# Heavy Isotope Abundances in Mike Thermonuclear Device*

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The November 1, 1952 thermonuclear explosion ("Mike") produced all of the uranium isotopes $U^{239}$, $U^{240}$, $\cdots U^{255}$ through multiple neutron capture by $U^{238}$. The long-lived products of successive $\beta^-$ decays from these isotopes were measured mass spectrometrically and radiometrically. The logarithms of the abundances decline smoothly with increasing mass number; the even-mass abundances slightly exceed the geometric mean of adjacent odd-mass abundances. Some nuclear properties of neutron-rich heavy nuclides, not subject to ordinary investigation are inferred.

## I. INTRODUCTION

ON November 1, 1952, the first large scale thermonuclear test explosion (Ivy Mike) took place in the Pacific Ocean. The device involved both the production of 14-Mev neutrons through deuterium-tritium fusion, and neutrons from the fission of uranium. The unusually high neutron flux of the device produced $U^{239}$, $U^{240}\cdots U^{255}$. These isotopes then decayed by beta emission to more stable isobars (Fig. 1). The device produced much of the earliest information about neutron-rich nuclides whose mass numbers exceed 243.[1–3] The abundances of the 17 successive uranium isotopes are of interest because of the insight into nuclear properties that may be obtained and because the nuclear reactions in the explosion constitute an analogy[4–7] to the currently postulated mechanism[4,7] of nucleogenesis on a fast time scale.

## II. MEASUREMENTS

Samples of airborne debris were collected on filter paper; more massive samples were obtained by collecting some of the condensed debris from an adjacent atoll. Elements 94 through 100, to which the uranium isotopes had decayed, were then chemically extracted and purified. It was assumed that the chemical yields in recoveries and separations for elements 95–100 were identical, all of them being actinide elements which are similar in the +3 oxidation state and behaving almost identically in most chemical procedures. The one exception to this, $Bk^{249}$, which exists also in a tetrapositive oxidation state, caused no difficulty since its abundance could be calculated from $Cf^{249}$ activity in californium fractions separated from the berkelium at known times after the detonation of the thermonuclear device.

### A. Isotopic Ratios

The isotopic composition of the plutonium, americium, and curium fractions were measured on 12-inch, 60° mass spectrometers with triple filament sources. The isotope $Cm^{250}$ was not discernible through the use of the mass spectrometer but its presence was inferred from an excess of spontaneous fission activity over that which would be expected from $Cm^{246}$ and $Cm^{248}$.[3] A minute amount of $Cm^{242}$ was detected through the observation of its characteristic alpha particle energy.

The abundances of all the transcurium elements were measured radiometrically. The isotope $Bk^{249}$ was observed by the measurement of its beta activity and quantitatively measured by the observations of the growth of its alpha-particle emitting daughter, $Cf^{249}$. The abundances of $Cf^{252}$ and $Cf^{249}$ were determined from their measured alpha activities and known half-lives.[8]

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TABLE I. Mass abundances at zero time.

| $A$ Mass number | Isobar[a] observed | $N_A$ [b] = Relative abundance | $C_A$ = Cumulative abundance $= N_A + N_{A+1} + N_{A+2} + \cdots$ | $K = \frac{C_{A(\text{even})}}{(C_{A-1}C_{A+1})^{\frac{1}{2}}}$ [h] |
|---|---|---|---|---|
| 239 | Pu | 1.000±0.004 | 1.424 | |
| 240 | Pu | 0.363±0.004 | 0.424 | 1.44 |
| 241 | Pu, Am | 0.039±0.005 | 0.061 | |
| 242 | Pu | 0.0191±0.003[c] | 0.0226 | 1.55 |
| 243 | Am | $(2.1\pm0.4)\times10^{-3}$ [d] | 0.0035 | |
| 244 | Pu | $(1.18\pm0.07)\times10^{-3}$ | 0.00135 | 1.71 |
| 245 | Cm | $(1.24\pm0.01)\times10^{-4}$ [e] | $1.77\times10^{-4}$ | |
| 246 | (Pu)Cm | $(4.78\pm0.11)\times10^{-5}$ | $5.30\times10^{-5}$ | 1.73 |
| 247 | Cm | $(3.9\pm0.2)\times10^{-6}$ | $5.3\times10^{-6}$ | |
| 248 | Cm | $(1.2\pm0.1)\times10^{-6}$ | $1.4\times10^{-6}$ [g] | 1.48 |
| 249 | (Bk)Cf | $(1.1\pm0.1)\times10^{-7}$ | $1.7\times10^{-7}$ [g] | |
| 250 | (Cm) | ... | | |
| 251 | (Cf) | ... | | |
| 252 | Cf | $(1.03\pm0.07)\times10^{-9}$ [f] | $1.5\times10^{-9}$ | |
| 253 | Cf, Es | $(4.0\pm1.2)\times10^{-10}$ | $5.0\times10^{-10}$ [g] | i |
| 254 | Cf | $(4.2\pm0.3)\times10^{-11}$ | $1.0\times10^{-10}$ | |
| 255 | Es, Fm | $(5.7\pm3)\times10^{-11}$ | $5.7\times10^{-11}$ | |

[a] Isobars in parentheses were not used to obtain quantitative data.
[b] Errors stated are standard errors for comparison of isotopes of the same element.
[c] Relative abundance of $Cm^{242} = 1.9\times10^{-8}$. This probably arose from a small amount of plutonium which contained a small amount of $Am^{241}$.
[d] This error is primarily that of comparing $Am^{243}$ to $Pu^{239}$.
[e] The error of comparing Cm to $Am^{243}$ is ~15%.
[f] The error of comparing Cf to $Am^{243}$ is ~10%.
[g] Includes interpolated abundances from Fig. 2.
[h] $K$ = ratio of even-mass cumulative abundances ($C_A$) to geometric mean of adjacent odd-mass $C_A$'s.
[i] The ratio of $C_A^{253}/(C_A^{252}C_A^{254})^{\frac{1}{2}} = 1.3$. Since here the odd $C_A$ is in the numerator, this datum is not consistent with the other five $K$ values.

The isotope $Cf^{251}$ was not observed directly, but its presence was indicated by the production of $Cf^{252}$ in a short neutron irradiation of the californium fraction. The yield of $Cf^{254}$ was obtained from the amount of 56-day[3] spontaneous fission activity. The relative yield of the nuclide with mass number 253 was inferred from the growth and decay of $Es^{253}$ in a purified californium fraction. Similarly, the growth and decay of $Fm^{255}$ in a purified einsteinium fraction gave the relative yield of mass number 255.

## B. Elemental Ratios

The amounts of the six perceived elements were normalized to each other by observing the variation of beta-decay daughter activities with time, by comparing alpha activities of the elements, and by determining the yield of the same mass numbers in two different elements.

The isotope $Pu^{241}$ undergoes beta decay with a 13-year half-life to $Am^{241}$. The isotope $Pu^{243}$ decays to $Am^{243}$ almost immediately (5-hour beta half-life). The mass spectrometric ratio of $Am^{241}:Am^{243}$ in samples where the time between the detonation of the device and the separation of americium from plutonium was known made it possible to derive the amount of $Pu^{241}$ associated with the americium, and served to connect the plutonium and transplutonium data. The accuracy of the joining of the plutonium data to the americium data is estimated to be about 15%.[9]

[9] In the three available mass spectrometric measurements, the $Am^{241}/Am^{243}$ ratio rises less rapidly than linear growth of $Am^{241}$ from $Pu^{241}$ would predict, indicating that a long-lived (ca 80 day) $\beta$-decay precursor of $Am^{243}$ such as a $Np^{243}$ isomer might exist. This uncertainty is responsible for much of the error in the joining of americium and plutonium data.

The alpha activities of the isotopes of curium, californium, einsteinium, and fermium were all compared to the alpha activity of $Am^{243}$ in the same sample. The curium activity was difficult to interpret since $Cm^{245}$ and $Cm^{246}$ both emit alpha particles whose energies are close to 5.35 Mev and the half-lives of these two nuclides are not known precisely. The mass spectrometric ratio of $Cm^{245}:Cm^{246}$ (Table I) and half-lives of 7500 years and 5500 years,[10] respectively, give a specific

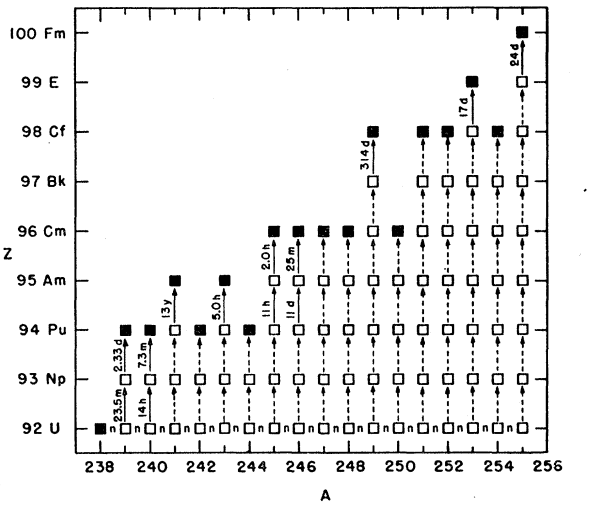


FIG. 1. Production of uranium isotopes in the November, 1952 Mike thermonuclear event, and their decay to beta stability. □ beta unstable nuclide. ■ beta stable nuclide.

[10] This half-life is chosen from unpublished work (ANL) and a consideration of the half-lives reported by J. P. Butler, T. A. Eastwood, H. G. Jackson, and R. P. Schuman, Phys. Rev. **103**, 965 (1956); and A. M. Friedman, A. L. Harkness, P. R. Fields, M. H. Studier, and J. R. Huizenga, Phys. Rev. **95**, 1501 (1954).

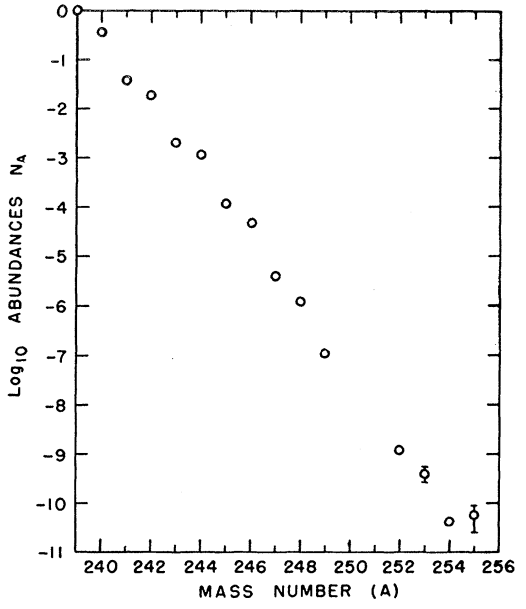


FIG. 2. The $\log_{10}$ of the abundances ($N_A$) of the various nuclides with mass numbers at the time of detonation of Mike.

activity of $(6.6\pm1.0)\times10^5$ 5.35-Mev alpha dis/min per microgram of $Cm^{245}$.

An early mass spectrometric measurement of plutonium gave a crude relative yield of 11-day $Pu^{246}$ that was in good agreement with the later determination of the $Cm^{246}$ relative yield obtained through comparison of activities.

Comparison of these different isotopes of the actinide elements rests upon the assumption that no important fractionation of the elements occurred during the interval between the time they were produced and the time they were deliberately separated and measured.

## III. RESULTS

The abundances $N_A$ determined as described above are shown in Table I. The stated errors are the standard deviations estimated from counting and mass spectrometric data, and the uncertainties in the half-lives used. The logarithms of these abundances are plotted as a function of their mass number ($A$) in Fig. 2.

Since the high-mass nuclides are produced by neutron capture it would be useful to consider not only the abundances $N_A$ but also the sum of the abundances of all nuclides whose mass numbers are higher than $A$; because nuclides of mass greater than $A$ represent the burnout of mass $A$ by neutron capture. We define therefore, the cumulative abundance $C_A$ as follows:

$$C_A=N_A+N_{A+1}+N_{A+2}+\cdots N_{255}.$$

The cumulative abundances $C_A$ are listed in Table I and their logarithms are plotted as a function of mass number ($A$) in Fig. 3.

It can be seen in Fig. 3 that the even mass number cumulative abundances are generally slightly above a line joining the cumulative abundances of odd-mass number isotopes. The approximately semilogarithmic linearity of the plot in Fig. 3 allows one to estimate quantitatively the extent of the excess of even-mass number isotopes over neighboring odd-mass number isotopes. We define $K$ as the ratio of an even-mass cumulative abundance to the geometric mean of its adjacent odd-mass cumulative abundances.

$$K=C_{A\,(\text{even})}/(C_{A-1}C_{A+1})^{\frac{1}{2}}.$$

The value of $K$ shown in Table I is quite constant through mass number 249. The mean value in this region is 1.58 and its mean deviation is 0.14. This generality is violated at masses 253 and 255.

## IV. DISCUSSION

### A. Nature of the Device

The principal thermonuclear reaction in the device is indicated by the equation $H^2+H^3=He^4+n$. The reaction produced neutrons whose energy is initially 14 Mev. In addition, neutrons were also produced by the fission of uranium. The duration in time of the reaction produced by the device was short compared to the shortest possible beta decay half-lives.[11] The temperature of ordinary atomic bombs is about $5\times10^7$°K;[12]

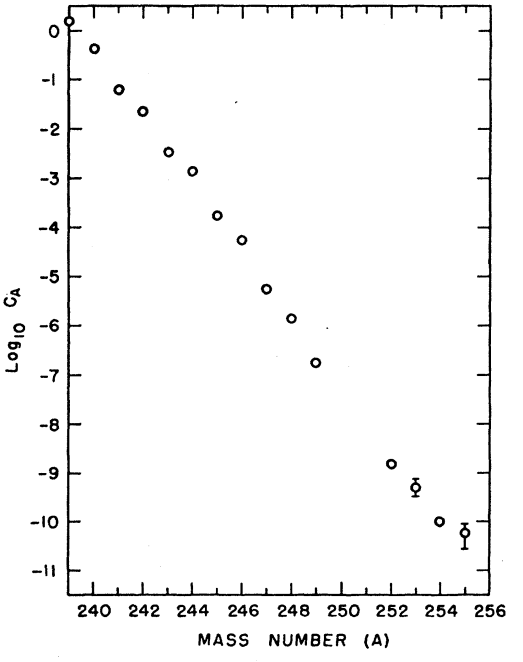


FIG. 3. The $\log_{10}$ of the cumulative abundances ($C_A$) at the time of the detonation of Mike. $C_A=N_A+N_{A+1}+N_{A+2}\cdots+N_{255}$.

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hence the thermal neutron energy must have exceeded four kev.

Cameron,[5] considering multiple neutron capture in Mike, has reasoned that the heavy uranium isotopes were produced after most of the neutrons had been reduced in energy to the point where $(n,f)$ and $(n,2n)$ reactions were unimportant. Devaney, Petschek, and Menzel[13] in their numerical computation of the buildup of heavy nuclides in a device such as Mike, have estimated the product of neutron concentration and time necessary to produce $U^{256}:U^{238}=10^{-11}$ would exceed one mole of neutrons per ml for $10^{-8}$ second.

### B. Neutron Capture

If the concentration of neutrons were uniform throughout the device and if the cross sections did not increase with increasing mass number, the curves in Figs. 2 and 3 would be convex. Cameron suggests that the yields of the heavier nuclides may be greater than would ordinarily be expected because of greater nuclear radii and greater neutron binding energies resulting in greater neutron capture cross sections. One can also assume the existence of zones and gradients of neutron concentration within the device, and by a suitable combination of the curves of Devaney, Petschek, and Menzel synthesize the observed mass abundance curve.

### C. Even-Odd Effect

The cumulative abundances are described by two smooth curves: that for the even-massed cumulative abundances lies uniformly above that for the odd-massed cumulative abundances (through mass 249) by a factor of $1.58\pm0.14$. The most plausible explanation of this even-odd effect is that it represents differences in the capture cross section for the thermonuclear device neutrons.

The even-odd effect is not directly comparable to experimental neutron cross sections for neutron energies close to those expected in the device, because most of these measurements[14–19] do not employ target nuclides having odd numbers of neutrons. Neutron cross sections do appear to be about a factor of 1.5 greater for odd $Z$ than for even $Z$ nuclides in the approximately 300-kev neutron flux in Zephyr.[15]

The analogy between the thermonuclear device and nucleogenesis on a fast time scale has been noted.[3–7] In nucleogenesis, it is unlikely that there was any single element in which many neutron captures by successive isotopes were responsible for the observed abundances. Looking at the Appendix to the review by Burbidge, Burbidge, Fowler, and Hoyle,[4] one can see that only the region between mass numbers 155 and 170 in nuclide abundances taken from Suess and Urey[20] represents the results of the neutron capture on a fast time scale untrammelled by other processes or the proximity of closed shells. There the ratio of even-mass abundances to the mean of adjacent odd-massed abundances is $1.36\pm0.21$. This then, supports the crude analogy between the thermonuclear event and the rapid neutron capture phase of nucleogenesis.

The neutrons responsible for the mass abundance curve, are energetic enough so that capture cross sections are at least partially dependent upon the density of energy levels in the compound nucleus. The small variation in the cross sections of successive even and odd uranium isotopes implied by the data might be taken as an extension of the experimental evidence for the observation (Hurwitz and Bethe)[21] that the significant excitation energy for determining level density is "not measured from the ground state, but from a characteristic level," which depends in a smooth way upon the total number of nucleons in the nucleus.

The relatively large abundances of mass number 253 and mass number 255 nuclides might have been due to alpha decay of higher mass precursors, while neighboring even mass precursors would have undergone fission before alpha decay.

### D. Level Densities

The survival of $Cf^{254}$ in the decay chain: $U\xrightarrow{254\ \beta^-}Np\xrightarrow{254\ \beta^-}Pu\xrightarrow{254\ \beta^-}\cdots Cf^{254}$ can be used to infer some of the nuclear properties of highly neutron-rich nuclides around this mass number. The beta-decay energy of $Np^{254}$ is predicted[22] to be 7 Mev: if the $Pu^{254}$ level density above approximately 4.6 Mev (the neutron binding energy[22]) were high enough to overcome the strong energy dependence favoring decay by beta emission to lower states, then such neutron emission would be expected to compete with beta decay. The fact that $Cf^{254}$ exists implies such competition is small.

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[14] R. L. Macklin, N. H. Lazar, and W. S. Lyon, Phys. Rev. **107**, 504 (1957), show the closed shell effect for 25-kev neutrons.

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It might be expected that the curves in Figs. 2 and 3 would show a dip after mass number 244 where the minor 152 neutron subshell observed in alpha decay energies would effect the level densities and the capture cross sections.[14] No such dip is seen.

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# Evidence for $0^+$ and $1^-$ Levels in $U^{234}$ Populated in the One-Minute Beta Decay of $Pa^{234}$

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A directional correlation measurement has been performed on the 250-kev vs (751+795)-kev composite cascade in $U^{234}$ following the 24-day beta decay of $Th^{234}$ and the subsequent one-minute beta decay of $Pa^{234}$. The "770"-kev (751+795 kev) composite photopeak spectrum coincident with the 250-kev gamma ray was displayed on a multichannel pulse-height analyzer for different positions of the scintillation counters. From these spectra the directional correlations between the 250-kev gamma ray and the lower and upper sides of the 770-kev composite line were measured. The results together with other measurements are consistent with assignments of multipolarity $E1$ to the 250-, 751-, and 795-kev gamma rays and spin and parity assignments $0^+-1^--2^+$ and $0^+-1^--0^+$ to the levels involved in the 250–751 kev and 250–795 kev gamma-gamma cascades, respectively. Thus new levels are proposed at 795 and 1046 kev with spin and parity $1^-$ and $0^+$, respectively.

## INTRODUCTION

SINCE $Th^{234}$ is obtained from natural uranium, it was one of the earliest radioactive nuclei to be studied. Nevertheless, there still exist uncertainties in the interpretation of this decay. One reason for this is that the electron conversion spectrum is masked by the presence of a very intense $Pa^{234} \rightarrow U^{234}$ ground-state beta-ray group (98% of all transitions) with end point 2300 kev. Another reason is that several transitions are strongly electron converted and, therefore, do not appear in the gamma-ray spectrum. For these reasons, it is especially important to complement beta- and gamma-ray spectroscopy of the $Th^{234}$ decay by additional measurements. Consequently, we have attempted a gamma-gamma directional correlation measurement in order to lead to a clearer interpretation of the $U^{234}$ level scheme.

Several outstanding features regarding the $Th^{234} \rightarrow Pa^{234} \rightarrow U^{234}$ decay are well known. The beta and gamma rays from the $Th^{234}$ decay are relatively low in energy and do not interfere with the radiations from the $Pa^{234}$ beta decay which are of higher energy. The $Pa^{234}$ decay proceeds from two isomeric states with half-lives 1.18 minutes and 6.66 hours, respectively. The radiation from the one-minute decay is the more intense and dominates the spectrum. From scintillation counter measurements of the beta- and gamma-ray spectrum, and beta-gamma coincidence measurements, Johansson[1] has established the presence of a transition of energy 810 kev which must be completely converted in $U^{234}$ and which is fed directly by beta decay from the one-minute isomer. Therefore, it is clear there must be a $0^+ \rightarrow 0^+$ ground state transition of this energy. DeHaan *et al.*[2] have measured the end-point energies of the beta-ray groups and their intensities in a beta-ray spectrometer. Ong Ping Hok *et al.*[3] have studied both the internal conversion electron spectrum with a beta-ray spectrometer and the gamma-ray spectrum with a scintillation counter. Complete references to earlier work are given by these authors.[1–3]

A decay scheme for the one-minute $Pa^{234} \rightarrow U^{234}$ decay has been proposed by Bjørnholm and Nielsen and is given in Fig. 1. The decay scheme is based principally on Bjørnholm and Nielsen's measurements[4] using the Copenhagen six-gap "orange" type beta-gamma coincidence spectrometer and the author's gamma-gamma coincidence measurements performed on a scintillation apparatus described in the next section.

The main pieces of evidence for proposing this decay scheme are as follows. The 811-kev transition is very strongly converted and, therefore, must have the multipolarity $E0$. Consequently, there must exist a level at 811 kev with spin and parity $0^+$. The same $0^+$ level has apparently been observed by Asaro and Perl-

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