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This report is a detailed review of published information on the isotopes of the transuranium elements.* The discussion of each element begins with a description of the experiments leading to the first synthesis and identification of an isotope of that element. Following this, each isotopes is discussed separately. The discovery of the isotope, the alternate methods for its synthesis, and the radiations emitted by it are discussed. In those cases where it seems warranted, a decay scheme is presented and interpreted according to current nuclear models, chiefly the unified model of Bohr and Mottelson. No attempt is made to review the interesting chemical properties of the transuranium elements.

This report has been prepared as part of a general review of the nuclear properties of the heavy elements being prepared under the joint authorship of E. K. Hyde, G. T. Seaborg and I. Perlman. The general review will appear at a later date. This review is being issued in the belief that it will prove useful in its present form and in the hope that those readers who detect errors or who have newer and better data on the matters discussed herein will help the author in the preparation of an improved edition in the future.

* The literature was reviewed up to November of 1960.

THE RADIOACTIVE DECAY OF THE ISOTOPES OF THE TRANSURANIUM ELEMENTS

Earl K. Hyde

9.1 THE ELEMENT NEPTUNIUM (ELEMENT 93)

9.1.1 The Discovery and Early History of Neptunium. The first attempts to prepare an isotope of neptunium were made in the 1930's by the bombardment of uranium with neutrons. This was a logical approach because experiments with many other elements had shown that an isotope produced by addition of a neutron to a stable target isotope was usually unstable and underwent beta decay to form an isotope of the next heavier element. We now know that when natural uranium is irradiated with slowed neutrons this expected capture process and subsequent beta-decay does occur with the predominant U^{238} isotope. During the neutron irradiation, however, the rare uranium isotope, U^{235}, undergoes nuclear fission resulting in the production of numerous radioactive fission product isotopes. This was not known at the time of the first attempts to prepare element 93 by the neutron capture method and of course caused considerable confusion. Another fact leading to confusion was that the element neptunium was not correctly placed in the Periodic System of the Elements so that the properties expected for element 93 were incorrect. Most experimentalists at the time thought that element 93 should resemble rhenium, and the radiochemical methods for isolation of element 93 were based on this false supposition. With the dual difficulties of an ignorance of the nuclear fission reaction and of the chemical properties of element 93 some mistaken conclusions were reached in the period before the discovery of fission. As early as 1934, FERMI[1] and his co-workers at Rome had isolated a 13-minute activity from a uranium sample after neutron irradiation and had separated it chemically from all elements of atomic number 82 to 92. This led them to the logical conclusion that this 13-minute activity was an isotope of element 93 particularly since it seemed to have chemical properties resembling rhenium. Additional experiments by the Italian investigators and by experimentalists in other countries resulted in the discovery of many additional activities which appeared to be isotopes of transuranium elements. Some of these were believed to have atomic numbers as large as 96. In addition to the transuranium element isotopes other activities were found which had the properties of thorium, protactinium, actinium and other elements of smaller atomic number than the target uranium. It was difficult to understand why such

a variety of products should come from the irradiation of uranium with neutrons when nothing of this nature had been observed with lighter element targets.[2] At this point HAHN AND STRASSMANN[3] performed their beautiful radiochemical experiments which proved beyond question that uranium underwent a unique nuclear reaction in which the nucleus was split to form radioactive isotopes of medium weight elements. It became evident at once that all the previous reports of trans-uranium elements had to be reinterpreted and the search for element 93 had to be begun once more.

Following the announcement of HAHN AND STRASSMANN'S[3] discovery of fission in 1939, McMILLAN[4] at the University of California carried out a study of the range of the radioactive fission products collected from a thin foil of uranium oxide irradiated with neutrons. The neutrons were produced by the reaction of 16 Mev deuterons with a beryllium target. One striking fact to emerge was that the recoil range of a 2.3-day activity was much less than that of the other activities produced. Furthermore, this activity was in much greater abundance than any activity of comparable half-life. This non-recoiling behavior would be expected of a heavy element isotope as distinguished from a fission product and proof was sought that the 2.3-day radioactivity was an isotope of a trans-uranium element. SEGRE'S[5] initial studies seemed to indicate that this was not the case and that the 2.3-day period was due to a heavy rare earth. McMILLAN AND ABELSON[6] teamed up to carry the investigation further. When the uranium was wrapped in cadmium to remove neutrons of thermal energy the fission product activity was greatly reduced but the 2.3 day period and a 23 minute period which had previously been identified by MEITNER, STRASSMANN AND HAHN[7] as a uranium isotope were only slightly reduced. Chemical studies showed a characteristic difference from the rare earths; namely, that the substance did not coprecipitate with insoluble fluorides in the presence of a strong oxidizing agent in strong acid. In the presence of a strong reducing agent it precipitated quantitatively with rare earth elements upon the addition of HF.

McMILLAN AND ABELSON then conclusively proved the genetic relationship of the 2.3-day activity to the 23-minute uranium beta emitter and hence established its identity as an isotope of element 93.

$$U^{238}(n,\gamma)\,U^{239};\ U^{239} \xrightarrow[23\ \text{min.}]{\beta^-} Np^{239} \xrightarrow[2.3\ \text{days}]{\beta^-}$$

Further chemical experiments showed a distinct similarity of the new element to hexapositive uranium in the oxidized state and to Th^{+4} and U^{+4} in the reduced state. There was no resemblance to rhenium, contrary to the expectations of most experimenters seeking at this time to identify element 93. McMILLAN AND ABELSON suggested that neptunium was to be considered a member of a second "rare earth" group of elements starting in this region of the periodic chart, probably with the element uranium. McMILLAN AND ABELSON originated the lanthanum fluoride oxidation-reduction cycle (coprecipitation of neptunium on an insoluble fluoride compound precipitated from reducing solution - non-coprecipitation of neptunium on an insoluble fluoride compound precipitated under aqueous oxidizing conditions.) which was to be a mainstay of neptunium purification for many years to come.

McMILLAN AND ABELSON had earned the right to name element 93 and chose the name neptunium from the planet Neptune which is the first planet beyond Uranus in our solar system. The symbol Np was suggested somewhat later.

STARKE[8] apparently made an independent discovery of Np^{239} at about the same time.

A second isotope of neptunium, the 2.0-day beta-emitter Np^{238}, was discovered later in 1940 by SEABORG, McMILLAN, KENNEDY AND WAHL[9] at the University of California. This isotope was produced by the bombardment of natural uranium with 16 Mev deuterons by the reactions:

$$U^{238}(d,2n)\,Np^{238};\ Np^{238} \xrightarrow[2.0\ \text{days}]{\beta^-} Pu^{238}$$

The beta decay of Np^{238} produces Pu^{238} which is an alpha emitter of some 90 years half life. The isolation of this long-lived daughter of Np^{238} by the above research team constituted the discovery of element 94.

Much of the initial tracer work on the chemistry of neptunium was performed with Np^{239} prepared by neutron irradiation of uranium or with the Np^{239} - Np^{238} mixture produced by the deuteron bombardment of uranium.

The longest-lived isotope of neptunium and the isotope which must be considered the most important from the standpoint of chemical investigations

of neptunium is Np^{237} which has a half life of 2.20×10^6 years. Its discovery in 1942 was an outgrowth of the study of U^{237}. In 1940, NISHINA AND COWORKERS[10] found a 6.5 day uranium period when uranium oxide was bombarded with fast neutrons from a cyclotron. This activity remained with the uranium when the fission product activities were removed. They assigned the new activity correctly to U^{237} produced by the reaction:

$$U^{238} (n,2n) U^{237}.$$

Since U^{237} is a beta emitter, it must decay into 93^{237} and NISHINA AND COWORKERS[10] attempted to isolate the new element. They were unsuccessful because of the weakness of their sources and because they assumed chemical properties for neptunium similar to rhenium.

Independently, at the same time, McMILLAN[11] had also studied the new isotope U^{237}. He too looked for the 93^{237} daughter using the fluroide oxidation-reduction coprecipitation cycle which he and Abelson had developed for their studies of Np^{239}. Using U^{237} sources of 80 microcurie strength they were unable to detect any radiations due to Np^{237} because of the long half-life of Np^{237}.

In 1942, WAHL AND SEABORG[12] in a continuation of the experiments of McMILLAN prepared a much larger source of U^{237} (97 Millicuries) by irradiating 1,200 grams of uranyl nitrate with the fast neutrons produced in a 15,000 microampere hour bombardment of beryllium with 16 Mev deuterons. Using ether extractions to remove the bulk of the uranium and numerous rare earth fluoride coprecipitation cycles to remove and purify the neptunium fraction these workers succeeded in isolating and identifying 300 alpha counts per minute of Np^{237}. Using the ratio of U^{237} to the daughter Np^{237} activity they calculated a half-life of 3×10^6 years which agrees within their experimental error with the presently accepted value of 2.20×10^6 years.

The principal source of Np^{237} is the chain reacting uranium pile. When natural uranium or uranium of only moderate enrichment is used the Np^{237} is produced almost entirely by the U^{238} (n,2n) reaction for which the cross-section is appreciable. In the original Hanford reactors, for example, the amount of Np^{237} produced was approximately 0.3 percent of the amount of Pu^{239} produced. Therefore, neptunium can be made available in kilogram quantities by suitable modification of plutonium chemical processing methods. This is fortunate for

it means that this element, whose complex chemical and physical properties are of such great scientific interest, can be subjected to careful study in many laboratories. In reactors operating on uranium highly enriched in U^{235} the principal reaction path giving rise to Np^{237} is the following reaction sequence which was first studied by MANNING AND BRITTAIN[13].

$$U^{235}\ (n,\gamma)\ U^{236}\ (n,\gamma)\ U^{237} \xrightarrow[6.7\ \text{days}]{\beta^-} Np^{237}.$$

The first isolation of a weighable amount of Np^{237} was carried out by MAGNUSSON AND LA CHAPELLE[14] in October 1944. These workers purified neptunium by the rare-earth fluoride coprecipitation technique and then eliminated the rare earth carrier by precipitating pure neptunium compounds such as sodium neptunyl acetate. Since they had only a few micrograms of material available to them these precipitations were carried out with ultramicro-chemical techniques. The half life was measured by determining the counting rates of accurately weighed samples of NpO_2 whose identity and purity were checked by x-ray crystallographic analysis. The weight of one of the NpO_2 samples was only 3.8 micrograms. These experiments constituted the first chemical isolation of a weighable amount of neptunium.

The isotope Np^{237} is beta-stable, i.e., is stable towards decay to its neighboring isobars either by negative beta-particle emission or by orbital electron capture. It is the only beta-stable isotope of neptunium, a fact which agrees with the general rule that all the odd elements have at most, one or two beta-stable isotopes. An isotope such as Np^{237}, which is unstable only because of its alpha-particle lability, is therefore analogous to the "stable" nuclides in the region below lead.

Identification of further isotopes of neptunium was not feasible until target materials other than U^{238} were available or cyclotron beam energies were increased beyond the 16 Mev deuteron maximum prevailing in 1942. By 1944 milligram quantities of U^{233} and U^{235} became available for cyclotron bombardments, and JAMES, FLORIN, HOPKINS AND GHIORSO[15] identified Np^{236}, Np^{235} and Np^{234} as products of deuteron bombardments of U^{235}. The isotope Np^{234} was also made[16] by deuteron bombardment of U^{233}. A few years later the Berkeley synchrocyclotron went into operation. This greatly extended the complexity of spallation reactions

which could be carried out. MAGNUSSON, THOMPSON AND SEABORG[17] took advantage of the new beam energy range to produce and identify Np^{233}, Np^{232} and Np^{231} by (d,xn) type reactions using U^{233}, U^{235} and U^{238} targets. At the other end of the mass range new studies have been aided by the high neutron fluxes available in modern nuclear reactors. The isotope Np^{240} can be prepared by successive capture of two neutrons in U^{238} as was first shown by HYDE, STUDIER AND MANNING.[18]

$$U^{238}\ (n,\gamma)\ U^{239}\ (n,\gamma)\ U^{240} \xrightarrow[14\ \text{hours}]{\beta^-} Np^{240}.$$

The isotope Np^{241} was first identified by LESSLER AND MICHEL[19] who prepared it by the reaction

$$U^{238}\ (\alpha,p)\ Np^{241}$$

In the sections which follow we discuss the properties of the isotopes of neptunium in a systematic fashion. The isotopes of neptunium follow the normal pattern with regard to beta instability; those with mass greater than 237 are unstable toward negative beta particle emission while those with smaller mass numbers are unstable toward decay by orbital electron capture. The isotopes Np^{238} and Np^{236} exhibit both types of beta-instability; this is the expected behavior for odd-odd nuclei near the bottom of the Bohr-Wheeler parabolas. All neptunium isotopes are alpha unstable but the alpha activity of Np^{238} and higher mass isotopes has not been detected because negative beta particle emission is so much more rapid. Similarly for Np^{236} and lighter-mass isotopes the rate of decay by capture of an orbital electron is so fast that alpha-particle emission is often difficult to detect. For isotopes which are extremely deficient in neutrons such as Np^{231}, the alpha decay-energy becomes so large and the partial alpha half-life so short that alpha-particle emission again becomes prominent.

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18. E. K. Hyde, M. H. Studier and W.M. Manning, Argonne National Laboratory Report, ANL-4143, April 15, 1948 and ANL-4182, August 4, 1948 (unpublished.)
19. R. M. Lessler and M. C. Michel, University of California Radiation Laboratory Report, UCRL-8757, August 1959.

9.1.2 Neptunium-231. MAGNUSSON, THOMPSON AND SEABORG[1] produced Np^{231} by bombardment of natural uranium with 100 Mev deuterons or of enriched U^{235} with 45-100 Mev deuterons.

$$U^{238}\ (d,9n)\ Np^{231}$$

$$U^{235}\ (d,6n)\ Np^{231}$$

This isotope decays principally by electron capture to U^{231} with a half life of about 50 minutes although the radiations corresponding to this decay cannot be observed directly because of interference from the electromagnetic radiation of the several other neptunium isotopes produced in the bombardment. It decays also by the emission of 6.28 Mev alpha particles. These alpha particles can be readily distinguished by measurements in an ion chamber because the alpha branching of the heavier neptunium isotopes present in the sample is so slight. The mass identification depends on the observation of the growth of Pa^{227} and of the Pa^{227} daughters comprising the Pa^{227} collateral series. (See section 7.2.3) The alpha branching is probably about one percent.

Reference - Np^{231}

1. L. B. Magnusson, S. G. Thompson and G. T. Seaborg, Phys. Rev. 78, 363 (1950).

9.1.3 Neptunium- 232 . In the experiments of MAGNUSSON, THOMPSON AND SEABORG cited above, preliminary evidence for Np^{232} was found indicating a half life of 13 minutes for decay by electron capture. Further confirmatory experiments are necessary; one essential experiment is the production of detectable yields of U^{232} daughter activity. No alpha radioactivity was observed. This is in agreement with alpha systematics which would predict that the degree of

alpha branching should be smaller than could have been detected in experiments described.

9.1.4 Neptunium-233. MAGNUSSON, THOMPSON AND SEABORG[1] produced 35 minute Np^{233} by bombardment of U^{235} targets with deuterons of 45-100 Mev energy. In the case of U^{233} targets the yield of Np^{233} was still detectable when the deuteron energy was lowered to as low as 15 Mev. The reactions involved are:

$$U^{235}\ (d,4n)\ Np^{233}$$

$$U^{233}\ (d,2n)\ Np^{233}$$

The decay is $\geq$ 99 percent by electron capture. Absorption curve measurements revealed the presence of K x-radiation and some gamma radiation and conversion electrons. It was possible to look for alpha particles emitted by Np^{233} in samples prepared by the bombardment of U^{233} with 15 Mev deuterons because at this beam energy no Np^{231} alpha activity was produced. Alpha particles of 5.53 Mev energy were observed in amounts corresponding to a branching ratio of only 10^{-5}.

LESSLER[2] prepared Np^{233} by the reaction:

$$U^{234}\ (d,3n)\ Np^{233},$$

and studied the gamma radiations in a NaI spectrometer. Peaks were observed at 95, 150, 170, 205 and 410 Kev. Broad peaks indicating two or more unresolved gamma rays were observed from 230-310 Kev and from 500 to 560 Kev. The most prominent photons were K x-rays. Other gamma rays are probably present, but the presence of some Np^{234} activity interfered with their measurement. No detailed decay scheme has been proposed for Np^{233}.

LESSLER set an upper limit of 3×10^{-5} for the α-decay branching.

Reference - Np^{233}

1. L. B. Magnusson, S. G. Thompson and G. T. Seaborg, Phys. Rev. 78, 363 (1950).
2. R. M. Lessler, University of California Radiation Laboratory Report, UCRL-8439, October 1958.

9.1.5 Neptunium-234. The 4.40 day isotope Np^{234} was discovered by JAMES, FLORIN, HOPKINS AND GHIORSO[1] who found that the neptunium fraction chemically isolated from a U^{235} target bombarded with 20 Mev deuterons contained in addition to Np^{238} and Np^{239} (from the U^{238} present) an activity decaying with a half life of about 4 days. This activity was characterized by soft electrons, x-rays and energetic gamma radiation. Shortly afterward HYDE, STUDIER AND GHIORSO[2] found that Np^{234} could be prepared in a purer state by bombarding U^{233} with 20 Mev deuterons. These authors reported high energy gamma radiation in high abundance. An upper limit of 0.01 percent was set on the alpha branching of Np^{234} by the failure to isolate Pa^{230} from the neptunium fraction. OSBORNE, THOMPSON AND VAN WINKLE[3] prepared Np^{234} by bombarding Pa^{231} with 40 Mev helium ions.

The reaction by which Np^{234} has been formed include:

U^{236} (d,4n) Np^{234}
U^{235} (d,3n) Np^{234}
U^{235} (α,p4n) Np^{234}
Pa^{231} (α,n) Np^{234}
U^{234} (d,2n) Np^{234}
U^{233} (d,n) Np^{234}
U^{233} (α,p2n) Np^{234}
U^{233} (α,3n) Pu^{234} $\xrightarrow{EC}$ Np^{234}
U^{235} (p,2n) Np^{234}

The cross sections for several of these reactions are given in Table 5.5 of Chapter 5.

The various modes of formation limited the mass number assignment to 234 or 233. The assignment 234 was confirmed by the fissionability measurements of HYDE, BENTLEY AND HAGEMANN[4] who found a slow neutron fission cross section of 900 barns for Np^{234}. The fissionability of the sample decreased with a half life of 4.4 days and did not level out as would have been expected if the highly fissionable U^{233} had been the daughter product. In the case of U^{233} bombarded with helium ions the formation of Np^{234} by the electron capture decay of Pu^{234} has been directly observed.[5] This bombardment yields a plutonium activity of approximately 8 hour half life that is known to be Pu^{234} which decays

principally (~96%) by electron capture. The genetic relationship between the 8-hour plutonium and the 4-day neptunium has been demonstrated.

A number of workers[6-10], have contributed to the measurement of the gamma ray photons, the x-rays and the conversion electrons emitted by Np^{234}. We quote here chiefly from the study of GALLAGHER AND THOMAS[12] which is considerably more comprehensive than those published earlier. Np^{234} emits a large number of gamma rays ranging in energy from 43 to 1606 Kev. Measurements by the NaI scintillation method do not resolve these gamma rays cleanly. The most fruitful approach has been the measurement of the conversion electrons in permanent magnet spectrographs of high resolution and in electromagnetic beta spectrometers of moderate to high resolution.

GALLAGHER AND THOMAS[12] measured more than 150 conversion lines and deduced the gamma rays listed in the table from the energies and relative intensities of these lines. In addition, a number of plutonium auger electrons were observed as well as a few lines which could not be definitely assigned to a gamma transition.

GALLAGHER AND THOMAS[12] constructed the decay scheme shown in the figure. In constructing this figure, they were able to use information on the low-lying levels obtained from the α-decay of Pu^{238} (See Section 9.2.8). They relied partially also on sum and difference relationships between their observed gamma rays and partially on some gamma-gamma coincidence data reported by HUIZENGA AND CO-WORKERS.[9] In many respects this decay scheme is incomplete and tentative. The spin of Np^{234} is unknown (although restricted to the values 0, 1, or 2) and the primary electron-capture branching to the levels of U^{234} is also unknown. The spin and parity assignments of several of the highest-lying levels are uncertain. Nonetheless this decay scheme contains a great deal of information on the level structure of U^{234} and it is worthwhile calling attention to several interesting points.

The lowest levels constitute a rotational band based on the 0^+ ground state. The spin sequence is 0+, 2+, 4+ ...., the energy spacing follows the I (I+1) rule, and the pattern of de-excitation is a cascade of E2 transitions. In the decay scheme of Pu^{238} which is shown in section 9.2.8 the 6+ and 8+ members of this rotational band of levels are also observed. Furthermore, the 4+ and 6+ levels are seen in the beta decay of Pa^{234} (UZ).

Table 1

Transitions in U^{234} following Np^{234} decay as reported by Gallagher and Thomas*. Transition assignments are based on internal conversion data. The isotope assignment is based on the decay of the lines observed in a series of five exposures at 4.4-day intervals over an approximately three week period. Visual intensities listed are vvs = very very strong, vs = very strong, s = strong, ms = moderately strong, m = moderate, mw = moderately weak, w = weak, vw = very weak, vvw = very very weak, ew = extremely weak, ew? = extremely weak and questionable, -d = diffuse.

Assignment	Energy of gamma (kev)	Subshell Conversion										Multipolarity
		K	L_I	L_{II}	L_{III}	M_I	M_{II}	M_{III}	N_I	N_{II}	N_{III}	
BA	43.49 ± .05			mw	m		m	ms		w	vw	E2
CB	99.7 ± .1			vw	ew?		ew?					E2
--	233.6 ± .2	ew?	ew									
MK	234.6 ± .2	vvw	w[f]			ew?			ew?			
IF	238.6 ± .4	ew?										
KI	247.9 ± .2	vvw-d	ew			ew						
NK	265.8 ± .5	ew?										
--	297.6 ± .5	ew?										
JD	450.5 ± .5	ms	vw	ew	ew?	ew?						
MI	482.8 ± 1[g]	ew?										
MH	485.1 ± 1[g]	ew?										
NH	515.7 ± .5	mw	ew									
MG	525.9 ± .5	ew-d										
NG	556.8 ± .6	m	ew-d									
DB	744.1 ± .7	w										
NF	751.7 ± .8	w	ew?									
EB	768.0 ± .8	ew-d	ew?									
DA	787.8 ± .8	vvw										
NE	793.8 ± .8	vw										
FB	810.0 ± .8	s	mw			ew						(EO)
EA	811.6 ± .8	vvs	ms			w			ew-d			EO
FA	853.6 ± 1.[3]	ew?										
--	1003 ±2	vvw	ew?									
--	1105 ±2	ew?										
JB	1196 ±2	ew										

Table (cont'd)

Assignment	Energy of gamma (kev)	K	L_I	L_{II}	L_{III}	M_I	M_{II}	M_{III}	N_I	N_{II}	N_{III}	Multipolarity
JA	1240 ±2	ew??										
LB	1395 ±3	ew??										
LA	1439 ±3	ew										
MB	1531 ±3	vw										
NB	1562 ±3	m	vvw			ew						
MA	1575 ±3	vw	ew									
NA	1606 ±3	w	ew									

Capital letters refer to levels in the decay scheme.

* Electron lines observed on a series of permanent magnet spectrographs with fields of 99, 160 and 350 gauss.

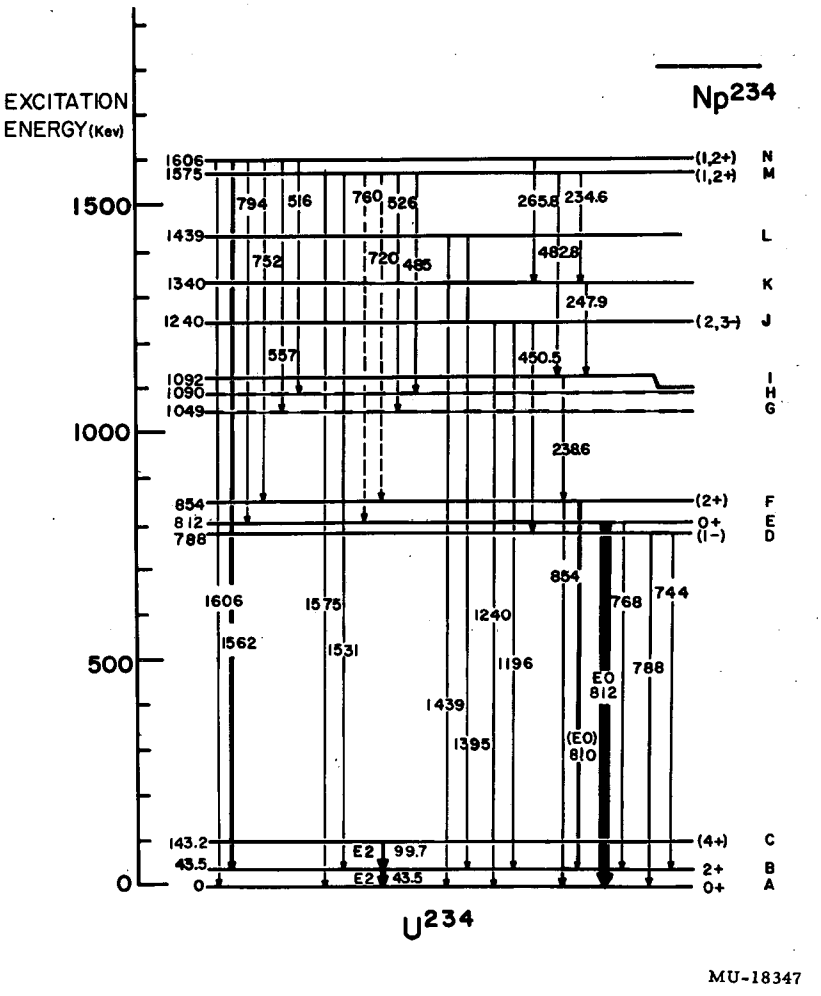

Figure 1. Decay scheme of the electron-capture decay of Np^{234} as formulated by Gallagher and Thomas.

The 1- level at 788 is the first 1- level to be reported in an even-even nucleus of uranium but it is a commonly observed level in the spectra of even-even nuclei of neighboring elements (see discussion in chapter 3). It is assigned to a collective mode of excitation. This level de-excites by a pair of E1 transitions to the 0+ and 2+ levels of the ground state rotational band. These gamma rays are also seen in the beta decay of Pa^{234} (UX_2); See discussion in section 8.2.12.

The level at 812 kev is given the spin assignment 0+ and is believed with some confidence to be the first β-vibrational level of excitation in U^{234}. (See chapter 3 for a discussion of collective β and γ vibrations in even-even nuclei). The level at 854 is believed to be the first rotational level in a band based on the 812 kev state. This interpretation for both levels is strongly based on the observance of the completely converted E0 transitions which de-excite them. The observation of these E0 transitions is one of the most interesting results of the study of the decay of Np^{234}. Further information on these particular levels and on the E0 transitions deexciting them comes from the work of DURHAM, REST ER AND CLASS[14] who excited the level at 854 kev by bombardment of U^{234} with 5 Mev protons.

E0 transitions proceed completely by emission of conversion electrons. Hence the dominant feature of the conversion elelctron spectrum of Np^{234} is the K and L electron lines of these prominent E0 transitions. The transition from the 0+ level to the ground state has also been identified in the α-decay of Pu^{238} and in the β-decay of Pa^{234}, (see sections 9.2.8 and 8.0.0, respectively). This particular transition connects an 0+ state with an 0+ state and is pure E0. The other E0 connecting the 2+ level at 854 kev with the 2+ level at 43.5 kev competes with E2 de-excitation. This was the first observation in the heavy element region of an E0-admixed transition between two states with $I \neq 0$. CHURCH AND WENESER[13] had predicted such transitions on theoretical grounds.

GALLAGHER AND THOMAS[12] also suggested that the levels at 1049 and 1090 (G and H) may be the expected 2+ and 3+ (K = 2) states corresponding to collective excitation of the first γ-vibrational mode. This interpretation is probably incorrect in view of the later studies of BJØRNHOLM AND NIELSEN[15] on the decay of Pa^{234} (UZ) which indicates that the γ-vibrational level falls at 922 kev. Refer to the discussion in section 8.2.12 of Chapter 8,

PRESTWOOD AND CO-WORKERS[10] have investigated the radiations of Np^{234} with a trochoidal analyzer and have identified positrons with an end point energy of 0.8 Mev. The ratio of positron emission to electron capture was found to be 4.6×10^{-4}. This is the first observation of the emission of positrons by a nuclide of atomic number higher than 80. This measurement set a lower limit of 1.8 Mev to the disintegration energy.

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9.1.6 Neptunium-235. JAMES, FLORIN, HOPKINS AND GHIORSO[1] found that the neptunium fraction from uranium bombarded with 20 Mev deuterons, after thorough decontamination, exhibits a long-lived activity characterized by low-energy electromagnetic radiation such as would be emitted by an electron-capturing species. Bombardments of uranium targets of varying isotopic composition have established the responsible target isotope as U^{235}. It is now certain that this activity is due to Np^{235}, produced by the reaction U^{235} (d,2n) Np^{235}.

Bombardment of U^{235} [1] and U^{233} [2] with 40 Mev helium ions also produces Np^{235} by the reactions:

$$U^{235}(\alpha,p3n)\,Np^{235}$$

$$U^{235}(\alpha,4n)\,Pu^{235} \xrightarrow{\text{electron capture}} Np^{235}$$

$$U^{233}(\alpha,pn)\,Np^{235}$$

$$U^{233}(\alpha,2n)\,Pu^{235} \xrightarrow{\text{electron capture}} Np^{235}$$

Failure to find long-lived neptunium activity from the bombardment of U^{233} with deuterons[2] is confirmation of the isotopic assignment of the activity as Np^{235}.

The best estimate of the half life,[3] obtained from the decay of several samples over a period of 2 years, is 410 ± 10 days; these particular samples were prepared by the bombardment of 95 percent U^{235} with 20 Mev deuterons. The observed radiations consist chiefly of the characteristic L x-rays of uranium.[3]

The total disintegration energy is low and the ratio of L-electron to K-electron capture is large. JAMES, GHIORSO AND ORTH[3] set the ratio at approximately 10. Later HOFF, OLSEN AND MANN[4] reported the value 30 ± 2. The most careful measurement was made by GINDLER, HUIZENGA AND ENGELKEMEIR[6] who arrived at the value of 36.7 for the L/K electron capture ratio. They also observed M x-rays and set 0.46 as the ratio of M-electron to L-electron capture. The L/K electron capture ratio is a sensitive measure of the decay energy for a nuclide with low disintegration energy. For the experimentally observed L/K ratio of 36.7 the calculated decay energy, based on an allowed

transition, is 123 kev. The estimates of decay energy from closed decay-energy cycles are imprecise by comparison. FOREMAN AND SEABORG[7] calculate a value of 210 kilovolts while GINDLER, HUIZENGA AND ENGELKEMEIR[6] estimate 70 kilovolts.

The electron capture decay of Np^{235} leads to U^{235} whose low lying levels are well known. They consist of a 7/2-, 9/2-, 11/2- rotational band based on a K = 7/2 ground state and a 1/2+, 3/2+ and 5/2+ rotational band based on a K = 1/2 base level lying within a few electron volts of the ground state. The K = 1/2, I = 1/2 state has a half life of 26.17 minutes as has been learned from a study of alpha decay of Pu^{239}. (See section 9.2.9) GINDLER, HUIZENGA AND ENGELKEMEIR[6] examined the radiations of Np^{235} very carefully to see whether any of the decay events led to the low-lying excited states of U^{235}. They attempted a chemical isolation of the 26 minute U^{235m} but found no evidence for decay to any state other than the ground state of U^{235}; they set a lower limit of 97.9 percent for decay to this state. Their conclusions are summarized in Figure 2. A ground state assignment of I = 5/2+, K = 5/2 for Np^{235} is consistent with the decay scheme.

Neptunium-235 also decays to a slight extent by alpha emission. JAMES, GHIORSO AND ORTH[3] first observed the emission of 5.06 ± 0.02 Mev alpha particles and reported a branching ratio of 5×10^{-5}. HOFF et al.[4] reported a value of the branching ratio of 3.5×10^{-5}. More recent values by THOMAS[5] and by GINDLER et al.[6] are $(1.23 \pm 0.10) \times 10^{-5}$ and 1.59×10^{-5} respectively. The latter corresponds to a partial alpha half-life of 7.0×10^{4} years. The alpha spectrum is complex as was first shown by HOFF, OLSEN AND MANN[4] who found gamma rays of 26 kev and 85 kev and L x-rays to be in coincidence with alpha particles. These gamma rays were shown to arise from a metastable state with a half-life of 37 millimicroseconds. GINDLER AND ENGELKEMEIR[8] confirmed these findings and also noted a coincidence with K x-radiation in small intensity. Intensities of (0.15 ± 0.02), (0.088 ± 0.008) and (0.006 ± 0.002) photon per alpha disintegration were found for the 26 kev, 84 kev and K x-radiation, respectively.

Direct measurement of the alpha spectrum is extremely difficult because of the low percentage of events going by alpha decay. Nonetheless, by means of precise technique with the ion chamber method GINDLER AND ENGELKEMEIR[8] achieved a partial resolution of the spectrum. Their results are displayed in Figure 3. It is possible to interpret these results in some detail because the low-lying excited levels of Pa^{231} are well-known from a study of the decay schemes of U^{231}, Th^{231} and by Coulombic excitation of Pa^{231} targets with low energy beams

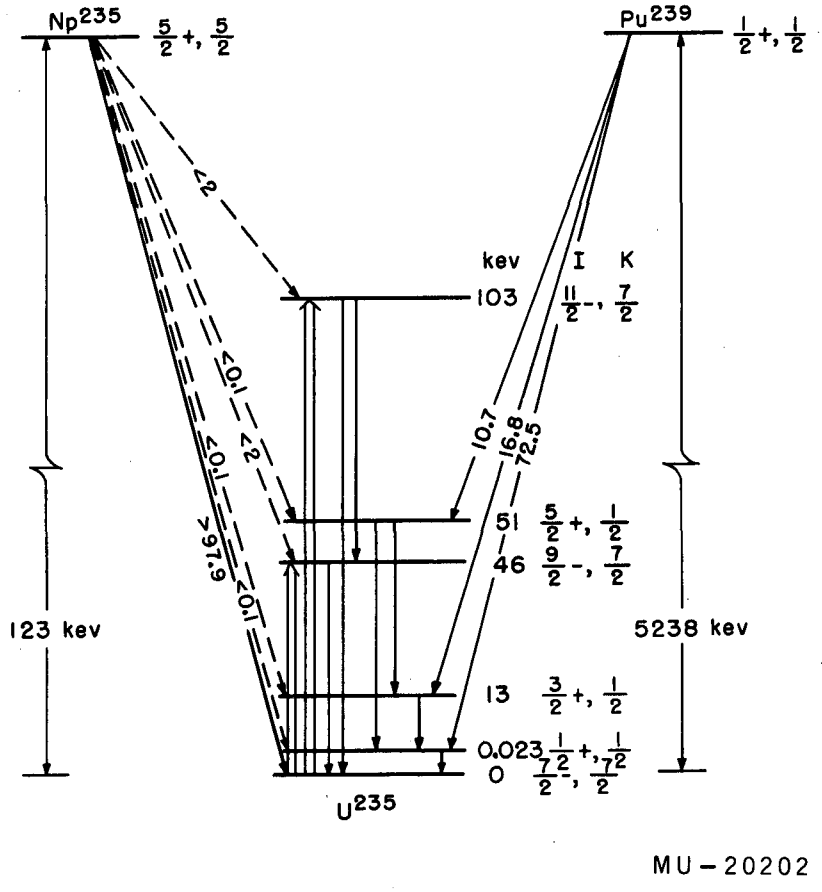

Figure 2. Decay scheme of Np^{235} showing quantitative decay (within experimental error) to the ground state of U^{235}. The three levels in U^{235} with spins and parities 1/2+, 3/2+ and 5/2+ comprise a rotational band and are populated by Pu^{239} alpha decay. The base state 1/2+ has a half life of 26 minutes. Levels at 46 and 103 kev are reached by Coulomb excitation of U^{235} and are assumed to have spins and parities of 9/2- and 11/2-, respectively.

of charged particles. The lower part of the excitation spectrum as deduced from these studies is shown as dotted lines in the figure. The detailed arguments on which these levels and their Nilsson wave function assignments are based are given in section 8.2.10 of Chapter 8.

It is apparent that the favored decay of Np^{235} leads to the Nilsson state 5/2+ 642 which lies at 84.2 kilovolts above ground. On the basis of the principle of favored alpha decay between parent and daughter nuclei bearing identical Nilsson wave function assignments, it is clear that the ground state of Np^{235} is $5/2+\ 642$. The quantum numbers being referred to here are K, π N n_z . It is rather likely that the alpha groups shown in the figure are still not completely resolved and that alpha population is distributed over several members of the rotational bands of each intrinsic state of Pa^{231}. The alpha decay of Np^{235} closely resembles that of Np^{237} discussed below in section 9.1.9 and it is reasonable that the greater complexity observed in Np^{237} decay is also in fact present in Np^{235} decay.

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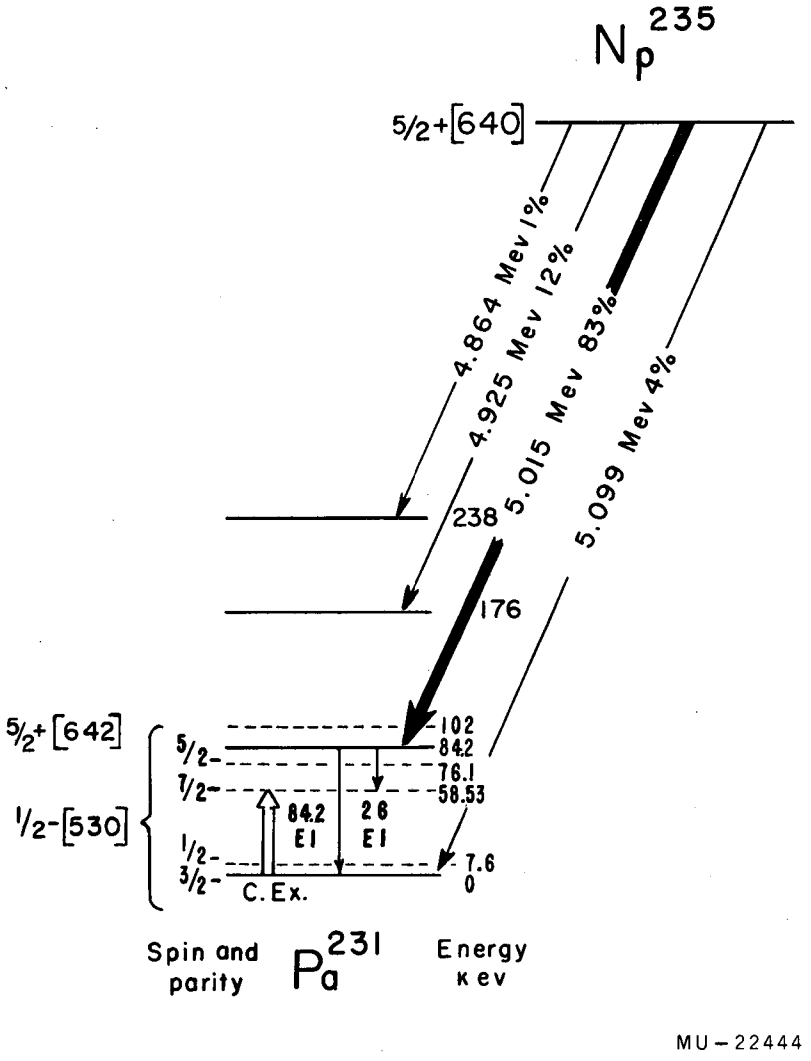

Figure 3. Alpha decay scheme of Np^{235}.
Dotted levels of Pa^{231} and Nilsson state assignments based on discussion of section 8.2.10 of chapter 8. Total alpha branching is 1.59×10^{-5}.

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9.1.7. Neptunium-236. The isotope Np^{236} was discovered by JAMES, FLORIN, HOPKINS AND GHIORSO.[1] In the neptunium fraction isolated from a target of U^{238}, depleted with respect to U^{235}, which had been bombarded with 20 Mev deuterons, they observed the growth of alpha particles of 4.3-cm range, known from other evidence to belong to Pu^{236}. Consequently, there was indicated the formation of Np^{236} and its beta decay to Pu^{236} as follows:

$$U^{238}\ (d,4n)\ Np^{236} \xrightarrow{\beta^-} Pu^{236}$$

The same phenomenon was found in the bombardment of U^{235} with 20 Mev deuterons and 40 Mev helium ions:[1]

$$U^{235}\ (d,n)\ Np^{236} \xrightarrow{\beta^-} Pu^{236}$$

$$U^{235}\ (\alpha,p2n)\ Np^{236} \xrightarrow{\beta^-} Pu^{236}$$

This isotope is also produced in 40 Mev helium-ion bombardments of Np^{237} [2] and U^{233}[3]:

$$Np^{237}\ (\alpha,\alpha n)\ Np^{236} \xrightarrow{\beta^-} Pu^{236}$$

$$U^{233}\ (\alpha,p)\ Np^{236} \xrightarrow{\beta^-} Pu^{236}$$

In the helium-ion bombardments of U^{235} and U^{233} it was possible to observe in the neptunium fraction not only the growth of Pu^{236} alpha particles but also the decay of the beta particles of Np^{236}. From the rate of growth of Pu^{236} alpha particles in a bombardment of U^{235} with helium ions, a half life of 22 hours was derived for Np^{236}. Analysis of the decay curve from the U^{235} experiment gave an independent value of the Np^{236} half life of 21 hours.[1] JAMES, FLORIN, HOPKINS AND GHIORSO[1] were unable to produce samples of Np^{236} free of excessive amounts of Np^{238} and Np^{239}, and therefore could set only wide limits on the beta energy. ORTH AND O'KELLEY,[4] using highly purified U^{235} (99.4 %),

repeated the deuteron bombardments and prepared sufficient activity for absorption measurements and a beta ray spectrometer analysis. At a later time JAFFE[5] AND PASSELL[6] prepared Np^{236} in a similar way for study of the radiations. The results of ORTH AND O'KELLEY had to be reinterpreted by the later workers and will not be quoted here. JAFFE[5] and PASSELL[6] represent the decay scheme as shown in the figure. The L_{II} and L_{III} conversion lines and the M and N conversion lines of the 43.4-kev transition in Pu^{236} and the 44.2 kev transition in U^{236} were resolved from the electron spectrum.[6] HOLLANDER,[7] studying the decay of Np^{236}, has remeasured the energy of the first excited states of U^{236} and Pu^{236} and found 45.28 ± 0.06 kev and 44.63 ± 0.1 kev, respectively. The beta decay of Np^{236} appears to proceed to the 0+ and 2+ levels of the ground state rotational band of Pu^{236}; the electron capture decay appears to proceed to the 0+ and 2+ levels of the ground state rotational band of U^{236} which are familiar from the alpha decay of Pu^{240} to the same nucleus. The log ft values for these four transitions are 7.1, 6.6, 7.5 and 7.0 respectively indicating first forbidden transitions. On this basis one can assign a spin and parity of 1- to 22 hour Np^{236}. This is consistent with the measured spins of Np^{237} (93 protons), which is 5/2, and of U^{235} (143 neutrons), which is 7/2 . The Nilsson orbital assignments of the odd nucleon in the ground states of these nuclei are known on pretty sure evidence to be 5/2+ [642] and 7/2- [743], respectively. Neptunium-236 with 93 protons and 143 neutrons might then be expected to be represented by (5/2+, 7/2-) leading to 1- or 6-. It is logical to assign 1- to the 22 hour form of Np^{236} and 6- to the long lived isomer. The M5 transition between the two isomers would have a very long lifetime and would account for the occurrence of isomerism. It is known, however, that a 5/2- level identified as the Nilsson state [523 5/2-] occurs at 59.6 kev above ground in Np^{237}. This state could possibly be the ground state for the 93rd proton in Np^{236}. This in combination with the 143rd neutron state 7/2- would lead to spins of 1+ and 6+ for the isomers of Np^{236}, which would be an alternative set of assignments. The negative parity set may be preferred on the basis of the log ft values.

GRAY[8] sets the ratio of negatron emission to K-electron capture as 57/43 in Np^{236}. GINDLER, HUIZENGA AND ENGELKEMEIR[9] suggests a 15 percent cor-

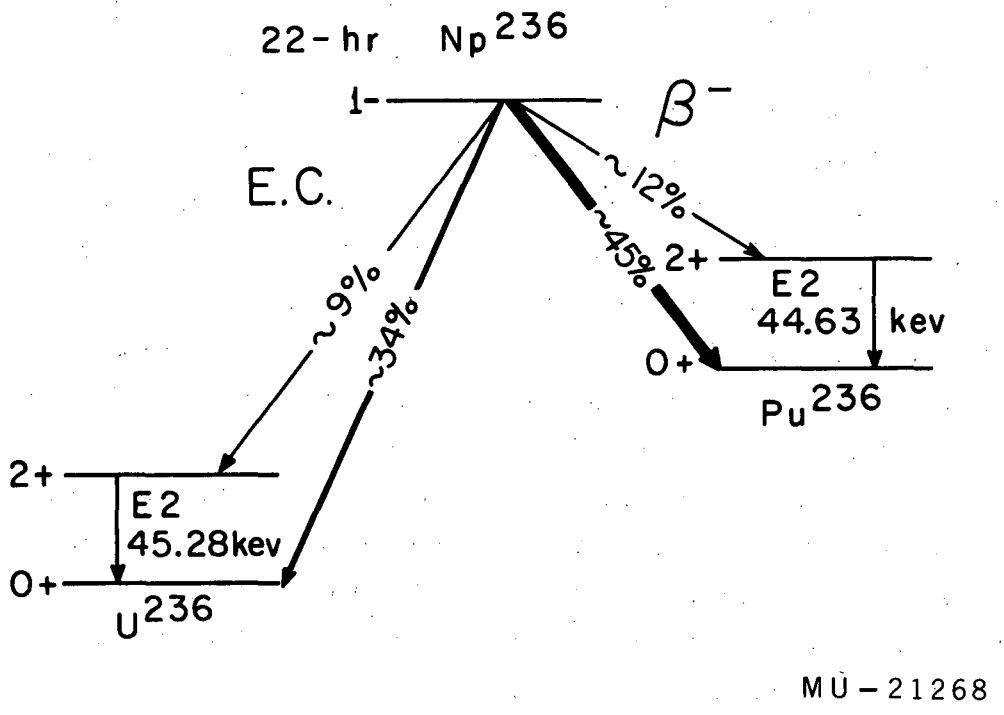

Figure 4. Partial decay scheme of 22 hour Np^{236}.

rection for the contribution of L-electron capture to the electron capture branch. This reduces the β^-/EC ratio to 1.2. GINDLER AND SJOBLOM[11] report the even lower value of 0.95.

The energy of the most energetic beta particle group is about 520 kev.[8] GALLAGHER AND THOMAS[10] observed conversion lines of gamma rays with 641.7 and 687.0 kev energy which defined a level at 687 kev in U^{236}. This may be a K = 0 I = 1- state of collective excitation.

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9.1.8 Long-lived Isomer of Np^{236}. Evidence for the existence of a long-lived isomer of Np^{236} was first obtained by STUDIER, HOPKINS, GHIORSO AND BENTLEY[1] from a 22 Mev deuteron bombardment of uranium metal in which the U^{235}/U^{238} ratio was only 1/2800. Neptunium prepared in a bombardment of long duration was isolated chemically and measurements were made of the fission rate of the sample in a calibrated flux of slow neutrons. These showed the fissionability to decrease with a two day half-life characteristic of Np^{238}. In addition to the fission events due to Np^{238} a long-lived fissionable material was present even after further chemical separations had been performed which should have removed any other element. Since the slow-neutron fission cross section of long-lived Np^{237} was too small to account for the observed number of fission events, and since the long-lived fissionability was found only in the upper 3 mil layer of the uranium metal target where the (d,4n) reaction was energetically possible, it had to be concluded that this fissionability was due to a Np^{236} isomer.

In a followup experiment STUDIER, GINDLER AND STEVENS[2] prepared a larger sample of the suspected isomer in a similar way. Fifteen months after bombardment the neptunium fraction was isolated from the U^{238} and subjected to analysis in a mass spectrometer. Masses 236 and 237 were observed in the mass ratio 0.062. The alpha activity of the sample was entirely due to Np^{237}. Beta activity was observed and assigned to Np^{236} (long-lived). A beta decay half life of $\geq$ 5000 years was derived. This is a highly-forbidden transition (log ft $>$ 12). The cross section for fission with slow neutrons was measured to be 2800 barns. GINDLER, HUIZENGA AND ENGELKEMEIR[3] repeated the experiment of STUDIER, GINDLER AND STEPHENS with the same results. They found that the 22 hour isomer was produced in a 7-fold greater yield than the long-lived isomer.

Further research is required to determine the relationship of the 5000-year Np^{236} to the 22-hour Np^{236} but a tentative spin of 6 can be assigned to the isomer on the basis of the considerations discussed in the preceding section.

References for long-lived isomer of Np^{236}

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9.1.9 Neptunium-237. The discovery of Np^{237} by WAHL AND SEABORG[1] is mentioned in the introductory section 9.1.1. There is also some discussion of Np^{237} in section 7.1.2 of Chapter 7 where the 4n + 1 decay chain is treated. Np^{237} is an alpha-emitter with a half-life reported by MAGNUSSON AND LA CHAPELLE[2] as $(2.20 \pm 0.10) \times 10^6$ years and by BRAUER[3] and his co-workers as $(2.14 \pm 0.01) \times 10^6$ years. This latter value corresponds to a specific activity of 1562 ± 7 disintegrations per minute per microgram. The spontaneous fission rate is less than 5 fissions per gram-hour, corresponding to a half-life limit of greater than 4×10^{16} years.[4] Np^{237} is beta stable. This isotope is the longest lived species of the element, neptunium and the most suitable for investigation of the chemical properties of the element. It is isolated in gram and kilogram amounts by the special processing of spent reactor fuel elements made of natural or slightly enriched uranium.

A careful measurement of the alpha spectrum is a matter of some difficulty because of the low specific activity. MAGNUSSON, ENGELKEMEIR, FREEDMAN, PORTER AND WAGNER[5] studied the spectrum with a gridded ionization chamber connected to a multichannel pulse height analyzer. Close attention was paid to important details[6], such as sample collimation, to get the best resolution of which this method is capable. Their results are summarized in Table 2. These same authors used scintillation, proportional and magnetic spectrometers to get information on transitions with the energies, 20, 29, 56.8, 86.9, 145, 175, and 200 Mev. Transition assignments derived from α-e^-, α-γ and γ-γ coincidence measurements were used to construct the decay scheme shown in the figure. STEPHENS[7] also contributed to the measurement of the gamma transitions by scintillation spectrometer determinations and γ-γ coincidence measurements.

Alpha spectrum measurements by KONDRAT'EV, NOVIKOVA, VOROB'EV AND GOLDIN[8] are also listed in the table. ASARO, STEPHENS AND PERLMAN[9] have unpublished results on the alpha spectrum of Np^{237} taken on a magnetic spectrometer which agree for the most part with those seen in the table with the exception that they include more groups.

Table 2 Alpha Particle Groups Emitted by Np^{237}

Magnusson et al.*			Kondrat'ev et al.**	
Alpha Particle Energy (Mev)	Energy of Pa^{233} Excited State (Kev)	Abundance (Percent)	Alpha Particle Energy (Mev)	Abundance (Percent)
4.872	0	3.1	4.867	2.5
4.816	57	3.5	4.803	3.4
4.787	86	53	4.781	54
4.767	107	29	4.761	29.5
4.713	162	1.7	4.701	2.3
4.674	201	3.3	4.643 (sev'l unresolved groups)	8.3
4.644	232	6.0		
4.589	288	0.5		
4.52	~360	0.02		

* Data of Magnusson and co-workers, Phys. Rev. 100, 1237A, 1955. Relative to Th^{230} E_α = 4.682 Mev and Po^{209} E_α = 4.877 Mev.

** Data of Kondrat'ev and co-workers relative to Po^{210} E_α = 5.2984 Mev.

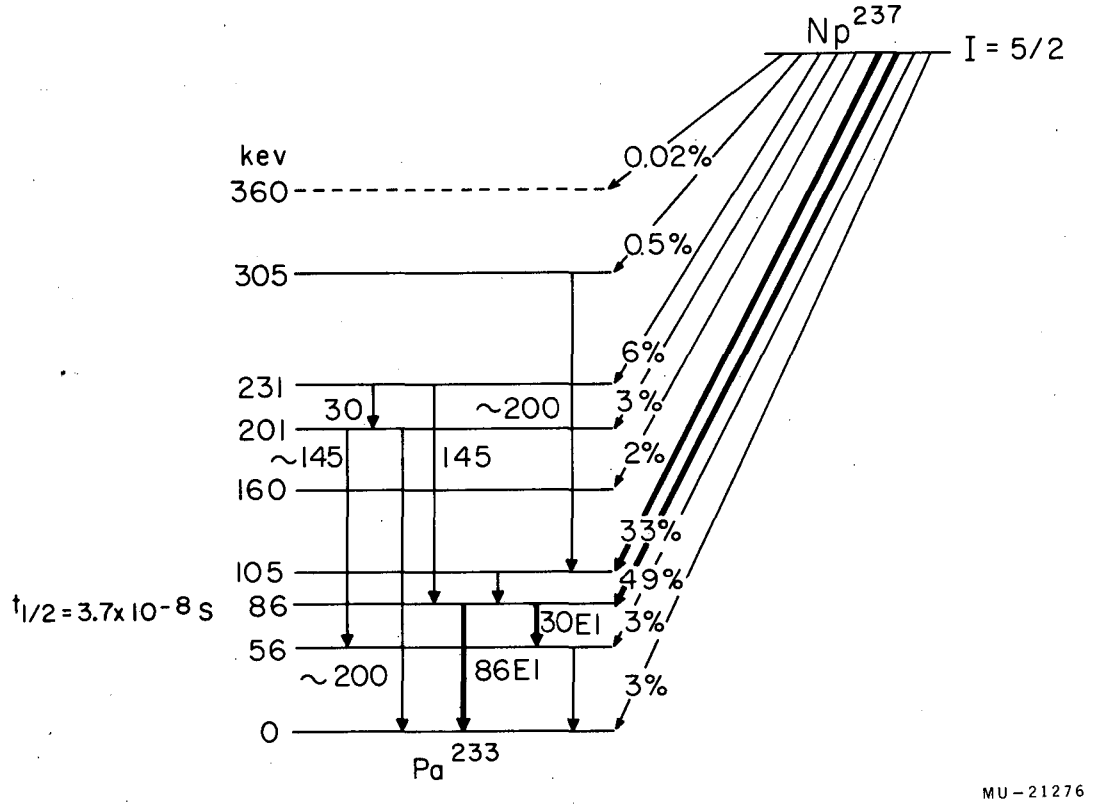

Figure 5. Decay scheme of Np^{237} derived chiefly from the experimental results of Magnusson and Co-Workers.

The decay scheme shown in the figure is incomplete and no detailed assignments of the spins, parities and excitation character has been given to the majority of the levels in the Pa^{233} daughter nucleus. There are interesting experimental facts and suggested interpretations of the lower-lying levels and of the transitions which de-excite them which we should like to mention. As an aid to the discussion we show a figure 6 which emphasizes the similarity in the lowest levels of Pa^{231} and Pa^{233}.

First let us consider the 86.3 kev level in Pa^{233} which plays a prominent role in the decay of Np^{237} since it is populated in 90± 5 out of every 100 alpha decay events. ENGELKEMEIR AND MAGNUSSON[10] have shown that this level is metastable by measuring an apparent half-life of $(3.69 \pm 0.04) \times 10^{-8}$ seconds for the 86.3 and 29.6 kev transitions. The true partial half-lives are 2.6×10^{-7} seconds and 3.0×10^{-7} seconds, respectively. STROMINGER AND RASMUSSEN[11] confirmed these results and speculated on their significance. Both the 86.3 and the 29.6 kev transitions are believed to be E1 in nature from the measured conversion coefficients and from other lines of evidence.[12] The half-lives of these transitions are enormously longer than would be predicted by the "single particle" transition probability formulas. The retardation in these cases in 1.4×10^{6} and 7.2×10^{4}, respectively. These transitions belong to a class of E1 transitions observed in the decay of heavy deformed nuclei of odd mass which have anomalously long decay periods and anomalous L-shell and M-shell conversion coefficients. The 84.2 kev and 25.7 kev transitions in the decay of the 84.2 kev level of excitation of Pa^{231} (shown in the figure) also show these anomalous characteristics. Experimental data on this class of E1 transitions is discussed in a paper by ASARO, STEPHENS, HOLLANDER AND PERLMAN[12]. Some of the theoretical implications are discussed by NILSSON AND RASMUSSEN[13].

Now let us discuss the spin and parity assignments of the Pa^{233} levels. The ground state spin of Pa^{233} is deduced to be 3/2 from the decay scheme of Np^{237} and of Pa^{233}. This assignment is confirmed by the measurement[19] of spin 3/2 for the ground state of Pa^{231}. In each case this ground state is probably the I = 3/2 member of a rotational band of levels with K = 1/2. In the discussion of the collective model in chapter 3 it is pointed out that K = 1/2 rotational bands can have an "anomalous" ordering of levels. The Nilsson orbital assignment for the ground state of both Pa^{231} and Pa^{233} is 1/2- 530 . For

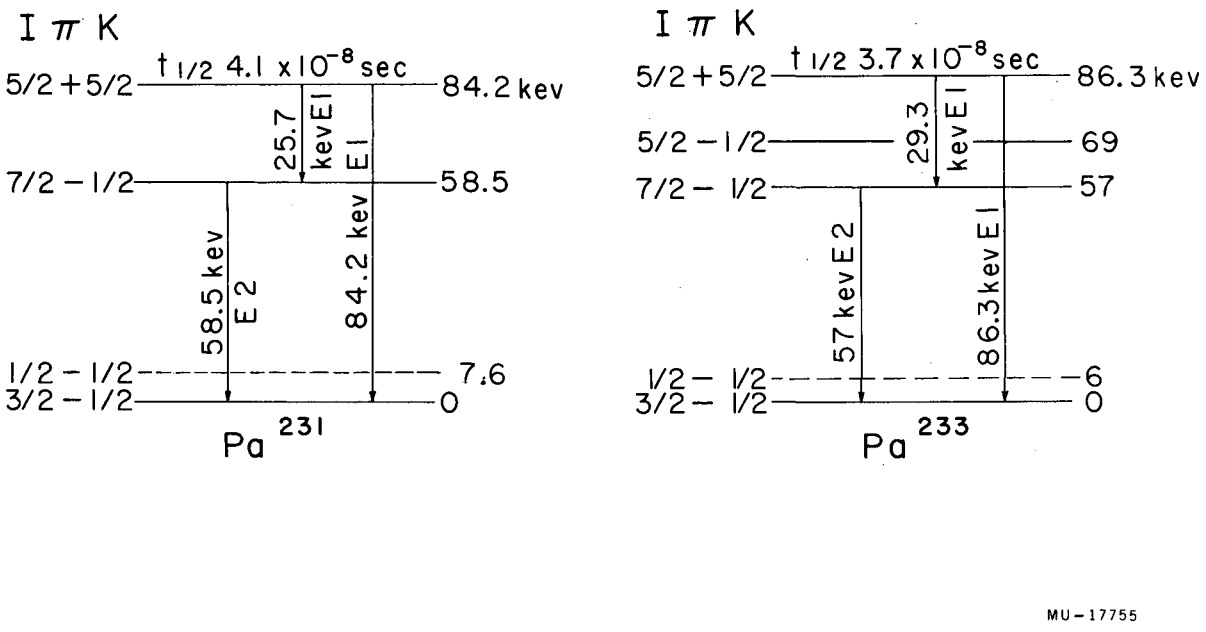

Figure 6. Partial level schemes of Pa^{231} and Pa^{233}. The lowest-lying levels of Pa^{233} as seen in the α-decay of Np^{237} are shown on the right.

details of these assignments see section 3.5.5 of chapter 3 or other references.[9,14,15] The levels at 6, 57 and 69 kev are assigned as I = 1/2, 7/2 and 5/2 members of this same band. Note the inversion of the 5/2 and 7/2 members. In the Pa^{231} case only the I = 3/2 and I = 7/2 members have been identified by decay scheme analysis but the others are believed to exist. The NILSSON state assignment of the 86.3 kev level in Pa^{233} is 5/2+ [642.] A somewhat uncertain assignment of 3/2+ [651] has been made for the 200 kev level.

The ground state spin has been determined to be 5/2 by hyperfine structure[16] analysis and by the method of nuclear paramagnetic resonance.[17] The Nilsson state assignment of the Np^{237} ground state is firmly made to 5/2+ [642] for reasons which are discussed in connection with the decay scheme of Am^{241} below. The identity of this Nilsson assignment with that of the 86.3 kev level in Pa^{233} accounts for the high percentage of the alpha transitions which go to this level.

The neptunyl ion, NpO_2^{++}, is paramagnetic because of its unpaired 5 f electron. It is possible to grow crystals of such compounds as rubidium neptunyl acetate and by cooling such crystals to temperatures in the vicinity of absolute zero to align the nuclei of Np^{237}. The angular distribtuion of the alpha particles of such aligned nuclei of Np^{237} have been studied and found to be anisotropic by ROBERTS, DABBS AND CO-WORKERS[18]. It was found that Np^{237} emits its alpha particles preferentially out through the tips of the Np^{237} nucleus, a prolate spheroid.

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9.1.10 Neptunium-238. It is mentioned in the historical introduction that 2.10-day Np^{238} was the second isotope of neptunium to be discovered and that it was first produced[1] by the cyclotron-induced reaction:

$$U^{238}\ (d,2n)\ Np^{238}$$

It can also be produced by helium ion bombardment of uranium:[2]

$$U^{238}\ (\alpha,\ p3n)\ Np^{238}$$
$$U^{235}\ (\alpha,\ p)\ Np^{238}$$

Cross sections for these reactions are listed in Table 5.9, Chapter 5. The first evidence for the production of Np^{238} by the reaction $Np^{237}\ (n,\gamma)\ Np^{238}$ was the discovery of Pu^{238} in pile produced plutonium.[3] This was produced by the neutron reaction sequence:

$$U^{238}\ (n,2n)\ U^{237} \xrightarrow{\beta^-} Np^{237}\ (n,\gamma)\ Np^{238} \xrightarrow{\beta^-} Pu^{238}$$

fast neutron; slow neutron

In reactors operating with enriched uranium another mechanism becomes important, namely:

$$U^{235}\ (n,\gamma)\ U^{236}\ (n,\gamma)\ U^{237} \xrightarrow{\beta^-} Np^{237}\ (n,\gamma)\ Np^{238} \xrightarrow{\beta^-} Pu^{238}$$

The rate of buildup of Pu^{238} by this latter mechanism in a high flux pile is discussed in section 9.28 of this chapter.

The production of Np^{238} from Np^{237} was first observed directly by JAFFEY AND MAGNUSSON.[4] The slow neutron capture cross section of Np^{237} is 172 barns.[5] An important feature of this reaction is that it provides a means of producing Np^{238} free from other short-lived neptunium isotopes, and Pu^{238} free from other plutonium isotopes, although in careful studies of the radiations of Np^{238} prepared in this way some attention must be paid to the growth of Pa^{233} activity from its Np^{237} parent.

The early studies of Np^{238} were complicated by the fact that the isotope, when produced by the deuteron bombardment of uranium, was always accompanied by comparable amounts of Np^{239}. Since the half-life of the latter, 2.33 days, is close to that of Np^{238} a differential analysis of decay and absorption curves was required. Extensive studies of the complex beta-rays, gamma-rays, x-rays and conversion electrons have been carried out in recent years using many modern techniques. These studies have been carried out on pure Np^{238} made from neutron irradiation of Np^{237} and with the Np^{238}-Np^{239} mixture made from cyclotron bombardment. We shall not try to review the experimental evidence exhaustively but will summarize the most important results. In this we follow closely the treatment of RASMUSSEN, SLATIS AND PASSELL,[6] of RASMUSSEN, STEPHENS, STROMINGER AND ASTROM,[7] and of ALBRIDGE AND HOLLANDER.[13] Others who have also done detailed work on this isotope are FREEDMAN, JAFFEY AND WAGNER[8] and BARANOV AND SHLYAGIN.[9] Data on the beta groups which may be resolved from the electron spectrum are summarized in Table 3. Data on the numerous conversion electron lines which are superimposed on the complex continuous beta spectrum are given in Table 4. The half life of Np^{238}, which was originally reported[1] as 2.0 days has since been revised slightly to 2.10 days.[8]

The gamma rays revealed by a NaI scintillation spectrometer include an unresolved photopeak near 1 Mev made up of 4 high energy gamma-rays, a photopeak near 100 kev made up of 102-kev gamma-rays plus K x-rays, and a low energy x-ray peak of about 20 kev. A number of gamma-gamma, gamma-x-ray, and beta-gamma coincidence measurements have been made[7] to clarify the decay scheme.

Figure 7c presents a decay scheme based on all these measurements. This decay scheme is not completely established in spite of the detailed experiments which have been done to test it. One of our purposes in presenting it here is to discuss a possible interpretation of the higher-lying levels of Pu^{238} since this nucleus was the first in the heavy element region to provide evidence for the vibrational states at ~1 Mev predicted by the unified model of the nucleus.

The lower-lying states of Pu^{238} are firmly established by the study of the alpha decay of Cm^{242}. A ground state rotational band with levels 0+, 2+, 4+, 6+, and 8+ has been observed. (See section 9.4.6 and the Figures therein.) These states are connected by a series of cascading E2 transitions which have

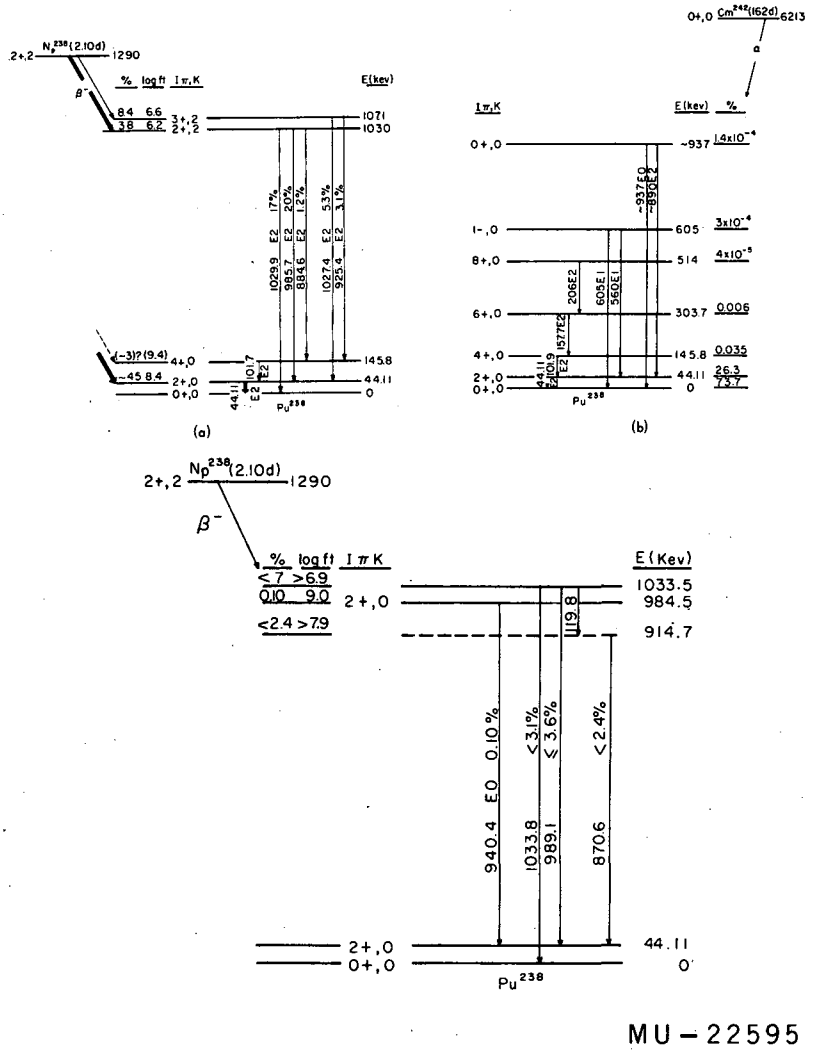

Figure 7. Decay scheme of Np^{238}. The more certain features are shown in part a, the more speculative in part c. The decay scheme of Cm^{242} is also shown in part b since it strengthens the interpretation of the level system seen in the decay of Np^{238}. All observed levels of Pu^{238} are interpreted as arising from a single (ground) state of intrinsic particle motion. The lowest levels clearly constitute a rotational band based on the ground state. Several of the 6 levels at around 1 Mev may be assigned to two rotational bands based on a K = 0, beta-vibrational level at 937 kev and a K = 2, gamma-vibrational level at 1030, but this interpretation is speculative.

Table 3

Beta groups of Np^{238}

End point energy (Mev)	Abundance (%)	Reference	log ft	Reference
Determinations of the most energetic beta group				
1.272	47	8		
1.25	45	6	8.5	7
1.236	38	9	8.6	9
Determinations of lower energy beta groups				
0.258	53	8		
0.27	55	6	5.8	7
0.28 and	20		6.6	9
0.250	31	9	6.2	
0.200 ?	8			
1.139 ?	2.8			

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Table 4

Np^{238} Conversion Electron Data

Electron energy (kev)	Conversion shell	Transition energy[a] (kev)	Selected transition energy[b] (kev)	Abundances per 100 beta disintegrations			Relative intensities[h]	
				Freedman et al.[c]	Slätis et al.[d]	Rasmussen et al.[e]	Baranov and Shlyagin[f]	Albridge and Hollander[g]
21.76	L_{II}	44.01		38	28.7	---	23.1	---
25.96	L_{III}	44.02		20	21.0	---	21.1	---
38.44	M_{II}	44.00		14	---	---	11.6	---
39.40	M_{III}	43.96		---	---	---	5.72	---
42.58	N_{II}	43.96		---	15.2	---	3.71	---
42.84	N_{III}	43.97		3.2	---	---	---	---
43.71	O	~44.0		---	---	---	---	---
			(44.11)[i]					
79.52	L_{II}	101.8		1.9	1.4	---	1.21	---
83.74	L_{III}	101.8		---	0.9	---	0.64	---
96.20	M_{II}	101.8		1.2	0.7	---	0.40	---
97.17	M_{III}	101.7		---	---	---	0.36	---
100.3	N_{II}	101.7		---	---	---	---	---
101.4	O	101.7		---	---	---	---	---
			101.7					
96.79	L_I	119.9		---	---	---	---	---
113.9	M_I	119.8		---	---	---	---	---
118.1	N_I	119.7		---	---	---	---	---
			119.8					

Table 4 (continued)

Electron energy (kev)	Conversion shell	Transition energy[a] (kev)	Selected transition energy[b] (kev)	Abundances per 100 beta disintegrations			Relative intensities[h]	
				Freedman et al.[c]	Slätis et al.[d]	Rasmussen et al.[e]	Baranov and Shlyagin[f]	Albridge and Hollander[g]
99.22	K	221.0	221.0	---	---	---	---	0.09
170.8	K	292.6	292.6	---	---	---	---	(?)
748.8	K	870.6	870.6	---	---	---	---	<0.01
762.8	K	884.6	884.6	---	---	---	---	0.014
803.6	K	925.4	925.4	---	0.05	0.04	---	0.034
818.8	K	940.6		---	0.10	0.07	0.09	0.072
917.9	L_I, L_{II}	940.2		---	0.06	---	---	0.021
934.4	M_I, M_{II}	940.0		---	---	---	---	0.0075
			940.4					
821.3	K	943.1	943.1	---	---	---	---	0.0093
864.0	K	985.8		0.3	0.26	0.20	0.20	0.20
963.1	L_I, L_{II}	985.4		---	0.13	0.06	0.06	0.059
967.8	L_{III}	985.9		---	---	---	---	0.010
980.2	M_I, M_{II}	985.8		---	---	---	---	0.020
985.3	N	~986		---	---	---	---	0.0090
			985.7					
867.3	K	989.1	989.1	---	---	---	---	0.012
905.6	K	1027.4		0.3[j]	0.22[j]	0.20[j]	0.16[j]	0.048
1005.1	L_I, L_{II}	1027.4		---	0.08	0.04	0.03	0.018
1021.7	M	~1027		---	0.06	0.02	---	extremely weak
			1027.4					

Table 4 - (continued)

Electron energy (kev)	Conversion shell	Transition energy[a] (kev)	Selected transition energy[b] (kev)	Abundances per 100 beta disintegrations			Relative intensities[h]	
				Freedman et al.[c]	Slätis et al.[d]	Rasmussen et al.[e]	Baranov and Shlyagin[f]	Albridge and Hollander[g]
908.1	K	1029.9		---	---	---	0.08	0.15
1007.3	L_I, L_{II}	1029.6		---	---	---	0.03	0.039
1024.8	M	~1030		---	---	---	---	0.011
			1029.9					
912.1	K	1033.8	1033.8	---	---	---	---	<0.01
973.5	K	1095.3	1095.3	---	---	---	---	?

(a) Electron binding energies were taken from Hill, Church, and Mihelich, Revs. Sci. Instr. 23, 523 (1952).

(b) The selected values are weighted averages of the experimental values.

(c) Reference 8

(d) Reference 10

(e) Reference 6

(f) Reference 9

(g) Reference 13

(h) For purposes of comparison, the relative intensities from Baranov and Shlyagin and from Albridge and Hollander were normalized to Rasmussen's value of 0.20 for the K line of the 985-kev transition. In the last column question marks indicate uncertainty as to the actual existence of the lines.

(i) The weighted average is 44.00. The value 44.11 had been reported previously by Smith and Hollander by measurement of Cm^{242} in which the same transition occurs and is considered more accurate.

(j) The intensities listed for the 1027-kev transition by Freedman, Slätis, and Rasmussen include those of the unresolved 1030-kev transition. The division of intensities by Baranov and Shlyagin between 1027 and 1030 is reported as only approximate.

been precisely measured and characterized. In agreement with the spin and parity assignments there are no crossover transitions. Only the first three members of this band are involved in the decay of Np^{238}. There is a 1-level of Pu^{238} at 605 kev but this plays no role in the decay of Np^{238}.

The levels at about 1 Mev which are populated by the lower energy beta groups of Np^{238} are interpreted as a gamma-vibrational band and a beta-vibrational band. Beta-vibration refers to oscillations of the spheroidal nucleus in which the eccentricity is changed but the shape is preserved; gamma-vibration refers to oscillations entailing shape changes. A general account of collective oscillations is given in chapter 3. An excited vibrational state can have rotational excitation superimposed on it so that in general each such state can give rise to a whole family of levels. A strong indication that rotational excitation of two of the higher lying levels may be occurring in the present case is the fact that two sets of these high-lying levels are separated in energy by about 45 kev or the same amount as the lowest members of the ground state rotational band. The higher levels of Pu^{238} are interpreted as a (0+,2+) rotational band based on a K = 0, beta-vibrational level at 937 and a (2+, 3+) rotational band based on a K = 2, gamma-vibrational level at 1030 kev. It has been suggested that two other levels exist at 914.7 and 1033.5 kev and are perhaps related to an octupole vibrational band[13] with a base state spin and parity 2-.

Several lines of evidence suggest the above interpretations: (1) the spins and parities as determined by the multipolarity of the gamma transitions. (2) The close spacing of the levels above 900 kev; in particular the 48 kev spacing in the beta-vibrational pair and the 41 kev spacing in the gamma-vibrational pair are quite close to the 44.11 kev spacing of the 2+ level in the ground state rotational band.

(3) Another strong piece of evidence concerns the ratio of the reduced transition probabilities for the energetic gamma rays which depopulate the level at 1030 kev. (The reduced transition probability is obtained by correcting for the fifth power energy dependence.) Since these gamma rays originate at a common level and go to levels in the same rotational band, the relative intensities of the gamma rays do not depend on the details of the nuclear wave functions but only on the squares of the appropriate vector addition coefficients (CLEBSCH-GORDAN coefficients) involving I- and K-quantum numbers. The experimental intensity ratios and the ratios calculated theoretically by the equation given in

Chapter 3 for three different values for $K_{initial}$ are compared in Table 5. The assignment, K = 2, is clearly indicated for the 1030-kev level. With this assignment the absence of M1 character in the 985.7 kev gamma ray finds a natural explanation in K-forbiddenness; the K=2 exceeds the multipolarity, L. The weakness of the gamma ray from the 1030 (2+) level to the 146 kev (4+) level also receives a natural explanation.

The spin of Np^{238} was determined to be 2 by the atomic beam magnetic-resonance method.[14] ALBRIDGE AND HOLLANDER calculated the log ft values of beta decay to the various levels which are given in the figure. It is to be emphasized that these calculations are not independent of the present assignment of multipole orders of the various transitions; however, the only ft values which are sensitively dependent are those of the beta decay to the 985- and 1089-kev levels. If the transitions which de-excite these states should prove to be of M1 character instead of E0 character, then the log ft values will decrease by one unit.

The log ft values are of importance as an indication of the parity of Np^{238}. The most reasonable choice, based upon the existing data, seems to be even parity. The log ft values of 6.2 and 6.6 to the 1030- and 1071-kev levels, respectively, indicate either slow allowed transitions (I = 0 or 1, no) or rather fast first-forbidden transitions (I = 0 or 1, yes). As pointed out by Rasmussen et al.[7], the nuclear rearrangement accompanying the decay to the vibrational levels may tend to hinder these transitions; thus the interpretation of the transitions as slow allowed, with even parity for Np^{238} has been chosen.

One can make use of the 2+ (or possibly 2-) assignment of Np^{238} to draw conclusions concerning the nature of the single-particle states that comprise the ground state of this nucleus. It is simplest to assume that the odd proton and odd neutron in Np^{238} have the same Nilsson orbitals as are seen in the ground state or the very low-lying states in the neighboring odd mass isotopes or isotones. The 93rd proton in Np^{235} in Np^{237} and in Np^{239} has the Nilsson assignment 5/2 + 642 so it seems likely that the odd proton in Np^{238} also is in this state. The 145th neutron in Pu^{239} is in the 1/2+ 631 Nilsson orbital so perhaps the 145th neutron in Np^{238} is also in this state. It is also possible that the proton is in the 5/2- 523 state found in Np^{237} at 60 kev and in Np^{239} at 75 kev. A second possibility for the odd neutron is 1/2- 501, which is the assignment favored for the 145th neutron in U^{237}. GALLAGHER AND MOSZKOWSKI[16] have formulated coupling rules for the angular momentum of individual particle states in odd-odd nuclei. According to these rules the possible

Table 5. Experimental and theoretical relative reduced E2 transition probabilities from the 1030-kev level (2+).

Final state I_f	Experimental $\frac{B(2 \longrightarrow I_f)}{B(2 \longrightarrow 0)}$		Theoretical $\frac{B(2 \longrightarrow I_f)}{B(2 \longrightarrow 0)}$		
			$K_i = 0$	$K_i = 1$	$K_i = 2$
	Rasmussen[7]	Albridge[13]			
2+ (44.11 kev)	1.3	1.5	1.4	0.36	1.4
4+ (145.8 kev)	<0.3	0.15	2.6	1.1	0.071

combinations of the above cited states which give a resultant ground-state spin of 2 are the following:

Proton	Neutron	Final State
5/2+ 642	1/2+ 631	2+
5/2+ 642	1/2- 501	2-

Thus the coupling rules predict for the proton state in Np^{238} the unique assignment 5/2+ 642 . If the even choice of parity is correct, the neutron state is 1/2+ 631.

Closed decay energy cycles (see chapter 2) indicate that Np^{238} is unstable toward electron capture decay to U^{238} but only by about 120 kev so that K-electron capture is energetically impossible. JAFFE[10] has looked for possible L-electron capture by studying the L x-rays using a Cauchois-type bent-crystal spectrometer. With the high resolution of this instrument the L x-rays of uranium should have been observable in the presence of L x-rays of plutonium. His failure to find L x-rays of uranium allowed him to set an upper limit of 4 percent for L_I electron capture and 2 percent for L_{II} electron capture.

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9.1.11 Neptunium-239. In the historical description of the element neptunium presented in section 9.1.1 we discussed the early experiments on Np^{239} by MC MILLAN AND ABELSON[1] which constituted the discovery of neptunium. The isotope Np^{239} is also made by a number of reactions carried out in a cyclotron. These synthetic methods are summarized by the reactions:

$$U^{238}\ (n,\gamma)\ U^{239} \xrightarrow[23\ min]{\beta^-} Np^{239}$$

$$U^{238}\ (d,n)\ Np^{239}$$

$$U^{238}\ (d,p)\ U^{239} \xrightarrow{\beta^-} Np^{239}$$

$$U^{238}\ (\alpha,p2n)\ Np^{239}$$

Some reaction cross section values are given in Table 5.5, Chapter 5. The isotope Np^{238} is an impurity in any neptunium sample prepared by deuteron or helium ion bombardment and it has to be considered in any study of the radiations of Np^{239} in such material.

The half-life of Np^{239} is 2.35 days.[20,21] The disintegration scheme is complex involving four or more partial beta spectra, nineteen or more gamma-rays, x-rays, a large number of conversion electron and Auger-electron lines. A number of the more complete studies are summarized in Tables 6 and 7. Early spectroscopic studies were carried out by FULBRIGHT[2], SLÄTIS[3], TOMLINSON, FULBRIGHT AND HOWLAND[4] AND FREEDMAN et al.[5] GRAHAM AND BELL[6] used a thin lens spectrometer to study the electrons. Delayed coincidence measurements by these experimenters established that the 210-, 227- and 276-kev transitions followed a state with a half-life of $(1.1 \pm 0.1) \times 10^{-9}$ seconds. ENGELKEMEIR AND MAGNUSSON[7] discovered a 193 millimicrosecond metastable level in Pu^{239} by the delayed coincidence technique in which anthracene and sodium iodide scintillation crystals were used as beta and gamma detectors, respectively. The delayed state is preceeded by a (343 ± 15) - Kev beta group and is de-excited mainly by E1 transitions of 61- and 105- Kev energy. LEFEVRE, KINDERMAN AND VAN TUYL[8] observed the gamma-ray photons with the aid of a NaI scintillation crystal and found 440 Kev and 490 Kev photons in low abundance which had not previously been reported. The abundance of these gamma rays is 1.6×10^{-4} and 1.9×10^{-4} per disintegration. HOLLANDER, SMITH AND MIHELICH[9] studied the conversion electron spectrum with 180° permanent magnet spectrographs utilizing

Table 6 Beta Ray Groups of Np^{239}

	Slätis[3] 1947	Graham and Bell[4] 1951	Tomlinson, Fulbright and Howland[5] 1951	Freedman et al[6] 1952	Baranov and Shlyagin[10] 1956	Connor and Fairweather[23] 1959
End point Energy (kev) and abundances (%)	288	310 (47%)	330	329 (52%)	327 (45%)	332 (28%)
	403	435 (46%)	440	380 (10%)	382 (27%)	393 (13.5%)
	676	705 (7%)	654	441 (31%)	439 (21%)	437 (48%)
			715	655 (1.7%)	655 } (7%)	654 (4%)
				718 (4.8%)	723 }	713 (6.5%)

Table 7: Gamma Ray Energies in Np^{239} Decay as Shown by Conversion Electron Data

Fulbright[2] 1944	Tomlinson, Fulbright and Howland[5] 1951	Graham and Bell[4] 1951	Freedman et al[6] 1952	Baranov and Shlyagin[10] 1956	Hollander Smith and Mihelich[9] 1956	Ewan, Geiger, Graham and Mac Kenzie[27] 1959
			13	12		
			19			
	44.2		44	44.4	44.64	44.65
49.1	49.0	49	49	49.3	49.40	49.41
57.3	57.0	57	57	57.2	57.25	57.26
61.2	61.4	61	61	61.4	61.4	61.46
67.5	67.5	67	67	67.9	67.82	67.86
			77			88.06
105.5	105		105	106.2	106.12	106.14
						106.47
						166.39
					181.8	181.71
209.3	209	210	209	210.3	209.9	209.76
227.8	228	227	228	[illegible]	226.4	226.42
				228.4	228.4	228.20
	254		254		254.6	254.41
					273.1	272.87
277.4	277	276	277	278.1	277.7	277.62
	286		285		285.6	285.47
			316	[illegible]	316.1	315.91
			334	334	334.5	334.33

A number of estimates of beta ray end-points and gamma ray energies were made by absorption techniques in early publications on Np^{239} but these may be regarded as superseded by the spectrometer work quoted above.

photographic recording. BARANOV AND SHLYAGIN[10] have studied Np^{239} with a double focussing spectrometer, a scintillation gamma spectrometer, a proportional counter and other equipment. EWAN, GEIGER, GRAHAM AND MAC KENZIE[27] have studied the conversion electron spectrum with an iron-free double-focussing spectrometer and the beta spectrum has been re-examined by CONNER AND FAIRWEATHER.[23]

A number of decay schemes which have been proposed on the basis of part of the data quoted above have had to be modified in the light of later more complete information. In the discussion which follows we accept the interpretation of HOLLANDER, SMITH AND MIHELICH[9]. (See also the review of PERLMAN AND RASMUSSEN.[11]) In constructing a level scheme for Pu^{239} based on the beta decay of Np^{239} one is aided by the fact that the electron capture decay of Am^{239} and, particularly, the alpha decay of Cm^{243} give independently-derived information on the excited levels of Pu^{239} which aids in the elimination or acceptance of proposed level schemes. Coulombic excitation of Pu^{239} has also been of considerable importance in setting a location of the ground state. A decay scheme consistent with all known information on Np^{239} is shown in Figure 8. For comparison the levels of Pu^{239} revealed by Np^{239} beta decay, Am^{239} electron capture decay, and Cm^{243} alpha-decay are shown in the figure.

The levels of Pu^{239} have been interpreted as 4 sets of rotational levels. The separation of the levels into rotational bands is shown more clearly in the second figure which is free of all the details concerning gamma transitions.

The ground state and the first five excited levels comprise one band. The measured ground state spin of 1/2 (BLEANEY, LLEWELLYN, PRYCE AND HALL[12]) identifies this band, with its irregular spacings, as an "anomalous" $K = \Omega = 1/2$ band with energy level spacings given by the special rotational formula, applicable to $K = 1/2$ cases.

$$E_I = \frac{\hbar^2}{2\mathcal{I}} \left[I\,(I+1) + (-)^{I+1/2}\, a\,(I + 1/2) \right]$$

The quantity a, the decoupling parameter, depends on details of the intrinsic nucleonic structure. The discussion of this paragraph follows the language of the Bohr-Mottelson unified model according to which K is the projection of the total angular momentum on the symmetry axis of the deformed (spheroidal) nucleus and Ω is the component of the total nucleonic angular momentum along the symmetry

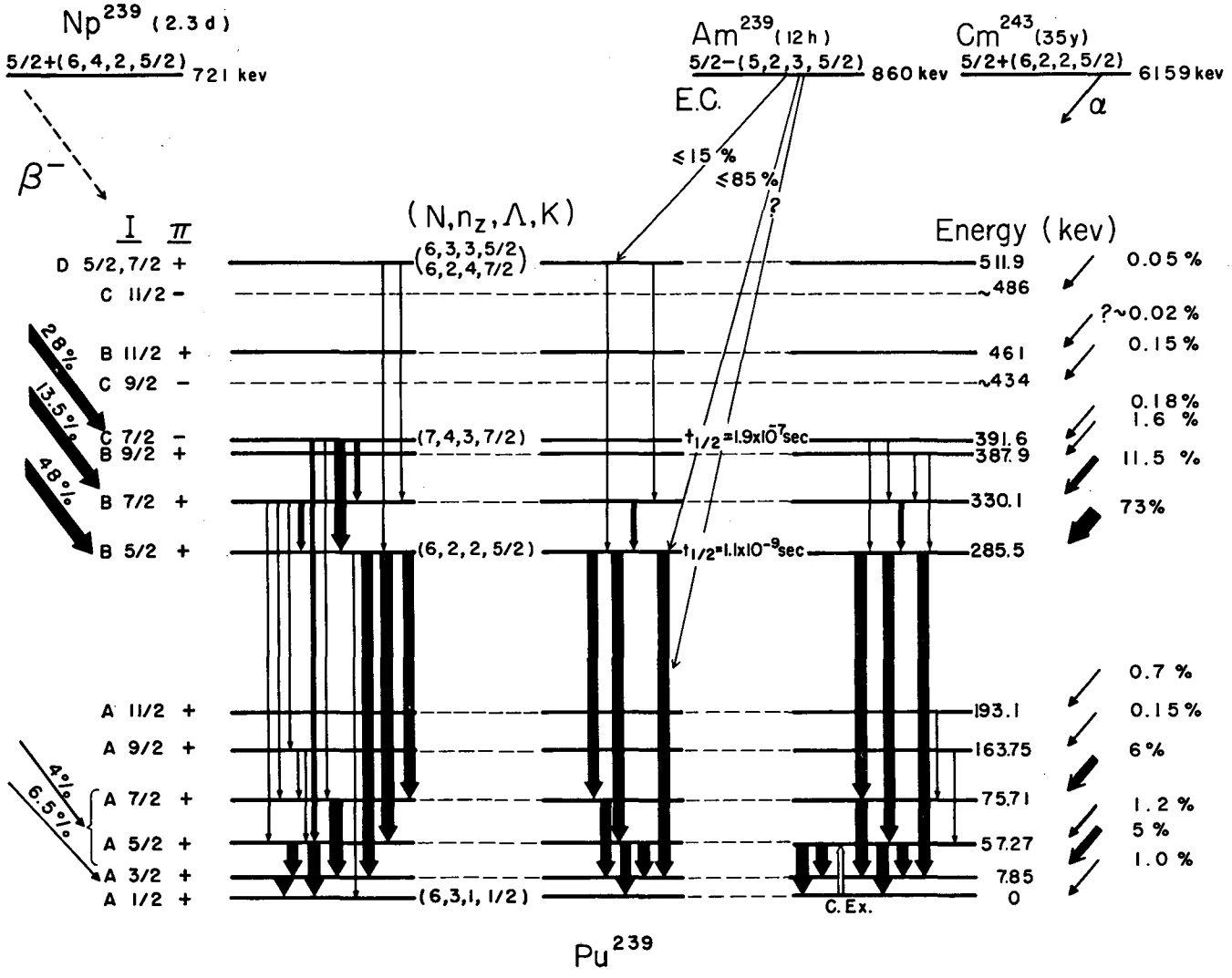

Figure 8. Decay schemes of Np^{239}, Am^{239}, and Cm^{243} shown on a common plot. The Coulombic excitation scheme of Pu^{239} is also shown. All known gamma rays of Np^{239} are placed with the exception of the very weak gamma rays of 440 and 490 kev energy found by Lefevre, Kinderman and Van Tuyl[8] which may originate at the 511.9 kev level.

The vertical arrows in each scheme are drawn only for experimentally observed transitions. Assignments of the asymptotic quantum numbers (N, n_z, Λ, K) are given as well as spins and parities. Rotational levels in a common band are designated by a common letter (A, B, C or D).

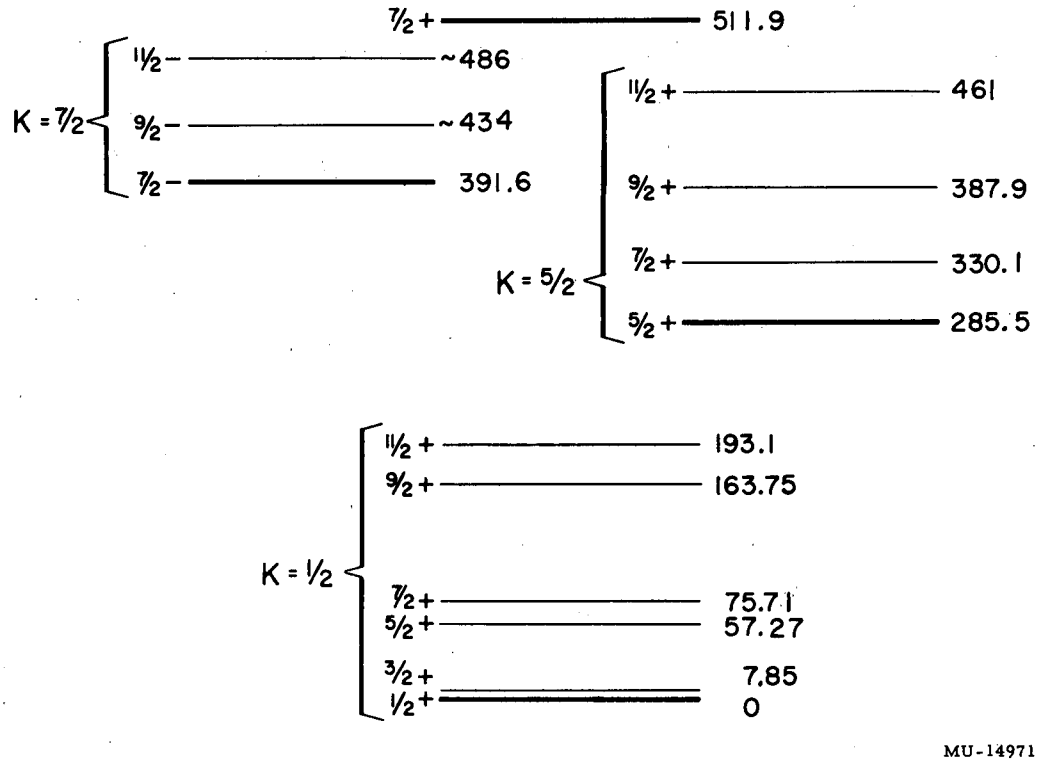

Figure 9. Energy levels of Pu^{239} displayed to emphasize their interpretation as rotational bands based on four intrinsic states.

axis. $\mathcal{J}$ is the moment of inertia of the collective wave motion of the nucleus and I is the angular momentum of a given level. These quantities are discussed in Chapter 3.

Ewan et al.[27] have shown that the simple rotational formula is inadequate to fit the precisely known energies of the ground state rotational bandaand that at least one additional parameter is required. They add a "rotation-vibration" term to the level spacing formula:

$$E_I = \frac{\hbar^2}{2\mathcal{J}}\left[I\ (I+1) + a(-1)^{I+1/2}\ I+1/2\right] - B\left[I(I+1) + a(-1)^{I+1/2}(I+1/2)\right]^2$$

Table 8 shows the experimental and calculated energy levels.

If the values 7.85 kev and 57.27 kev, respectively, are substituted for the energies of the 3/2 and 5/2 levels into the simpler rotational formula given above the value of the rotational splitting constant $\frac{\hbar^2}{2\mathcal{J}}$ turns out to be 6.28 kev which is close to the value, 6.20 kev, found for the K = 5/2 band in Np^{237} but somewhat smaller than the 7.37-kev value found for the K = 0 band in Pu^{238}. This is in accord with the expectation that this quantity for odd A nuclei should be less than for neighboring even-even nuclei. The value of the decoupling parameter a is -0.58 from the above substitution.

In early versions of the decay scheme of Np^{239} the ground state of Pu^{239} was placed 7.85 kev above its designated position in the figures shown here. Later, however, it was found that the most reasonable interpretation[9] of the gamma ray cascades could be made if the location of the ground state was lowered by 7.85 kev. This implied that a transition of 7.85 kev had been overlooked. This assignment was later confirmed by the Coulombic excitation experiments of NEWTON[13], who observed 57.5 kev and 49.6 kev gamma rays (difference 7.8 Kev). BARANOV AND SHLYAGIN[10] directly observed the conversion electrons of the 7.85 kev transition, but these workers interpreted the observed lines as due to M and L conversion lines of a 12.3 kev transition; they are here reinterpreted as N and O conversion lines of the 7.85 kev transition.[19]

The levels labeled B in figure 8 are rotational levels based on an excited state with a K-value different from that of the ground state. The tentative assignment of K is 5/2. The 285.5 kev base level of the K = 5/2 rotational band has a measured half-life of 1.1×10^{-9} seconds. This is interesting since this represents a slowdown from the value calculated from the

Table 8. Energy Levels in Ground State K = 1/2 band, in kev

Level	Theoretical (Simple Rotational Model Formula)[a]	Theoretical (Including Rotation-Vibration Term)[b]	Experimental
1/2	0	0	0
3/2	7.85	7.88	7.85
1/2	57.27	57.37	57.27
7/2	75.59	75.65	75.71
9/2	164.51	163.75	163.75

From Ewan et al.[27]

a. $\frac{\hbar^2}{2\mathfrak{I}}$ = 6.284 kev, a = -0.581

b. $\frac{\hbar^2}{2\mathfrak{I}}$ = 6.290 kev,

a = -0.582, and

b = 0.0024 kev

Weisskopf single-particle formula of about a factor of 10^4 for the M1 transitions to the ground state band. The long half-life of these transitions finds a natural explanation in the fact that transitions between the upper (K = 5/2) and lower (K = 1/2) bands involve a ΔK of two. This violates the K-selection rule which states that ΔK must be equal to or less than the multipolarity of the transition.[14,18]

The level at 391.6 kev has a measured half-life[7] of 1.9×10^{-7} seconds representing a retardation of $\sim 2 \times 10^6$ from the single-proton lifetime estimate for an E1 transition. This slowness is rather commonly observed for E1 transitions in the heavy element region and may be associated with a violation of selection rules in the asymptotic quantum number N_Z or Λ . The experimental data for these anomalous transitions between Pu^{239} levels and for many other E1 transitions in odd mass heavy deformed nuclei is summarized by ASARO, STEPHENS, HOLLANDER AND PERLMAN[24]. (See Chapter 3 for discussion of these quantum numbers). It has been suggested that the long life of the level at 391.6 kev may be attributed to a violation of the selection rule in N_Z since on the basis of reasonable assignments of Nilsson wave functions to the intrinsic particle states ΔN_Z is 3 which is greater than the multipolarity of the transition.

The decay scheme of Np^{239} provides several other opportunities to test the selection rules for beta and gamma transitions for strongly-deformed nuclei. A number of these are discussed in detail elsewhere.[9,18]

The spin of 5/2 is required for the ground state of Np^{239} in order to make a consistent interpretation of the observed transitions. A suitable orbital with $\Omega = 5/2$ is predicted by Nilsson's calculations; specifically the favored assignment is $\Omega\, \pi(Nn_z \Lambda) = 5/2 + (642)$. The log ft values for the beta transitions are readily interpreted[18] using Alaga's selection rules[14] together with the orbital assignments given in the figure; the log ft values definitely cannot be understood in terms of the ordinary selection rules of beta decay. This 5/2 spin value for Np^{239} has been confirmed by the atomic beam measurement of HUBBS AND MARRUS.[17] The value 1/2 obtained earlier by the optical experiments of CONWAY AND MC LAUGHLIN[15] is probably in error. Another early assignment of the spin value 1/2 by a paramagnetic resonance technique[16] has been withdrawn[25] after a restudy of the determination.

The following Nilsson orbital assignments have been made to the

important levels of Pu^{239}. The ground state is 1/2 + (631); the level at 285.5 kev is 5/2 + (622); the level at 391.6 kev is 7/2 - (743) and the level at 511.9 is 7/2 + (624). The arguments on which these assignments are based are covered elsewhere.[18,26]

A close inspection of figure 8 will reveal that Pu^{239} levels at 193.1, 387.9, ~434 and ~486 kev are populated in the α-decay of Cm^{243} but not in the decay of Np^{239}.

References 9 and 27 should be consulted for a more detailed discussion of the radiations of Np^{239} and their interpretation in terms of the unified model.

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9.1.12 Neptunium-240. The isotopes U^{240} and Np^{240} were found by HYDE, STUDIER AND MANNING[1] in natural uranium which had been irradiated in the high neutron flux of one of the Hanford piles. U^{240} was produced by the capture sequence,

$$U^{238}\ (n,\gamma)\ U^{239}\ (n,\gamma)\ U^{240}$$

U^{240} and its daughter, Np^{246} were identified by standard radiochemical techniques and found to be beta-emitters with half lives of 14 ± 2 hours and 7.3 ± 0.3 minutes, respectively. No further characterization of these nuclides were made with these samples because of their low intensity and because of interference from the radiations of U^{237}, which isotope was prepared by a side reaction,

$$U^{238}\ (n,2n)\ U^{237}$$

The most careful study of the radiations of U^{240} and Np^{240} has been made by BUNKER, DROPESKY, KNIGHT, STARNER AND WARREN[2,3] who made use of U^{240} sources of high strength and high specific activity produced by irradiation of U^{238} with the very high instantaneous neutron fluxes available during nuclear weapons testing.

The half life of U^{240} was more precisely determined as 14.1 ± 0.2 hours.

The β-spectrum of the U^{240}-Np^{240} mixture was studied in a β-spectrometer and the observed complex spectrum was analyzed by Fermi plots into the groups shown in Table 9. From the gamma ray studies and the decay scheme to be discussed below it is certain that these observed β groups contain unresolved components. The right hand side of the table shows a postulated breakdown of the Np^{240} β spectrum into its primary components. An intense low-energy component of the U^{240}-Np^{240} mixture with an end-point energy of 0.36 ± 0.02 Mev was assigned to U^{240}.

BUNKER, DROPESKY, KNIGHT, STARNER AND WARREN[3] studied the conversion electron spectrum in 180° permanent magnet spectrographs and in a uniform-field, ring-focusing solenoidal spectrometer. They studied the γ-ray photons in a NaI scintillation spectrometer. γ-γ coincidence measurements were made with two NaI crystal spectrometers in coincidence. β-γ coincidences were also studied. ASARO[5] and his co-workers also studied the radiations of Np^{240}. We first tabulate the principal results, then present the derived decay scheme and discuss its interesting features.

Tables 10 and show the conversion electron lines seen in the 180° permanent magnet spectrographs by ASARO[4] and by BUNKER[3]. Table 4 shows con-

Table 9. Beta-ray transitions of Np^{240}

Observed groups[a]		Postulated transitions[b]			
End-point energy (Mev)	Intensity (%)	End-point energy (Mev)	Intensity (%)	Log ft	I, π final state
2.18±0.02	52±3	2.18	41 ±5	6.68	0+
		2.14	12 ±4	7.18	2+
1.60±0.03	31±2	1.60	32 ±2	6.28	1-
1.30±0.05	10±1	1.32	3.9±0.5	6.86	0+
		1.28	2.3±0.3	7.05	2+
		1.24	3.3±0.5	6.86	(2+)
0.65±0.10	7±3	0.76	0.3±0.1	7.15	(1±,2±)
		0.65	3.8±0.5	5.83	(1,2)+
		0.57	0.7±0.1	6.33	(1±,2+)

[a] Deduced from Fermi-Kurie analysis of the β spectrum.

[b] Deduced from analysis of γ-ray spectra.

Table 10. Conversion electron lines from 7.3 minute Np^{240} (as observed by Asaro et al.[4])

Electron Energy (Kev)	Conversion Shell (Pu)	Transition Energy (Kev)	Relative electron intensity (Visual estimate)
20.6	L_{II}	42.9	Strong
24.8	L_{III}		
435.3	K	557	100
	L_I		10
477.7	K	599.5	50
576.4	L_I		
638.3	K	760.1	
670.1	(K)	(791.9)	
722.6	(K)	(844.4)	
738.1	K	859.9	40
836.8	L_I		4
740.9	(K)	(862.7)	20
823.6	(K)	(945.4)	8
969.3	(K)	(1091.9)	4

Table II. Internal-conversion electron lines as measured on Permanent Magnet Spectrograph by Bunker.

Electron Energy (Kev)	Conversion Shell	Transition Energy (Kev)	Relative intensity (visual estimate)
20.6	L_{II}(Pu)	42.9	~ 2
21.6	L_{I}(Np)	44.0	~ 2
22.4	L_{II}(Np)	44.0	~0.5
24.8	L_{III}(Pu)	42.9	~ 2
38.3	M_{III} Pu	42.9	~ 1
	M_{I} Np	44.0	

Table 12. Internal-conversion electron data obtained by Bunker et al.[3] with a beta-ray spectrometer.

Electron energy (Kev)	Assigned shell	Transition energy (Kev)	Intensity[a] x 10^4
432.4	K	554.1	19.6±1.5
530.7	L_I	553.8	3.8±0.7
474.8	K	596.5	9.8±1.0
573.3	L_I	596.4	2.0±0.6
636.5	K	758.2	1.8±0.6
694.3	K	816.0	2.2±0.6
736.2	K	857.9	14.0±1.2
835.4	L_I	858.5	2.7±0.6
776.0	K	897.7	1.5±0.7
819.9	K	941.6	3.5±0.7

[a] Area of the conversion line relative to the area of the total Np^{240} β-ray spectrum.

version electron data taken by the latter authors in a beta-spectrometer and Table 13 shows their summary of the data and assignments for all the gamma ray transitions of 7.3 minute Np^{240}.

From these data and from coincidence data and detailed arguments not reproduced here the decay scheme of figure 10 was constructed. Pu^{240} is an even-even nucleus and like all other even-even nuclei in this mass region has for its first levels of excitation a rotational band of levels with a familiar 0+, 2+, 4+ .... sequence and the familiar energy spacing proportional to I (I+1). This is very well established in the case of Pu^{240} because the 2+ and 4+ levels have been observed in the electron capture decay of Am^{240}. (see section 9.3.5) and the 2+, 4+ and 6+ levels have been seen in the α-decay of Cm^{244} (see section 9.3.8). The 4+ and 2+ levels are de-excited by E2 transitions with energy 98.9 and 42.9 kev energy. These are almost completely converted in the L_{II} and L_{III} shells so that these transitions are revealed chiefly through the electrons and through L x-rays. These abundant L x-rays are very useful in fixing the placement of the higher-energy gamma rays in the decay scheme through a consideration of L x-ray versus γ-ray coincidence spectra. γ-rays which decay directly to ground do not show an L x-ray coincidence, while those which populate the 42.9 kev level show a strong L x-ray coincidence.

At 597 kev there occurs a 1- level which is strongly populated in the β-decay of Np^{240}. This 1- level is believed to represent collection oscillations of the Pu^{240} nucleus of the octupole type. Such 1- collective states have been observed at about this energy above ground for several other even-even nuclei. See discussion in chapter 3. The spin and parity are firmly fixed by the proved E1 character of the 597 and 554 kev gamma rays which de-excite it. The ratio of the reduced transition probabilities of these two transitions is 0.47 ± 0.05.

$$\frac{B\ (E1;\ 1- \longrightarrow 0+)}{B\ (E1;\ 1- \longrightarrow 2+)} = 0.47$$

This fixes the K-quantum number of the 1- level as K = 0.

The level at 858 has spin and parity 0+ with near certainty because of the occurrence of the completely-converted E0 transition which goes directly to ground. This level is believed to be a vibrational state of the β-classification discussed in chapter 3. ($n_\beta = 1$, $n_\gamma = 0$ and K = 0). Such states have been

Table 13. Transitions in Pu^{240} which follow the decay of 7.3-min Np^{240}. As reported by Bunker et al.[3]

Energy (Mev)	Transition	Photon intensity[a]	K conversion coefficient x 10^2 Exptl.	K conversion coefficient x 10^2 Theor.[b]	Assigned multipolarity	Total estimated transition intensity[a]
0.0429	BA				E2	39 ±5
0.0989[c]	CB				E2	1.3 ±0.2
0.260	ED	1.9 ±0.3		4.3	E1	2.0 ±0.3
0.304	FD	0.9 ±0.2		3.1	E1	0.9 ±0.2
0.554	DB	21.4 ±1.5	0.92±0.09	0.94	E1	21.6 ±1.5
0.597	DA	12.6 ±1.4	0.78±0.12	0.81	E1	12.7 ±1.4
0.758	FC	1.3 ±0.2	1.4 ±0.5	1.50	E2	1.3 ±0.2
0.816	EB	1.6 ±0.3	1.4 ±0.5	1.32	E2	1.6 ±0.3
0.82	HD	0.3 ±0.1		(0.45)	(E1)	0.3 ±0.1
0.858	EA	<0.17	>80		E0	0.14±0.01
0.898	GB	1.2 ±0.3	1.2 ±0.7	1.16	E2	1.2 ±0.3
0.936	ID	0.3 ±0.1		(0.36)	(E1)	0.3 ±0.1
0.942	GA	1.9 ±0.5	1.8 ±0.7	0.94	E2	1.9 ±0.5
1.49	IB	1.5 ±0.3		(0.46)	(E2)	1.5 ±0.3
1.53	IA	1.9 ±0.5		(0.44)	(E2)	1.9 ±0.5
1.62	JA	0.7 ±0.1		(0.40)	(E2)	0.7 ±0.1

[a] Per 100 disintegrations.

[b] Values obtained from the tables of L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57 ICC K1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)] for the multipolarities shown in the next column.

[c] Not observed in the present experiments.

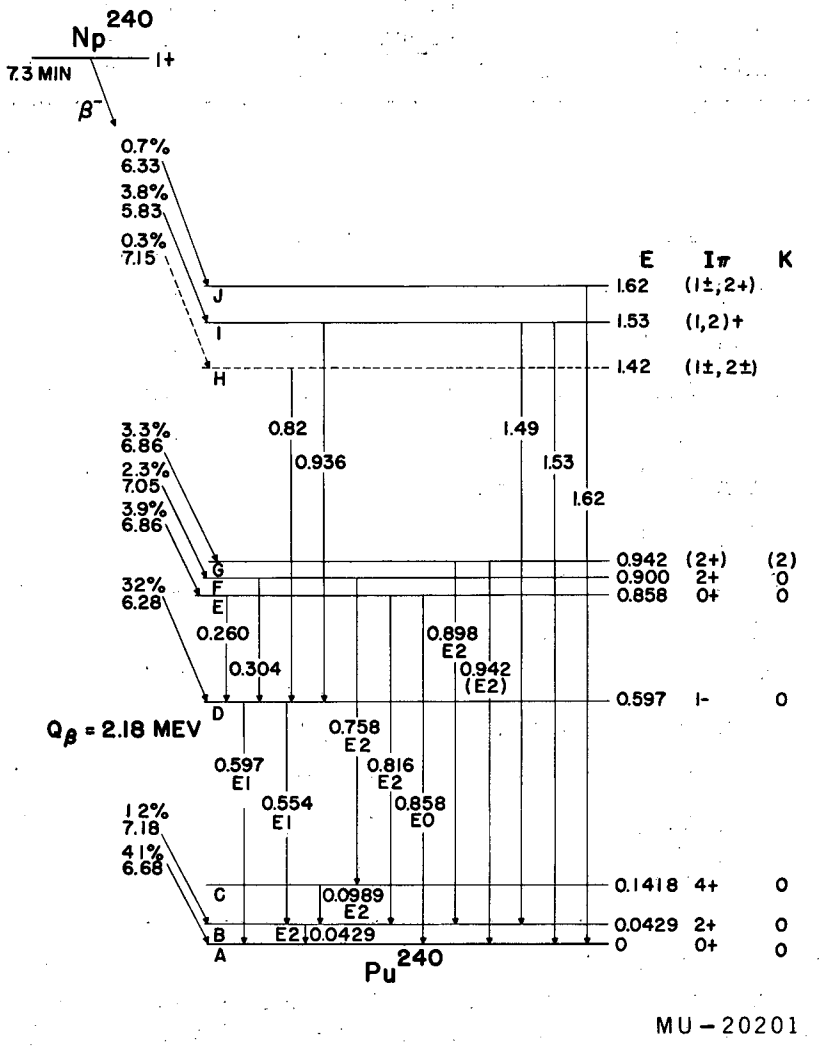

Figure 10. Decay scheme of 7.3 minute Np^{240} (as drawn by Bunker, Dropesky, Knight, Starner and Warren).

observed at about this level of excitation in Pu^{238} and U^{234}. With considerably less certainty the level at 900 kev is given the spin and parity 2+ and assigned as the first rotational state in a band of rotational levels based on the 0+ level at 858 kev.

The level at 942 is given the tentative assignment of 2+ and may be the first Bohr-Mottelson γ-vibrational level with quantum numbers $n_\beta = 0$ $n_\gamma = 1$ and $K = 2$.

The experimental evidence does not point unambiguously to a definite spin and parity for any one of the upper three levels (H, I and J). The possible choices are shown in the figure.

The beta spectrum of U^{240} as resolved with some difficulty from the β and conversion electron spectrum of the U^{240} - Np^{240} - U^{237} mixture has an end-point energy of 0.36 Mev. The occurence of electron lines of a 44 kev transition converted in neptunium rather than plutonium L and M electron shells (see table11) means that the U^{240} beta spectrum contains two components. BUNKER AND CO-WORKERS[3] estimated that 25 percent of the U^{240} betas decay to a 44 kev level in Np^{240} (log ft = 6.0) while 75 percent decayed to the ground state (log ft = 5.7). The log ft value suggests a spin and parity assignment for 7.3 minute Np^{240} of 1+. The most reasonable Nilsson orbital assignment for the odd proton in Np^{240} is 5/2+[642] . A possible choice for the odd neutron is 7/2 + [624]. These choices are tentative. A final settlement on the Nilsson states for Np^{240} will have to take into account and explain the existence of the 63 minute isomer discussed in the next section. This isomer is not observed in the decay of U^{240} and presumably has a spin $\geq$ 2.

References Np^{240}

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9.1.13 Isomer of Neptunium-240. ORTH AND STREET[1] produced a 60 ± 2 minute beta-emitting activity with the chemical properties of neptunium by bombarding U^{238} with 35 Mev helium ions. This activity was not produced in proton or deuteron bombardments of U^{238}; hence this 60 minute neptunium activity can be assigned to mass number 240 or 241. ORTH AND STREET[1] tentatively chose Np^{241} but later work of LESSLER AND MICHEL[2] favored Np^{240}. These latter authors carried out a mass separation of neptunium activities produced in a helium ion bombardment of U^{238} using a mass spectrometer based on a time-of-flight principle. If this assignment is correct the 60 minute activity is an isomer of 7.3 minute Np^{240}.

RITSEMA[3] determined the excitation function for the production of 60-minute neptunium from U^{238} bombarded with helium ions. LESSLER[5] determined an excitation function for neptunium produced by the (d,γ) reaction on U^{238}. LEFEVRE, KINDERMAN, AND VAN TUYL[6] produced both Np^{240} isomers by bombarding Np^{239} with pile neutrons and set an upper limit of < 5% for the genetic linkage between the two isomers.

Measurements with a beta-ray spectrometer[1] showed a beta spectrum of upper energy limit 0.89 ± 0.03 Mev and conversion electrons corresponding to gamma rays of energies 0.15, 0.20, 0.26 and 0.58 Mev. Gamma ray measurements by LESSLER AND STEPHENS[4] with a NaI scintillation spectrometer showed gamma rays of energies 1160, 1000, 915, 580 and 435 kev, but additional gamma rays of less than 350 kev were obscured by the Np^{239} background. By gamma-gamma coincidence techniques LESSLER AND STEPHENS[4] found additional gamma rays of 160, 245 and 85 kev. The 580-kev gamma ray was shown to be a complex with components of 595 and 565 kev in coincidence with each other. The most energetic beta particle group had an end point energy of 900 kev and is in coincidence with 1160 kev gamma radiations. Further work remains to be done before the decay scheme of the 60-minute isomer and the relationships between the isomers can be written with certainty. LESSLER estimated the decay energy as 2.06 Mev; this is consistent with the mass assignment since from closed decay-energy cycles one would estimate only 1.32 Mev for the decay energy of Np^{241}, whereas over 2 Mev is available in the case of Np^{240}.

References for 60-minute Np^{240}

1. D. A. Orth and K. Street, Jr., unpublished data (1951).
2. R. Lessler and M. C. Michel, University of California Radiation Laboratory Quarterly Report, UCRL-2709 (September 1954), unpublished, and Phys. Rev. 118 263 (1960.)
3. S. E. Ritsema, University of California Radiation Laboratory Report, UCRL-3266 (January 1956); also included in Phys. Rev. 111, 1358 (1958).
4. R. Lessler and F. S. Stephens, Jr., University of California Radiation Laboratory Quarterly Reports, UCRL-2531 (April 1954), and UCRL-2647 (July 1954). See also reference 2.
5. R. M. Lessler, University of California Lawrence Radiation Laboratory Report, UCRL-8439 (October 1955).
6. H. W. Lefevre, E. M. Kinderman and H. N. van Tuyl, Bull. Am. Phys. Soc. II, No. 1, 62 (1956).

9.1.14 Neptunium 241. LESSLER AND MICHEL[1,2] identified a 16 minute beta-emitter in the mass 241 fraction of a neptunium sample carefully isolated from a U^{238} target bombarded with 35 Mev helium ions. The mass separation was performed on a time-of-flight isotope separator. In addition, another beta activity of 3.4 hours half-life was found in the 241 fraction and assigned tentatively to an isomeric form of Np^{241}.

VANDENBOSCH[3] confirmed the existence of a 16 minute species of neptunium of high mass by isolating and identifying such an activity from an uranium target bombarded with helium ions.

$$U^{238}\ (\alpha,p)\ Np^{241}$$

The 60 minute Np^{240} and 2.3 day Np^{239} were also produced in this bombardment but no 3.4 hour neptunium activity was seen (upper limit to production cross section 0.2 mb). The beta spectrum of the 16 minute Neptunium activity was studied with an anthracene crystal-photomultiplier tube detector. The end point of the beta spectrum was 1.36 Mev. No gamma rays or K x-rays were observed in a NaI crystal spectrometer which could be attributed to the 16 minute neptunium activity. Hence the 1.36 Mev beta group may represent decay to the ground state of Pu^{241}. This is consistent with the 1.32 Mev decay energy

estimated for Np^{241} from closed decay-energy cycles. (see chapter 2). The log ft value is 5.8 on this assumption.

The measured spins of Np^{237} and Np^{239} are both 5/2 and the Nilsson orbital assignments with considerable certainty are 5/2+ [642] for the ground states of these two nuclei. Hence one might predict the same spin and orbital assignment for the ground state of Np^{241}.

References Np^{241}

1. R. M. Lessler and M. C. Michel, University of California Radiation Laboratory Report, UCRL-2709, September 1954, unpublished, and Phys. Rev. 118 263(1960.)
2. R. M. Lessler, Ph.D. thesis, University of California, October 1958; also available as report UCRL-8439, October 1958, unpublished.
3. R. Vandenbosch, Phys. Rev. 113, 259, (1959).

9.2 THE ELEMENT PLUTONIUM (ELEMENT 94)

9.2.1 The Discovery and Early History of Plutonium. MC MILLAN AND AND ABELSON[1] found that Np^{239}, formed by the beta decay of U^{239}, underwent radioactive decay by the emission of beta particles, but they were unable to detect the 94^{239} daughter in their samples because of the long half-life of this daughter isotope. The first isotope of element 94 to be identified was Pu^{238} produced in the experiments of SEABORG, MC MILLAN, KENNEDY AND WAHL[2] by the bombardment of uranium with 16-Mev deuterons in the University of California 60-inch cyclotron:

$$U^{238}\ (d,2n)\ Np^{238} \xrightarrow[2.1\ days]{\beta^-} Pu^{238}$$

The isotope Pu^{238} was shown to emit alpha particles with a half-life of approximately 50 years (best present value = 86.4 years). The discoverers[7] later proposed the name plutonium (symbol Pu) taken from the planet Pluto, at that time believed to be the second planet beyond Uranus in our solar system.

In 1941, KENNEDY, SEABORG, SEGRÈ AND WAHL[3] isolated and identified Pu^{239} as the decay product of a very intense source of Np^{239}. A target of about 1.2 kilograms of uranyl nitrate hexahydrate was distributed in a large paraffin block and irradiated with the neutrons produced when a beryllium target was struck with 16 Mev deuterons from the Berkeley 60-inch cyclotron. It was necessary to bombard for a total of 3500 microampere-hours of deuteron current to obtain a sufficiently active sample of Np^{239}, about 125 millicuries. This was isolated by the fluoride coprecipitation method of MC MILLAN AND ABELSON[1] to free it from all impurities. Alpha particles were observed to grow into this neptunium sample with a growth half-life of 2.3 days. From the number of alpha particles which grew in, the half-life of Pu^{239} was estimated to be 30,000 years. (The presently accepted value is 24,400 years.) These experimenters used this sample of Pu^{239} (about 0.5 micrograms) to investigate the slow neutron fissionability of this isotope. It was established that Pu^{239} undergoes fission with slow neutrons and that the fission cross section is somewhat larger than that of U^{235}. This measurement immediately gave this isotope great importance because it meant that if Pu^{239} could be produced and isolated in appreciable

quantities, it might possibly be used in the release of huge amounts of nuclear energy. It was recognized that the slow neutron fission of large amounts of U^{235} in a suitably designed assembly might provide a neutron source of sufficient strength to allow the conversion of appreciable quantities of natural uranium to Pu^{239} via the neutron capture reaction:

$$U^{238}\ (n,\gamma)\ U^{239} \xrightarrow[23\ min]{\beta^-} Np^{239} \xrightarrow[2.3\ days]{\beta^-} Pu^{239}$$

On the strength of this hope, the secret plutonium project was set up by the Uranium Committee, an agency of the United States Government, established just before World War II to explore the possibility of utilizing the energy of the fission reaction for military purposes. During 1941 and the early part of 1942 cyclotron-produced Pu^{238} was used at the University of California to investigate the chemical properties of plutonium by the tracer method. Following this initial work, the investigation of plutonium was taken up by other laboratories. Chief among these were the Metallurgical Laboratory of the University of Chicago, where the program got underway during the first half of 1942, the Los Alamos Laboratory in New Mexico, where work began early in 1943, the Clinton Laboratories in Oak Ridge, Tennessee, which opened late in 1943, and the Hanford Engineer Works in Washington, which began operation in the latter half of 1944. The goal of these investigations was to design a chemical process workable on an industrial scale for the isolation of plutonium from uranium fuel elements irradiated in a chain-reacting pile. Other groups of investigators at these same laboratories were concerned with the demonstration of the feasibility of a chain-reacting pile and the design of the large reactors ultimately constructed at Hanford, Washington.

The organizational and administrative history of this effort is recorded in the SMYTH REPORT[4]. The scientific findings of these American laboratories on the nuclear and chemical properties of plutonium and other transuranium elements are detailed in the volumes of the National Nuclear Energy Series - Plutonium Project Record.[5] SEABORG[11] has traced the history of the wartime developments in plutonium isolation from the Hanford pile fuel elements in another volume. Very considerable contributions by Canadian workers at their Chalk River Laboratory are described in other publications.

Prior to the establishment of the organized attack on plutonium chemistry some of the important features of the chemical behavior of plutonium were established by SEABORG, WAHL AND KENNEDY[6] and SEABORG AND WAHL[7] working at the University of California. Plutonium could be carried from acid solution on rare earth fluorides, on thorium fluoride, on the peroxyhydrate of thorium, and on insoluble iodates. In basic solution it would coprecipitate on insoluble hydroxides. However, in the presence of a powerful oxidizing agent such as peroxydisulfate ion the plutonium tracer could be oxidized to a higher oxidation state which did not coprecipitate with insoluble fluorides in acid solution. In order to estimate oxidation-reduction potentials, a number of oxidizing and reducing agents were tested for their ability to oxidize or reduce tracer plutonium. An important difference between neptunium and plutonium in trace concentrations is the faster rate of oxidation of neptunium to the fluoride-soluble higher oxidation state by bromate ion at room temperature. Neptunium is quantitatively oxidized in 20 minutes whereas plutonium is not and can be removed from solution by precipitation of an insoluble fluoride such as lanthanum fluoride. This bromate-lanthanum fluoride cycle was a mainstay of neptunium and plutonium separation for some time to come.

SEABORG AND WAHL[7], in the first detailed paper on the chemistry of plutonium, emphasized the conclusion that plutonium was a member of a new rare earth-like series which included neptunium and uranium. It was regarded as an open question whether this series began with actinium, thorium or uranium.

Very soon after the discovery of plutonium, G. T. SEABORG AND M. PERLMAN[8] made a search for Pu^{239} in natural sources. Their work and the later work of others which is described in the section entitled "The Natural Occurrence of Transuranium Elements in Trace Amounts" in the chapter on the Natural Radioactivities shows that traces of Pu^{239} are present in minerals containing uranium resulting from the action of naturally occurring neutrons on U^{238}.

During 1942, when the main effort to develop a process for plutonium isolation was centered at the Metallurgical Laboratory, a process which later came to be known as the bismuth-phosphate process, was originated by the tracer scale experiments of S. G. THOMPSON.[12] Since the chemical behavior of an element on the tracer scale and the macroscopic scale is not always the same, it was important to test the behavior of plutonium at the concentrations expected in the chemical plant. At this time only microgram quantities of plutonium were available and hence such experiments had to be done by ultramicrochemical techniques. P. Kirk was responsible for the development of many of the methods and much of the specialized equipment employed in these studies. The first chemically pure plutonium, free from carrier material and all other foreign

matter, was prepared by CUNNINGHAM AND WERNER[9] in August, 1942. This was the first isolation of any synthetic element. The half-life of plutonium was determined by these authors by direct weighing of a few micrograms of plutonium oxide and direct counting of the alpha particle emission rate. A value of 24,300 years was obtained. By direct analysis of a 1.7 microgram sample of plutonium iodate using microscale analytical methods CUNNINGHAM AND WERNER established the formula $Pu(IO_3)_4$. This was the first direct chemical proof of any of the oxidation states of plutonium.

A year or two later plutonium became available in milligram quantities from the operation of the Clinton, Tennessee and Hanford, Washington reactors and it was possible to initiate chemical studies by ordinary bench-top techniques (albeit modified somewhat because of the physiological hazard). The chemistry of plutonium on the tracer scale and at ordinary concentrations is summarized by KATZ AND SEABORG[13] and by HYDE AND SEABORG.[14]

For a few years after the discovery of plutonium the only isotopes available for study were Pu^{238} and Pu^{239}; then the isotope Pu^{240} was found by CHAMBERLAIN, FARWELL AND SEGRÈ[10] in samples of Pu^{239} subjected to long neutron bombardment. Cyclotron bombardment of uranium targets with high-energy helium ions resulted in the identification of Pu^{237}, Pu^{236} and lighter isotopes. When higher-flux piles were built the production of a number of higher isotopes of plutonium by multiple neutron capture reaction became feasible. Some of the plutonium isotopes of very high mass number were first identified during nuclear weapons test programs. In the following pages the individual isotopes of plutonium will be discussed in detail.

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1. E. M. Mc Millan and P. Abelson, Phys. Rev. 57, 1185 (1940).
2. G. T. Seaborg, E. M. Mc Millan, J. W. Kennedy and A. C. Wahl, Phys. Rev. 69, 366 (1946); G.T.Seaborg, A.C. Wahl and J.W.Kennedy, Phys. Rev. 69, 367 (1946). Original reports written early in 1941.
3. J. W. Kennedy, G. T. Seaborg, E. Segrè, and A. C. Wahl, Phys. Rev. 70, 555 (1946) Original report written in May 1941.
4. H. D. Smyth, "Atomic Energy for Military Purposes." Princeton University Press, Princeton, New Jersey, 1945.
5. The National Nuclear Energy Series - Plutonium Project Record (McGraw-Hill

Book Co., Inc., New York) Volume 14A "The Actinide Elements" G. T. Seaborg and J. J. Katz, editors, 1954.
Volume 14B "The Transuranium Elements" G. T. Seaborg, J. J. Katz and W. M. Manning, editors, 1949.

6. G. T. Seaborg, A. C. Wahl and J. W. Kennedy, Phys. Rev. 69, 367 (1946).
7. G. T. Seaborg and A. C. Wahl, Paper No. 1.6, page 25, "The Transuranium Elements", National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Co., Inc., New York, 1949. Original report written March, 1941.
8. G. T. Seaborg and M. L. Perlman, Paper No. 1.3, "The Transuranium Elements," National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Company, Inc., New York, 1949.
9. B. B. Cunningham and L. B. Werner, Paper No. 1.8, "The Transuranium Elements," National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Co., Inc., New York, 1949; see also J. Am. Chem. Soc. 71, 1521 (1949).
10. O. Chamberlain, G. Farwell and E. Segre, unpublished information, 1944.
11. G. T. Seaborg, "The Transuranium Elements" Silliman Lectures, Yale University Press, 1958.
12. For a description of the historical bismuth phosphate process for plutonium isolation see a paper by S. G. Thompson and G. T. Seaborg in Progress in Nuclear Energy, Ser. 3, Vol. 3 "Process Chemistry", edited by F. R. Bruce, J. M. Fletcher, H. H. Hyman and J. J. Katz, Pergamon Press, Ltd. 1956.
13. J. J. Katz and G. T. Seaborg, "The Chemistry of the Actinide Elements" Wiley and Sons, New York, 1957.
14. E. K. Hyde and G. T. Seaborg, "The Transuranium Elements" in Volume 42, Handbuch der Physik, S. Flügge, editor, Springer-Verlag, Berlin, 1957.

9.2.2 Plutonium-232. This isotope was first produced by ORTH AND STREET[1] who bombarded uranium targets enriched in U^{235} to make it by the reaction:

$$U^{235}\ (\alpha,7n)\ Pu^{232}$$

The maximum yield occurred when the energy of the helium ions was 110 Mev. At such an energy several other isotopes of plutonium are produced and pure samples of Pu^{232} can not be made. Pu^{232} has also[2] been prepared by the reaction:

$$U^{233}\ (\alpha,5n)\ Pu^{232}.$$

The half-life of Pu^{232} is 36 minutes. The alpha decay of Pu^{232} produces 9.3 minute U^{228} which in turn decays through a series of short-lived alpha emitting descendents; this U^{228} collateral series is discussed in Chapter 7 (Section 7.2.7). The growth of this series of isotopes into the plutonium fraction is strong evidence for the presence of Pu^{232}. The U^{228} can be separated chemically, and observed to decay with a 9.3-minute half-life. The alpha particle energy of Pu^{232} was found to be 6.58 Mev in a gridded ion chamber; the resolution of this peak from the 6.67-Mev peak of U^{228} presents some difficulty.

The isotope Pu^{232} is very far on the neutron-deficient side of beta stability and decays also by capture of an orbital electron although it is quite difficult to establish this mode of decay by direct measurement of x-rays because of the presence of Pu^{235} and Pu^{234} in the available samples. Attempts were made to determine the amount of electron capture by separating the 13 minute Np^{232} daughter but no positive results were obtained. An upper limit of 60 was placed on the EC/α branching ratio.[1]

REFERENCES - Pu^{232}

1. D. A. Orth, Ph.D. Thesis, University of California, January 1951 (unpublished); also published as University of California Radiation Laboratory Report UCRL-1059 (Rev.) March, 1952.
2. T. D. Thomas, R. Vandenbosch, R. A. Glass and G. T. Seaborg, Phys. Rev. 106, 1228 (1957).

9.2.3 Plutonium-233. The isotope Pu^{233} was discovered years after the first identification of Pu^{232}, Pu^{234} and Pu^{235} even though it is always produced when Pu^{232} is made and frequently produced when Pu^{234} or Pu^{235} is made. This delay was due to the fact that the radiations of Pu^{233} are obscured by those of the other plutonium isotopes and are unobserved unless careful experiments are performed. THOMAS, VANDENBOSCH, GLASS AND SEABORG[1] identified Pu^{233} in the plutonium fraction from targets of U^{233} bombarded with helium ions of 40-45 Mev energy:

$$U^{233}\ (\alpha,4n)\ Pu^{233}.$$

Figure 11 shows a pulse-height analysis of the output of a gridded ionization chamber used to examine the alpha activity of the mixture of plutonium isotopes produced in such a bombardment. (The Pu^{239} was added to the target after the end of the bombardment to serve as a monitor of the chemical yield.) An alpha particle group was regularly observed at 6.3 Mev as a shoulder on the high energy side of the Pu^{234} alpha peak. This 6.30-Mev activity, which decayed with a 20 ± 2 minute half-life, was assigned to Pu^{233}.

The mass assignment is based on four lines of evidence: (1) the change in the yield of the new activity with the helium ion energy is in agreement with that expected for an $(\alpha,4n)$ reaction, (2) the identification of U^{229} daughter activity, (3) agreement of the alpha-particle energy and half-life with the values expected from alpha-decay systematics, (4) the identification of Np^{233} daughter activity.

The Np^{233} daughter activity was isolated quantitatively and the x-radiation of this 35-minute isotope was counted. From the amount of Np^{233} isolated the alpha-decay branching of Pu^{233} was set at $(1.2 \pm 0.5) \times 10^{-3}$, the major mode of decay being orbital electron capture. The partial alpha half-life is 11 ± 4 days.

Experimental difficulties make difficult a more detailed study of the decay scheme of Pu^{233}.

REFERENCES-Pu^{233}

1. T. D. Thomas, R. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. 106, 1228 (1957).

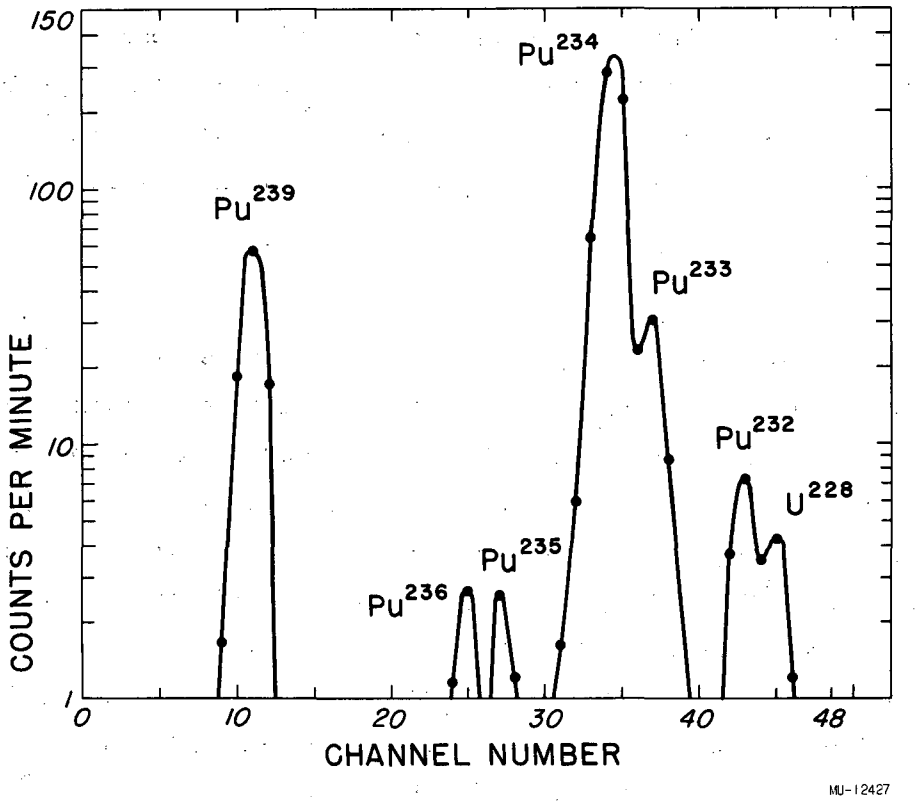

Figure 11. Alpha spectrum of plutonium fraction isolated from a U^{233} target bombarded with 45.2 Mev helium ions.

9.2.4 Plutonium-234. HYDE, STUDIER AND GHIORSO[1] produced Pu^{234} by bombardment of U^{233} with 40 Mev helium ions:

$$U^{233}(\alpha,3n)\ Pu^{234}$$

The isotope was shown to decay partially by the emission of alpha particles of 6.0 Mev energy to produce U^{230} and partially by orbital electron capture to produce Np^{234}; the observed half-life was 8 hours. The U^{230} and the alpha-emitting daughters of U^{230} had previously been well characterized (see the discussion in Section 7.3, Chapter 7) so that identification of such isotopes as U^{230} and Th^{226} could be made with certainty. PERLMAN, O'CONNOR AND MORGAN[2] extended these measurements using larger samples and revised the half-life to 8.5 hours, the alpha-particle energy to 6.2 ± 0.1 Mev. They isolated and identified the 4.40-day Np^{234} produced by orbital electron capture. ORTH AND STREET[3] made additional experiments on Pu^{234} produced by the above reaction and also prepared it from U^{235} as follows:

$$U^{235}(\alpha,5n)\ Pu^{234}$$

These authors report a half-life of 9.0 ± 0.5 hours and an alpha-particle energy of 6.19 ± 0.01 Mev.

HOFF AND ASARO[4] have shown that 25 percent of the alpha decay populates a level at 47 kev in U^{230}. They have further found that there are 0.09 alpha disintegrations per K-electron capture and 0.3 L-electron captures per K-electron capture. From these figures and an estimated value of the amount of electron capture from the M shell and higher shells, it is possible to calculate a ratio of electron capture decay to alpha particle emission of 16, corresponding to a partial alpha half-life of 6 days. The electron capture decay goes directly to the ground state[6] of Np^{234}. For a calculated decay energy of 460 kev this corresponds[6] to a log ft value of 5.6.

Pu^{234} has been observed as the daughter of 2.5-hour Cm^{238} by HIGGINS.[5]

REFERENCES - Pu^{234}

1. E. K. Hyde, M. H. Studier, and A. Ghiorso, Paper No. 22.15, "The transuranium Elements", National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Company, Inc., New York, 1949.

2. I. Perlman, P. R. O'Connor, and L. O. Morgan, Paper No. 22.30, "The Transuranium Elements", National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Company, Inc., New York, 1949.
3. D. A. Orth, Ph. D. Thesis, University of California, January, 1951; also published as University of California Radiation Laboratory Report, UCRL-1059 (Rev.), March 1952. D. A. Orth and K. Street, Jr., unpublished results.
4. R. W. Hoff and F. Asaro, unpublished data, University of California, 1956.
5. G. H. Higgins, Ph.D. Thesis, University of California, June, 1952; also published as University of California Radiation Laboratory Report, UCRL-1796, June 1952.
6. R. W. Hoff and S. G. Thompson, Phys. Rev. 96, 1350 (1954).

9.2.5 Plutonium-235. ORTH AND STREET[1] found that a 26 ± 2-minute plutonium isotope was produced by helium-ion bombardments of U^{233} together with 8.5-hour Pu^{234} and 2.7-year Pu^{236}. The best relative yield of the 26-minute plutonium isotope occurred when the helium ion energy was in the range 28-30 Mev. The most reasonable mass assignment is Pu^{235}. It is possible to produce Pu^{235} from U^{235} targets as well. The reactions are:

$$U^{233}\ (\alpha, 2n)\ Pu^{235}$$
$$U^{235}\ (\alpha, 4n)\ Pu^{235}$$

From the work of ORTH AND STREET[1] the radiations appear to consist primarily of L x-rays, a small amount of harder electromagnetic radiation and few, if any, conversion electrons of energy greater than 100 kev. Alpha activity of energy 5.85 ± 0.03 Mev was observed in amounts corresponding to about 0.002 percent branching decay by this process.

Further work on the radiations of Pu^{235}, done by THOMAS, VANDENBOSCH, GLASS AND SEABORG[2], has confirmed the half-life and alpha particle energy measured by ORTH AND STREET. By measuring the K x-rays accompanying the electron capture and using an estimated value of 0.23 for the ratio of L-electron to K-electron capture, these authors were able to calculate an alpha branching ratio of $(3.0 \pm 0.6) \times 10^{-5}$, corresponding to a partial alpha half-life of 1.7 ± 0.4 years.

REFERENCES - Pu^{235}

1. D. A. Orth, Ph. D. Thesis, University of California, January, 1951; also published as University of California Radiation Laboratory Report, UCRL-1059 (Rev.) March 1952. D. A. Orth and K. Street, unpublished results, 1952.

2. T. D. Thomas, R. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. 106, 1228 (1957).

9.2.6 Plutonium-236. JAMES, FLORIN, HOPKINS AND GHIORSO[1] observed, in the plutonium fraction from the bombardment of natural uranium with 40-Mev helium ions, an alpha-particle group of 5.75-Mev energy and proved it to be due to Pu^{236}, formed by the reaction:

$$U^{235}\ (\alpha,3n)\ Pu^{236}$$

That this is the principal reaction was confirmed by the relative yields from targets of enriched U^{235} and of U^{238}, depleted with respect to U^{235}, but the evidence indicated in addition the occurrence of the reaction:

$$U^{238}\ (\alpha,6n)\ Pu^{236}$$

Several other cyclotron reactions can be used to produce Pu^{236}:

$$\left.\begin{array}{l} Np^{237}\ (d,3n)\ Pu^{236} \\ Np^{237}\ (\alpha,p4n)\ Pu^{236} \\ Np^{237}\ (\alpha,5n)\ Am^{236} \xrightarrow{EC} Pu^{236} \\ U^{233}\ (\alpha,n)\ Pu^{236} \end{array}\right\} \begin{array}{l} \text{Ref. 2} \\ \\ \\ \text{Ref. 3,4} \end{array}$$

This isotope has repeatedly been observed as the daughter of the 22-hour beta-emitter, Np^{236}, produced by the several reactions discussed in Section 9.1.7. Similarly this plutonium isotope has been observed as the alpha decay daughter of 27-day Cm^{240} which is discussed in Section 9.4.4. Confirmation of the mass assignment of Pu^{236} comes from the isolation and identification of U^{232} daughter activity.[1]

Small amounts of Pu^{236} have been detected in reactor-produced plutonium and are presumably accounted for by the following sequence of fast neutron reactions.

$$U^{238}\ (n,2n)\ U^{237} \xrightarrow[6.8\ days]{\beta^-} Np^{237}\ (n,2n)\ Np^{236} \xrightarrow[22\ hour]{\beta^-} Pu^{236}$$

The half-life of Pu^{236} as determined by direct decay is 2.85 years.[5] The spontaneous-fission rate as measured by GHIORSO AND COWORKERS[6] is $(5.8 \pm 2) \times 10^7$ fissions per gram-hour corresponding to a half-life of $(3.5 \pm 1) \times 10^9$ years for this process. Pu^{236} is beta stable.

The alpha particle spectrum of Pu^{236} has been measured on a magnetic spectrograph by HUMMEL AND COWORKERS:[7] 5.763 Mev (68.9 percent), 5.716 Mev (30.9 percent) and 5.610 Mev (0.18 percent). By alpha-gamma coincidence experiments it was established that a further group must exist with energy 5.448 Mev and 0.002 percent abundance.

The isotope Pu^{236} is an <u>even-even</u> nucleus and the energy levels of U^{232} observed in its decay follows a familiar pattern for <u>even-even</u> nuclei in this mass region. The lowest lying levels of U^{232} defined by the alpha spectrum of Pu^{236} are the following: Ground state (0+), 47.5 kev (2+), 156 kev (4+) and 321 (6+). A decay scheme is given in Figure 12. Pa^{232} decays to the same daughter nucleus, U^{232}, and several of the same E2 gamma rays are seen in its decay. Refer to Section 8.3.8.

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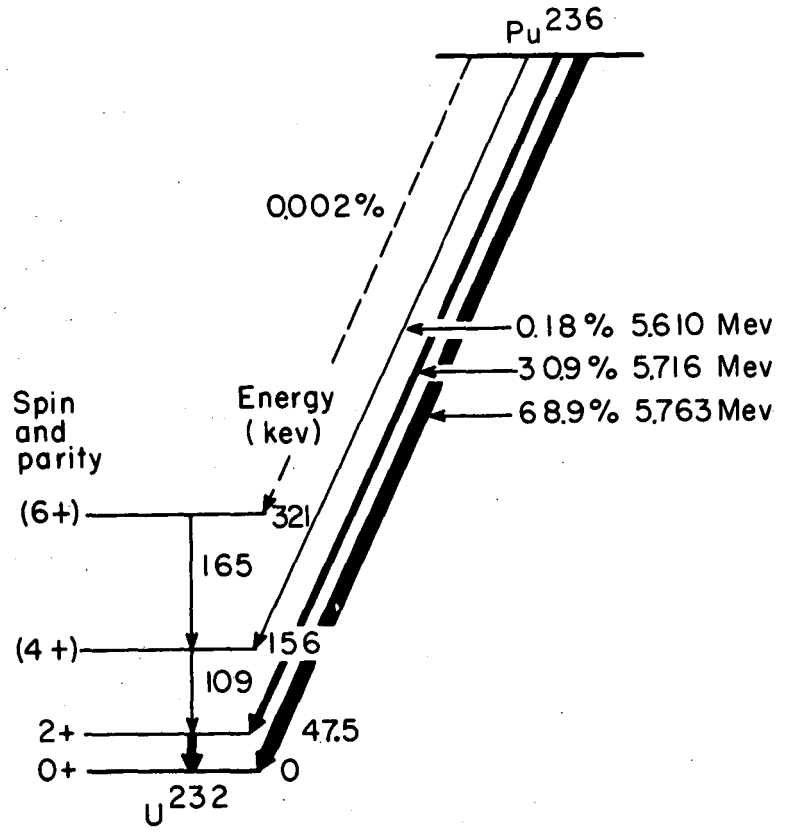

Figure 12. Decay scheme of Pu^{236}.

7. J. P. Hummel, F. Asaro, G. H. Higgins and I. Perlman, unpublished results (1956); see J. P. Hummel, Ph.D. Thesis, University of California, August, 1956; also published as University of California Radiation Laboratory Report, UCRL-3456, August, 1956.

9.2.7 Plutonium-237. JAMES, FLORIN, HOPKINS AND GHIORSO[1] found that the plutonium fraction from natural uranium bombarded with 40-Mev helium ions emitted a considerable amount of electromagnetic radiation of energy similar to characteristic K and L x-rays of a heavy element which suggested the presence of a nuclide decaying by electron capture. This activity decayed with a half-life of about 40 days. By bombardments of different uranium targets, some enriched and some depleted with respect to U^{235}, it was established that the U^{235} was the target isotope principally responsible for the formation of the activity but that it originated to a small extent from U^{238} as well. The same activity was produced later[2] by the action of 20-Mev deuterons on Np^{237} but was not found when U^{233} was bombarded with helium ions.[3] Neptunium isolated from an aged plutonium fraction[1] failed to show radioactivity due to Np^{235} indicating that the 40-day plutonium cannot be Pu^{235}. Consequently this activity is assigned to Pu^{237} produced by the reactions:

$$U^{235}\ (\alpha, 2n)\ Pu^{237}$$
$$U^{238}\ (\alpha, 5n)\ Pu^{237}$$
$$Np^{237}\ (d, 2n)\ Pu^{237}$$

Recent work by HOFFMAN[4] has established the over-all half-life as 45.63 ± 0.20 days.

For several years after its discovery no alpha radiation was found for Pu^{237} although it was known to be alpha unstable. The reason for this was the low branching ratio for this mode of decay and the unavoidable presence of Pu^{238} and often of Pu^{239} and Pu^{236} in the plutonium samples available for examination. Later search for the alpha radiation by THOMAS, VANDENBOSCH, GLASS AND SEABORG[5] and by HOFFMAN[4] was successful. The former reported alpha particles of 5.65 ± 0.02 Mev in 21 ± 4 percent abundance and of 5.36 ± 0.02 Mev in 79 ± 8 percent abundance. HOFFMAN[4] has observed an alpha group at 5.34 ± 0.01

Mev and has reported evidence for a group at 5.60 - 5.65 Mev and for a group at 5.20 - 5.25 Mev. Her , value for the branching ratio of the 5.34 Mev group is $(2.0 \pm 0.4) \times 10^{-5}$ in agreement with that determined by THOMAS AND CO-WORKERS, $(2.6 \pm 0.3) \times 10^{-5}$.

HOFFMAN AND DROPESKY[6] have studied in detail the conversion electron spectrum, the K-Auger electron spectrum and the photon spectrum of Pu^{237}. They found evidence for K and L x-rays and for gamma rays of energies 26.36, 33.20, 43.46, 59.57, 55.56 and 76.4 kev. The data indicate that the previously established levels of Np^{237} at 33.20, 59.57, 103.0 and 158.5 kev are fed by the electron-capture decay of Pu^{237}. These authors measured the following intensity ratios: k x-radiation; L x-radiation; 59.6-kev gamma radiation = 1.00; 0.75; 0.14. On the basis of their results, they have calculated a value of 0.06 ± 0.01 for the K-Auger coefficient. Both K- and L-electron capture populate the level at 59.6 kev, with a ratio of L- to K-electron capture to this level of 2.8. From this ratio HOFFMAN AND DROPESKY[6] have calculated an electron-capture decay energy of 0.21 Mev, in good agreement with the value of 0.22 Mev calculated from closed cycles. The relative intensities of the electron capture transitions and the log ft values for these transitions are shown in the decay scheme for Pu^{237} given in Figure 13. The spins, parities and asymptotic quantum numbers are those assigned by HOLLANDER, SMITH AND RASMUSSEN[7] on the basis of the alpha decay of Am^{241} and by RASMUSSEN, CANAVAN AND HOLLANDER[8] on the basis of the beta decay of U^{237}. The paper of HOFFMAN AND DROPESKY represents a very thorough study of the electron capture decay of Pu^{237} and should be consulted for details. GINDLER, GRAY AND HUIZENGA[11] found the ratio of K-electron capture to total electron capture to be 0.38 ± 0.06.

STEPHENS, ASARO, AMIEL AND PERLMAN[9] have identified an isomer of Pu^{237} with a half-life of 0.18 ± 0.02 seconds. This nuclide was isolated by collecting recoils from the alpha decay of Cm^{241}. The isomer is 145 kev above the ground state and is thought to have spin 1/2. A more detailed description of this isomer is found in the discussion of the decay of Cm^{241}, Section 9.4.5.

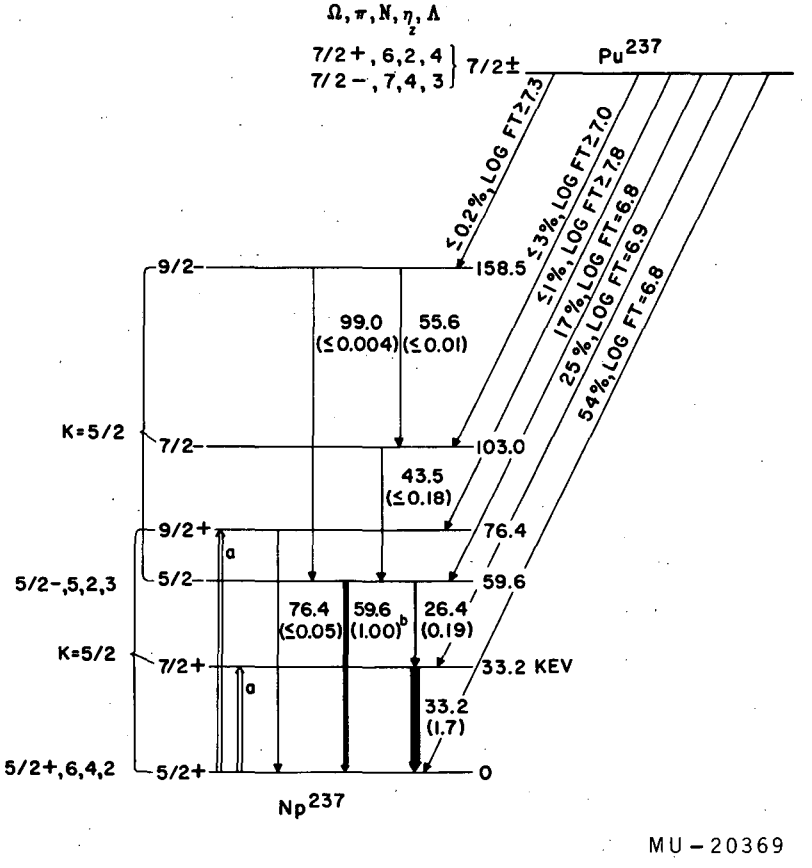

Figure 13. Proposed decay scheme for the electron capture decay of Pu^{237} as given by HOFFMAN and DROPESKY.[6] The spins, parities and asymptotic quantum numbers assigned to the levels of Np^{237} are those given by HOLLANDER et al[7] and RASMUSSEN et al[8] from a study of the decay of Am^{241} and U^{237} (Refer to the discussion of these nuclides). The symbol a denotes coulomb excitation experiments of NEWTON.[10] The values in parentheses give estimates of the intensities of the various transitions relative to the 59.6 kev transition.

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9.2.8 Plutonium-238. In the description of the discovery and early history of plutonium, which appears earlier in this chapter, it is mentioned that Pu^{238} was the first isotope of element 94 to be identified. SEABORG, MC MILLAN, KENNEDY AND WAHL[1] prepared it by bombarding normal uranium with 16 Mev deuterons. KENNEDY, PERLMAN, SEGRÈ AND WAHL[2] made the mass assignment certain by bombarding U^{238}, depleted with respect to U^{235}, and noting that the yield was proportional to the U^{238} content. ENGLISH AND JAMES[3] determined the yield as a function of deuteron energy and showed that it varied in a manner expected for the reaction:

$$U^{238}\ (d,2n)\ Np^{238} \xrightarrow[2.1\ days]{\beta^-} Pu^{238}$$

Subsequent experiments with helium ion bombardments of U^{238} and U^{235} targets have resulted in the formation of Pu^{238} by the following reactions:[4]

$$U^{238}\ (\alpha,4n)\ Pu^{238}$$
$$U^{238}\ (\alpha,p3n)\ Np^{238} \xrightarrow{\beta^-} Pu^{238}$$
$$U^{235}\ (\alpha,n)\ Pu^{238}$$
$$U^{235}\ (\alpha,p)\ Np^{238} \xrightarrow{\beta^-} Pu^{238}$$

Preparation of Pu^{238} by Neutron Reactions. This isotope is also produced[5,6] as a result of neutron capture by Np^{237}:

$$Np^{237}\ (n,\gamma)\ Np^{238} \xrightarrow{\beta^-} Pu^{238}$$
$$\sigma = 172\ barns$$

This is a convenient method for the preparation of isotopically pure Pu^{238}. A small amount of Pu^{238} is formed in reactor-produced plutonium via the reaction sequence:

$$U^{238}\ (n,2n)\ U^{237} \xrightarrow[6.8\ days]{\beta^-} Np^{237}\ (n,\gamma)\ Np^{238} \xrightarrow[2.1\ days]{\beta^-} Pu^{238}$$

The amount of Pu^{238} so produced is small on a weight ratio basis but it is readily detectable because its alpha half-life is short compared to Pu^{239}. The calculation of the yield of Pu^{238} by this mechanism depends on knowledge of the distribution of neutron energies in the reactor. In high-flux reactors containing plutonium operating at neutron fluxes of the order of 10^{14} neutrons cm^{-2} sec^{-1} and higher measurable amounts of Pu^{238} may be produced by another mechanism. If the integrated flux is high enough to produce Cm^{242} via the multipole neutron-capture sequence given below, some Pu^{238} will appear in the sample as a result of its formation by the alpha decay of Cm^{242}.

$$Pu^{239}\ (n,\gamma)\ Pu^{240}\ (n,\gamma)\ Pu^{241} \xrightarrow[13\ years]{\beta^-} Am^{241}\ (n,\gamma)\ Am^{242m} \xrightarrow[16\ hours]{\beta^-} Cm^{242}$$
$$Pu^{238} \xleftarrow[162\ days]{\alpha}$$

Figure[14] shows the production of Pu^{238} by this mechanism. If the flux is very high very little Pu^{238} is formed because the Pu^{241} is burned out quickly

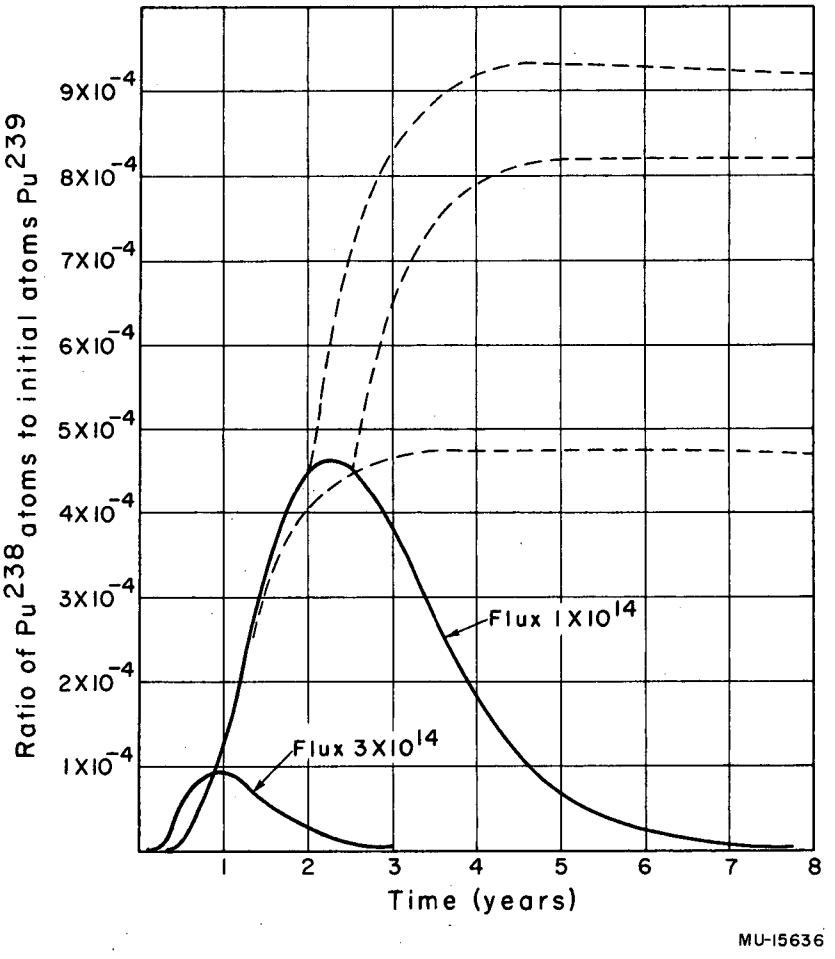
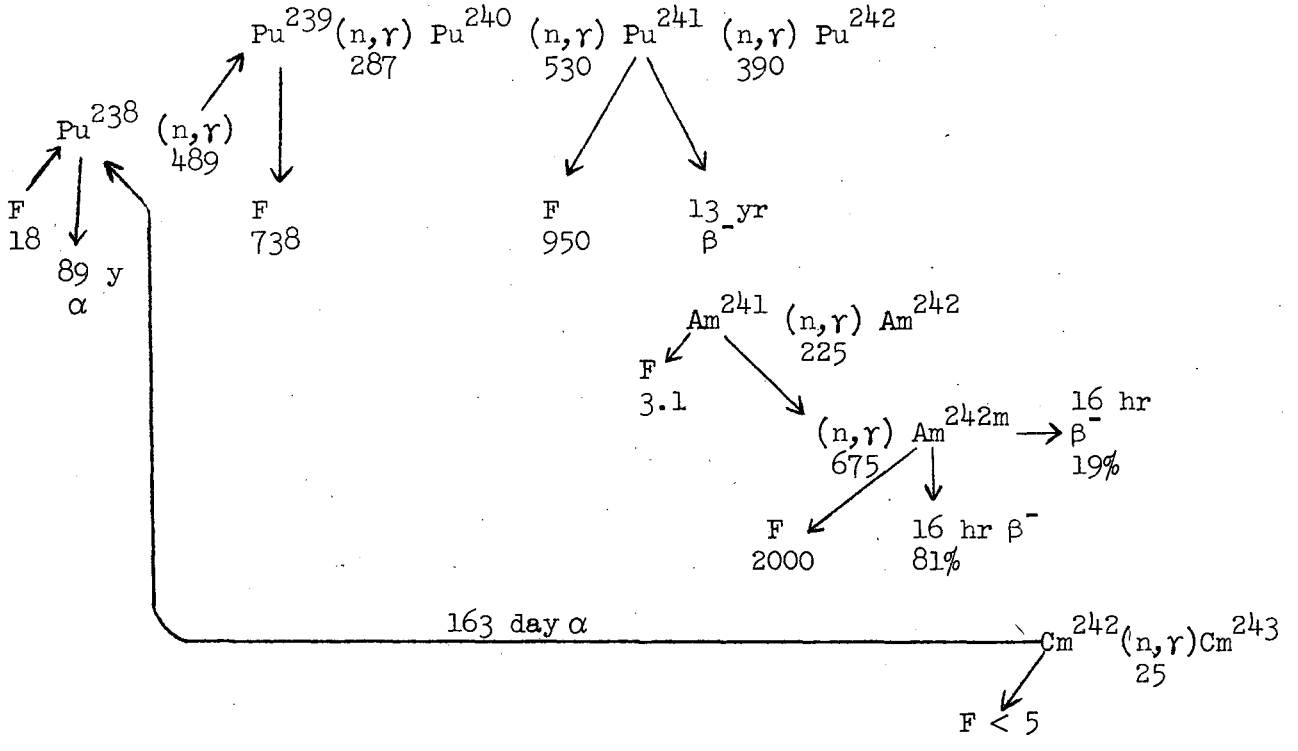

Ratio of Pu^{238} atoms to initial atoms Pu^{239}
9X10^{-4}
8X10^{-4}
7X10^{-4}
6X10^{-4}
5X10^{-4}
4X10^{-4}
3X10^{-4}
2X10^{-4}
1X10^{-4}
Flux 1X10^{14}
Flux 3X10^{14}
1
2
3
4
5
6
7
8
Time (years)
MU-15636

Figure 14. Production of Pu^{238} by multiple neutron capture to produce Cm^{242} followed by alpha decay of Cm^{242} at neutron fluxes of 3×10^{14} and 1×10^{14} neutrons cm^{-2} sec^{-1}. The constants used in this calculation and the complicated sequence of capture and decay steps are given in the following. F refers to removal of the isotope by fission. The numbers are cross sections in barns:



When the sample is removed from the reactor the Pu^{238} continues to increase because of the Cm^{242} inventory in the sample. This is shown by the dotted lines in the 1×10^{14} flux case for samples removed after one year, two years and 2.5 years or irradiation.

to form higher mass isotopes before much can decay to Am^{241}. If the irradiation is continued for several years at lower fluxes the production of Pu^{238} also goes down when most of the matter is converted to higher mass nuclides or to fission products and the Pu^{238} itself is converted back to Pu^{239} by neutron capture. It is interesting to note the dotted lines in the figure which show that the amount of the Pu^{238} may increase by more than a factor of two after the sample is removed from the reactor because of the subsequent decay of Cm^{242}. At a flux of 1×10^{14} neutrons cm^{-2} sec^{-1} the maximum production of Pu^{238} occurs for a radiation of about 2 years time. The chief isotopes of plutonium in the sample at this time will be Pu^{242} and Pu^{240}.

The amount of Pu^{238} by mass will be roughly one-half percent. In Fig. 1 we neglected the possibility of producing Pu^{238} by the Pu^{239} (n,2n) Pu^{238} path since the Pu^{239} is burned out quickly and any Pu^{238} so produced during the first two weeks of irradiation is converted back to Pu^{239} by neutron capture.

Gram quantities of Am^{241} are now available in many laboratories so that pure Pu^{238} can be made from Am^{241} by conversion of the Am^{241} to Cm^{242} as shown in Fig. 15. For the highest isotopic purity the Cm^{242} should be first isolated and purified and the Pu^{238} separated after decay; the reason for this indirect procedure is that Pu^{242} is produced by the electron capture decay of Am^{242m} and some Pu^{238} is converted to Pu^{239} in the reactor.

In reactors operating on U^{235} some Pu^{238} is produced via the reaction sequence:

$$U^{235}\ (n,\gamma)\ U^{236}\ (n,\gamma)\ U^{237} \xrightarrow[6.8\ \text{days}]{\beta^-} Np^{237}\ (n,\gamma)\ Np^{238} \xrightarrow[2.1\ \text{days}]{\beta^-} Pu^{238}$$

The rate of buildup of Pu^{238} by this sequence is shown in Fig. 16.

The whole question of the formation of heavy element isotopes by multiple neutron-capture reactions in high flux reactors is discussed thoroughly in Chapter 5.

The Radiations of Pu^{238}. The best measurements of the half-life of Pu^{238} have come from direct observation of Pu^{238} alpha activity over a period of a few years. An accurate determination is that of JAFFEY and LERNER[7] who followed four samples over a period of 31 months and obtained a value of 89.59 ± 0.37 years. A later determination was made by HOFFMAN, FORD and LAWRENCE[19] who measured the

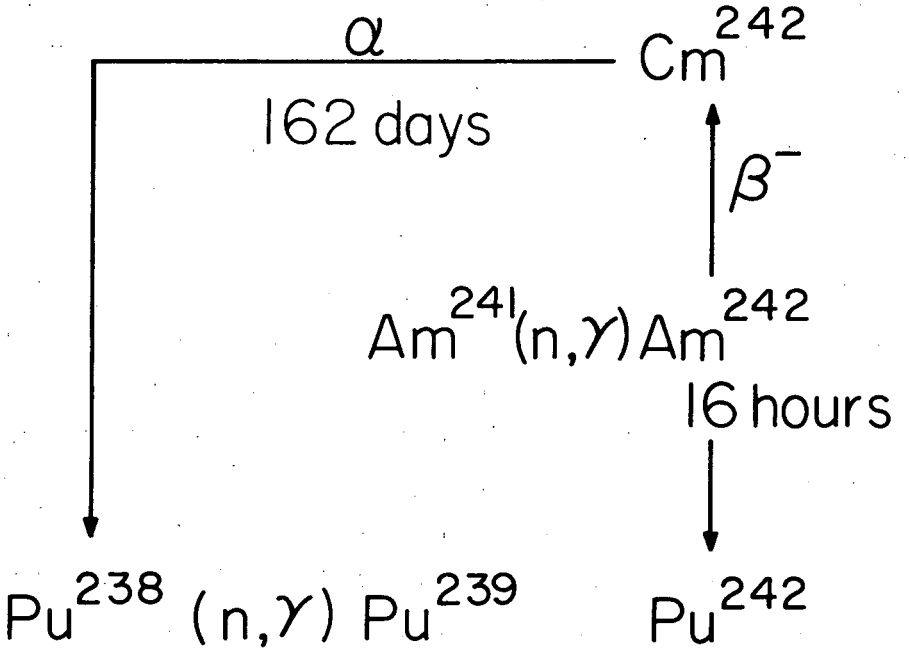


Figure 15. Path of conversion of Am^{241} to Pu^{238}.

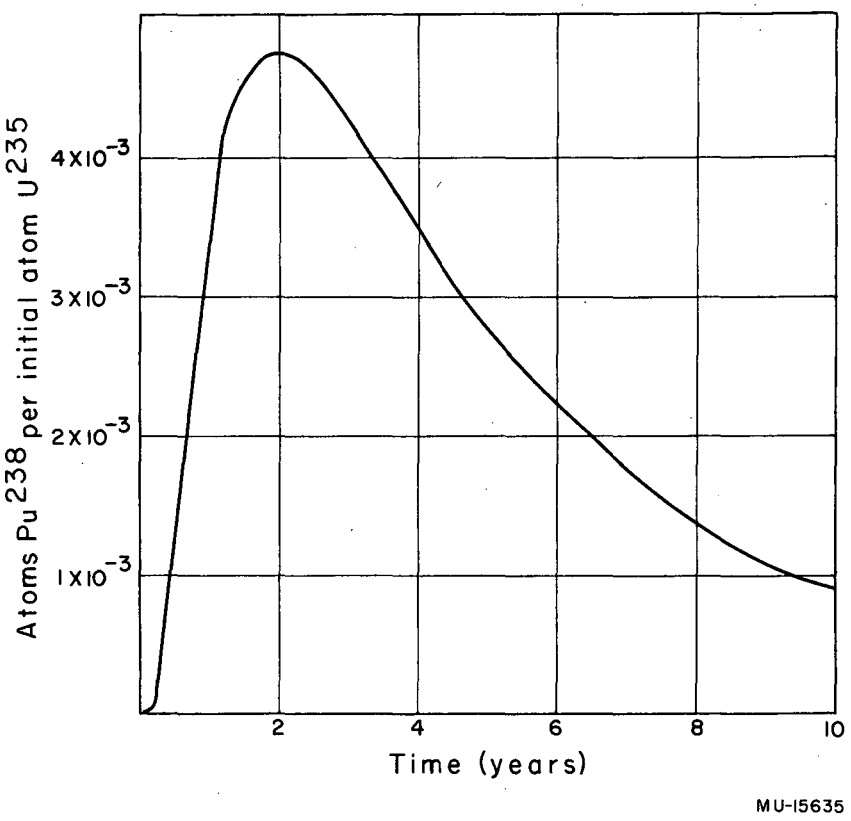


Figure 16. Buildup of Pu^{238} in a thin sample of pure U^{235} irradiated at a neutron flux of 3×10^{14} neutrons cm^{-2} sec^{-1}. The neutron capture and beta decay sequence and the reaction cross sections are as follows:

$$U^{235} \xrightarrow{(n,\gamma)\ 112} U^{236} \xrightarrow[24]{(n,\gamma)} \xrightarrow[6.8\ d]{\beta^-} Np^{237} \xrightarrow[170]{(n,\gamma)} Np^{238} \xrightarrow[2\ d]{\beta^-} Pu^{238} \xrightarrow[489]{(n,\gamma)}$$

U^{235} Fission 582; Np^{238} Fission 1600; Pu^{238} Fission 18

Pu^{238} activity growing into a Cm^{242} sample. They reported a half-life of 86.41± 0.3 years for Pu^{238} based on a Cm^{242} half-life of 162.7 days.

Several early reports on the energy of the alpha particles of Pu^{238} are superceded by those of ASARO and PERLMAN[8] and of GOL'DIN, TRET'YAKOV and NOVIKOVA.[9] The former group reported the following energies and abundances: 5.495 kev[†] (72 percent), 5.452 (28 percent) and 5.352 (0.095 percent). The latter group reported 5.4909 Mev (69 percent) and 5.4499 (31 percent).

ASARO and PERLMAN[8] studied the gamma radiation of Pu^{238} with a scintillation crystal spectrometer and found a large number of 17-kev L x-rays (from the conversion of gamma rays) and gamma rays at 43.8, 99, and 150 kev. NEWTON, ROSE and MILSTED[10] measured these gamma rays with somewhat greater accuracy by the proportional counter pulse-height analysis technique and reported gamma ray energies of 43.49 ± 0.08, 99.8 ± 0.4 and 153.1 ± 0.6 kev with relative intensities of 100: 28: 3. HOLLANDER[11] examined the conversion electrons of the lowest energy transition. His value of 43.50 kev checks closely the value of NEWTON, ROSE and MILSTED.[10] THE L_{II} L_{III} M_{II} M_{III} pattern of conversion confirms the E2 nature of the transition.

DUNLAVEY and SEABORG[12] found by the photographic plate technique, with emulsions soaked with a solution containing Pu^{238}, that 23 percent of the Pu^{238} alpha particles are in coincidence with electrons corresponding to an ~40-kev gamma ray. ASARO and PERLMAN[8] conclude from comparison of the conversion coefficients, estimated from the experimental data, with the theoretical conversion coefficients that these radiations are E2 in nature. CHURCH and SUNYAR[13] confirmed the electric quadrupole character of these gamma rays by detailed study of relative conversion in the L and M subshells. They report the values 43.6 and 100.0 kev for the gamma ray energies. MILTON and FRAZER[14] measured the angular correlations of the 43.6-kev photons with the 5.45 Mev alpha particles of Pu^{238} and obtained the correlation expected for a $0+ \xrightarrow{\alpha} 2+ \xrightarrow{\gamma} 0+$ sequence. The 153-kev gamma ray is also believed to be an electric quadrupole radiation although with less certainty. These results are summarized in the decay scheme shown in Fig. 17. TRETYAKOV, KONDRATEV, KHLEBNIKOV and GOLDIN[9a] measured conversion electrons in coincidence with the alpha-particles of Pu^{238} and found evidence for a gamma transition of energy 152.6 ± 0.3 kev with a subshell conversion ratio typical of an E2 transition.

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† A restandardization of α_0 by Asaro and Perlman in 1961 led to the result 5.497 Mev.

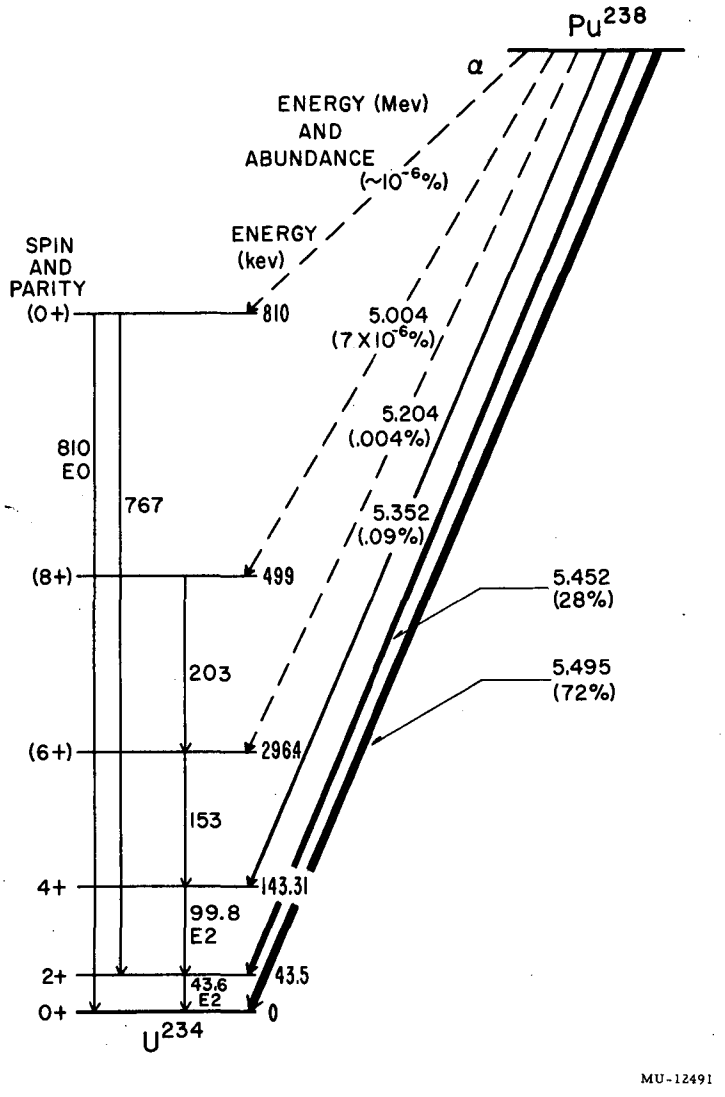

Figure 17. ^{234} Decay scheme of Pu^{238}. The decay of the UX_2 - UZ complex and of Np^{234} also involves these levels of U^{234} plus other levels not shown which are not reached in the alpha decay of Pu^{238}.

They interpret this as evidence for a 6+ level at 295.9 ± 0.4 kev populated in $(4.3 \pm 0.4) \times 10^{-3}$ percent of the alpha events.

By gamma-gamma coincidence experiments it was established by ASARO, STEPHENS and PERLMAN that the 153 kev gamma ray is in cascade with the 99.8-kev gamma ray.[8,15] Placement of this gamma ray in the scheme as shown in the figure requires that an unboserved alpha particle group of 5.204 Mev energy and 0.004 percent abundance be emitted by Pu^{238}; an alpha particle group with this low intensity would have been missed in the direct examination of the alpha spectrum. Later work by ASARO, STEPHENS and PERLMAN[15] led to the discovery of a 203 ± 5 kev gamma ray in very low abundance (4×10^{-6}%) which is in coincidence with the 150-kev gamma rays. This gamma ray is shown as a transition from an 8+ level at 499 kev to a 6+ level at 296.4 kev in the figure. This assignment, which is very reasonable but not definitely proved, requires that an undiscovered alpha-particle group of 5.004 Mev energy and 7×10^{-6} percent abundance be emitted by Pu^{238}.

ASARO, STEPHENS and PERLMAN[15] have found strong evidence for an electric monopole (E0) transition occurring in low abundance in the decay of Pu^{238}. An examination of the conversion electron spectrum using an anthracene crystal as detector showed K and L electrons of an 0.82-Mev transition. The total electron abundance was roughly 10^{-6} electrons per alpha disintegration. No gamma-ray photons of this energy were seen with a sodium iodide crystal detector. In an electric monopole transition photon emission is forbidden and the transition occurs entirely by electron emission. This 0.820-Mev transition is in all likelihood identical with the 810 kev (or 803 kev) 0+ —> 0+ transition to the ground state of U^{234} which has been identified in the beta decay of UX_2 (Pa^{234}) to U^{234} (see Section 5.2.12) and it is so drawn in the decay scheme. This same E0 transition is seen in the electron capture decay of Np^{234}. See figure 7 in Section 9.1.5. If this is correct one would expect to observe an E2 transition of 767 kev energy from the 0+ level at 810 kev to the 43.5-kev 2+ level. ASARO[15] observed photons of 763-kev energy in abundance corresponding to $\sim 5 \times 10^{-7}$ photons per alpha. These photons were shown to be in coincidence with L x-rays, presumably from the conversion of the 43.5-kev gamma ray, and not to be in coincidence with photons of 100-kev energy. In addition, photons of an 875-kev gamma ray were observed in about 10^{-6} % abundance but this transition has not been placed in the decay scheme.

The energy levels of U^{234} observed from the alpha decay of Pu^{238} resemble those of several other even-even nuclei in this mass region in conforming in a remarkable fashion with the predictions of the unified nuclear model as developed by BOHR and MOTTELSON. The 0+, 2+, 4+ sequence has the correct energy spacings for the first members of the ground state rotational band with the splitting constant $\hbar/2\Im$ set equal to 7.30 kev. The levels at 296 and 499 kev have the correct energy values for the 6+ and 8+ levels of this rotational band. The 0+ level at 810 represents a collective vibrational excitation of the nucleus.

Early measurements by SEGRÈ and CO-WORKERS[16,17] on the rate of spontaneous fission of Pu^{238} gave a value of $(5.1 \pm 0.8) \times 10^6$ fissions per gram-hour corresponding to a spontaneous fission half-life of about 3.8×10^{10} years. A more recent measurement by JAFFEY and HIRSCH[18] gave a value of $(4.0 \pm 0.34) \times 10^6$ fissions per gram-hour corresponding to a half-life of 4.9×10^{10} years.

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9.2.9 Plutonium-239. The discovery of Pu^{239} through the work of McMILLAN and ABELSON[1] and of KENNEDY, SEABORG, SEGRÈ and WAHL[2] is described in Section 9.2.1. The fission properties of Pu^{239} are discussed in Chapter 11.

The most important method of preparation of Pu^{239} is the neutron capture reaction involved in its original synthesis:

$$U^{238}\ (n,\gamma)\ U^{239};\ U^{239} \xrightarrow[23.5\ min]{\beta^-} Np^{239} \xrightarrow[2.33\ days]{\beta^-} Pu^{239}$$

Plutonium-239 is made in large quantities in nuclear reactors by this reaction for industrial or military purposes. Reactor-produced plutonium contains small amounts of other plutonium isotopes as a result of side reactions. In reactors operating at very high fluxes the amounts of higher-mass plutonium isotopes produced by multiple neutron capture can be quite sizeable. (See Chapter 5)

Plutonium-239 is produced by a number of cyclotron reactions with U^{238} as a target:

$$U^{238}\ (d,p)\ U^{239};\ U^{239} \xrightarrow{\beta^-} Np^{239} \longrightarrow Pu^{239}$$

$$(d,n)\ Np^{239};\qquad Np^{239} \longrightarrow Pu^{239}$$

$$(\alpha,3n)\ Pu^{239};$$

$$(\alpha,p2n)\ Np^{239};\qquad Np^{239} \longrightarrow Pu^{239}$$

The yields for several of these reactions are given in Table 5.5 in Chapter 5.

The Half-life of Pu^{239}. Considerable effort has been expended in the determination of the half-life of Pu^{239}. The first determination[2] of 30,000 years was obtained by counting the beta particles of Np^{239} and the alpha particles of its daughter and was subject to considerable uncertainty because of the complexity of the radiations of Np^{239}. A later refinement of the same method[3] gave the value 23,000 years; in this determination the Pu^{239} alpha activity was compared with the corresponding beta activity of its parent, U^{239}, whose radiations are simpler than those of Np^{239} and can be easily interpreted.

The first accurate value for the half-life was derived from the experiments of CUNNINGHAM and WERNER,[4] who determined the specific alpha activity by weighing a pure compound of Pu^{239} and counting its alpha particles. These experiments constituted the first isolation of plutonium and the first preparation and isolation of a weighable amount of a synthetic atomic species. In spite of the small quantity of material available, which was of the order of 100 μg, the accuracy of weighing and volumetric aliquoting was so great that the limiting factor in these experiments was the uncertainty in the alpha-counting yield (ratio of counts to disintegrations) of the instruments used. This quantity is needed in order to convert the observed alpha-particle counting rate of the samples to the absolute alpha-disintegration rate. Experimental work of CUNNINGHAM, GHIORSO and HINDMAN[5] and theoretical calculations of CRAWFORD[6] led to the discovery that, in a parallel-plate ionization chamber, the alpha-particle counting yield may be greater than 50 percent as a result of back-scattering of some of the particles from the sample mount. The early specific-activity measurements were in error because counting yields of 47 to 50 percent were assumed, whereas measurements[5] indicate that the counting yield of an extremely thin sample of Pu^{239} on a flat platinum plate in a parallel-plate counting ionization chamber operating in air is about 52 percent. However, most of the backscattering alpha particles leave the plate at low angles, and the calculations indicate that, normal to the surface of the plate, there are virtually no backscattering particles. Consequently, in a counter with a low geometry factor, in which only those particles traversing a small aperture at the end of an evacuated collimating tube normal to the plane of the sample mount are counted, the counting yield is identical with the geometry factor and can be

calculated by careful geometrical measurements. Such counters were therefore used for calibration in connection with the accurate specific-activity determinations.

The weighing experiments of CUNNINGHAM and CO-WORKERS[4,5] led to a half-life of 24,300 years. Careful redeterminations by WESTRUM, HINDMAN and GREENLEE[7] have given 24,400 ± 70 years, and FARWELL, ROBERTS and WAHL[8] have also obtained the value of 24,400 years. The calorimetric measurements of JONES and STOUT[9] have resulted in the value 24,110 ± 240 years, based on 5.23 Mev as the disintegration energy of the alpha decay. A later specific activity determination, carried out by WALLMANN[10] yielded the value 24,360 ± 100 years. DOKUCHAEV[39] made specific activity measurements on 12 specimens of Pu^{239} having isotopic composition varying from 91.26 to 99.11% Pu^{239} and obtained a value of 24,390 ± 30 years. MARKIN[40] solved the problem of the correction for Pu^{240} by using electromagnetically separated Pu^{239} with an isotopic purity of 99.92 percent. He prepared and weighed a series of samples as the compounds, $Pu(SO_4)_2$ and $PuCl_3$, and by specific activity measurements found a half-life value of 24,413 ± 30 years.

It should be pointed out that one of the chief complications in the determination of the specific activity of Pu^{239} is the presence of small amounts of Pu^{240} which contribute to the alpha activity. The Pu^{240} content must be measured in some manner such as by measuring the spontaneous-fission rate of the sample. The Pu^{240} content of Pu^{239} is dependent on the neutron irradiation history of the sample. Small amounts of Pu^{238} may also be present in Pu^{239} prepared in a reactor because of the side reactions discussed in the last section. Because of the 89-year half-life of Pu^{238} its contribution to the specific activity can be appreciable even when its weight percent is quite low.

The Alpha-Particle Spectrum of Pu^{239}. A number of measurements of the energy of the main alpha-particle group of Pu^{239} have been carried out but the earliest measurements will not be reviewed here. Three groups have made careful measurements by the ionization chamber method; JESSE and FORSTAT[11] obtained a value 5.137 Mev, CRANSHAW and HARVEY[12] found 5.159 ± 0.005 Mev and CONJEAUD and NAGGIAR[13] gave the value 5.134 ± 0.015 Mev for the main group. The most accurate measurements have been made by the magnetic-deflection method and these have revealed complex structure in the alpha spectrum. ROSENBLUM,

VALADARES and GOLDSCHMIDT[14] report a main group at 5.147 Mev in 70 percent abundance and a second group at 5.097 Mev in 30 percent abundance. ASARO and PERLMAN[15] report three groups with the energies and abundances, 5.150 Mev† (69 percent), 5.137 Mev (20 percent) and 5.099 Mev (11 percent). GOL'DIN, TRET'YAKOV and NOVIKOVA[16] report the same three groups, their figures being 5.1474 Mev (72.5 percent), 5.1344 Mev (16.8 percent) and 5.0963 Mev (10.7 percent); no other alpha groups in greater than 0.2 percent abundance in the energy region 4.9 to 5.33 Mev were found. NOVIKOVA, KONDRAT'EV, SOBOLEV and GOL'DIN[16a] reported additional groups with the following energies and intensities: 5.064 ± .002 Mev (0.037 ± 0.005%), 4.999 ± 5 Mev (0.013 ± 0.005%), and 4.917 ± 0.005 Mev (0.005 ± 0.001%).

The Gamma Radiations of Pu^{239}. A number of studies have been made of the electromagnetic and electron radiations of Pu^{239}. Absorption curve measurements carried out by early workers showed that the electromagnetic radiation included uranium K and L x-rays and apparently some gamma rays of 400 kev and lower energy. The most energetic gamma ray was reported to exist in approximately 10^{-4} of the disintegrations and x-rays were reported to be emitted in about one-tenth to one-hundredth of the disintegrations. The electrons are of low energy and consist of internal conversion and Auger electrons. Using photographic emulsions, ALBOUY and TEILLAC[17] found conversion electrons corresponding to gamma rays of 50 kev and also found 100-kev electrons (0.1 to 1 percent); by the same technique DUNLAVEY and SEABORG[18] found the conversion electrons of 35- and 50-kev gamma rays (0.12 gamma ray per alpha particle) and also 100-kev conversion electrons (0.5 percent). WEST, DAWSON and MANDELBERG,[19,20] using a proportional counter, found the corresponding gamma ray photons with energies 52.0 ± 0.3 kev (7×10^{-5} gamma ray per alpha particle) and 38.5 ± 0.4 kev (2×10^{-5} gamma ray per alpha particle). FREEDMAN, WAGNER and ENGELKEMEIR[21] have assigned gamma rays of 39, 53.1, 100, 124 and 384 kev to the alpha decay of Pu^{239} from conversion-electron and scintillation-spectrometer measurements. The ratio of L x-rays of uranium to the last four of these is about 100:0.56:0.25:0.14:0.08. These investigators,[19,20,21] as well as ISRAEL,[22] have made measurements of sufficient accuracy to assign the L x-rays to the daughter uranium. ASARO and PERLMAN[23] have studied the gamma rays with a scintillation spectrometer and found photons with energies of 37, 52, 120, 207, 340, 380 and 415 kev in

† In 1961 Asaro and Perlman remeasured α_0 and obtained the energy value 5.152 Mev.

addition to K_α and K_β x-rays. The relative intensity of the gamma ray photons is 130:320:70:20:30:60:40. Gamma-gamma coincidence studies established that L x-rays are in coincidence with the 207-, 340- and 380-kev gamma rays and that the 120- and 207-kev gamma rays are in coincidence with each other. SHLIAGEN[24] studied the conversion electrons of gamma rays with energies 12.5, 38.3, 50.8 and 117 kev. ALBRIDGE AND HOLLANDER[41] report precise values of 13.00 Kev and 51.67 for these two important transitions.

Decay Scheme of Pu^{239} and the Discovery of U^{235m}. It is simple to construct a reasonable partial decay scheme of Pu^{239} incorporating the known alpha groups and the most prominent gamma transitions of energy 38 and 51 kev which have been reported by most experimentalists. This partial decay scheme is shown on the right side of Fig. 18. It is evident, however, from the repeated observation of low intensity gamma rays of higher energy that one or more alpha groups in very low abundance (less than 0.1 percent) with lower energy than any yet reported, must be present in the alpha spectrum of Pu^{239}. It is tempting to identify the observed spectrum of U^{235} levels as an "anomalous" rotational band based on a $K = 1/2$ base level. This is consistent with the spacings of the levels, the known ground state spin of 1/2 for Pu^{239} and the low hindrance factor of 3 for the highest energy alpha group suggesting favored alpha decay to a daughter nucleus of very similar intrinsic particle structure. This interpretation was first suggested by BOHR, FROMAN and MOTTELSON.[25]

The formula given by BOHR and MOTTELSON (see Chapter 3) for the level spacings of an anomalous $K = \Omega = 1/2$ band is

$$E_I = \frac{\hbar^2}{2\mathfrak{J}} \left[I(I+1) + (-)^{I+1/2} a(I+1/2) \right] .$$

The lowest 5 levels of U^{235} populated by the alpha-groups of Pu^{239} fit this formula closely if the constant, a, is set equal to -0.276 and $\hbar^2/2\mathfrak{J}$ is 6.1 kev.[16a] ALBRIDGE AND HOLLANDER[41] give slightly different values of -0.28 and 6.04 Kev based on their values of 13.00 and 51.67 Kev for the first two levels.

For a long period of time this very reasonable interpretation of the alpha decay of Pu^{239} gave rise to a very puzzling discrepancy. The difficulty was that the measured spin[26] of the ground state of U^{235} is 7/2 instead of 1/2. Furthermore, the experiments of NEWTON[27] on the Coulombic excitation of U^{235} with helium ions clearly proved that levels with energy 46.2 kev and 103 kev were among the low-lying levels of U^{235}. He observed the gamma rays of 46.7, 57.8 and 103.8 kev shown in the figure. These experiments constitute strong

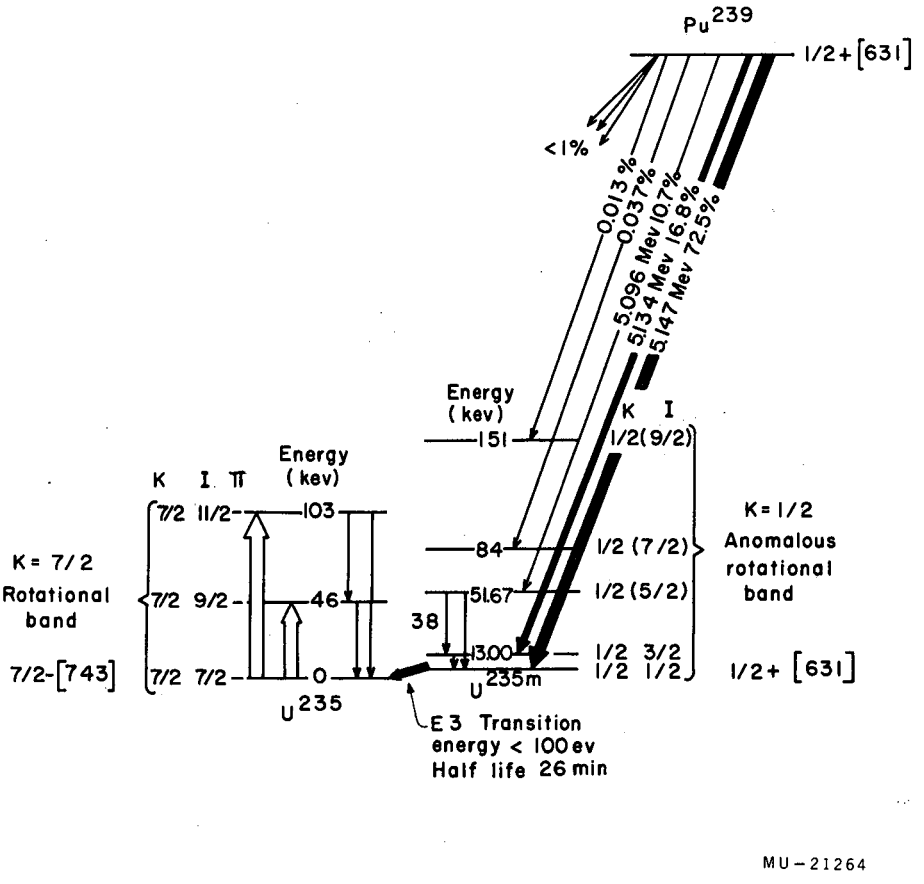

Figure 18. Decay scheme of Pu^{239}. The alpha decay populates five levels of a K = 1/2 "anomalous" rotational band. The ground state of this band is an isomeric state of U^{235} lying only a few volts above the I = 7/2- ground state. The isomeric state decays by a highly converted E3 isomeric transition with a half-life of 26 minutes. The higher-lying levels of the K = 7/2 ground state rotational band are known from coulombic excitation studies. Gamma rays (not shown) and α-groups in low abundance indicate that some low intensity alpha groups of Pu^{239} populate higher-lying levels. The Nilsson wave function assignments of U^{235} (ground state) and U^{235m} are 7/2-[743] and 1/2+[631].

evidence that a (7/2, 9/2, 11/2) rotational band based on a K = 7/2 ground state is being excited. It was realized by workers in several laboratories that the discrepancy between the two sets of observations could be resolved if some transition were observed in nearly 100 percent abundance between the K = 1/2 and K = 7/2 states. This transition would be expected to be of the $\Delta I = 3$ type. A diligent search for the photons, the conversion electrons or the x-rays from this transition lead at first only to the conclusion that the energy of the transition must lie below the L-shell binding energies of uranium. The possibility that this unobserved transition had a very long half-life was checked by HUIZENGA, ENGELKEMEIR and TOMKINS[28] who separated U^{235} which had grown into a 10-year old sample of Pu^{239}, and measured its spin. The optical spectrum was the same as that of normal uranium. From the limits of the experiment an upper limit of 4 months was set on the half-life for the transition between the two forms of U^{235}.

Following these observations the investigation took the form of a search for a short-lived transition of extremely low energy and was rewarded with immediate success. ASARO and PERLMAN[29] and HUIZENGA, RAO and ENGELKEMEIR[30] almost simultaneously reported the separation by a recoil collection technique of an activity emitting very soft conversion electrons and decaying with a half-life of 26.5 minutes. The electron activity could also be isolated chemically using separation methods specific for uranium. Later measurements showed that the energy of the electrons was extremely low indeed. FREEDMAN, PORTOR, WAGNER and DAY[31] set an upper limit of 19 electron volts and an upper limit of 23 electron volts to the transition energy. MICHEL, ASARO and PERLMAN[32] found electrons of energy up to 70 volts. The great majority of the electrons are of much less energy. They are nearly completely absorbed in 2 micrograms of plastic film, which accounts for their non-observance in the numerous studies of the radiations of Pu^{239} which had been carried out earlier. This is by a large margin the lowest transition energy ever reported for an isomeric state. The short half-life is accounted for by the extremely large conversion coefficient which is estimated to fall in the range 10^{19} - 10^{21}.

From a consideration of the nuclear level systems of Np^{239}, Pu^{239} and U^{235} it is possible to make a tentative assignment of Nilsson wave functions to the odd 143rd neutron in U^{235} (ground state) and in U^{235m}. In the $(N\ n_z\ \Lambda\ K\ \pi)$ nomenclature of the unified nuclear model (see Chapter 3) these assignments are

(743 7/2-) and (631 1/2+) respectively. The isomeric transition would accordingly be E3. The Nilsson orbital assignment of the ground state of Pu^{239} is (631 1/2+). It is interesting to note that another case of E3 isomerism resulting from the close proximity of these same two Nilsson states is observed[33] in Pu^{237} which, like U^{235}, has 143 neutrons. See Section 9.4.5.

Not all of the levels of U^{235} populated in the alpha decay of Pu^{239} are shown in the figure. There is evidence for very slight branching to higher-lying levels. ASARO and PERLMAN[23] reports levels at 172, 379 and ~430 kev populated in 0.02%, 0.006% and 0.006% of the decay events from a study of gamma-gamma coincidences. NOVIKOVA, KONDRATEV, SOBOL'EV and GOL'DIN[16a] report weak alpha-groups populating levels at 172, 234, 336, 373, 426 and 497 kev with the following percentage intensities; ~0.02%, 0.005%, ~0.035%, ~0.002%, ~0.005% and ~0.0015%. Spins, parities and Nilsson quantum numbers have not been assigned to these levels.

Level System of Pu^{239}. The excited levels of Pu^{239} are discussed in connection with the decay of Np^{239} (see Section 9.1.10). The excitation of the 57 kev level of Pu^{239} by Coulombic excitation techniques is also covered there rather than here.

Spin of Pu^{239}. The nuclear spin of Pu^{239} has been measured to be 1/2 by nuclear paramagnetic resonance,[34] by analysis of optical spectra,[35,36,37] and by atomic beam resonance studies.[38]

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9.2.10 Plutonium-240. CHAMBERLAIN, FARWELL and SEGRE[1] provided the first direct evidence for the isotope Pu^{240}; this evidence was the observation that samples of Pu^{239} that had been exposed to strong neutron fluxes exhibited spontaneous fission activity and that the spontaneous fission rate was proportional to the irradiation received. From this relation it was deduced that the isotope responsible was Pu^{240} formed in the reaction Pu^{239} (n,γ) Pu^{240}; and this has been firmly established by subsequent experiments. BARTLETT, SWINEHART and THOMPSON[2] established the presence of Pu^{240} in pile produced plutonium by mass spectrographic analysis.

The alpha half-life of Pu^{240} has been measured several times by direct specific activity measurements although this approach is made difficult by the presence of Pu^{239} in the samples available for analysis. The method consists in the measurement of the specific activity of a weighed plutonium sample of known composition and correcting for the Pu^{239} contribution. The first careful studies had of necessity to be carried out on samples of plutonium containing only a few percent of Pu^{240}. WESTRUM, HINDMAN and GREENLEE[3] obtained the value 6240±120 years. FARWELL, ROBERTS and WAHL[4] reported a value of 6300±600 years. DOKUCHAEV[23] published a value of 6620 ± 50 years.

At a later period, plutonium with a much higher Pu^{240} content became available. In Chapter 5 it is shown that plutonium samples containing as much as 45 atom percent of Pu^{240} can be made in high flux reactors. CUNNINGHAM, THOMPSON and GHIORSO,[5] using samples containing as much as 34 percent Pu^{240} redetermined the half-life and reported a value of 6650 ± 150 years. WALLMANN[6] repeated these experiments with somewhat more care and obtained the value 6760 years. Corrections were applied for appreciable amounts of Pu^{238} alpha activity in the sample. BUTLER and CO-WORKERS[7] report a value of 6600 ± 100 years.

An independent method of determining this quantity has been employed by INGHRAM, HESS, FIELDS and PYLE.[8] These workers carried out a mass spectrographic analysis of a plutonium sample which contained a substantial percentage of Pu^{240} and also mass analyzed the uranium daughters U^{235} and U^{236} which were formed by alpha decay of the plutonium isotopes over a several year period. From these ratios and from the known half-life of Pu^{239}, the half-life of Pu^{240} was calculated to be 6580 ± 40 years.

In uranium reactors some of the observed Pu^{240} results from the alternate reaction sequences:

$$U^{238}\ (n,\gamma)\ U^{239}\ (n,\gamma)\ U^{240} \xrightarrow[14\ h]{\beta^-} Np^{240} \xrightarrow[7.3\ m]{\beta^-} Pu^{240}$$

$$U^{238}\ (n,\gamma)\ U^{239} \xrightarrow[23.5\ m]{\beta^-} Np^{239}\ (n,\gamma)\ Np^{240} \xrightarrow[7.3\ m]{\beta^-} Pu^{240}$$

The first sequence, observed by HYDE and STUDIER[10] in their experiments on the discovery of U^{240} and Np^{240}, opens up the possibility of preparing isotopically pure samples of Pu^{240} by purification of neutron-irradiated uranium at the proper time after bombardment and isolation thereafter of the Pu^{240} formed. This method was used by HULET and CO-WORKERS[19] to prepare small samples of Pu^{240} for measurement of its fission cross section. The second sequence has never been specifically observed, although it undoubtedly occurs to some extent.

During the bombardment of uranium with 40-Mev helium ions, JAMES, FLORIN, HOPKINS and GHIORSO[9] observed the production of alpha activity attributed to Pu^{240} as a result of the reaction $U^{238}\ (\alpha,2n)\ Pu^{240}$ and presumably:

$$U^{238}\ (\alpha,pn)\ Np^{240} \xrightarrow[7.3\ m]{\beta^-} Pu^{240}.$$

These alpha particles had an energy hardly distinguishable from the energy of the alpha particles of Pu^{239}, but in a fission chamber the sample yielded a number of neutron-induced fissions different from a sample of pure Pu^{239}.

The energy of the main alpha-particle group of Pu^{240} is so close to that of the main 5.150 Mev group of Pu^{239} that the two cannot be distinguished when the ionization chamber – pulse height analysis method is applied. Hence, mass spectrographic or spontaneous-fission counting methods must be used to assay mixtures of the two. ASARO and PERLMAN,[11] using a magnetic spectrometer, found a main group of 5.162 ± 0.004 Mev[≠] (76 percent) and a lower energy group of 5.118 Mev (24 percent). ASARO[12] has also observed a third group at 5.014 Mev of 0.1 percent abundance. GOL'DIN, TRET'YAKOV and NOVIKOVA[13,14] report three groups: 5.1589 (75.5 percent), 5.1147 Mev (24.5 percent) and 5.004 (0.085 percent). This same Russian group[15] later restudied the Pu^{240} alpha spectrum and found an additional low intensity group plus two more tentatively assigned to Pu^{240}. Their revised alpha spectrum is given in Table 14.

It is expected that Pu^{240} will resemble all other even-even alpha emitters in this mass region. On this basis the first alpha groups undoubtedly populate the 0+, 2+ and 4+ states, respectively, of the daughter nucleus U^{236}. In addition, the excited state at 313 kev reached by the 4.851 Mev alpha group has the correct energy to be the 6+ state. These states should be de-excited by a cascade of electric quadrupole (E2) gamma rays with energies 166 kev, 102 and 45 kev. The intensity of the first two as directly deduced from the alpha group intensities are very low. TRET'YAKOV and CO-WORKERS[22] have seen conversion electrons of a gamma ray of 160 ± 1.5 kev energy in coincidence with the alpha particles of Pu^{240}. From this theory they deduce an energy of 309 kev for the 6+ level and an alpha population of (2±1) x 10^{-3}%. The 45 kev transition from the 2+ state to the ground state has been observed by several research groups.[16-18] HOLLANDER[18] found the expected conversion electron pattern for an E2 transition and measured a precise value of 45.28 ± 0.06 kev for the transition. This same transition is prominent in the electron capture decay of Np^{236}.

KONDRATEV and CO-WORKERS[15] tentatively interpret the possible levels at 210 and 239 kev (see table 14) as 1- and 3- states.

Plutonium-240 is beta stable. The spontaneous fission half-life has been reported to be 1.2 x 10^{11} years by CHAMBERLAIN, FARWELL and SEGRÈ[20], and 1.22 x 10^{11} years by BARCLAY and CO-WORKERS.[21]

≠ Asaro and Perlman in 1961 remeasured the energy of α_0 and obtained the value 5.164 Mev.

Table 14

Alpha Spectrum of Pu^{240}

Group	Energy (Mev)	Percent abundance	Energy of excited state
α_o	5.159	75.5	0
α_{45}	5.115	24.4	45
α_{147}	5.014	.091	147
α_{210}	4.952 ?	2.7×10^{-3}	210 ?
α_{239}	4.924 ?	3.1×10^{-3}	239 ?
α_{313}	4.851	3.2×10^{-3}	313

Data of Kondratev, Novikova, Sobolev and Goldin, see references 13, 14, 15.

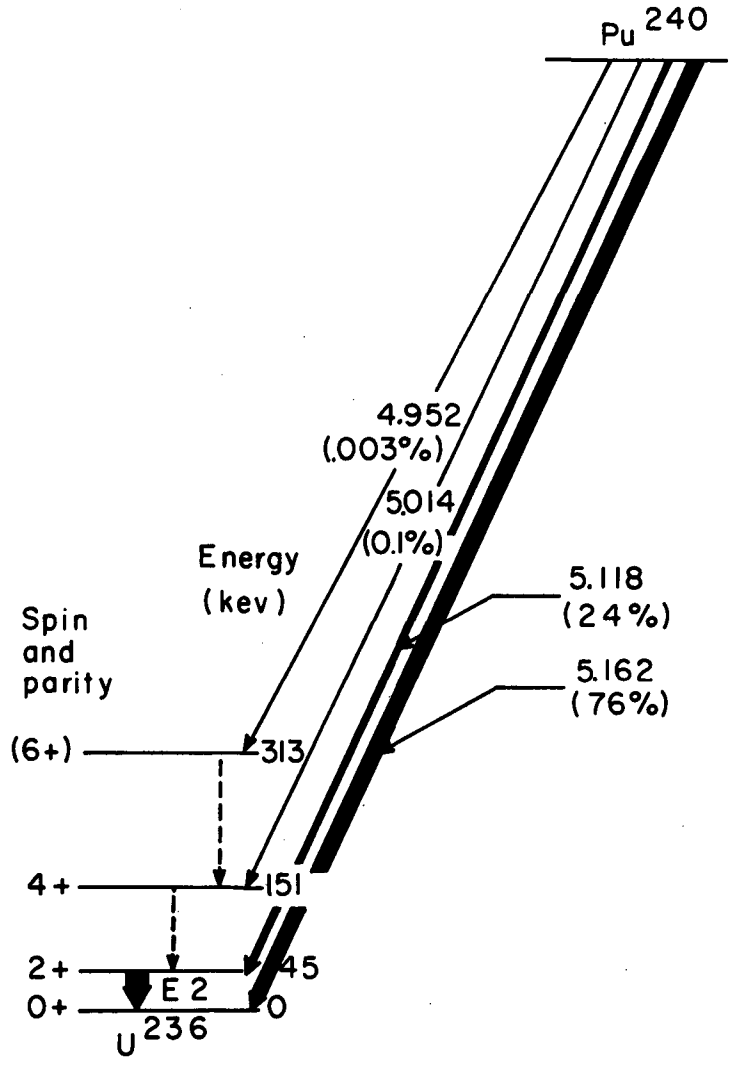

Figure 19. Decay scheme of Pu^{240}.

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9.2.11 Plutonium-241. Evidence for the presence of Pu^{241} in samples of plutonium which had been strongly irradiated with neutrons was first obtained by SEABORG, JAMES, MORGAN and GHIORSO[1,2] in the late fall of 1944. These workers isolated two new alpha emitting radioactivities from the plutonium and attributed them to Am^{241} and Cm^{242}. It was postulated that beta-decaying Pu^{241}, formed by the reaction sequence:

$$Pu^{239}\ (n,\gamma)\ Pu^{240}\ (n,\gamma)\ Pu^{241},$$

was an intermediate in the reaction sequence leading to these activities. Subsequent experiments showed that the activity attributed to Am^{241} continued to grow in the irradiated plutonium after purification of the latter, indicating that the parent Pu^{241} was long-lived. Observations on the rate of Am^{241} growth in various plutonium samples led to an early value[2] of about 10 years for the half-life of Pu^{241}. Later studies based on values determined mass spectrographically for the isotopic abundance of Pu^{241} in samples of neutron-irradiated plutonium, together with the best half-life of Am^{241}, led to the better values of 14 ± 1 years (THOMPSON and CO-WORKERS)[3] and 13.0 ± 0.2 years (MacKENZIE, LOUNSBURY and BOYD)[4] and 12.77 ± 0.28 years (ROSE and MILSTED)[5]. HALL and

MARKIN[6] remeasured the half-life of Am^{241} on which the Pu^{241} half-life directly depends and revised the last two values of the Pu^{241} half-life to 13.32 ± 0.12 and 13.04 ± 0.28 years, respectively. BROWN AND CO-WORKERS[16] report the value 13.24 ± 0.24 years.

It is possible to prepare samples of plutonium containing up to about 20 atom percent of Pu^{241} by bombardment of Pu^{239} to an integrated flux of approximately 4×10^{21} neutrons per square centimeter. Such a sample will contain about 40 atom percent of Pu^{240}, 25 atom percent of Pu^{242} and 15 atom percent of Pu^{239}. This is illustrated in Fig. 5.7 in Chapter 5. With further neutron irradiation the Pu^{241} isotopic abundance decreases and in order to obtain samples of Pu^{241} of isotopic purity approaching 100 percent it is necessary to resort to electromagnetic separation. Samples of 78 percent isotopic purity have been reported by English workers.[16,17]

The isotope Pu^{241} has also been prepared[2] in the cyclotron by the reaction:

$$U^{238}(\alpha,n)Pu^{241}.$$

It has also been isolated[12] as the daughter product of Cm^{245}.

The beta particles of Pu^{241} are of low energy. The first estimate of the maximum energy was approximately 20 kev.[2] Observations of the Pu^{241} beta particles is difficult in samples of low isotopic abundance because of interference from the intense alpha activity, the conversion electrons and the Auger electrons accompanying the disintegration of the Pu^{239}. FREEDMAN, WAGNER AND ENGELKEMEIR[7] have studied the electrons of Pu^{241} in a double lens spectrometer and report an endpoint energy of 20.5 kev; their Kurie plot exhibits an allowed shape down to 14 kev below which instrumental effects distort it. These workers also detected gamma rays of 100 and 145 kev energy which they assigned to Pu^{241}. The 100 kev peak may be K x-rays from the conversion of the 145 kev gamma ray. The intensity of the 145 kev gamma ray is only 2 photons per 10^6 Pu^{241} beta disintegrations which probably means the gamma ray is to be associated with the alpha branching of Pu^{241}. SHLIAGIN[8] reports an endpoint energy of 20.8 kev for the beta particles.

The alpha branching of Pu^{241} was established by KOHMAN, SWARTOUT and SULLIVAN[9] and by SEABORG, JAMES AND MORGAN[2] who isolated and characterized the daughter U^{237} from plutonium produced at high neutron-irradiation levels. THOMPSON, STREET, GHIORSO AND REYNOLDS[3] have observed a low-abundance group of alpha particles by analysis of ionization chamber pulses with the energy

4.91±0.03 Mev. This group was present in the plutonium samples in the amount expected for the isotope Pu^{241}. From the alpha particle intensity and the isotopic abundance the alpha branching was calculated to be ~ 0.003 percent and the alpha half-life ~ 4×10^5 years. These values are in agreement with the predictions of alpha systematics. ASARO[10] applied the more accurate magnetic deflection method to samples of plutonium with a higher Pu^{241} content and found two alpha-particle groups, 4.893 Mev (75 percent) and 4.848 Mev (25 percent) for Pu^{241}. His estimate of the alpha branching is 0.005 percent corresponding to an alpha half-life of 2.7×10^5 years. These older values for the α-half life of Pu^{241} have been improved by more recent measurements of BROWN, GEORGE, GREEN AND WATT[16] on samples containing 77 percent Pu^{241}. They reported a value of $(5.72 \pm 0.10) \times 10^5$ years.

Two groups[13,14] have calculated the alpha branching by determining the amount of U^{237} daughter activity in equilibrium with a plutonium sample of known Pu^{241} content. Both report a partial alpha half-life of $(2.9 \pm 0.5) \times 10^5$ years.

The nuclear spin of Pu^{241} has been determined to be 5/2 by the paramagnetic resonance method.[11]

HORROCKS AND STUDIER[15] describe the use of liquid scintillation techniques to measure the Pu^{241} content of plutonium samples. The lower limit of detection is 10^{-15} grams.

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9.2.12 Plutonium-242. THOMPSON, STREET, GHIORSO AND REYNOLDS[1] first observed the isotope Pu^{242} in a mass-spectrographic analysis of neutron-irradiated plutonium. The Pu^{242} content of this original sample was quite low. In modern high-flux reactors, however, it is rather simple to prepare Pu^{242} with isotopic purity of 98 percent or higher. For example, in Fig. 5.9 of Chapter 5 we note that a Pu^{239} sample irradiated at a flux of 3×10^{14} neutrons per cm^2 per second for 1.5 years is converted to plutonium which is nearly pure Pu^{242}. A typical analysis[2] for a plutonium sample irradiated to a total integrated flux of 1.4×10^{22} neutrons per centimeter^2 is given in Table 15.

Another method of preparation of Pu^{242}, the method which historically was used to provide samples for the first examination of the properties of this isotope, is based on the conversion of Am^{241} to Am^{242m} by neutron irradiation; the reaction sequence is illustrated in Figure 15. The complete decay schemes of Am^{242m} and Am^{242} are discussed later in this chapter. The Am^{241} starting material is carefully purified from plutonium and then placed in a reactor. After the irradiation the plutonium is isolated chemically from the americium. The Pu^{242} will be contaminated somewhat with Pu^{238} formed by the alpha decay of Cm^{242}. Samples of Pu^{242} 98 or higher by atom percent are formed when the irradiation time is short (i.e., a few days or less) and the plutonium isolation

Table 15. Isotopic Composition of Plutonium Prepared by Irradiation of Pure Pu^{239} to an Integrated Flux of 1.4×10^{22} neutrons/cm^2

Isotope	Abundance in Mole Percent	Composition by Alpha Activity
Pu^{238}	0.16 ± 0.02	81.9 ± 0.3
Pu^{239}	0.068 ± 0.004	5.3 ± 0.3
Pu^{240}	0.633 ± 0.006	
Pu^{241}	0.308 ± 0.006	12.8 ± 0.2
Pu^{242}	98.77 ± 0.03	
Pu^{244}	0.052 ± 0.004	

From W. C. Bentley et al., see reference 2.

step is carried out quickly after the irradiation before much of the Cm^{242} has decayed. Larger samples of Pu^{242} are produced by longer irradiation of Am^{241} at the expense of increased Pu^{238} content. This method is well described by BUTLER, LOUNSBURY and MERRITT.[3]

THOMPSON and CO-WORKERS[1] first prepared Pu^{242} by this mechanism and obtained samples which were 50 percent Pu^{242} and 50 percent Pu^{238}. Alpha-particle analysis using the ionization chamber method showed the presence of alpha particles of 4.88 Mev attributable to Pu^{242} and in an abundance corresponding to a half-life of roughly 5×10^5 years. ASARO[4] redetermined the alpha spectrum in a magnetic spectrograph and found two groups of 4.898 Mev (80 percent) and 4.854 Mev (20 percent). HUMMEL[5] checked these energies but revised the abundances to 74 percent and 26 percent, respectively. Three accurate determinations of the half-life of Pu^{242} have been made: $(3.73 \pm 0.05) \times 10^5$ years,[6] $(3.79 \pm 0.05) \times 10^5$ years,[7] and $(3.88 \pm 0.10) \times 10^5$ years.[8]

The two alpha groups of Pu^{242} define an excited state of the U^{238} daughter lying 45 kev above ground. This state is unquestionably a 2+ state representing the first state of rotational excitation of the U^{238} nucleus. When the alpha decay of Pu^{242} is re-examined it probably will be possible to detect some slight alpha branching to higher-lying levels of the ground-state rotational band of U^{238}. It is known[9] from the Coulombic excitation of U^{238} with energetic ions of A^{40} that U^{238} levels of rotational excitation are present at 44.7 kev (2+), 148 kev (4+), 310 kev (6+), 520 kev (8+), 790 kev (10+) and 1100 kev (12+).

The spontaneous fission half-life of Pu^{242} as measured by BUTLER, LOUNSBURY and MERRITT[3] on samples of 98.2 percent isotopic purity is $(6.64 \pm 0.10) \times 10^{10}$ yeras. MECH and CO-WORKERS[8] report a value of $(7.06 \pm 0.19) \times 10^{10}$ years from measurements on a sample with a similar isotopic purity.

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9.2.13 Plutonium-243. When samples of plutonium containing Pu^{242} are irradiated with slow neutrons, the isotope Pu^{243}, which decays by beta emission with a 4.98 ± 0.02 hour half-life, is formed. This was first established by SULLIVAN and CO-WORKERS[1] and confirmed by THOMPSON and CO-WORKERS[2] and O'KELLEY.[3] The cross section for formation of Pu^{243} is 18.6 barns for thermal neutrons.[4]

Determination of the beta-particle energy by absorption curve techniques[1] gave a rough value of 0.5 Mev while beta-ray spectrometer measurements[2,3] gave an early value of 0.4 Mev and a later value[5] of 0.560 Mev. Gamma rays of 95 and 120 kev were reported by early investigators.[2,3] A more detailed study by ENGELKEMEIR, FIELDS and HUIZENGA[5] showed that the beta spectrum is complex. The most energetic group, present in 53 percent abundance, has an end point energy of 566 kev. No gamma radiation is in coincidence with it. A second beta group in about 35 percent abundance has an energy of 468 kev and is followed by 85-kev gamma ray. Gamma-gamma coincidence studies showed that the 85-kev gamma ray is in coincidence with gamma rays of about 92-,107-, and 160-kev energy. The abundance of these higher energy gamma rays suggests that lower energy beta transitions with total abundance approximately 12 percent must be present. The total disintegration energy appears to be 566 kev, the energy of the most energetic beta-ray group.

STEPHENS and ASARO[6] have studied the gamma rays of Pu^{243} by scintillation spectrometer techniques and have measured conversion electrons in a permanent magnet spectrograph. The exact energy of the ~85 kev transition is 83.9 kev and from the conversion coefficient and the subshell conversion ratios it is an electric dipole transition. Another transition with energy 42.2 kev was found to be electric quadrupole in nature from the subshell conversion coefficient ratios. Gamma ray photons of a 340 kev and a 381.2 kev gamma ray were observed. Only the K-conversion line of the 381.2 kev transition was seen in the conversion electron spectrum.

STEPHENS, ASARO and PERLMAN[7] have interpreted the decay scheme of Pu^{243} in terms of the unified nuclear model and have made Nilsson orbital assignments to the ground state of Pu^{243} and to several of the levels of the Am^{243} daughter. These assignments were made on the basis of the incomplete published and unpublished information on Pu^{243}, on the data for the alpha decay of Bk^{247} to the same daughter nucleus and also to a large part on the systematic trends in the Nilsson orbitals in neighboring nuclei. These assignments are reviewed very briefly here and are summarized in the figure. More details are given in the reference cited.[7]

States in Am^{243} having spins 5/2, 7/2, and probably 9/2 seem to receive direct beta population from Pu^{243} so that a spin of 7/2 for Pu^{243} seems most reasonable. The expected Nilsson orbital is 7/2+ [624] expressed in the quantum numbers K, parity $[N\ n_z\ \Lambda]$. The 84 kev level decays by a prominent E1 transition to the ground state and by a very weak E1 transition to an ~40 kev level – presumably the first member of the ground state rotational band. Since the ground state has a measured spin of 5/2[8] this fixes the spin and parity of the 84 kev level at 5/2+ or 7/2+. Assignment is made to K = 5/2, in particular to 5/2+ [642], because the energy spacing with respect to the ground state, 5/2- [523], is similar to that seen in neptunium except that the ordering of the states is reversed. The 465 level decays by a predominantly M1 transition to the 84 kev level and to the band based on the 84 kev level. Thus the parity of the 465 kev level is even and the spin is probably 7/2 or 9/2, although 5/2 is also a possibility. The assignment 7/2+ [633] is consistent with these data and with the proposed 7/2+ spin of Pu^{243}. There is no other Nilsson level that seems to be satisfactory for this state. The level at 265 is not seen in the β decay

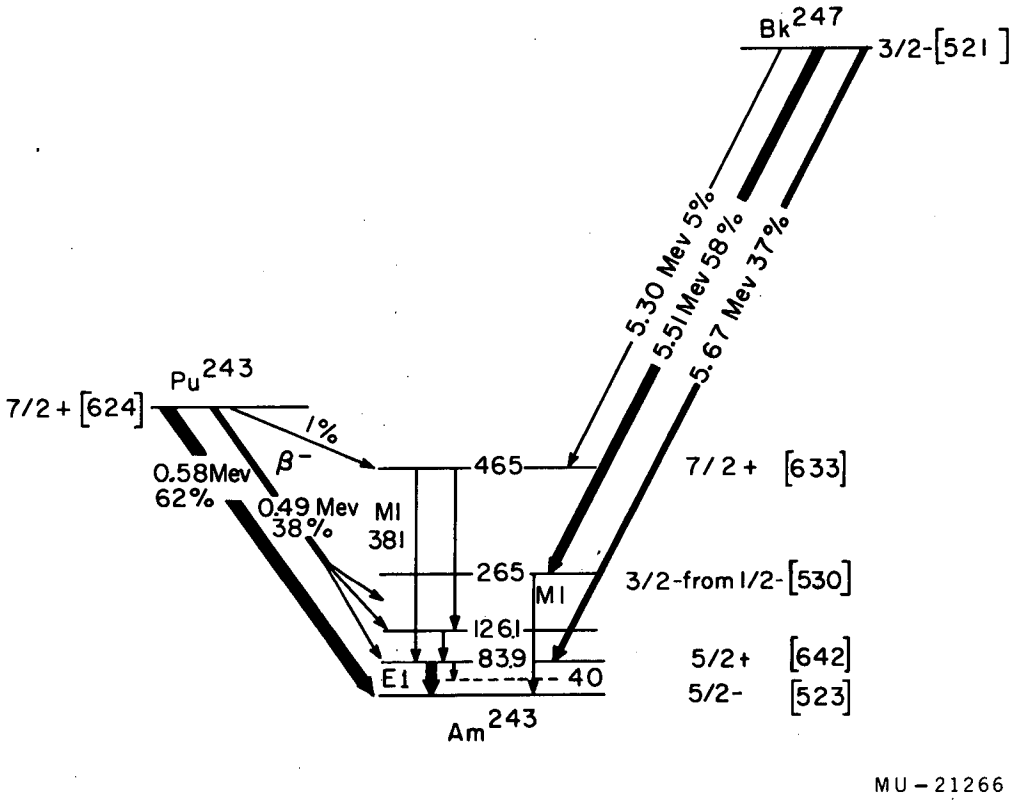

Figure 20. Decay scheme of Pu^{243} and Bk^{247} as interpreted by Stephens, Asaro and Perlman.

of Pu^{243}. From arguments not given here this level is assigned spin and parity 3/2- and may be the 3/2- level of the Nilsson state 1/2- [530]. The decay scheme is shown in the figure.

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9.2.14 Plutonium-244. In the course of an examination of the plutonium fraction isolated from the debris of a November 1952 thermonuclear test explosion the isotope Pu^{244} was detected by mass spectrographic methods by HESS, PYLE, FRIED and INGHRAM.[1] This experiment constituted the discovery of Pu^{244}. The neutron capture sequence in explosive devices of this type is discussed in Section 5.4.3 of Chapter 5. Because the entire synthesis takes place in a very brief period no beta emitting steps occur until after the initial reaction so that the usual path of buildup of higher-mass isotopes observed in nuclear reactors cannot be followed. It is concluded that Pu^{244} was formed by the successive additions of neutrons to U^{238} until the isotope U^{244} was produced; after the explosion the beta decay chain

$$U^{244} \xrightarrow[\text{short}]{\beta^-} Np^{244} \xrightarrow[\text{short}]{\beta^-} Pu^{244}$$

gave rise to the long-lived Pu^{244}. The isotope Pu^{244} has also been found in small isotopic abundance by the mass spectrographic analysis of Pu^{239} samples which had been irradiated with an integrated flux of 4×10^{21} neutrons.[2] The radiations of Pu^{244} could not be studied with this sample because of its long

half-life and the presence of the other plutonium isotopes. The reaction sequence leading to the formation of Pu^{244} is summarized in Fig.21. It will be observed that Pu^{244} is off the main course of the neutron capture sequence and is formed only by side reactions. The greater part of the yield comes from the $Pu^{243}(n,\gamma)Pu^{244}$ side reaction for which the cross section is about 170 barns, but the contribution of the electron-capture decay of Am^{244} is appreciable. This contribution increases as the irradiation is prolonged and the Am^{243} builds up in the sample.

Table 16 indicates how the Pu^{244} content builds up in Pu^{239} irradiated at a flux of 3×10^{14} nucleons per cm^2.

The presence of Pu^{244} in such samples of neutron-irradiated plutonium has been proven[5,6] by isolation of the U^{240} formed by its alpha decay. Uranium-240 could be identified by the known radiations of U^{240} in equilibrium with its Np^{240} daughter. An interfering activity in this "milking" experiment is U^{237} which forms from the alpha decay of Pu^{241} present in the plutonium in much greater abundance. This is taken care of by use of sufficient absorber over the uranium daughter fraction to cut out the weak beta radiation of U^{237} and U^{240}; while permitting the hard radiations of Np^{240} to pass through. Alternatively, the Np^{240} can be chemically isolated.

It is possible[3] to prepare plutonium samples with a much higher percentage of Pu^{244} by neutron irradiation of Am^{243} (see Fig.22).

By quantitative separation and counting of the U^{240} daughter activity in equilibrium with a plutonium sample whose Pu^{244} content was known from mass spectrographic analysis the alpha half-life of Pu^{244} has been found by DIAMOND and BARNES[5] to be $(7.6 \pm 2) \times 10^7$ years, and by BUTLER and CO-WORKERS[6] to be $(7.5 \pm 2) \times 10^7$ years.

Since Pu^{244} is an <u>even-even</u> isotope it is expected to have an appreciable half-life for decay by spontaneous fission; this quantity has been determined[3,4] as $(2.5 \pm 0.8) \times 10^{10}$ years.

The cross section of Pu^{244} for capture of thermal neutrons has been measured as 1.5 ± 0.3 barns[4] and as 2.1 ± 0.3 barns.[6]

DIAMOND and BARNES[5] conclude from the limits of error on their measurement of the half-life of Pu^{244} and from an estimate of its primeval abundance that Pu^{244} might still be present in barely detectable amounts in the earth's crust. They also conclude that Pu^{244} might have been an important heat source in the early history of the earth. Later considerations and calculations by KOHMAN[7] indicate that the heat contribution by Pu^{244} was never very significant compared to other radionuclides.

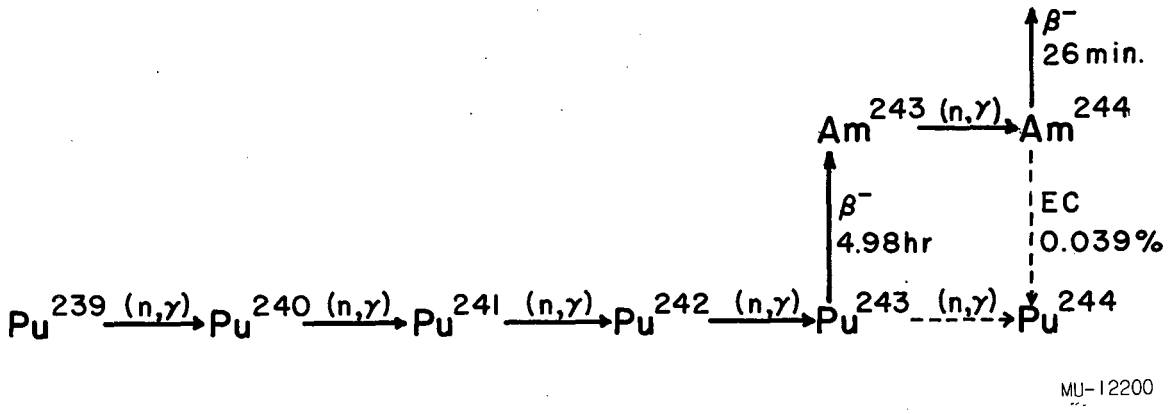

Figure 21. Reaction sequence leading to the formation of Pu^{244}. Note that Pu^{244} does not lie on the main course of the reaction sequence shown by dark arrows.

Table 16

Pu^{244}/Pu^{242} Ratios in Irradiated Pu^{239}.
Data from references 3 and 4

Integrated flux	Pu^{242} abundance in atom percent	Pu^{244} abundance in atom percent	Pu^{244}/Pu^{242}
4×10^{21}	34.1	0.0018	5.28×10^{-5}
1.1×10^{22}	96.33	0.037	3.84×10^{-4}
1.4×10^{22}	98.77	0.052	5.26×10^{-4}

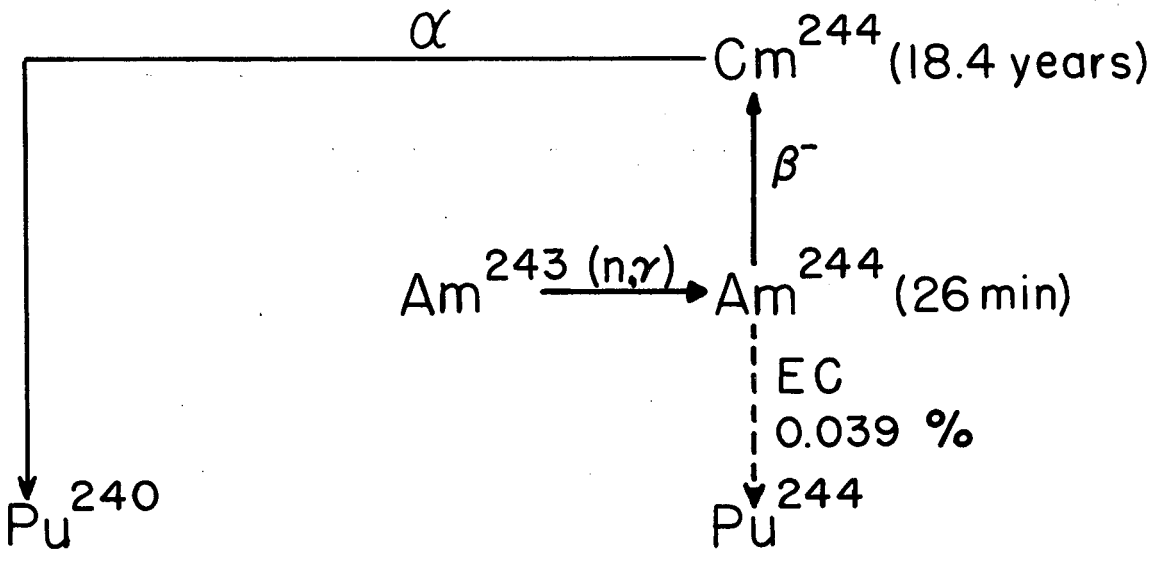

Figure 22. Preparation of Pu^{244} by neutron irradiation of Am^{243}.

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7. T. P. Kohman, paper presented at 1960 Spring meeting of Amer. Chem. Soc., Cleveland, Ohio, April 1960.

9.2.15 Plutonium-245. BROWNE and CO-WORKERS[1] and FIELDS and CO-WORKERS[2] first reported the existence of a 10.6-hour beta emitter assignable to Pu^{245}. This isotope is prepared by neutron irradiation of plutonium targets containing Pu^{244} or of other heavy element targets which can be converted to Pu^{244} and Pu^{245} by multiple neutron-capture reactions. The radiation of Pu^{245} itself were not studied by these authors because of conflicting radiations in the plutonium fractions. The half-life was measured by repeated isolation of the daughter, 2.08 hour Am^{245}, whose radiations are discussed in Section 9.3.10. BUTLER and CO-WORKERS[3] confirm the production of Pu^{245} from plutonium samples containing Pu^{244}.

REFERENCES - Pu^{245}

1. C. I. Browne, D. C. Hoffman, W. T. Crane, J. P. Balagna, G. H. Higgins, J. W. Barnes, R. W. Hoff, H. L. Smith, J. P. Mize, and M. E. Bunker, J. Inorg. Nuclear Chem. 1, 254 (1955).
2. P. R. Fields, M. H. Studier, A. M. Friedman, H. Diamond, R. Sjoblom and P. A. Sellers, J. Inorg. Nuclear Chem. 1, 262 (1955).
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9.2.16 Plutonium-246. During the course of the examination of the plutonium fraction isolated from the debris of the November, 1952, thermonuclear test explosion, the previously-unknown isotope Pu^{244} was detected by mass spectrometric methods.[1] This stimulated further investigation of the plutonium fraction for isotopes of higher mass and resulted in the discovery of the 11.2-day isotope, Pu^{246}, by groups of scientists at the Los Alamos Scientific Laboratory and at the Argonne National Laboratory.[2]

Examination of the radiations of the plutonium fraction indicated the presence of two beta activities, one of approximately 0.15 Mev and the other of 1.2 Mev energy. By chemical separation procedures it was possible to show that the more energetic beta-emitter was an isotope of americium with a half-life of 25 ± 0.2 minutes. The chemical identification was made certain by detailed chemical experiments during which the new activity was not separated from added Am^{241} tracer. The 25-minute americium isotope was proved to be the daughter of the 11.2-day beta emitter. This latter activity was proved to be a plutonium isotope by exhaustive chemical steps.

From the known properties of the plutonium isotopes it was certain that the mass number of the new 11.2-day isotope could not be less than 244. Several lines of evidence conclusively ruled out the mass number 244 and made the most likely assignment, 245 or 246. The mass spectrographic analysis results reported later[3] established that the correct mass assignment was 246.

ENGELKEMEIR and CO-WORKERS[2] reported that Pu^{246} emits gamma rays of 43, 111, 175 and 224 kev energy. The gamma rays of the 25-minute Am^{246} are also emitted by a sample of Pu^{246} because the daughter activity quickly comes to equilibrium with the parent.

HOFFMAN and BROWNE[4], and SMITH and CO-WORKERS[5] restudied the radiations of Pu^{246} using scintillation spectrometers and gamma-gamma and beta-gamma coincidence spectrometers. They report complexity in the beta spectrum with 73 percent of the disintegrations going by means of a 150-kev beta transition to a 249-kev level in Am^{246}. This level is deactivated mainly through a 175 kev - 47 kev - 27 kev gamma ray cascade, although alternate gamma ray sequences are also detected. This leads to a total disintegration energy of 400 ± 30 kev. In 27 percent of the transitions the beta ray energy is 330 kev. There may be higher energy beta particles in low abundance. Evidence for gamma rays of 100 kev and 225 kev was also found.[5] No decay scheme has been formulated for Pu^{246}. HOFFMAN AND BROWNE[4] report the value 10.85 ± 0.02 days for the half life.

REFERENCES - Pu^{246}

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2. D. Engelkemeir, P. R. Fields, S. Fried, G. L. Pyle, C. M. Stevens, L. B. Asprey, C. I. Browne, H. Louise Smith and R. W. Spence, J. Inorg. Nuclear Chem. 1, 345 (1955).
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4. D. C. Hoffman and C. I. Browne, Nuclear Chem. 2, 209 (1956).
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9.3 THE ELEMENT AMERICIUM (ELEMENT 95)

9.3.1 The Discovery of Americium. The element americium was discovered in experiments conducted late in 1944 and early in 1945 at the Wartime Metallurgical Laboratory (now Argonne National Laboratory) of the University of Chicago by SEABORG, JAMES, MORGAN, and GHIORSO.[1,2] The discovery of element 95 followed shortly after the discovery of curium (element 96). These investigators found that a long-lived, alpha-emitting isotope could be separated chemically from plutonium which had received extensive irradiation with neutrons. From the known flux and time of irradiation and the predicted properties of Pu^{239} and Pu^{240} it was clear that the reaction sequence responsible for the results was the following:

$$Pu^{239}(n,\gamma)Pu^{240}(n,\gamma)Pu^{241} \xrightarrow[\text{long}]{\beta^-} 95^{241} \xrightarrow[\text{long}]{\alpha}$$

The plutonium was purified repeatedly and the growth of the new alpha emitter into the purified plutonium was clearly established. Later irradiation of samples of the supposed 95^{241} with neutrons resulted in the production of the known isotope Cm^{242}.

Further proof of the mass assignment came from helium ion bombardments of U^{238} targets. A beta-emitting plutonium isotope of long half-life was produced with a yield which varied in a manner characteristic of an (α,n) reaction as the energy of the helium ions striking the target was changed.

$$U^{238}(\alpha,n)Pu^{241} \xrightarrow[\text{long}]{\beta^-} 95^{241}$$

Numerous chemical experiments were carried out to determine the properties of the new long-lived alpha-emitter assigned to 95^{241} and to provide the necessary proof that this activity could be separated chemically from all known elements. The chemical properties in aqueous solution were found to be those of a tripositive ion and in almost every particular to be very similar to those of lanthanide element ions. Indeed, considerable time elapsed before a completely satisfactory method for the separation of elements 95 and 96 from each other and from the rare earth elements was developed. Vigorous attempts to convert 95^{241} to a lower or a higher oxidation state were unsuccessful although many years later it was found that it could be converted to the oxidation states

V and VI under suitable conditions. This oxidation is easier to perform with macro quantities of the element but it can be done with tracer quantities.

The similarity of elements 95 and 96 to the rare-earth elements provided the first substantial clue that the new transition series of elements in the heavy element region should be considered to be an actinide series of elements. Indeed it was the adoption of this view as a working hypothesis[3] that lead directly to the identification of Cm^{242} and Am^{241} after a number of fruitless experiments had been carried out under the assumption that elements 95 and 96 would resemble neptunium and plutonium in being readily oxidized to the (VI) oxidation state.

Element 95 was named americium (symbol Am) after the Americas on the basis of its position as the sixth member of an actinide rare-earth series, analogous to europium of the lanthanide rare earths.

Americium was first isolated in the form of a pure compound by CUNNINGHAM[4] who carried out the first measurement of the half-life by direct measurement of specific activity. This work was done with only a few micrograms of material. From the weight of this sample and from its alpha counting rate a half-life of 498 years was determined, which is very close to the modern value of 458 years. Since then americium in the form of 458 year Am^{241} has been produced in gram quantities from reactor produced plutonium. The many studies of the chemistry of the element which have been performed with this isotope are described elsewhere.[5,6]

The specific activity of Am^{241} is 7×10^{13} disintegrations per minute per gram so that the study of the chemistry of americium is severely hindered by the remarkable chemical effects caused by the intense alpha radiations. The isotope Am^{243} with a half-life of 7930 years is more suitable for chemical studies and is certain to come into more widespread use as milligram and larger quantities become more generally available.

The nuclear properties of the individual isotopes of americium are described in the following pages of this chapter.

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9.3.2 Americium-237. HIGGINS[1] found evidence for a 1.3 hour isotope Am^{237} formed with 30 to 50 Mev deuterons by the reaction $Pu^{239}(d,4n)Am^{237}$. It decays chiefly by electron capture, but it also emits 6.01 Mev alpha particles to the extent of about 0.005 percent. This isotope has not been carefully studied.

REFERENCES Am^{237}

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9.3.3 Americium-238. STREET, GHIORSO and SEABORG[1] found that the bombardment of Pu^{239} with 50-Mev deuterons results in the production of an americium activity of about 1.2-hour half-life in addition to 12-hour Am^{239} and 50-hour Am^{240}. Later work by HIGGINS[2] suggested that this activity was mixed with Am^{237} and that the half-life of Am^{238} is actually about 2.1 hours. CARR, GLASS, GIBSON and COBBLE[3] report a half-life value of 1.86 ± 0.09 hours. Reactions by which Am^{238} is prepared include:

$$Pu^{239}\ (d,3n)\ Am^{238}$$
$$Pu^{239}\ (p,2n)\ Am^{238}$$
$$Np^{237}\ (\alpha,3n)\ Am^{238}$$

Peak yields are listed for these reactions in Table 5.5, Chapter 5.

The principal mode of decay of Am^{238} is orbital electron capture for which ~2.24 Mev of decay energy is available. An upper limit of 3×10^{-4}% has been set on the alpha branching.[2] CARR, GLASS, and GIBSON[3] have studied the gamma rays of Am^{238} with scintillation spectrometers by use of gamma-gamma coincidence techniques. They observed gamma rays with energies 370, 580, 950, 980 and 1350 kev. There is a 580-370-980 cascade, a 580-1350 cascade and a 950-980 cascade. These results define the decay scheme shown in the figure. Electron capture proceeds mainly to the Pu^{238} levels at 980 and 1930 but a few percent of the transitions may lead to other high-lying levels.

A striking feature of this decay scheme compared to the decay of Np^{238} is that none of the several known excited levels of Pu^{238} below 900 kev appear to be reached directly or indirectly. A great deal of information on the levels of Pu^{238} is available from the beta decay of Np^{238} (Section 9.1.11) and the alpha decay of Cm^{242} (Section 9.4.6).

REFERENCES Am^{238}

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3. R. J. Carr, R. A. Glass, and W. M. Gibson, [illegible], J. Inorg. Nuclear Chem. 13 181 (1960).

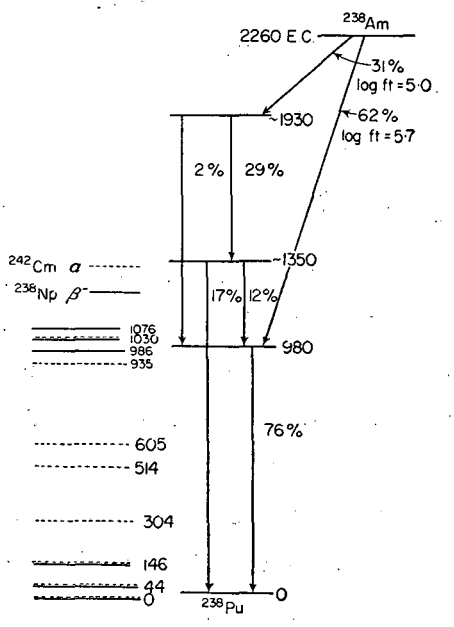

Figure 23. Preliminary decay scheme of Am^{238}, as formulated by Glass, Carr and Gibson.[3] Gamma-ray intensities and electron capture branching intensities are based on the assumption that one K X-ray is emitted per disintegration. Seven percent of the transitions are unaccounted for because of errors in the data or in the above assumption or because of unobserved branching to a low-lying state of Pu^{238}. The Pu^{238} levels on the left are those populated from Np^{238} β^- decay or from Cm^{242} α-decay. The exact alignment of these levels with those seen in the decay of Am^{238} is not certain due to uncertainties in gamma-ray energies and in the precise levels near the ground state populated by the high energy gamma-rays.

9.3.4 Americium-239. The 12-hour radioactivity Am^{239} was first prepared by SEABORG, JAMES and MORGAN[1] by the following reactions:

$$Pu^{239}\ (d,2n)\ Am^{239}$$
$$Np^{237}\ (\alpha,2n)\ Am^{239}.$$

The Am^{239} so prepared is not pure because it is contaminated with 53 hour Am^{240}. Other reactions which have been used for the preparation are:[2]

$$Pu^{239}\ (p,n)\ Am^{239}$$
$$Pu^{239}\ (\alpha,p3n)\ Am^{239}$$

Some cross sections for these reactions are listed in Table 5.5 of Chapter 5.

This isotope decays almost entirely by orbital electron capture although an alpha branching of 0.003 percent was reported by HIGGINS.[3] This was confirmed by GLASS, CARR AND GIBSON[10] who redetermined the branching as $(5.0 \pm 1.0) \times 10^{-3}$ corresponding to a partial alpha half-life of 28 ± 5 years. The alpha particle energy has been reported as 5.78 Mev[4] and as 5.77 Mev[10]. STEPHENS AND CO-WORKERS[5,7] found the alpha particles of Am^{239} to be in coincidence with a 48 kev E1 transition indicating that the alpha decay of Am^{239} is similar to that of Am^{241} and Am^{243}. This similarity is shown in Fig. 24 The presence of low-lying states of opposite parity from that of the ground state is an interesting feature of the daughter neptunium isotopes in these three cases. The nuclear states of Np^{235} and of Am^{239} have been assigned to specific quantum states of single particle motion by STEPHENS, ASARO AND PERLMAN[9]. The assignments are based on the great similarities of the decay of Am^{239} to that of Am^{241} for which Nilsson state assignments have been made on a variety of firm evidence. These Nilsson assignments are given in the figure. In all probability the alpha spectrum of Am^{239} is complex but a detailed study is made difficult by the low alpha branching and short half-life of the isotope.

Americium-239 is the daughter product of the isotope, 2.5-hour Cm^{239}, and can be prepared in small quantities free of contaminating Am^{240} activity by isolation of americium from a curium sample containing Cm^{239}, since the Cm^{240} shows no observable decay by orbital electron capture to Am^{240}.

The radiations of Am^{239} include K and L x-rays and a number of gamma ray photons and conversion electrons. SMITH, GIBSON and HOLLANDER[6] have made precision measurements of the conversion electrons with permanent magnet spectrographs and have discussed the decay scheme of Am^{239}. GLASS, CARR AND GIBSON[10] used scintillation spectrometers to measure the gamma rays singly and in coincidence. The electron capture decay of Am^{239} proceeds to a number of the same excited levels of Pu^{239}

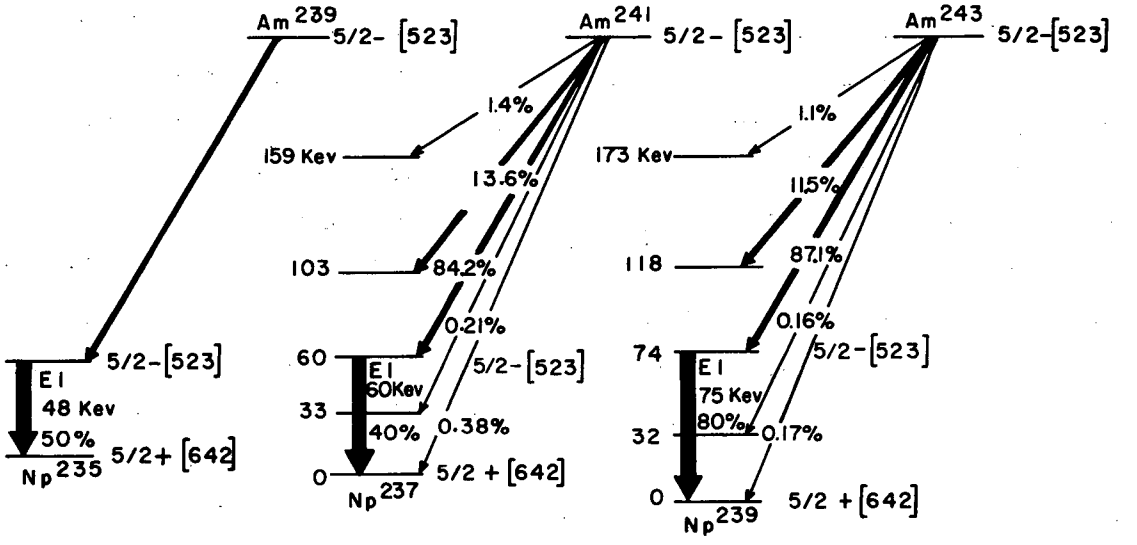

Figure 24. Comparison of the decay schemes of Am^{243}, Am^{241} and Am^{239}. Some Nilsson state assignments are given. Quantum numbers of these states are K, parity $[N\ n_z\ \Lambda]$.

that are reached in the beta decay of Np^{239} and the alpha decay of Cm^{243}. This is shown clearly in Fig. 8 in Section 9.1.11. The interpretation of the level scheme of Pu^{239} is discussed earlier in this chapter in connection with the radiations of Np^{239}.

The gamma rays of Am^{239} are listed in the table. The placement of these gamma rays in the decay scheme is shown in Fig. 8 accompanying the description of Np^{239} in section 9.1.11. HOLLANDER[8] has discussed the operation of selection rules in the asymptotic quantum numbers as they affect the electron capture decay of Am^{239}.

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10. R. A. Glass, R. J. Carr, and W. M. Gibson, J. Inorg. Nucl. Chem. 13, 181 (1960).

Table 17. Gamma Rays of Am^{239} from Conversion Electron Data*

Energy (kev)	Multipole order
44.70	M1 + E2
49.47	M1 + E2
57.31	E2
67.91	E2
181.8	M1
209.9	M1
226.5	M1
228.3	M1
277.6	M1

*See SMITH, GIBSON and HOLLANDER, reference 6.

9.3.5 Americium-240. The 53-hour isotope Am^{240} was first produced by deuteron bombardment of Pu^{239} and helium ion bombardment of Np^{237}.[1,3]

$$Pu^{239}\ (d,n)\ Am^{240}$$
$$Np^{237}\ (\alpha,n)\ Am^{240}$$

It may also be produced by the reaction:[1,2]

$$Pu^{239}\ (\alpha,p2n)\ Am^{240}.$$

The Am^{240} prepared in these ways is contaminated with 12-hour Am^{239}.

The main mode of decay of Am^{240} is orbital electron capture. Alpha decay is energetically possible but has not been observed; an upper limit of 0.2 percent has been set.[2] This is consistent with an estimated partial alpha half-life of 10^3 years. The Am^{240} is believed to be stable with respect to β^- decay to the extent of a few kilovolts. Because of the uncertainty regarding this, CARR[6,7] has searched for the Cm^{240} product of a possible beta transition in Am^{240} and has set an upper limit of 6×10^{-6} for the β^- branching; this corresponds to a lower limit of 1×10^5 years for the beta half-life.

The radiations of Am^{240} consist of x-rays, gamma rays and conversion electrons. The gamma transitions are of interest since they delineate the excited levels of the even-even nucleus Pu^{240}. The results can be compared to the level system deduced from the alpha decay of Cm^{244} and the β^- decay of Np^{240}. (See Sections 9.4.8 and 9.1.12). SMITH, GIBSON and HOLLANDER[4] reported measurements on the 4+ —> 2+ —> 0+ ground state gamma ray cascade in Pu^{240} following electron capture of Am^{240}. They saw L_{II}, L_{III}, M_{II}, M_{III}, N and O electrons corresponding to a 42.87-kev transition (the 2+ —> 0+ transition) and L_{II}, L_{III}, M_{II}, M_{III}, N and O electrons from a 98.90 ± 0.2 kev transition (the 4+ to 2+ transition). Both transitions are E2. The levels of Pu^{240} deduced from this information are a 2+ first excited state at 42.88 kev and a 4+ second excited state at 141.8 kev. These are members of a rotational band of levels based on the 0+ ground state. The constant $h^2/2\Im$ for the rotational spacing is 7.16 kev. GLASS[CARR AND GIBSON 5,7] have found additional gamma rays of 0.90, 1.00 and 1.40 Mev by scintillation spectrometer studies. These are similar to gamma rays seen in the beta decay of Np^{240} to the same daughter nucleus Pu^{240}. These gamma rays are believed to de-excite levels of Pu^{240} at

0.900, 1.000 and 1.40 Mev populated directly by electron capture. It is a striking feature of the decay of Am^{240} that all or nearly all of the transitions proceed to levels lying above 900 kev even though many lower-lying levels of the ground state rotational band covering a wide range of spins are available. In this respect Am^{240} resembles Am^{238}.

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5. R. A. Glass, Ph.D. Thesis, University of California, April, 1954; also published as University of California Radiation Laboratory Report, UCRL-2560, April, 1954.
6. R. J. Carr, Ph.D. Thesis, University of California, April, 1956; also published as University of California Radiation Laboratory Report, UCRL-3395, April 1, 1956.
7. R. A. Glass, R. J. Carr and W. M. Gibson, J. Inorg. Nucl. Chem. 13, 181 (1960)

9.3.6 Americium-241. This isotope was first[1] produced by the reaction sequence: $Pu^{239}(n,\gamma)\,Pu^{240}(n,\gamma)\,Pu^{241} \xrightarrow[13\ year]{\beta^-} Am^{241}$, and this remains the best method for its preparation. In some laboratories a sample of plutonium which has been strongly irradiated with neutrons is set up as a source or "Cow". The Am^{241} which builds up in this source is periodically removed by some simple chemical operation and the plutonium is set aside to produce more americium.[5] Plutonium sources containing several percent Pu^{241} can be made by neutron irradiation of Pu^{239}. The Am^{241} can also be isolated from the reactor fuel elements during chemical processing but this, for many reasons, is a more troublesome way to obtain large samples of this isotope. The amount of Am^{241} formed in a sample of plutonium containing one gram of Pu^{241} is shown in Fig. 25 as a function of the growth time.

The half-life of Am^{241} was determined originally by CUNNINGHAM[2] as 510 years. HALL AND MARKIN[3] have revised this value to 458.1 ± 0.5 years on the basis of careful determinations of the specific alpha activity of the compounds $Am_2(SO_4)_3$ and $AmCl_3$. /WALLMANN, GRAF AND GODA[4] check this closely with a value of 457.7± 1.8 obtained by specific activity measurements on a weighed sample of pure americium metal. For some reason the value obtained using the compound AmO_2 was slightly higher. Am^{241} is stable with respect to orbital electron capture or beta decay and decays entirely by the emission of alpha particles.

The Alpha Spectrum of Am^{241}. The complex alpha particle spectrum of Am^{241} has been measured carefully by the magnetic deflection technique by three groups of experimentalists.[6,9,10] The closely agreeing results are summarized in Table 18.

These alpha data define several excited levels in the daughter nucleus Np^{237} and assist in the placement of the observed gamma rays in a decay scheme. Additional assistance in this task is obtained from a study of the β^- decay of U^{237} (see section 8.4.10) and the decay of Pu^{237} by orbital electron capture (see section 9.2.000), both of which lead to the same Np^{237} daughter nucleus. In addition, the gamma rays observed in the Coulombic excitation of Np^{237} have been of great help in the interpretation of the decay scheme. Figs. 26 and 27 summarize the level scheme of Np^{237} as deduced from these four distinctly different studies. The work of many experimentalists on the gamma radiations of Am^{241} are summarized in the decay schemes and in the following paragraphs. All the observed low-lying levels of Np^{237} are neatly explained as consisting of rotational bands of levels based on four states of intrinsic excitation. The discussion of the decay scheme will be reserved until some of the more important

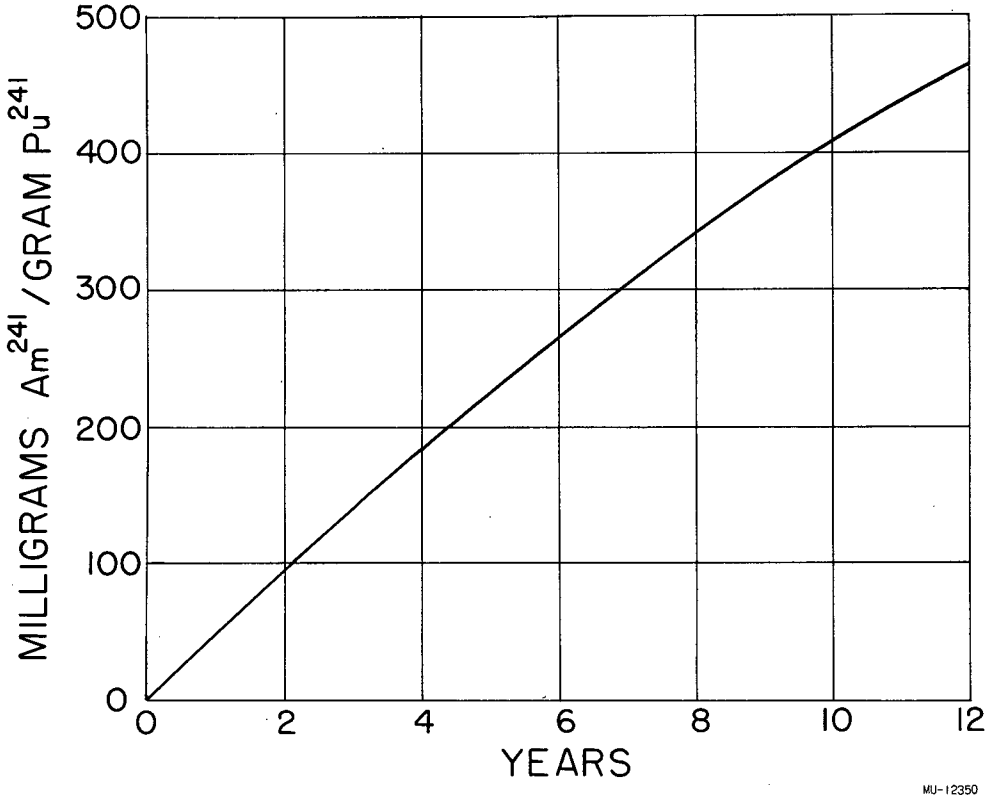

Figure 25. The number of milligrams of Am^{241} produced in a sample of plutonium containing one gram of 13-year Pu^{241} as a function of time since last removal of americium.

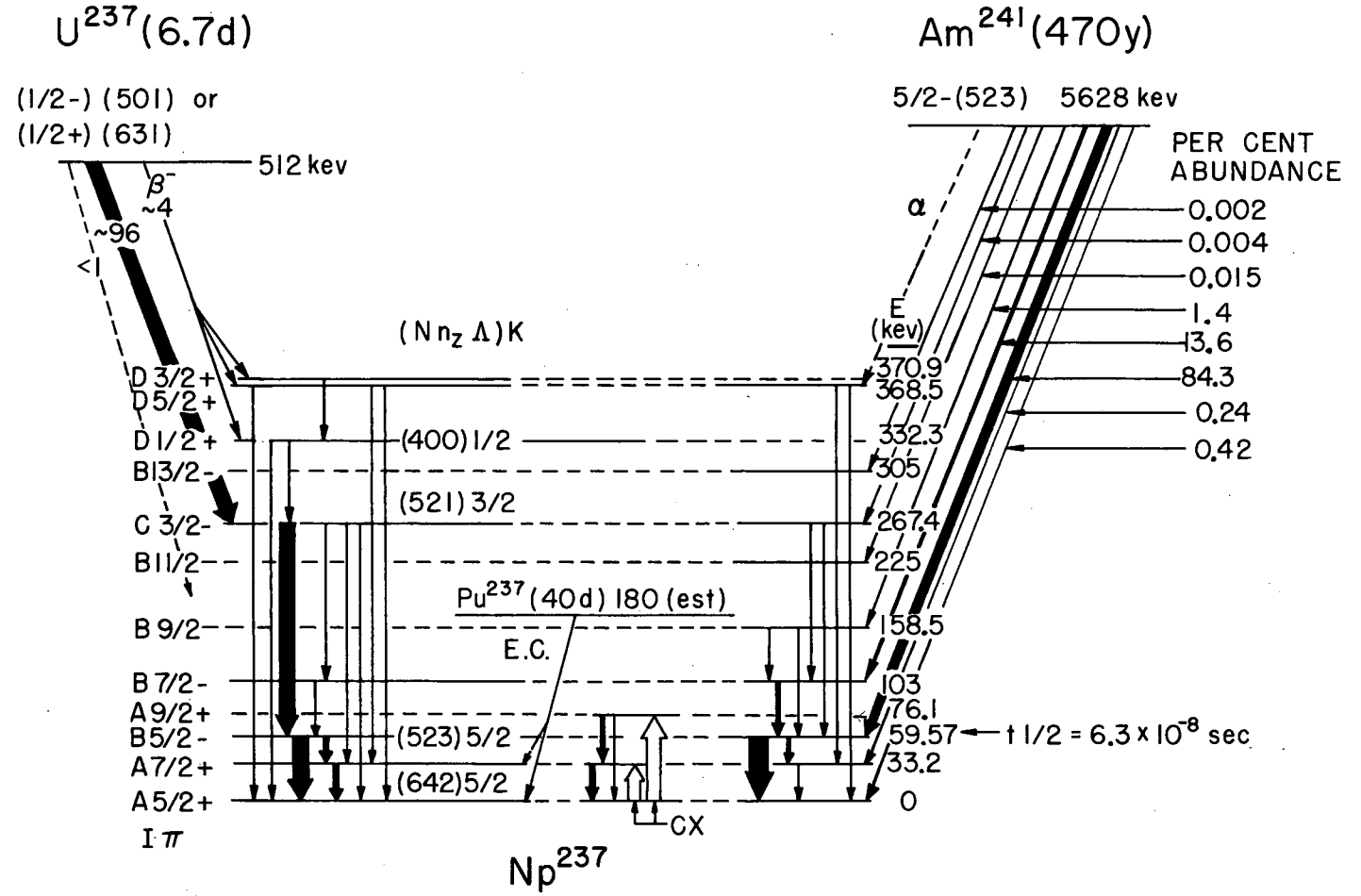

Figure 26. Alpha decay scheme of Am^{241}, beta decay scheme of U^{237} and electron capture decay scheme of Pu^{237} to the common daughter nucleus Np^{237}. Also shown is the coulombic excitation scheme. The vertical arrows in each scheme are drawn only for experimentally observed transitions. Proposed assignments of the asymptotic quantum numbers (N, n_z, Λ, K) are given as well as spins and parities. The decay scheme of U^{237} is displayed in somewhat more detail in 8.27 of chapter 8.

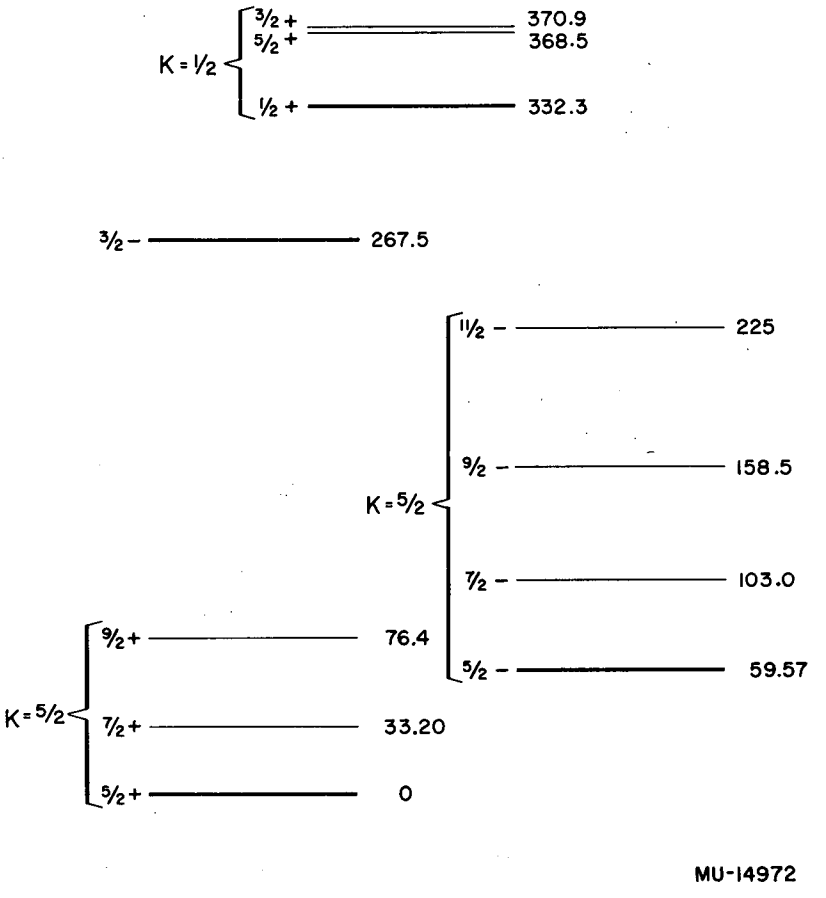

Figure 27. Level scheme of Np^{237} as drawn by Hollander, Rasmussen, Albridge and Smith.

TABLE 18: Alpha Particle Groups of Am^{241}

ASARO, REYNOLDS AND PERLMAN[9]			GOLDIN, NOVIKOVA AND TRETYAKOV[10]			ROSENBLUM, VALADARES AND MILSTED[6]	
Alpha particle energy (Mev)	Relative abundance (%)	Excited state energy	Alpha particle energy (Mev)	Relative abundance (%)	Excited state energy	Alpha particle energy (Mev)	Relative abundance (%)
5.535	0.42	0	5.541	0.39	0	5.534	0.35
5.503	0.24	33	5.508	0.24	33.1	5.500	0.23
5.476	84.3	59.6	5.482	85.0	59.8	5.477	85.1
5.433	13.6	103	5.439	12.8	103.2	5.435	12.6
5.379	1.4	158.6	5.386	1.66	157.2	5.378	1.74
---	---	---	5.321	0.015	224	5.311	0.013
5.27*	0.004	267.4	---	---	---		
---	---	---	5.241	0.002	305		

* Not directly observed - deduced from α - γ coincidence.

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experimental data on the gamma transitions are summarized.

The Gamma Transitions of Am^{241}. The low energy gamma rays have been studied carefully several times with a bent crystal spectrometer, an instrument which permits measurement of photon energies with great precision.[11,12] The results of DAY[12] are given in Table 19. The conversion electrons of these same transitions have been studied by several groups. The precise measurements of BARANOV AND SHLYAGIN[14] with a double-focusing spectrometer as well as the precise measurements of HOLLANDER, SMITH AND RASMUSSEN[16] and of ROSENBLUM, VALADARES AND MILSTED[6] made with permanent magnet spectrographs are summarized in the same table. Other pertinent references are listed in the bibliography.[13,15] The proportional counter measurements of BELING, NEWTON AND ROSE[17] were also important in determining photon energies and abundances. In particular, the absolute abundance of the 59.57-kev gamma ray was determined by them to be 0.40 ± 0.015 photons per alpha. This measurement is important because this transition is the most prominent gamma ray in the decay of Am^{241} and because the 59.57-kev gamma ray is a widely used standard in nuclear spectroscopy for calibrating the energy scale and efficiency (or geometry) of counting equipment. MAGNUSSON[22] remeasured this quantity and got the slightly lower value of 0.359 photons per alpha.

The discussion in several published articles[6,11,16] may be consulted for detailed arguments leading to the following multipolarity assignments for these low energy gamma rays: 26.28 kev (E1), 33.2 kev (M1 + E2), 43.4 kev (E2 + M1), 59.57 kev (E1) and 99.0 kev (E2).

L x-rays of neptunium are produced in considerable abundance as a result of the emission of conversion electrons in the decay of Am^{241}. The accurate measurements of DAY[12] made on a bent-crystal spectrometer are presented in Table 20. When instruments of lesser resolution such as the proportional counter spectrometer are used, only the more abundant groups appear. The measurements of BELING, NEWTON AND ROSE[17] and of MAGNUSSON[22] on the NpL_α, NpL_β and NpL_γ groups of x-rays are summarized in Table 20. Scintillation spectrometer measurements of high energy gamma rays are listed in Table 21.

The Decay Scheme of Am^{241}. We are now in a position to discuss the decay scheme of Figs. 26 and 27. We follow the discussion of PERLMAN AND RASMUSSEN[21] and others and interpret the decay scheme in terms of the Bohr-Mottelson unified model. The levels at 33.20 and 76.4 kev constitute a series of rotational levels based on the 5/2+ ground state. In the theory of Bohr and Mottelson the coupling of single particle motions and collective motions in a region of strong interaction leads to a limiting equation representing the energies of rotational levels in odd A nuclei (valid except for K = = 1/2) which is

TABLE 19: Gamma Transitions of Am^{241} (Low Energy Region)

Energy "Best" Value	Cauchois Bent-Crystal Spectrometer Results (Reference 12)		Conversion Electron Measurements				Proportional Counter Results		
	Energy (kev)	Photon Relative Intensity	Transition Energy	Conversion Electrons seen	Transition Type	Ref.	Energy	Photon Intensity per alpha	Ref.
	26.363±0.014	8.2	26.38	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{I}$	E1	14			
26.36			26.36	M_{I}		16	26.3	0.028	17
			26.34	$L_{I\ II\ III}\ M_{I}\ N_{I}$	E1	6	26.4	0.025	22
33.20	33.199±0.021	0.5	33.22	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{I}$	M1+E2	14			
			33.20	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{II}\ N_{I\ II\ III\ IV}\ O_{I\ II}$		16			
			33.14	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{I}\ O_{I}$	M1+E2	6			
43.46	43.463±0.085	0.6	43.43	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{I}\ O_{I}$	E2+M1	14	43.4	0.00073	22
			43.20	$L_{I\ II\ III}\ M_{I\ II\ III}\ N_{I\ II\ III}\ O_{I\ II\ III}$		16			
			43.34	$L_{I\ II\ III}\ M_{III}\ N_{III}\ O_{I}$	M1+E2	6			
			55.52	$L_{I\ II}\ M_{I\ III\ IV}$		14			
			55.56	$L_{I\ II\ III}\ M_{I\ II\ III}$		16			
			55.46	$L_{I\ II}\ M_{I\ II\ III}$	M1+E2	6			
59.57	59.568±0.017	100	59.62	$L_{I\ II\ III}\ M_{I\ II\ III\ IV}\ N_{I}\ O_{I}$	0.9E1 0.1M2	14	59.7	0.40	17
			59.57	$L_{I\ II\ III}\ M_{I\ II\ III\ IV\ V}\ N_{I\ II\ III\ IV\ V}\ O_{I\ II}\ P$	E1	16	59.6	0.359	22
			59.54	$L_{I\ II\ III}\ M_{I\ II\ III\ IV}\ N_{I\ II}\ O_{I\ II}$	E1	6			

TABLE 19: Gamma Transitions of Am^{241} (Low Energy Region) (cont'd)

98.9	99.80	$L_{I\ II\ III}\ M_{II\ III}$		14
	99.0	$L_{II\ III}$		
	98.85	$L_{II\ III}\ M_{II\ III}\ N_{II\ III}$	E1	6

TABLE 20: Summary of the intensities of the Np L x-rays and gamma rays

(As published by L. B. Magnusson)

Group	Line	Energy[a,i] kev	Relative Intensity[b] Bent Crystal spect. Day[c]	Bent Crystal spect. Day[d]	Proportional spect. Beling et al.[e]	Proportional spect. Present[f]	Absolute Intensity[g] photon/alpha	
L		11.89	0.83	1.32	...	2.2_4	0.008	0.008
	α_2	13.78	1.89	2.61				0.013
	α_1	13.96	17.2	24.4				0.119
	$\frac{1}{2}$	15.88	0.39	0.49				0.0024
L_α			19.5	27.5	42.0	37.5	0.135	
	$\frac{1}{2}$	15.88	0.39	0.49				0.0018
	β_6	16.14	0.38	0.50				0.0018
	β_2	16.86	4.60	6.03				0.022
	β_4	17.08	3.04	3.90				0.015
	β_5	17.52	0.74	0.99				0.0037
	β_1	17.76	26.1	34.2				0.127
	β_3	18.00	2.41	3.17				0.012
	$\frac{1}{2}\gamma_5$	20.12	0.11	0.15				0.0006
L_β			37.8	49.5	66.0	51.2	0.184	
	$\frac{1}{2}\gamma_5$	20.12	0.10	0.13				0.0005
	γ_1	20.80	6.42	7.92				0.031
	γ_2	21.11	0.71	0.86				0.0033
	γ_3	21.34	0.81	0.96				0.0037
	γ_6	21.48	1.73	2.01				0.0078
	γ_4	22.20	0.78	0.92				0.0036

TABLE 20: Summary of the intensities of the Np L x-rays and gamma rays (cont'd)

Group	Line	Energy[a,i] kev	Relative Intensity[b] Bent Crystal spect. Day[c]	Bent Crystal spect. Day[d]	Proportional spect. Beling et al.[e]	Proportional spect. Present[f]	Absolute Intensity[g] photon/alpha
L_γ			10.6	12.8	17.7	13.8	0.050
		26.36	5.93	7.5	7.5	7.0	0.025
		33.20	0.40	0.40	...	...	0.0011
		43.46	0.22	0.26	...	0.20[h]	0.0007
		59.56	100	100	100	100[h]	0.359
		99.0	0.10[h]		...	0.064[h]	0.00023
		103.0				0.053[h]	0.00019

a. P. F. Day, Phys. Rev. 97, 689 (1955).

b. Photons per 100 59.6-kev photons.

c. Relative intensities of reference a recalculated by Day with the topaz reflectivity dependence equal to $1/E^{1.35}$.

d. Relative intensites of column 4 recalculated with mass absorption coefficients for the sample self-absorption extrapolated from the data in Phys. Rev. 24, 1 (1924), 27, 266 (1926), 28, 907 (1926), and 43, 527 (1933).

e. Beling, Newton and Rose, Phys. Rev. 86, 797 (1952).

f. The preliminary intensites for the L x-ray groups were normalized to the absolute intensities of 0.376 L x-ray/ α and 0.359 59.6-kev gamma-ray/α determined by NaI crystal spectrometry.

g. The relative group intensities of column 7 were distributed by the relative line intensities within each group as given by column 5 and normalized to give the absolute total of 0.376 L x-ray/α.

h. These values were measured by scintillation spectrometry.

i. Better values of the energies of the neptunium L x-rays were later measured by J. J. Merrill by fluorescent excitation of x-rays in a sample of neptunium and the measurement of these x-rays in a carefully calibrated 2-crystal spectrometer. See Ph.D. thesis, Calif. Inst. of Technology (1960).

TABLE 21. Gamma Transitions of Am^{241} (High Energy Region)

Energy	Relative Abundance	Reference
99	2.3×10^{-4}	22
103	1.9×10^{-4}	"
113^{+}	3.3×10^{-5}	12
130	5.3×10^{-5}	"
159	4.4×10^{-6}	"
210	8.4×10^{-6}	"
270	1.1×10^{-6}	"
328	3.0×10^{-6}	"
370	1.7×10^{-6}	"

22. L. Magnusson, Phys. Rev. 107, 161 (1957).

12. P. Day, Phys. Rev. 97, 689 (1955).

$$E_i = \frac{h^2}{2\mathcal{I}} \quad I(I+1) - I_o(I_o+1)$$

where $\mathcal{I}$ = moment of inertia, I = spin, I_o = spin of base level. The ground state rotational band comprising the three levels mentioned has a spacing constant $h^2/2\mathcal{I}$ of 4.75 kev. The effective moment of inertia $\mathcal{I}$ is by far the largest one known for a rotational band. The value of $h^2/2\mathcal{I}$ is more than 20 percent less than the corresponding value for the band based on the 59.6 kev level in the same nucleus. The 76.4-kev 9/2+ level is not observed in the alpha decay of Am^{241} or the beta decay of U^{237}, but it was revealed by the coulombic excitation results of NEWTON[19]. His excitation of the 33.20 and 76.4 kev levels by bombardment of Np^{237} with low energy helium ions makes certain the collective nature of these levels. The base level, i.e. the ground state of Np^{237} is given the Nilsson orbital assignment 5/2 + 642 where the numbers in brakkets refer to the asymptotic quantum numbers $Nn_z\Lambda$.

The level at 59.57 kev which is so prominently observed in the decay of Am^{241}, being involved in more than 99 percent of the transitions, is the base state of a second rotational band of levels. The associated levels, which are labeled with the letter B in Fig. 26 are 59.57 kev (5/2-), 103 kev (7/2-), and 158.5 kev (9/2-). This level spacing corresponds to a value of 6.21 kev for the spacing constant. The next higher levels in this rotational band would be expected to be 225 kev (11/2-) and 305 kev (13/2-). Alpha particle groups corresponding to such levels were found by GOLDIN, NOVIKOVA AND TRETYAKOV[10] as listed in Table 19 and interpreted as the higher members of this rotational band. The Nilsson assignment for this band is 5/2- 523 .

It is noteworthy that Am^{241}, with a ground state spin 5/2, is so highly hindered in alpha decay to the ground state rotational band of Np^{237} whose ground state spin is 5/2, while, on the other hand, alpha decay to the family of levels based on the 5/2 state at 59.57 kev accounts for the great majority of the transitions. RASMUSSEN[20] interpreted this as meaning that the wave function of the odd-proton (Nilsson eigenstate) in Am^{241} is the same as that for the 59.57 kev state in Np^{237} but distinctly different from the ground state of Np^{237}. Specifically, the 59.57 kev state in Np^{237} and the ground state of Am^{241} are both given the Nilsson assignment 5/2- 523 .

BOHR, FROMAN AND MOTTELSON[23] have found this to be a general phenomenon in the alpha decay of odd mass nuclei and have developed a theory showing why certain alpha transitions are favored while others are hindered. These matters are discussed further in Chapter 3.

The 59.37 kev level in Np^{237} took on special significance with the discovery by BELING, NEWTON AND ROSE[17] that it had a half-life of 6.3×10^{-8} seconds, a value which is $\sim 4 \times 10^{5}$ longer than that expected through the use of the Weisskopf single-proton formula for estimating the lifetimes of E1 transitions. It has been suggested that this retardation is caused by the violation of a selection rule in the asymptotic quantum number n_z. The general occurrence of highly retarded E1 transitions for heavy element nuclei is discussed in Section 3.5.7 of Chapter 3.

In addition to its unexpectedly long-half-life the 59.37 kev (and the 26.4 kev) transition in Np^{237} is anomalous in another respect. The L-shell conversion coefficients do not agree with the theoretical conversion coefficients as given by ROSE[30] or by SLIV AND BAND[31]. The disagreement is in a direction which cannot be removed by postulating some suitable admixture of other multipole types. ASARO, STEPHENS, HOLLANDER AND PERLMAN[8] have critically examined the data on 13 such anomalous E1 transitions in the region of heavy element deformed nuclei. Their summary of the transitions appearing in the decay of Am^{241} is presented in Table 22.

KROHN, NOVEY, AND RABOY[28] measured the gyromagnetic ratio of the 59.57 kev level as $\pm 0.8 \pm 0.2$ by the attenuation of the alpha-gamma angular correlation in an applied magnetic field. The magnetic moment of the state is $+2.0 \pm 0.5$ nuclear magnetons.

The ground state spin of Am^{241} has been proved to be 5/2 from a study of atomic spectra.[24,25,33] This assignment is fully confirmed by atomic beam experiments.[34] The decay scheme is also strengthened by the determination of the ground state spin of Np^{237} as 5/2 by atomic spectrum analysis[26] and paramagnetic resonance studies.[27] MANNING, FRED AND TOMKINS[33] report a quadrupole moment of +4.9 barns and a dipole moment of +1.4 nuclear magnetons.

The half life of Am^{241} for spontaneous fission has been found by MIKHEEV, SKOBELEV, DRYIN AND FLEROV[29] to be 2×10^{14} years.

TABLE 22: Summary of L-Shell E1 Conversion Data for the "Anomalous" Transitions in Np^{237}.

Transition Energy (kev)		$\alpha(L_I)$	$\alpha(L_{(II)})$	$\alpha(L_{III})$	$\alpha(L_{Total})$
59.57	Experimental	0.22±0.02	0.46±0.05	0.12±0.03	0.80±0.08
	Theory -Rose	0.11	0.10	0.125	0.34
	Theory -Sliv and Band	0.13	0.12	0.13	0.38
26.4	Experimental	2.0	3.9	1.2	7.1
	Theory - Rose	0.22	0.55	1.25	2.0
	Theory - Sliv and Band	0.55	1.1	1.4	3.1

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9.3.7 The Isomers of Am^{242}. When Am^{241} is irradiated with slow neutrons two isomeric forms of Am^{242} are produced independently. The first to be discovered was a 16-hour β^- -emitter (MANNING AND ASPREY[1]) whose half-life was later determined more precisely as 16.01±0.02 hours.[3] Experiments by SEABORG, JAMES AND MORGAN[2] showed that a long-lived form of Am^{242} was also produced. This isomer was identified by the isolation of its radioactive decay products (Cm^{242} and Np^{238}) and by mass spectrographic analysis.[2,4] The half-life was given as ~100-years[4], later determined more precisely as 152±7 years.[12]

The β^- and conversion electron spectra associated with both isomers have been investigated [5,7,8,11] and the puzzling conclusion was reached that both had almost identical decay schemes involving transitions to the 0+ and 2+ states of Cm^{242}. The ratio of β^- emission/electron capture also appeared to be similar. This, of course, is out of keeping with their existence as isomers. On the basis of a small difference in the β^- spectra and points the 16-hour isomer was taken to be the metastable state.

This unsatisfactory situation was cleared up by ASARO, PERLMAN, RASMUSSEN AND THOMPSON[14] who showed that the 152-year isomer is the metastable state and that its principal mode of decay is by a 48.6 kev isomeric transition to the ground state which is the 16-hour β-emitter. This explains the similarity of β-decay properties because in both cases only the decay associated with the ground state was under observation. The proof of the decay sequence consisted of an isomer separation in which it was shown that 16-hour isomer was present in equilibrium with the 152-year isomer. The minor differences observed[5] in the decay properties (β^- populations to different energy levels; β^-/EC ratios) have disappeared in the light of later measurements[12,15] and only the apparent difference in β^- end points (which have not been remeasured) remains.

The decay scheme embodying all available information is shown in Fig. 28. The percentages of branching decay by orbital electron capture and by β^--emission are taken from the work of BARNES, HENDERSON, HARKNESS AND DIAMOND.[12] A slight alpha decay of the long lived isomer was noted by SEABORG, JAMES AND MORGAN[2] as demonstrated by the chemical separation of the daughter Np^{238}. An accurate value of 0.476±0.014% corresponding to a partial alpha half life of 32,000±1600 years was measured by BARNES, HENDERSON, HARKNESS AND DIAMOND.[12] The alpha particles have not been observed directly because of the interference of overwhelmingly greater Am^{241} activity, but the alpha decay energy is estimated as 5.52 Mev.

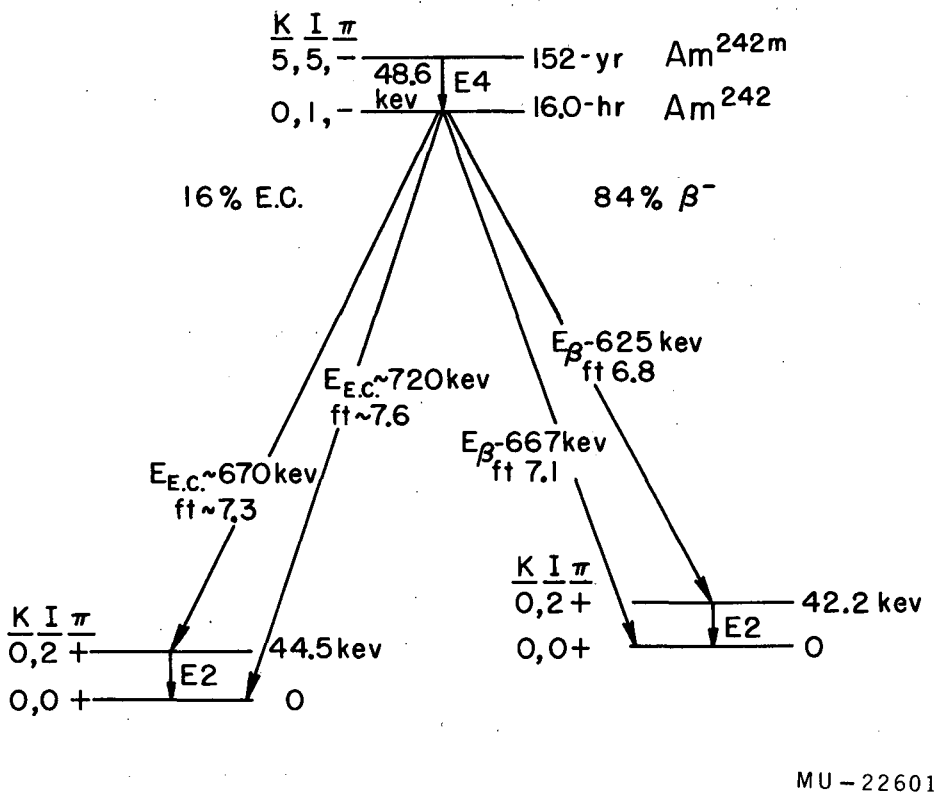

Figure 28. Decay scheme of isomers of Am^{242}. The alpha decay of Am^{242m}, known to occur to the extent of 0.48%, is not shown.

It may be worthwhile to point out the difficulty responsible for the long delay in the discovery of the correct relationship of the two isomers. It happens that the long-lived Am^{242} isomer has a large neutron capture cross section so that a maximum concentration of only a few percent by activity can be built up in a sample of Am^{241}. Since Am^{241} has abundant low-energy photon transitions associated with its alpha decay, the electrons associated with the isomeric transition are obscured unless special pains are taken to look for them. In addition, Am^{243} is always present and its decay product Np^{239} also obscures the picture.

Table 23 summarizes the precision measurements of the gamma transitions seen in the decay of the 16 hour isomer. The authors cited therein used electron spectrometers to measure the L,M,N and O electrons of the 2 chief gamma transitions. The subshell conversion ratios clearly identify both transitions as E2. ASARO, PERLMAN, RASMUSSEN AND THOMPSON measured the electrons associated with an aged americium sample which contained an appreciable content of 152 year Am^{242} and found L,M,N and O conversion lines of a 48.63 kev transition converted in americium subshells. This is the isomeric transition by which the 152-year isomer is chiefly de-excited. The multipolarity of the transition was determined unambiguously as E4 by comparison of the experimental ratios of conversion lines, particularly those from the M-shell, with ratios calculated from theoretical conversion coefficients.

The spin of the 16 hour state was measured by the atomic beam resonance technique and found to be 1 by WINOCUR, MARRUS AND NIERENBERG.[16] In combination with the E4 multipolarity assignment of the 48.63 kev transition this establishes a spin value of 5 for the 152 year Am^{242m}. The atomic beam experiment emphasized strongly the shortcomings of the earlier decay schemes for the Am^{242} isomers and provided a strong stimulus for the work of ASARO AND CO-AUTHORS.[14]

The beta decay properties of Am^{242} and Am^{242m} are summarized in Table 24.

The radiations of Am^{242} include a complex spectrum of X-rays resulting from the electron capture branching, and the gamma conversion process. Some precise measurements of the L x-ray spectrum on a best crystal type x-ray spectrometer are given in Table 25 . Since these measurements were made with samples of the 16 hour isomer, the L x-rays of americium can not be attributed to the isomeric transition but only to the fluorescent excitation

TABLE 23: Gamma Transitions in Am^{242} from Conversion Electron Measurements

Transition	Energy	Ref.
16 hr $Am^{242} \longrightarrow Cm^{242}$ conversion in Cm subshells	42.3	5
	42.2	8
	42.12	11
	42.18	7
	42.20	14
16 hr $Am^{242} \longrightarrow Pu^{242}$ conversion in Pu subshells	44.8	5
	44.6	8
	44.50	11
	44.52	7
	44.50	14

TABLE 24: Beta Decay Properties of Am^{242} and Am^{242m}

Isomer	Decay mode	Product -state	Decay Energy	Relative Intensity %	Log ft
16-hour Am^{242}	β^- 83.6%	Cm^{242}, 0+	0.667	34	7.1
		Cm^{242}, 2+	0.625	50	6.8
	EC 16%	Pu^{242}, 0+	0.72	~6 (K-cap.,4.4)	~7.6
		Pu^{242}, 2+	0.67	~10 (K-cap.,~7.6)	~7.3
152-year Am^{242m}	β^-	Cm^{242}, 4+	0.578	<2% of IT	>13

As summarized in reference 14.

TABLE 25: X-rays following the decay of Am^{242} (From Hoff, reference 5)

Line	Transition	Observed energy (kev)	Siegbahn's extrapolated energy (kev)	Corrected relative intensity
Cm $L\alpha_2$	$L_3 - M_4$	14.75 ± 0.03	14.74	4
Cm $L\alpha_1$	$L_3 - M_5$	14.97 ± 0.03	14.96	27
Cm $L\beta_2$	$L_3 - N_5$	18.09 ± 0.02	18.10	16
Cm $L\beta_1$	$L_2 - M_4$	19.47 ± 0.02	19.38	100
Cm $L\gamma_1$	$L_1 - N_3$	22.79 ± 0.04	22.63	40
Cm $L\gamma_3$	$L_1 - N_3$	23.30 ± 0.06	23.25	6
Cm $L\gamma_6$	$L_2 - O_4$	23.62 ± 0.12	23.46	12
Am $L\alpha_2$	$L_3 - M_4$	14.44 ± 0.06	14.41	1
Am $L\alpha_1$	$L_3 - M_5$	14.61 ± 0.03	14.61	4
Am $L\beta_1$	$L_2 - M_4$	18.89 ± 0.02	18.80	6
Pu $L\alpha_2$	$L_3 - M_4$	14.08 ± 0.03	14.08	2
Pu $L\alpha_1$	$L_3 - M_5$	14.28 ± 0.03	14.28	10
Pu $L\beta_2$	$L_3 - N_5$	17.29 ± 0.03	17.25	9
Pu $L\beta_1$	$L_2 - M_4$	18.33 ± 0.03	18.27	27
Pu $L\beta_3$	$L_1 - M_3$	18.62 ± 0.04	18.52	7
Pu $L\gamma_1$	$L_2 - N_4$	21.46 ± 0.04	21.38	16
Pu $L\gamma_3$	$L_1 - N_3$	22.06 ± 0.10	21.97	7
Pu $L\gamma_6$	$L_2 - O_4$	22.24 ± 0.10	22.13	7
Pu K x-rays		102		37

of americium x-rays in the bulk sample by the L x-rays of curium; all the samples subjected to study contained large amounts of Am^{241} because of the method of preparation.

Several features of the decay scheme were interpreted in terms of the unified model of nuclear structure by ASARO, PERLMAN, RASMUSSEN AND THOMPSON.[14] Americium-242 lies within a group of nuclei which have large prolate nuclear deformation. It has one odd neutron and one odd proton and it is believed that the wave functions of these odd nucleons should be identifiable with Nilsson wave functions for nucleons in a deformed nucleus. The most likely Nilsson state for the odd neutron is the 5/2 + 622 state, which appears as the ground state of the isotonic nuclei Pu^{241} and Cm^{243}. The quantum numbers are the usual ones $K\ \pi\ N\ n_z$ appropriate to particles in spheroidal nuclei; see Chapter 3. The most likely proton orbital is 5/2- 523 , the ground state of Am^{241} and Am^{243}. The orbital 5/2 + 642 , however, is rather near-lying and is also a possibility.

The K-quantum number is the projection of the total angular momentum on the nuclear symmetry axis and with two unpaired particles of = 5/2+ and = 5/2- the values K = 0- and K = 5- are possible. According to the Nordheim coupling rules as modified by GALLAGHER AND MOSKOWSKI[17], in this particular case, the K = 0- state should lie below the 5-states.

Thus the spin 5 of the long-lived form of Am^{242} receives a natural explanation. Also the high log ft value, > 13, for the beta transition from this isomer to the 4+ level of Cm^{242} receives a ready explanation in 5-fold change in K which this transition would entail. On the other hand, it seems hard to reconcile the K = 0- prediction for the ground state with the observed spin of 1. The reconciliation is made by asserting that the spin 1 member of the rotational band of levels based on a K = 0 ground state appears as the lowest member of the K = 0 band. This surprising feature is strongly supported by the close correspondence of several of its theoretical implications with experimental data.

For example, a traditional test for the K-quantum number involves comparisons of branching ratios of beta or gamma radiation to different members of the same rotational band, the reduced transition probabilities being proportioned to the square of a Clebsch-Gordan coefficient. This test is fully described in section 3.4.7 of Chapter 3. When this criterion is applied to the relative population of the 2+ and 0+ state of Cm^{242} in the beta decay of 16

hour Am^{242} the K = 0 assignment for Am^{242} favors the population of the 2+ first excited state by a factor of two, whereas the K = 1 assignment favors the transition to the ground state by a factor of 2. Experiment (see Table 24) clearly favors the K = 0 choice.

Even more striking evidence comes from consideration of quantities derived from atomic beam measurements.[16] These have fixed precisely the absolute value of the ratio of the magnetic moments of Am^{242} and Am^{241}, 0.236, and also the absolute value of the ratio of their spectroscopic quadrupole moments, 0.562. Furthermore, the measurements have established that either the magnetic moment or the quadrupole moment of Am^{242} is of opposite sign to the corresponding moment of Am^{241}. The optical spectrographic data of MANNING, FRED AND TOMKINS[20] for Am^{241} gives μ = +1.4 nuclear magnetons (nm) and Q_{spec} = +4.9 barns. Combining this information with that from the atomic beam measurements, we obtain for Am^{242}, $\mu = \pm$ 0.33 nm and Q_{spec} = +2.75 barns.

These nuclear moments may now be compared with the theoretical expectations for the I = 1, K = 0 assignment. An abundance of evidence has established the intrinsic quadrupole deformations of nuclei in the region of americium as prolate (positive). However, for nuclear states with $K^2 < I(I + 1)/3$ the signs of the intrinsic and spectroscopic moments will be opposite. The relation between these moments is as follows:

$$Q_{spec} = \frac{3K^2 - I(I + 1)}{(I + 1)(2I + 3)} Q_o$$

$$\text{for } I = 1,\ K = 0,\ Q_{spec} = \frac{Q_o}{5}$$

If we assume that Q_o for Am^{242} is the same as that for Am^{241}, the K = 0 assignment gives Q_{spec} = -2.74 barns, which is in excellent agreement with the value +2.75 barns obtained as mentioned above. If, on the other hand, we assume K = 1 for Am^{242}, then Q_{spec} should be +1.4 barns.

The magnetic moment may also be analyzed. For the K = 0, I = 1 assignment the angular momentum is directed perpendicular to the symmetry axis, and there is no specific contribution to the magnetic moment from the odd nucleons. In this case we would expect a magnetic moment $\mu = g_R I$, where g_R is the gyromagnetic ratio for collective motion, usually estimated as +Z/A, the fraction of protons in the nucleus. From this we get μ = +0.39 nm, which agrees in sign and magnitude with the measured value (μ = +0.33 nm) if the sign for Q_{spec} is taken to be negative in accord with our theoretical prediction.

If we consider the assignment I = 1, K = 1, we must first of all postulate some possible orbital assignments for the neutron and proton. The most likely are 5/2- 523 (or 5/2+ 642) for the proton as before and 7/2+ 624 for the neutron. This neutron orbital appears as a state at 172 kev in Pu^{241}. If the magnetic moments are calculated from Nilsson wave functions,[18,19] (with a deformation parameter, , of 4.8), the values for 5/2- 523 and 5/2+ 642 proton orbtials are 0.2 nm and -1.2 nm respectively. Thus the measured magnetic moment is not consistent with a K = 1 assignment with proton orbital 5/2+ 642 but cannot be used by itself to rule out K = 1 with proton orbital 5/2- 523 . With the latter assignment, however, the ratio of μ/Q_{spec} would be positive, in disagreement with the experimental results. It is seen, therefore, that not only is the magnitude of the measured quadrupole moment in better agreement with theory for K = 0 than for K = 1, but the sign of the ratio μ/Q_{spec} (as determined from theory) can only be negative for K = 0. Thus, the atomic beam measurements reinforce the K = 0 assignment.

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9.3.8 Americium-243. The neptunium fraction from a sample of Am^{241} that had been irradiated with slow neutrons in a reactor was shown by STREET, GHIORSO and SEABORG[1] to contain equilibrium amounts of both Np^{238} and Np^{239}. The presence of Np^{239} indicated the existence of the nuclide Am^{243} prepared by the reaction sequence:

$$Am^{241}\ (n,\gamma)\ Am^{242}\ (n,\gamma)\ Am^{243}$$

$$Am^{243} \xrightarrow{\alpha} Np^{239}$$

Mass spectrographic analysis of the irradiated americium sample confirmed the presence of Am^{243} in an amount of about 0.5 percent. This, together with the chemical yield of Np^{239}, gave a partial half-life for alpha emission of about 10^4 years. Better values are cited below. It has also been possible[2] to produce Am^{243} by the reaction:

$$Pu^{242}\ (n,\gamma)\ Pu^{243}$$

$$Pu^{243} \xrightarrow[5.0\ hours]{\beta^-} Am^{243}$$

In high flux reactors Am^{243} can be produced in large quantity in the following way. Plutonium of mass number 239 or 240 is converted to Pu^{242} of high isotopic purity (> 98 percent) by prolonged neutron bombardment. The Pu^{242} can be repurified, principally to remove Am^{241}, and reinserted in a reactor to produce Am^{243} by the reaction outlined above.

This synthesis is important because Am^{243} is the longest-lived of the isotopes of americium; it is about 19 times longer in half-life than Am^{241}. The study of the chemistry of americium using Am^{241} is severely hindered by the remarkable chemical effects caused by the intense alpha radiation of Am^{241}, since its specific activity is 7×10^{13} disintegrations per minute per gram. The 19 times smaller specific activity of Am^{243} extends the range of possible studies of the properties of americium.

The half-life of Am^{243} has been reported as 7600 years,[3,7] 8800 ± 600 years,[4] 7600 ± 370 years,[5] 7720 ± 160 years,[11] 7650±50 years,[19] and 7951 ± 48 years.[6] The last cited measurement was made by specific activity measurements on two weighed samples of americium metal in which the alpha activity was 99.35% Am^{243}.

A careful study of the alpha spectrum of Am^{243} in a magnetic spectrograph was made by STEPHENS, HUMMEL, ASARO and PERLMAN[5] who report the following groups; 5.339 Mev (0.17 percent), 5.308 Mev (0.16 percent), 5.266 Mev (86.9 percent), 5.224 Mev (11.5 percent) and 5.169 Mev (1.3 percent). The decay scheme based on these data is shown in the figure. The nuclear spin 5/2 has been assigned to Am^{243} on the basis of a study of the hyperfine structure of atomic spectral lines.[8] The quadrupole moment and the magnetic moment have been measured as +4.9 barns and +1.4 nuclear magnetons.[12]

The pattern of alpha group abundances and level spacings is remarkably similar to that of Am^{241} as can be seen in Figure 24 appearing in Section 9.3.4. This suggests that the interpretation of the decay scheme of Am^{243} should follow closely that of Am^{241}, about which much more evidence has been gathered. In particular the levels at 74, 117 and 173 kev in the daughter nucleus Np^{239} may constitute a rotational band of levels with spins 5/2, 7/2 and 9/2 respectively based on the 5/2 level. The Nilsson assignment of this rotational band is 5/2- [523] where the numbers have the meanings K,parity $[N,n_z,\Lambda]$.

A prominent E1 gamma ray with energy 75 kev has been studied[3] by alpha-gamma coincidence techniques and shown to follow about 80 percent of the alpha transitions. HOLLANDER[13] measured the L shell conversion electrons which characterize the transition as E1 and determined the energy to be 74.6 kev. This gamma ray is shown in the figure. The half-life of this gamma ray is less than 2×10^{-9} seconds.[9] ASARO, STEPHENS, HOLLANDER and PERLMAN[14] have made a detailed comparison of the characteristics of this 74.6 kev transition with many other E1 transitions in heavy element nuclei. Many of these are anomalous in their conversion and half life characteristics. Other low intensity gamma rays must certainly be present.

The ground state spin of Np^{239} is known to be 5/2 from the atomic beam experiments of HUBBS and MARRUS.[10] The ground state of Np^{239} is given the ~~a~~ther firm Nilsson orbital assignment of 5/2+ [642]. This assignment is based on a study of the decay scheme of Np^{239}, and on a comparison of Np^{239} with Np^{237} as well as on a study of Am^{243}. The ground state of Am^{243} is given the Nilsson assignment 5/2- [523] on the basis of a study of Am^{243} itself and

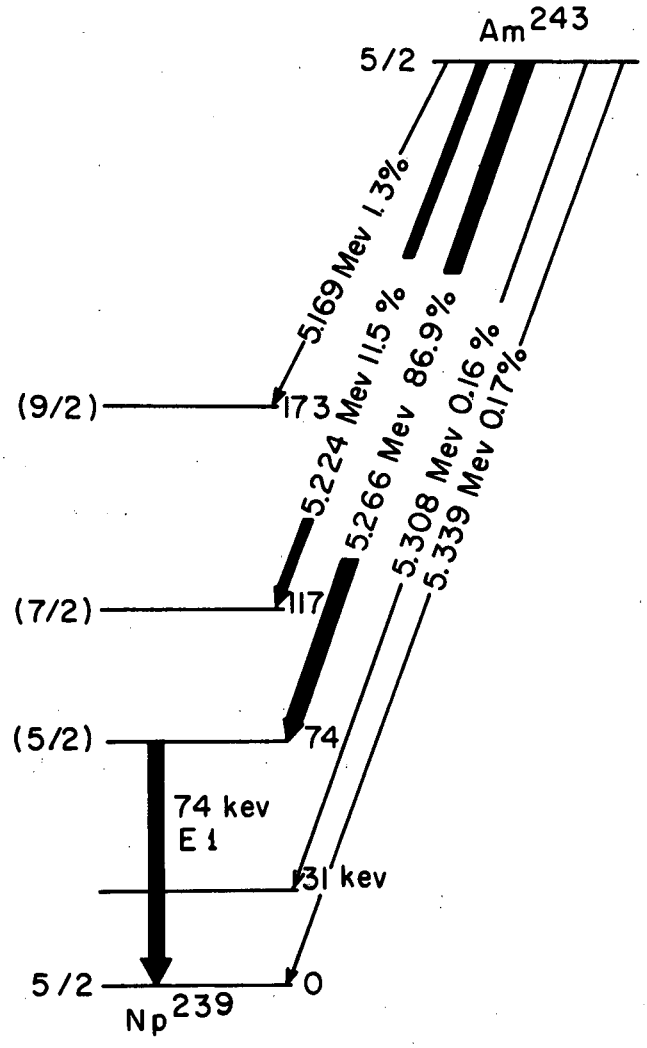

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Figure 29. Decay scheme of Am^{243}.

of the decay of Pu^{243} and Bk^{247}. For a review of Nilsson orbital assignments see STEPHENS, ASARO and PERLMAN.[15]

We note that the favored alpha decay proceeds to a rotational band of levels in Np^{239} which has a Nilsson orbital assignment identical with that of the ground state of the parent in accordance with the ideas of BOHR, FROMAN and MOTTELSON.[16]

For some time it was not certain whether Am^{243} was stable with respect to beta decay to Cm^{243}. Using the decay energy cycle involving the alpha-disintegration energies of Am^{243} and Cm^{243} and the beta-disintegration energy of Np^{239}, FOREMAN and SEABORG[17] found a value of the mass difference of Am^{243} and Cm^{243} which was smaller than the errors involved, and hence it was uncertain which member of this isobaric pair was the heavier. But CHOPPIN and THOMPSON[18] found evidence for an electron capture decay of Cm^{243} to Am^{243} with a partial half-life of $(1.0 \pm 0.1) \times 10^4$ years.

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9.3.9 The Isomers of Americium-244. STREET, GHIORSO, and SEABORG[1] found that neutron irradiation of a sample of americium containing approximately 10 per cent Am^{243} produced a new americium activity. This activity decays by beta emission with a half-life of about 25 minutes. The isotope was assigned to Am^{244} produced by the reaction:

$$Am^{243}\ (n,\gamma)\ Am^{244}.$$

Later work of GHIORSO, THOMPSON, CHOPPIN, and HARVEY[2] confirmed this result. These workers studied the beta rays with an anthracene crystal spectrometer and found an endpoint energy of 1.5 Mev. They found no prominent gamma rays in a study carried out with the aid of a sodium iodide crystal spectrometer.

FIELDS and CO-WORKERS[3] suspected that Am^{244} was unstable toward electron capture from a consideration of closed, decay-energy cycles. By isolation of the Pu^{244} daughter from samples of Am^{243} which had received extensive irradiation with neutrons and comparing the amount of Pu^{244} to the amount of Cm^{244} found in the sample they measured a value of 0.039 ± 0.003 per cent for the electron capture branching of Am^{244}.

At a later date DAY and VANDENBOSCH[4] showed that the neutron irradiation of Am^{243} results in the formation of an 11 hour isomer of Am^{244} as well as the 26 minute species. Their study of the beta and gamma radiations of the isomers led to the decay schemes shown in figure 30. The 11-hour isomer lies higher in energy. In view of this result the percentage of electron capture decay measured by FIELDS et al[3] is an average for the mixture of isomers formed by neutron irradiation of their sample.

Since large amounts of Am^{243} can be produced by prolonged neutron irradiation of plutonium (as was discussed in the previous section), it is possible to prepare high intensity samples of Am^{244} with considerable ease.

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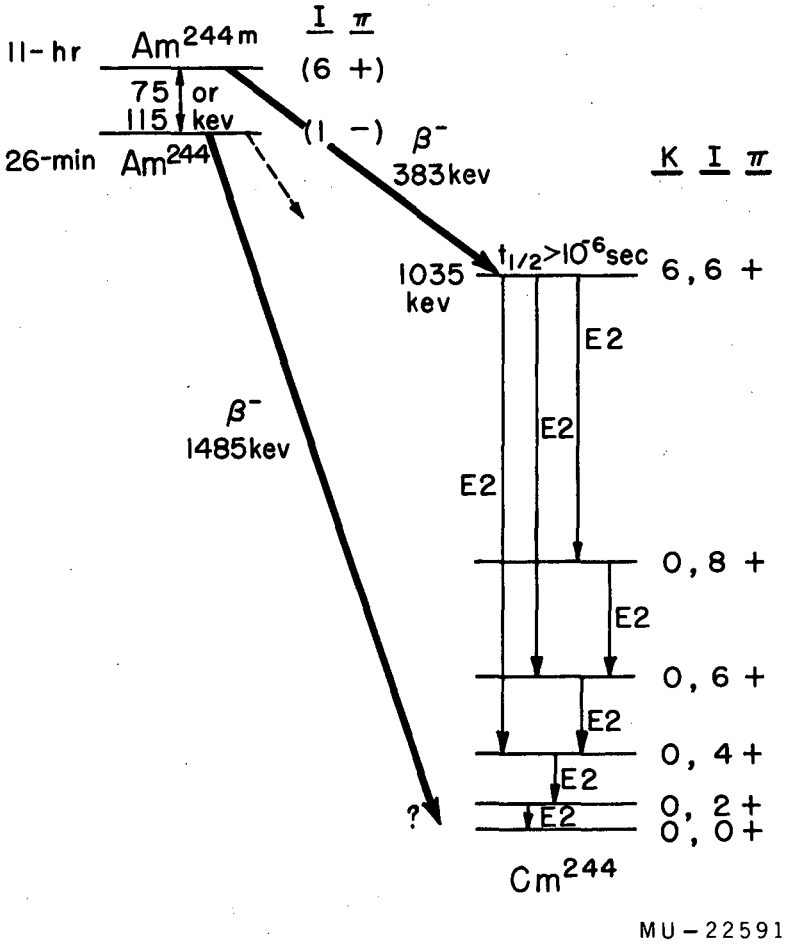

Figure 30. Decay scheme of the isomers of Am^{244}.

9.3.10 Americium-245. The isotope Am^{245} is a beta emitter with a half-life of 2.08 hours. It is produced by the beta decay of Pu^{245} which has a half-life of 10.5 hours. The Pu^{245} parent can be produced by the neutron irradiation of plutonium samples containing appreciable concentrations of 4×10^5 year Pu^{244}; or by the irradiation of a heavy element with an extremely high instantaneous flux of neutrons such as is available in the explosion of thermonuclear devices. The first reports on Am^{245} were provided by BROWNE and CO-WORKERS[1] and by FIELDS and CO-WORKERS.[2] Confirmatory evidence was provided by BUTLER and CO-WORKERS.[3] The mass assignment was made on the basis of the method of preparation, the systematics of isotope properties in this mass region and the isolation of Cm^{245} daughter activity.

The beta spectrum of Am^{245} has been studied with a thin lens magnetic spectrometer. It has single beta component of endpoint energy 905 ± 5 kev. The log ft value is 6.2 indicating an allowed or first forbidden transition. The K and L conversion electrons of a 255 kev gamma ray are believed to correspond to an E1 transition. Scintillation counter studies show prominent gamma rays of 108 kev (K x-rays) and 248 kev. By gamma-gamma coincidence methods several other gamma rays are found;[1] their energies are 36, 120, 143, 156 and 232 kev. A tentative decay scheme[1] leads to a total disintegration energy of 1.32 Mev. This energy leads to a gross discrepancy with the decay-energy cycle involving the nuclides Bk^{249}, Cf^{249}, Cm^{245} and Am^{245} since only 860 kev should be available for the decay of Am^{245}. The decay of Bk^{243} by electron capture also produces Cm^{245} as a daughter product but there is not much correspondence between the radiations seen in the two cases except for the common occurrence of a 255 kev gamma ray. Compare section 9.5.4 where Bk^{243} is described.

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9.3.11 Americium-246. The isotope Am^{246} was first found in a study of the plutonium fraction isolated from the debris of the explosion of a thermonuclear device.[1] This was discussed under Plutonium-246 in Section 9.2.16 of this chapter. The plutonium fraction was found to contain 10.8-day Pu^{246} decaying by beta emission to Am^{246}. The Am^{246} has a half-life of 25 ± 0.2 minutes for the emission of beta particles. The genetic relationship of Am^{246} and Pu^{246} is firmly established.

The beta and gamma ray spectrum of Am^{246} has been studied in equilibrium mixtures of Pu^{246} and Am^{246} and in samples of radiochemically pure Am^{246}. The following resolution of the beta spectrum has been reported:[2] 1.35 Mev, 79 percent, log ft = 6.1; 1.60 Mev, 14 percent, log ft = 7.3; 2.10 Mev, 7 percent, log ft = 8.0. Gamma rays were identified which have energies of 18.5, 103, 245, 795 (complex) and 1069 kev (complex). The 18.5- and 103-kev radiations are probably L and K x-rays, respectively. The beta ray spectrum in coincidence with 1069-kev gamma rays has an endpoint energy of 1.222 Mev. Hence, the beta decay energy is at least 2.29 Mev. SMITH and CO-WORKERS[2] have made an extensive study of gamma-gamma coincidences. More extensive studies are required before a complete decay scheme can be formulated.

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9.4 THE ELEMENT CURIUM (Element 96)

9.4.1 The Discovery of Curium. The first isotope of curium was prepared by SEABORG, JAMES and GHIORSO.[1] in mid-1954. These men worked at the Metallurgical Laboratory of the University of Chicago on cyclotron targets of plutonium which had been bombarded with helium ions at the 60-inch cyclotron at the University of California. Their goal was to identify an isotope of element 96 prepared by reactions of the (α,xn) type. At first these experiments were unsuccessful because the chemical steps chosen for the isolation of the element 96 fraction were based on the premise that this element should resemble uranium, neptunium and plutonium in having a stable hexapositive oxidation state. The first successful experiments on element 96 occurred shortly after it was realized that this element might be a member of an actinide series of elements and as such might be oxidized beyond the tripositive state only with extreme difficulty if at all. A new activity emitting alpha particles with a range of 4.75 centimeters of air and a half-life of five months was then isolated by coprecipitation on lanthanum fluoride and other carrier precipitates suitable for a rare-earth-like tripositive ion. This activity was Cm^{242} produced by the reaction:

$$Pu^{239}\ (\alpha,n)\ Cm^{242}$$

Shortly thereafter, late in 1944, the identification of element 95 followed as a result of the bombardment of Pu^{239} with pile neutrons, the production reactions being as follows:

$$Pu^{239}\ (n,\gamma)\ Pu^{240}\ (n,\gamma)\ Pu^{241}$$

$$_{94}Pu^{241} \xrightarrow[\text{long}]{\beta^-} {}_{95}Am^{241}$$

In the same samples the isotope Cm^{242} was formed by an additional neutron-capture step.

$$Am^{241}\ (n,\gamma)\ Am^{242}$$

$$_{95}Am^{242} \xrightarrow{\beta^-} {}_{96}Cm^{242}$$

The irradiated plutonium was processed chemically to separate a transplutonium element fraction with rare earth carriers. Alpha particles of 4.75 cm range and five months half-life were assigned to 96^{242} and alpha particles of 4.0 cm range were assigned to 95^{241}. This assignment was partially based on the ratio of the yields of the two alpha particle groups in plutonium samples bombarded with differing total numbers of neutrons; The Cm^{242} is produced by a second-order reaction whereas the Am^{241} is produced by a first-order reaction. The activity assigned to Cm^{242} was also identified by separating Pu^{238} daughter activity. A satisfactory chemical separation of curium from its neighboring element americium did not come until about one year later when the method of selective elution of the elements with a buffered citric acid complexing agent from a cation exchange resin was adapted for this purpose based on the methods previously used in rare-earth chemistry.

The name of element 96 was chosen by SEABORG, JAMES and GHIORSO[1] to honor Marie and Pierre Curie and to emphasize the analogy of element 96 as the seventh member of the actinide series of elements to gadolinium, the seventh member of the lanthanide elements, whose name honored the Finnish chemist, J. Gadolin.

The first isolation of curium in a weighable quantity was carried through by WERNER and PERLMAN.[2] Microgram quantities of curium were made by the intense neutron irradiation of 4.5 milligrams of americium in a nuclear reactor. The chief separation was made by the ion exchange method. The final sample was 40 micrograms of Cm_2O_3 of about 90 percent purity.

Most chemical studies of curium up until the mid-nineteen fifties were carried out with samples of Cm^{242} prepared in this way. There are severe difficulties in working with this isotope with its short 162.5 day half-life because of the high specific activity. Each milligram of Cm^{242} emits about 10^{14} alpha particles of 6.110 Mev energy per minute. Water solutions of Cm^{242} are decomposed by this radiation; some of the decomposition products interfere with attempted chemical reactions or the complete study of the absorption spectrum. Precise temperature control of solutions or compounds as for example in magnetic susceptibility experiments is difficult because of heating effects. A massive chunk of such curium metal would be heated rapidly to incandescence,

if it could be prepared, since the energy released is 1230 watts per gram. Crystal lattices of compounds are disarranged by recoil effects of the alpha particles.

It is for these reasons that the isotope Cm^{242} is being replaced with longer-lived isotopes for laboratory studies of the chemical and physical properties as the longer-lived isotopes become available. The isotope Cm^{244} has a particular importance at present because it can be prepared with rather high isotopic purity in milligram or greater quantities. The half-life is 19.4 years and the specific alpha activity is 43 times less than that of Cm^{242}. It is also possible to prepare Cm^{245}, Cm^{246}, Cm^{247} and Cm^{248} in milligram-or-larger quantities by the neutron irradiation procedures described later in this chapter. These isotopes are even longer-lived than Cm^{244} and hence are even more suitable candidates for laboratory study of many chemical properties.

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9.4.2 Curium-238. The isotope Cm^{238} was first prepared by the reaction,[1,2]

$$Pu^{239}\ (\alpha,5n)\ Cm^{238}.$$

This isotope decays by the emission of 6.52-Mev alpha particles and also by orbital electron capture. The alpha branching has been estimated to be about 0.4 percent.[3] The observed half-life is 2.5 hours.

The Cm^{238} prepared by the above reaction is contaminated with 12-hour Cm^{239} and with higher mass curium isotopes. GLASS, CARR, COBBLE and SEABORG[2] have studied the yield of Cm^{238} as a function of helium ion energy for the reaction

$$Pu^{238}\ (\alpha,4n)\ Cm^{238}.$$

They report a cross section of only 0.26 millibarns at 47.4 Mev the highest energy for which they could obtain data.

REFERENCES - Cm^{238}

1. K. Street, Jr., A. Ghiorso, D. A. Orth and G. T. Seaborg, unpublished results (1948).
2. R. A. Glass, R. J. Carr, J. W. Cobble and G. T. Seaborg, Phys. Rev. 104, 434 (1956).
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9.4.3 Curium-239. By bombardment of Pu^{239} with high energy (70-80 Mev) helium ions the 3-hour isotope Cm^{239} is produced.[1]

$$Pu^{239} (\alpha,4n) Cm^{239}$$

A number of other curium isotopes are produced at the same time and interfere with the study of the radiations of Cm^{239}. This isotope decays by orbital electron capture to 12-hour Am^{239}. The genetic relationship of these two isotopes has been established by radiochemical experiments. An upper limit of 0.1 percent has been set on alpha branching.

REFERENCES - Cm^{239}

1. G. H. Higgins, Ph.D. Thesis, University of California, June 1952; also published as University of California Radiation Laboratory Report, UCRL-1796 (1952).

9.4.4 Curium-240. The second isotope of curium to be discovered[1] was the 26.8-day Cm^{240} produced when Pu^{239} was bombarded with 40 Mev helium ions:

$$Pu^{239}\ (\alpha,3n)\ Cm^{240}.$$

GLASS, CARR, COBBLE and SEABORG[2] report a cross section for this reaction at several values of the helium ion bombardment energy: .09 millibarns at 27.5 Mev, 0.22 millibarns at 33.3 Mev and 1.6 millibarns at 37.2 Mev. The Cm^{240} is not produced in a pure state by the above reaction since other curium isotopes are formed at the same time.

Curium-240 decays by the emission of alpha particles of 6.26 Mev energy.[6] The mass assignment has been confirmed by the isolation of Pu^{236} daughter activity.[1] Curium-240 is nearly beta stable but the best estimate from closed decay-energy cycles is that it is unstable toward orbital electron capture decay to Am^{240} by about 90 kev. HIGGINS and STREET[3] have set an experimental upper limit of 0.5 percent to this mode of decay. Curium-240 is the daughter of the alpha-emitter, 45-minute Cf^{244}.[4]

The spontaneous fission half-life of Cm^{240} has been measured by GHIORSO, HIGGINS, LARSH and SEABORG[5] to be $(1.9 \pm 0.4) \times 10^6$ years.

REFERENCES - Cm^{240}

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9.4.5 Curium-241. The isotope, Cm^{241}, was first produced by SEABORG, JAMES AND GHIORSO[1] by bombardment of Pu^{239} with 40 Mev helium ions:

$$Pu^{239}\ (\alpha, 2n)\ Cm^{241}$$

These investigators reported a half-life of about 55 days for decay by orbital electron capture. HIGGINS AND STREET[2] redetermined a half-life of 35 ± 2 days and detected a small branching decay by the emission of alpha particles. GLASS, CARR, COBBLE AND SEABORG[3] obtained a value of 0.96 percent for the alpha branching which corresponds to a partial alpha half-life of 10 years. They reported a value of 5.95 ± 0.02 Mev for the alpha-particle energy.

The discovery of an interesting case of isomerism in U^{235} during a study of the α-decay of Pu^{239} led to the speculation[4] that a similar isomerism might be involved in the alpha decay of Cm^{241} to Pu^{237}; U^{235} and Pu^{237} both have 143 neutrons. This isomerism in the U^{235} case is completely discussed under the description of the decay of Pu^{239} in section 9.2.9 where it is shown that the chief α-decay of Pu^{239} goes to a state of spin 1/2 located less than one kilovolt above ground, and that this state decays by an E3 transition of 26 minute half-life to the 7/2-ground state. It was thought that Pu^{237} might have a similar 1/2- isomeric form, particularly since the ground state of Pu^{237} can be assigned[6] the same Nilsson orbital, namely 7/2 - [743], as the ground state of U^{235}. The decay scheme of Po^{237} is described in section 9.2.7.

Experiments of STEPHENS, ASARO, AMIEL AND PERLMAN[4] did indeed prove that a 0.18 second isomeric state exists in Pu^{237}. Furthermore the 145 Kev delayed transition was shown to be E3 from its total K and L conversion coefficients. Thus, the analogy to U^{235} seems complete except that the energy of the transition is much larger in the case of Pu^{237}. The isomeric state of Pu^{237} is assigned to the 1/2 - [631] Nilsson orbital on the basis of the above data. Since the favored alpha decay is to this state, it seems logical to identify this same Nilsson orbital with the ground state of Cm^{241}. These assignments are summarized in the decay scheme shown here.

We now consider the electron-capture decay of Cm^{241}. Our account follows the experimental findings and theoretical interpretation of STEPHENS, ASARO, AMIEL AND PERLMAN.[5]

Two gamma rays have been shown to accompany the electron capture decay of Cm^{241} and coincidence data indicate that other weaker transitions are probably also present.[5] The two gamma rays have energies of 470 and 600 kev. The 470 kev transition has also been seen accompanying the alpha decay of Bk^{245} to Am^{241}.[7] (See section 9.5.4). This transition is quite strong in Cm^{241}, accounting for most of the decay. It is assigned on the basis of its K conversion coefficient as principally M1, although the data indicate that there is some E2 admixture. Since no strong transitions are in coincidence with this gamma ray, and also because of the data from Bk^{245} decay, it is thought to terminate at the ground state of Am^{241}, placing the excited state at 470 kev. Less is known about the weaker 600 kev transition, but because there are no strong coincident transitions, it too is thought to terminate at the ground state, indicating a 600 kev level.

The ground state of Am^{241} has a measured spin of 5/2 and is known from its alpha decay properties to have the Nilsson configuration, 5/2 - [523]. (See discussion of Am^{241} in section 9.3.6). Another level in Am^{241} at 206 kev, populated from Bk^{245} decay, is thought to have the 5/2 + [642] assignment. No decay to either of these levels from Cm^{241} is observed; this is consistent with the K = 1/2 assignment for Cm^{241}. The 470 kev level in Am^{241} must have negative parity since it is connected with the ground state by an M1 (E2) transition; and the 600 kev level probably has negative parity also since higher-energy positive-parity levels would probably decay to the 206 kev level. We would expect levels with K values of 1/2 and 3/2 to receive most of the decay since they are populated from a K = 1/2 parent. Two such Nilsson states are available, the 1/2 - [530] which is the ground state for Pa^{231} and Pa^{233}, and the 3/2 - [521] which probably occurs as the ground state for the berkelium isotopes (except Bk^{249}). It is not easy to decide which level in Am^{241} has which of these two assignments, particularly since this 1/2 - [530] band has its 3/2 member as the lowest state in the protactinium isotope. (See discussion of Th^{231} decay in chapter 8, section 8.2.10). Because of arguments hinging on a comparison of excited levels in Am^{239}, Am^{241} and Am^{243} which are summarized in section 3.5.5 of chapter 3, we preferred the K = 1/2 assignment for the 470 kev level, and the K = 3/2 assignment for the 600 kev level, but these arguments are not conclusive. The electron capture decay scheme is also shown in the accompanying figure 31.

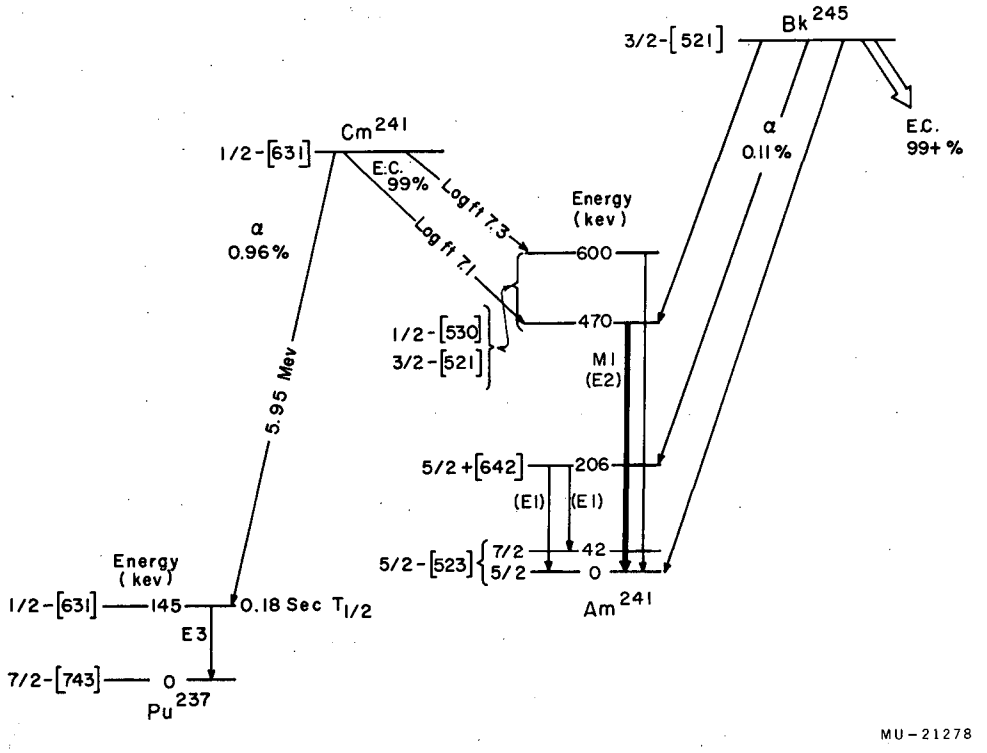

Figure 31. Decay scheme of Cm^{241}. The α-decay of Bk^{245} is also shown since knowledge gained from its study was essential to an understanding of the EC decay of Cm^{241}. The Nilsson orbital assignments are given to the left of the levels of intrinsic excitation. In order, the quantum numbers are K, parity $[N\ n_z\ \Lambda]$.

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9.4.6 Curium-242. The first isotope of curium to be identified was Cm^{242}. SEABORG, JAMES and GHIORSO[1] found that this alpha-emitting isotope was produced when Pu^{239} was bombarded with 32 Mev helium ions:

$$Pu^{239}\ (\alpha,n)\ Cm^{242}.$$

The cross section for this reaction is close to one millibarn for helium ion energy in the range 20-37 Mev.[2] Another cyclotron reaction useful in the preparation of Cm^{242} is the bombardment of Am^{241} with deuterons:

$$Am^{241}\ (d,n)\ Cm^{242}.$$

The Cm^{242} is prepared in much larger quantities by neutron irradiation of plutonium or americium. When Pu^{239} is irradiated with neutrons, Cm^{242} is formed via the reaction sequence:

$$Pu^{239}\ (n,\gamma)\ Pu^{240}\ (n,\gamma)\ Pu^{241} \xrightarrow[13\ yrs]{\beta^-} Am^{241}\ (n,\gamma)\ Am^{242m} \xrightarrow[16\ hrs]{\beta^-} Cm^{242}$$

The curium isolated from neutron-irradiated Pu^{239} may be contaminated with higher mass curium isotopes produced by further addition of neutrons to Cm^{242} or by the sequence:

$$Pu^{241}\ (n,\gamma)\ Pu^{242}\ (n,\gamma)\ Pu^{243} \xrightarrow[5\ hrs]{\beta^-} Am^{243}\ (n,\gamma)\ Am^{244} \xrightarrow[25\ min]{\beta^-} Cm^{244}\ (n,\gamma)\ Cm^{245},\ \text{etc.}$$

The isotopic composition of the curium will depend on the neutron flux and on the total integrated flux through the sample. However, it is possible to prepare samples which are almost pure Cm^{242} in terms of alpha activity.

Perhaps the best way to prepare milligram quantities of Cm^{242} is to irradiate Am^{241} with thermal neutrons.

$$Am^{241}\ (n,\gamma)\ Am^{242m} \xrightarrow[16\ hrs]{\beta^-} Cm^{242}$$

The cross section for this reaction is ~600 barns.

The half-life of Cm^{242} has been measured as 162.5 ± 2 days[3,4,5] and as 162.7 ± 0.1 days.[21] This isotope is beta stable so that all disintegrations occur by the emission of alpha particles except for a small but measurable decay by spontaneous fission to be discussed later. The alpha particle groups are summarized in Table 26. These alpha particle groups represent transitions to excited levels in Pu^{238} which are de-excited by gamma ray transitions.

TABLE 26 Alpha Particle Groups of Cm^{242}

Results of Asaro and Co-workers 6,7,12			Results of Kondrat'ev and co-workers 22		
	α-particle energy (Mev)	Energy of Pu^{238} state (kev)	Abundance (%)	α-particle energy (Mev)	Energy of Pu^{238} state (kev)	Abundance (%)
	6.110	0	73.7	6.110	0	73.5
a	6.066	44.11[b]	26.3	a 6.0656	45.1	26.5
	5.965	146	0.035	5.967	145	0.03
	5.811	303.7	6×10^{-3}	5.809	306	4.6×10^{-3}
	5.605	514	4×10^{-5}			
c	5.515	605	3.2×10^{-4}			
	5.20	~935	1.4×10^{-4}			

a. measured in magnetic α-spectrometer

b. this exact energy is the γ-transition energy

c. deduced from gamma ray measurements[6,7,12]

Since the beta decay of Np^{238} and the electron capture decay of Am^{238} result in the same endproduct nucleus, it is instructive to compare the decay schemes of the three isotopes. This is done in Fig. 32. The decay scheme of Cm^{242} is shown separately in another form in Fig. 33.

By far the most prominent gamma transition is the 44.11 kev E2 transition from the first excited state to the ground state. ASARO, THOMPSON and PERLMAN[6] found the L-shell conversion coefficient of this gamma ray to be 520. O'KELLEY[8] and PASSELL[9] measured the conversion electrons of this transition in a double focusing beta ray spectrometer and found a transition energy of 44.9 kev. NEWTON, ROSE and MILSTED[19] used a proportional counter to measure a gamma energy of 44.03 ± 0.06 kev and an abundance of 3.9×10^{-4} photons per alpha particle. SMITH and HOLLANDER[10] measured the conversion electrons in a permanent magnet spectrograph and obtained a value of 44.11 ± 0.05 kev. BARANOV and SHLYAGIN[20] report an energy of 44.1 kev. HUMMEL[13] measured an abundance of 2.9×10^{-4} 44 kev photons per alpha particle. These values are in close agreement with those reported for the identical transition which occurs in the beta decay of Np^{238}. (See Section 9.1.10). From the conversion coefficient and the L_{II}/L_{III} subshell ratio this transition is clearly electric quadrupole in nature.

The conversion of the 44.1-kev transition gives rise to L x-rays of plutonium which have been measured carefully in a Cauchois-type bent crystal spectrometer by BARTON, ROBINSON and PERLMAN.[11] Table 27 lists the x-rays which were observed.

While the plutonium L x-radiation constitutes the principal electromagnetic radiation of Cm^{242} the photons of several gamma-ray transitions have been observed in scintillation spectrometer measurements. These are summarized in Table 28. It will be noted that these gamma rays are in very low abundance corresponding to very small intensities of alpha groups to excited levels of Pu^{238}. The intensity of some of these gamma rays is so low that gamma rays from the spontaneous fission of Cm^{242} interfere with the measurements. The spontaneous-fission gamma-ray background may be decreased appreciably by anti-coincidence techniques. If the Cm sources are placed on Al backing plates, the radiations from P^{30}, formed by the (α,n) reaction, obscure all gamma rays with energy above that of the 158 kev transition.

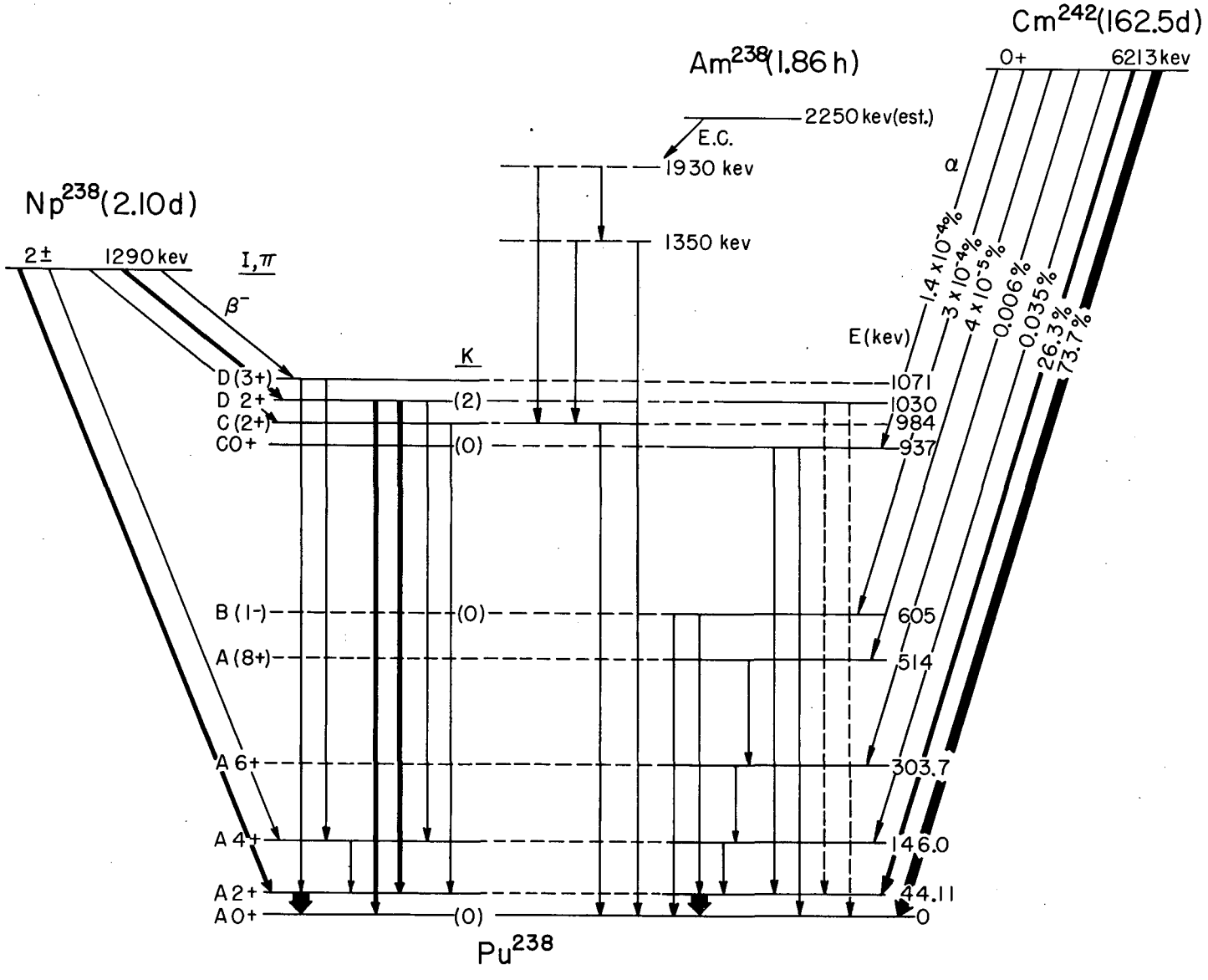

Figure 32. Level scheme of Pu^{238} deduced from decay schemes of Cm^{242}, Np^{238} and Am^{238}.

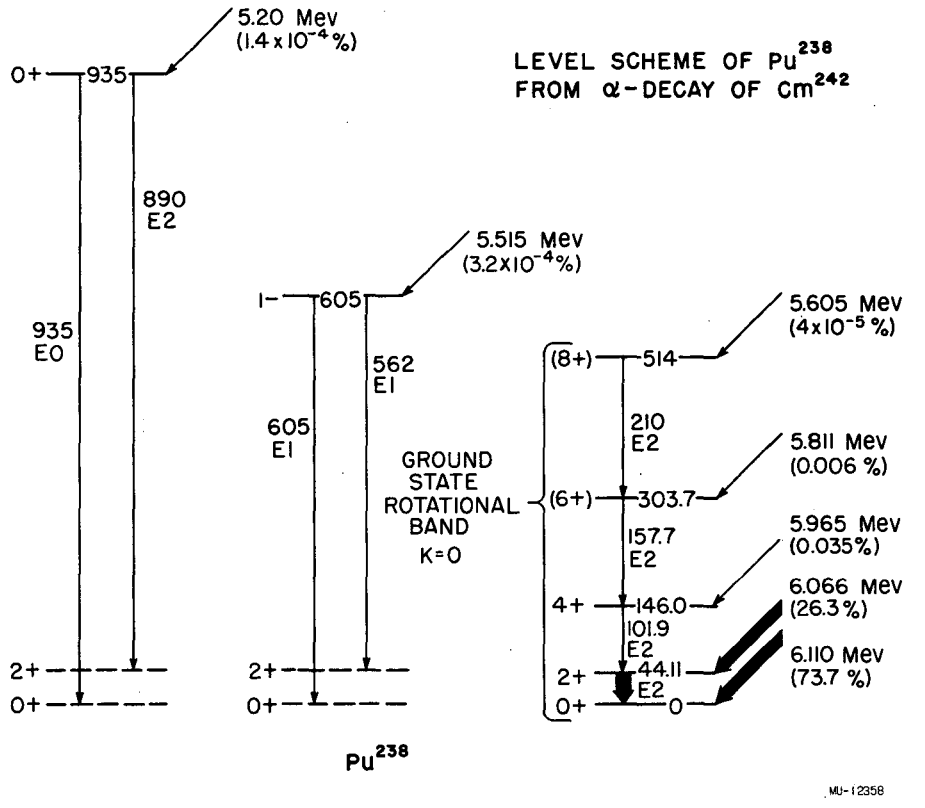

Figure 33. Decay scheme of Cm^{242} with gamma transitions isolated to clarify different features of the level scheme of Pu^{238}.

Table 27. Plutonium X-rays Emitted in the Decay of Cm^{242}

X-ray disintegration	Energy
L_{α_1}	14.31 ± 0.01
L_{α_2}	14.14 ± 0.01
L_{β_1}	18.35 ± 0.02
L_{β_5}	17.91 ± 0.02
L_{β_2}	17.28 ± 0.02
L_{γ_1}	21.46 ± 0.04
L_{γ_6}	22.20 ± 0.04

Barton, Robinson and Perlman, Phys. Rev. 81, 208 (1951).

Table 28. Gamma Rays of Cm^{242}

Energy (kev)	Photon abundances relative to total alphas	Multipole order	Comments	Reference
44.11	2.9×10^{-4}	E2		6,13,8,9,10,19
101.9	4.1×10^{-5}	E2		6,10,13,19
157.7	1.8×10^{-5}	E2	in cascade with 101.9 kev γ	6,10,13,19
210	1.5×10^{-7}	E2	in cascade with 157.7, 101.9 kev σ's	7
562	1.8×10^{-6}	E1	de-excites 1- level at 605 kev, in cascade with L x-rays of 44 kev γ	12
605	1.4×10^{-6}	E1	de-excites 1- level at 605 kev	12
890	8.6×10^{-7}	E2	de-excites 0+ level at 935 kev, in cascade with L x-rays of 44 kev γ	7
935	none observed	E0	completely converted, de-excites 0+ level at 935 kev	7

These gamma rays are placed in the decay scheme of Fig. 32 and 33. Several interesting features of the level scheme of Pu^{238} may be mentioned. A well developed rotational band based on the 0+ ground state is observed with the levels at 44.11 (2+), 146.0 (4+), 303.7 (6+) and 514 kev (8+) following closely the rotational formula given in Chapter 3 where $\hbar^2/2\Im$, is set equal to 7.37 kev and the constant B of the second order correction term is set equal to 0.0033 kev. De-excitation by the predicted cascade of E2 transitions with no observable cross over transitions is verified.

The level at 605, assigned spin and parity 1-, was deduced[12] from a pair of gamma rays at 605 kev and about 560 kev. From the electron spectrum taken with the aid of an anthracene crystal,[7] the average K conversion coefficient for these gamma rays was found to be $\lesssim 1\%$, indicating E1 and E2 transitions. The higher-energy component was not in coincidence with any photons and the lower energy component was in coincidence with L x-rays proving that it leads to the 2+ level at 44.11 kev. Such pairs of gamma rays have been seen[14] in other heavy even-even nuclei and have been proved to be E1 transitions arising from 1- states. In all cases examined in which the 1- assignment was established the reduced transition probabilities of the competing E1 transitions were found in the ratio, 0.5, expected for the K-quantum number assignment of 0 for the 1- state as well as for the rotational band based upon the ground state. This same relationship of the reduced transition probabilities was found in the present case. (Actual experimental value = 0.6). A general discussion of these 1- states is given in Section 3.6.3 of Chapter 3.

The 890 kev E2 transition and the 935-kev E0 transition shown in Table 28 and Fig. 32 and 33 are quite interesting. The electric monopole transition goes entirely by the emission of conversion electrons. Hence this transition was overlooked until considerations of excited-level systematics in heavy element nuclei (see discussion in Section 3.6.2 in Chapter 3) suggested that such an electric monopole transition might exist. Such a transition exists for example in the de-excitation of excited levels of U^{234}. (The 806-kev transition of the decay of UX_2). CHURCH and WENESER[18] emphasized the probable importance of E0 transitions in heavy nuclei. For these reasons and also because photons of an 890-kev E2 transition had been found in cascade with the L x-rays of the 44.11-kev transition discussed above, it seemed worthwhile to look for

conversion electrons of a completely converted transition from a 935 kev 0+ level to the 0+ ground state. ASARO and CO-WORKERS[7] found these conversion electrons using an anthracene crystal spectrometer. The assignment of the multipolarity is unambiguous. The ratio of the total transition rates of the 890-kev E2 gamma ray to the 935-kev E0 transition is 1.67.

The other higher levels of Pu^{238} are hard to characterize by studying α decay of Cm^{242} because of the extremely low α-branching to these levels. These are better studied in the β-decay of Np^{238}. (See Section 9.1.12).

GHIORSO and ROBINSON[15] made the observation that the isotope Cm^{242} undergoes spontaneous fission at a high rate (3×10^{10} fissions per gram-hour), corresponding to a half-life for this process of about 7×10^{6} years. HANNA, HARVEY, MOSS and TUNNICLIFFE[16] report this half-life to be $(7.2 \pm 0.2) \times 10^{6}$ years corresponding to 2.7×10^{10} fissions per gram-hour. Many characteristics of the spontaneous fission of Cm^{242} have been measured. These experiments are thoroughly discussed in Chapter 11.

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9.4.7 Curium-243. When a sample of Am^{241} or Cm^{242} is strongly irradiated with slow neutrons, some Cm^{243} is produced via the reactions

$$Am^{241}(n,\gamma)Am^{242m} \xrightarrow[16\ hr]{\beta^-} Cm^{242}(n,\gamma)Cm^{243}$$

REYNOLDS, HULET and STREET[1] first produced this isotope in this manner and identified it by mass spectroscopic analysis. The conversion of Am^{241} to Cm^{242} and higher isotopes of curium is illustrated graphically in Fig. 34.

The isotope Cm^{243} has also been identified experimentally as the daughter of 4.6-hour Bk^{243}.

Curium-243 decays by the emission of alpha particles. An early published half-life value associated with the decay of three of the most intense groups was about 35 years, determined from mass spectrographic and alpha spectrum measurements.[2] Later measurements[3] discussed below showed that these groups accounted for only 90% of the alpha emission, thus reducing the half life to 32 years.

ASARO, THOMPSON, STEPHENS and PERLMAN[3] measured the alpha spectrum of samples of curium containing varying amounts of Cm^{242}, Cm^{243} and Cm^{244} and assigned to Cm^{243} the groups listed in Table 29.

The alpha particle groups define excited levels of Pu^{239} as shown in Fig. 35. An interpretation of the level scheme of Pu^{239} is given in Section 9.1.12 where the decay of Np^{239} is described. The complex structure of the α-spectrum of Cm^{243} implies the existence of many low-intensity gamma rays. Experimental difficulties have retarded the investigation of many of these. The data of NEWTON, ROSE and MILSTED[5] on the gamma spectrum are summarized in Table 30. The 278- and 228-kev gamma rays have been shown to be in coincidence with 5.777-Mev alpha particles.[3]

ASARO, THOMPSON, STEPHENS and PERLMAN[3] have interpreted the alpha decay to the levels shown in Fig. 35 in terms of the theory of BOHR, FROMAN and MOTTELSON[6] for unhindered (favored) alpha decay.

Table 31 shows the calculated and experimental alpha particle populations to the rotational states with the same intrinsic configuration as the parent nucleus, Cm^{243}. The decay probabilities for alpha particles with various angular momenta were taken from the adjacent even-even nuclides. Table 32 shows the data for hindered (unfavored) alpha decay to states with a different

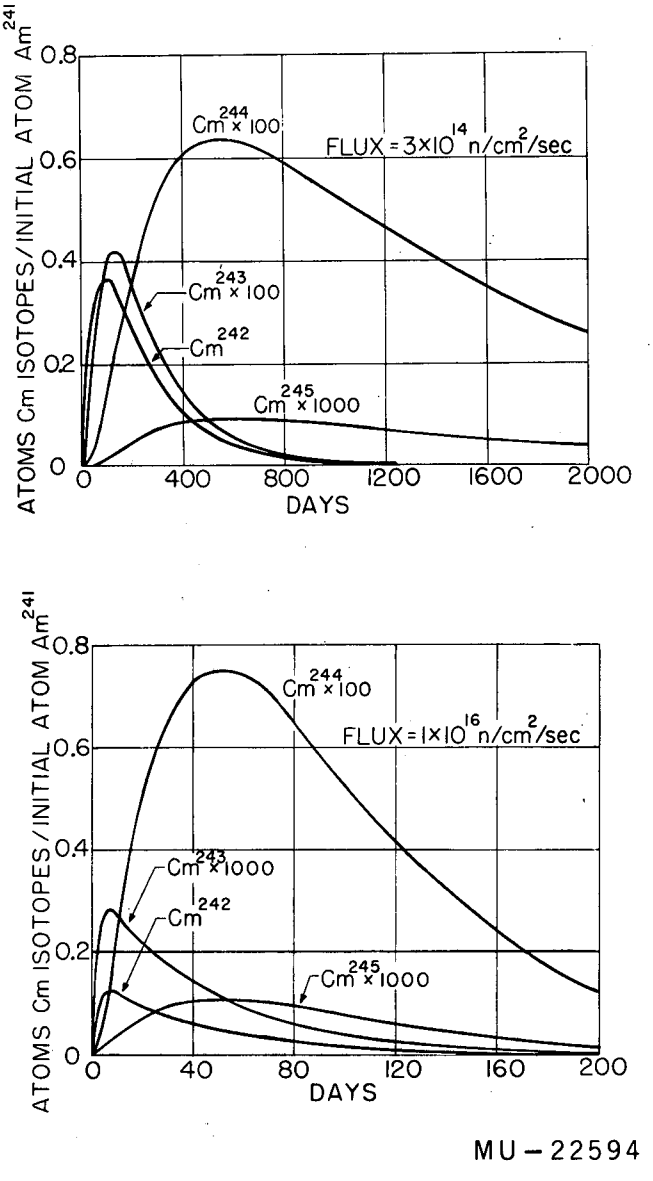

Figure 34. Buildup of curium isotopes in Am^{241} by the neutron capture sequence

$$Am^{241}\ (n,\gamma)\ Am^{242m} \xrightarrow[16\ hr]{\beta^-} Cm^{242}\ (n,\gamma)\ Cm^{243}\ (n,\gamma)\ Cm^{244}\ (n,\gamma)\ Cm^{245}.$$

Curves are given for two neutron fluxes.

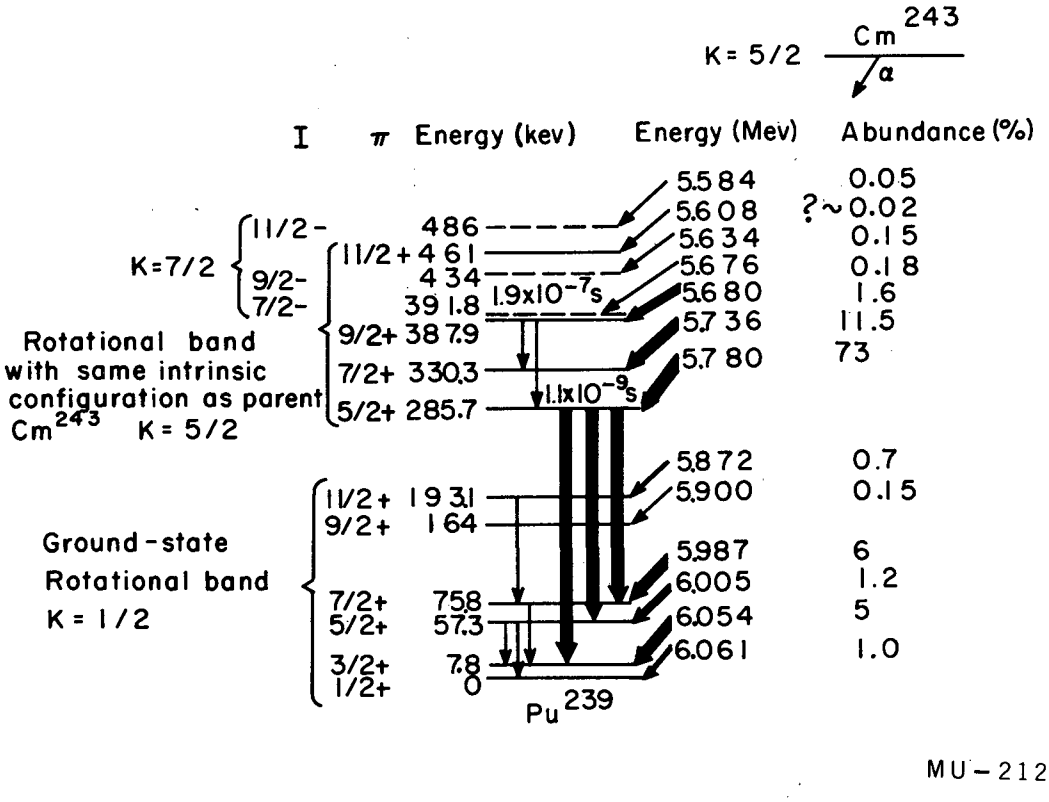

Figure 35. Decay scheme of Cm^{243} including interpretation of Pu^{239} levels as three sets of rotational levels based on three states of intrinsic excitation (Nilsson orbitals).

Table 29. Alpha Groups of Cm^{243}

6.061 Mev	1.0%
6.054 Mev	5%
6.005 Mev	1.2%
5.987 Mev	6%
5.900 Mev	0.15%
5.872 Mev	0.7%
5.780 Mev	73%
5.736 Mev	11.5%
5.680 Mev	1.6%
5.676 Mev	0.18%
5.634 Mev	0.15%
5.608 Mev ?	~0.02%
5.584 Mev	0.05%

From Asaro, Thompson, Stephens and Perlman, Phys. Rev. 92, 694 (1953); Bull. Am. Phys. Soc. 8, 393 (1957); and unpublished results, 1956-1957.

Table 30. Gamma Rays of Cm^{243}

Gamma ray energy (kev)	Relative intensity	
102.15 ± 0.2	3.6 ± 0.2	K_α Pu x-ray
117.3 ± 0.4	1.4 ± 0.2	K_β Pu x-ray
210 ± 1.5	0.5 ± 0.15	
228 ± 2	0.65 ± 0.15	
277 ± 2	1.00	

From Newton, Rose and Milsted, Phil. Mag. 1, 981 (1956).

Table 31. Cm^{243} Alpha Decay to 5/2+ Band of Pu^{239}

Unhindered decay $P_\alpha \propto \sum_L \frac{1}{H.F.} \begin{bmatrix} I_i & L & I_f \\ K_i & K_f - K_i & K_f \end{bmatrix}^2$

I	E	Calculated abundances (%) L = 0	L = 2	L = 4	L = 6	Sum	Experimental abundances
5/2	286	57.0 (norm)	16.0	0.0	--	73	73
7/2	330	--	9.2	0.0	0.0	9.2	11.5
9/2	388	--	1.57	0.01	0.0	1.58	1.6
11/2	461	--	--	0.0037	0.0026	0.006	~0.02?
13/2	547 (calc)	--	--	0.0004	0.0015	0.002	<0.02
Assumed relative hindrance factors from Cm^{242} and Cm^{244} decay		1.00	1.82	590	580		

Table 32. Cm^{243} Alpha Decay to 7/2- Band

$P_\alpha(\text{reduced}) \propto \sum_L \frac{1}{H.F.} \begin{bmatrix} I_i & L & I_f \\ K_i & K_f - K_i & K_f \end{bmatrix}^2 \quad L < K_i + K_f$

I	E (kev)	Calculated abundances L = 1	L = 3	Sum	Experimental abundances
7/2	392	0.07 (norm)	0.11	0.18 (norm)	0.18
9/2	434	--	0.15 (norm)	0.15 (norm)	0.15
11/2	485	--	0.042	0.042	0.05
Hindrance factor		4×10^2	59		

Table 33. Alpha Decay of Cm^{243} to 1/2+ Band of Pu^{239}

$$P_{\alpha,L}(\text{reduced}) \propto \left[C^{I_i\ L\ I_f}_{K_i\ K_f-K_i\ K_f} + b(-1)^{I_f+K_f} C^{I_i\ L\ I_f}_{K_i,\ -K_f-K_i,\ -K_f} \right]^2 \quad L \geq K_f+K_i$$

I	E (kev)	Calculated abundances L = 2	L = 4 b = 1.9	L = 6 b = 2.2 b = 0.3	Sum	Experimental abundances
1/2	0	1.00% (norm)	--	--	1.00% (norm)	1.0%
3/2	8	1.05	3.78 (norm)	--	4.83 (norm)	4.8
5/2	57	0.33	0.86 (norm)	--	1.19 (norm)	1.2
7/2	76	0.08	5.22	0.45	5.75	6.1
9/2	164	0.00	0.03	0.12 (norm)	0.15 (norm)	0.15
11/2	193	--	0.21	0.49 (norm)	0.70 (norm)	0.70
Hindrance factor		1100	170	400		

intrinsic configuration than the parent nucleus. For this type of decay the relative decay probabilities are not known for alpha particles with different angular momenta and are arbitrarily normalized to give the proper experimental ratio for the 392 and 434 kev levels. The calculated population to the 485 kev level serves as an independent check of the method and is in satisfactory agreement with the experimental value. The hindered decay to the ground state rotational band is more complicated because an appreciable portion of the decay takes place by alpha particles whose angular momenta are equal or less than the sum of $K_i + K_f$. Waves of a given angular momentum then populate levels with $\pm K_f$ and can interfere with each other. Table 33 shows the calculated and experimental abundances. Five of the experimental abundances were used to evaluate the parameters in the equations and the sixth value served as an independent check of the method.

CHOPPIN and THOMPSON[7] found that Cm^{243} is slightly unstable toward electron capture decay with an electron capture half life of $(1 \pm 0.1) \times 10^4$ years. Previously[4] a lower limit of 50,000 years had been placed on the electron capture half life.

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9.4.8 Curium-244. The long-lived curium isotope Cm^{244} was first identified[1] by mass spectrographic analysis of a curium sample produced by neutron irradiation of Am^{241}. The Cm^{244} is produced in such an irradiation through four separate paths, because of the different modes of decay of Am^{242} and the existence of isomeric forms of Am^{242}.

$$Am^{241}(n,\gamma)Am^{242}(100\ yr);\ Am^{242}(n,\gamma)Am^{243}(n,\gamma)Am^{244} \xrightarrow{\beta^-} Cm^{244}$$

$$Am^{241}(n,\gamma)Am^{242}(16\ hr);\ Am^{242}(n,\gamma)Am^{243}(n,\gamma)Am^{244} \xrightarrow[25\ min]{\beta^-} Cm^{244}$$

$$Am^{242} \xrightarrow[16\ hr]{\beta^-} Cm^{242}(n,\gamma)Cm^{243}(n,\gamma)Cm^{244}$$

$$Am^{242} \xrightarrow[16\ hr]{EC} Pu^{242}(n,\gamma)Pu^{243} \xrightarrow[5\ hr]{\beta^-} Am^{243}(n,\gamma)Am^{244} \xrightarrow[25\ min]{\beta^-} Cm^{244}.$$

Figure 34 in the previous section illustrates the rate of build-up of Cm^{244} as a function of neutron irradiation. These curves are approximate because of uncertainties in the neutron capture cross sections of some of the isotopes in the chain.

The most satisfactory way to make large amounts of Cm^{244} in high isotopic purity is to irradiate Pu^{239} or Pu^{242} with neutrons in a high flux pile. The main sequence of reactions leads directly to Cm^{244} as follows:

$$Pu^{239}(n,\gamma)Pu^{240}(n,\gamma)Pu^{241}(n,\gamma)Pu^{242}(n,\gamma)Pu^{243} \xrightarrow[5\ hr]{\beta^-} Am^{243}(n,\gamma)Am^{244} \xrightarrow[25\ min]{\beta^-} Cm^{244}.$$

The build-up of Cm^{244} by this mechanism when the neutron flux is 3×10^{14} $n/cm^2/sec$ is shown in Chapter 5. In an experiment[2,3] in which Pu^{239} was irradiated at this flux until the total integrated flux was 1.4×10^{22} neutrons/cm^2 it was found by mass spectrographic analysis that the isotopic composition of the curium fraction was the following:

Cm^{242}	0.68 %
Cm^{244}	96.29 %
Cm^{245}	1.14 %
Cm^{246}	1.86 %
Cm^{247}	0.028%

If it is desirable to achieve a higher isotopic purity the above neutron capture sequence can be interrupted when a good yield of Am^{243} has been achieved, the

Am^{243} can be chemically purified and then reinserted in the reactor for the final transmutation to curium via the short sequence:

$$Am^{243}(n,\gamma)Am^{244} \xrightarrow[25\ min]{\beta^-} Cm^{244}$$

The half life of Cm^{244} has been determined in three ways. THOMPSON, HULET AND GHIORSO[5] followed the decay directly to obtain a value of 19 years. STEVENS AND CO-WORKERS[2] determined the atom ratio and alpha activity ratio of Cm^{244} to Cm^{242} in a mixture of the two isotopes and from the known half life of 162.5 days for Cm^{242} were able to calculate an alpha half life of 19.2 ± 0.6 years for Cm^{244}. FRIEDMAN AND CO-WORKERS[4] measure the atom ratio of Cm^{244} to Cm^{242} in a mixture of the two isotopes. After a certain decay period the plutonium daughters Pu^{240} and Pu^{238} were isolated and the ratio of the two determined mass spectrographically. From these data and the known half-life of Cm^{242} the half life of Cm^{244} was calculated to be 17.9 ± 0.5 years. CARNALL, FRIED AND HARKNESS measured the specific activity of a curium sample (in the form of $CmCl_3$) which was 94.57 percent Cm^{244} in isotopic content and almost pure Cm^{244} by alpha activity. They reported a half-life of 17.59 ± 0.06 years.

The alpha spectrum of Cm^{244} was first determined on a magnetic spectrograph by ASARO, THOMPSON, AND PERLMAN[6] who reported alpha groups of 5.798 (75 percent) and 5.755 Mev (25 percent). WHITE AND CO-WORKERS[8] later remeasured the energy of the main transition more carefully relative to Po^{210} and obtained a better value of 5.8025 ± 0.002 Mev. HUMMEL, ASARO AND PERLMAN[7] measured the energies and abundances of the alpha groups of Cm^{244} relative to the ground state group. Their results combined with those of WHITE[8] leads to the results given in Table 34. Some later results of ASARO AND PERLMAN[13] on low intensity groups are also included.

HUMMEL[7] has studied the gamma rays of Cm^{244} using a scintillation spectrometer. He found three gamma rays with the following abundances relative to the alpha disintegration rate: 43 kev (2.1×10^{-4}), 100 kev (1.3×10^{-5}) and 150 kev (1.4×10^{-5}). SMITH AND HOLLANDER[9] measured the conversion electrons of the first of these gamma rays. They found a transition energy of 42.88 ± 0.05 kev and defined the transition as E2 from the absence of L_I electrons and from an L_{II}/L_{III} conversion ratio of 1.10 in agreement with the theoretical prediction. In the electron capture decay of Am^{240} to the same Pu^{240} daughter nucleus the 42.88-kev gamma ray also appears as shown by SMITH, GIBSON AND HOLLANDER.[10] The 100 kev gamma ray occurs also in Am^{240} decay and the conversion electron measurements show it to be a 98.90 ± 0.2 kev E2 transition.

Table 34. Alpha Particle Groups of Cm^{244}

	Energy (Mev)	Abundance	Energy of excited level in Pu^{240} daughter (kev)
	5.802±0.002	76.7	0
a	5.760	23.3	42.9
	5.662	0.017	142.2
	5.507±002	$(3.6\pm0.3)\times10^{-5}$	295
b	4.950±005	$(1.4\pm0.3)\times10^{-6}$	861
	4.910±.005	$(6\pm2)\times10^{-7}$	901

Data of Hummel, Asaro and Perlman (1956)

Data of Asaro and Perlman (1960)

These data on the alpha and gamma transitions of Cm^{244} lead to the decay scheme shown in the figure. This resembles the decay scheme of all other even-even nuclei in this mass region. The excited levels of Pu^{240} are members of a rotational band of levels based on a 0+ (K=0) ground state. Other information on the levels of Pu^{240} comes from the beta decay of Np^{240}. See Section 9.4.13.

Curium-244 is beta stable.

The spontaneous fission rate of Cm^{244} is quite high compared to isotopes of lighter elements. GHIORSO and CO-WORKERS[11] measured a spontaneous fission rate of $(1.4 \pm 0.2) \times 10^{10}$ fissions per gram hour corresponding to a half life of $(1.4 \pm 0.2) \times 10^{7}$ years.

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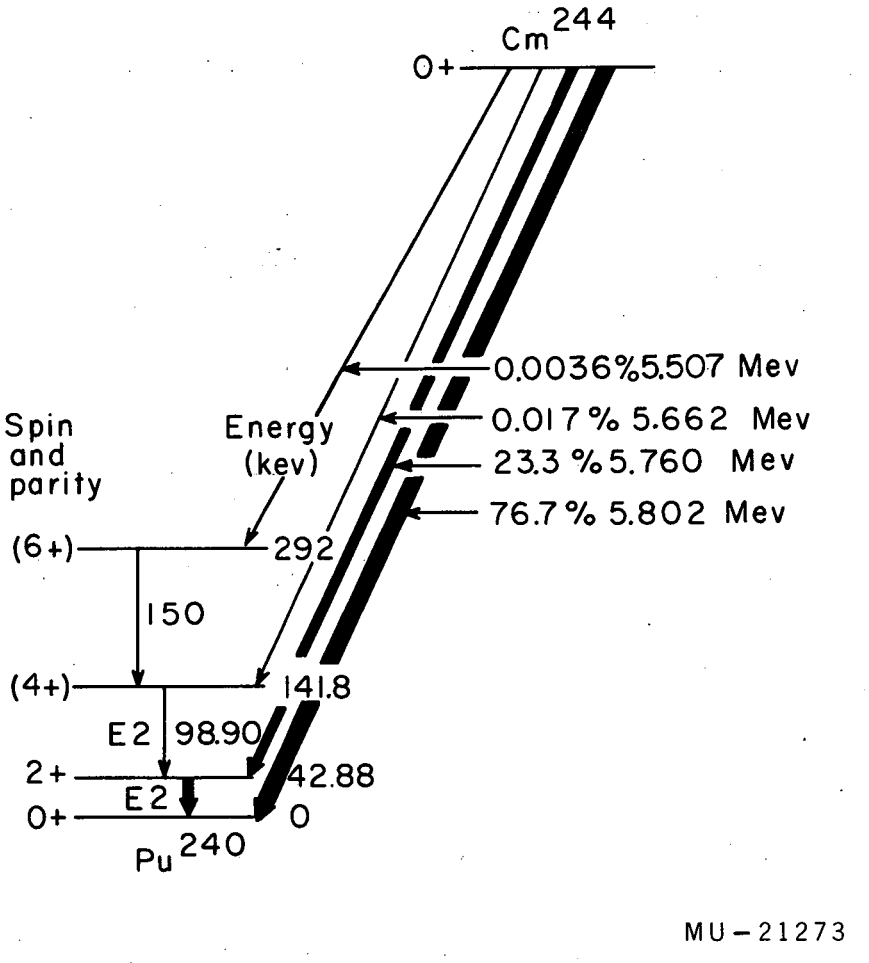

Figure 36. Decay scheme of Cm^{244}. Two additional alpha groups leading to levels at 861 and 907 kev in the intensities 1.4×10^{-6} and 6×10^{-7} respectively are not shown.

9.4.9 Curium-245. The existence of Cm^{245} was first proved by REYNOLD'S [1] mass spectrographic analysis of samples of curium subjected to prolonged neutron irradiation in the Chalk River reactor. Pure but small samples of Cm^{245} were first obtained[2,3] by examining the curium daughter fraction from a sample of berkelium which contained 4.95-day Bk^{245}, an isotope which decays chiefly by electron capture. The curium fraction from Pu^{239} subjected to prolonged neutron irradiation also contains Cm^{245}. STEVENS AND CO-WORKERS[4] report the values quoted in Table 35 given in the next section for the isotopic composition of a curium fraction isolated from a sample of Pu^{239} irradiated at a flux of 3×10^{14} neutrons/cm^2/sec for a total irradiation of 8×10^{21} neutrons/cm^2.

Curium-245 has been found in the curium fraction isolated from the debris of thermonuclear test explosions. In one case the curium fraction was mass analyzed and found to contain 68.7 mole percent Cm^{245}, the rest of the sample being higher mass isotopes.[8] (See Table 35 in the next section).

FRIEDMAN AND CO-WORKERS[5] measured the half-life of Cm^{245} by mass spectrographic techniques and determined a value of $(1.15 \pm 0.5) \times 10^4$ years. BROWNE AND CO-WORKERS[9] report a value of 14,300 years ± 20 percent. DIAMOND[12] reports a value of $(7.5 \pm 1.9) \times 10^3$ years. CARNALL, FRIED AND HARKNESS[13] obtained a value of 9320±280 based on mass spectrometric assay of the isotopic composition of plutonium growing into a mixed sample of Cm^{245} and Cm^{244}. This value also depends on a choice of 17.59 years for the half-life of Cm^{244}.

The energies of the alpha particles of Cm^{245} have been measured roughly by the ionization chamber method. HULET AND CO-WORKERS[3] report a main group at 5.36 Mev and a smaller intensity group[6] at 5.45 Mev. ASARO, THOMPSON AND PERLMAN[7] find that the prominent features of the gamma ray spectrum of Cm^{245} as determined by a scintillation spectrometer are K x-rays and 130-kev and 172-kev gamma rays. The gamma ray results indicate the existence of an alpha particle group at 5.31 Mev. They also suggest that the most intense alpha group proceeds to an excited state at 172 kev in Pu^{241} which de-excites to the ground state via a 172 kev M1 transition. The alpha spectrum is given as 5.45 Mev (~15 percent), 5.36 Mev (~77 percent) and 5.31 (~8 percent). ASARO AND PERLMAN[14] measured the energy 5.352±0.005 for the energy of the most intense group.

Curium-245 is beta stable.

More experimental data are required before a reliable decay scheme can be constructed for Cm^{245} but a few features can be interpreted in terms of Nilsson orbital assignments. This has been done in the following way by STEPHENS, ASARO AND PERLMAN[10] and by NILSSON AND MOTTELSON.[11] The ground state spin of Pu^{241} has been measured as 5/2. The 5/2+ 633 orbital was the ground

state for neutron number 141 so the ground state of Pu^{241} (neutron number 147) is almost surely 5/2+ [622]. This follows from an examination of the Nilsson diagram for neutrons in the region $126 \leq N \leq 160$. See Chapter 3. The favored alpha decay of Cm^{245} leads to a state 172 kev above ground. Another state of 58 kev higher energy has been observed and interpreted as the first member of the rotational band based upon the 172 kev state.[7] This spacing suggests that the band has K = 7/2 or higher. The parity is fixed as even from the observation that the 172 kev state decays to the ground-state band by M1 transitions. This fact not only fixes the parity of the 172 kev state but also is consistent with the spin assignment of 7/2. The only Nilsson assignment in this region with these properties is 7/2+ [624] and the assignment is considered to be reasonably certain. On the basis that favored alpha decay occurs between parent and daughter states with the same Nilsson orbital for the odd nucleon (BOHR, FROMAN and MOTTELSON, 1955), the ground state assignment of Cm^{245} is also 7/2+ [624]. Several excited levels of Cm^{245} are seen in the α-decay of Cf^{249}, the electron capture decay of Bk^{245} and the beta decay of Am^{245}. The interpretation of these excited levels is given in the discussion under Cf^{249}, Section 9.6.7.

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9.4.10 The Curium Isotopes Cm^{246}, Cm^{247}, Cm^{248}, Cm^{249} and Cm^{250}: The first evidence for Cm^{246}, Cm^{247}, and Cm^{248} came from mass spectrographic analysis of a curium fraction isolated from the debris of a thermonuclear test explosion.[1] Some U^{238} present in the test device was transmuted in the very high instantaneous flux to higher mass isotopes up through mass number 255. Beta decay which occurred subsequent to the explosion resulted in the production of isotopes of higher elements. The analysis of the curium fraction is given in Table 35.

No detectable amounts of Cm^{242}, Cm^{243} and Cm^{244} appeared in the bomb debris samples because the beta-decay chains were interrupted at Pu^{242}, Am^{243} and Pu^{244} respectively. For the higher-mass chains the sequences are as follows:

$$U^{245} \xrightarrow[\text{short}]{\beta^-} Np^{245} \xrightarrow[\text{short}]{\beta^-} Pu^{245} \xrightarrow[12 \pm 1 \text{ hr}]{\beta^-} Am^{245} \xrightarrow[2.08 \text{ hr}]{\beta^-} Cm^{245} \quad \text{(ref. 4)}$$

$$U^{246} \xrightarrow[\text{short}]{\beta^-} Np^{246} \xrightarrow[\text{short}]{\beta^-} Pu^{246} \xrightarrow[11 \text{ day}]{\beta^-} Am^{246} \xrightarrow[25 \text{ min}]{\beta^-} Cm^{246} \quad \text{(ref. 6)}$$

$$U^{247} \xrightarrow[\text{short}]{\beta^-} Np^{247} \xrightarrow[\text{short}]{\beta^-} Pu^{247} \xrightarrow[\text{short}]{\beta^-} Am^{247} \xrightarrow{\beta^-} Cm^{247}$$

$$U^{248} \xrightarrow[\text{short}]{\beta^-} Np^{248} \xrightarrow[\text{short}]{\beta^-} Pu^{248} \xrightarrow[\text{short}]{\beta^-} Am^{248} \xrightarrow{\beta^-} Cm^{248}$$

The isotopes, Cm^{246} and Cm^{247} have also been prepared by intensive neutron irradiation of Pu^{239} in high flux reactors.[2] Column 3 of the table shows a mass spectrographic analysis of a curium fraction produced in such a way.[2] The mole percentages of the higher mass isotopes can be increased substantially by longer irradiation of such preparations.

The isotopes Cm^{246} and Cm^{248} have also been isolated as the daughter products of Cf^{250} and Cf^{252} respectively.[5]

The alpha half-life of Cm^{246} was determined by FRIEDMAN and CO-WORKERS[3] by a mass spectrometric technique. A curium sample with the isotopic composition given in the last column of Table 35was purified thoroughly from plutonium daughter activity. Then after a two week interval the newly-formed plutonium daughter activity containing a mixture of $Pu^{238\text{-}242}$ was mass analyzed. The half-life of Cm^{246} was obtained from the following relationship:

$$t_{1/2}\,(Cm^{245}) = t_{1/2}\,(Cm^{242}) \frac{\text{mole \% } Cm^{245}}{\text{mole \% } Cm^{242}} \times \frac{\text{mole \% } Pu^{238}}{\text{mole \% } Pu^{241}}$$

Table 35. Mass Spectrographic Analysis of Curium Samples Containing Isotopes of High Mass Number

Isotope	Composition (in mole percent)	
	Thermonuclear "Mike" test debris. Ref. 1	Curium[2] from Pu^{239} irradiated with 8×10^{21} neutrons per cm^2
Cm^{242}	---	1.84
Cm^{244}	---	99.51
Cm^{245}	68.7 ± 0.4	1.27
Cm^{246}	28.4 ± 0.4	1.36
Cm^{247}	2.2 ± 0.1	0.016
Cm^{248}	0.7 ± 0.2	---
Cm^{249}	$0.5 \pm \begin{matrix} 0.2 \\ 0.5 \end{matrix}$	

A value of 4000 ± 600 years was obtained.

BROWNE AND CO-WORKERS[4] obtained the value 2300 ± 460 years and BUTLER AND CO-AUTHORS[5] obtained 6620 ± 320 years for the same half life by quite different methods. CARNALL, FRIED AND HARKNESS[11] redetermined the Cm^{246} half-life by a mass spectrometric method identical with that used by FRIEDMAN AND CO-WORKERS[3] with the difference that Cm^{244} rather than Cm^{242} was used as the reference isotope. They reported a value of 5480 ± 140 years for Cm^{246} based on a Cm^{244} half-life of 17.59 years.

The energy of the principal alpha group has been reported as 5.373 ± 0.010 Mev[5] and as 5.378 ± 0.005 Mev[12]. The abundance of this group is 79% and that of a second group at 5.334 ± 0.005 Mev is 21%. The spontaneous fission half-life is (2.0 ± 0.8) x 10^7 years.[6] Curium 246 is beta stable.

DIAMOND, FRIEDMAN, GINDLER AND FIELDS[7] have measured the half-life of Cm^{247} and have discussed the interesting possibility that it may exist in nature. The measurement of the half-life is difficult because of the lack of curium samples containing much Cm^{247}, but, on the other hand, the 4.98 hour Pu^{243} daughter provides a sensitive method for the detection of alpha decay. By searching for Pu^{243} daughter activity in a curium sample of known Cm^{247} content and finding none, these investigators set a conservative lower limit of 4 x 10^7 years and a probable lower limit of 9 x 10^7 years to the half life. Other modes of decay were considered. Curium-247 should be beta stable and spontaneous fission systematics would predict a fission half-life of more than 6 x 10^8 years.

With a half-life of the order of 10^8 years it is possible that Cm^{247}, or daughter material attributable to the decay of Cm^{247}, may be identified in terrestrial material. If one makes a reasonable guess as to the primordial abundance of Cm^{247} and sets a limit of detection for curium by mass spectrometric methods of one atom of Cm^{247} per 10^{15} atoms of rare earth one concludes that there is some hope of isolating curium in old rare earth minerals. Curium-247 is a nuclide of the 4n + 3 type decaying into U^{235} by the following decay chain:

$$Cm^{247} \xrightarrow{\alpha} Pu^{243} \xrightarrow[4.98\ hr]{\beta^-} Am^{243} \xrightarrow[8600\ yr]{\alpha} Np^{239} \xrightarrow[2.33\ days]{\beta} Pu^{239} \xrightarrow[24,400\ yr]{\alpha} U^{235}.$$

Hence the effects of Cm^{247} might be detected in very old rare earth minerals by finding an abnormally high ratio of U^{235} to U^{238}. Curium-247 and its daughters should have been an appreciable source of radioactive heat for at least the first half-billion years after the formation of the elements. The decay of one atom of Cm^{247} into U^{235} releases (6.23 ± 0.13) x 10^{-13} calories.

The isotope Cm^{248} has not been made in sufficiently high isotopic composition by the neutron irradiation technique that significant measurements of

its radiations could be made. It is a product of the alpha decay of Cf^{252}, however, and since sizable amounts (micrograms and larger) of Cf^{252} can be made, it is possible to isolate Cm^{248} alpha activity in a relatively pure state. Californium produced by neutron irradiation of plutonium consists of Cf^{249}, Cf^{250}, Cf^{251} and Cf^{252}, but most of the alpha activity is due to Cf^{252} since it has the shortest half-life and is prepared in high abundance. Most of the remaining alpha activity is assignable to Cf^{250}. Thus, the curium daughter fraction will consist mainly of Cm^{248} and Cm^{246} with very small amounts of Cm^{247} and Cm^{245}. By this "milking" technique BUTLER and CO-AUTHORS[5] determined an alpha half-life for Cm^{248} of $(4.7 \pm 0.4) \times 10^5$ years, based on a half-life of 2.2 ± 0.2 years for Cf^{252}. The alpha particle energy is 5.054 ± 0.015 Mev. They measured a spontaneous fission half-life of $(4.6 \pm 0.5) \times 10^6$ years. It is interesting that the spontaneous fission accounts for 11% of the total decay events. THOMPSON, GHIORSO, HARVEY and CHOPPIN[10] first found evidence for a beta-emitting Cm^{249} produced by the prolonged neutron irradiation of plutonium. The isotope Cm^{249} was also prepared[1] by neutron irradiation of a part of the curium fraction from a thermonuclear test explosion whose composition is given in Table 3.5. Curium-249 has a half-life of 65 minutes for the emission of 0.9 Mev beta particles.[1] EASTWOOD and SCHUMAN[9] report a half life of 64 ± 3 minutes and a maximum beta energy of 0.86 ± 0.10 Mev.

The only information we have on Cm^{250} is a crude estimate of the spontaneous fission half-life which HUIZENGA and DIAMOND[8] report to be roughly 2×10^4 years. From the absence of Cf^{250} in the debris of the 1958 "Mike" test explosion a lower limit of 130 years has been set on the β^- decay half-life.

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9.5 THE ELEMENT BERKELIUM (Element 97)

9.5.1 The Discovery of Berkelium. The first isotope of element 97 was identified by THOMPSON, GHIORSO and SEABORG[1] in 1949. This isotope was Bk^{243} produced in the 60-inch Berkeley cyclotron by the reaction:

$$Am^{241} (\alpha, 2n) Bk^{243}$$

The isotope Bk^{243} decays predominantly by orbital electron capture with a half-life of 4.6 hours although in approximately 0.1% of its disintegrations it emits alpha particles.

The preparation of element 97 by this method represents a straightforward extrapolation of experiments used in the initial preparation of other transuranium elements. However, a number of experimental difficulties made it necessary to modify considerably the previous experimental techniques.

The search for transcurium elements was begun in the fall of 1945. SEABORG[2] anticipated that element 97, as eka-terbium in the actinide transition series, would possess oxidation states (III) and (IV) with properties similar to curium in the (III) oxidation state and to plutonium (IV) in its (IV) oxidation state. It was more difficult to estimate the oxidation potential of the (III) —> (IV) couple but it was expected that element 97 would be somewhat easier to oxidize than terbium (III) which is not oxidizable to higher states in aqueous solution. The salient point is that if element 97 could not be converted to an oxidation state higher than (III) in solution it would be difficult, with the techniques than available, to separate it in a short period of time from rare earth fission product elements and from the actinide elements target material from which it was produced. It appeared that it might be necessary to use tedious rare earth separation procedures in order to separate and identify the new element, whose isotopes produced by cyclotron bombardments would have short half-lives as a result of considerable instability toward alpha-particle emission and electron-capture decay.

In view of the fact that sufficiently intense beams of energetic particles of nuclear charge greater than two were not available, or at least were not thought to be available, at the time of this research, there were only two methods of approach to the production of element 97. The first method was the bombardment of americium with helium ions or the bombardment of curium with

deuterons or helium ions. The second was through intensive neutron irradiations of curium in order to produce through successive (n,γ) reactions a curium isotope decaying by emission of a beta particle to form an isotope of element 97.

Both methods were employed in attempting to observe element 97, and for each, different chemical procedures were used. Some of these procedures were designed to separate the new element in oxidation states greater than (III) and others were used on the assumption that element 97 existed in solution in the tripositive oxidation state.

Although the broad assumptions made when the work was started were all correct, the experiments done prior to December 1949, were unsuccessful for many reasons which may be grouped into three classes. First, the methods of predicting the properties of the new isotopes were relatively undeveloped and the experiments were never done with sufficient speed. The further development of the alpha-decay systematics[3] made it possible to estimate energies and half-lives for alpha-particle decay more reliably. The resulting estimated alpha-particle decay energies could be used in calculating the total energies for electron-capture decay or beta-particle decay by the method of closed decay-energy cycles. An empirical method of estimating electron-capture half-lives from disintegration energies was developed by THOMPSON.[4]

The second major difficulty was that of obtaining sufficiently large amounts of americium and curium as sources for the production of element 97. Eventually, americium became available in milligram amounts through neutron irradiation of plutonium. The curium was produced in smaller amounts by the irradiation of americium with neutrons.

The intense radioactivity of the americium and curium source materials presented the third major difficulty. The specific activity of a milligram of americium and curium is 7×10^{10} and $\sim 10^{14}$ disintegrations per minute, respectively. This radioactivity necessitated not only the design and development of advanced techniques and equipment for its safe handling, but also made it necessary to attain enormous separation factors in the isolation of the new element from the target material in order to be able to detect the small amounts of radioactivity due to the new element. Furthermore, this high degree of separation had to be carried out in good yield in a short length of time.

Essentially, three chemical steps were developed to solve these chemical problems. The conversion of the americium to a hexapositive oxidation state according to the method of ASPREY, STEPHANOU and PENNEMAN[5] was used in the rapid removal of the bulk of the bombarded americium. An ion exchange method involving the separation of the actinide elements as a group from the rare earth fission products by elution with concentrated HCl from a cation exchange resin column was exploited for the rapid separation of the tripositive actinide elements.[6] An ion exchange method involving elution from a cation exchange resin at elevated temperature with ammonium citrate solution was used in the rapid separation of the tripositive actinide elements from each other. These steps had been worked out adequately by December 1949 and the first successful experiment was done on December 19, 1949

A few milligrams of americium oxide were placed in the special target assembly indicated schematically in Fig. 37 which was designed to prevent alpha radioactivity from entering the cyclotron and to eliminate its spread to the surroundings during transportation. In this assembly the particle beam from the cyclotron was passed through two thin duralumin foils (each 1.5 mil in thickness) before entering the evacuated compartment containing the sample. The compartment was isolated from the surroundings. The beam was also passed through a thin platinum foil placed directly in contact with and over the target dish. The back of the platinum disk containing the sample was cooled directly with a water jet. The intensity of the beam striking the sample was about 2 microamperes per cm^2. After the irradiation the element 97 fraction was separated by the chemical techniques mentioned above.

Examination of the radiations in the element 97 fraction showed alpha and x-radiation decaying with a half-life of 4.6 hours. The alpha spectrum determined in an ionization chamber had three distinct groups: 6.20 Mev (17%), 6.55 Mev (53%) and 6.72 Mev (30%). Measurements of the L x-radiation in a proportional counter spectrometer showed it to be curium L x-rays which indicated that the berkelium was decaying principally by orbital-electron capture. Chemical separation and examination of americium and curium daughter activity resulted in the identification of radiations believed to be, and subsequently proved to be, those of Am^{239} and Cm^{243}. A complete discussion of the properties of Bk^{243} is given in the next section of this chapter.

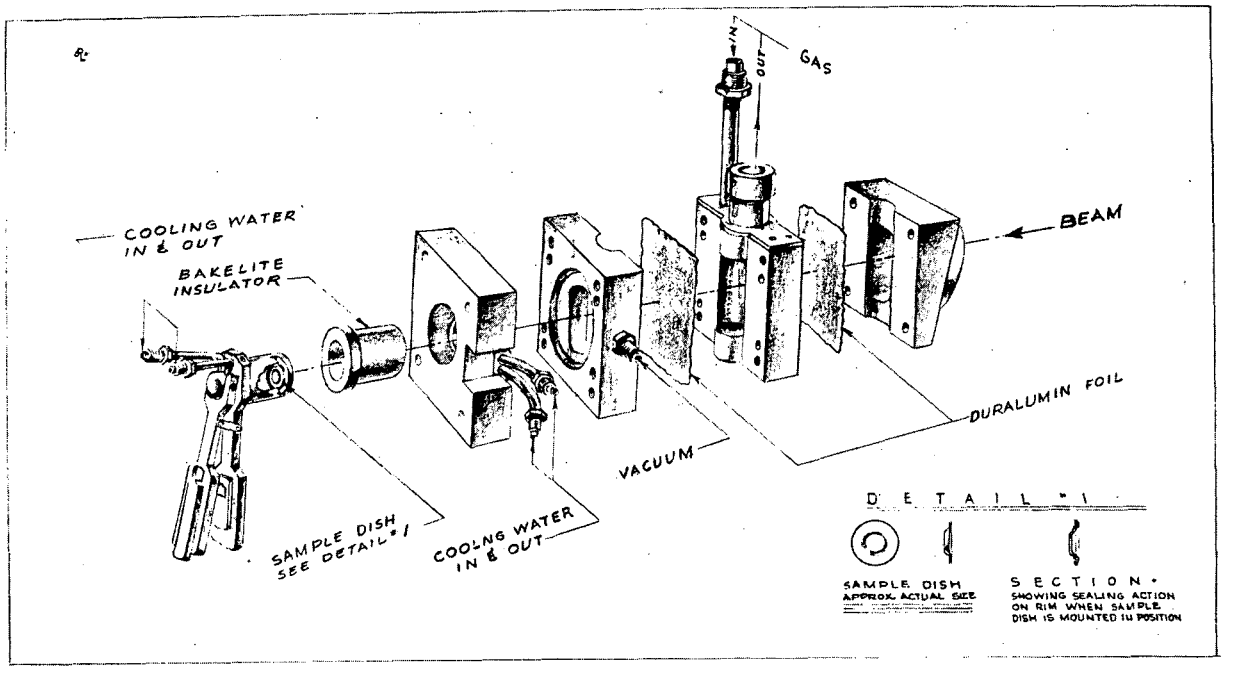

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Figure 37. Schematic sketch of the special target assembly for the bombardment of Am^{241} with helium ions in the 60-inch cyclotron. This assembly confines the radioactive target material during bombardment and during transportation to and from the cyclotron.

Additional cyclotron experiments have been carried out with targets of Am^{241}, Am^{243}, Cm^{242} and Cm^{244} which have resulted in the production and identification of Bk^{244}, Bk^{245}, Bk^{246} and Bk^{247}. The irradiation of Pu^{239} or of other materials such as Am^{241}, Am^{243} or Cm^{244} in a high flux reactor for long term neutron irradiation results ultimately in the production of the curium isotope Cm^{249} which decays by beta emission to Bk^{249}, a 290 day beta-emitter. The first preparation of isotopes of berkelium via the neutron-capture route was reported by THOMPSON, GHIORSO, HARVEY and CHOPPIN.[8] The isotope Bk^{249} is noteworthy because of its half-life and of the fact that it can be prepared in quantity; hence it is suitable for investigations of the chemical properties of this element. The first microgram-scale sample of berkelium was isolated by THOMPSON and CUNNINGHAM[7] in the sping of 1958 from 8 grams of Pu^{239} which had been irradiated in the Materials Testing Reactor for several years. With this 0.4 microgram sample, it was possible to measure several physical and chemical properties of berkelium using ultra-microchemical techniques. A consideration of all the isotopes of berkelium reveals that there are probably no beta stable isotopes of this element, although the situation is borderline for Bk^{247} and Bk^{250}.

Berkelium has the predicted properties of an eka-terbium element. In solution the most stable oxidation state is (III). In addition, however, it can be oxidized to the (IV) oxidation state. This reflects the special stability of the half-filled f electron shell which terbium achieves in the (IV) state.

The name berkelium (symbol Bk) was chosen[1] after the city of Berkeley in a manner similar to that used in naming the chemical homologue terbium (atomic number 65) whose name was derived from the town of Ytterby, Sweden where many rare earth minerals were first found.

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9.5.2 Berkelium-243. The isotope Bk^{243} can be made by the following reactions:[1-4]

$$Am^{241}\ (\alpha,2n)\ Bk^{243}$$
$$Am^{243}\ (\alpha,4n)\ Bk^{243}$$
$$Cm^{242}\ (d,n)\ Bk^{243}$$
$$Cm^{244}\ (d,3n)\ Bk^{243}$$

This isotope was the first isotope of element 97 to be reported.[1] It decays chiefly by orbital-electron capture[1] with a 4.5 hour half-life forming Cm^{243} as its daughter product. The identification of the alpha activity of the Cm^{243} daughter confirms the mass assignment.[1,3] In 0.15 percent[3] of its disintegrations Bk^{243} decays by emission[1] of alpha particles with the following energies and abundances: 6.20 Mev (17 percent), 6.55 Mev (53 percent) and 6.72 Mev (30 percent).

THOMPSON, GHIORSO and SEABORG[1] studied the L x-rays in a xenon-filled proportional counter connected to a pulse height analyzer and identified the L_α, L_β and L_γ x-rays of curium. CHETHAM-STRODE[4] has studied the gamma radiation of Bk^{243} in a scintillation spectrometer and found K and L x-rays as well as gamma rays of energy 740 ± 40 kev, 840 ± 40 kev and 960 ± 40 kev. The gamma rays have the relative abundances 1.0, 0.3 and 0.3 respectively. These gamma rays accompany the electron capture decay of Bk^{243} but the decay scheme is not known in detail. The estimated total decay energy of 1.5 Mev for the electron capture process precludes a cascade arrangement of the observed gamma rays. Careful study of the gamma rays of Bk^{243} is made difficult if Bk^{244} is also present because Bk^{244} decays with the same half life.

CHETHAM-STRODE[4] used alpha-gamma coincidence techniques to study the gamma rays accompanying the small alpha branching decay of this isotope. In addition to americium K x-rays, gamma rays of 536, 187, 146 and 42 kev were observed in coincidence with alpha particles. The abundances per alpha

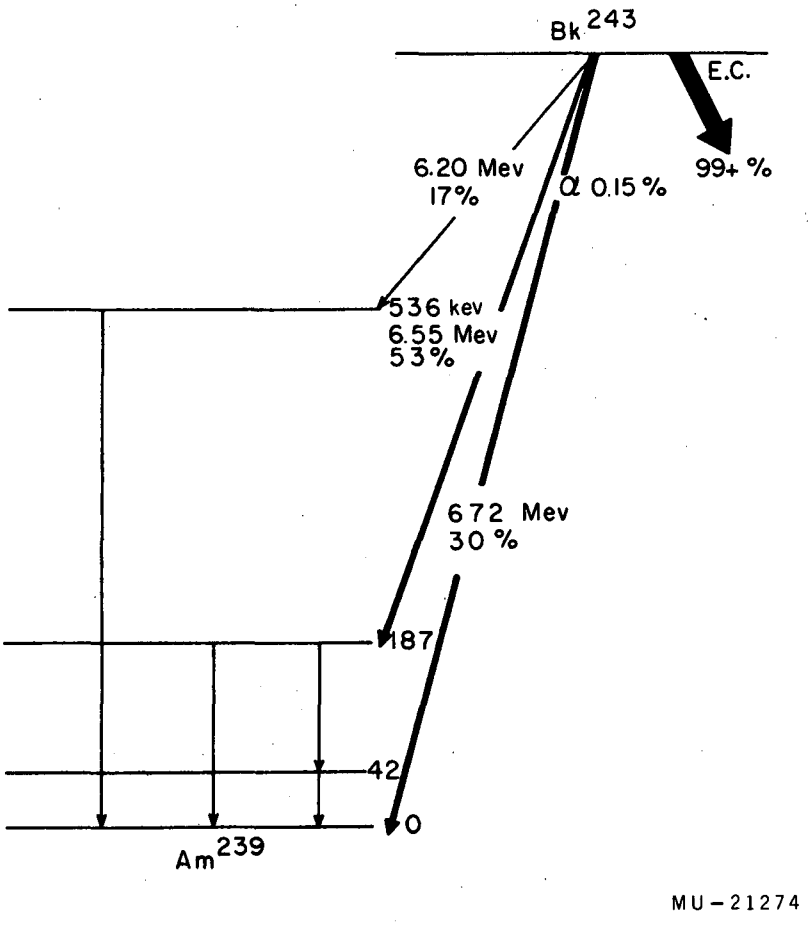

Figure 38. Alpha branching decay of Bk^{243}.

disintegration of those five radiations was found to be 0.09, 0.10, 0.34, 0.08 and 0.04 respectively.

Information on the alpha decay of Bk^{243} is summarized in Figure 38.

From a consideration of the similarities in the alpha decay patterns of Bk^{243}, Bk^{245} and Bk^{247}, STEPHENS, ASARO and PERLMAN[5] have made Nilsson orbital assignments to the ground states of the berkelium isotopes and to the ground state and excited levels of the americium daughter isotopes. The quantum numbers K, parity $[N,n_z,\Lambda]$ for Bk^{243} are 3/2- [521] while that for the ground state of Am^{239} is 5/2- [523]. The level at 187 kev in Am^{239} is assigned as 5/2+ [642] while that at 540 kev is either the 3/2 member of a rotational band based on 1/2- [530] or is the Nilsson state 3/2- [521]. The considerations on which these assignments are based are given in Chapter 3. See also the publication of NILSSON and MOTTELSON.[6]

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9.5.3 Berkelium-244. The isotope Bk^{244} decays almost entirely by orbital-electron capture with a half-life of ~4.5 hours. Since this half-life is identical with the half-life of Bk^{243} it is difficult to make meaningful measurements of its radiations unless special care is taken during the preparative reaction to minimize the production of Bk^{243}. HULET[1] obtained

indirect evidence for the production of Bk^{244} along with Bk^{243} in the bombardment of Am^{241} with helium ions. The pertinent reaction was:

$$Am^{241}\ (\alpha,n)\ Bk^{244}.$$

HULET showed that the curium fraction isolated after the decay of such berkelium samples differed from that found in the decay of samples of pure Bk^{243} made by the deuteron bombardment of Cm^{242}.

CHETHAM-STRODE[2] prepared Bk^{244} by the reaction:

$$Am^{243}\ (\alpha,3n)\ Bk^{244}.$$

By carrying out this reaction with helium ions in the range 28 to 32 Mev (see Fig. 39) the amount of Bk^{244} relative to Bk^{243} is greatly enhanced. The longer-lived isotopes, Bk^{245} and Bk^{246}, are also formed but do not interfere to the same extent as does Bk^{243}. The 5.79 Mev alpha particles of Cm^{244} were observed to grow into the purified berkelium sample which confirmed the mass assignment.

CHETHAM-STRODE[2] studied the gamma radiations of Bk^{244} in samples containing varying amounts of Bk^{243}, Bk^{245} and Bk^{246}. The most prominent gamma rays are of energy 900 and 200 kev and these are emitted in cascade. Six other gamma rays were observed with energies in the region 0.9 Mev to 1.72 Mev but in much lower intensity; these are listed in the table. These gamma rays have not been placed in a decay scheme.

Alpha particles of 6.67 ± 0.0015 Mev energy have been observed[2] for Bk^{244} in an abundance corresponding to an alpha branching of only 6×10^{-5}. This corresponds to a partial alpha half-life of 8 ± 3 years.

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Table 36. Gamma Rays of Bk^{244}

Energy (Mev)	Relative intensity
0.200	---
0.900	1.0
1.06	0.07
1.16	0.11
1.23	0.05
1.37	0.007
1.50	0.02
1.72	0.002

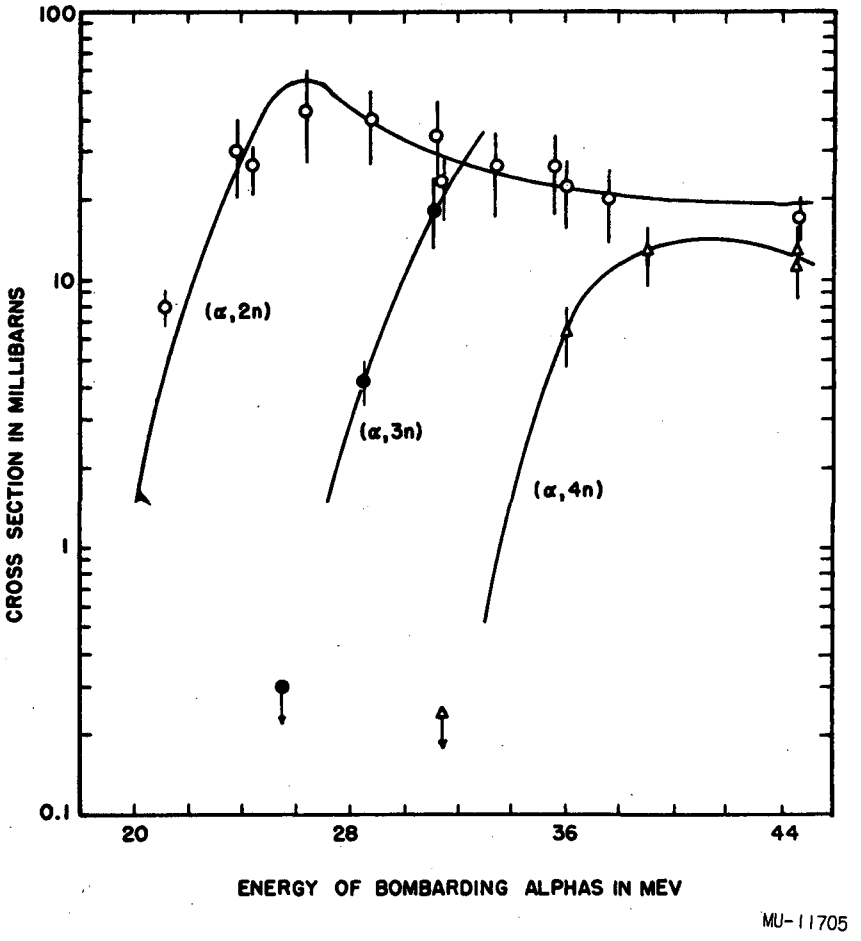

Figure 39. Excitation functions for the reactions $Am^{243}(\alpha,2n)Bk^{245}$, $Am^{243}(\alpha,3n)Bk^{244}$ and $Am^{243}(\alpha,4n)Bk^{243}$, Chetham-Strode[2].

9.5.4 Berkelium-245. HULET, THOMPSON, GHIORSO and STREET[1,2] first identified the isotope, Bk^{245}, produced in bombardments of a curium target with helium ions and with deuterons. The targets contained a mixture of Cm^{242}, Cm^{243} and Cm^{244} and the principal reactions leading to the production of Bk^{245} were:

$$Cm^{242}\ (\alpha,p)\ Bk^{245}$$
$$Cm^{244}\ (d,n)\ Bk^{245}$$

Since milligram quantities of nearly pure Am^{243} and Cm^{244} have become available. The principal reactions used in the preparation of Bk^{245} have been the following:

$$Am^{243}\ (\alpha,2n)\ Bk^{245}$$
$$Cm^{244}\ (d,n)\ Bk^{245}$$

A rough excitation function for the Am^{243} $(\alpha,2n)$ reaction is given in figure 39c accompanying Section 9.5.3.

HULET and CO-WORKERS[1,2] found that Bk^{245} decays chiefly by orbital electron capture with a half-life of 4.95 ± 0.1 days. It decays also in about 0.1 percent of its disintegrations by the emission of alpha particles distributed among three prominent groups. MAGNUSSON and CO-WORKERS[4] and CHETHAM-STRODE[3] have studied Bk^{245} in more detail. MAGNUSSON[4] gives the value 4.98 ± 0.02 days for the half-life.

The alpha spectrum as given by MAGNUSSON and CO-WORKERS[4] consists of three groups: 5.89 ± 0.02 Mev (26 percent), 6.17 ± 0.02 Mev (41 percent), and 6.37 ± 0.02 Mev (33 percent). These are shown in the decay scheme. The gamma rays following alpha decay were studied by CHETHAM-STRODE[3] by alpha-gamma coincidence techniques using scintillation crystal spectrometers. He reported gamma rays with energies 115 kev (K x-rays), 164 kev, 206 kev and 480 kev. The number of photons of these radiations per total alpha disintegration was measured to be 0.11, 0.07, 0.28 and 0.18 respectively. These transitions are shown in figure 40.

The isotope Cm^{241} also decays into Am^{241}. The decay is known to proceed to the ground state and to the 480 kev level. There may be some slight branching decay to a level at 600 kev not observed in the alpha decay of Bk^{245}. There is no conflict in the information derived from these two isotopes. The

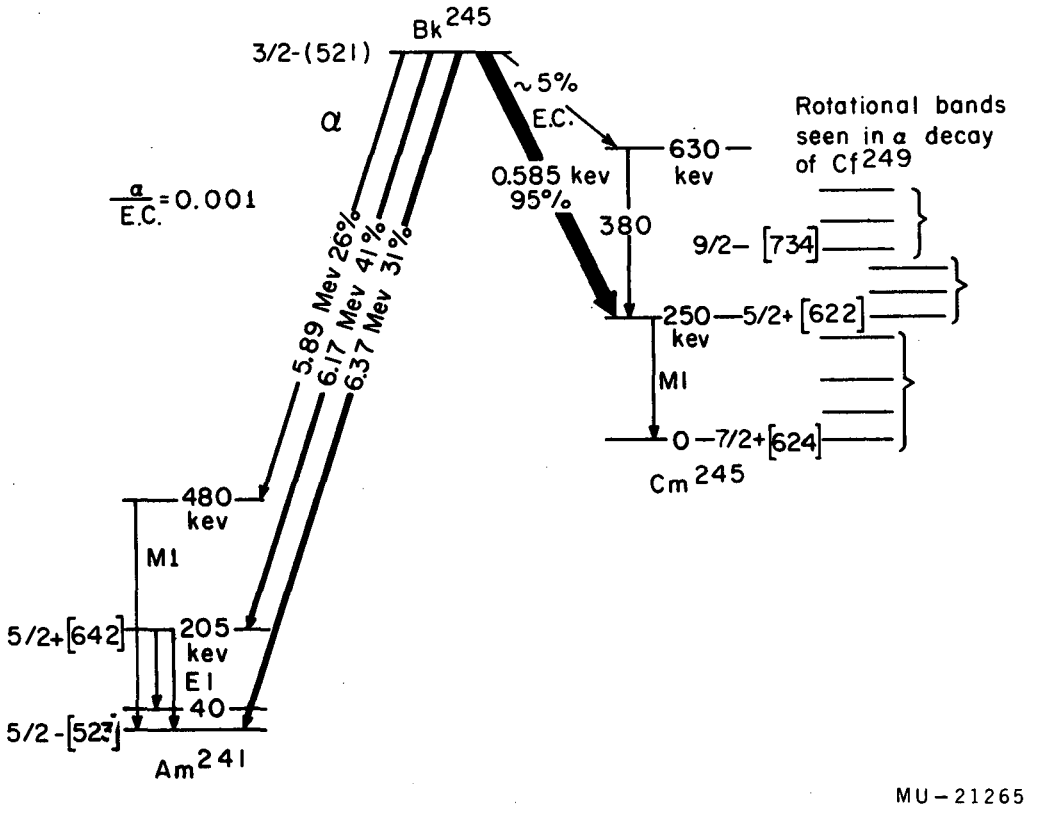

Figure 40. Decay scheme of 4.98 day Bk^{245}. Nilsson orbital assignments are given to most of the levels. The number given in front of and within the square brackets refer to the following quantum numbers: K, parity $[N\ n_z\ \Lambda]$. For further information on the Cm^{245} level structure see section 9.6.7.

beta emitter Pu^{241} also decays to Am^{241} but the decay energy is so low that only the ground state is reached.

The electromagnetic radiation accompanying electron capture has also been studied. In addition to K and L x-radiation, some photons of 250 kev and 380 kev gamma rays are observed. The conversion electrons of the 250 kev gamma rays were also measured. From the conversion coefficient the 250 kev transition is chiefly M1 in nature. The 250 and 380 kev transitions are in cascade as shown in the figure. Approximately 94% of the electron capture decay proceeds to the 250-kev level, and the rest goes to a level at 600 kev. There is no evidence for decay directly to the ground state.

The interpretation of the decay scheme of Bk^{245} and in particular the assignment of Nilsson orbitals is aided by a detailed consideration of the decay schemes of Cm^{241}, Am^{241}, Am^{245}, Cf^{249} and other nuclides. STEPHENS, ASARO and PERLMAN as well as NILSSON and MOTTELSON give detailed arguments (many of which are reproduced in Section 3.5.5 of Chapter 3) which lead to the Nilsson assignments given in the decay scheme figure.

The level system of Cm^{245} can also be studied by examining the radiations of the beta emitter Am^{245} and the alpha emitter Cf^{249}. The radiations of Am^{245} have been studied by BROWNE and CO-WORKERS[7] and by FIELDS and CO-WORKERS[8] (see Section 5.3.10) but there appears to be little correspondence with the decay scheme of Bk^{245} except for the mutual occurrence of the 250 kev gamma ray. Alpha decay of Cf^{249} proceeds to the ground state of Cm^{245} and to the levels shown at the right side of the figure. Hence, only the 250-kev level is common to the decay of Cf^{249} and Bk^{245}.

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8. P. R. Fields, M. H. Studier, A. M. Friedman, H. Diamond, R. Sjoblom and P. A. Sellers, J. Inorg. Nuclear Chem. 1, 262 (1955).

9.5.5 Berkelium-246. The 1.8 day isotope, Bk^{246}, is prepared by the reactions:[1-3]

$$Am^{243}\ (\alpha,n)\ Bk^{246}$$
$$Cm^{244}\ (\alpha,pn)\ Bk^{246}.$$

When prepared in this manner it is contaminated with 4.98 day Bk^{245} and with lighter isotopes of berkelium in amounts depending on the energy of the bombarding particles.

Berkelium-246 decays[1,2] by orbital electron capture to Cm^{246}. CHETHAM-STRODE[3] examined the gamma ray spectrum of mixtures of Bk^{245} and Bk^{246} and found a prominent 800 kev gamma ray assignable to Bk^{246} and a composite photopeak at 1.09 Mev which could be partially resolved into components with energies of 0.980, 1.08 and 1.13 Mev. Coincidence experiments indicate that the principal electron capture decay is to levels in Cm^{246} at 840 kev and higher energies. CHETHAM-STRODE[3] has constructed the tentative decay scheme shown in the figure. This scheme relies on the information given by the alpha decay of Cf^{250} for the low-lying levels of Cm^{246}. These levels constitute the rotational band of levels familiar in all even-even nuclei in this mass region. The level at 1130 kev apparently de-excites by gamma transitions to the first three members of the ground state rotational band. Further study is required before this level can be characterized as an excitation of collective vibrational motion. This is similar to the situation in the decay scheme of Bk^{250}, discussed in Section 6.5.9.

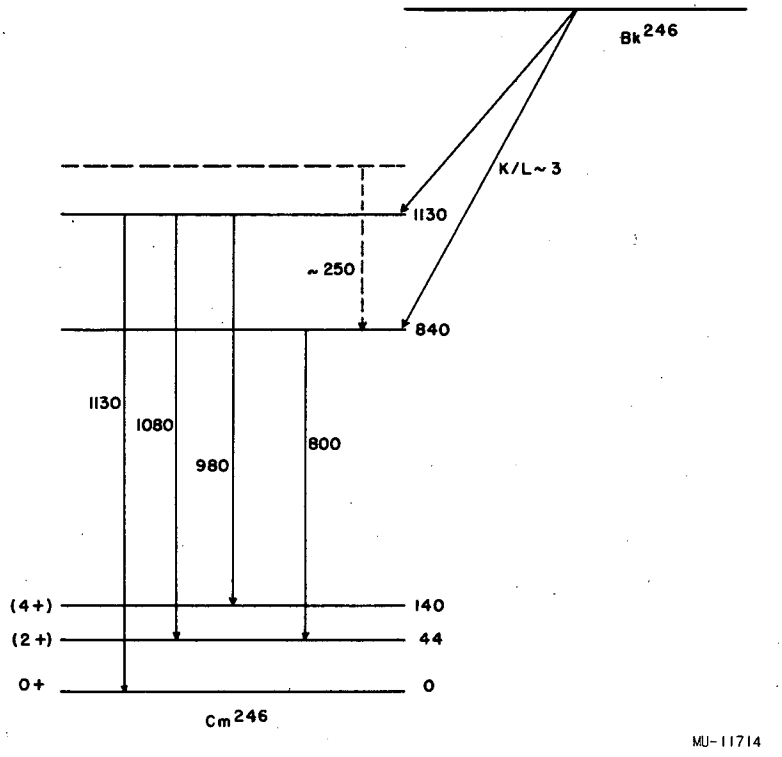

Figure 41. Tentative decay scheme for the electron capture decay of Bk^{246}. Note similarities to decay scheme of Bk^{250} shown in figure 42.

REFERENCES - Bk^{246}

1. E. K. Hulet, S. G. Thompson and A. Ghiorso, Phys. Rev. 95, 1703 (1954).
2. E. K. Hulet, Ph.D. Thesis, University of California, July, 1953; also published as University of California Radiation Laboratory Report UCRL-2283, July, 1953.
3. A. Chetham-Strode, Jr., Ph.D. Thesis, University of California, June, 1956; also published as University of California Radiation Laboratory Report UCRL-3322, June, 1956.

9.5.6 Berkelium-247. The isotope Bk^{247} has a half-life, on the basis of yields and estimated cross section for its production, of 7000 years, which makes it the most long-lived of all the isotopes of element 97. Hence the preparation by cyclotron reactions of a sample large enough for convenient measurement of its radiations requires larger amounts of target material and more extensive bombardment than does the preparation of the lighter berkelium isotopes we have just discussed. Cyclotron-induced reactions which can be used include:

$$Cm^{244}\ (\alpha,n)\ Cf^{247} \xrightarrow[2.4\ hrs]{EC} Bk^{247}$$
$$Cm^{244}\ (\alpha,p)\ Bk^{247}$$
$$Cm^{245}\ (\alpha,2n)\ Cf^{247} \longrightarrow Bk^{247}$$
$$Cm^{245}\ (\alpha,pn)\ Bk^{247}$$
$$Cm^{246}\ (\alpha,3n)\ Cf^{247} \longrightarrow Bk^{247}$$
$$Cm^{246}\ (\alpha,p2n)\ Bk^{247}$$

In a sample containing a mixture of higher mass curium isotopes all these reactions occur simultaneously.

Berkelium-247 is not produced by the intensive neutron irradiation of plutonium, americium and curium because the curium isotopes of mass numbers 242 through 248 are beta stable; it is only at Cm^{249} that a beta decay path opens up leading to the production of berkelium. This is unfortunate as it would be desirable to have a means for the preparation of microgram or larger quantities of this long-lived isotope.

Berkelium-247 can be isolated in a pure state from cyclotron targets once sufficient time has elapsed for the lighter isotopes to decay provided the curium target is principally Cm^{244}. When a chief constituent of the curium

target is Cm^{246} or higher isotopes of curium, the 280-day isotope Bk^{249} is also produced.

CHETHAM-STRODE[1] first identified Bk^{247}. He prepared small samples of Bk^{247}, and purified them thoroughly by the ion-exchange elution technique. The alpha spectrum determined by ion chamber measurements has three principal groups: 5.67 Mev (37 percent), 5.51 Mev (58 percent), and 5.30 Mev (5 percent). The gamma spectrum was studied by alpha-gamma coincidence techniques and the following electromagnetic radiation was found: K x-rays, 84 ± 3 kev and 265 ± 10 kev. The corrected abundances were 0.2, 0.4 and 0.3 per alpha decay.

The alpha decay scheme of Bk^{247} is given in a joint figure with the β^- decay of Pu^{243}. This figure appears in Section 9.2.13. See figure 20.[3]. The radiations de-exciting the 465-kev state have not been observed directly in the case of Bk^{247} because of the low intensity of the available samples. Some of the evidence for the Nilsson assignments given in the figure is discussed in the Pu^{243} summary (Section 9.2.13) but a more complete interpretation of the evidence is given elsewhere.[2,3] A lot of the interpretation is based on the great similarities in the α-decay schemes of Bk^{243}, Bk^{245} and Bk^{247}.

It is not known definitely whether Bk^{247} is beta unstable. From con-considerations of closed decay energy cycles it should be unstable by about 40 kev toward electron capture but the uncertainty in this estimate is greater than 40 kev.

REFERENCES - Bk^{247}

1. A. Chetham-Strode, Jr., Ph.D. Thesis, University of California, June, 1956; also published as University of California Radiation Laboratory Report UCRL-3322, June, 1956.
2. F. S. Stephens, F. Asaro and I. Perlman, Phys. Rev. 113, 212 (1959)
3. S. G. Nilsson and B. R. Mottelson, Mat. Fys. Skr. Dan. Vid. Selsk. Vol 1, No. 8 (1959).

9.5.7 Berkelium-248. The isotope Bk^{248} is best prepared[1] by the neutron irradiation of Bk^{247}:

$$Bk^{247}\ (n,\gamma)\ Bk^{248}.$$

It can also be prepared[2] by cyclotron reactions but the simultaneous production of other berkelium isotopes of comparable half-life intereferes with a study of its radiations. HULET[2] prepared the isotope for the first time through the use of these reactions:

$$Cm^{245}\ (\alpha,p)\ Bk^{248}$$
$$Cm^{246}\ (\alpha,pn)\ Bk^{248}$$

HULET established a half-life value of 23 ± 5 hours through radiochemical measurements of the Cf^{248} daughter activity growing into his samples.

CHETHAM-STRODE[1] determined a better half-life of 16 ± 3 hours. The decay goes partially to Cf^{248} by beta emission, and the growth of the alpha particles of this daughter activity has been observed. An alternate path of decay is by orbital electron-capture to Cm^{248}. A beta-to-electron-capture ratio of 2.4 was determined from the observed beta disintegration rate and the K and L x-ray intensities. This ratio corresponds to log ft values of 6.75 and 6.7 for the electron capture and beta decay respectively. The endpoint of the beta spectrum is at 650 ± 50 kev as determined from measurements with an anthracene crystal spectrometer.

No evidence has been reported for gamma transitions in either the electron capture or beta decay branches so it appears that the principal decay proceeds to the ground state in both branches.

REFERENCES - Bk^{248}

1. A. Chetham-Strode, Jr., Ph.D. Thesis, University of California, June, 1956; also published as University of California Radiation Laboratory Report UCRL-3322, June, 1956.
2. E. K. Hulet, Phys. Rev. 102, 182 (1956).

9.5.8 Berkelium-249. This isotope was first isolated from dust samples collected after a test explosion of the "Mike" thermonuclear reaction device in November 1952.[1] In this explosion, an extremely-high momentary flux of neutrons was generated and the uranium present in the device was converted to higher-mass uranium isotopes by the capture of many neutrons. The Bk^{249} resulted from the instantaneous production of U^{249} which then decayed through a chain of short-lived beta-emitters until Bk^{249} was reached.

This isotope has also been prepared[2,3,4] by the prolonged irradiation of samples of plutonium, americium or curium in a high-flux reactor. The course of the main neutron-capture sequence starting with Am^{243}, for example, is the following:

$$Am^{243}(n,\gamma)Am^{244} \xrightarrow{\beta^-} Cm^{244}(n,\gamma)Cm^{245}(n,\gamma)Cm^{246}(n,\gamma)Cm^{247}(n,\gamma)Cm^{248}(n,\gamma)Cm^{249} \xrightarrow{\beta^-} Bk^{249}$$

The entire neutron-capture sequence beginning with Pu^{239} is discussed in Chapter 5. The total yield of Bk^{249} in such a long sequence of steps is low but significant.

EASTWOOD and CO-WORKERS[5] report a yield of 3.2×10^{-9} grams from a 348 milligram sample of Pu^{239} irradiated to an integrated flux of 1.46×10^{22} neutrons per cm^2. THOMPSON, PARSONS and CO-WORKERS[6] isolated 0.7 micrograms of Bk^{249} from 8 grams of Pu^{239} which had been irradiated for approximately 5 years at a flux of 4-5 x 10^{14} neutrons cm^{-2} sec^{-1} in the Materials Testing Reactor, Arco, Idaho. Larger samples of berkelium can be made by a sustained program of irradiation of kilogram quantities of transuranium element samples in high flux reactors. The production of Bk^{249} by this method is important since this is the only method for the preparation of microgram and larger samples of element 97 for extensive measurements of chemical and physical properties. Berkelium-249 has a half-life very close to those of polonium and Cm^{242}, for which elements extensive macroscopic chemical investigations have proved to be possible. The isotope Bk^{247} with its half life of $\sim 10^4$ years

would be more suitable for this purpose but it cannot be made in quantity by the neutron capture method because Cm^{247} does not decay to form Bk^{247} at any appreciable rate, if at all, before it is converted to Cm^{248}.

The half-life of Bk^{249} has been reported as 290 days[4] and 314 days.[5] The endpoint energy of the beta particles is reported as 80±20 kilovolts,[4] 110±20 kilovolts,[3] 114±15 kilovolts[5] and 125±2 kilovolts.[8] The log ft value for the last-named energy is 7.1. A study of the gamma spectrum in coincidence with beta particles showed no gamma rays above a limit of about one percent of the disintegrations.[7] MAGNUSSON and CO-WORKERS[4] noted a slight alpha branching of $\sim 10^{-3}$ percent for the emission of 5.40 Mev alpha particles. EASTWOOD and CO-WORKERS[5] confirm this but report complex structure. Their figures are: total alpha-branching = $(2.2 \pm 0.3) \times 10^{-3}$ percent; alpha particle energies 5.417 ± 0.015 (96 percent) and 5.03 ± 0.03 (4 percent). This complex structure is borne out by the studies of gamma radiation in coincidence with alpha particles carried out by CHETHAM-STRODE.[7] These studies showed that a 320 kev gamma-ray was in coincidence with about 4 percent and that L x-rays were in coincidence with about 20 percent of the alpha particles. These fragmentary data are insufficient for the construction of a decay scheme.

STEPHENS, ASARO and PERLMAN assign the Nilsson state 7/2+ [633] to the ground state of Bk^{249} largely from a consideration of the alpha decay of E^{253}. The favored alpha decay of E^{253} goes to the ground state of Bk^{249} and four members of the ground state rotational band have been observed to receive alpha population. This means that the ground-state configuration (Nilsson states) of Bk^{249} and E^{253} are identical. The spacing of the band gives best agreement with a 7/2 spin.

These authors also give a ground state assignment of 9/2- [734] to Cf^{249}. This is consistent with the log ft value of 7.1 for the beta decay of Bk^{249}.

Conclusions identical to the above are reached by NILSSON and MOTTELSON.[10] These latter authors also point out that the favored alpha decay of Bk^{249} is to a level about 400 kev above ground in Am^{245} and that this level should be given the 7/2+ [633] assignment identical to the Bk^{249} ground state.

A lower limit of 1.5×10^9 years has been set[5] on the spontaneous fission half-life of Bk^{249}

REFERENCES - Bk^{249}

1. P. R. Fields, et al., Phys. Rev. 102, 180 (1956).
2. Thompson, Ghiorso, Harvey and Choppin, Phys. Rev. 93, 908 (1954).
3. H. Diamond, et al., Phys. Rev. 94, 1083 (1954).
4. L. B. Magnusson, et al., Phys. Rev. 96, 1576 (1954).
5. T. A. Eastwood, et al., Phys. Rev. 107, 1635 (1957).
6. S. G. Thompson, T. Parsons, et al., unpublished results (1958).
7. A. Chetham-Strode, Jr., Ph.D. Thesis, University of California, June, 1956; also published as University of California Radiation Laboratory Report UCRL-3322, June, 1956.
8. S. E. Vandenbosch, H. Diamond, R. K. Sjoblom and P. R. Fields, Phys. Rev. 115, 115 (1959).
9. F. S. Stephens, F. Asaro and I. Perlman, Phys. Rev. 113, 212 (1959).
10. S. G. Nilsson and B. R. Mottelson, Mat. Fys. Skr. Dan. Vid. Selsk., Vol. 1, No. 8 (1959).

9.5.9 Berkelium-250. When a sample of 290-day Bk^{249} is irradiated with neutrons the isotope Bk^{250} produced.[1]

$$Bk^{249}\ (n,\gamma)\ Bk^{250}$$

This isotope decays by beta-emission with a half-life reported originally as 3.13 hours and later as 193.3 ± 0.3 minutes.[4] Berkelium-250 can also be formed[3,5] as the alpha-decay product of 320 day E^{254}.

ASARO, STEPHENS and THOMPSON[2] studied the gamma rays of Bk^{250} in an equilibrium mixture of E^{254} plus Bk^{250}, and in a daughter fraction prepared by alpha recoil collection. Their measurements of the singles gamma spectrum and of the gamma spectrum in coincidence with beta particles and L x-rays lead to the conclusion that three gamma rays with energies 1.04 Mev, 1.02 Mev and 0.91 Mev (in low intensity) are present. Their interpretation was that these three gamma rays represented decay from a common parent level at 1.04 Mev in the daughter Cf^{250} to the three lowest levels of the ground-state rotational band. The lower members of the ground state rotational band are known from the α-decay of Fm^{254} as shown in the figure.

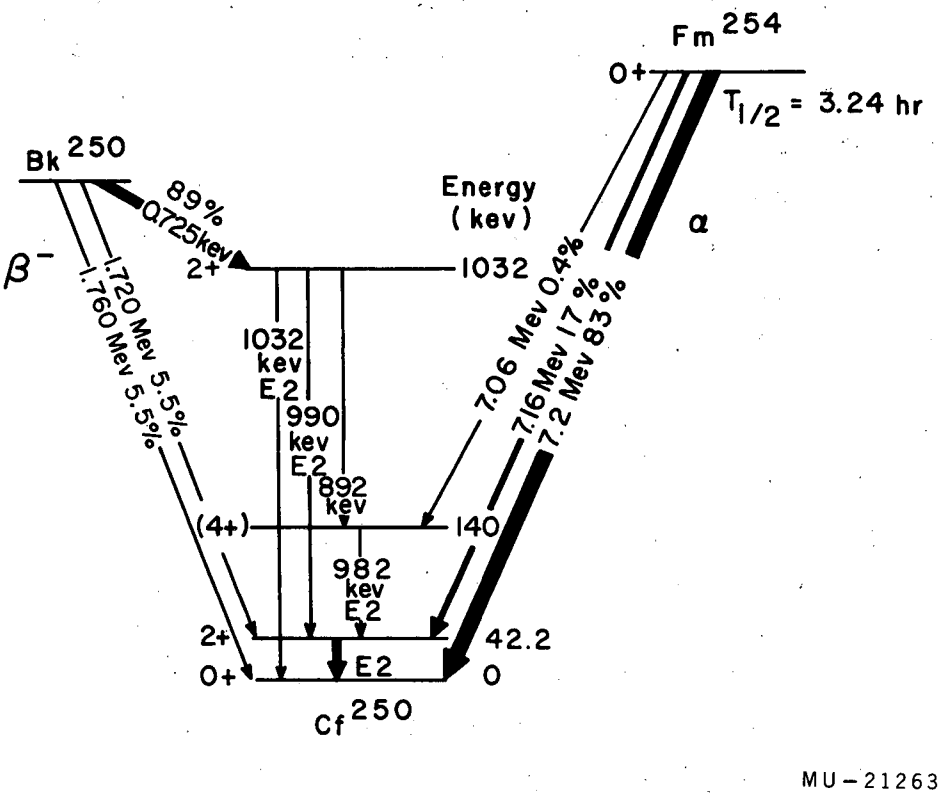

Figure 42. Decay scheme of Bk^{250} and of Fm^{254} to the common daughter product, Cf^{250}.

These results were confirmed and extended by VANDENBOSCH, DIAMOND, SJOBLOM and FIELDS[4] who made a more thorough study of the radiations of Bk^{250} in samples prepared by neutron irradiation of Bk^{249}. The beta spectrum was shown to contain two groups: 725 ± 15 kev (89 ± 1% abundant) and 1760 ± 50 kev (11 ± 1% abundant). Conversion lines corresponding to transitions with energies 42.2, 98.2, 890, 930, 990 and 1032 kev energy were measured with a double-lens magnetic spectrometer. These radiations were placed in the decay scheme as shown in the accompanying figure with the aid of quantitative $\beta\gamma$, $\gamma\gamma$ and L x-ray γ coincidence measurements. These measurements required the interpretation that the 1760 kev beta ray group observed in 11% abundance be an unresolved mixture of two beta transitions in about equal abundance going to the ground state and the first excited state of Cf^{250}.

The lowest levels of Cf^{250} consist of a rotational band of levels following the I(I+1) energy-spacing rule and the 0+, 2+, 4+ spin sequence. These levels are de-excited with a cascade of E2 gamma rays. A level at 1032 kev receives the main beta population. It is interpreted as a vibrational level of collective excitation. VANDENBOSCH, DIAMOND, SJOBLOM and FIELDS suggest at K = 2, I = 2 and parity positive assignment for this level. This level is de-excited by a triplet of gamma rays (probably E2 transitions) to the first three levels of the ground state rotational band. These authors find some evidence for a level at 1074 kev which may be the first level of rotational excitation of the 1032 kev level. This level is not shown in the figure. The log ft value for the beta transitions suggest a spin and parity of 2- for Bk^{250}.

Mass spectrometric measurements by C. M. STEVENS[6] of the upper limit of Cm^{250} produced by the electron-capture decay of Bk^{250} present in a neutron-irradiated sample of plutonium lead to a lower limit of 50 hours for the partial half-life of Bk^{250} for decay by capture of an orbital electron.

REFERENCES - Bk^{250}

1. A. Ghiorso, S. G. Thompson, G. R. Choppin and B. G. Harvey, Phys. Rev. 94, 1081 (1954).
2. F. Asaro, F. S. Stephens, Jr., and S. G. Thompson, unpublished results, 1956.
3. B. G. Harvey, S. G. Thompson, G. R. Choppin and A. Ghiorso, Phys. Rev. 99, 337 (1955).
4. S. E. Vandenbosch, H. Diamond, R. K. Sjoblom and P. R. Fields, Phys. Rev. 115, 115 (1959).
5. Jones, Schuman, Butler, Cowper, Eastwood and Jackson, Phys. Rev. 102, 203 (1956).
6. C. M. Stevens, as reported by Vandenbosch et al., in reference 4.

9.6 THE ELEMENT CALIFORNIUM (Element 98)

9.6.1 The Discovery of Californium. In section 9.5.1 we discussed how the discovery of berkelium hinged on the development of new experimental techniques and the refinement of methods for the prediction of radioactive characteristics of the isotopes to be investigated. These new experimental and theoretical methods were directly applicable to the problem of the preparation and identification of element 98. Hence after the discovery of an isotope of berkelium in December, 1949 the same group of investigators, THOMPSON, GHIORSO AND SEABORG, together with K. Street, Jr., undertook an immediate attack on the problem of element 98 and within two months were successful.

The preparative method selected by these investigators was to bombard the element curium with helium ions to prepare isotopes of element 98 by (α, xn) reactions. The only curium isotope available in sufficient quantity at that time was the 162-day isotope, Cm^{242}, which they prepared in microgram quantities by the neutron irradiation of Am^{241}. The highly radioactive curium target material ($\sim 10^{11}$ alpha disintegrations per minute) was confined in a target of the type illustrated in the figure accompanying the description of the discovery of berkeleium (section 9.5.1).

Following the bombardment of this target with 35-Mev helium ions in the Berkeley 60-inch cyclotron, the few thousands of atoms of element 98 were separated by ion exchange techniques tailored to the assumption that element 98 should be "eka-dysprosium." Element 98 was expected to have only the tripositive oxidation state in aqueous solution. Its elution position in the elution of tracer elements with buffered citric acid from a Dowex-50 cation exchange resin was estimated from the known elution behavior of gadolinium, terbium and dysprosium on the one hand and that of curium and berkelium on the other. The requirements for decontamination were severe since the few thousands of atoms of californium expected in the cyclotron bombardment had to be separated from the $\sim 10^{11}$ disintegrations per minute of Cm^{242} and from considerable amounts of fission product radioactivity. Consideration of nuclear systematics indicated that the longest-lived isotopes of element 98 to be expected in this experiment had half-lives of the order of 30 minutes to a few hours; hence the chemical separations were designed for complete separation within a period of an hour.

Using these techniques, THOMPSON, STREET, GHIORSO AND SEABORG[1] in February, 1950 produced, purified and chemically identified an isotope of element 98. This isotope had a half-life of 45 minutes. The observed mode of decay was the

emission of alpha particles with energy of about 7.1 Mev. The mass assignment was tentatively made to 98^{244} but the later work of CHETHAM-STRODE, CHOPPIN AND HARVEY[2] established that the correct mass assignment was 245.

In the selection of names for elements 95, 96 and 97 the guiding principle had been that the chosen name should emphasize the observed chemical homology to the rare earth elements 63, 64 and 65. In the case of element 98 it was difficult to suggest a suitable name stressing its relationship to the rare earth element, dysprosium (number 66). Instead, the name californium (symbol Cf) was proposed to honor the University and state of California where the work was done.

The isotopes of californium which can be produced by helium ion bombardment of Cm^{242} lie far on the neutron deficient side of beta stability and the only isotopes [1,2] to be produced with Cm^{242} as a target are 25-minute Cf^{244} and 44-minute Cf^{245}. Other heavier isotopes of californium were produced later by several types of experiments. For example, when it became possible to use cyclotron targets of curium containing appreciable percentages of Cm^{243}, Cm^{244} and higher isotopes of curium, it was possible to prepare and identify Cf^{246}, Cf^{247} and Cf^{248} by helium-ion bombardment of such mixtures.[3,4]

The development of methods for the acceleration of charged ions of carbon and nitrogen to energies above 100 Mev in moderate beam intensity made it possible to carry out reactions of the following type:[5,6]

$$U^{238}\ (C^{12},\ 4n)\ Cf^{246}$$

A number of heavier californium isotopes have been prepared by intense irradiation of lighter elements in high-flux nuclear reactors. The irradiation of Pu^{239} or of other materials such as Am^{241}, Am^{243} or Cm^{244} in a high neutron flux results ultimately in the production of the curium isotope Cm^{249} which decays by beta emission to Bk^{249}; this isotope in turn decays by beta emission to Cf^{249}. From this point, the capture of more neutrons produces Cf^{250}, Cf^{251}, and Cf^{253}.[7] (see MAGNUSSON AND CO-WORKERS[7] and GHIORSO AND CO-WORKERS[8] and also chapter 5). This is an important method of synthesis because these longer-lived isotopes can be prepared in weighable amounts and used in more detailed investigations of the chemistry of californium. The longest-lived isotopes are Cf^{249} and Cf^{251}, with a half-lives of about 400 years. The first isolation of californium in weighable quantities occurred in the summer of 1958

when THOMPSON AND CUNNINGHAM[9] isolated a few micrograms from 8 grams of plutonium which had been irradiated for several years in the highest flux of the Materials Testing Reactor. With this sample they were able to initiate a program of investigation of the bulk properties of californium using ultra-microchemical techniques.

Another method of preparation of californium which is related to the buildup of higher elements by the neutron irradiation of lower elements is observed in connection with nuclear explosions as described in section 5.4.3 of chapter 5.

One feature of the californium isotopes which is of great interest is the high rate of decay by spontaneous fission exhibited by the heavy even-even isotopes. The Cf^{250} has an alpha-decay-to-fission ratio of only 400, in Cf^{252} this ratio is only 30 and Cf^{254} decays almost entirely by spontaneous fission.

The radioactive decay properties of the individual isotopes are critically reviewed in the following sections.

References - Discovery of Californium

1. S. G. Thompson, K. Street, Jr., A. Ghiorso and G. T. Seaborg, Phys. Rev. 80, 790 (1950).
2. A. Chetham-Strode, Jr., Gregory R. Choppin and B. G. Harvey, Phys. Rev. 102, 747 (1956).
3. E. K. Hulet, S. G. Thompson, A. Ghiorso and K. Street, Jr., Phys. Rev. 84, 366 (1951).
4. E. K. Hulet, S. G. Thompson and A. Ghiorso, Phys. Rev. 95, 1702 (1954).
5. A. Ghiorso, G. B. Rossi, B. G. Harvey and S. G. Thompson, Phys. Rev. 93, 257, (1954).
6. A. Ghiorso, S. G. Thompson, K. Street, Jr., and G. T. Seaborg, Phys. Rev. 81, 154 (1951).
7. L. B. Magnusson, M. H. Studier, P. R. Fields, C. M. Stevens, J. F. Mech, A. M. Friedman, H. Diamond and J. R. Huizenga, Phys. Rev. 96, 1576 (1954).
8. A. Ghiorso, S. G. Thompson, G. R. Choppin and B. G. Harvey, Phys. Rev. 94, 1081 (1954).
9. S. G. Thompson and B. B. Cunningham, Proceedings of the Second U.N. Conference on the Peaceful Uses of Atomic Energy, Geneva 1958.

9.6.2 Californium-244. The 25 ± 3 minute isotope Cf^{244} was produced by CHETHAM-STRODE, CHOPPIN AND HARVEY[1] by the reactions:

$$Cm^{244} \quad (\alpha,4n) \quad Cf^{244}$$
$$Cm^{242} \quad (\alpha,2n) \quad Cf^{244}$$

The excitation function for the first reaction is given in figure 43. The mass assignment was confirmed by this determination of the yield as a function of the helium ion energy and by establishment of the genetic relationship of the 25 minute activity to the 26.8-day Cm^{240} daughter. Californium-244 emits alpha particles with an energy of 7.17 ± 0.01 Mev. Branching decay by electron capture is expected to be small since the decay energy available is only 0.6 Mev. Californium-244 is difficult to distinguish from Cf^{245} because of similarities in half-life and alpha particle energy, their alpha particle energies differing by only 55 kev.

It is possible to make Cf^{244} by the bombardment of uranium with carbon ions.[2]

$$U^{238} \quad (C^{12}, 6n) \quad Cf^{244}$$

References Cf^{244}

1. A. Chetham-Strode, Jr., G. R. Choppin and B. G. Harvey, Phys. Rev. 102, 747 (1956).
2. T. Sikkeland, A. Ghiorso and S. G. Thompson, unpublished results; see abstract in Bull. Am. Phys. Soc. Series II 2, No. 8, 385 (1957); see University of California Radiation Laboratory Report UCRL-8142 (1958).

9.6.3 Californium-245. The isotope Cf^{245} was first prepared and studied by THOMPSON, STREET, GHIORSO AND SEABORG[1] in the experiments which constituted the discovery of element 98. The original mass number assignment was 244 but later work by CHETHAM-STRODE, CHOPPIN AND HARVEY[2] based largely on the excitation function for the activity when Cm^{242} and Cm^{244} targets were bombarded with helium ions, made certain that the correct assignment was 245. These preparative reactions are:

$$Cm^{242} \quad (\alpha,n) \quad Cf^{245}$$
$$Cm^{244} \quad (\alpha,3n) \quad Cf^{245}$$

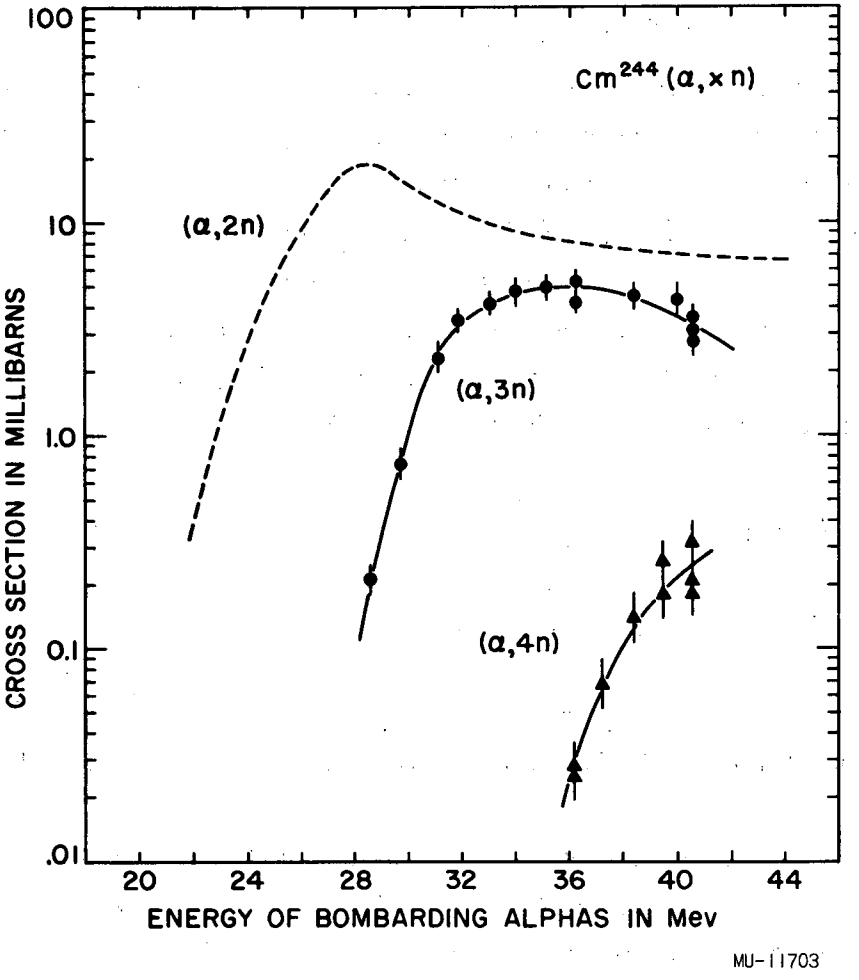

Figure 43. Excitation functions for the reactions $Cm^{244}(\alpha,xn)$ as given by Chetham-Strode, Choppin, and Harvey.[1] Dashed line: $(\alpha,2n)$ as determined from Cf^{246} alpha activity. Circles: $(\alpha,3n)$ as determined from Cf^{243} alpha activity. Triangles: $(\alpha,4n)$ as determined from alpha activity of Cm^{240} daughter.

The excitation function for the second reaction is given in the figure. Californium-245 decays by emission of 7.11 ± 0.01 Mev alpha particles with a half-life of 44 minutes. Alpha-gamma coincidence experiments indicated no lower energy alpha group in greater than 2 percent abundance. The Cf^{245} decays by orbital electron capture to Bk^{245} in 70 percent of its disintegrations and the berkelium daughter has been isolated and identified.[2]

This isotope can also be made by the bombardment of uranium with carbon ions or by other reactions involving heavy ions and heavy element targets:[3,4]

$$U^{238}\ (C^{12},5n)\ Cf^{245}$$

References Cf^{245}

1. S. G. Thompson, K. Street, Jr., A. Ghiorso and G. T. Seaborg, Phys. Rev. 80, 790 (1950).
2. A. Chetham-Strode, Jr., G. R. Choppin and B. G. Harvey, Phys. Rev. 102, 747, (1956).
3. A. Ghiorso, S. G. Thompson, K. Street, Jr., and G. T. Seaborg, Phys. Rev. 81, 154 (1951).
4. A. Ghiorso, G. Bernard Rossi, Bernard G. Harvey and S. G. Thompson, Phys. Rev. 93, 257 (1954).

9.6.4 Californium-246. GHIORSO, THOMPSON, STREET AND SEABORG[1] produced Cf^{246} by the bombardment of natural uranium with carbon ions accelerated above 100 Mev in the Berkeley 60-inch cyclotron:

$$_{92}U^{238} + {}_{6}C^{12} \longrightarrow {}_{98}Cf^{246} + 4\,{}_{0}n^{1}.$$

SIKKELAND, GHIORSO AND THOMPSON[8] later studied the excitation function for this reaction using monoenergetic carbon ions from a linear accelerator. They found a peak yield of a few microbarns when the beam energy was about 63 Mev. HULET AND CO-WORKERS[2] prepared Cf^{246} by the more conventional reactions:

$$Cm^{243}\ (\alpha,n)\ Cf^{246}$$
$$Cm^{244}\ (\alpha,2n)\ Cf^{246}.$$

CHETHAM-STRODE, CHOPPIN AND HARVEY[3] observed the excitation function shown in section 9.6.3 for the latter reaction. The radioactivity of the californium

fraction isolated from curium targets bombarded with helium ions is nearly entirely due to Cf^{246} once the shorter-lived Cf^{244} and Cf^{245} have decayed. The isotope Cf^{246} decays with a half-life of 35.7 hours.[1,2] The alpha spectrum as measured in a magnetic spectrograph indicates groups at 6.753 and 6.711 Mev with intensities of 78 percent and 22 percent respectively.[4,5] Alpha-gamma coincidence experiments [4,5,7] revealed gamma rays of 42, 98 and 151 kev energy. The alpha decay of the even-even nucleus Cf^{246} populates a rotational band of levels in Cm^{242}. The experimental data lead to the decay scheme shown in the figure which closely resembles that of all other even-even alpha emitters in the transuranium region. The energy of the first excited state of Cm^{242} is known to be exactly 42.12±kev from studies of the beta decay of Am^{242m}. Measurements of HULET, THOMPSON AND GHIORSO[6] indicate a half-life for spontaneous fission of 2100 ± 300 years.

References Cf^{246}

1. A. Ghiorso, S. G. Thompson, K. Street, Jr., and G. T. Seaborg, Phys. Rev. 81, 154 (1951).
2. E. K. Hulet, S. G. Thompson, A. Ghiorso and K. Street, Jr., Phys. Rev. 84, 366 (1951).
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5. J. P. Hummel, Ph. D. Thesis, University of California, September 1956; also published as University of California Radiation Laboratory Report, UCRL-3456, July 1956.
6. E. K. Hulet, S. G. Thompson and A. Ghiorso, Phys. Rev. 89, 878 (1953).
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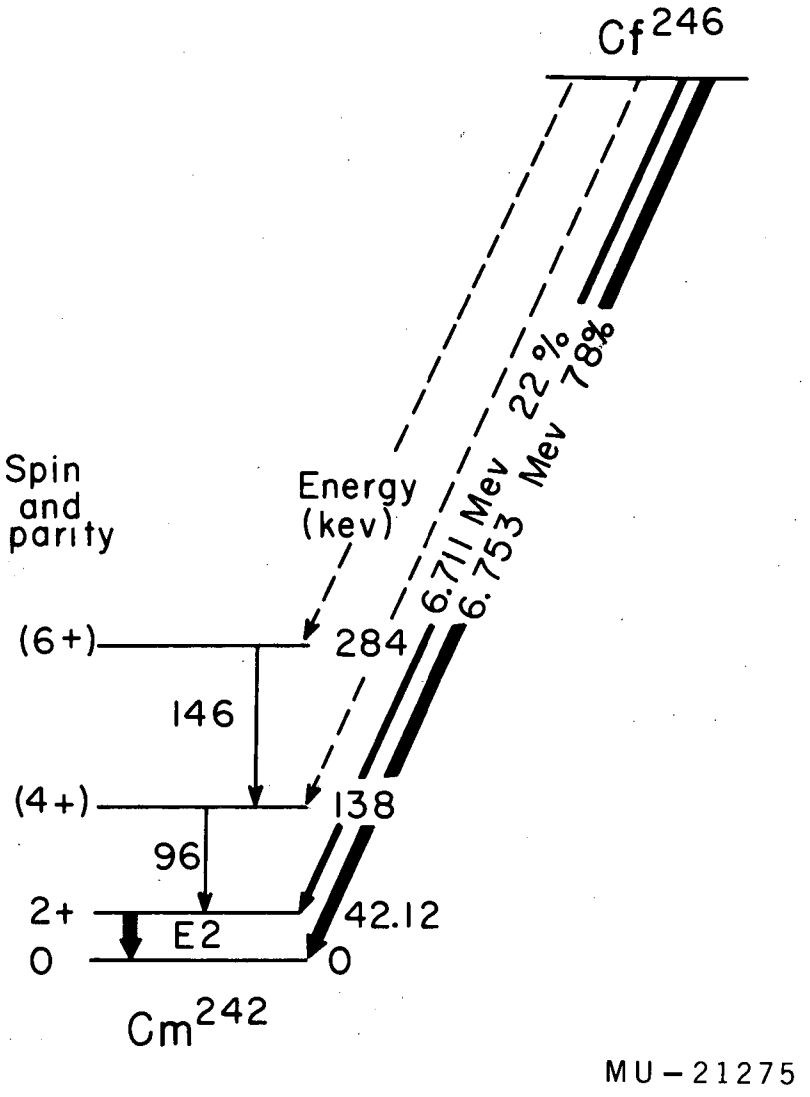


Figure 44. Decay scheme of Cf^{246}.

9.6.5 Californium-247. The 2.5-hour Cf^{247} was first identified by HULET, THOMPSON AND GHIORSO[1] who prepared it by bombardment of Cm^{244}:

$$Cm^{244}\ (\alpha,n)\ Cf^{247}.$$

It has also been produced, for example, by bombardment of uranium with energetic nitrogen ions[2]:

$$_{92}U^{238} + {}_{7}N^{14} \longrightarrow {}_{98}Cf^{247} + 4\,{}_{0}n^{1} + {}_{1}p^{1}$$

These methods of preparation result in samples containing Cf^{246} as well, which interferes somewhat with the study of the radiations of Cf^{247}.

CHETHAM-STRODE[3] has reported a half life of 2.45 ± 0.15 hours for the decay of this isotope by orbital electron capture. Examination of the electromagnetic radiations revealed K and L x-rays plus gamma rays with energies of 295 ± 15 kev, 417 ± 8 kev and 460 ± 10 kev. Gamma-gamma coincidence experiments show that the 295-kev gamma ray decays directly to the ground state and that the 417 and 460 kev gamma rays originate at a common 460 kev level in the daughter nucleus. Approximately 90 percent of the electron capture transitions lead directly to the ground state of Bk^{247}. Approximately 4 percent lead to the 295 kev level and approximately 2 percent lead to the 460 kev level.

References Cf^{247}

1. E. K. Hulet, S. G. Thompson and A. Ghiorso, Phys. Rev. 95, 1703 (1954).
2. A. Ghiorso, G. B. Rossi, B. G. Harvey and S. G. Thompson, Phys. Rev. 93, 257 (1954).
3. A. Chetham-Strode, Jr., Ph. D. Thesis, University of California, September 1956; also published as University of California Radiation Laboratory Report, UCRL-3322, June 26, 1956.

9.6.6 Californium-248. Bombardment of curium containing appreciable amounts of Cm^{245} and/or higher mass curium isotopes gives rise to Cf^{248} via one or more of the following reactions:

$$Cm^{245}\ (\alpha,n)\ Cf^{248}$$
$$Cm^{246}\ (\alpha,2n)\ Cf^{248}$$
$$Cm^{247}\ (\alpha,3n)\ Cf^{248}$$
$$Cm^{248}\ (\alpha,4n)\ Cf^{248}$$

HULET, THOMPSON AND GHIORSO[1] first produced the isotope by the first of these reactions and proved the mass assignment by observing the growth of the 5.80 Mev alpha particle of Cm^{244}.

This isotope is not produced by the intense neutron irradiation of lighter elements either on a fast time scale (nuclear explosions) or on a slower time scale (nuclear reactors) because its formation is blocked by the beta stability of Cm^{248}. It can be produced by bombardment of lighter elements with heavy ions as in the example written below.[2]

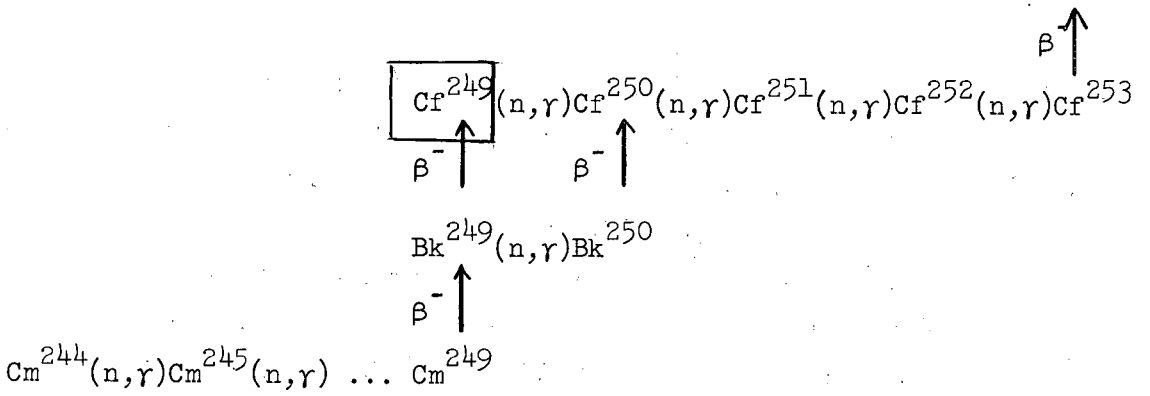
$$_{92}U^{238} + {}_{7}N^{14} \longrightarrow {}_{98}Cf^{248} + {}_{1}p^{1} + 3\,{}_{0}n^{1} .$$

The isotope, Cf^{248} decays by the emission of alpha particles with a half-life of 350 days,[3] the main group of alpha particles having an energy of 6.26 ± 0.03 Mev. HULET[3] has found that the main alpha-group abundance is approximately 82 percent while a second group in about 18 percent abundance has an energy 45 kilovolts lower in energy. This result conforms to the pattern of decay shown by all the neighboring even-even alpha-emitters. A measurement of the spontaneous fission half-life gave a lower limit of 15,000 years.[3] Small amounts of Cf^{248} have been isolated as the daughter products of Bk^{248}, E^{248} and Fm^{252} decay. Cf^{248} is beta stable. The half-life for spontaneous fission is greater than 15,000 years.

References Cf^{248}

1. E. K. Hulet, S. G. Thompson and A. Ghiorso, Phys. Rev. 95, 1703 (1954); E. K. Hulet, Ph. D. Thesis, University of California, August 1953; also published as University of California Radiation Laboratory Report, UCRL-2283, August 1953.
2. A. Ghiorso, G. Bernard Rossi, B. G. Harvey and S. G. Thompson, Phys. Rev. 93, 257 (1954).
3. E. K. Hulet, unpublished results, 1956-57.

9.6.7 Californium-249. The beta-decay of 314-day Bk^{249} has been observed[1,2,3] to give rise to the long-lived alpha-emitter Cf^{249}. This isotope of californium can be produced by the intense and prolonged irradiation of uranium, plutonium, americium or curium starting with neutrons. The neutron-capture sequence proceeds through the isotopes of curium until mass 249 is reached; Cm^{249} then decays to Bk^{249} which decays to Cf^{249}; Cf^{249} in turn captures neutrons to give rise to several heavier isotopes of californium. The pertinent sequence is the following:



The isotopic composition of the californium samples prepared by intense neutron irradiation of Pu^{239} has been determined mass spectrometrically by MAGNUSSON and CO-WORKERS.[3] Typical results are shown in Table 37.

In order to obtain pure samples of Cf^{249} it is necessary to isolate and purify the berkelium fraction from such neutron-irradiated samples and at a later date remove the daughter Cf^{249} activity. The Cf^{249} was observed for the first time in the debris of a thermonuclear test explosion.[4]

Two early determinations of the half-life of Cf^{249} resulted in the values of ~400 years[1] and 470 ± 100 years.[3] A more recent result is 360 ± 40 years.[5] All three values were determined by measuring the growth of Cf^{249} daughter activity into a known amount of Bk^{249}. The half-life for spontaneous fission has been reported as 1.5×10^9 years[1] and as $\geq 4.5 \times 10^8$ years.[5]

The alpha spectrum of Cf^{249} has been studied by MAGNUSSON[6] and by ASARO, STEPHENS and PERLMAN.[7,8] We quote the results of the latter group[8] in Table 38. This spectrum was measured in a magnetic spectrometer with special shaping of the field to accomplish double-focusing with the purpose of achieving higher transmission. The alpha groups are also indicated in the decay scheme shown in the accompanying figure.

Table 37

Isotopic Composition of Californium Fraction Isolated from Pu^{239} Sample Irradiated to an Integrated Flux of 1.14×10^{22} neutrons/cm^2. (reference 3)

Isotope	Abundance - Mole(%)
Cf^{249}	4.3 ± 0.5
Cf^{250}	49 ± 6
Cf^{251}	11 ± 3
Cf^{252}	36 ± 5

Table 38

Alpha Spectrum of Cf^{249}

Energy of Cm^{245} daughter level (kev)	Alpha intensity (percent)
0	1.9
56	1.1
124	0.4
207(?)	0.08
257	3.3
301	3.0
357	1.2
394	83.7
422(?)	0.5
450	4.4
511	0.4

The energy of the intense alpha group leading to the 394 kev level was determined to be 5.806 Mev relative to a Cm^{244} standard taken as 5.801 Mev. The energies of the other levels of Cm^{245} were determined by a joint consideration of the experimental gamma-ray and alpha-ray spectra.

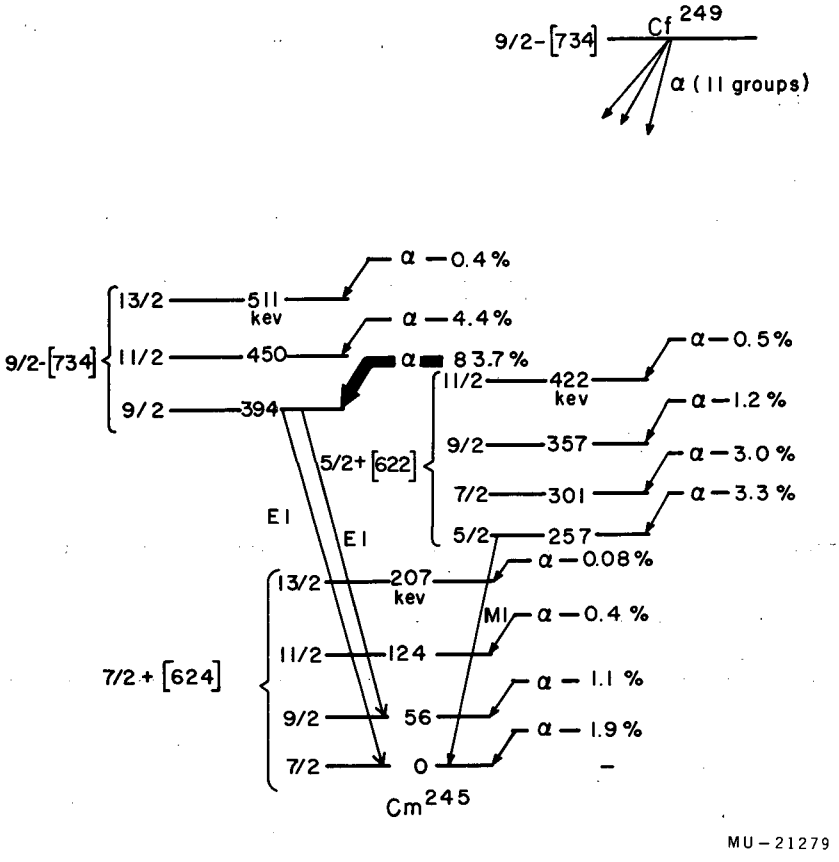


Figure 45. Decay scheme of Cf^{249} drawn in a way to emphasize the rotational band interpretation of the observed levels of Cm^{245}. The Nilsson level quantum numbers are given to the left of each set of levels: K, parity $[N\ n_z\ \Lambda]$. The energy of each level is given in kev in the center of the line representing that level.

Three gamma rays have been reported:[8] 255 kev (3 percent), 340 kev (15 percent) and 394 kev (72 percent). The 255 kev gamma ray has been seen also from Bk^{245} electron-capture decay[9,10] (see section 9.5.4) and from Am^{245} beta decay.[11-13] It has clearly been identified as an M1 transition. Conversion coefficient arguments as well as some angular correlation data indicate that the 340 and 394 kev transitions are probably E1. A rather satisfactory analysis of all the data in terms of three rotational bands has been given[6-8] and is reproduced in the decay scheme shown here. The principal data lacking are conversion electron spectra, which are difficult to obtain with the limited amounts of Cf^{249} usually available. In the following paragraphs we review briefly the properties of the three rotational bands, the choice of Nilsson states and finally the interpretation of the alpha decay intensity pattern.

According to the Nilsson diagram of neutron states the ground state of Cm^{245} with 149 neutrons should be 7/2+ [624] or 9/2- [734]. Four members of the ground state rotational band are known at 0, 56, 124 and 207 kev. These energies fit the rotational formula,

$$E_I = E_o + \frac{\hbar^2}{2\Im} [I(I+1) - I_o(I_o+1)],$$

quite well if we set $E_o = 0$, $\frac{\hbar^2}{2\Im} = 6.2$ kev and $I_o = 7/2$. Hence the indicated ground state assignment is 7/2+ [624].

The levels at 257, 301, 357 and 422 are also interpreted as a rotational band. These energies agree well with by the above rotational formula if $E_o = 257$ kev, $\frac{\hbar^2}{2\Im} = 6.3$ kev and $I_o = 5/2$. The Nilsson assignment is 5/2+ [622]. This level is the ground state for Pu^{241} and Cm^{243}, both of which have 147 neutrons, and its reappearance in Cm^{245} represents the opening of a closed shell. A spin assignment of 5/2 to the 257 kev level is also supported by the following evidence.

Americium-245 has spin 5/2 and in its decay to Cm^{245} populates both the ground state and the level at 257 kev. Berkelium-245, which presumably has spin 3/2, does not populate the ground state of Cm^{245} but only the 257 kev level. This 257 kev level decays to the ground state with an M1 transition and does not decay to the higher spin members of the ground state rotational band. (See discussion of Bk^{245} in section 9.5.4). These data , taken together with

rotational band spacings, strongly support the 5/2+ assignment to the 257 kev level. The 5/2+ [622] Nilsson assignment seems almost certain.

The favored alpha decay of Cf^{249} populates a rotational band whose base level is 394 kev above the ground state of Cm^{245}. According to the favored alpha decay concept introduced by BOHR, FROMAN and MOTTELSON[14]. this means that the Nilsson assignment of the 394 kev level in Cm^{245} and the ground state of Cf^{249} are identical. The 394 kev level decays by E1 transitions to the 7/2 and 9/2 members of the ground state rotational band with no detectable branching to the 5/2+ [622] band at 255 kev. No branching to this 394 kev level was observed in the decay of either Am^{245} or Bk^{245} (spins 5/3 and 3/2). The conclusion from these facts is that the 394 kev level has spin and parity 7/2- or 9/2- with 9/2- more likely. Alpha-gamma correlation data[8] favor the 9/2- choice. Three members of a rotational band based on the 394 kev level appear at 394, 450 and 511 kev. These energy values are consistent with a 9/2 assignment to the base state if E_o = 394 kev and $\frac{\hbar^2}{2\Im}$ = 5.1 kev are substituted in the rotational formula given above. The most likely Nilsson choice for a 9/2+ state in a nucleus with 149 or 151 neutrons is 9/2- [734] and this assignment is made to the 394 kev state in Cm^{245} and the ground state of Cf^{249}.

The appreciably lower value of $\frac{\hbar^2}{2\Im}$ for the 9/2- [734] band compared to the other two bands can be explained by invoking the coriolis interaction which is larger for this band because it comes from the $j_{15/2}$ shell model orbital. RASMUSSEN[15] has pointed out that the rotational bands based on Nilsson states derived from shell-model orbitals of high j will have larger coriolis interactions and hence will be more compressed than other bands. The 9/2- [734] band is a good example of this situtation.

We now turn to a discussion of the alpha intensity patterns. We consider first the alpha decay to the four members of the ground state rotational band of Cm^{245}. This decay involves a change of parity or, equivalently the emission of alpha particle waves of odd angular momentum. ASARO, STEPHENS and PERLMAN[8] analyzed this decay according to the method outlined by BOHR, FROMAN and MOTTELSON[14] and determined that the alpha waves going to the ground state band are a mixture of 25 percent $\ell = 1$, 67 percent $\ell = 3$ and 8 percent $\ell = 5$. Such an analysis assumes that there is no interference between the various alpha waves. This mixture reproduces, well within the experimental error, the

four alpha intensities to the band members. The hindrance factors (reciprocal of the reduced alpha transition probability) for the ℓ = 1,3 and 5 waves are 3.1×10^4, 6.9×10^3 and 2.2×10^4, respectively. The analysis into orbital angular waves is considered to be essentially correct but the various wave hindrances are not understood. This situation holds true generally for "unfavored" or "hindered" alpha groups.

The alpha decay which populates the four levels of the rotational band whose base level lies 257 kev above the ground state has been analyzed similarly into angular momentum components. In this case the alpha waves are 87 percent ℓ = 3 and 13 percent ℓ = 5. The corresponding hindrance factors are 150 and 350. It is not known why the wave hindrance factors are so much lower than for the ground state/band.

The favored alpha decay to the 9/2- [734] band has a composition 80 percent ℓ = 0 and 20 percent ℓ = 2 with corresponding hindrance factors of 4.5 and 14. The ℓ = 0 wave can of course only populate the 9/2- level itself, while the ℓ = 2 can populate the 11/2 and 13/2 levels as well. It is perhaps worthwhile to point out again that this "favored" alpha decay is believed to be much like the ground-state-to-ground-state alpha transitions in even-even nuclei.

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1. A. Ghiorso, S. G. Thompson, G. R. Choppin and B. G. Harvey, Phys. Rev. 94, 1081 (1954).
2. H. Diamond, L. B. Magnusson, J. F. Mech, C. M. Stevens, A. M. Friedman, M. H. Studier, P. R. Fields and J. R. Huizenga, Phys. Rev. 94, 1083 (1954).
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11. C. I. Browne, et al., J. Inorg. Nuclear Chem. 1, 254 (1955).
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9.6.8 Californium-250. In the previous section we reported how Cf^{249} is produced by the build up of lighter isotopes by neutron capture reactions. The isotope Cf^{250} is produced in the same way when the irradiation proceeds for a sufficiently long time. In the table printed in the previous section it is shown that a californium fraction containing 49 mole percent Cf^{250} has been prepared[1] by neutron irradiation of Pu^{239}. When the neutron buildup occurs on a very fast time scale, as in thermonuclear explosions,[2] Cf^{250} is not produced because of the beta stability or long β^- half-life of Cm^{250}. It is also possible to prepare Cf^{250} by the following reactions:

$$Bk^{249}\ (d,n)\ Cf^{250}$$

$$Bk^{249}\ (n,\gamma)\ Bk^{250} \xrightarrow[3.1\ \text{hours}]{\beta^-} Cf^{250}$$

Still another method is bombardment of a sample of curium containing appreciable amounts of Cm^{247} or higher-mass isotopes with helium ions:

$$Cm^{247}\ (\alpha,n)\ Cf^{250}$$

$$Cm^{248}\ (\alpha,2n)\ Cf^{250}.$$

GHIORSO AND CO-WORKERS[3] observed that chemically pure samples of 3.13-hour Bk^{250} decayed to a daughter product which emits 6.05-Mev alpha particles. The number of alpha particles corresponded to a half-life of about 12 years for the Cf^{250}. The ratio of the rate of alpha decay to the rate of spontaneous fission was set at 400 from which the spontaneous fission half-life of Cf^{250} was estimated as about 5000 years.

MAGNUSSON AND CO-WORKERS[1] used data from the mass spectrographic analysis and alpha spectrum analysis of a sample of californium containing 49 isotope percent Cf^{250} to calculate a half-life of 10.0 ± 2.4 years. The alpha to spontaneous fission rate was set at 1460 ± 350 leading to a spontaneous fission half-life of $1.5 \pm 0.5 \times 10^4$ years. These workers measured[1] the alpha decay energy of the main alpha group to be 6.031 Mev. By alpha-gamma coincidence studies it was shown that an alpha group of approximately 40 kev lower energy (Ca 5.99 Mev) was in coincidence with L x-rays (from the conversion of a gamma ray of 40 kev). EASTWOOD AND CO-WORKERS[4] analyzed a somewhat similar sample of mixed californium isotopes and found for Cf^{250} a half-life of 10.9 ± 0.8 years and an alpha particle energy for the main group of 6.020 ± 0.010 Mev.

ASARO, STEPHENS, HARVEY AND PERLMAN[5] studied a mixed sample of californium in which the alpha radioactivity was 85% due to Cf^{252} and 15% due to Cf^{250}. Measurements in a magnetic alpha spectrograph showed a main alpha group for Cf^{250} of 6.024 Mev (83 percent abundance) and a smaller group at 5.980 Mev (17 percent abundance). Alpha-gamma coincidence experiments showed that L x-rays were in coincidence with alpha particles. From their interpretation of their quantitative measurements the 44 kev transition from the first excited state in the Cm^{246} daughter is an E2 transition; the L x-rays arise from its nearly complete conversion. HARVEY AND HOLLANDER[6] have measured the L_{II} and L_{III} conversion electrons of this transition and report an energy value of 42.9 ± 0.1 kev. The decay scheme for Cf^{250} is shown in the figure. ASARO AND CO-WORKERS[5] point out that a lower-energy alpha particle group leading to a 4 + level at ~145 kev is to be expected, but in abundance too low (~ .01 percent) for detection in their experiments. A detailed study of the gamma radiations of Cf^{250} in a mixed source of this type is somewhat hindered by the gamma radiations of other californium isotopes present and by an appreciable amount of gamma activity arising from the spontaneous fission of Cf^{252}. Cf^{252} is stable toward beta decay processes.

References Cf^{250}

1. L. B. Magnusson, M. H. Studier, P. R. Fields, C. M. Stevens, J. F. Mech, A. M. Friedman, H. Diamond, and J. R. Huizenga, Phys. Rev. 96, 1576 (1954).
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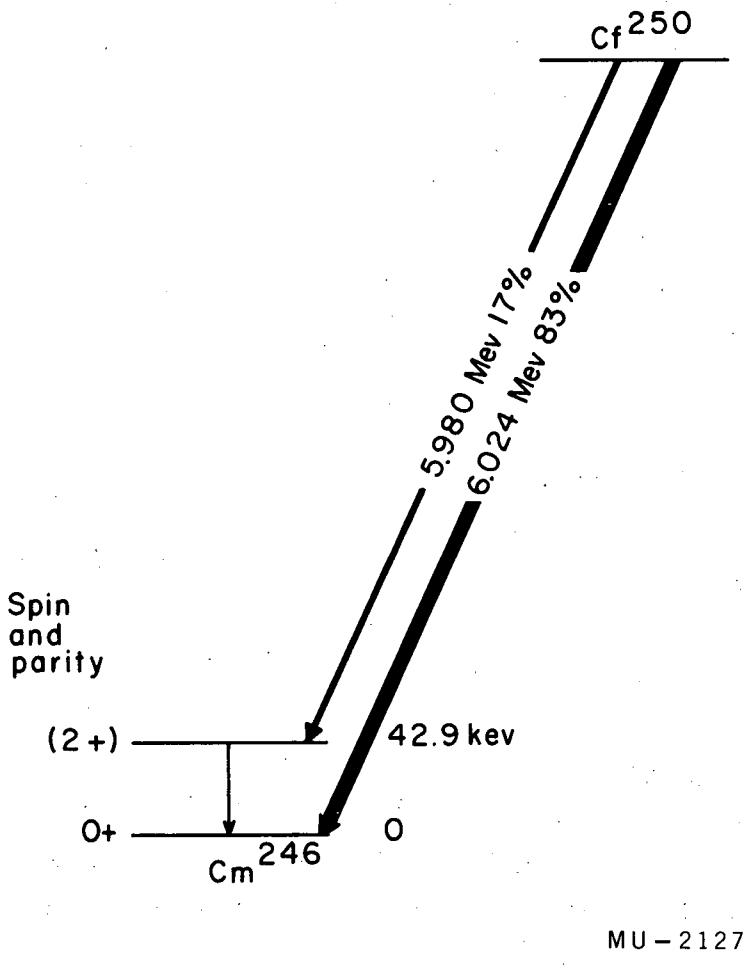

Figure 46. Decay scheme of Cf^{250}.

9.6.9 Californium-251. The isotope Cf^{251} has been identified mass-spectrometrically in the californium fraction isolated from neutron irradiated plutonium.[1,2,3] See for example the analysis of one sample given in the table accompanying the write-up of Cf^{249}. Very little additional information is available on this isotope. From the systematics of alpha decay it is expected to be beta stable. The half-lives for alpha decay and spontaneous fission are so long compared to the neighboring even-even californium isotopes present in all samples so far investigated that no radiations directly attributable to it have been observed. A half-life of approximately 800 years was estimated[3] by mass spectrographic measurements of the ratio Cm^{246}/Cm^{247} in the daughter curium fraction separated from a californium fraction containing Cf^{250} and Cf^{251} in the ratio 7/2.

References Cf^{251}

1. H. Diamond, L. B. Magnusson, J. F. Mech, C. M. Stevens, A. M. Friedman, M. H. Studier, P. R. Fields and J. R. Huizenga, Phys. Rev. 94, 1083, (1954).
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3. T. A. Eastwood et al., Phys. Rev. 107, 1635 (1957).

9.6.10 Californium-252. The 2.2 year isotope, Cf^{252}, is prepared by intense neutron irradiation of Pu^{239} [1-3] or of other heavy element materials and has been identified mass spectrometrically in californium fractions isolated from such irradiated samples.[2,3,8] It can also be prepared by multiple neutron capture on a fast time scale (i.e., lifetimes for neutron capture small compared to beta decay lifetimes) such as occurs in connection with thermonuclear reactions; it was in fact first identified in samples taken from the "mike" thermonuclear test explosion.[4]

The production of Cf^{252} in high-flux reactors is a matter of considerable scientific importance and special reactors have been proposed for the production of Cf^{252} and other transplutonium nuclides. Refer to discussion in chapter 5.

Cf^{252} decays by alpha emission and by spontaneous fission. It is beta stable. The ratio of alpha decay to spontaneous fission is only 30. The best method of measurement of the alpha decay half-life is to follow the decay of

the spontaneous fission rate, since the observed rate is controlled by the rate of alpha decay. One group[3] reports an alpha half-life of 2.2 years and a spontaneous fission half-life of 66 ± 10 years. A second laboratory[8] reports 2.55 ± 0.15 years and 82 ± 6 years for these half-lives.

Many features of the spontaneous fission of Cf^{252} have been studied. An account is given in the later chapter on fission (chapter 11) of radiochemical studies of the fission products, of the number of neutrons emitted in fission, and other important features of the spontaneous fission of Cf^{252}. This isotope together with other even-even isotopes of very high atomic number such as Cf^{254}, Fm^{256}, etc., is highly suited for a detailed study of the spontaneous fission process. One milligram of Cf^{252} gives rise to 5×10^{10} spontaneous fission events per minute. It has been suggested[7] that Cf^{252} might be suitable as a portable neutron source. The average number of neutrons released in this spontaneous fission is 3.7; hence one gram of Cf^{252} would release $\sim 10^{12}$ neutrons per second. This would be a much stronger neutron source than any of the (αn) or photoneutron sources listed in the special tables of chapter 5.

The energy of the main alpha particle group was given as 6.117 Mev by MAGNUSSON AND CO-WORKERS[3] who also showed by the measurement of alpha-L-x-ray coincidences that a second group with energy about 6.08 Mev was present. ASARO, STEPHENS, HARVEY AND PERLMAN[5] studied a mixture of californium isotopes in which Cf^{252} accounted for 83 percent and Cf^{250} for 17 percent of the alpha activity. Their measurements on a magnetic spectrograph showed two alpha groups of Cf^{252}: 6.112 Mev (84.5 percent)and 6.069 Mev (15.5 percent). By alpha-gamma coincidence experiments L x-rays and gamma rays of 42 kev (0.014 percent) and 100 kev (0.01_3 percent) were assigned to Cf^{252}. The 42-kev transition is certainly E2 in nature and the 100 kev transition is probably E2. Direct examination of the gamma rays is hindered by the presence of gamma rays from the products of spontaneous fission, but this background is eliminated by suitable coincidence techniques. The decay scheme of Cf^{252} as given in the figure is similar to that of all even-even alpha emitters in this mass region. The L_{II} and L_{III} conversion electrons of the lower energy gamma ray were measured by HOLLANDER[6] who found a transition energy of 43.4 ± 0.1 kev. The alpha group of 5.970 Mev leading to the 4+ level was not directly observed. From the gamma ray measurements its abundance is estimated at 0.2 percent corresponding to an alpha hindrance factor of > 100.

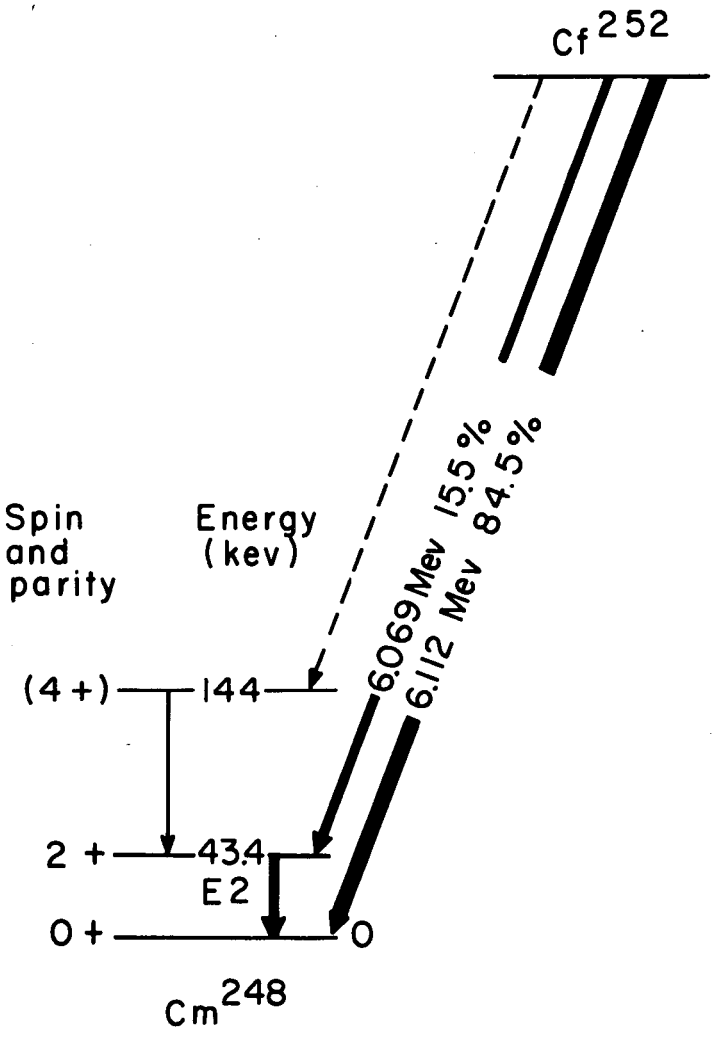


Figure 47. Decay scheme of Cf^{252}.

A californium sample containing a substantial percentage of Cf^{252} is a convenient source of its daughter product Cm^{248} [9].

References Cf^{252}

1. A. Ghiorso, S. G. Thompson, G. R. Choppin and B. G. Harvey, Phys. Rev. 94, 1081 (1954).
2. H. Diamond, L. B. Magnusson, J. F. Mech, C. M. Stevens, A. M. Friedman, M. H. Studier, P. R. Fields and J. R. Huizenga, Phys. Rev. 94, 1083 (1954).
3. L. B. Magnusson, M. H. Studier, P. R. Fields, C. M. Stevens, J. F. Mech, A. M. Friedman, H. Diamond and J. R. Huizenga, Phys. Rev. 96, 1576 (1954).
4. P. R. Fields, A. Ghiorso and co-workers, Phys. Rev. 102, 180 (1956).
5. F. Asaro, F. S. Stephens, Jr., B. G. Harvey, and I. Perlman, Phys. Rev. 100, 137 (1955).
6. J. M. Hollander, Phys. Rev. 103, 1590 (1956).
7. W. C. Bentley, et al., Paper No. P/809 "Proceedings of the International Conference on the Peaceful Uses of Atomic Energy," Vol. 7, United Nations, New York, 1956. See also P. R. Fields, et al., Nature 174, 265 (1954).
8. T. A. Eastwood et al., Phys. Rev. 107, 1635 (1957).
9. J. P. Butler, T. A. Eastwood, H. G. Jackson and R. P. Schuman, Phys. Rev. 103, 965 (1956).

9.6.11 Californium-253. Intense neutron irradiation of plutonium[1,2] or other heavy element samples results ultimately in the formation of californium fractions containing appreciable amounts of Cf^{253}. This isotope has been observed to decay by β^- emission with a half-life reported as 18 ± 3 days[1] or ~20 days[3] or 17 days.[4] These values were obtained by observing the growth of the 6.61-Mev alpha particles of the daughter product, E^{253}, into a californium sample. The beta decay of Cf^{253} has not been observed directly because of the interference of other radiations in the samples available for study. The isotope Cf^{253} has been found in the debris from thermonuclear explosions,[3] and this in fact is where the isotope was discovered.

The isotope Cf^{253} is the lightest isotope of californium to decay by beta emission; hence it is the key isotope leading to the production of einsteinium and heavier elements in neutron-capture reactions.

Preliminary measurements[5] of the beta-particles indicate an end-point energy of 270 kilovolts. An upper limit of one percent has been set on gamma radiation between 100 and 700 kilovolts.[6]

References Cf^{253}

1. L. B. Magnusson, M. H. Studier, P. R. Fields, C. M. Stevens, J. F. Mech, A. M. Friedman, H. Diamond and J. R. Huizenga, Phys. Rev. 96, 1576 (1954).
2. S. G. Thompson, A. Ghiorso, B. G. Harvey and G. R. Choppin, Phys. Rev. 93. 908 (1954); Phys. Rev. 94, 1080 (1954).
3. P. R. Fields, A. Ghiorso and co-workers, Phys. Rev. 102, 180 (1956).
4. T. A. Eastwood et al., Phys. Rev. 107, 1635 (1957).
5. C. J. Gallagher, Jr., unpublished data (1957).
6. F. Asaro, F. S. Stephens, Jr., unpublished results (1957).

9.6.12 Californium-254. The isotope Cf^{254} was first found[1] during the examination of the californium fraction from the debris of the "mike" thermonuclear test explosion. The decay curve for the rate of spontaneous fission could be resolved into two components with half-lives of 2.1 years and 55 days. The 2.1 year component could be identified with Cf^{252} spontaneous fission events and the 55-days component was assigned to Cf^{254} on the basis of yields and decay systematics. This assignment was later confirmed[2] by noting that spontaneous fission activity of this half-life grew into the californium daughter fraction of a sample of 36-hour E^{254} prepared by neutron irradiation of E^{253}.

The decay of Cf^{254} is entirely by spontaneous fission. No decay by alpha particle emission has been observed and the isotope is beta stable.

The first values which were reported for the half-life for spontaneous fission were somewhat rough; the values 55 days,[1] 85 ± 15 days[2] and 60 ± 10 days[3] were reported. The importance of this half-life in astrophysics prompted HUIZENGA AND DIAMOND[4] to re-examine the data by the least-squares method. They obtained a value of 56.2 ± 0.7 days in good agreement with the astrophysical half-life of 55 ± 1 nights deduced from the light intensity curves of type I supernovae.

In chapter 5 section 5.4.3, we have outlined the possible path of forma-

tion of heavy elements in stars by neutron capture processes and referred to the suggestion that the formation of Cf^{254} by neutron capture on a fast-time scale might possibly explain the light intensity curves seen in the supernovae explosions.

References Cf^{254}

1. P. R. Fields, et al., Phys. Rev. 102, 180 (1956); Phys. Rev. 119, 2000 (1960)
2. B. G. Harvey, S. G. Thompson, G. R. Choppin, and A. Ghiorso, Phys. Rev. 99, 337 (1955).
3. Bentley, et al., Paper No. P/809, "Proceedings of the International Conference on the Peaceful Uses of Atomic Energy," Vol. 7, p.261, United Nations, New York, 1956.
4. J. R. Huizenga and H. Diamond, Phys. Rev. 107, 1087 (1957).

9.7 THE ELEMENT EINSTEINIUM (ELEMENT 99)

9.7.1 Discovery of Einsteinium. The first identification of an isotope of element 99 and of element 100 came about as the result of careful analysis of heavy element samples from the "Mike" thermonuclear explosion staged by the Los Alamos Scientific Laboratory in November, 1952. The uranium in this device was subjected to a very intense, instantaneous, neutron flux. Multiple neutron-capture reactions gave rise to very heavy uranium isotopes which rapidly decayed by beta emission into previously uninvestigated heavy isotopes of neptunium, plutonium, americium, curium, berkelium, californium, and of the elements 99 and 100. Three teams of nuclear chemists[1] working at the University of California Radiation Laboratory, the Argonne National Laboratory and the Los Alamos Scientific Laboratory contributed to the chemical isolation and identification of the new elements in December 1952 and January 1953. The scientists involved were A. GHIORSO, S. G. THOMPSON, G. H. HIGGINS AND G. T. SEABORG of the University of California; M. H. STUDIER, P. R. FIELDS, S. M. FRIED, H. DIAMOND, J. F. MECH, G. L. PYLE, J. R. HUIZENGA, A. HIRSCH AND W. M. MANNING of the Argonne National Laboratory; and C. I. BROWNE, H. L. SMITH, AND R. W. SPENCE of the Los Alamos Laboratory.

The names which have been suggested for these elements are einsteinium (symbol E), after Albert Einstein for element 99 and fermium (symbol Fm) after Enrico Fermi, for element 100.

Einsteinium and fermium are actinide elements with chemical properties very similar to americium, curium, berkelium and californium. Einsteinium and fermium apparently exist only in the tripositive state in aqueous solution. Hence, as in the identification of berkelium and californium, the ion-exchange elution technique played a prominent role in their purification and identification. Rare earth contaminants were removed by adsorbing the contaminated heavy-element fractions on Dowex-50 cation resin and eluting with 13 molar hydrochloric acid. The partially-purified, heavy-element fraction was then adsorbed on another column of Dowex-50 resin and eluted carefully with buffered ammonium citrate solution to effect separation of the individual actinide elements. These experiments showed the elution of a 6.6 Mev alpha activity ahead of the element californium and in the expected eka-holmium (element 99) position.

Also a new 7.1-Mev alpha activity was found to elute in the eka-erbium (element 100) position. The conclusions from the detailed experiments were the following: An appreciable amount of U^{253} had formed in the initial nuclear explosion. This rapidly decayed by a chain of short-lived beta emissions to Cf^{253} which decayed by beta emission with a half-life of approximately 20 days to E^{253} which decayed with the emission of 6.6 Mev alpha particles with a half-life of about 20 days. Similarly, an appreciable amount of U^{255} had formed instantaneously and had decayed via a long chain of short-lived beta-emitters to E^{255} which decayed by beta emission with a half-life of approximately 30 days to Fm^{255} which decayed by the emission of 7.1-Mev alpha particles with a half-life of approximately 16 hours.

Since these initial experiments other methods of synthesis of isotopes of einsteinium have been employed. Neutron irradiation of Pu^{239} in high flux reactors for extended periods of time results in the production of californium isotopes by the neutron-capture, beta-decay paths discussed in chapter 5. The lowest mass isotope of californium capable of negative beta decay is Cf^{253}; when Cf^{253} is formed it decays with a 17 day half-life to produce 20-day E^{253}. The E^{253} can capture neutrons to produce higher mass isotopes such as 37 hour E^{254m}, 480 day E^{254}, 24 day E^{255} and the short-lived E^{256}. A number of lighter isotopes ranging in mass number down to 246 have been produced by helium ion or deuteron bombardment of targets containing berkelium and californium isotopes or by bombardment of heavy element targets with carbon ions, nitrogen ions or other heavy ions. All of the isotopes are discussed completely in the sections which follow.

The longest-lived of the einsteinium isotopes is 480 day E^{254} and this isotope will eventually be the most important from the standpoint of chemical study of the element. In the early studies of the element the most suitable isotope for this purpose was 20 day E^{253}.

References - Discovery of Einsteinium and Fermium

1. A. Ghiorso et al., Phys. Rev. 99, 1048 (1955).

2. P. R. Fields et al., Phys. Rev. 102, 180 (1956).

3. H. Diamond et al., Phys. Rev. 119, 2000 (1960).

9.7.2 Einsteinium-246. The bombardment of uranium targets with nitrogen ions accelerated above 100 Mev leads to the production in low yield of the neutron deficient isotopes of einsteinium. The low yield is attributable to the high probability for fission of the compound nucleus before it can be de-excited by neutron emission. GHIORSO, ROSSI, HARVEY AND THOMPSON[1] have identified E^{246} among the products of such a bombardment.

$$_{92}U^{238} + {}_{7}N^{14} \longrightarrow {}_{99}E^{246} + 6\ {}_{0}n^{1}.$$

The information on this isotope is scanty. Einsteinium-246 decays by orbital electron capture: its decay was noted only through the growth of 36-hour Cf^{246} daughter[1]. Alpha particles of 7.35 Mev were observed to decay with a half-life of 7.3 minutes.[1] These alpha particles were originally assigned tentatively to E^{247} but they are perhaps more properly assigned to E^{246}. GUSEVA AND CO-WORKERS have also observed these alpha particles.

References E^{246}

1. A. Ghiorso, G. B. Rossi, B. G. Harvey and S. G. Thompson, Phys. Rev. 93, 257 (1954).
2. Guseva, Filippova, Gerlit, Druin, Myasoedov and Tarantin, Atomic Energy (USSR) No. 2, 50 (1956); Soviet J. Atomic Energy (Consultants Bureau Trans.) No. 2, 193 (1956).

9.7.3 Einsteinium-247. No definite information is available on E^{247}.

9.7.4 Einsteinium-248. The isotope E^{248} was first prepared by the deuteron bombardment of Cf^{249} with 18 to 22 Mev deuterons:[1]

$$Cf^{249}\ (d,3n)\ E^{248}.$$

The targets available in the original study of this reaction contained only 10^{13} atoms of Cf^{249} so the specialized bombardment techniques discussed in the next section were employed.

Einsteinium-248 decays principally by electron capture with a half-life of 25 ± 5 minutes and also by the emission of 6.87 ± 0.02 Mev alpha particles. The ratio of electron capture to alpha decay is estimated at 400 from measurements of the alpha counting rate of E^{248} and of the Cf^{248} which grew into the sample.

Reference E^{248}

1. A. Chetham-Strode and L. W. Holm, Phys. Rev. 104, 1314 (1956).

9.7.5 Einsteinium-249. The two-hour isotope E^{249} is conveniently prepared by bombardment of Bk^{249} or Cf^{249} with helium ions[1] or deuterons:[2]

$$Bk^{249}\ (\alpha,4n)\ E^{249}$$
$$Cf^{249}\ (\alpha,p3n)\ E^{249}$$
$$Cf^{249}\ (d,2n)\ E^{249}$$

In the original tests of this method the total amount of Bk^{249} available for use as a target was only 1.3×10^{-8} grams or about 3×10^{13} atoms. Special target methods were worked out to use effectively this small amount of material and to conserve it for reuse. Since these methods are of general application we shall describe them briefly.

The Bk^{249} was electrodeposited in a micro cell over a small rectangular area upon a 0.005 cm. thick gold foil. This target area was exposed to a deflected collimated helium-ion beam from the 60-inch Berkeley cyclotron. The beam intensity was approximately 50 microamperes per square centimeter. The helium ions passed through the gold foil and then through the Bk^{249} target material so that nuclear reaction products were ejected from the target in the forward direction. They were caught on a 0.00025 cm. thick gold "catcher" foil placed about 5.5 millimeters from the target foil. By this method it was possible to bombard the Bk^{249} target a large number of times without the necessity of dissolving it. Some 100 bombardments, amounting to a total irradiation of roughly 1000 microampere hours were made with no loss of target material.

The energy of the berkelium ions was varied by inserting aluminum degrading foils ahead of the target. The helium ion beam intensity was measured with a Faraday cup placed behind the thin gold catcher foil. The recoil energy of the products of (α,xn) reactions is sufficiently great that essentially complete removal of these products from the target is achieved if the target thickness is less than about 30 micrograms per square centimeter.

After bombardment the thin gold "catcher" foil is dissolved and the reaction products are chemically separated. This technique not only conserves the target material for reuse but simplifies the chemical treatment of the

products since none of the target material has to be removed.

The target arrangement can be visualized in the expanded schematic view of the target assembly shown in Figure 48. The helium ion beam was first collimated by means of the water cooled copper and graphite collimator A. The second collimator B reduced the beam to the required size. Stainless steel holder C supported a 0.04 cm. thick duraluminum foil which sealed the rest of the probe assembly from the cyclotron vacuum. Aluminum absorber strips could be mounted behind the duraluminum foil. Holder D supported the target. The foils C and D were cooled by means of rapid circulation of helium between them. The gold "catcher" foil E was mounted on the end of rod F with scotch tape. The rod F terminated in a water cooled aluminum block which served as a Faraday cup. The space between the target and the catcher foil was evacuated. The several cooling and vacuum systems were interlocked so that the cyclotron could not be operated until proper conditions were obtained.

The rod F could be very rapidly withdrawn after completion of a bombardment so that no time was lost in examining the products. Cooling of the foils was adequate to permit a beam as large as 200 microamperes per square centimeter to be used.

When Bk^{249} is bombarded with helium ions several isotopes of einsteinium are produced. The yield of these isotopes as a function of the helium ion energy is given in figure 49. These excitation functions were important in confirming the mass assignments of the products.

Einsteinium-249 has a half-life of 2 hours. It decays partly by emission of alpha particles with an energy of 6.76 Mev. The chief decay is by orbital electron capture to produce 360-year Cf^{249}. The ratio of orbital electron capture to alpha decay is estimated at 760. Study of the radiations of E^{249} is hindered by the presence of the other einsteinium isotopes. The decay energy for electron capture is estimated at 1.4 Mev by the use of closed decay cycles.

References E^{249}

1. B. G. Harvey, A. Chetham-Strode, Jr., A. Ghiorso, G. R. Choppin, and S. G. Thompson, Phys. Rev. 104, 1315 (1956).
2. A. Chetham-Strode and L. W. Holm, Phys. Rev. 104, 1314 (1956).

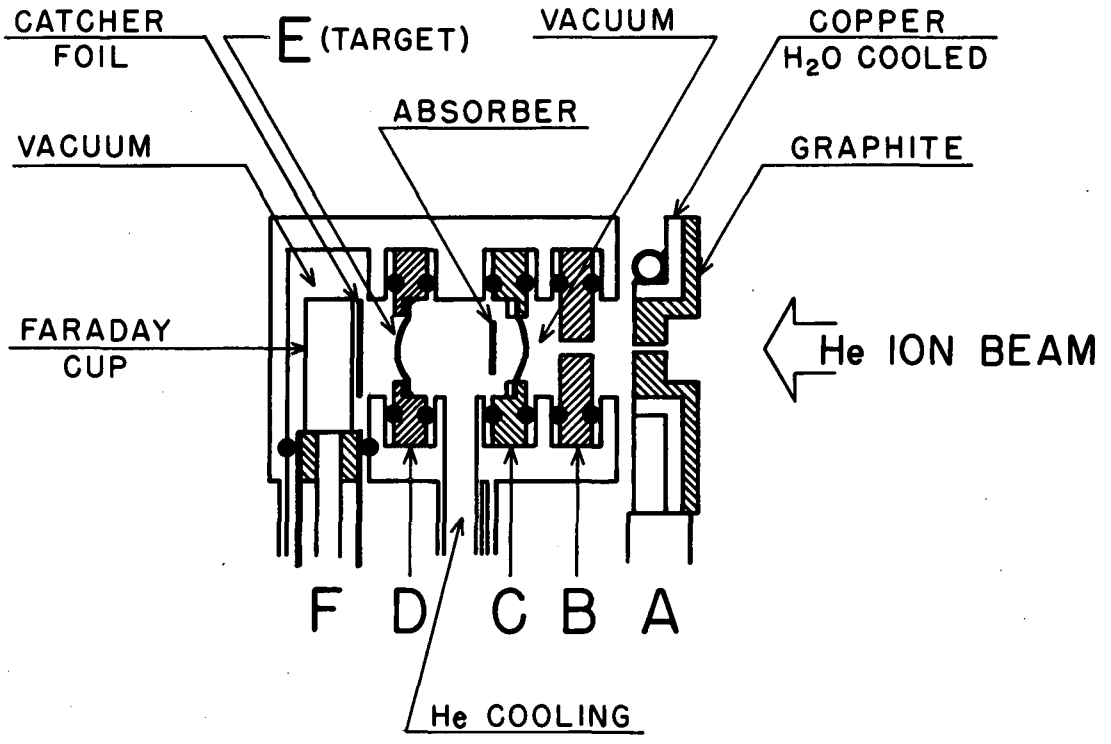


Figure 48. Schematic view of target assembly used by Harvey and Co-Workers, Phys. Rev. 104, 1315 (1956). The labeled parts are described in the text.

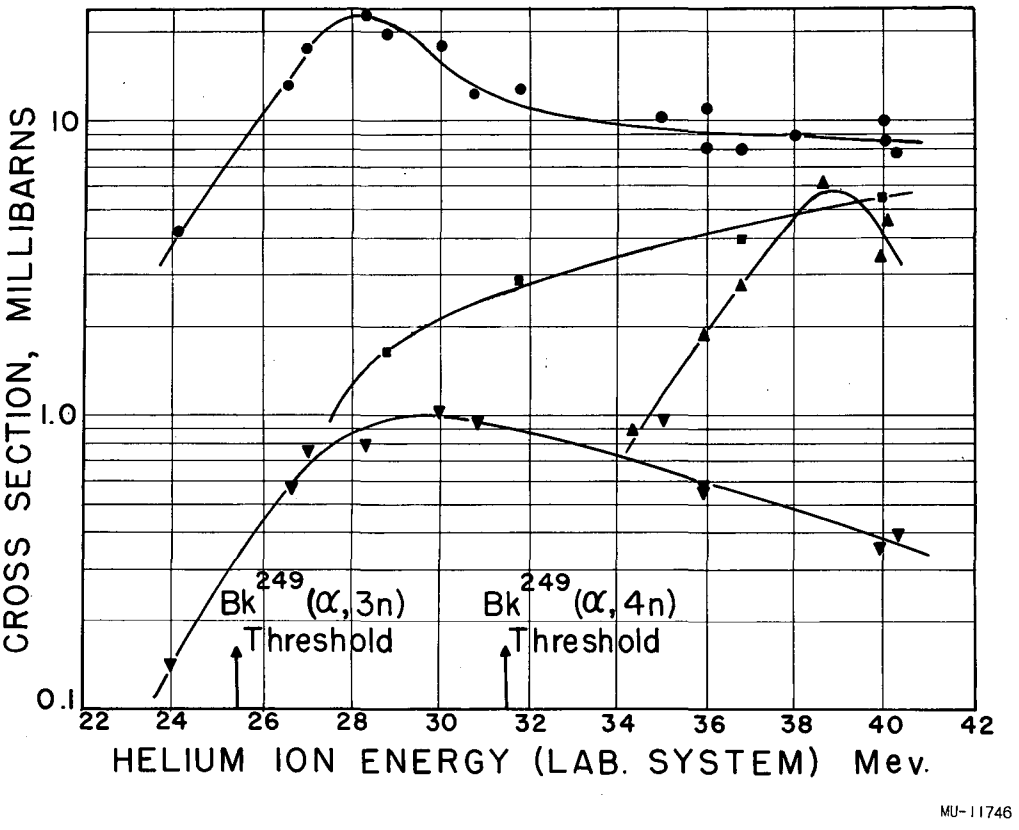


Figure 49. Excitation functions for formation of einsteinium isotopes. The target is Bk^{249} plus some Cf^{249} daughter.

Key:

▼ Bk^{249} (α,n) E^{252}

● Bk^{249} $(\alpha,2n)$ E^{251}

■ Bk^{249} $(\alpha,3n)$ E^{250} + $Cf^{249}(\alpha,3n)Fm^{250} \rightarrow E^{250}$ + $Cf^{249}(\alpha,p2n)E^{250}$ + $Cf^{249}(\alpha,t)E^{250}$

▲ Bk^{249} $(\alpha,4n)$ E^{249} + Cf^{249} $(\alpha,p3n)$ E^{249}.

This figure is reproduced from the paper of Harvey and Co-Workers.[1]

9.7.6 Einsteinium-250. It is possible to prepare E^{250} by the following reactions:

$$Bk^{249}\ (\alpha,3n)\ E^{250}$$
$$Cf^{249}\ (\alpha,3n)\ Fm^{250} \xrightarrow{EC} E^{250}$$
$$Cf^{249}\ (\alpha,p2n)\ E^{250}$$
$$Cf^{249}\ (\alpha,t)\ E^{250}$$
$$Cf^{249}\ (d,n)\ E^{250}.$$

A composite excitation function for the production of E^{250} by a helium ion bombardment of a mixed target of Bk^{249} and Cf^{249} is given in the last shown figure. Suitable technique for carrying out such bombardments is discussed in the preceeding section. A number of reactions of heavy ions with heavy element target materials lead to the formation of E^{250}.

Einsteinium-250 decays by orbital electron capture with a half-life of 8 hours. The branching decay by alpha emission is slight and has not been observed. The available decay energy for electron capture and alpha-particle emission have been estimated as 1.94 Mev and 6.72 Mev respectively.

References E^{250}

1. B. G. Harvey, A. Chetham-Strode, Jr., A. Ghiorso, G. R. Choppin and S. G. Thompson, Phys. Rev. 104, 1315 (1956).

9.7.7 Einsteinium-251. The first preparation[1] of E^{251} was carried out by the reaction:

$$Bk^{249}\ (\alpha,2n)\ E^{251}.$$

The excitation function for this reaction is shown in section 9.7.5. An experimental technique suitable for carrying out such a reaction is discussed in that section.

Einsteinium-251 decays by orbital electron capture and by the emission of alpha particles with 6.48-Mev energy. The observed half-life is 1.5 days and the ratioof electron capture to alpha decay is 190. The decay energy available for electron capture has been estimated as 0.41 Mev from closed decay cycles.

Reference E^{251}

1. B. G. Harvey, A. Chetham-Strode, Jr., A. Ghiorso, G. R. Choppin and S. G. Thompson, Phys. Rev. 104, 1315 (1956).

9.7.8 Einsteinium-252. The isotope, E^{252}, was discovered as a result of the bombardment of Bk^{249} with helium ions. A suitable experimental technique is discussed in section 9.7.5. The excitation function for the preparative reaction is also given in Figure 49 which appears in that section.

$$Bk^{249}\ (\alpha,n)\ E^{252}$$

Einsteinium-252 decays by the emission of 6.64-Mev alpha particles with a half-life of ~140 days. Einsteinium-252 is expected to be unstable toward electron capture by 1.23 Mev and toward β^- decay by ~0.24 Mev but no radiations corresponding to either transition have been reported.

Reference E^{252}

1. B. G. Harvey, A. Chetham-Strode, Jr., A. Ghiorso, G. R. Choppin and S. G. Thompson, Phys. Rev. 104, 1315 (1956).

9.7.9 Einsteinium-253. The first isotope of element 99 to be identified was E^{253}. The discovery of this isotope in the debris of a thermonuclear test explosion is discussed earlier in section 9.7.1. This nuclide can also be prepared by the intense irradiation of plutonium, americium, curium or californium samples with neutrons.[2,3,4] The pertinent neutron capture sequence starting with Cm^{244} is the following:

$$Cm^{244}(n,\gamma)Cm^{245}(n,\gamma)\ldots.Cm^{249} \xrightarrow{\beta^-} Bk^{249}(n,\gamma)Bk^{250} \xrightarrow{\beta^-} Cf^{249}(n,\gamma)Cf^{250}(n,\gamma)Cf^{251}(n,\gamma)Cf^{252}(n,\gamma)Cf^{253} \xrightarrow{\beta^-,\ 20\ \text{days}} E^{253}$$

Very prolonged irradiation[2,3,4] results in the production of higher mass isotopes of einsteinium but it is possible to obtain pure samples of E^{253} by isolating a californium fraction containing an appreciable amount of the 20 day beta-emitter Cf^{253}. After a suitable decay period pure E^{253} can be separated chemically.

Einsteinium-253 is beta stable. The half-life for alpha emission has been measured several times;[1,4,5,6] we quote here the precise value of 20.03 ± 0.01 days determined by JONES AND CO-WORKERS.[4] A number of workers have measured the energy of the main alpha particle group by the ion chamber technique.[4,5,6] When larger samples became available it was possible to carry out more precise measurements with magnetic spectrographs. The alpha spectrum was studied at Berkeley with a 60°-sector single focussing spectrograph and with a 180° double-focussing spectrograph.[8-11] Twelve alpha groups which seem clearly established are listed in Table 39, together with their relative intensities and hindrance factors. (The hindrance factors are defined as the reciprocal of the reduced alpha transition probabilities compared to those of the neighboring even-even nuclei. The term reduced transition probability refers to the corrected transition probability after the energy dependence is removed.) These alpha groups are placed in the decay scheme in Figure 50; this figure also contains several tentative groups not listed in Table 39, which are deduced from the gamma ray measurements and other considerations.

Table 39. Alpha Groups of E^{253}

Energy (Mev)	6.633	6.624	6.594	6.592	6.552	6.540	6.497	6.479	6.429	6.249	6.209	6.158
Intensity (%)	90	0.8	0.7	6.6	0.75	0.85	0.26	0.08	0.1	0.04	0.04	0.015
Hindrance Factor	1.93	200	165	17	100	77	160	430	200	70	40	60

Table 40. Gamma-rays of E^{253}

Energy Gamma-ray	Conversion electron	Multipolarity
	30.7	M1-E2
41.7	41.7	M1-E2
	42.9	M1 (E2?)
51	51.7	M1-E2
	54.8	M1 (E2?)
	73.5	E2
	93.3	E2
	97.6	E2
110 K-x-ray (0.04%)		
393 (0.05%)		M1
439 (~0.01%)		

The conversion electrons of E^{253} have been studied on a 180° permanent-magnet electron spectrograph,[11] with the results listed in Table 40. Several conversion lines were seen for each of the gamma rays listed, and from the sub-shell conversion intensity ratios the multipolarity assignments listed in Table 40 were made. In most cases these assignments were unambiguous; but in a few cases questionable assignments arose which are indicated in Table 40 by question marks. Studies of the gamma rays accompanying E^{253} decay have also been made using NaI scintillation counters;[4,8,11] the results of which have been included. The intensities for the higher-energy gamma rays are given, but for the lower energy ones these intensities are not very certain and have not been included. The M1 assignment for the 393 kev gamma ray is based on the large number of K x-rays relative to the 393- and 439-kev gamma rays, whose conversion is presumably the only source of these x-rays. All the gamma rays listed in Table 40 have been placed in the decay scheme of Fig. 50. The following discussion of this decay scheme is taken from a report by ASARO, THOMPSON, STEPHENS AND PERLMAN.[11]

As is indicated in Fig. 50, the energy levels of Bk^{249} have been divided into three rotational bands, and the base levels of each band have been assigned as a particular Nilsson state. The basis for the division into three bands is fairly clear; the energy separations, the transitions, and the alpha populations all indicate such a division. The assignment of particular Nilsson orbitals is not so obvious and will be discussed in some detail below.

The relative energy spacings of the assigned states in the 8.8-kev band are in excellent agreement with a spin of 3/2 for the base state. In addition the rotational constant, $\hbar^2/2\mathfrak{I}$, for spin 3/2 is 6.1 kev, in good agreement with the usual values for odd mass nuclides. Although the parity of this band cannot be determined simply from the experimental data, inspection of a Nilsson diagram (see figure 3.25 in Chapter 3) readily shows the most probable state is 3/2 - [521]. This assignment is supported by the fact that even-parity, odd-proton levels in this region would be expected to have rotational constants smaller than 6.1. It will be shown later that the ground state spin of Bk^{249} is 7/2+. Thus an M2 transition might be expected between the 3/2- and 7/2+ states. Alpha-electron delayed coincidence measurements showed a half-life of 0.3 milliseconds following about 2% of the E^{253} alpha transitions, in good agreement with the total alpha population to the 3/2- band. This lifetime is retarded by about a factor of 50 over the single proton M2 value.[12]

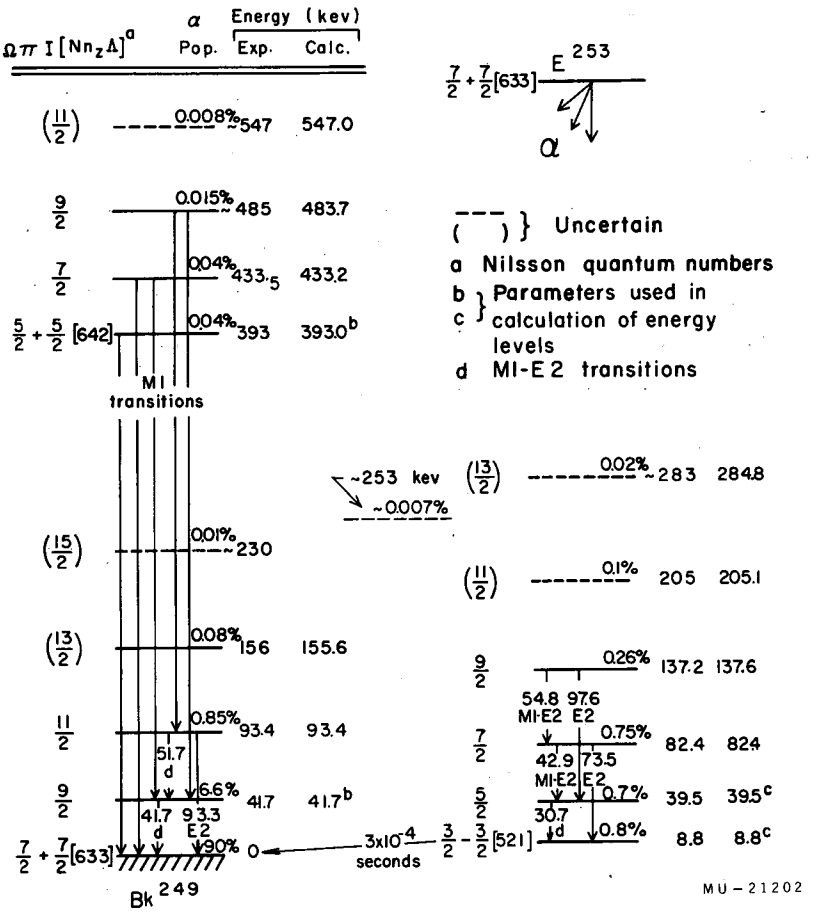


Figure 50. Decay scheme of $_{11}E^{253}$ as formulated by Asaro, Thompson, Stephens, and Perlman.[11] Nilsson state assignments are given.

The ground state and 393-kev bands are discussed together as they are closely related. From the level spacing of this ground state band, the spin of the ground state is calculated to be 3.16 ± 0.01. This value is closer to 7/2 than any other spin, but the agreement is not very good. Also, the rotational constant for this band has a value of 4.63-kev, which is unusually low. STEPHENS[11] calculated the properties of this band, however, considering the Coriolis interaction[13] as modified by the pairing correlations[14] and these quantities are both well explained provided the assignment 7/2+ [633] is made for this band. The Nilsson diagram shows that this assignment is proper for element 99. The decay characteristics and spacing of the 393-kev band are also well accounted for if the 5/2+ [642] assignment is made, which, again is an acceptable choice from the Nilsson diagram. This case is a particularly favorable one for detailed Coriolis calculations for the following reasons: (1) within the entire 82-126 proton shell the only positive parity levels are those originating from the $i_{13/2}$ shell-model level and these can all be taken into account explicitly; the Coriolis interaction occurs only between states of the same parity. (2) The Coriolis matrix elements are large and do not depend on details of the Nilsson wave functions, and (3) the positions and moments of inertia of two bands from the $i_{13/2}$ orbital are known. Two parameters were used in the calculation, which fix the positions of the 393 and 41.7 kev levels. The other calculated energies are given in Fig. 50, and agree well with the experimental values. It is interesting to note that the mixing causes positive deviations from the I(I+1) formula in the ground (K=7/2) band and irregular deviations from the I(I+1) formula in the 393-kev (K=5/2) band. The experimental data confirm the former trend but are not sufficiently accurate to test the latter. The transition between the two bands are M1 as would be expected from calculation using the mixed Nilsson wave functions.

The theoretical alpha populations to various members in a rotational band are readily calculated for an alpha wave of a given angular momentum.[15] In addition, if the parent and daughter states have the same configuration, the alpha transition to the base state of the band is essentially unhindered, and the relative alpha populations to the various band members can be calculated explicitly.[15] This is the phenomenon of favored alpha decay discussed in Chapter 4. As seen in Table 41, the above conditions are well met by the ground state band, resulting in the same assignment for the E^{253} ground state, 7/2+ [633], as found for Bk^{249}.

Table 41. Alpha decay of E^{253} to the 7/2+ band in Bk^{249}.

Excited state Energy (kev)	I	Calculated Abundances[a] (%) L=0	L=2	L=4	Σ_L	Experimental Values (%)	Hindrance Factors[b]
0	7/2	79.6	10.0	0.1	89.7	90	1.9
41.7	9/2	--	5.9	0.3	6.2	6.6	17
93.4	11/2	--	0.9	0.3	1.2	0.85	7.7
156	13/2	--	--	0.11	0.11	0.08	430
230	15/2	--	--	0.009	0.009	0.01	~1500

[a]The abundances are calculated from the expression

$$P_E = P_{oE} \sum_L C_L \begin{pmatrix} I_i & L & I_f \\ K_i K_f & -K_i K_f & \end{pmatrix} ; \quad \sum_E P_E$$ is the total population to the rotational band, P_{oE} is the unhindered population to a state E calculated from simple spin independent alpha decay theory, C_L is the reciprocal of the hindrance factors for the various L waves in the adjacent even-even nuclides ($C_0 : C_2 : C_4$ = 1:0.27:0.023 from Fm^{254} and Cf^{252})[16] and the last factor is a Clebsch Gordan coefficient.

[b]The hindrance factors as used here are the ratio of the experimental partial half life of a given α group to the value calculated from spin independent alpha decay theory[17] using the radii of adjacent even-even nuclides.

In the hindered alpha decays to the other bands, the relative alpha populations are also in good agreement with the calculated values, consistent with the spin and parity assignments.

References - E^{253}

1. A. Ghiorso et al., Phys. Rev. 99, 1048 (1955), 102, 180 (1956), 119, 2000 (1960).
2. S. G. Thompson, A. Ghiorso, B. G. Harvey, and G. R. Choppin, Phys. Rev., 93, 908 (1954).
3. M. H. Studier et al., Phys. Rev. 93, 1428 (1954).
4. M. Jones et al., Phys. Rev. 102, 203 (1956).
5. G. R. Choppin, S. G. Thompson, A. Ghiorso, and B. G. Harvey, Phys. Rev. 94, 1080 (1954).
6. P. R. Fields et al., Phys. Rev. 94, 209 (1954).
7. A. Ghiorso and B. G Harvey, unpublished data, 1954 reported in reference 8.
8. F. S. Stephens, University of California Radiation Laboratory Report, UCRL-2970, June 1955.
9. J. P. Hummel, University of California Radiation Laboratory Report, UCRL-3456, 1956.
10. J. P. Hummel, G. Choppin, F. Asaro and I. Perlman, unpublished data (1955) reported in reference 8.
11. F. Asaro, S. G. Thompson, F. S. Stephens and I. Perlman, University of California Radiation Laboratory Report, UCRL-9382 (1960); presented in somewhat different form by I. Perlman in Proceedings of the International Conference on Nuclear Structure at Kingston, Ontario, Aug. 29-Sept. 3, 1960, University of Toronto Press, pp. 553-557.
12. S. A. Moszkowski, Beta and Gamma Ray Spectroscopy, p. 391, edited by Kai Siegbahn (1955), Interscience Publishers Inc., New York and North Holland Publishing Company, Amsterdam.
13. A. Kerman, Mat. Fys. Medd. Dan. Vid. Selsk., 30, No. 15. (1955)
14. S. T. Belyaev, Mat. Fys. Medd. Dan. Vid. Selsk. 31, No. 11 (1959).
15. A. Bohr, P. O. Fröman and B. R. Mottelson, Dan. Mat. Fys. Medd, 29, No. 10, (1955)
16. H. V. Michel, University of California Lawrence Radiation Laboratory Report, UCRL-9229, May 1960.
17. M. A. Preston, Phys. Rev. 71, 865 (1947).

9.7.10 The Isomers of Einsteinium-254. Einsteinium of mass 254 exists in two isomeric forms both produced by neutron irradiation of E^{253}. One decays chiefly by emission of a beta particle with a half life of 38.5 ± 1.0 hours to form Fm^{254}. The second decays by alpha particle emission to form Bk^{250}. The relationship of the two isomers to each other is unknown. For convenience the 38.5 hour species will be referred to here as E^{254m}. The 38.5 hour isomer was first produced and identified independently by two groups of investigators[1,2] who irradiated small samples of 20 day E^{253} with neutrons and noted the growth into the repurified einsteinium fraction of a 3[illegible]3 hour fermium alpha emitter. The half life of the einsteinium beta emitter supporting this alpha emitter was found to be 38 hours. The long-lived form was first reported by HARVEY, THOMPSON, CHOPPIN, AND GHIORSO[3] who noted the presence of 6.44 Mev alpha particles in the einsteinium fraction many months after the neutron irradiation of a sample of E^{253}. The mass assignment was proved by the recoil collection of the daughter product, identified as the 3-hour beta emitter, Bk^{250}. The work on both isomers has been confirmed by JONES AND CO-WORKERS.[4]

Information on the radiations of 38.5 hour E^{254m} is incomplete. Values of 1.1 Mev[2] and 1.04 ± 0.04 Mev[4] have been reported for the main beta particle group. A gamma ray of 660 ± 15 kev was observed in an abundance of 0.6 to 0.8 per decay event by JONES AND CO-WORKERS.[4] STEPHENS[6] found the 660 kev gamma peak to be in coincidence with beta particles of ~0.45 Mev. He also showed that the 1.04 Mev beta group was in coincidence with L x-rays.

Later studies by ASARO AND PERLMAN with a scintillation spectrometer showed that the gamma spectrum was complex. Gamma ray photons with energy 529, 575, 636, and 680 were resolved. L x-rays were prominent, indicating conversion of a lower energy transition. A series of beta-gamma, K x-ray-gamma, and L x-ray-gamma coincidence experiments led to the tentative decay scheme shown in the figure. This scheme was constructed on the assumption that the spectrum of collective excitations universally observed in even-even nuclei throughout the transuranium element groups of nuclides would also appear in Fm^{254}. HARVEY, THOMPSON, GHIORSO, AND CHOPPIN[3] proved the existence of a 0.1 percent electron capture branch by detecting the growth of spontaneous fission activity of Cf^{254} into a purified einsteinium fraction.

The half life of the long-lived form of E^{254} has been reported as 272 days[5] and 480 ± 70 days.[9] This is the longest half-life for any isotope of

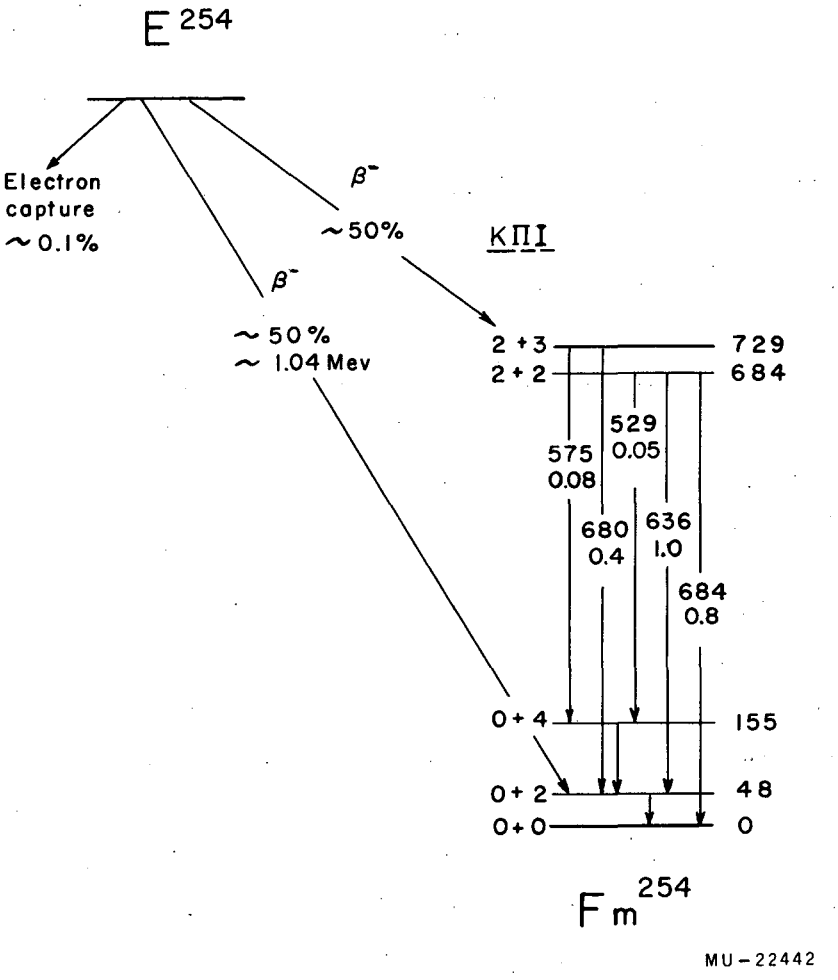


Figure 51. Tentative decay scheme of 38.5 hour E^{254} as formulated by Asaro and Perlman. Gamma transition energies are given in kev. These are accurate to about 5 kev. Intensities relative to the 636 kev peak are given beneath the transition energy. The KπI assignments are not proved. If they are correct the probable spin for E^{254} is 2 or 3. The photons of the 48 kev and 107 kev transitions were not directly observed. The presence of the transitions was deduced from L x-ray-gamma coincidence measurements.

einsteinium and hence this isomer is the most suitable material for the study of the chemical properties of the element. Preliminary measurements of the alpha particle energy by the ion-chamber method yielded the values 6.44 ± 0.01 Mev[3] and 6.42 ± 0.02 Mev.[4] ASARO AND PERLMAN[7] made some preliminary measurements in a magnetic spectrometer and determined an energy of 6.430 ± 0.005 Mev for the main group. The intensity of this group is about 90 percent. STEPHENS, ASARO, AND PERLMAN[8] report the presence of a 62 ± 2 kev gamma ray in the alpha decay of long-lived E^{254}. By alpha-gamma delayed coincidence measurements the half life of this transition was measured to be 3.9×10^{-8} seconds.

REFERENCES - Isomers of E^{254}

1. P. R. Fields, M. H. Studier, J. F. Mech, H. Diamond, A. M. Friedman, L. B. Magnusson, and J. R. Huizenga, Phys. Rev. 93, 1428 (1954) and Phys. Rev. 94, 209 (1954).

2. S. G. Thompson, A. Ghiorso, B. G. Harvey, and G. R. Choppin, Phys. Rev. 93, 1129 (1954) and Phys. Rev. 94, 1080 (1954).

3. B. G. Harvey, S. G. Thompson, A. Ghiorso, and G. R. Choppin, Phys. Rev. 99, 337 (1955).

4. M. Jones, R. P. Schuman, J. P. Butler, G. Cowper, T. A. Eastwood, and H. G. Jackson, Phys. Rev. 102, 203 (1956).

5. B. G. Harvey and co-workers, unpublished results, December 1955.

6. F. S. Stephens, Jr., University of California Radiation Laboratory Report UCRL-2970, June 1955.

7. F. Asaro and I. Perlman, unpublished results, 1960.

8. F. S. Stephens, Jr., F. Asaro, and I. Perlman, unpublished results, 1957.

9. R. P. Schuman, T. A. Eastwood, H. G. Jackson, and J. P. Butler, J. Inorg. Nuclear Chem. 6, 1 (1958).

9.7.11 Einsteinium-255. The isotope E^{255} was discovered during the initial studies[1] of the heavy element fractions from the thermonuclear test which led to the discovery of elements 99 and 100. In this instance U^{238} was converted to U^{255} which decayed by a long series of beta transitions to form E^{255}. This isotope has also been produced by the intense neutron irradiation of samples containing appreciable amounts of 20 day E^{253} or of isotopes of lighter elements which can be converted to E^{253}.

$$E^{253}\ (n,\gamma)\ E^{254}\ (n,\gamma)\ E^{255}$$

This isotope decays by beta emission with a half-life given as 30 days[1,3] and 24 ± 2 days.[2] No additional information is available on its decay scheme. The decay energy available for β^- emission is estimated as 0.38 Mev.

The decay product of E^{255} is the 21-hour alpha-emitter, Fm^{255}, which can be observed to grow into a freshly purified sample.

References E^{255}

1. A. Ghiorso, M. H. Studier, C. I. Browne, and co-workers, Phys. Rev. 99, 1048 (1955); Phys. Rev. 102, 180 (1956); Phys. Rev. 119, 2000 (1960).
2. M. Jones, R. P. Schuman, J. P. Butler, G. Cowper, T. A. Eastwood and H. G. Jackson, Phys. Rev. 102, 203 (1956).
3. G. R. Choppin, S. G. Thompson, A. Ghiorso and B. G. Harvey, Phys. Rev. 94, 1080, (1954).

9.7.12 Einsteinium-256. The only information available on E^{256} is that it is a beta emitter with a short half-life (hours or less). It is produced by neutron irradiation of samples containing E^{255}.

Reference E^{256}

1. G. R. Choppin, B. G. Harvey, S. G. Thompson and A. Ghiorso, Phys. Rev. 98 1519, (1955).

9.8 THE ELEMENT FERMIUM (ELEMENT 100)

9.8.1 General Comments. The first isotope of fermium (element 100) and the first isotope of einsteinium (element 99) were identified in the same investigation. Hence the discovery of fermium is described together with the discovery of einsteinium in section 9.7.1. The first isotope of element 100 to be discovered was 21.5 hour Fm^{255}. Subsequent to this work a number of other isotopes were identified in heavy element targets bombarded with neutrons, deuterons, helium ions, and heavier ions. The longest-lived of the known isotopes is 4.5 day Fm^{253}. On the basis of the predictions which have been made for the half lives of undiscovered species it is unlikely that an isotope of longer half life will be found and essentially certain that no long-lived form will be discovered. Spontaneous fission instability plays a large role in shortening the lifetime of some of the heavier isotopes which might otherwise be prepared in weighable amounts. Fm^{258}, for example, is beta stable and has an estimated alpha-decay half-life of 300 days. The half-life of this species for spontaneous fission is, however, expected to be only 30 seconds.

9.8.2 Fermium-248. GHIORSO, SIKKELAND, WALTON AND SEABORG[1] prepared Fm^{248} by the bombardment of Pu^{240} with C^{12} ions accelerated in the Berkeley heavy ion linear accelerator (Hilac). It was shown that Fm^{248} decays by alpha particle emission to produce Cf^{244}. The measured half life was 0.6 minutes. GUSEVA AND CO-WORKERS[2] found a little evidence for Fm^{248} in a bombardment of an uranium target with 120 Mev oxygen ions. In a chemically-separated transplutonium element fraction the 7.0 Mev alpha particles of Cf^{254} were seen and it was concluded that the Cf^{254} had been formed by the alpha decay of a short-lived Fm^{258}.

References Fm^{248}

1. A. Ghiorso, T. Sikkeland, J. R. Walton and G. T. Seaborg, Phys. Rev. Letters 1, 18 (1958).
2. Guseva, Filippova, Gerlet, Druin, Myasoedov and Tarantin, Soviet J. Atomic Energy (Consultants Bureau transl.), 2, 193 (1956).

9.8.3 Fermium-250. Some tentative evidence for an isotope of element 100 with mass 250 was obtained by a Swedish research team[1] who bombarded uranium with ions of O^{16} accelerated in the 225-cm cyclotron of the Nobel Institute for Physics. After the bombardment, heavy element fractions were separated by ion exchange techniques. In the element 100 fraction a few atoms of an isotope emitting 7.7 Mev alpha particles with a half-life of about a half hour were observed.

More evidence for Fm^{250} was found by AMIEL AND CO-WORKERS[2] in the bombardment of a small sample ($\sim 10^{13}$ atoms) of Cf^{249} with helium ions. Fermium-250 was produced by the reaction:

$$Cf^{249}\ (\alpha, 3n)\ Fm^{250}.$$

The recoil collection technique described in section 9.7.5 was used. The observed half life was 30 minutes for the emission of 7.43-Mev alpha particles. The mass assignment is made on the basis of the excitation function shown in the figure and also on the basis of the alpha particles of Cf^{246} which were observed in the sample after decay. Fermium-250 is unstable toward electron capture by 0.94 Mev of energy but this mode of decay was not directly observed. Fermium-250 has also been observed as the daughter of 102^{254} prepared by the bombardment of Cm^{246} with carbon ions. (See section 9.10)

References Fm^{250}

1. H. Atterling, W. Forsling, L. W. Holm, L. Melander and B. Astrom, Phys. Rev. 95, 585 (1954).
2. S. Amiel, A. Chetham-Strode, Jr., G. R. Choppin, A. Ghiorso, B. G. Harvey, L. W. Holm and S. G. Thompson, Phys. Rev. 106, 553 (1957).

9.8.4 Fermium-251. The 7-hour isotope Fm^{251} has been prepared by the bombardment of Cf^{249} with helium ions using the recoil "catcher" foil technique discussed in section 9.7.5.

$$Cf^{249}\ (\alpha, 2n)\ Fm^{251}$$

The ratio of orbital electron capture to alpha decay of Fm^{251} is about 100. The energy of the alpha particles is 6.89 Mev.

The mass assignment is based on the yield curve shown in Figure 52.

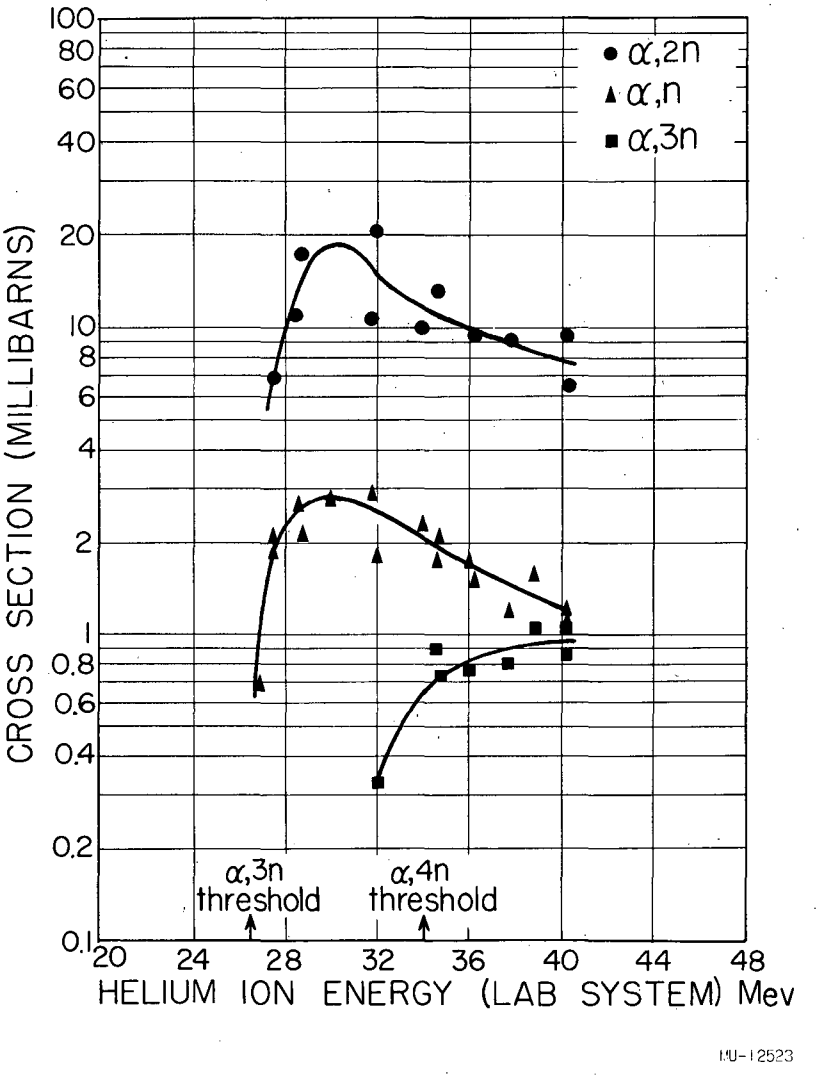


Figure 52. Excitation function for fermium isotopes prepared by helium ion bombardment of Cf^{249}. The (α,n), (α,2n), and (α,3n) products are Fm^{252}, Fm^{251}, and Fm^{250}, respectively, from Amiel et al.[2]

References Fm^{251}

1. S. Amiel, A. Chetham-Strode, Jr., G. R. Choppin, A. Ghiorso, B. G. Harvey, L. W. Holm and S. G. Thompson, Phys. Rev. 106, 553 (1957).

9.8.5 Fermimum-252. FRIEDMAN AND CO-WORKERS[1] first prepared Fm^{252} by bombarding a mixed target of Cf^{250}, Cf^{251} and Cf^{252} with helium ions of 34 Mev and 42 Mev energy. AMIEL AND CO-WORKERS[2] prepared it by bombardment of a pure sample of Cf^{249} with helium ions of several energies. The preparative reactions involved in these two studies were:

$$Cf^{250}\ (\alpha,2n)\ Fm^{252}$$
$$Cf^{251}\ (\alpha,3n)\ Fm^{252}$$
$$Cf^{252}\ (\alpha,4n)\ Fm^{252}$$
$$Cf^{249}\ (\alpha,n)\ Fm^{252}$$

The half-life was reported as 22.7 hours[1] and 30 hours.[2] The energy of the alpha particles is given as 7.04[1] and as 7.05[2] Mev. The mass assignment is based on the observation of the alpha particles of the Cf^{248} daughter and on the excitation function shown in Figure 52 accompanying section 9.8.3. A lower limit of 3000 days was set on the half-life for spontaneous fission.[1] Fermium-252 is beta stable.

References Fm^{252}

1. A. M. Friedman, J. E. Gindler, R. F. Barnes, R. Sjoblom and P. R. Fields, Phys. Rev. 102, 585 (1956).
2. S. Amiel, A. Chetham-Strode, Jr., G. R. Choppin, B. G. Harvey, L. W. Holm and S. G. Thompson, Phys. Rev. 106, 553 (1957).

9.8.6 Fermimum-253. FRIEDMAN AND CO-WORKERS[1] made a tentative identification of Fm^{253} which was confirmed by the work of AMIEL[2]; both groups prepared the isotope by the reaction:

$$Cf^{252}\ (\alpha,3n)\ Fm^{253}$$

AMIEL[2] isolated the product of this reaction by the recoil "catcher" foil technique discussed in section 9.7.5. In the first few days after bombardment the major activity in the fermium fraction was 30-hour Fm^{252} produced by the $(\alpha,4n)$ reaction. At a later period the radiations of the longer-lived Fm^{253} became more prominent.

Fermium-253 has a half-life of 4.5 ± 1.0 days.[2] In 10 percent of its disintegrations Fm^{253} emits alpha particles with an energy of 6.94 ± 0.04 Mev.[2] In 90 percent of its disintegrations it decays by orbital electron capture to E^{253}. This latter decay is made eivdent by the growth of the 6.64 ± 0.3 Mev alpha particles of E^{253}. This is the longest-lived of the known isotopes offermium. There is a good likelihood that no longer-lived isotopes of fermium will be found.

References Fm^{253}

1. A. M. Friedman, J. E. Gindler, R. F. Barnes, R. Sjoblom and P. R. Fields, Phys. Rev. 102, 585 (1956).
2. S. Amiel, Phys. Rev. 105, 1412 (1957).

9.8.7 Fermium-254. The isotope, Fm^{254}, is observed to grow into a sample of E^{253} which has been irradiated with neutrons.[1,2] The sequence of reactions is:

$$\text{20-day } E^{253}\ (n,\gamma)\ E^{254m} \xrightarrow[38.5 \text{ hours}]{\beta^-} Fm^{254}.$$

The half life of Fm^{254} is 3.24 ± 0.0.1hours for the emission of alpha particles. Fermium-254 is beta stable. The energy of the alpha particles has been determined by the ion chamber method. Values of 7.17 ± 0.01 Mev,[1,3] 7.22 ± 0.03 Mev[2] and 7.20 ± 0.01 Mev[4] have been reported. In a freshly purified sample of einsteinium containing E^{254m}these 7.2 Mev alpha particles are observed to grow rapidly to a maximum and then to decay with the 38.5 hour half life of the einsteinium parent.

Fermium-254 is an even-even isotope and from the great regularity of the decay scheme of even-even alpha emitters in the heavy element region it may be expected that lower energy alpha-particle groups are present leading to a 2+ excited state of about 40-kev energy and a 4+ excited state at about 140 kev. ASARO AND CO-WORKERS[5] have studied the gamma radiations of Fm^{254} and found two gammas consistent with this interpretation. A gamma ray of 41 ± 2 kev energy was seen. From alpha-gamma and alpha-L x-ray measurements the intensity of the 2+ state was set at 17 ± 2 percent. The conversion coefficient was established for the 41 kev transition and found to agree with that expected from an E2 transition. A second gamma ray of 96 ± 2 kev energy eas seen in an intensity of 2.8×10^{-2} percent. This is assumed to correspond to an E2 transition

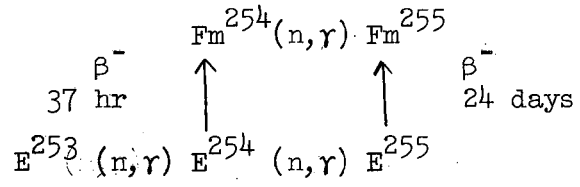
between a 4+ level at 138 kev and the 2+ level at 41 kev. This interpretation is reinforced by a study of the beta decay of Bk^{250} to the same daughter nucleus. More precise energies of 42.2 and 98.2 kev were determined from a study of that nuclide. The decay scheme of Fm^{254} is shown together with that of Bk^{250} in the figure presented in section 9.5.9. See figure 42.

Fermium-254 undergoes spontaneous fission at a high rate. Three values of the half-life for spontaneous fission have been reported: 200 days,[2] 220 ± 40 days[3] and 246 days.[4]

References Fm^{254}

1. M. H. Studier, P. R. Fields, H. Diamond, J. F. Mech, A. M. Friedman, P. A. Sellers, G. Pyle, C. M. Stevens, L. B. Magnusson and J. R. Huizenga, Phys. Rev. 93, 1428 (1954).
2. G. R. Choppin, S. G. Thompson, A. Ghiorso and B. G. Harvey, Phys. Rev. 93, 908 (1954); 93, 1129 (1954); 94, 1080 (1954).
3. P. R. Fields, M. H. Studier, J. F. Mech, H. Diamond, A. M. Friedman, L. B. Magnusson and J. R. Huizenga, Phys. Rev. 94, 209 (1954).
4. M. Jones, R. P. Schuman, J. P. Butler, G. Cowper, T. A. Eastwood and H. G. Jackson, Phys. Rev. 102, 203 (1956).
5. F. Asaro, F. S. Stephens, Jr., S. G. Thompson and I. Perlman, Phys. Rev. 98, 19 (1955) and later unpublished results, 1956.

9.8.8 Fermium-255. The discovery[1] of Fm^{255}, which constituted the discovery of element 100 is described in section 9.7.1. Fermium-255 can be produced by the prolonged neutron irradiation of E^{253} or of lighter isotopes such as Pu^{239} [2,3,4]. The complex chain of neutron capture and beta decay steps starting with Pu^{239} is discussed in chapter 5. Starting with E^{253} the synthesis of Fm^{255} occurs as follows:



The Fm^{255} has a half-life of only 21.5 ± 0.1 hours[4] but is supported by its longer lived E^{255} parent. It decays by the emission of alpha particles whose energy has been reported as 7.1 Mev,[1] 7.08 Mev[4] and 7.03 Mev.[5] Fermium-255 is

beta stable. Unpublished data of GHIORSO set a lower limit of 60 years on the half-life for spontaneous fission.

References Fm^{255}

1. A. Ghiorso, M. H. Studier, C. I. Browne, and co-workers, Phys. Rev. 99, 1048 (1955); Phys. Rev. 102, 180 (1956); Phys. Rev. 119, 2000 (1960).
2. G. R. Choppin, S. G. Thompson, A. Ghiorso and B. G. Harvey, Phys. Rev. 94, 1080 (1954).
3. M. H. Studier, P. R. Fields, H. Diamond, J. F. Mech, A. M. Friedman, P. A. Sellers, G. Pyle, C. M. Stevens, L. B. Magnusson and J. R. Huizenga, Phys. Rev. 93, 1428 (1954).
4. M. Jones, R. P. Schuman, J. P. Butler, G. Cowper, T. A. Eastwood and H. G. Jackson, Phys. Rev. 102, 203 (1956).
5. F. Asaro, F. S. Stephens, Jr., and I. Perlman, unpublished data (1957).

9.8.9 Fermium-256. Small samples of Fm^{256} have been prepared by the neutron irradiation of einsteinium fractions containing small amounts of E^{255}. The reaction sequence is:

$$E^{255}(n,\gamma)E^{256} \xrightarrow[\text{short}]{\beta^-} Fm^{256}.$$

Fermium-256 has also been formed by the electron-capture decay of Mv^{256} as discussed in the next section. Fermium-256 has been observed to decay by spontaneous fission with a half-life of 160 ± 10 minutes. The ratio of alpha decay to spontaneous fission is estimated to be 0.04 but the alpha decay (expected energy = 6.9 Mev) was not directly observed because of the presence of large amounts of Fm^{254} and Fm^{255} in the samples. Fermium-256 should be beta stable.

References Fm^{256}

1. G. R. Choppin, B. G. Harvey, S. G. Thompson and A. Ghiorso, Phys. Rev. 98, 1519 (1955).
2. L. Philipps, R. Gatti, A. Chesné, L. Muga and S. G. Thompson, unpublished information., Phys. Rev. Letters 1, 215, 1958,

9.9 THE ELEMENT MENDELEVIUM (ELEMENT 101)

By intense helium ion bombardment of tiny targets of E^{253}, GHIORSO, HARVEY, CHOPPIN, THOMPSON AND SEABORG[1] succeeded in preparing a few atoms with the chemical properties of element 101. The isotope so produced has the mass number 256:

$$_{99}E^{253}\ (\alpha,n)\ _{101}Mv^{256}$$

In this series of experiments the reaction products were collected by the recoil "catcher" foil technique discussed in section 6.7.5 which was developed for and used for the first time in this research. At the end of the bombardment the thin gold catcher foil was dissolved and the gold was removed by extraction into ethyl acetate and by use of an anion exchange resin. The remainder of the solution, containing element 101 and tracer amounts of Cf^{246} and E^{253}, was separated by adsorption of the radioactive atoms on a cation-exchange resin column and by elution of these atoms with the complexing agent, alpha-hydroxy-isobutyrate. The position of the californium and einsteinium elution peaks served to define the fraction containing elements 100 and 101. Atoms decaying by spontaneous fission with a half life of 3.5 hours were found in both fractions. These results were interpreted as follows: 101^{256} was produced in the nuclear reaction and separated chemically in the element 101 elution position; it then decayed by orbital electron capture with a half-life of the order of 30 minutes to produce Fm^{256} which decays by spontaneous fission with a half-life of 3.5 hours. The spontaneous fission events observed in the element 100 elution position represented atoms of Fm^{256} formed by the decay of 101^{256} before the chemical separation was complete. The proof that these experiments resulted in the identification of element 101 is the following:

1. Only the very heaviest elements decay by spontaneous fission with such short half-lives.

2. The elution of the new element from the ion-exchange column immediately ahead of element 100 shows that the chemical properties are those of an element heavier than fermium and probably 101 rather than 102.

3. By the method of preparation it would not be possible to produce an element above 101.

The name mendelevium, symbol Mv, was suggested for the new element in

recognition of the pioneering role of the great Russian chemist Dmitri Mendeleev, who was the first to use the periodic system of the elements to predict the chemical properties of undiscovered elements, a principle which was the key to the discovery of most of the transuranium elements.

In the original experiments a total of only 17 atoms of Mv^{256} were chemically isolated, an average of one atom per bombardment. This work was repeated by PHILLIPS AND CO-WORKERS[2] who produced several hundred atoms of Mv^{256} and identified them by the spontaneous fission of the Fm^{256} daughter. These workers deduced a half-life of ~1.5 hours for Mv^{256}.

PHILLIPS AND CO-WORKERS also found evidence for Mv^{255} produced by the reaction:

$$E^{253}\ (\alpha,2n)\ Mv^{255}.$$

The chief evidence was the isolation of a fermium daughter activity emitting 7.08 Mev alpha particles with a half-life of 21 hours. These are just the characteristics of Fm^{255}. It was deduced that Mv^{255} decays by capture of an orbital electron with a half-life of about 30 minutes. There was some evidence for the emission of 7.34 Mev alpha particles by Mv^{255}.

Mendelevium is the twelfth of the actinide series of elements and may be regarded as the homologue of thulium (element 69) of the lanthanide series. In its chemical properties it may be assumed to resemble the other tripositive actinide elements.

References - Mendelevium

1. A. Ghiorso, B. G. Harvey, G. R. Choppin, S. G. Thompson and G. T. Seaborg, Phys. Rev. 98, 1518 (1955).
2. L. Phillips, R. Gatti, A. Chesne, L. Muga and S. Thompson, Phys. Rev. Letters 1, 215 (1958).

9.10 ELEMENT 102

The first report of the identification of an isotope of element 102 was published in 1957. An international team of investigators including members from the Argonne National Laboratory of the United States, of the Atomic Energy Research Establishment in Harwell, England and the Nobel Institute for Physics in Sweden bombarded targets of curium in the 225 cm cyclotron at the Nobel Institute with high-energy carbon ions (C^{13}). The curium target had the composition 95 percent Cm^{244}, 1 percent Cm^{245} and 4 percent Cm^{246}. The reaction products which recoiled out of the curium target were caught on a tygon catcher foil which was ignited on a platinum plate to make a thin source suitable for alpha-particle analysis in an ionization chamber. In some experiments the tygon collector foil was dissolved and in one case the radioactivity was chemically separated by the same ion exchange-elution system previously used to separate and identify mendelevium. A few atoms decaying by the emission of 8.5± 0.1 Mev alpha particles with a half life of approximately 10 minutes were observed. In the one elution experiment in which such alpha particles were observed four atoms of this activity were observed in an elution position close to that expected for an element 102 fraction. It was concluded that the new activity was an isotope of element 102 produced most likely by one of these reactions:

$$Cm^{244} (C^{13}, 4n) 102^{253}$$
$$Cm^{244} (C^{13}, 6n) 102^{251}$$

The discovery of the new element 102 was claimed and the name nobelium, symbol No, was suggested in recognition of Alfred Nobel's support of scientific research and after the institute where the work was done.

This report was contested by a later publication of GHIORSO, SIKKELAND, WALTON AND SEABORG[2] who found they were unable to reproduce these results after scores of attempts under experimental conditions which were much more favorable than those confronting the original investigators. In particular GHIORSO AND CO-WORKERS[2] were able to use the high intensity beam of monoenergetic heavy ions delivered by a linear accelerator specially designed to accelerate heavy ions. Their report should be consulted for a statement of the reasons they were forced to conclude that the earlier claims to the discovery of element 102 were erroneous.

GHIORSO, SIKKELAND, WALTON AND SEABORG[3] then turned to problems of the positive identification of an isotope of element 102 using the hypothesis that the half-lives of those isotopes which could be made by bombardment of the available curium targets would be very much shorter than 10 minutes. A radically new method was developed based on the apparatus shown in the figure. The method was essentially a continuous milking experiment wherein the atoms of the daughter element 100 were separated from the parent element 102 by taking advantage of the recoil caused by the element 102 alpha particle decay. By this method the isotope, 102^{254}, having a half-life of 3 seconds for alpha decay to produce 30 minute Fm^{250} was unambiguously identified.

The target consisted of a mixture of isotopes of curium (95% Cm^{244} and 4.5% Cm^{246}) mounted on a very thin nickel foil. The target was approximately 0.5 mg/cm^2 thick and was covered with 75 $\mu gm/cm^2$ aluminum to prevent curium "knockover". The curium was bombarded with mono-energetic C^{12} ions at energies from 60 to 100 Mev. The transmuted atoms were knocked into helium gas to absorb the considerable recoil energy. It was found that with a sufficient electric field strength practically all of these positively charged atoms could be attracted to a moving negatively charged metallic belt placed directly beneath the target. These atoms would then be carried on this conveyer belt under a foil which was charged negatively relative to the belt. Approximately half of the atoms undergoing alpha decay would cause their daughter atoms to recoil from the surface of the belt to the catcher foil. The catcher foil was cut transversely to the direction of the belt motion into five equal length sections after a time of bombardment suited to the half-life of the daughter atom to be examined. The five foils were then alpha-pulse-analyzed simultaneously in a multiplex assembly consisting of five Frisch-grid ion-chambers, amplifiers, a single Wilkinson-type "kick-sorter", and a printer. With this equipment it was easily possible to make all the desired measurements for identifying the atoms caught on the catcher foils and thus to measure the half-life of the parent of the recoiling atoms. The method was first successfully used in bombardments of Pu^{240} with C^{12} ions to identify a new isotope of element 100, Fm^{248}. It was shown to have a half-life of 0.6 minutes by analysis of the amounts of the 20-minute Cf^{244} caught on the catcher foils.

Experiments were then started which were aimed at finding a short-lived isotope of element 102. The most likely isotope of element 102 that could be

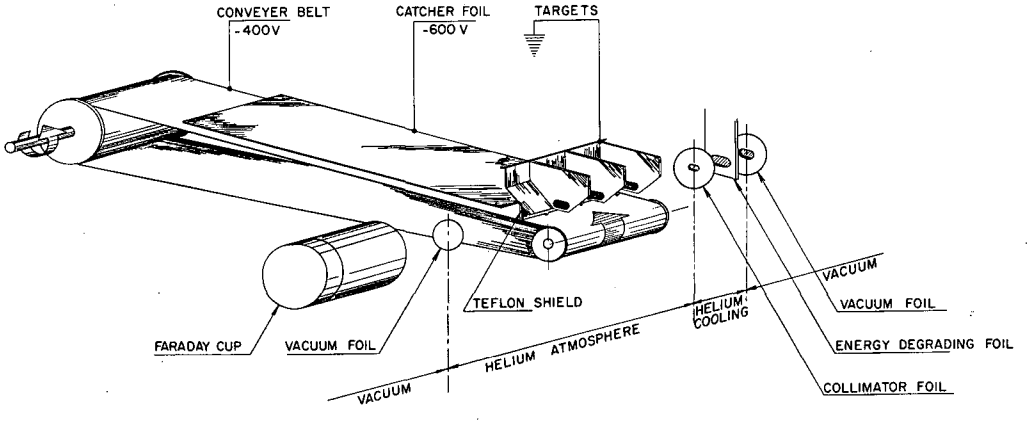


Figure 53. Schematic diagram of target assembly used by Ghiorso, Sikkeland, Walton, and Seaborg in the preparation and identification of 102^{254}. Reaction products recoiling out of the curium target were collected on the electrically-charged belt and transported from the region of the target. When the atoms of 102^{254} later decayed by alpha-particle emission a fraction of the Fm^{250} daughter atoms were ejected from the moving belt and collected on the charged catcher foil.

detected with this method was deemed to be 102^{254} with a predicted half-life of seconds leading to the 30-minute, 7.43-Mev, alpha-particle-emitter, Fm^{250}. In a series of experiments it was found that Fm^{250} could be collected on the catcher foils in relative amounts corresponding to a parent half-life of 3 seconds. The parent was produced in the reaction Cm^{246} (C^{12}, 4n) 102^{254}. The excitation function for producing Fm^{250} in this manner was found to peak sharply at 70 ± 5 Mev corresponding to a (C^{12},4n) reaction in accordance with a theoretical method for the calculation of (C,xn) reaction cross-sections.[4] Changing the belt speed was found to change the distribution of the Fm^{250} on the catcher foil in a manner conforming to a three-second parent. The number of Fm^{250} counts observed in a single experiment was as great as 40 and corresponded to a maximum cross-section of a few microbarns for the reaction with Cm^{246}.

The final identification of the activity ascribed to Fm^{250} was carried out by dissolving the activity from the catcher foil and separating it from the other actinide elements by elution with ammonium α-hydroxyisobutyrate from a column packed with Dowex-50 cation exchange resin. In one experiment 2 atoms of Fm^{250} were identified and in another 9 atoms were observed in the element 100 position.

From these results the authors concluded that there was no doubt that the existence of an alpha emitting isotope, 102^{254}, with a half-life of 3 seconds, had been demonstrated.

At the 1958 Geneva Conference G. N. Flerov reported on some experiments carried out in his laboratory in Moscow on the production of element 102. Thin targets of Pu^{241} were bombarded with ions of O^{16} accelerated to 100 Mev in a cyclotron. Atoms of element 102 were formed by the interaction of the oxygen ions with the plutonium. These atoms were ejected from the thin target and caught in an aluminum catcher foil. This collector foil was mechanically moved a distance of 200 cm from the target in a time of 2 seconds and placed in front of a nuclear emulsion. Some of the alpha particles from the activity in the catcher foils entered the emulsion and were recorded. Alpha particles of 8.5 ± 0.5 Mev were detected and attributed to an isotope of element 102 having a half-life much less than 4 minutes. The activity was given the probable mass assignment of 253 or 254. The possibility of the formation of an isotope of a lighter element emitting alpha particles with these characteristics was ruled out by many control experiments.

References - Element 102

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2. A. Ghiorso, T. Sikkeland, J. R. Walton and G. T. Seaborg, Phys. Rev. Letters Vol. 1, p.17, July 1958.
3. A. Ghiorso, T. Sikkeland, J. R. Walton, and G. T. Seaborg, Phys. Rev. Letters Vol. 1, p. 18, July 1958.
4. T. Sikkeland, S. G. Thompson and A. Ghiorso, Phys. Rev. 112, 543, (1958).
5. G. N. Flerov, Paper P/2299, Volume 15, Proceedings of the Second U.N. Conference on the Peaceful Uses of Atomic Energy, Geneva 1958.

9.11 GENETIC RELATIONSHIPS OF TRANSURANIUM NUCLIDES

For some purposes it is useful to trace out the genetic relationships of the nuclides throughout the entire transuranium group of nuclides. We present here four summary diagrams (figures 54-57) which permit one to see at a glance these family relationships for the four main series of nuclides; namely for the 4n, 4n+1, 4n+2, and 4n+3 series. It will be recalled that the term 4n signifies that the mass number is evenly divisible by 4, while in the other cases the mass number is divisible by 4 with the remainders 1, 2, and 3, respectively. These figures can be regarded as an extension to higher mass numbers of the thorium series, the artificial 4n+1 series beginning with U^{233}, the uranium-radium series, and the actinouranium series.

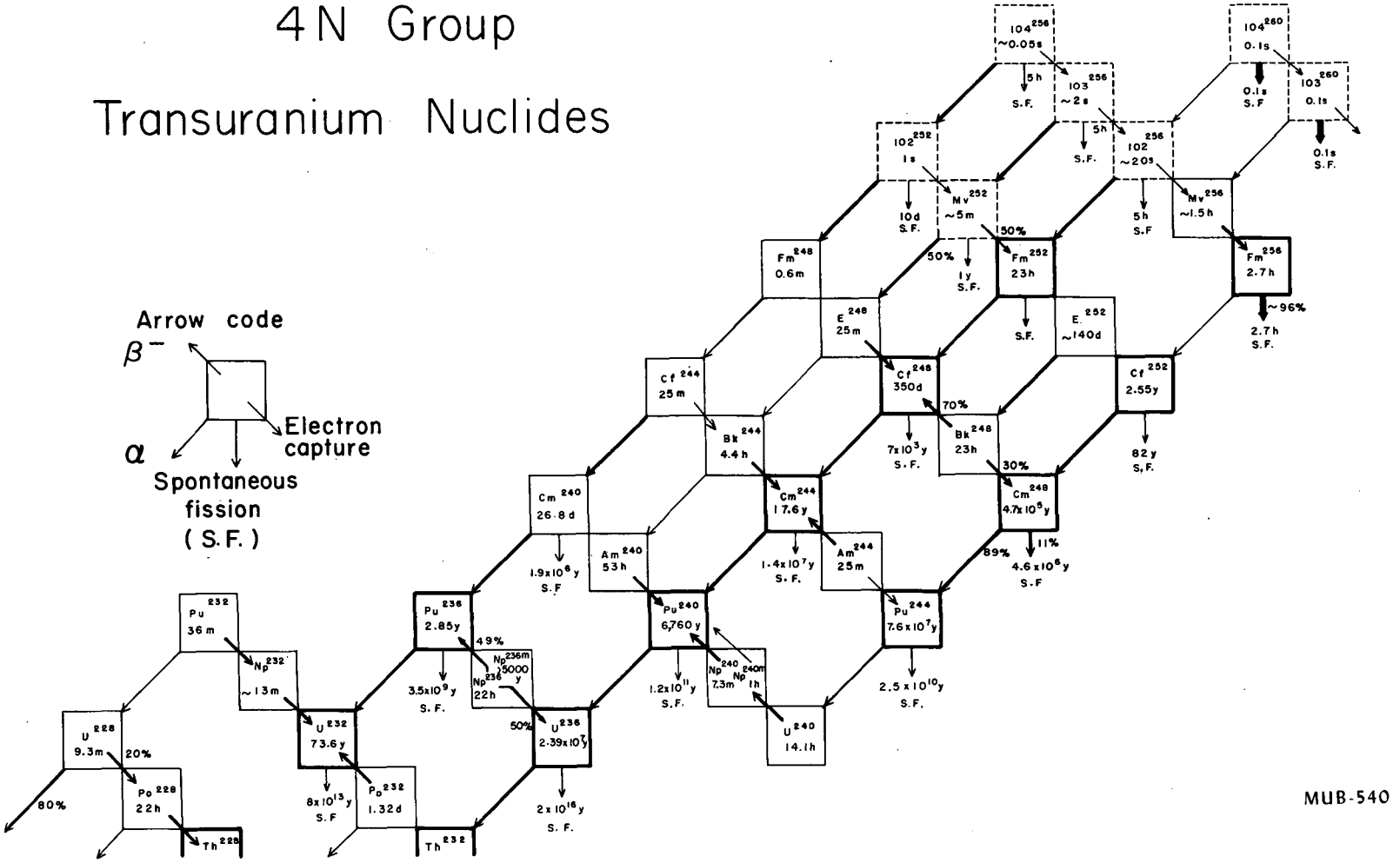

Figure 54. Genetic relationships of the 4n group of transuranium nuclides. Decay modes of each nuclide are indicated by arrow code. A heavy arrow indicates 10 percent or greater branching by that decay mode. A light arrow indicates slight branching. Heavily outlined squares indicate beta stability. Only experimentally measured decay modes are shown except in the case of nuclides whose squares are outlined in broken lines. In these latter cases all indicated decay modes and half lives are predictions.

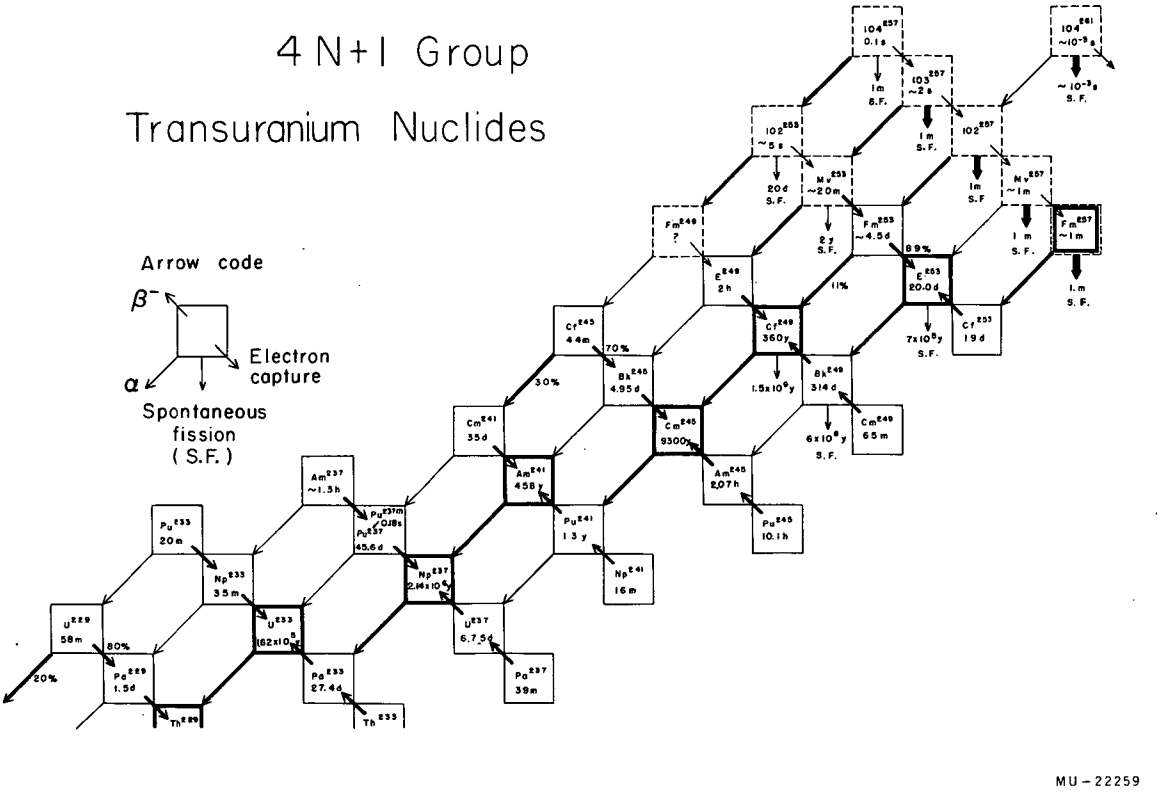

Figure 55. Genetic relationships of the 4n+1 group of transuranium nuclides. Decay modes of each nuclide are indicated by arrow code. A heavy arrow indicates 10 percent or greater branching by that decay mode. A light arrow indicates slight branching. Heavily outlined squares indicate beta stability. Only experimentally measured decay modes are shown except in the case of nuclides whose squares are outlined in broken lines. In these latter cases all indicated decay modes and half lives are predictions.

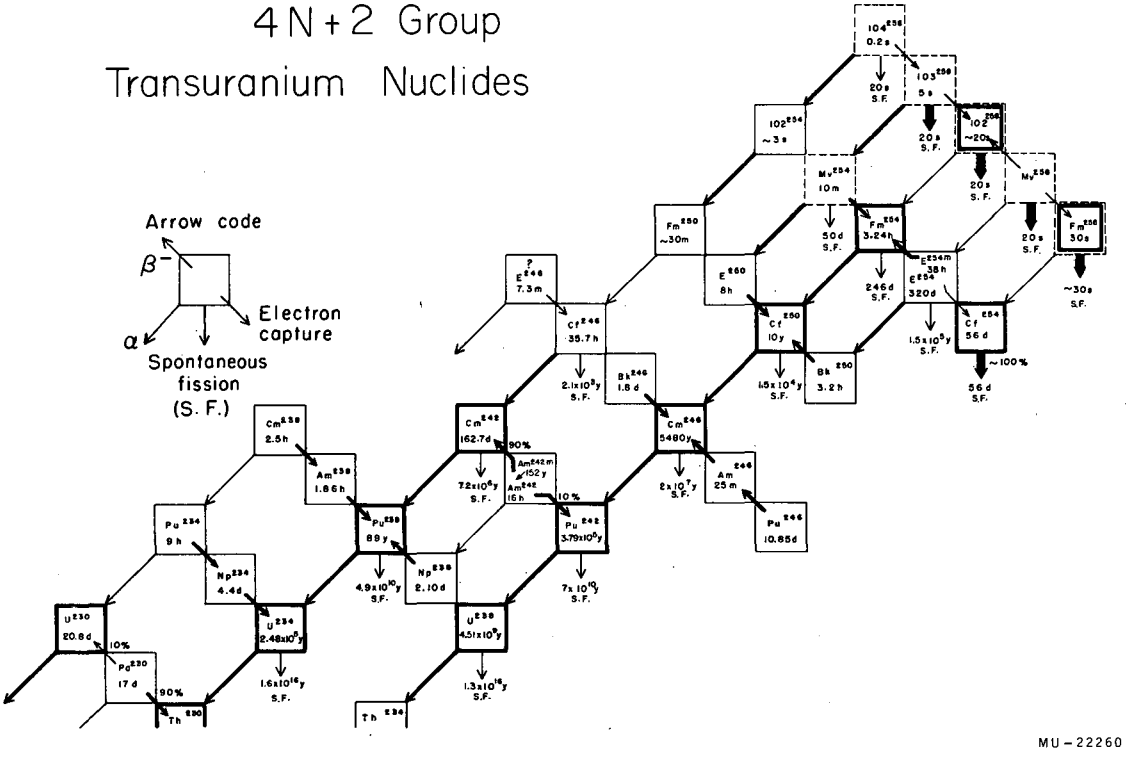

Figure 56. Genetic relationships of the 4n+2 group of transuranium nuclides. Decay modes of each nuclide are indicated by arrow code. A heavy arrow indicates 10 percent or greater branching by that decay mode. A light arrow indicates slight branching. Heavily outlined squares indicate beta stability. Only experimentally measured decay modes are shown except in the case of nuclides whose squares are outlined in broken lines. In these latter cases all indicated decay modes and half lives are predictions.

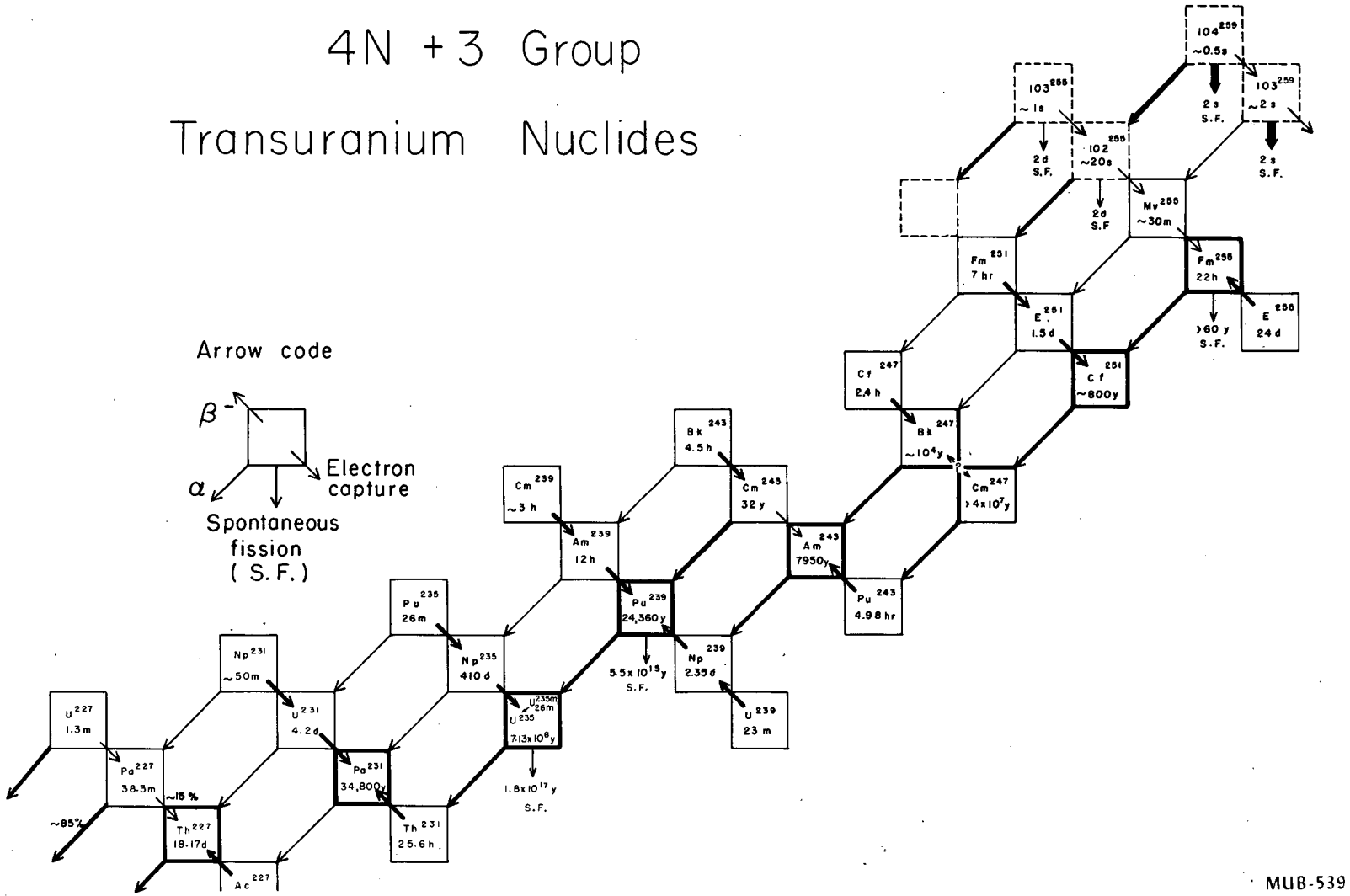

Figure 57. Genetic relationships of the 4n+3 group of transuranium nuclides. Decay modes of each nuclide are indicated by arrow code. A heavy arrow indicates 10 percent or greater branching by that decay mode. A light arrow indicates slight branching. Heavily outlined squares indicate beta stability. Only experimentally measured decay modes are shown except in the case of nuclides whose squares are outlined in broken lines. In these latter cases all indicated decay modes and half lives are predictions.

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