Transplutonium Elements in Thermonuclear Test Debris*

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The isotopes of curium, berkelium, and californium found in the thermonuclear debris of the November, 1952 thermonuclear test are discussed. The instantaneous buildup of the heavy elements in the thermonuclear device is compared with the buildup during pile irradiation. The alpha-particle energy (5.4 Mev) and the spontaneous fission half-life ($<1.2\times10^7$ years) of Cm^{246} are reported. The spontaneous fission half-life of Cf^{254} was found to be 55 days. No other mode of decay was observed for this isotope.

ONE of the very interesting results of the November, 1952 thermonuclear test was the discovery of elements 99 and 100, einsteinium and fermium,[1] in the resulting debris. In addition to these elements many new isotopes of plutonium, americium, curium, berkelium, and californium were produced and isolated. Many of these nuclides were made later by intense neutron irradiations of plutonium in the Materials Testing Reactor (MTR).[2–6] It is the purpose of this communication to report the properties of the nuclides formed in the thermonuclear test which have not been measured in reactor irradiation products. The discovery of Pu^{244}, Pu^{246}, and Am^{246} in thermonuclear debris is reported elsewhere.[7]

The thermonuclear and the MTR experiments were similar to each other in that, in each case, neutron-excess nuclides were formed which decayed by beta emission. However, they differed in the time scale. In the thermonuclear experiment, the neutron irradiation was essentially instantaneous in that the uranium was subjected to an intense neutron irradiation of such short duration that the absorption process was complete before beta emission could take place. Thus uranium isotopes of mass number of at least 255 were formed. Each of these high-mass uranium isotopes then decayed to isotopes of higher atomic number until the first beta-stable nuclide of each mass number was formed. Figure 1 shows the nuclides which were first characterized in the thermonuclear experiment. Some nuclides (e.g., Cf^{250}, E^{254}, and Fm^{254}) were shielded by beta-stable ones and were not found in the original thermonuclear debris but were found in the MTR irradiations because the longer time scale in the latter allowed some intervening beta decays to take place between successive neutron captures as shown in Fig. 1. On the other hand, activities which have been assigned to Cf^{254}, E^{255}, and Fm^{255} were present in relatively smaller amounts in the MTR material, apparently because of unfavorable capture and/or fission cross sections for thermal neutrons, and because of competition of radioactive decay with neutron capture.

The specific results are given below:

Curium.—Curium samples of sufficient size to permit mass spectrometric analyses were chemically separated from the thermonuclear debris. The isotopic compositions of the curium samples were measured in the Argonne 12-inch, 60° mass spectrometer with a multiple filament surface ionization source.[8] The curium was found to contain, in addition to the previously known

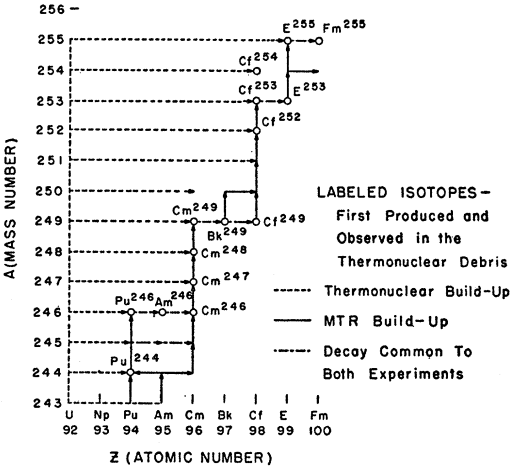

FIG. 1. The processes of isotope build up in thermonuclear test and Materials Testing Reactor.

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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Cm^{245},[9] the isotopes Cm^{246}, Cm^{247}, and Cm^{248}, in the mole percentages given in column 2 (sample I) of Table I. This was the first identification of Cm^{246}, Cm^{247}, and Cm^{248}. The isotopes Cm^{246} and Cm^{247} were later identified among the products of the neutron irradiation of plutonium in the MTR,[2] but Cm^{248} has not as yet been detected in such material.

A fraction of sample I was irradiated in the MTR for six weeks (total flux 9×10^{20} neutrons). The isotopic composition of this curium is given in column 3 of Table I (sample II). The size of sample II was too small to permit detection of curium masses higher than 247.

Alpha-pulse analysis of curium sample I showed a rather broad alpha peak with energy of about 5.4 Mev due to Cm^{245} and Cm^{246}. The ratios of the 5.4-Mev alpha particles (Cm^{245} and Cm^{246}) to spontaneous fission disintegrations were $(5.8\pm0.3)\times10^3$ and $(2.7_4\pm0.1_4)\times10^3$, respectively, for curium samples I and II. The percentages of Cm^{245} and Cm^{246} alpha particles in the 5.4-Mev alpha peak of sample I, calculated from the mass spectrometric data and known Cm^{245} and Cm^{246} alpha half-lives[10] of $(1.15\pm0.05)\times10^4$ and $(4.0\pm0.6)\times10^3$ years, were (46_{-15}^{+10}) and (54_{-10}^{+15}) percent, respectively. Similarly, in sample II the percentages of Cm^{245} and Cm^{246} alpha particles were calculated to be 9 ± 4 and $91\pm3\%$, respectively. By employing the foregoing data, the ratios of the Cm^{246} alpha particles to spontaneous fission disintegrations (spontaneous fission events may presumably be associated with even-even isotopes Cm^{246}, Cm^{248}, etc.) for curium samples I and II were calculated to be 3.1×10^3 and 2.5×10^3, respectively. The smaller ratio for sample II indicates that Cm^{248} is making a greater contribution to the spontaneous fissions in this sample. From sample I, a lower limit of $1.2_4\times10^7$ years could be placed on the spontaneous fission half-life of Cm^{246}.

TABLE I. Mass spectrometric analysis of two curium samples. (Isotopic abundances given in mole percent.)

Cm isotope	Sample I	Sample II (Sample I irradiated in MTR for six weeks)
245	68.7±0.4	20.7±0.5
246	28.4±0.4	75.1±1.0
247	2.2±0.1	4.2±1.0
248	0.7±0.2	a
(249)	$0.5\pm^{0.2}_{0.5}$	a

[a] Insufficient quantity of sample II for detection of these mass numbers

A thermal-neutron fission cross section of 1800±300 barns was calculated for Cm^{245} based on the fissionability of a portion of sample I. This is in agreement with the value of 2000±150 barns derived from curium produced in the MTR.[2] The small contribution of Cm^{247} to the total fissionability of the curium sample gave rise to a negligible correction.

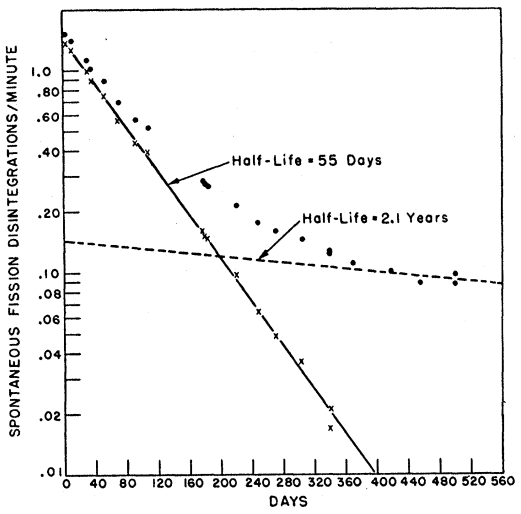

FIG. 2. Spontaneous fission decay curve of californium fraction from thermonuclear debris.

The isotope Cm^{249} was made by MTR neutron bombardment of a curium fraction from the debris and found to have a half-life of 65 minutes and a beta energy of 0.9 Mev.

The absence of Cf^{250} from the thermonuclear debris suggests that Cm^{250} is either beta-stable or has a half-life longer than 130 years.

Berkelium.—The isotope Bk^{249} was isolated for the first time from the debris and its nuclear properties and mass number were determined. The nuclear properties of this isotope have previously been reported elsewhere.[3,5]

Californium.—The isotopes Cf^{249}, Cf^{252}, Cf^{253}, and Cf^{254} were likewise characterized for the first time. The Cf^{249} was observed to grow in Bk^{249} samples. As mentioned above, no Cf^{250} could be found, probably because Cm^{250} is beta stable. The properties of Cf^{249}, Cf^{252}, and Cf^{253} have been reported elsewhere.[3–6]

Figure 2 shows the spontaneous fission decay curve of a californium fraction isolated from the thermonuclear debris. The decay curve for the spontaneous fission activity was resolved into a 55-day component and a 2.1-year component. The 2.1-year component was due to Cf^{252}, and the 55-day component was assigned to Cf^{254} on the basis of yields and decay systematics. It was concluded that Cf^{254} decayed predominantly by spontaneous fission since neither its alpha particles were detected nor those of daughters formed by beta decay. The mass assignment of 254 was later confirmed by observing the growth of Cf^{254} from electron-capture decay of E^{254}, made in the MTR work.[11]

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New Isotope of Berkelium*

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A new isotope of berkelium, Bk^{248}, formed by a 25-Mev helium-ion bombardment of a mixture of curium nuclides has been found. Its existence and a half-life of 23±5 hours were determined by periodically separating the β^- decay daughter, Cf^{248}, from the mixture of berkelium isotopes isolated after the bombardment.

FURTHER bombardments of curium isotopes of mass 244 and greater with 25-Mev helium ions have resulted in the formation of Bk^{248}, hitherto an unknown isotope.

The target was made from 800 micrograms of Cm^{244} which contained a few percent each of Cm^{245} and Cm^{246}, the mixture being produced from a seventeen-month neutron irradiation of Pu^{238} and Pu^{242}. After bombarding the curium with helium ions for ~700 microampere-hours, the berkelium fraction was separated from all curium, californium, and fission products by precipitation and ion-exchange methods.[1] Direct measurement of the decay of Bk^{248} was impractical as all observable radioactivity in the berkelium fraction was from the 4.95-day Bk^{245}.[2] However, as calculations from closed decay cycles have indicated that Bk^{248} would have considerable energy available for β^- decay as well as electron capture,[3] a californium fraction was periodically separated from the purified berkelium to observe the growth of Cf^{248}, a 6.26-Mev alpha emitter.[4]

Four such "milks" were performed within an eight-day interval, and Cf^{248} (~2 dis/min) and a much smaller amount of Cf^{249} from the β^- decay[5] of Bk^{249} were found to have grown into the berkelium sample between each separation. The existence of Cf^{248} and the amount formed were determined by alpha-particle pulse analysis. A 23±5 hour half-life was calculated for Bk^{248} from the amount of Cf^{248} grown, the time intervals associated with the growths and decays, and estimated chemical yields. The value for the half-life of Bk^{248} appears quite reasonable when its calculated disintegration energy[3] (1.45 Mev) is compared with that of the 1.8-day Bk^{246} (1.40 Mev),[3] a similar odd-odd nuclei. No value could be obtained for a possible β^-/electron capture branching ratio since alpha particles from the electron capture daughter, Cm^{248}, could not be observed. Because the mass assignment of Cf^{248} has been proven,[4] the mass assignment of Bk^{248} is certain.

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* This work was performed under the auspices of the U. S. Atomic Energy Commission.

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