

correcting the data by this method, a few materials were measured with a series of sample thicknesses. The results of these calculations for carbon and uranium are shown in Fig. 4, with detailed explanation in the figure captions.

It is of interest to point out that the functions  $Q_k$  are very rapidly varying functions of  $h$ , varying about as

$h^n$ ,  $2 < n < 3$ . As a result,  $\Phi_2(a_2, \varphi_2)$  is far more sensitive to the value of  $h$ , and therefore to  $\sigma_T$ , than to the values of  $\omega_l$ . Consequently, it was found that most often iteration was necessary almost exclusively in order to obtain a suitably corrected  $\sigma_T$ . Therefore if  $\sigma_T$  is measured independently, accurate results could be obtained with fewer iterations.

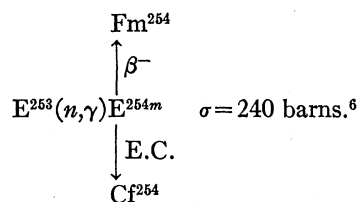
### Spontaneous-Fission Half-Lives of $\text{Cf}^{254}$ and $\text{Cm}^{250}$ †

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(Received May 3, 1957)

Least-squares analysis of spontaneous-fission data obtained over the last four years from a californium sample produced in the November, 1952 thermonuclear test gives a  $\text{Cf}^{254}$  spontaneous-fission half-life of  $56.2 \pm 0.7$  days in agreement with the astrophysical value of  $55 \pm 1$  days. Some comments are made on the neutron buildup in Type I supernovae and the influence of a possible double magic number,  $N=184$ ,  $Z=82$ , at  $A=266$  on the yield curve near  $A=254$ . An approximate spontaneous-fission half-life of  $2 \times 10^4$  years for  $\text{Cm}^{250}$  is deduced from a curium sample also produced in the November, 1952 thermonuclear test.

#### CALIFORNIUM-254

A SUGGESTION<sup>1,2</sup> has recently been made that the exponential decay of the light curves ( $t_3 = 55 \pm 1$  days) of Type I supernovae is due to the spontaneous-fission decay of  $\text{Cf}^{254}$ . In the interval 100 days to 640 days after maximum, the light curve of the supernova in IC 4182 shows no deviation from an exponential decline.<sup>1</sup> If the above light curve represents the spontaneous-fission decay of  $\text{Cf}^{254}$ , the astrophysical half-life of  $\text{Cf}^{254}$  is more precise than the reported half-life values of  $60 \pm 10$  days,<sup>3</sup>  $85 \pm 15$  days,<sup>4</sup> and 55 days<sup>5</sup> determined by spontaneous-fission counting. The first two values were obtained by following the decay of a chemically separated californium fraction containing  $\text{Cf}^{254}$  produced by the following reactions:



† Based on work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> Baade, Burbidge, Hoyle, Burbidge, Christy, and Fowler, *Astron. Soc. Pacific* **68**, 296 (1956).

<sup>2</sup> Burbidge, Hoyle, Burbidge, Christy, and Fowler, *Phys. Rev.* **103**, 1145 (1956).

<sup>3</sup> Bentley, Diamond, Fields, Friedman, Gindler, Hess, Huizenga, Inghram, Jaffey, Magnusson, Manning, Mech, Pyle, Sjolom, Stevens, and Studier, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August 8-20, 1955* (United Nations, New York, 1956), Vol. 7, p. 261.

<sup>4</sup> Harvey, Thompson, Choppin, and Ghiorso, *Phys. Rev.* **99**, 337 (1955); A. Ghiorso, *Proceedings of the International Conference on*

The electron capture to  $\beta^-$  decay ratio of  $\text{E}^{254m}$  is small,  $\sim 10^{-3}$ , and as a result only very low activities of  $\text{Cf}^{254}$  have been produced to date by this method.

The other value (55 days) of the  $\text{Cf}^{254}$  half-life was measured by following a californium fraction<sup>7</sup> separated from the debris of the November, 1952 thermonuclear test ("Mike").

Since the laboratory measurements of the  $\text{Cf}^{254}$  half-life are in such poor agreement and the recent interpretation of the light curves of Type I supernovae depends on the  $\text{Cf}^{254}$  half-life being close to 55 days ( $\pm 1$  day),<sup>1</sup> we have analyzed our thermonuclear test data by the least-squares method.

Californium isotopes of mass number 249, 251, 252, 253, and 254 were produced in the test by the reaction paths illustrated in Fig. 1 and were present initially in the sample which has been periodically counted for its spontaneous-fission activity over the last four years. The more recent measurements indicate that the residual activity is decaying with the known half-life of  $\text{Cf}^{252}$  ( $t_3 = 2.2 \pm 0.2$  years; spontaneous-fission half-life of  $66 \pm 10$  years).<sup>8</sup> The spontaneous-fission activity on November 15, 1956 was  $0.043 \pm 0.001$  counts/min which,

*the Peaceful Uses of Atomic Energy, Geneva, August 8-20, 1955* (United Nations, New York, 1956), Vol. 7, p. 15.

<sup>6</sup> Fields, Studier, Diamond, Mech, Inghram, Pyle, Stevens, Fried, Manning, Ghiorso, Thompson, Higgins, and Seaborg, *Phys. Rev.* **102**, 180 (1956).

<sup>7</sup> Fields, Studier, Mech, Diamond, Friedman, Magnusson, and Huizenga, *Phys. Rev.* **94**, 209 (1954).

<sup>8</sup> A sample from which the californium and curium fractions were separated was supplied by the Los Alamos Scientific Laboratory.

<sup>9</sup> Magnusson, Studier, Fields, Stevens, Mech, Friedman, Diamond, and Huizenga, *Phys. Rev.* **96**, 1576 (1954).

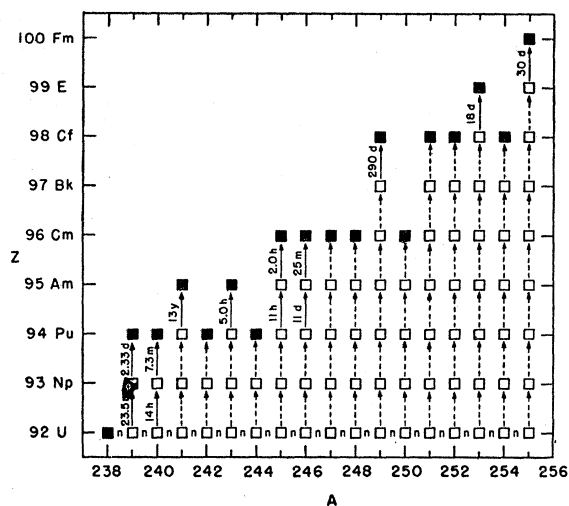


FIG. 1. Production of uranium isotopes in the November, 1952 thermonuclear test, and their decay to the beta-stable valley.  $\square$  beta-unstable nuclide;  $\blacksquare$  beta-stable nuclide.

corrected for decay, gives 0.15 counts/min of  $\text{Cf}^{252}$  at the time of our first count. At zero time on Fig. 2 (approximately two months after the test) about 90% of the spontaneous-fission events were due to  $\text{Cf}^{254}$ . The spontaneous-fission half-life of  $\text{Cf}^{249}$  is  $1.5 \times 10^9$  years, and the other odd- $A$  isotopes,  $\text{Cf}^{251}$  and  $\text{Cf}^{253}$ , are expected to have half-lives which are very much longer than the neighboring even-even isotopes and as a result their contribution is entirely negligible.

Subtracting out the extrapolated  $\text{Cf}^{252}$  activities for the first 400 days of the decay gives the  $\text{Cf}^{254}$  activities listed in Table I. The error associated with each counting rate in Table I includes the counting error and the

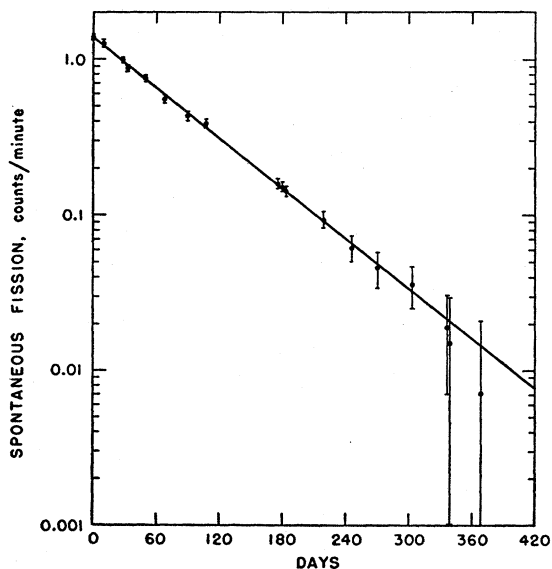


FIG. 2.  $\text{Cf}^{254}$  spontaneous-fission counts/minute vs time. The plotted error includes the uncertainty in the  $\text{Cf}^{252}$  activity which has been subtracted.

uncertainty in the  $\text{Cf}^{252}$  activity which is 0.01 counts/min.

A least-squares analysis of the data with the weighting factors derived from the listed errors gives a half-life of  $56.2 \pm 0.7$  days, where the error is the standard error derived from the least-squares analysis.

#### CURIUM-250

Recent determinations<sup>9,10</sup> of the spontaneous-fission half-lives of  $\text{Cm}^{246}$  and  $\text{Cm}^{248}$  make it possible to estimate the spontaneous-fission half-life of  $\text{Cm}^{250}$ . The curium<sup>7</sup> produced in the "Mike" November, 1952 thermonuclear test (see Fig. 1) was analyzed in a 12-inch 60° mass spectrometer; the results are given in Table II.<sup>11</sup>

The mole percent of  $\text{Cm}^{250}$  in the above curium sample was too small to detect in the mass spectrometer but its presence is established by the following observations:

(1) The "Mike" explosion generated a neutron flux sufficient to produce uranium isotopes at least up to  $A = 255$  by multiple-order neutron capture on  $\text{U}^{238}$ . The presence of  $\text{U}^{255}$  was established by detecting its beta-decay products  $\text{E}^{255}$  and  $\text{Fm}^{255}$ .<sup>12</sup>

TABLE I. Californium-254 spontaneous-fission activity.

Time (days)	Spontaneous fission (counts/min)	Time (days)	Spontaneous fission (counts/min)
0	$1.39 \pm 0.05$	179	$0.153 \pm 0.012$
9	$1.27 \pm 0.06$	183	$0.144 \pm 0.012$
28	$0.99 \pm 0.04$	219	$0.094 \pm 0.012$
33	$0.88 \pm 0.03$	246	$0.062 \pm 0.012$
50	$0.746 \pm 0.03$	269	$0.046 \pm 0.012$
68	$0.560 \pm 0.03$	303	$0.036 \pm 0.011$
91	$0.434 \pm 0.03$	337	$0.019 \pm 0.012$
107	$0.389 \pm 0.024$	338	$0.015 \pm 0.016$
176	$0.160 \pm 0.012$	368	$0.007 \pm 0.014$

(2)  $\text{Cf}^{250}$  (10 years) was not detected in the "Mike" californium fraction. Since  $\text{Pu}^{250}$  is certainly beta-unstable, the absence of  $\text{Cf}^{250}$  is evidence that  $\text{Cm}^{250}$  is either beta-stable, or has a half-life for beta emission of greater than 130 years.<sup>5</sup>

(3) The specific spontaneous-fission activity of the "Mike" curium exceeds the value calculated for the even-mass isotopes  $\text{Cm}^{246}$  and  $\text{Cm}^{248}$ . Since the odd-mass isotopes,  $\text{Cm}^{245}$  and  $\text{Cm}^{247}$ , are expected to make only a negligible contribution to the specific spontaneous-fission activity and other even-mass isotopes can be ruled out on stability arguments (as shown in Fig. 1, beta-decay in the 244 mass chain terminates at  $\text{Pu}^{244}$  and the 252 mass chain decays through curium and terminates at  $\text{Cf}^{252}$ ), it seems reasonable to assign the additional spontaneous-fission activity to  $\text{Cm}^{250}$ .

<sup>9</sup> Fried, Pyle, Stevens, and Huizenga, *J. Inorg. Nuc. Chem.* **2**, 415 (1956).

<sup>10</sup> Butler, Eastwood, Jackson, and Schuman, *Phys. Rev.* **103**, 965 (1956).

<sup>11</sup> Mass spectrometric analysis by C. E. Stevens.

<sup>12</sup> Ghiorso, Thompson, Higgins, Seaborg, Studier, Fields, Fried, Diamond, Mech, Pyle, Huizenga, Hirsch, Manning, Browne, Smith, and Spence, *Phys. Rev.* **99**, 1048 (1955).

The spontaneous-fission disintegration/min per 5.4-Mev alpha-particle disintegration/min for "Mike" curium was measured to be  $(1.72 \pm 0.09) \times 10^{-4}$ . The alpha particles of Cm<sup>245</sup> and Cm<sup>246</sup> were not resolved and both isotopes contribute to the 5.4-Mev alpha-particle intensity. The number of Cm<sup>246</sup> atoms per 5.4-Mev alpha particle is derived in two ways, both of which use the mass-spectrometer data of Table II; (1) a calculation making use of the alpha half-life data of Table III, (2) a determination of the Cm<sup>245</sup> atoms per 5.4-Mev alpha particle through thermal-neutron fission counting, making use of the previously measured thermal-neutron fission cross section for Cm<sup>245</sup> of 2000 barns.<sup>5</sup> For our "Mike" curium sample we calculate  $(1.50 \pm 0.15) \times 10^9$  atoms of Cm<sup>246</sup> per 5.4-Mev alpha disintegration/min. Combining the latter quantity with the spontaneous-fission half-lives of Table III and the mass analysis data of Table II, gives  $(0.99 \pm 0.41) \times 10^{-4}$  and  $(0.10 \pm 0.06) \times 10^{-4}$  spontaneous fissions per 5.4-Mev alpha particle, respectively, for Cm<sup>246</sup> and Cm<sup>248</sup>. The difference between the above sum  $(1.09 \pm 0.47) \times 10^{-4}$  and the experimental (spontaneous fission/5.4-Mev alpha particle) ratio of  $(1.72 \pm 0.09) \times 10^{-4}$  is  $(0.63 \pm 0.56)$

TABLE II. Mass spectrometric analysis of curium from the debris of the November, 1952 thermonuclear test.

Cm isotope	Mole percent
Cm <sup>245</sup>	70.1 ± 0.6
Cm <sup>246</sup>	27.0 ± 0.6
Cm <sup>247</sup>	2.2 ± 0.1
Cm <sup>248</sup>	0.7 ± 0.07

$\times 10^{-4}$  and is assumed to arise from the presence of Cm<sup>250</sup>.

The mass of Cm<sup>250</sup> in our sample was estimated by interpolating between the thermonuclear test yields of Cm<sup>248</sup> and Cf<sup>252</sup>. A Cm<sup>246</sup>/Cm<sup>250</sup> atom ratio of 27.0/0.02<sup>13</sup> is used in the calculation which gives a most probable spontaneous-fission half-life of  $2.3 \times 10^4$  years for Cm<sup>250</sup>. In the light of the above errors and assumptions the Cm<sup>250</sup> spontaneous-fission half-life lies within the range  $(1-10) \times 10^4$  years.

#### DISCUSSION

Our measured half-life value of  $56.2 \pm 0.7$  days for Cf<sup>254</sup> is in good agreement with the astrophysical half-life of  $55 \pm 1$  days for Cf<sup>254</sup> deduced from the light curves of Type I supernovae by assuming the light intensity to be a direct measure of the Cf<sup>254</sup> spontaneous-fission activity in the region of exponential decay.

Cf<sup>254</sup> decays predominantly by spontaneous fission; no other radiations have been observed. This is not too surprising, although it is the first such nuclide discovered, in that Cf<sup>254</sup> is beta-stable and has a predicted alpha-decay half-life of approximately  $10^2$  years.

<sup>13</sup> Argonne National Laboratory (unpublished data).

TABLE III. Half-lives of some curium isotopes.

Isotope	Alpha-disintegration half-life (years)	Spontaneous-fission half-life (years)
Cm <sup>245</sup>	$(7.5 \pm 1.9) \times 10^3$ <sup>a</sup>	
Cm <sup>246</sup>	$(4.0 \pm 0.6) \times 10^3$ <sup>b</sup>	$(2.0 \pm 0.8) \times 10^7$ <sup>c</sup>
Cm <sup>248</sup>		$(4.2_{-2.1}^{+0.6}) \times 10^6$ <sup>d</sup>

<sup>a</sup> H. Diamond (unpublished data).

<sup>b</sup> Friedman, Harkness, Fields, Studier, and Huizenga, Phys. Rev. **95**, 1501 (1954).

<sup>c</sup> See reference 9.

<sup>d</sup> See reference 10.

The nuclide Cm<sup>250</sup> is in many ways similar to Cf<sup>254</sup>. The predicted alpha-decay half-life of Cm<sup>250</sup> is about  $10^5$  years. With a spontaneous-fission half-life of  $(1-10) \times 10^4$  years, Cm<sup>250</sup> probably also decays predominantly by spontaneous fission (this assumes that Cm<sup>250</sup> is beta-stable).

Cf<sup>254</sup> has a spontaneous-fission half-life  $4 \times 10^2$  times shorter than Cf<sup>252</sup>. Cm<sup>250</sup> is similarly related to Cm<sup>248</sup> with a spontaneous-fission half-life approximately  $2 \times 10^2$  times shorter than the Cm<sup>248</sup> value. These results are consistent with the trends observed in other elements where, after a maximum half-life, the heavier even-even isotopes have a greater probability of decaying by spontaneous fission with increasing  $A$ .<sup>14</sup>

The fact that the light curve for the supernova in IC 4182 does not deviate from an exponential decline between the period 100 to 640 days after maximum is difficult to explain. If initially the Cf<sup>254</sup>/Cf<sup>252</sup> atom ratio was 1, at 640 days the spontaneous-fission activity of Cf<sup>254</sup> would be only one-third that of Cf<sup>252</sup>. In comparison, the November, 1952 thermonuclear test produced an initial Cf<sup>254</sup>/Cf<sup>252</sup> atom ratio of 0.04. The absence of a tail on the light curve for the supernova in IC 4182 at 640 days implies that the Cf<sup>254</sup>/Cf<sup>252</sup> atom ratio is greater than 10.

One possible explanation is that the peak yield is strongly influenced by the major shell of 184 neutrons.<sup>15</sup> As pointed out earlier<sup>1,2</sup> the neutron-capture processes take place faster than beta decay, and the original Fe builds up to an atomic weight at which it becomes neutron unstable. Smart<sup>16</sup> estimates that neutrons can no longer be captured when  $N \approx 0.7A$ ,  $Z \approx 0.3A$  and at this point the neutron-capture process stops until a beta decay occurs. The beta-decay lifetimes for such neutron-rich nuclides is estimated to be 0.1 second.<sup>16</sup>

If one assumes that the heavy elements were produced by neutron capture on the lighter elements, evidence for high neutron fluxes for short times is reflected in the abundances of the elements. The cosmological abundance curve based on the data of Suess and Urey<sup>17</sup> has

<sup>14</sup> J. R. Huizenga, Phys. Rev. **94**, 158 (1954).

<sup>15</sup> M. G. Mayer, Phys. Rev. **78**, 16 (1950); Haxel, Jensen, and Suess, Z. Physik **128**, 295 (1950). Beyond the 126 nuclear shell the next major shell occurs after the following orbits are filled:  $1i_{11/2}$ ,  $2g_{9/2}$ ,  $2g_{7/2}$ ,  $3d_{5/2}$ ,  $3d_{3/2}$ ,  $4s_{1/2}$ , and  $1j_{15/2}$ .

<sup>16</sup> J. S. Smart, Phys. Rev. **75**, 1379 (1949).

<sup>17</sup> H. E. Suess and H. C. Urey, Revs. Modern Phys. **28**, 53 (1956).

peaks at Br, Xe, and Pt in addition to the peaks at Y (50 neutrons), La (82 neutrons), and Pb (126 neutrons). Coryell<sup>18</sup> has explained the Br, Xe, and Pt peaks as beta-decay products of unstable nuclides with 50, 82, and 126 neutrons, respectively, which are formed in large abundance, because of the small cross sections of nuclei with magic numbers, in a neutron-capture process of short time-duration. The unstable nuclides with magic neutron numbers giving rise to the peaks at Br, Xe, and Pt have  $N \approx (0.6-0.7)A$  and lie close to the point at which Smart<sup>16</sup> estimates that neutrons cannot be captured until beta decay occurs. The extra stability of the magic number nuclei causes a drop in the beta-decay energies and the resulting longer beta-decay lifetimes may be partially responsible for the observed peaks.

Similarly one might expect a peak at  $N \approx 0.7A = 184$ , or  $A \approx 263$ . It so happens that this is also in the vicinity of a proton shell for a double shell occurs at  $A = 82 + 184 = 266$ . The neutron-capture process in a supernova (Type I) may terminate near  $A \approx 266$  because of (1) the small neutron-capture cross sections of nuclei near closed shells, (2) the fact that neutrons are no longer bound and further capture depends on a beta decay, the lifetime of which is increased due to the extra stability of the nuclei with magic numbers, and (3) the onset of very short spontaneous-fission half-lives. The peak yield predicted on this basis would be several mass numbers beyond Cf<sup>264</sup> and considerable energy would be released by the short-lived spontaneous-fission emitters of  $A > 254$ . Since Cf<sup>264</sup> produces about  $10^{47}$  ergs of energy out of a total release of  $10^{49}$ – $10^{50}$  ergs per supernova outburst,<sup>1,2</sup> additional spontaneous-fission energy from short lived components presents no difficulties, with regard to total energy. Assuming an incidence of one Type I supernova per 500 years for approximately  $5 \times 10^9$  years, the energy per outburst arising from

spontaneous fission is limited by the cosmic abundance curve since the mass of the heavy nuclides which undergo spontaneous fission is distributed among the fission products, and the nuclei resulting from neutron capture on the fission products. On this basis, however, considerably more than  $10^{47}$  ergs per supernova outburst may be due to spontaneous fission.

The energy from the decay of the fission products produced by the short-lived spontaneous-fission emitters will not noticeably alter the observed exponential part of the light curve.<sup>19</sup> A peak in the yield curve beyond  $A = 254$  for a supernova explosion appears to be consistent with the experimental data, and would be consistent with the production of more Cf<sup>264</sup> than Cf<sup>262</sup>.

Nuclides like Fm<sup>267</sup> probably make a measurable contribution to the light curve during the early days. The unstable nuclei produced by neutron capture with  $A \gtrsim 260$  probably do not reach the beta stability valley since spontaneous fission will compete favorably with  $\beta^-$  emission for some nuclides in each constant- $A$  chain. For example, with  $A = 260$ , Cm<sup>260</sup> will have a spontaneous-fission half-life comparable to its  $\beta^-$  decay half-life.

The spontaneous-fission decay of Cm<sup>260</sup> may be an important source of energy in old remnants of Type I supernova explosions, e.g., the Crab Nebula. Crab Nebula presently radiates about  $5.7 \times 10^{42}$  ergs/year<sup>20</sup> or about a factor of  $10^5$  less than the initial decay rate of the exponential part of a Type I supernova light curve. If initially the amount of Cm<sup>260</sup> equalled the amount of Cf<sup>264</sup>, then at present the Cm<sup>260</sup> spontaneous-fission activity would be  $10^{-5}$  times the initial Cf<sup>264</sup> activity and would account for all the present radiation of the Crab Nebula. This difficulty is overcome when one assumes the yield of mass 254 to be greater than mass 250 from a Type I supernova outburst.

We wish to express our thanks to D. W. Engelkemeir for several helpful discussions.

<sup>18</sup> C. D. Coryell, Massachusetts Institute of Technology Laboratory for Nuclear Science Annual Report, May 31, 1956 (unpublished); P. Fong (private communication).

<sup>19</sup> K. Way and E. P. Wigner, *Phys. Rev.* **73**, 1318 (1948).

<sup>20</sup> Calculated from data given in *The Observer's Handbook For 1956* (Royal Astronomical Society of Canada).