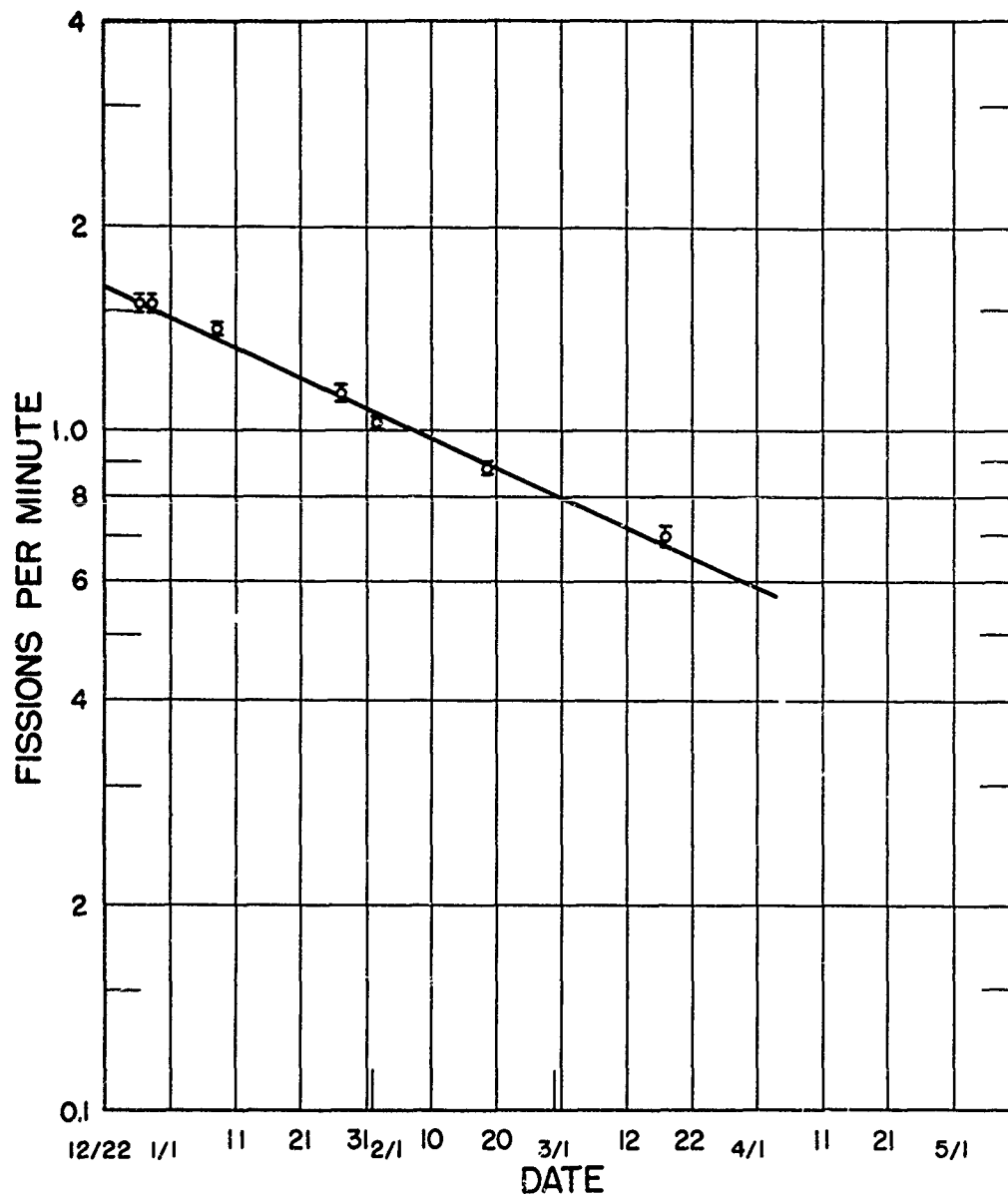


Fig. 8.15—ANL elution No. 1; spontaneous-fission decay of drops 18, 19, 20, 21, and 22 (combined).

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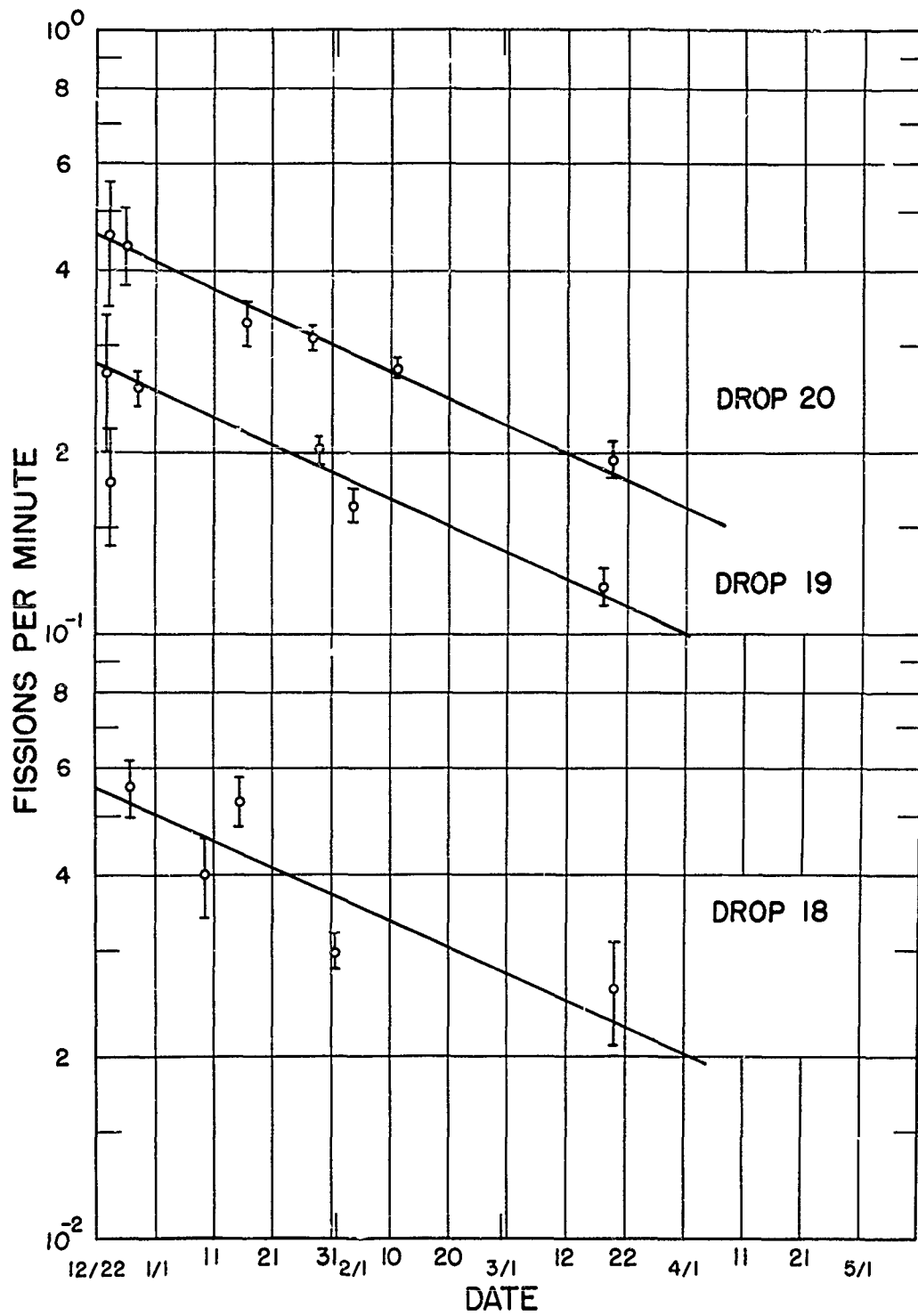


Fig. 8.16—ANL elution No. 1; spontaneous-fission decay of drops 18, 19, and 20.

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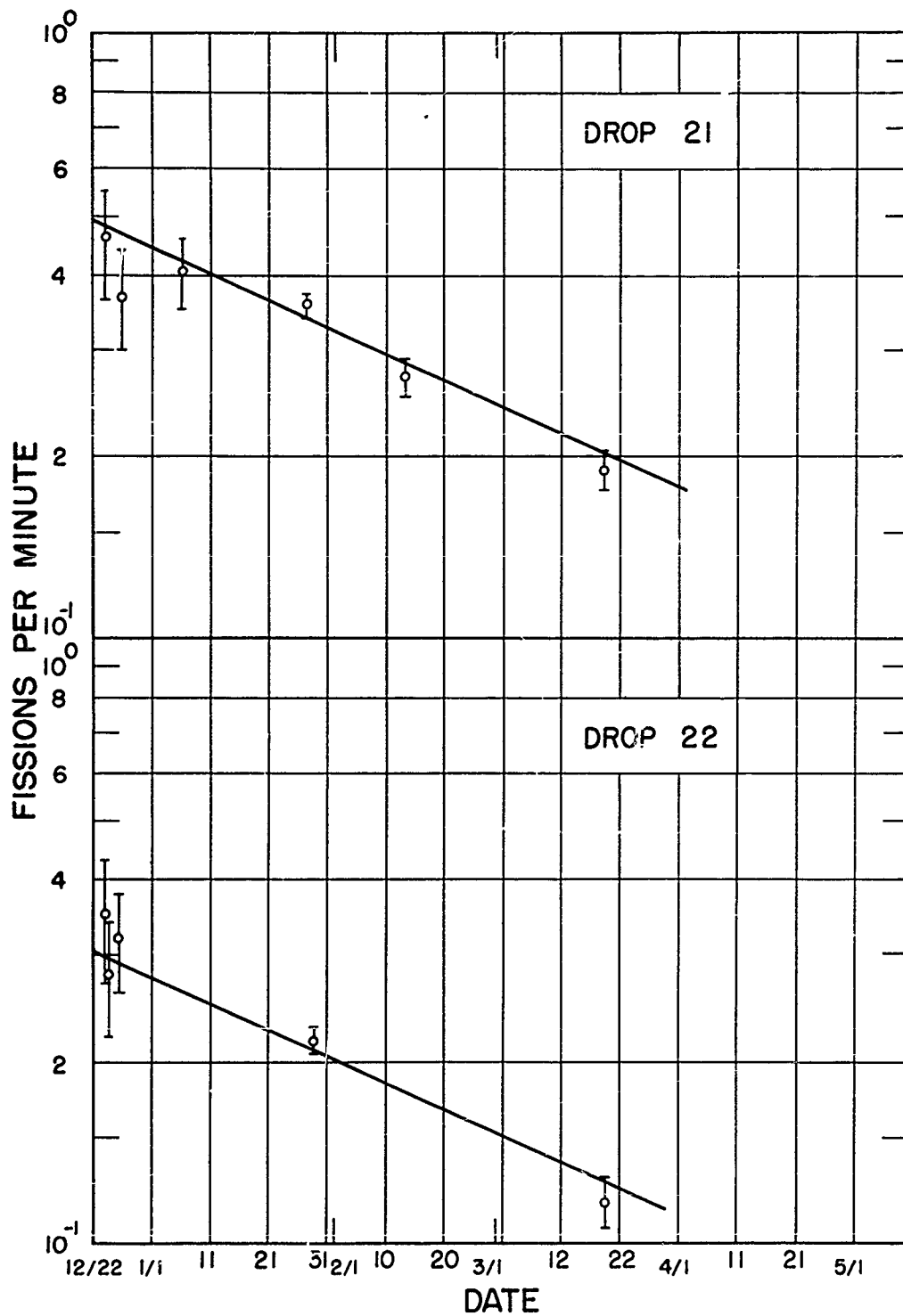


Fig. 8.17—ANL elution No. 1; spontaneous-fission decay of drops 21 and 22.

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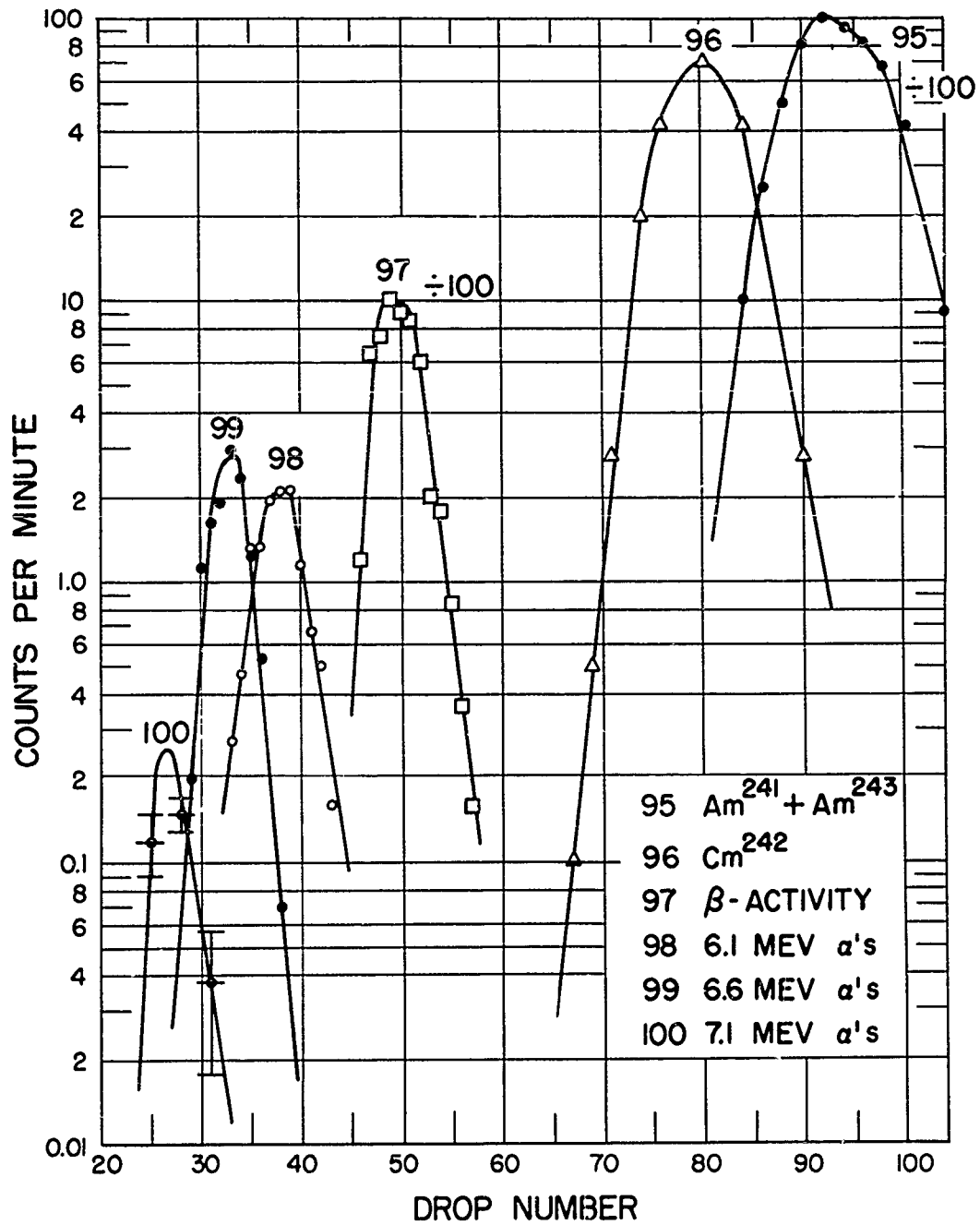


Fig. 8.18—UCRL elution curve; coral sample, pulse analysis.

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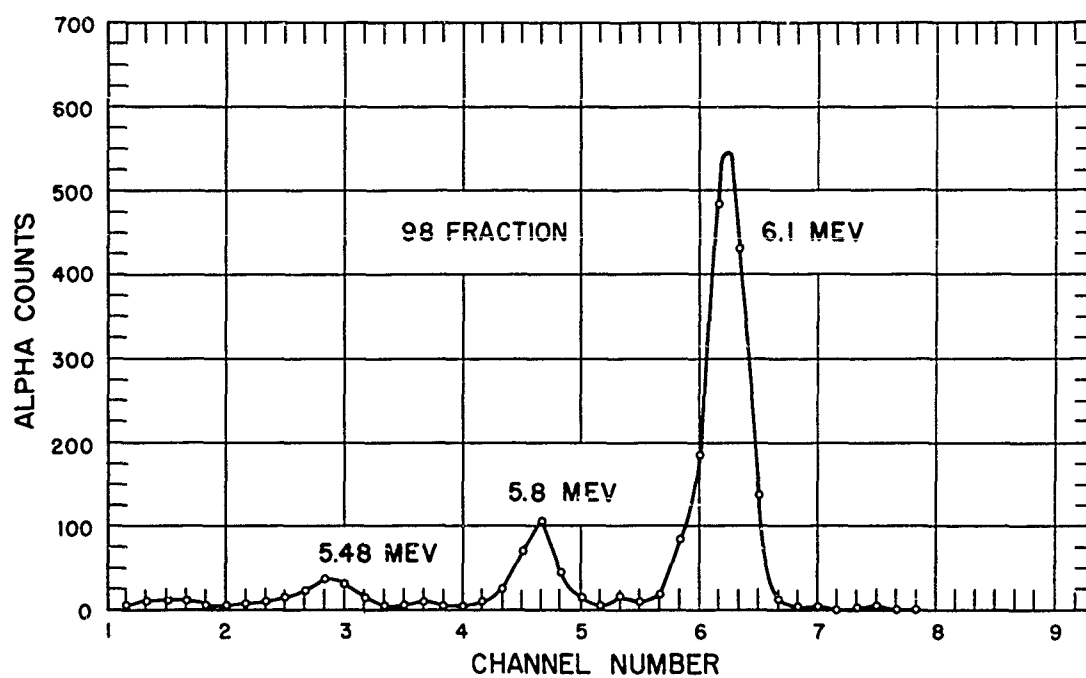


Fig. 8.19—UCRL elution curve; coral sample, pulse analysis of californium fraction, 525 min.

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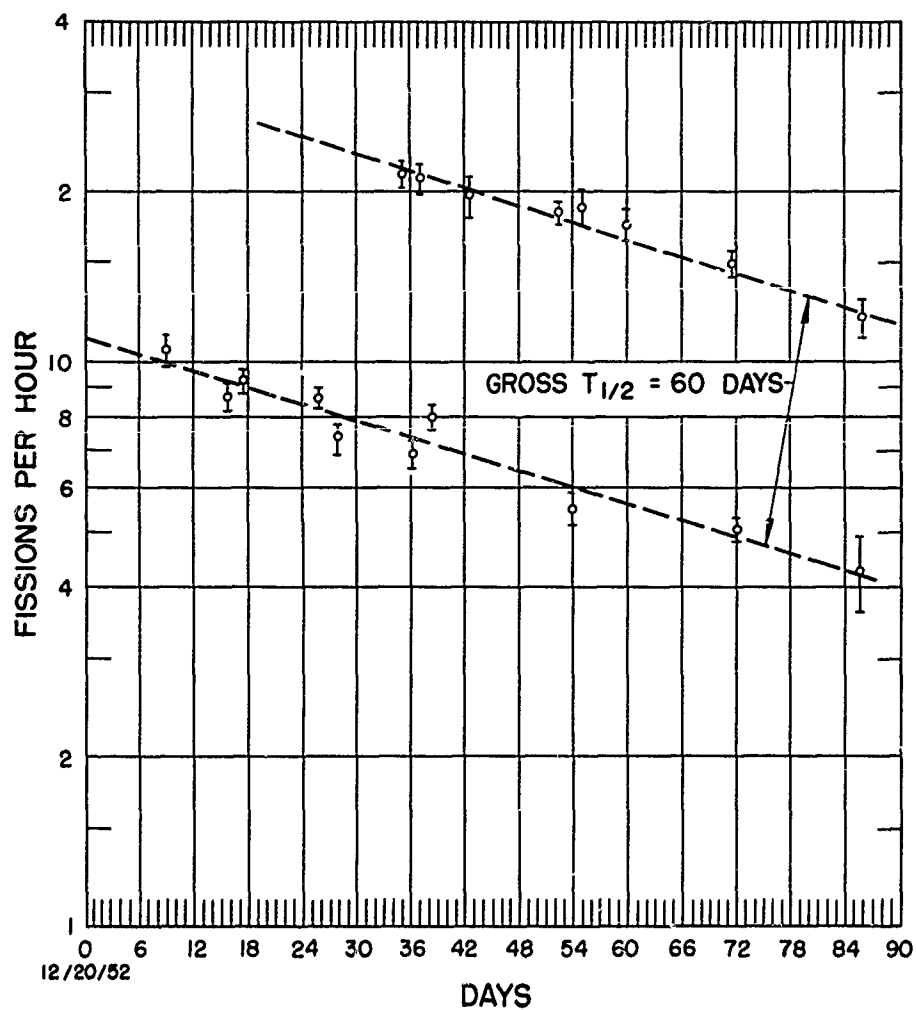


Fig. 8.20—UCRL elutions; spontaneous-fission decay in californium fraction.

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followed in the pulse analyzer for about three months, with the results shown in Figs. 8.21 and 8.22. Since the sample of Fig. 8.21 contained an overlapping portion of the element 99 fraction, the growth of the 6.6-Mev alpha particle was not observable but may be seen in Fig. 8.18 with a sample from a subsequent experiment in which great care was taken in purification. The two experiments indicate a half-life of approximately one month for the parent californium beta emitter; for the daughter element 99 alpha emitter, the data are consistent with the 22-day half-life found in the 99 fraction.

In contrast with these data the ANL group has been unable to observe any growth of activity of 6.56-Mev alpha particles in the elution fractions which they attribute to californium. Figures 8.23 to 8.26 present the gross-decay data on drops of their second elution. They remark, however, that there is some indication in the data of their first run that there may have been some growth of 6.56-Mev alpha activity from a short-lived (approximately 10-day) californium isotope. No resolution of this discrepancy is available at this time.

8.3 ELEMENT 99 ACTIVITIES

In an early elution the UCRL group came to the conclusion that a transcalifornium isotope emitting 6.6-Mev alpha particles had been chemically separated from the other activities. It was not possible, however, to distinguish chemically between elements 99 and 100 in the earliest experiments. To confirm the chemical identification of the new element, the UCRL group performed another elution on new material using cyclotron-produced 1.5-day Cf^{246} , which emits a 6.76-Mev alpha particle, as a tracer. The 6.6-Mev alpha-particle activity was found to elute just ahead of the californium tracer activity in a position analogous to the rare-earth position of holmium and was thus concluded to be an isotope of element 99. Figure 8.27 shows the separation of the two peaks on the column as shown by the pulse analyses of adjacent elution fractions. In the same experiment most of the spontaneous-fission activity was shown to follow closely the Cf^{246} elution peak.

The decay of the 6.6-Mev alpha activity was observed over a period of about two months and revealed, after corrections, a decay corresponding to a half-life of 22 days. Figure 8.28 illustrates the data of these observations. The yield of this activity corresponded closely to that given by the mass-yield curve for mass 253, which, in conjunction with the patterns of beta stability established from closed decay cycles, led the UCRL chemists to assign a mass number of 253 to this nuclide. A difficulty arose in the UCRL interpretation in that the yield of the 6.6-Mev alpha activity was too high by a factor of 10 to arise from a californium parent with a one-month half-life. It was thus postulated that either there is a short-lived beta-emitting isomer of Cf^{253} or there are two isotopes of element 99 emitting alpha particles of the same energy.

The data of the ANL group led to a different interpretation of the situation. Their observation that the 6.56-Mev alpha activity eluted before the peak which they attributed to californium (Figs. 8.2 and 8.6) led to the conclusion that the activity was due to the presence of one or more elements of atomic number higher than 98. Observations of the decay of the elution fractions across the 6.56-Mev alpha elution peak led to the conclusion that the activity which eluted first decayed more rapidly than that which eluted late in the peak. Figures 8.29 to 8.34 show the gross decay of drops 17 to 22 of ANL elution 1. These data were interpreted by the ANL group to mean that there were two alpha activities with alpha-particle energies close to 6.56 Mev and with different half-lives. The shorter lived nuclide was considered an isotope of element 100, and the longer lived was considered an isotope of element 99. Efforts to resolve the 6.56-Mev alpha activity into two peaks by pulse analysis were inconclusive, as was an attempt to detect an energy shift in the 6.56-Mev peak in samples from the two extremes of the elution peak. It was concluded that the energy difference of the alpha particles of the two nuclides was less than 80 kev.

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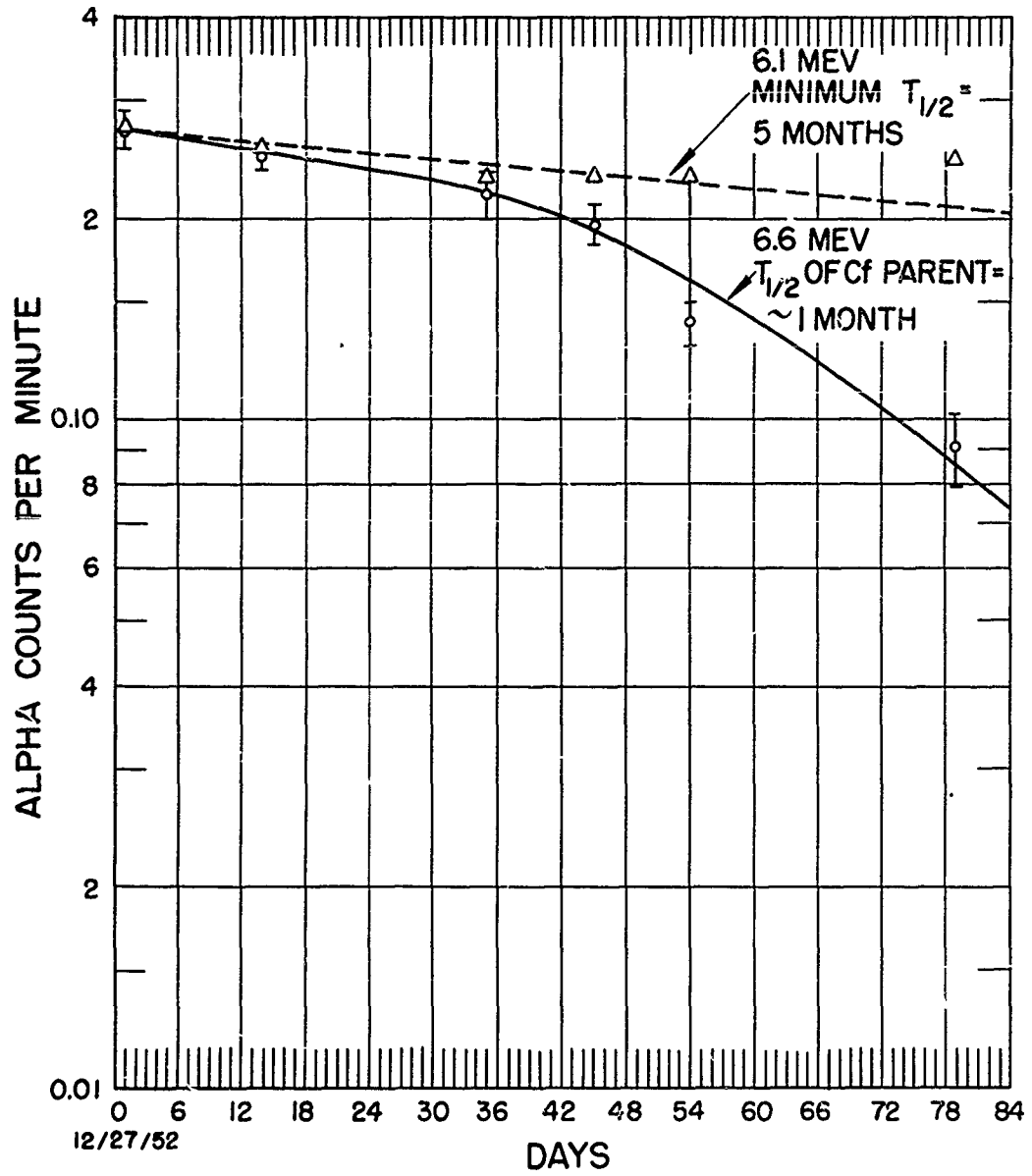


Fig. 8.21—UCRL elutions; decay of 6.6-Mev alpha activity in californium fraction.

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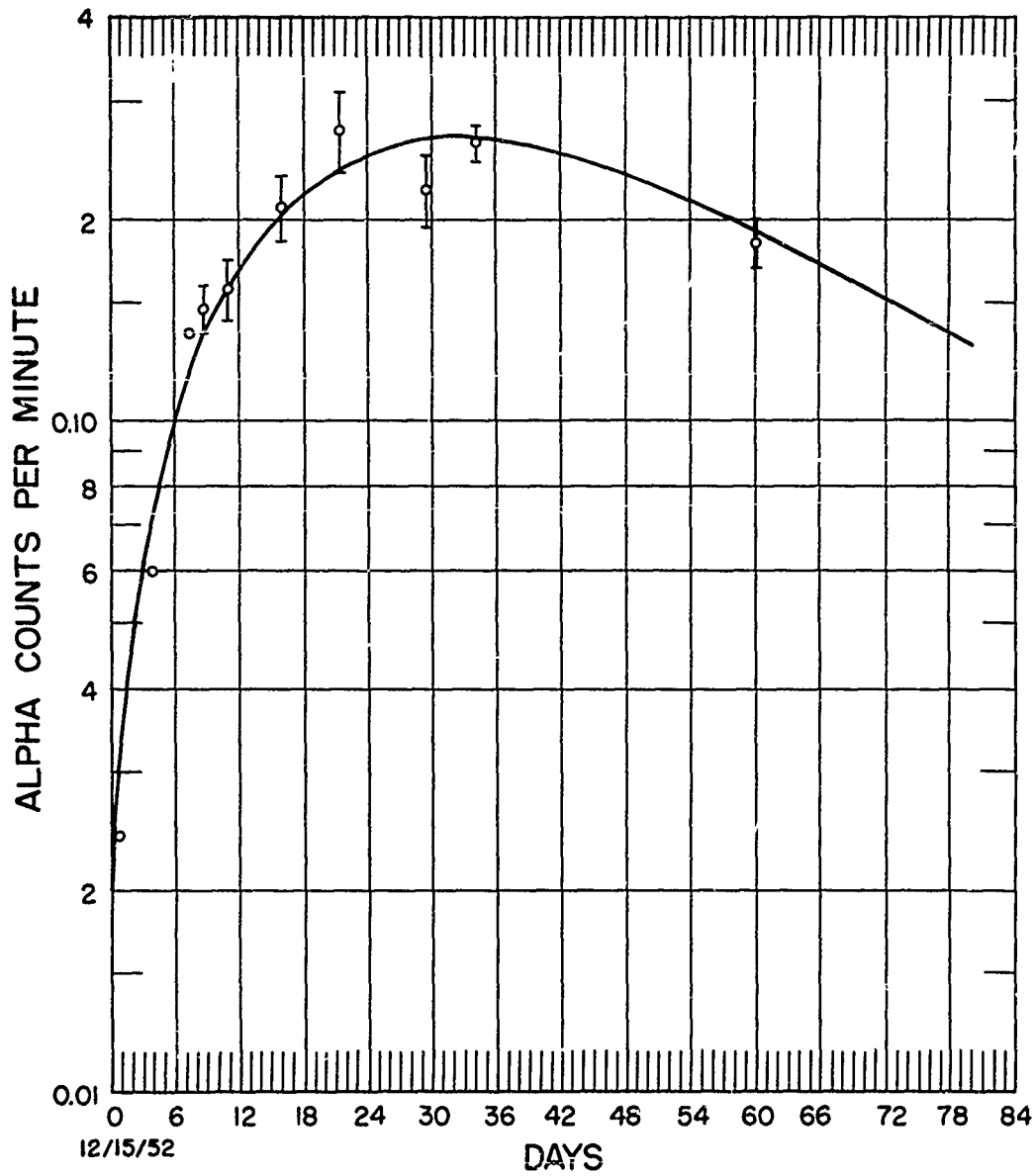


Fig. 8.22—UCRL elutions; growth and decay of 6.6-Mev alpha activity in californium fraction.

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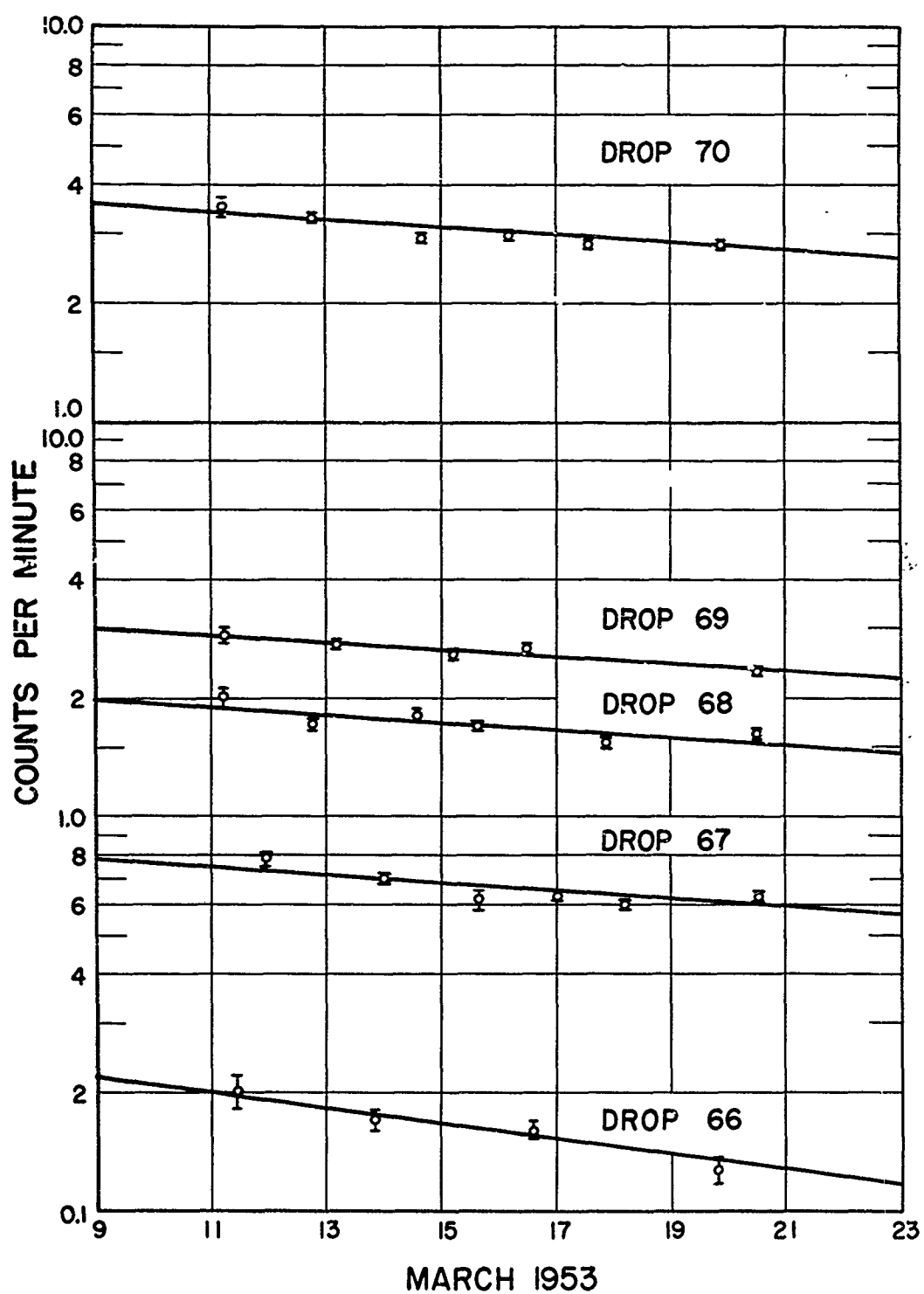


Fig. 8.23—ANL elution No. 2; decay of drops 66, 67, 68, 69, and 70.

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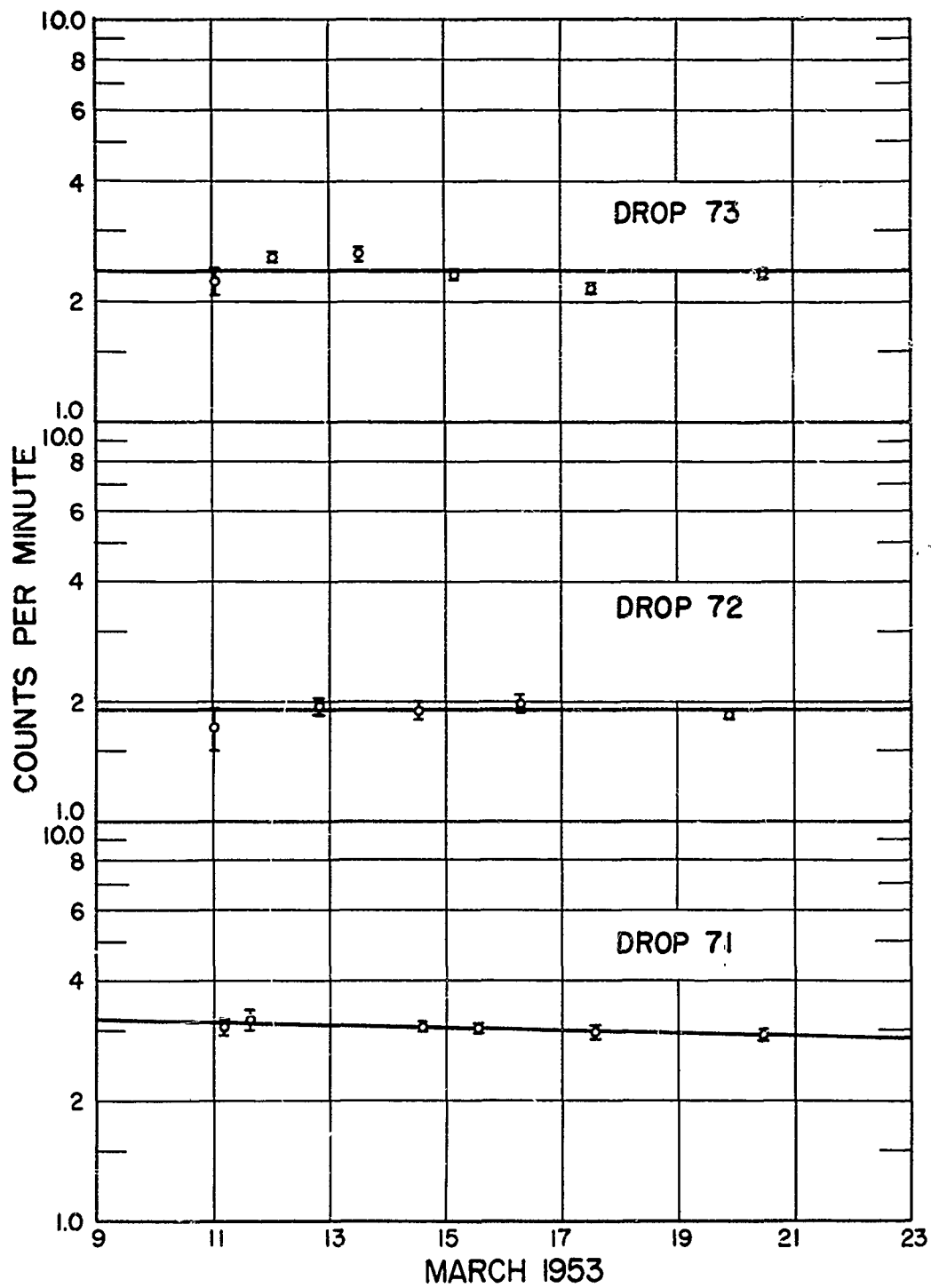


Fig. 8.24—ANL elution No. 2; decay of drops 71, 72, and 73.

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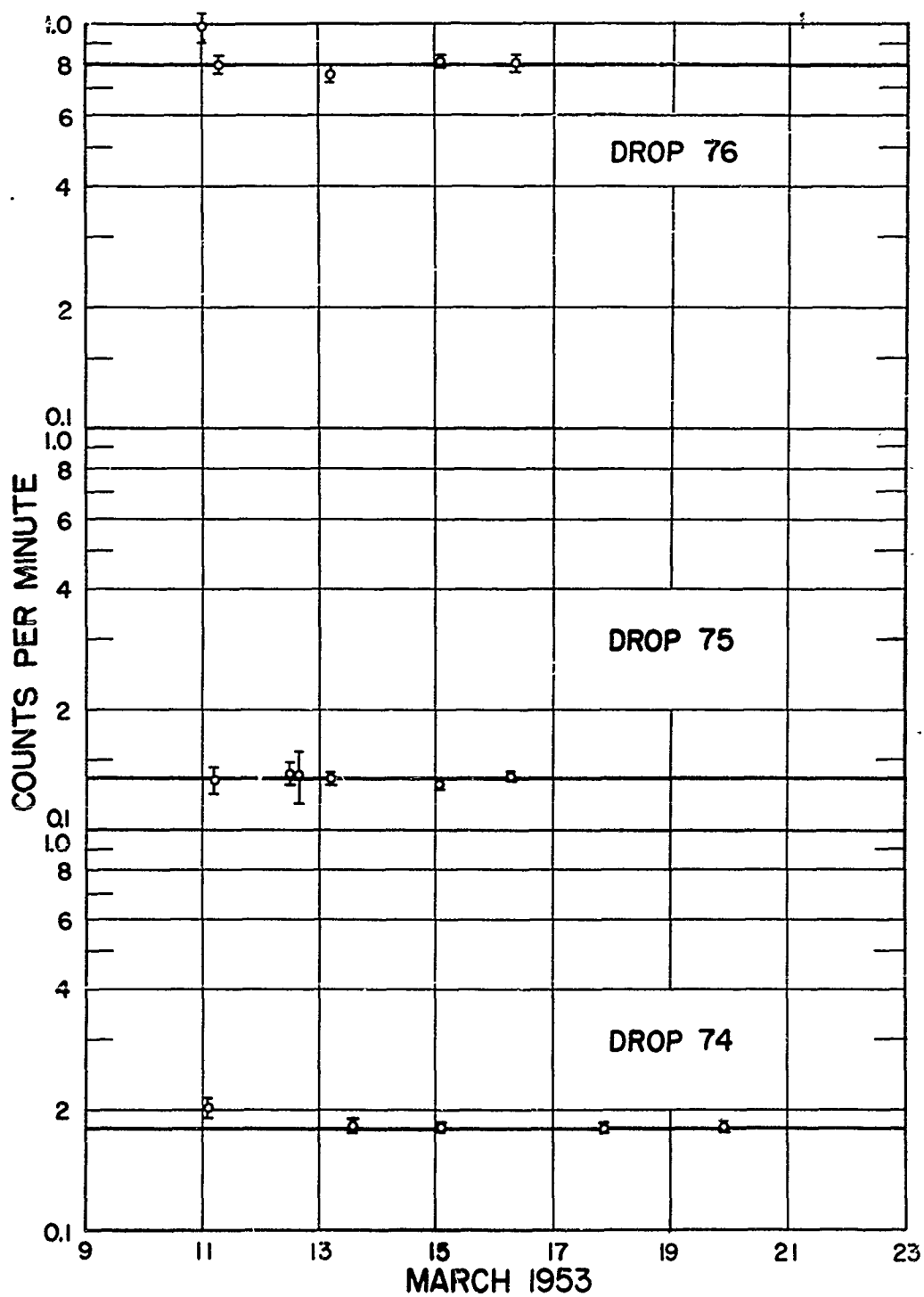


Fig. 8.25—ANL elution No. 2; decay of drops 74, 75, and 76.

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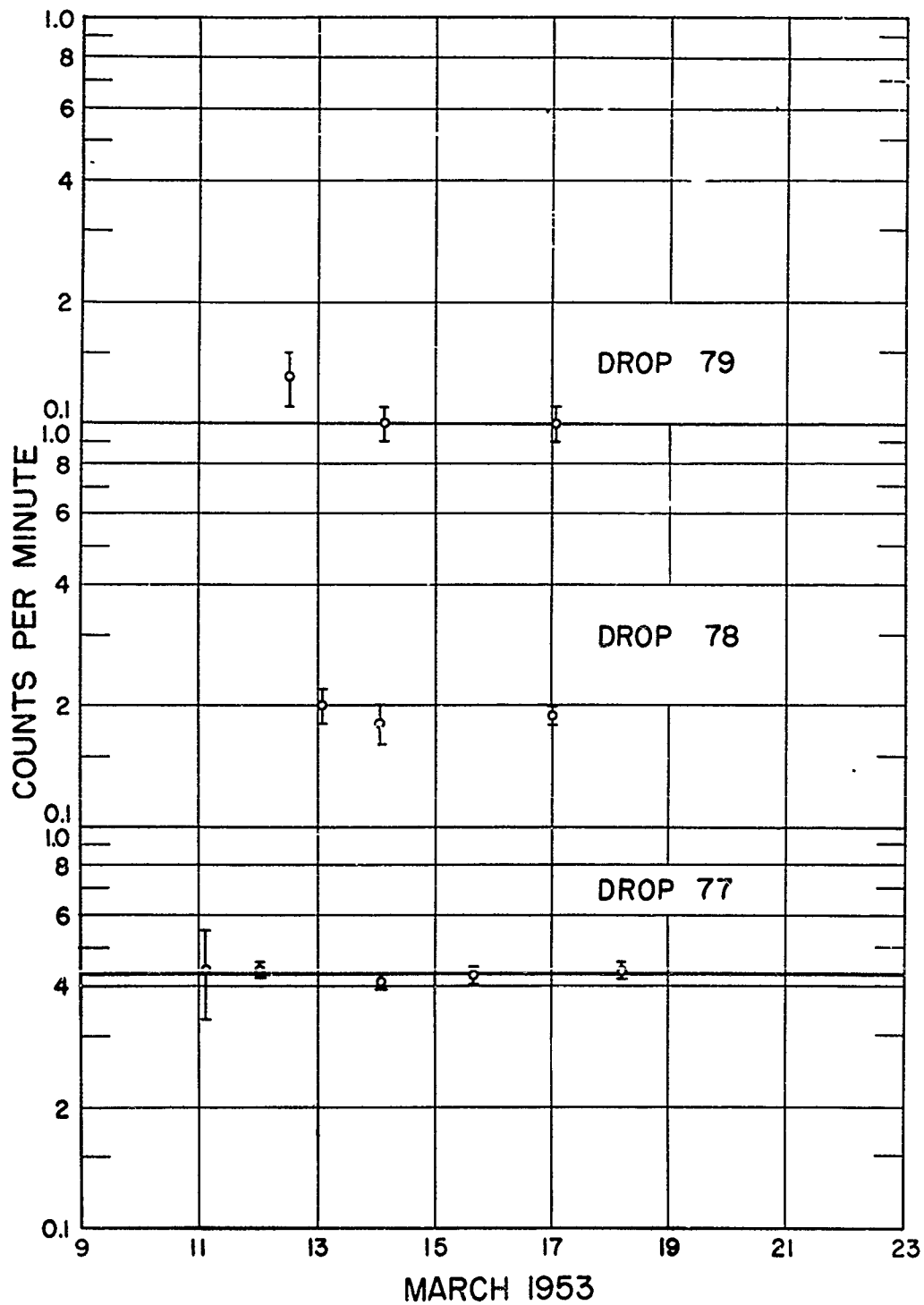


Fig. 8.26—ANL elution No. 2; decay of drops 77, 78, and 79.

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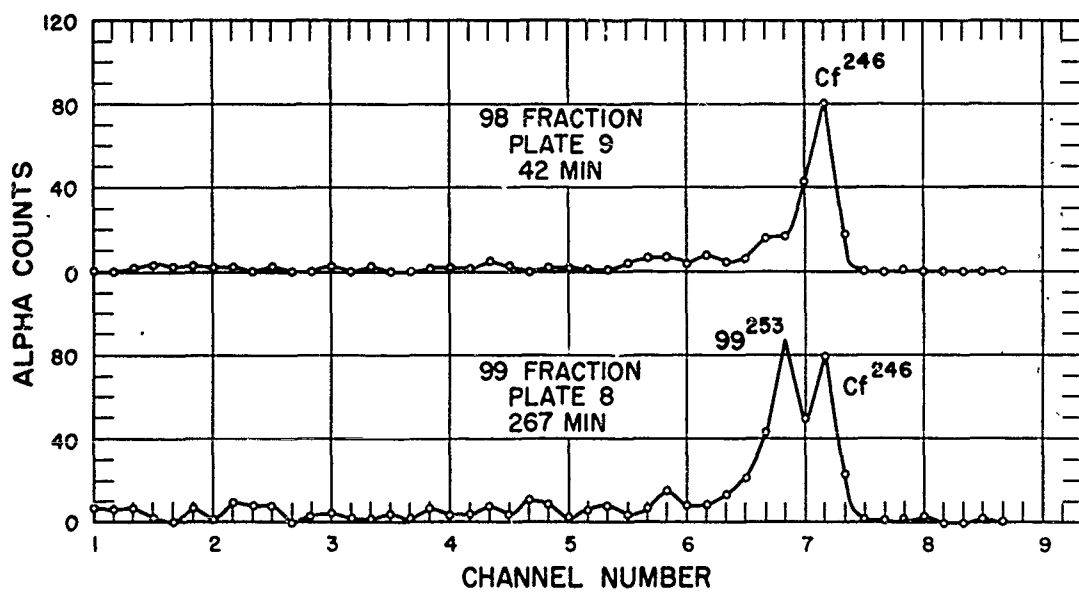


Fig. 6.27—UCRL elutions; pulse analyses of californium and 99 fractions.

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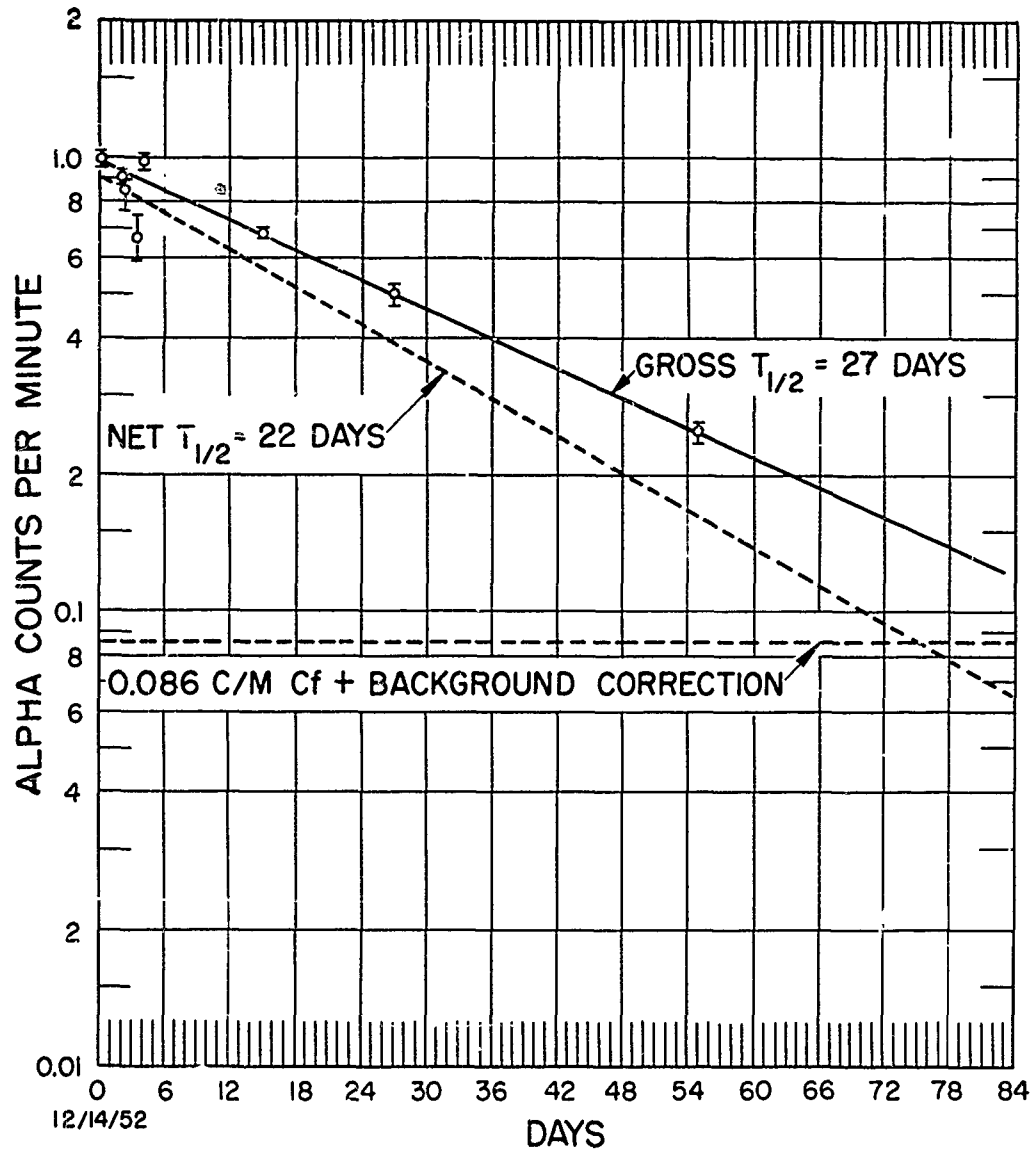


Fig. 8.28—UCRL elutions; decay of 6.8-Mev alpha activity in 99 fraction.

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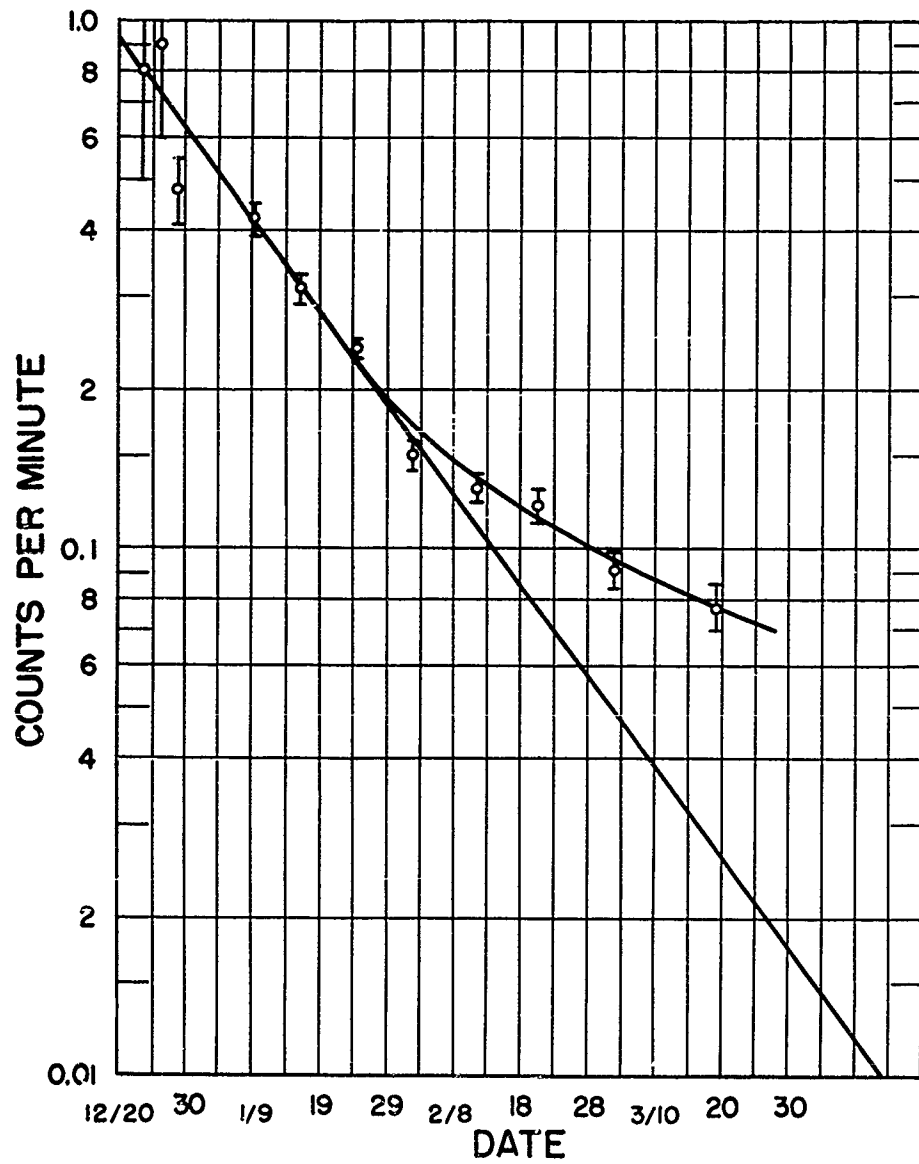


Fig. 8.29—ANL elution No. 1; decay of drop 17.

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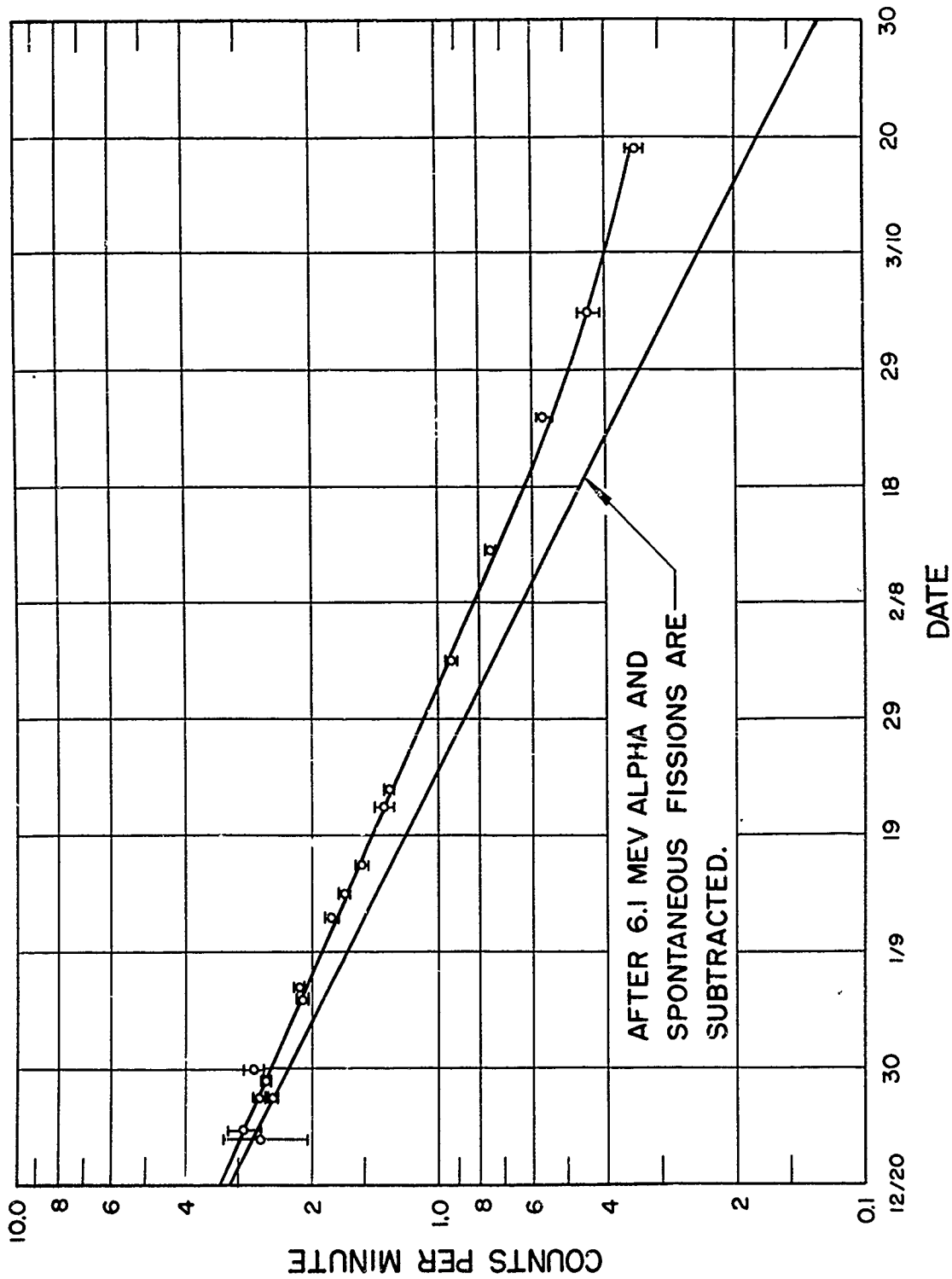


Fig. 8.30—ANL elution No. 1; decay of drop 18.

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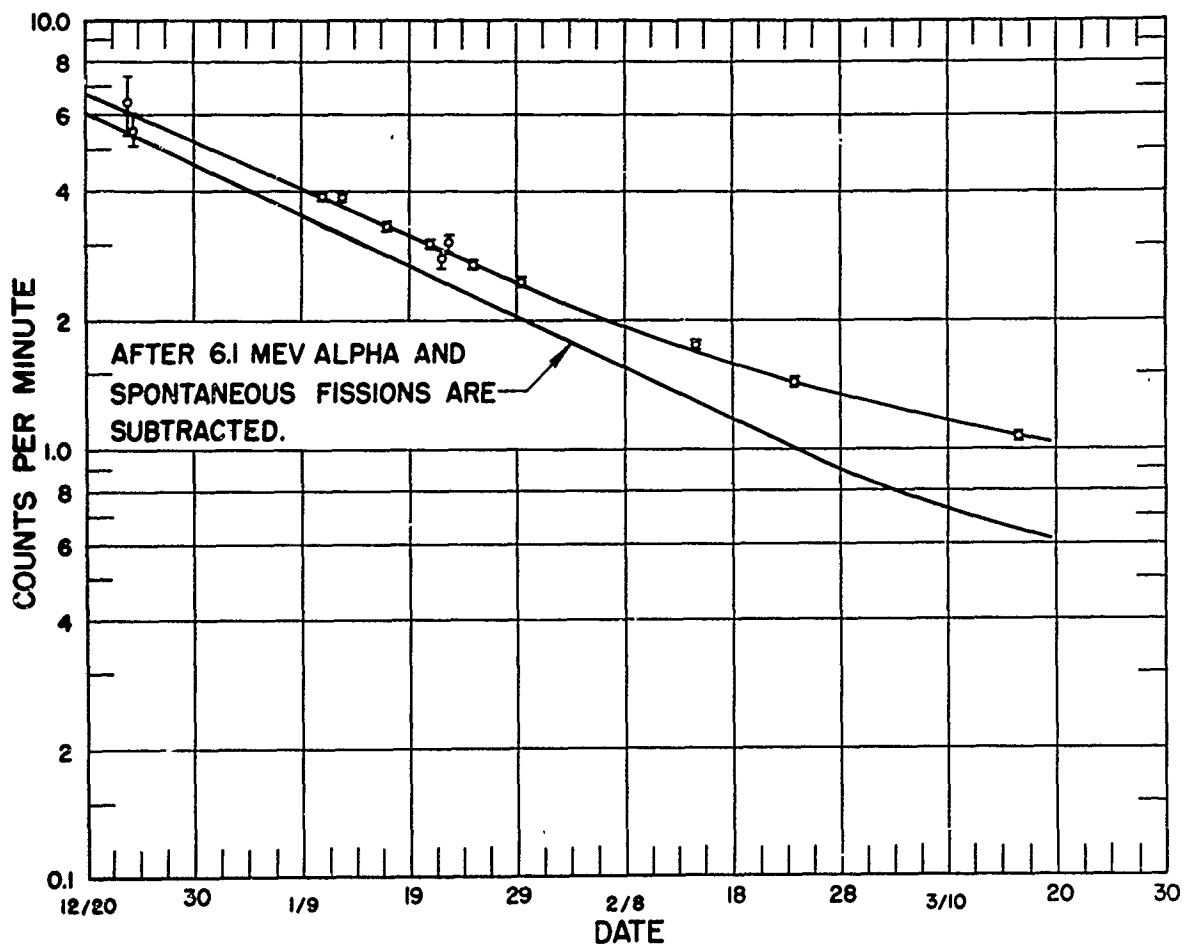


Fig. 8.31—ANL elution No. 1; decay of drop 19.

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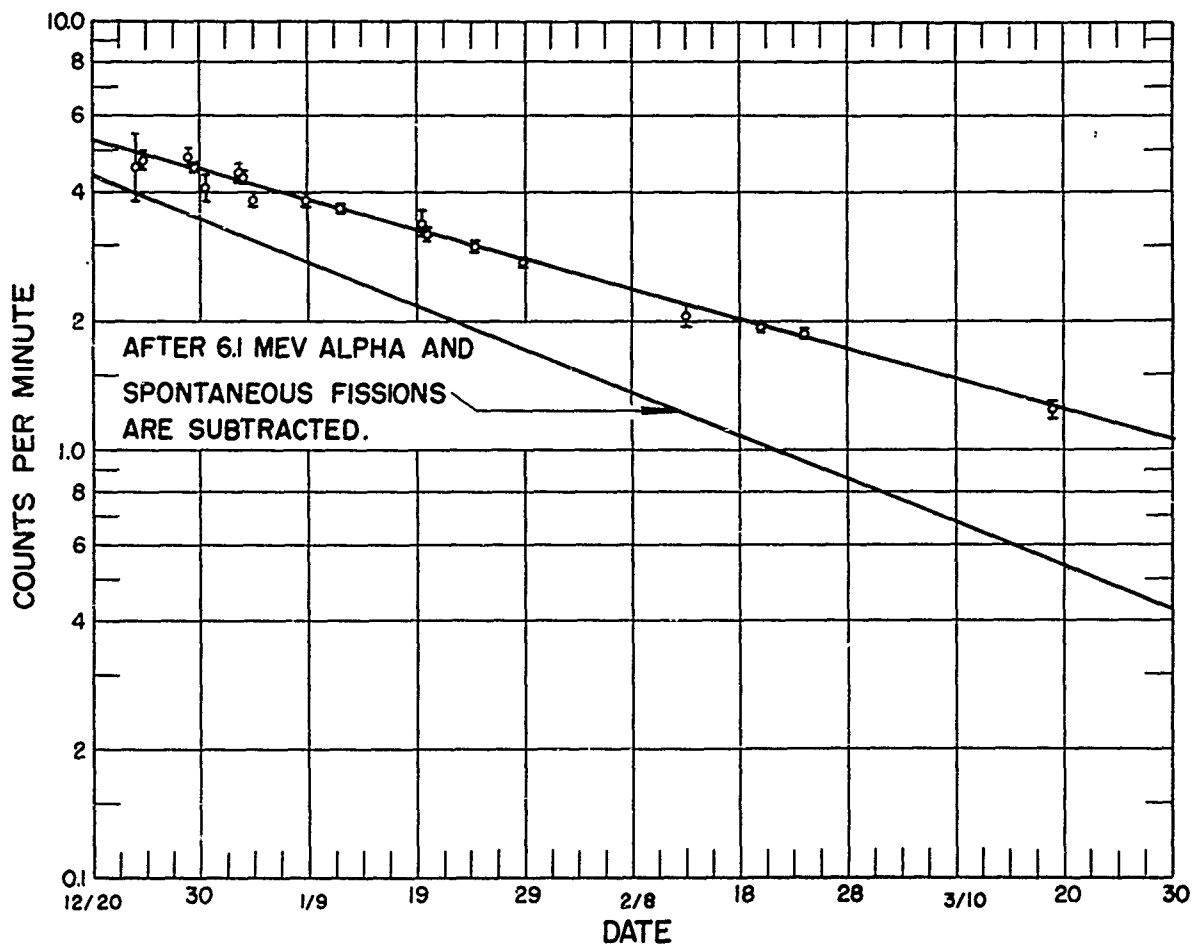


Fig. 8.32—ANL elution No. 1; decay of drop 20.

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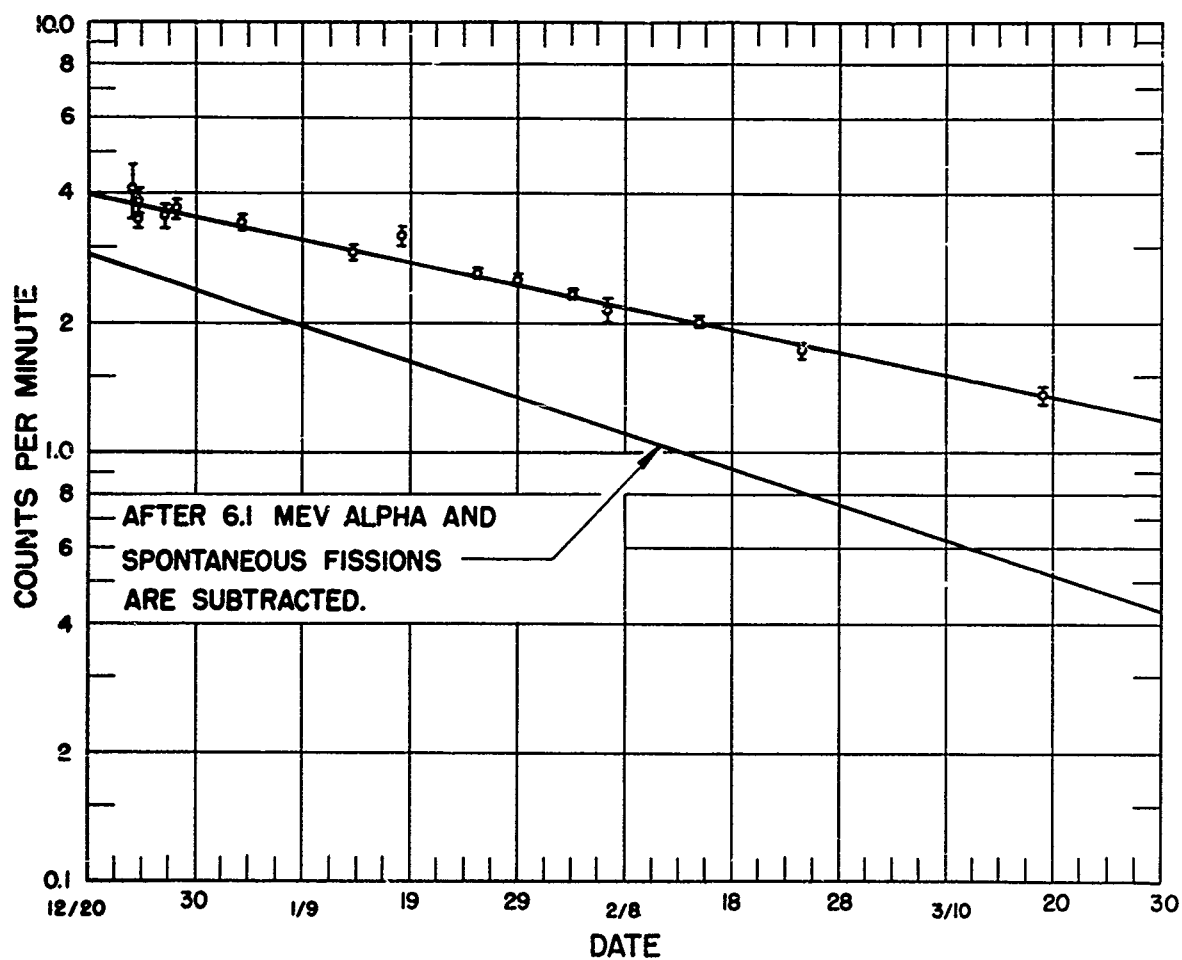


Fig. 8.33—ANL elution No. 1; decay of drop 21.

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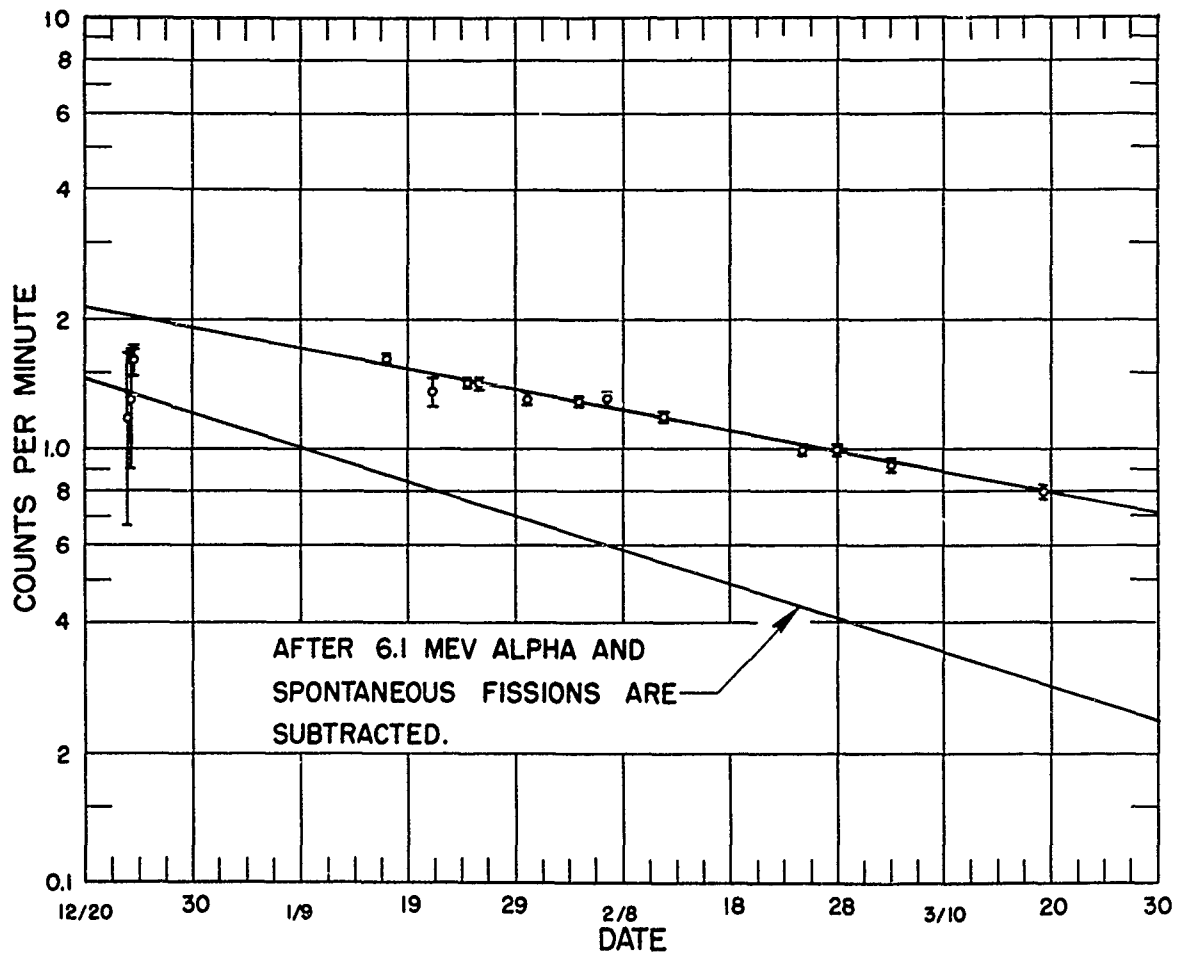


Fig. 8.34—ANL elution No. 1; decay of drop 22.

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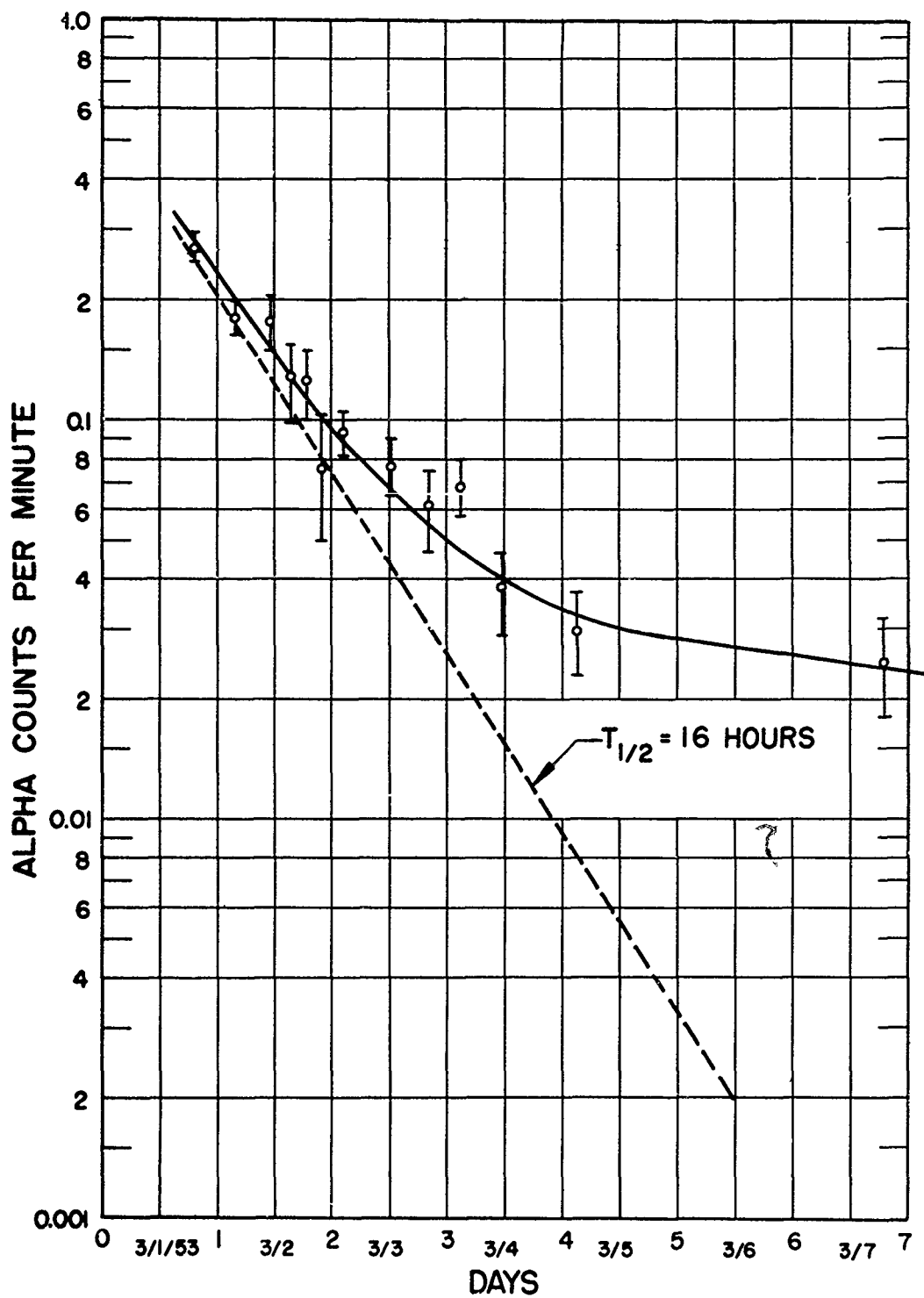


Fig. 8.35—UCRL elutions; decay of 7.1-Mev alpha activity in 100 fraction.

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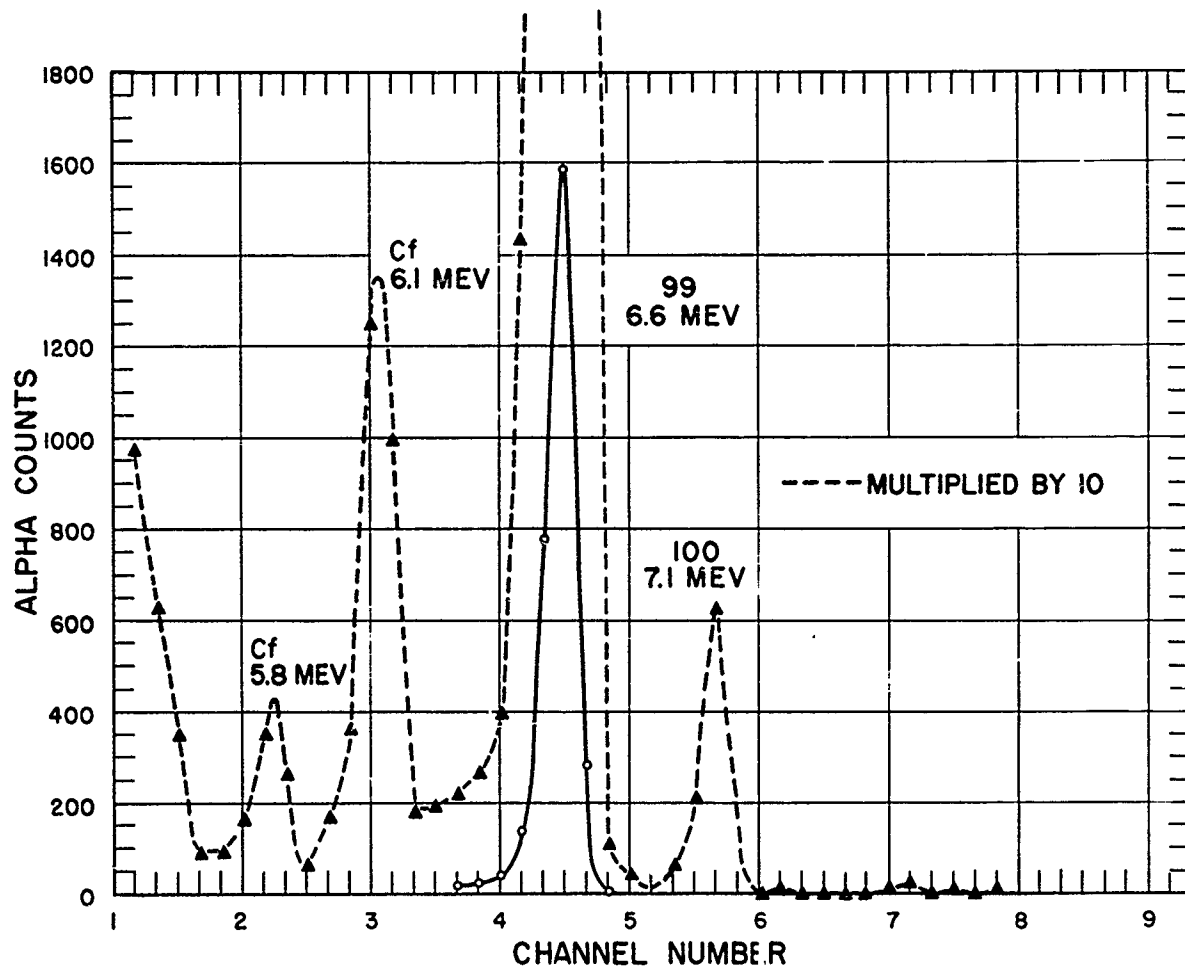


Fig. 8.36—UCRL elutions; pulse analysis of 99 fraction, 902 min.

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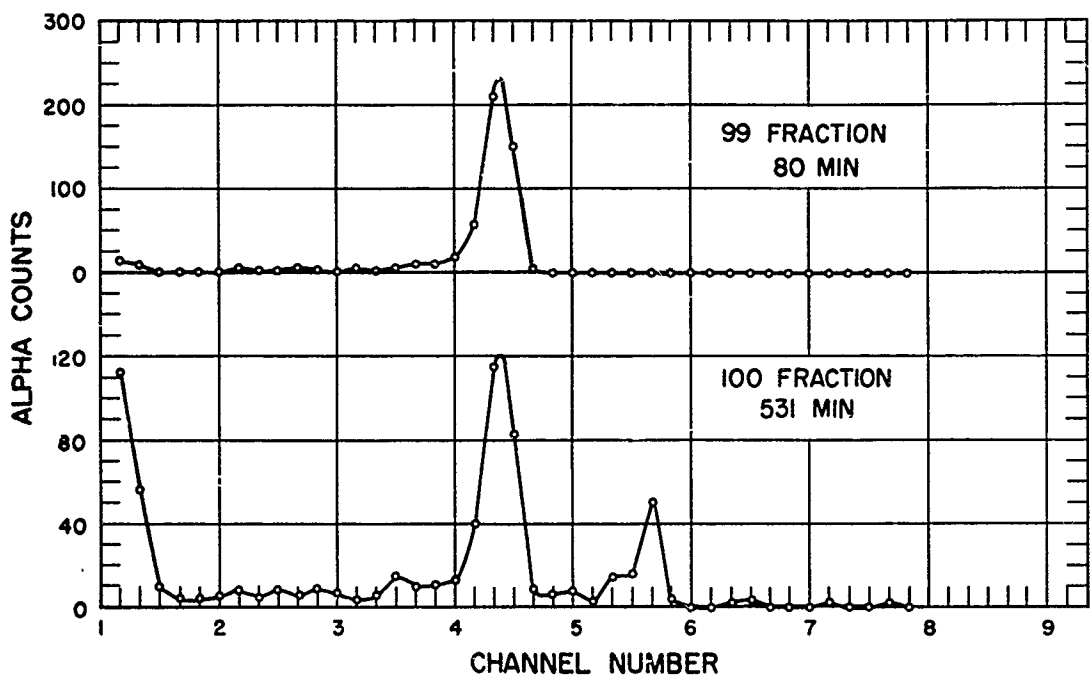


Fig. 8.37—UCRL elutions; separation of 99 and 100.

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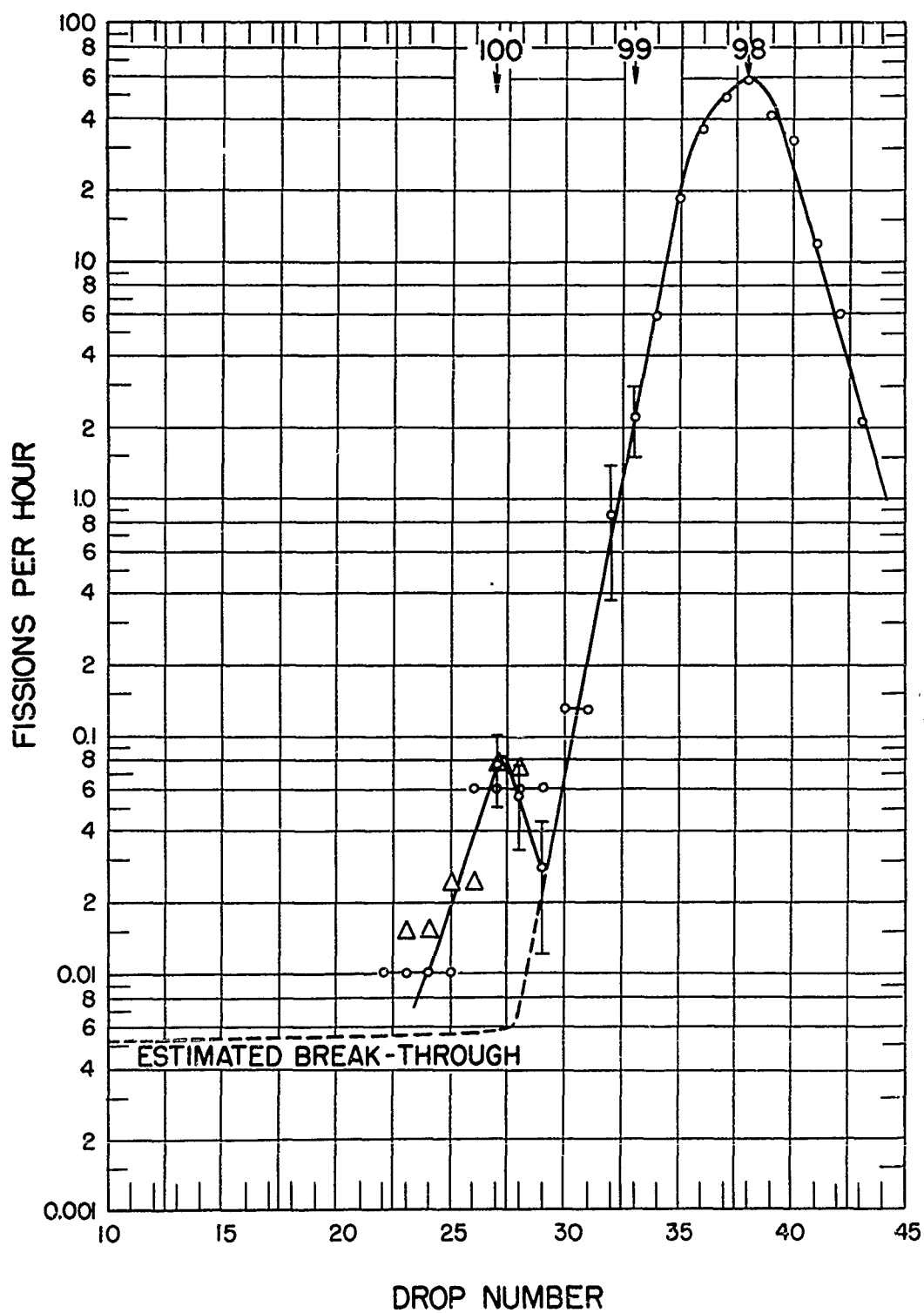


Fig. 8.38—UCRL elutions; spontaneous-fission elution in the 98, 99, and 100 region.

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The data from this elution and the second ANL elution indicate a range of half-lives from 15 to 40 days for the 6.56-Mev activity, with the fraction of the shorter activity much less on the second elution than the first. The interpretation of the data combined from the two runs was that there existed two activities which decayed by emission of a 6.56-Mev alpha particle; one of these activities, of about a 15-day half-life, was due to an isotope of element 100, probably of mass 255; the other activity, of about a 35- to 50-day half-life, was due to an isotope of element 99, probably of mass 253.

8.4 ELEMENT 100 ACTIVITIES

A 7.1-Mev alpha activity in transplutonium fractions was first observed at LASL and shortly thereafter at UCRL. In an experiment at UCRL on filter-paper samples, long counts revealed the presence of a 7.1-Mev alpha-particle activity in the elution fraction just ahead of the fraction attributed to element 99. This activity had essentially disappeared two days after the initial count which led to the conclusions that its half-life was of the order of one day and that its presence at this time could only result from its production by a beta emitter in the 99 fraction. Further analysis of the 99 fraction revealed the presence of the 7.1-Mev alpha activity in an abundance of about 4 per cent. The 99 fraction was then recombined, and the experiment was repeated, with the result that the 7.1-Mev peak was once again observed to elute in advance of the 99 fraction.

A later experiment on samples from coral yielded considerably more 7.1-Mev activity; therefore the UCRL group was able to derive a more accurate half-life for this activity. Figure 8.35 illustrates the data which led to an assignment of 16 hr as the half-life value. Figure 8.36 shows the 6.6-Mev alpha activity in the 99 fraction, together with the equilibrium amount of 7.1-Mev element 100 activity; the 7.1-Mev activity was not observable in the 99 fraction immediately after separation. The californium activity in Fig. 8.36 arises from incomplete separation from the neighboring californium peak and is a small (approximately 5 per cent) fraction of the total californium activity. Figure 8.37 shows the chemical separation between elements 99 and 100 as determined by alpha pulse analyses on adjacent elution fractions.

The UCRL group could not identify alpha activity other than the 7.1-Mev activity as being that of an isotope of element 100 in experiments to the level of 0.02 c/m. Figure 8.38 shows a spontaneous-fission elution curve over the region of elements 98, 99, and 100. The small elution peak, which corresponded to a total of some 2 fissions/day, was considered as possibly arising from element 100, but the statistical difficulty of locating the exact position of this very small peak precluded the elimination of the possibility that the fissions arose from isotopes of elements 101 or 102. For a heavy isotope of element 100, such as 100^{258} or 100^{260} , the UCRL chemists considered that the alpha half-life might be expected to be longer than that for spontaneous fission, and therefore the very low yield of fissions would be consistent with a mass number of 258 or 260. They reasoned that an odd-neutron isotope of element 100 could not be ruled out on the basis of the present knowledge of spontaneous fission but that it might be expected that even-odd isotopes should be hindered by factors of 100 or more in the probability that they would undergo this type of decay. It was concluded that the alpha to fission ratio of greater than 50 exhibited by the 7.1-Mev activity was consistent with a tentative mass assignment of 255.

The ANL group observed alpha groups of 6.35, 6.7, and 7.0 Mev. On the basis of pulse analyses of elution samples, it was concluded that the 6.35-Mev alpha activity was associated with an emitter of atomic number higher than that of the 6.56-Mev alpha emitter interpreted as element 99. On this basis the 6.35-Mev alpha activity was assigned to an isotope of element 100. The 6.7-Mev emitter was considered to be a nuclide of atomic number higher than 98. The 7.0-Mev activity could not be found in advance (by elution) of the 6.56-Mev elution peak but was observed with that activity in an abundance of about one-fiftieth of the 6.56-Mev activity as of Mar. 20, 1953.

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CHAPTER 9

SPONTANEOUS-FISSION SYSTEMATICS

9.1 ANL SYSTEMATICS

In the interpretation of the spontaneous-fission data in the experiments reported here, systematics of different types were employed at ANL and UCRL. At ANL a systematic relation was noted in plots of the log of the ratio of spontaneous-fission half-life to the alpha half-life vs Z^2/A . The section of the report from the ANL group concerning these systematics is quoted below.

"Kramish⁵ has plotted the logarithm of the ratio of spontaneous fission half-life to alpha half-life against Z^2/A for even-even nuclides. He drew straight lines through the points representing nuclides which differ by an alpha particle. Figure 9.1 is a similar plot containing additional points based on spontaneous fission data not available to Kramish. The isotopes differing in mass number by six units and in charge by two units appear to fall approximately on straight lines. It will be noted that there are three points based on measurements for each of three out of the six lines shown in Fig. 9.1. The isotopes of a given element seem to fall on a smooth curve. The family of curves indicated that Pu^{242} should have a shorter spontaneous fission half-life than Pu^{240} . To check this, samples of plutonium with widely different $\text{Pu}^{242}/\text{Pu}^{240}$ ratios were measured for spontaneous fissions. The spontaneous fission rate of Pu^{242} was found to be 1.8 ± 0.2 times that of Pu^{240} . The Cm^{246} point was calculated on the assumption that all the fissions and all 5.4 Mev alpha activity are due to Cm^{246} . The data of Fig. 9.1 suggest that Cf^{254} may have a spontaneous fission rate high enough to account for the decay. It falls on a family with Th^{230} , U^{238} , and Pu^{242} . Since the spontaneous fission half-life used for Th^{230} is only a limit that point should probably be higher. The true slope of the line through these points may be such that the predicted fission to alpha rate for Cf^{254} may be as high as 10^4 or 10^5 . The alpha half-life one would predict from systematics is not so long but that a spontaneous fission half-life of 68 days for this isotope is reasonable. Cf^{256} , if present, would also show a high spontaneous fission rate. The spontaneous fission systematics indicate that the ratio of fission to alpha decay for the corresponding 100 isotopes would be much less."

9.2 UCRL SYSTEMATICS

At UCRL the systematics employed plots of the spontaneous-fission half-life vs the neutron number or the ratio of the atomic number to the neutron number. Huizenga has pointed out that spontaneous-fission rates at constant atomic number increase with mass number at sufficiently high values of mass number; this is in disagreement with the simpler systematics of Seaborg which predict for a given atomic number a continually decreasing spontaneous-fission rate with increasing mass number.⁶ The relations revealed are shown in Figs. 9.2 and 9.3. The sections of the UCRL report which discuss these systematics are quoted below.

"To extend the spontaneous fission data for purposes of correlating the results from these experiments a measurement of the half-life of Pu^{242} for this process was made on a highly enriched sample prepared by neutron irradiation of Am^{241} at the Materials Testing Reactor in Idaho. The isotopic composition

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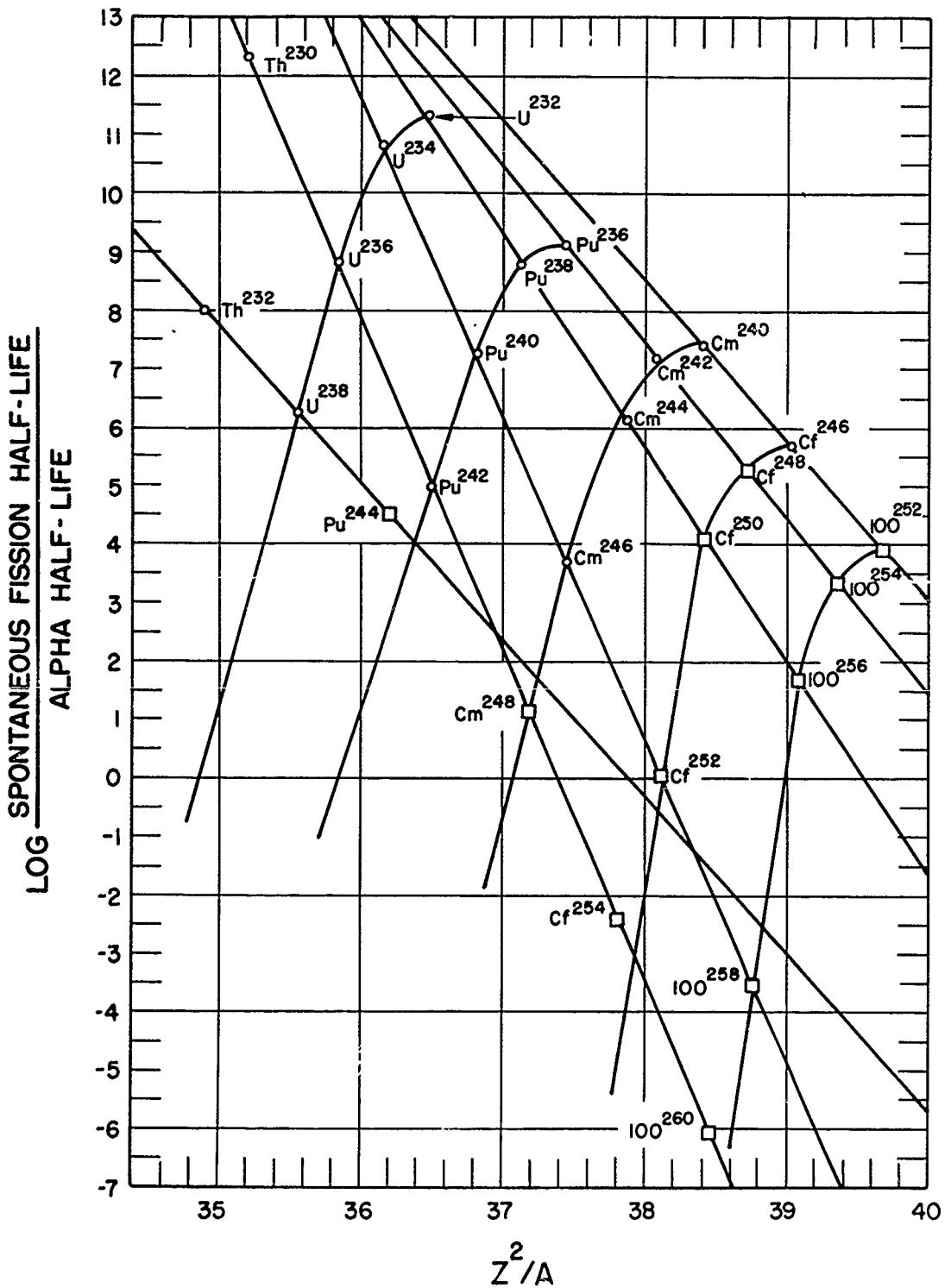


Fig. 9.1—ANL spontaneous-fission systematics.

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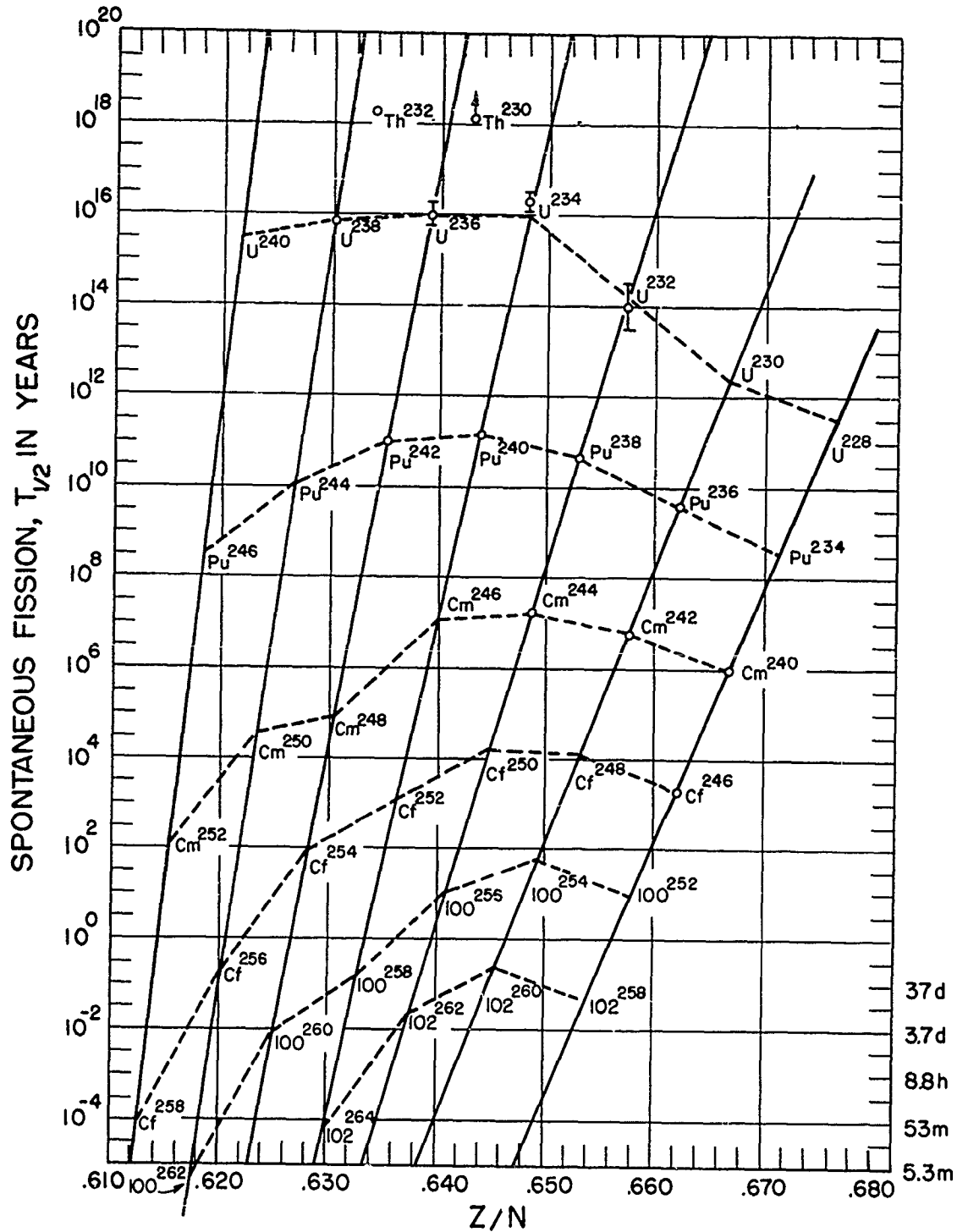


Fig. 3.2—UCRL spontaneous-fission systematics (I).

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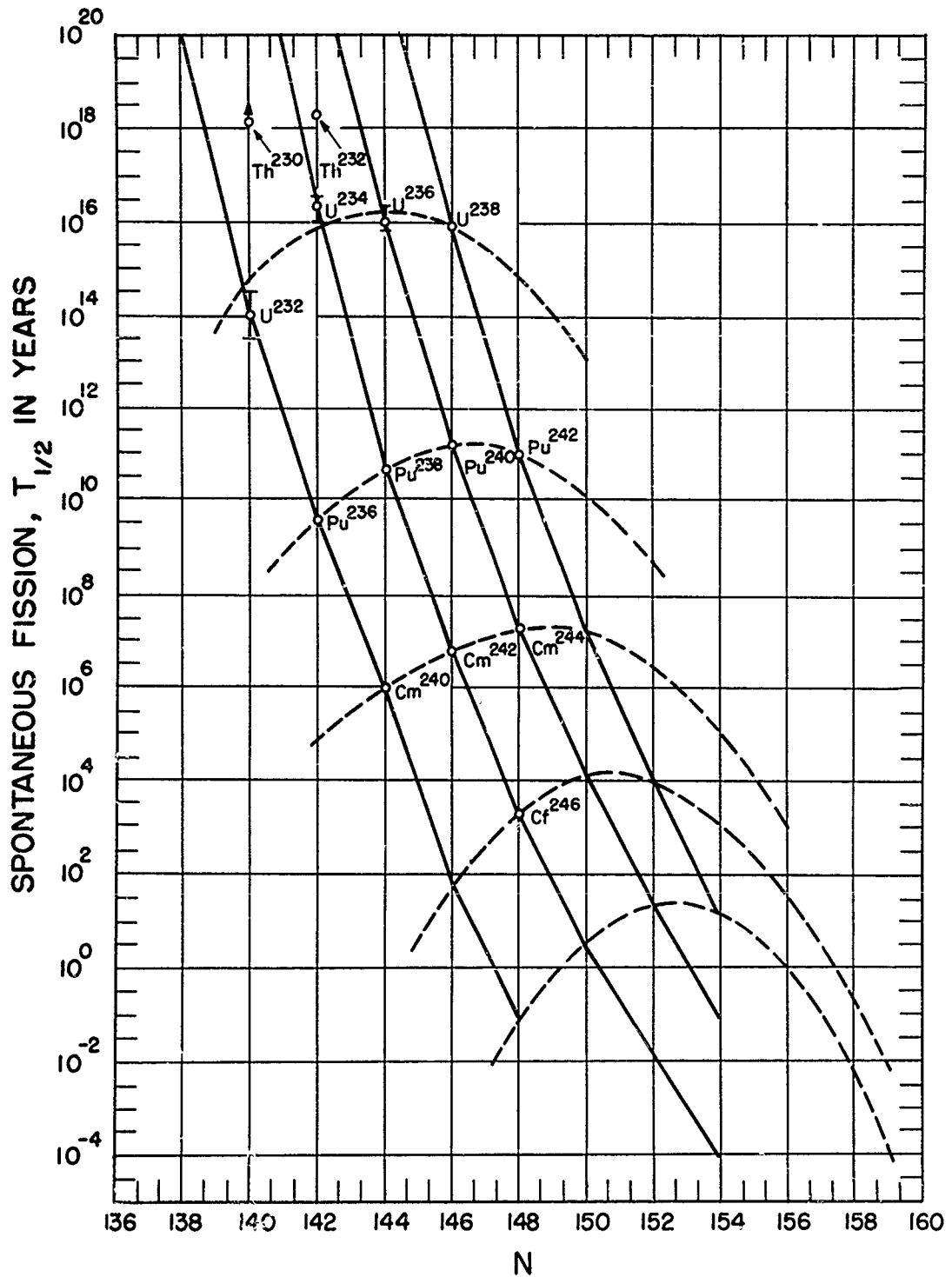


Fig. 9.3—UCRL spontaneous-fission systematics (II).

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of the sample was found to be: 238 - 1.5%, 239 - 79.9%, 240 - 4.2%, 241 - 0.5% and 242 - 13.9%. The spontaneous fission rate of a sample of this material was found to be about three times as high as the combined effect calculated for the Pu^{238} and Pu^{240} present. This information leads to a spontaneous fission half-life for Pu^{242} of about 8×10^{10} years. A previous measurement at the Argonne National Laboratory gives approximately the same value.

"Preliminary measurements on a sample of Cf^{248} produced in the Berkeley 60" Crocker Laboratory cyclotron by helium ion bombardment of curium containing small amounts of Cm^{245} indicate a spontaneous fission half-life for Cf^{248} of roughly 10^4 years.⁴ Since the fission rate of this sample is very low (about 1 fission in five days) no great confidence can yet be placed in this value.

"Some of the new heavy isotopes show relatively high spontaneous fission rates and are useful in extending the systematics of spontaneous fission. Two of the many different ways of plotting the data are illustrated in Figs. 9.2 and 9.3 and are largely self-explanatory. However, the final results of this phase of the investigation depend on better mass assignments and more spontaneous fission data."

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CHAPTER 10

IDENTIFICATION OF NEW ELEMENTS

The position of elution peaks is the critical factor in the chemical identification of new actinide elements. The comments of the UCRL group on this point are as follows:

"The elements immediately beyond californium are expected to continue the sequence of other tri-positive actinide elements with respect to ionic radius so that this property may be applied in identifying these elements by the order of elution from ion exchange resins. The same principle was applied in identifying elements 97 (berkelium)⁷ and 98 (californium)⁸. In the latter case it was shown that the relative spacing of the elution positions from ion exchange resins (due to change in ionic radius) between the members of the actinide series, californium, berkelium and curium was very similar to the corresponding spacing between the rare earth homologues dysprosium, terbium and gadolinium. This principle is merely extended in identifying the two new elements observed in this work. Since the elution position of MIKE produced californium has been proved by the addition of Cf^{246} tracer, the two elution peaks preceding californium are proved to be due to trans-californium elements. That these peaks correspond to elements 99 and 100 is best shown by comparison with an elution diagram (Fig. 10.1) showing the relative spacing between the corresponding rare earths. The actinide elution curve is a duplication of Fig. 8.18 with the heights of the peaks normalized to facilitate comparison. The rare earth elution curve is from an actual separation which was not run under the same conditions (not on standard hot citrate column) and is only used to compare the relative spacing. Previous work in this laboratory has indicated the relative spacing is essentially independent of the particular conditions of operating the columns although the sharpness of separation and the overall spacing is greatly affected. The rare earth elution curve is also normalized for comparison.

"Aside from minor variations in relative spacing between the positions of rare earths and corresponding actinides, it will be seen that the elution position of the 6.6 Mev alpha particles, indeed, corresponds to element 99 and that the position of the 7.1 Mev alpha particles corresponds to element 100."

Of considerable additional aid are the relations known as the alpha systematics. A portion of these relations is shown in Fig. 10.2, and the comments of the UCRL group on possible explanations of the data of this report as they affect these systematics are as follows:

"There are some possible difficulties and inconsistencies in the preceding interpretation of results, particularly with regard to mass assignments, which can be remedied by the assumption of a minor magic number at 152 neutrons. Figure 10.2 is a plot of alpha particle energy versus mass number; the dotted lines illustrate what might occur if 152 neutrons is an unusually stable configuration. In the absence of closed shell effects and assuming the mass assignment of 99^{253} is correct, it will be noted that there is an unusually large increase in alpha energy between the californium isotopes and those of element 99; however, if 152 neutrons is taken as an especially stable combination the lighter isotopes of 99 and 100 would have normal alpha energies and those of 154 neutrons or more would have abnormally high energies. If this hypothesis were correct, the 6.1 Mev californium activity could be assigned to mass 252 rather than to mass 249. This assumption would also tend to increase the beta instability of Cm^{250} and increase the beta stability of Bk^{248} . A magic number of 154 neutrons would give similar results with mass assignments shifted still further to the higher mass numbers."

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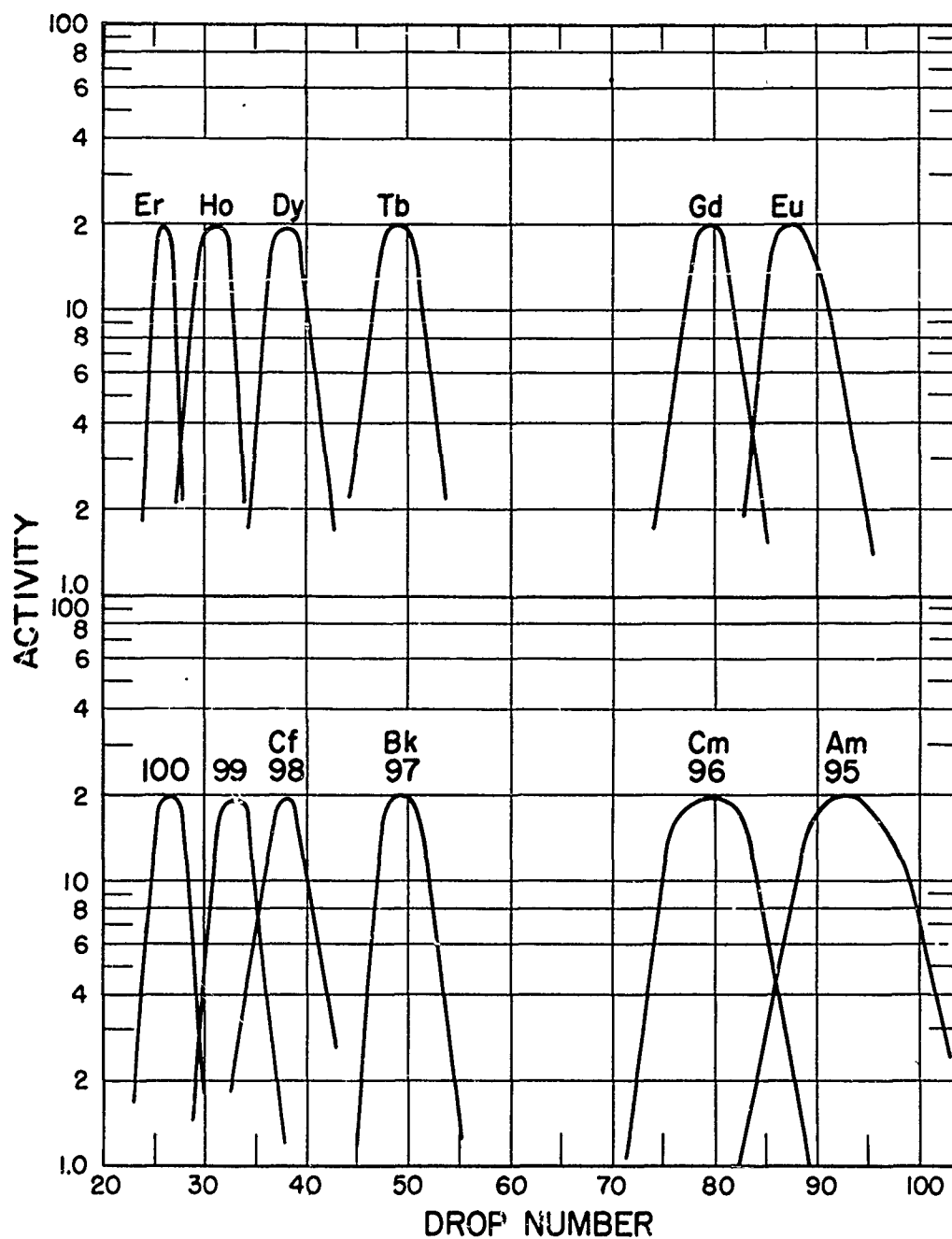
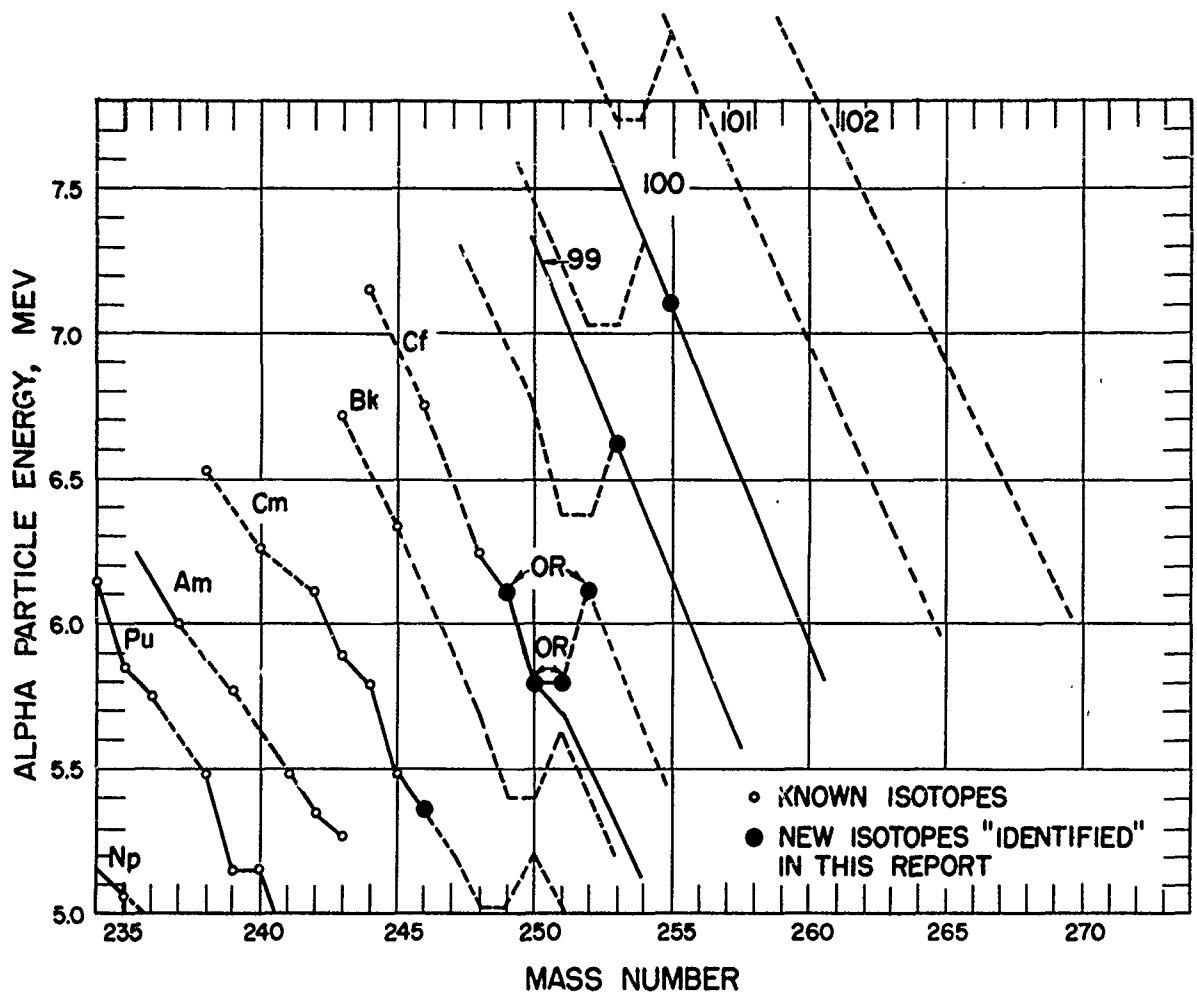


Fig. 10.1—UCRL comparison of rates of elution of actinides with rare-earth homologues.

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CHAPTER 11

SUMMARY

The investigations reported here have shown that the extreme neutron flux present at the detonation of the Ivy Mike bomb resulted in the production of a number of new heavy nuclides, including isotopes of at least two new elements (Fig. 11.1). The nuclear characteristics and tentative mass-number assignments of these nuclides are presented in the following tabulation:

Nuclide	Radiations	Half-life	Remarks
Pu ²⁴⁴	Probably beta stable	Beta half-life greater than 1000 years	
Pu ²⁴⁶	0.15 Mev beta; 0.043, 0.103, 0.175, 0.220 Mev gamma	11.2 days	
Am ²⁴⁵	Beta	<2 days	Limit set only
Am ²⁴⁶	1.22 Mev beta; 0.0185, 0.103, 0.795, 1.069 Mev gamma	25.0 min	Decay energy, 2.29 Mev
Cm ²⁴⁶	5.48 Mev alpha	1.3 × 10 ⁵ years	Half-life discrepancy with cyclotron-produced material
Cm ²⁴⁶	5.36 Mev alpha	Alpha, 2000 years; spontaneous fission, 3 × 10 ⁷ years	
Cm ²⁴⁷			Beta stable or very long beta lived
Bk ²⁵⁰⁻²⁵¹	Beta	>1 month	
Cf ²⁵⁰⁻²⁵¹	5.8 Mev alpha	~400 years	
Cf ²⁴⁹ or ²⁵²	6.1 Mev alpha	>5 months	
Cf ²⁵³	Beta (particles not observed)	~1 month	UCRL interpretation
Cf ²⁵⁴ or ²⁵⁶		Spontaneous-fission half-life, 68 days	No detectable alpha
99 ²⁵³	6.6 Mev alpha	35 to 50 days	ANL interpretation
99 ²⁵³	6.6 Mev alpha	22 days	UCRL interpretation (Cf ²⁵³ → 99 ²⁵³)

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Nuclide	Radiations	Half-life	Remarks
99^{255}	Beta (particles not observed)	~1 month	UCRL interpretation
100^{255}	6.6 Mev alpha	15 days	ANL interpretation
100^{255}	7.1 Mev alpha	16 hr	UCRL interpretation
100	6.35 Mev alpha		ANL data

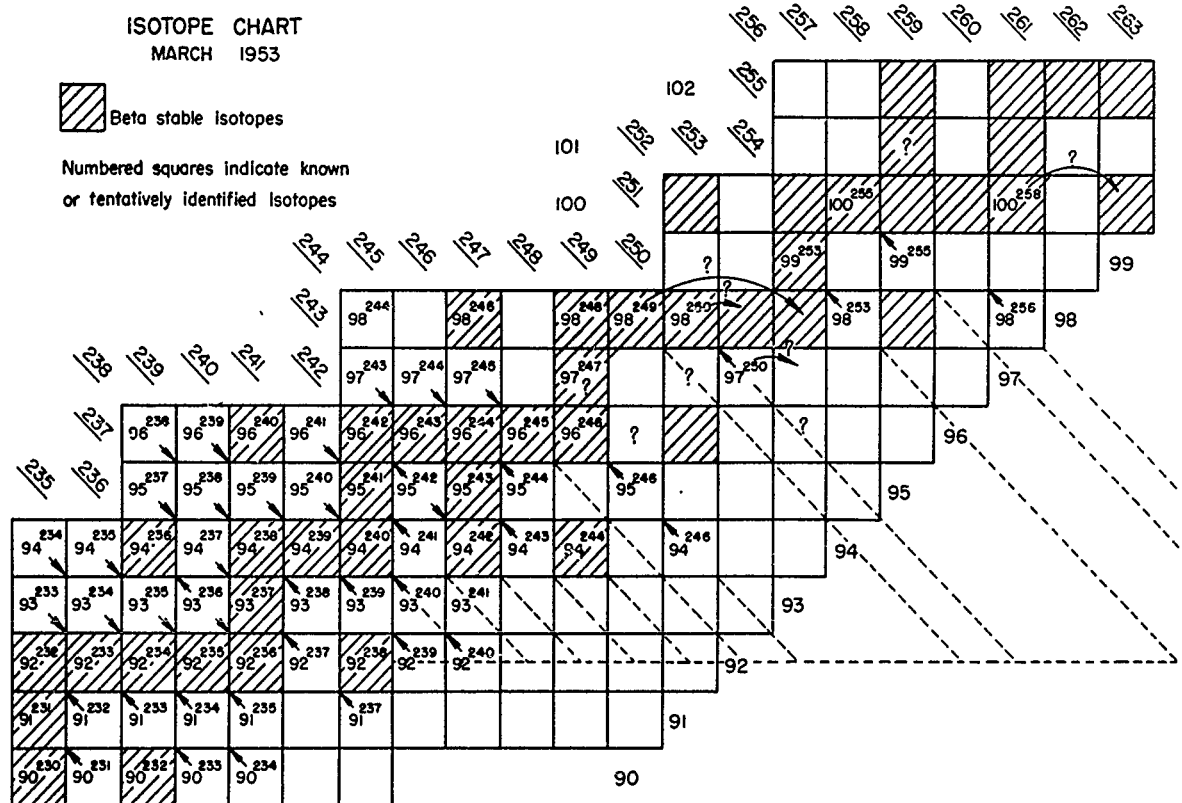


Fig. 11.1—UCRL isotope chart of heavy nuclides.

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