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NUCLEAR PROPERTIES OF TRANSPLUTONIUM ISOTOPES

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Introduction

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Several isotopes of transplutonium elements, all radioactive, are known. This chapter is concerned with the radioactive properties of these isotopes and the ways in which they are produced. At the present time there are two transplutonium elements known, the element with atomic number 95 (americium) and the element with atomic number 96 (curium).

A number of matters of interest in connection with this chapter have been discussed in Chapter II which is devoted to the nuclear properties of the isotopes of plutonium, and to which reference should be made. Also for a discussion of the chemical and instrumental techniques used in the study of the radioactive and nuclear properties of transplutonium isotopes, Chapters XVI and XVIII of this Volume should be consulted.

Table 1 summarizes the radioactive properties of the transplutonium isotopes.

1. Americium

1.1 Am²⁴¹. The most important isotope of americium is the one of mass 241. As a result of work on the ultramicrochemical scale of investigation, this isotope is available in weighable quantities and its chemical properties have been studied by working with weighable amounts of material.

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29
17M

TABLE 1

Radioactive Properties of Transplutonium Isotopes

Atomic Number	Mass Number	Half-Life	Mode of Disintegration, Radiations, Energies	Modes of Formation
95-Am	238	50 hr.	K, γ (~ 1.4 Mev), e^-	Pu-d-3n Np- α -3n Pu- α -5n
	239	12 hr.	K ($\sim 99.9\%$), γ (0.285 Mev) α ($\sim 0.1\%$)	Pu-d-2n Np- α -2n
	241	510 ± 20 yr.	α , 5.45 Mev (4.05 cm.)	Pu- $^{241}\beta$ decay
	242	16 ± 3 hr.	β^- , 1.0 ± 0.3 Mev	Am-n- γ
	242	~ 600 yr.*	β^- ($\sim 99.8\%$), ~ 0.5 Mev α ($\sim 0.2\%$)	Am-n- γ
96-Cm	240	26.8 ± 0.3 days	α , 6.3 Mev (4.95 cm.) K	Pu- α -3n
	241	~ 55 days	K	Pu- α -2n
	242	162 ± 3 days	α , 6.1 Mev (4.75 cm.)	Pu- α -n Am- 242 (16 hr.) β^- decay

* On basis of entirely arbitrary assumption that cross section for formation from Am-n- γ reaction is 550 barns; actually, recent rough measurements indicate 400 barns for this cross section corresponding to a half-life of about 400 years.

The element americium was discovered in late 1944 and early 1945 at the Metallurgical Laboratory of the University of Chicago by G. T. Seaborg, R. A. James, and L. O. Morgan.⁽¹⁾ These investigators found that the isotope Am²⁴¹ decays with the emission of alpha-particles which have a range of 4.05 ± 0.1 cm. (energy 5.45 ± 0.05 Mev).

The isotope Am²⁴¹ also emits gamma-rays⁽¹⁾ of energy about 62 Kev and in the amount of some 0.6 per alpha-particle. There are also low

27³⁷

energy x-rays, in the range 10 to 20 Kev and hence consistent with the view that they are the L x-rays of a heavy element such as neptunium. These radiations are probably best explained by assuming that most or all of the alpha-particles are followed by the emission of a partially converted (in the L electron shell) gamma-ray with an energy of about 62 Kev.

The isotope Am^{241} is produced as the result of the bombardment of U^{238} with 4.0 Mev helium ions according to the reactions $\text{U}^{238}(\alpha, n)\text{Pu}^{241}$, $\text{Pu}^{241} \xrightarrow{\beta^-} \text{Am}^{241}$. (1) Although the bombardment of Pu^{239} with helium ions of various energies has been performed, production of Am^{241} by the reaction $\text{Pu}^{239}(\alpha, pn)\text{Am}^{241}$ has not been directly established in these relatively short bombardments due to the long half-life of Am^{241} . (1)

The naming of element 95 was done on the basis of its chemical properties. Chemical and other evidence indicate that the heavy elements form a new "rare earth" series in which the 5f electron shell is being filled. This series of "synthetic earths" which might be called the "actinide" series on the same basis as the rare earths are referred to as the "lanthanide" series, is discussed in detail in Chapter XXI of this volume. On this basis americium contains six 5f electrons and is analogous to the element europium which contains six 4f electrons, and the name comes from the Americas in a manner analogous to the naming of europium after Europe. (1) The chemical symbol is Am.

The first pure americium in the form of the isotope Am^{241} was isolated early in 1946 by B. B. Cunningham. (2) Since this first isolation a number of samples of the pure isotope have been isolated and the spe-

cific alpha-activity has been determined by direct weighing, leading to a value for the half-life of about 510 ± 20 years.⁽²⁾

1.2 16-hour Am^{242} . The neutron irradiation of Am^{241} produces Am^{242} as a result of the reaction $\text{Am}^{241}(n, \gamma)\text{Am}^{242}$. The half-life of Am^{242} was determined as 16 ± 3 hours in the work of W. M. Maming and L. B. Asprey⁽³⁾ who irradiated a sample of pure Am^{241} with neutrons in the Argonne heavy water-uranium pile. These investigators found beta-particles due to Am^{242} , with an upper energy limit of about 1.0 Mev decaying with a half-life of 16 hours and they found that alpha-particles of 4.75 cm. range, due to Cm^{242} (see below) grew in at the rate expected for this half-life. The investigation included chemical work which proved that the 16-hour activity was an isotope of americium and the assignment of this activity to Am^{242} seems certain.

1.3 Long-lived Am^{242} . It has been shown⁽¹⁾ that an intense and lengthy slow neutron irradiation of Am^{241} leads to a long-lived isomeric form of Am^{242} . This was first shown conclusively in the summer of 1947 in experiments with several months old neutron irradiated Am^{241} in which the growth of the 2-day Np^{238} was demonstrated, thus giving indirect evidence for the presence of a long-lived alpha-particle-emitting Am^{242} . It was shown that the Np^{238} could be repeatedly "milked" from old samples of americium which had received strong irradiations with slow neutrons.

Following these initial observations the same investigators made a careful search, by means of the absorption method, for beta-particles in these samples in order to establish whether this long-lived Am^{242} might also decay to the 5-months Cm^{242} in a manner similar to its 16-hour

47^m 39

isomer. The presence of such beta-particles was demonstrated and the energy could be roughly determined as about 0.5 Mev by the experiments in which their absorption in aluminum was measured.

The half-lives for the two modes of decay of the long-lived Am^{242} cannot be calculated with certainty because the n, γ cross section for the formation of this isomer from Am^{241} is not well known. If it is assumed, entirely for the sake of discussion and with no thought that this need be the case, that the cross section for the formation of the long-lived Am^{242} is the same as that for the 16-hour isomer, the half-life for alpha-particle emission is calculated to be approximately 3×10^5 years on the basis of the observed Np^{238} activity and the half-life for beta-particle emission is calculated to be about 600 years on the basis of the directly observed beta-particles. The observed branching ratio is 99.8% beta-particles and about 0.2% alpha-particles in the decay of this long-lived isomer.

1.4 Am^{239} . An early attempt to produce isotopes of mass less than 241 was made by irradiating the isotope Np^{237} with helium ions. (4) Although some evidence was obtained it was not conclusive.

Later experiments, performed largely during the first half of 1947, show that the irradiation of Pu^{239} with 20-Mev deuterons and of Np^{237} with 40-Mev helium ions leads to a 12-hour radioactivity (1) which can now be assigned with reasonable certainty to the isotope Am^{239} . The nuclear reactions are $\text{Pu}^{239}(\text{d}, 2\text{n})\text{Am}^{239}$ and $\text{Np}^{237}(\alpha, 2\text{n})\text{Am}^{239}$. (The same irradiations also produce 50-hour Am^{238} which will be discussed in the next section.) This isotope undergoes branching decay, about 99.9% by orbital electron capture and about 0.1% by alpha-particle emission

39
5m

(corresponding to a partial half-life of about 500 days for the latter process.) The orbital electron capture decay is accompanied by partially converted gamma-rays of energy about 0.285 Mev, and K and L x-rays each of about equal intensity and each of about twice the intensity of the unconverted gamma-rays. The energy of the alpha-particles has not yet been measured and the growth of the daughter 400-day Np^{235} has not yet been observed due to its relatively long half-life and the relatively small intensities of Am^{239} which can be conveniently produced in the cyclotron irradiations. The growth of the daughter Pu^{239} has not been observed for the same reasons. Since another americium isotope, the 50-hour orbital electron capture Am^{238} , is produced at the same time, all of the above mentioned radiation characteristics had to be determined in the presence of this other isotope.

Since it hasn't been possible to observe the growth of the daughter activities the isotopic assignment of this 12-hour activity (and also the 50-hour activity to be discussed in the next section) had to be made by indirect means. It was necessary to use a combination of arguments involving the yields of these isotopes and the variations of these yields with change in energy of the bombarding deuterons and helium ions. However, such experiments have led to rather definite results, so it is felt that the isotopic assignment is fairly certain.

1.5 Am^{238} . Experiments performed during the first half of 1947 show that the irradiation of Pu^{239} with 20-Mev deuterons and of Np^{237} with 40-Mev helium ions produces a 50-hour activity⁽¹⁾ which must be assigned to the isotope Am^{238} . The corresponding nuclear reactions are

67m³⁷

$\text{Pu}^{239}(\text{d}, 3\text{n})\text{Am}^{238}$ and $\text{Np}^{237}(\alpha, 3\text{n})\text{Am}^{238}$. This isotope has also been produced in the helium ion irradiation of Pu^{239} , probably according to the reaction $\text{Pu}^{239}(\alpha, \text{pn})\text{Am}^{238}$.

This isotope decays by the orbital electron capture mechanism accompanied by the emission of gamma-rays with energy about 1.3 to 1.4 Mev, converted to the extent of about 5%. The corresponding K and L x-rays have also been observed and the rough measurements show that the intensities of each of these x-rays and of the gamma-rays are about equal.

Due to the relatively long half-life of the daughter Pu^{238} and the relatively small intensities which could conveniently be prepared in cyclotron irradiations it has not yet been possible to observe the growth of the daughter Pu^{238} . As mentioned in the previous section the isotopic assignment to the mass 238 has been made on the basis of yield considerations taken together with the variation of the yield with the energy of the deuterons and helium ions. These experiments have given rather good results and the isotopic assignment is considered to be fairly certain. Further, it may be pointed out that the assignment of the low energy orbital electron capturing 12-hour activity to Am^{239} and the much higher energy orbital electron capturing activity to Am^{238} is consistent with expectations from the mass types (that is, odd-even and odd-odd character) of these two isotopes.

1.6 Other Americium Isotopes. There have as yet been no observations which would indicate that the radiations from the isotope Am^{240} have been observed and the only evidence which we have concerning this isotope is very indirect. There are some indications that the isotope Cm^{240} (to be discussed in a later section) decays by the orbital electron

capture mechanism as well as by the emission of alpha-particles.⁽⁵⁾ This would indicate that Am^{240} is unstable toward decay by the orbital electron capture process, in addition, of course, to its instability toward alpha-particle emission. Since no irradiation which can be ascribed to Am^{240} has been observed, although this isotope would have been produced in the deuteron irradiation of Pu^{239} and the helium ion irradiation of Np^{237} , it follows that it must be either quite short or quite long-lived. Considerations involving the general regularities as to nuclear stability indicate that it is very probably long-lived.

The americium isotopes with mass less than 238 are all probably quite short-lived, decaying by the orbital electron capture or alpha-particle emission process or both.

It does not seem possible to predict with certainty whether the isotope Am^{243} will be beta-stable although careful consideration of the stability curve seems to indicate that this should be the case. The indicated instability of Am^{240} toward decay by orbital electron capture together with the extremely small energy of the Pu^{241} beta-particles (about 10 Kev⁽¹⁾) certainly points toward a beta-stable Am^{243} .

There seems to be no doubt that the isotopes of mass greater than Am^{243} will all be unstable toward decay by negative beta-particle emission.

2. Curium

2.1 Cm^{242} . The element curium was discovered in the summer of 1944 at the Metallurgical Laboratory of the University of Chicago by Seaborg, James and A. Ghiorso⁽⁵⁾ who found that the alpha-emitting isotope Cm^{242} was produced from the reaction $\text{Pu}^{239}(\alpha, n)\text{Cm}^{242}$ as the result of the

37
8 m

bombardment of plutonium with 36-Mev helium ions in the Berkeley cyclotron. It is likely, of course, that this isotope is also produced by the reactions $\text{Pu}^{239}(\alpha, p)\text{Am}^{242}$ and $\text{Am}^{242} \xrightarrow{\beta^-} \text{Cm}^{242}$ in this bombardment. The radioactivity formed in this manner decays with the emission of alpha-particles of 4.75 ± 0.1 cm. range (energy 6.1 Mev) and with a half-life of 5.0 ± 0.1 months. The isotope Cm^{242} also emits a small number of 65-Kev gamma-rays (about one per 500 alpha-particles) and a small number of conversion electrons and the corresponding L x-rays. The isotopic assignment of this radioactivity has been confirmed as the result of the proof that the isotope Pu^{238} is its decay product.⁽⁵⁾ The latest decay measurements of James indicate that 162 ± 3 days is a better value for the half-life.

As mentioned above the isotope Cm^{242} is also formed as the result of the strong neutron irradiation of Am^{241} . The Am^{241} absorbs neutrons to form the 16-hour, beta-particle emitter, Am^{242} , which decays to Cm^{242} . Using this method of production, L. B. Werner and I. Perlman,⁽⁶⁾ after irradiating a sample of Am^{241} with the neutrons in one of the Hanford piles for a period of 15 months, isolated the element curium in the form of the isotope Cm^{242} , in weighable amounts and studied its chemical properties using the methods of ultramicrochemistry.

This element was given its name on the same basis as that used for the naming of americium. Since curium contains seven 5f electrons it was named after Pierre and Marie Curie in a manner analogous to the naming of gadolinium with its seven 4f electrons which honors Gadolin, the Finnish rare earth chemist.

2.2 Cm^{240} . A second isotope of curium was discovered early in 1945⁽⁵⁾ as the result of the bombardment of Pu^{239} with higher energy helium

97m 37

ions, namely, 14 Mev rather than 36 Mev. In addition to the production of Cm^{242} under these conditions, the isotope Cm^{240} is produced by the reaction $\text{Pu}^{239}(\alpha, 3n)\text{Cm}^{240}$.⁽⁵⁾ This isotope decays with a half-life of 26.8 ± 0.3 days by the emission of alpha-particles with a range of 4.95 ± 0.1 cm. (energy 6.3 Mev). There is some evidence that the decay of this isotope is accompanied by the L and K x-rays of a heavy element indicating, in addition to the alpha-particle emission, a branching decay by the orbital electron capture mechanism.⁽⁵⁾ An attempt has been made, which can be adjudged as very rough at best, to compare the number of x-rays with the number of alpha-particles and hence to obtain a value for the branching ratio; such a rough comparison yields the preliminary value of about 5 times as many x-rays as alpha-particles, indicating that Cm^{240} decays mainly by orbital electron capture.⁽⁷⁾

The isotopic assignment of this activity to Cm^{240} has been checked by the growth as a daughter of the isotope Pu^{236} whose radioactive properties have been definitely established.⁽⁸⁾

2.3 Cm^{241} . The irradiation of Pu^{239} with 14-Mev helium ions also leads to the production of an x-ray-emitting radioactivity, which decays with a half-life of about 55 days, and which may possibly be ascribed to the isotope Cm^{241} produced by the reaction $\text{Pu}^{239}(\alpha, 2n)\text{Cm}^{241}$. The x-rays indicate decay by the orbital electron capture mechanism. There may also be some electrons associated with this activity but no hard gamma-rays are observed. It should be noted that this isotopic assignment is very tentative.

2.4 Other Curium Isotopes. Very little is known about curium isotopes other than those discussed above. An irradiation of Pu^{239} with

10m³⁷

40-Mev helium ions, with a subsequent fast americium-curium chemical separation so as to obtain a pure curium fraction, showed that Am^{238} and Am^{239} did not grow in the curium fraction, thus setting an upper limit of about ten years for the decay of Cm^{238} and Cm^{239} by the orbital electron capture process (9) on the assumption that Am^{238} and Am^{239} are sufficiently long-lived to have been detected. It seems likely that all of the isotopes of curium with mass less than 240 will be unstable toward decay by orbital electron capture and that isotopes which decay by this mechanism as well as by alpha-emission will be found in this region. The isotope Cm^{243} could very likely be unstable toward decay by orbital electron capture if Am^{243} is beta-stable which is indicated to be possible above. The isotopes Cm^{244} and Cm^{246} should be beta-stable while it is not so certain whether or not this is the case for Cm^{245} . The isotope Cm^{247} should be unstable toward decay by negative beta-particle emission, leading to an isotope of the element with atomic number 97, namely, the isotope 97^{247} .

117m

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