

volcano rising in a time corresponding to $\tau = 2.5 \times 10^{-25}$ sec, in which about 1% of the neutrons originally present in the square well nucleus are ejected (Table VI). For Cf^{252} with 154 neutrons, this gives about 1.5 neutrons ejected. The number of scission neutrons found for Cf^{252} would be roughly 10–20% of the average number of neutrons per fission which is about 4 for Cf^{252} ; that is, the number of scission neutrons per fission would be about 0.4 to 0.8. Our specific case does not exactly fit these data, but by a slight increase in the adiabaticity of the rate of rise of the volcano, the number of neutrons ejected by the mechanism of nonadiabatic potential change can be reduced to within the experimentally observed range for still quite reasonable rates of rise of the volcano (i.e., times of scission).

A rough estimate of the energy of the neutrons emitted is also available from the figures computed here. We know that if the volcano rises adiabatically the particles in the nucleus will gain in energy simply because of the reduced phase space available to them. To determine the energy of an emitted particle we must subtract off this artificial energy gain. Thus in the case

of Table V ($\tau = 2.5 \times 10^{-22}$ sec) for which 1% of the particles in the well are ejected, we see that almost all of this 1% is raised in energy to state $m=10$ which has an energy of 66.5 Mev (Table IV). But from this energy must be subtracted the energy of the state to which the adiabatic volcano raises the particles, that is, state $m=9$, with energy 58.0 Mev. Thus about 8.5 Mev is given to 1% of the particles which for Cf^{252} is about 1.5 neutrons. Each scission neutron would have an energy of about 5–6 Mev. For a slightly less gentle volcano one could find four neutrons ejected of approximately 2 Mev each.

The mechanism discussed here in relation to neutron production could perhaps also be applied to the problem of heavier particles (tritons, alphas) in fission.

ACKNOWLEDGMENTS

I would like to thank Professor John A. Wheeler who suggested this problem and guided me through it.

Also, I would like to acknowledge the many valuable discussions with H. Bowman, S. Thompson, N. Newby, and W. Swiatecki.

Mike Results—Implications for Spontaneous Fission*

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(Received November 9, 1961)

The November 1, 1952, thermonuclear explosion "Mike" produced isotopes through mass number 255. Since neutron irradiation time is short compared with possible beta-decay lifetimes, we conclude that, through $A=255$, nuclei far from the line of beta stability on the neutron-rich side are at least as likely to decay by negatron emission as by spontaneous fission.

I. INTRODUCTION

THE heavy isotope yields of the Mike thermonuclear device¹ afford a unique opportunity for the examination of nuclear properties far from the stability curve. Both the occurrence of high-mass-number isotopes and the shape of the yield curve are of significance. In this paper, we examine implications of these factors for spontaneous fission.

II. SYSTEMATICS OF SPONTANEOUS FISSION HALF-LIVES

Early interpretations of spontaneous fission lifetimes were based on the liquid-drop model.² Marked devia-

tions from such a simple model were evident and many attempts were made to improve the model as well as to formulate new approaches. One of the most successful was that of Swiatecki who was able to show a regular dependence of the fission half-life on ground state masses.³ His work has recently been slightly modified and extended by the author.⁴ This modification predicts spontaneous fission half-lives which, for a given z , decrease very rapidly with increasing A (Fig. 1).

Foreman and Seaborg⁵ plotted the logarithm (base 10) of the spontaneous fission half-life vs mass number. They observed that all the half-life curves approached the same linear dependence on mass number at the line characterizing 152 neutrons (Fig. 2).

Recently, Johansson⁶ has analyzed spontaneous fis-

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

¹ H. Diamond, P. R. Fields, C. S. Stevens, M. H. Studier, S. M. Fried, M. G. Inghram, D. C. Hess, G. L. Pyle, J. F. Mech, W. M. Manning, A. Ghiorso, S. G. Thompson, G. H. Higgins, G. T. Seaborg, C. I. Browne, H. L. Smith, and R. W. Spence, *Phys. Rev.* **119**, 2000 (1960).

² E. K. Hyde, University of California Radiation Laboratory Report UCRL-9036, 1960 (unpublished).

³ W. J. Swiatecki, *Phys. Rev.* **100**, 937 (1955).

⁴ D. W. Dorn, *Phys. Rev.* **121**, 1740 (1961).

⁵ B. Foreman and G. T. Seaborg, *J. Inorg. & Nuclear Chem.* **7**, 305 (1958).

⁶ S. A. E. Johansson, *Nuclear Phys.* **12**, 449 (1959).

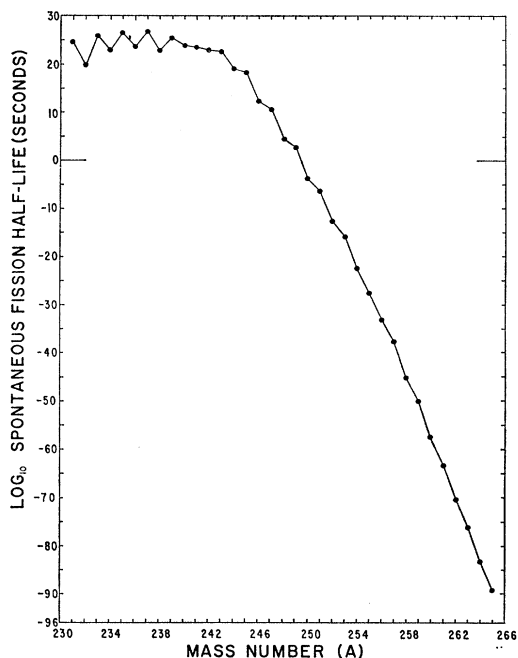


FIG. 1. Spontaneous fission half-lives for isotopes of uranium. (From Dorn.⁴)

sion lifetimes on the basis of single-particle orbitals in a deformed potential. The most interesting implication of his work, as pointed out by Hoyle and Fowler,⁷ is

that the precipitous drop in lifetimes at 152 neutrons⁵ can be interpreted as a single-particle effect which should be "healed" by the time 156 neutrons are reached. Nuclei with $n > 156$ should then have a lifetime roughly as predicted by the liquid-drop model. Johansson's analysis, however, should be modified by a correction for neutron-proton asymmetry which changes the nuclear surface energy and thus decreases the spontaneous fission lifetime.⁸

III. CALCULATION OF ELEMENT PRODUCTION

The system of partial differential equations used in the calculation is

$$\begin{aligned} \frac{dN_{z,n}}{dt} = & (\lambda_{\beta}N)_{z-1,n+1} + (\lambda_{\alpha}N)_{z+2,n+2} - (\lambda_{\beta}N)_{z,n} - (\lambda_{\alpha}N)_{z,n} \\ & - (\lambda_{f}N)_{z,n} + \int_0^{\infty} \phi(E,t)dE \{ [N\sigma_{n,\gamma}(E)]_{z,n-1} \\ & + [N\sigma_{n,2n}(E)]_{z,n+1} - [N\sigma_{n,\gamma}(E)]_{z,n} \\ & - [N\sigma_{n,2n}(E)]_{z,n} - [N\sigma_{n,f}(E)]_{z,n} \}, \quad (1) \end{aligned}$$

where Eq. (1) represents the time rate of change of the number of nuclei with z protons and n neutrons. Processes considered are β^- decay, α decay, spontaneous fission, and the (n,γ) , $(n,2n)$, (n,f) reactions. For machine calculation, the integral over energy from zero to infinity is approximated by a sum over ten

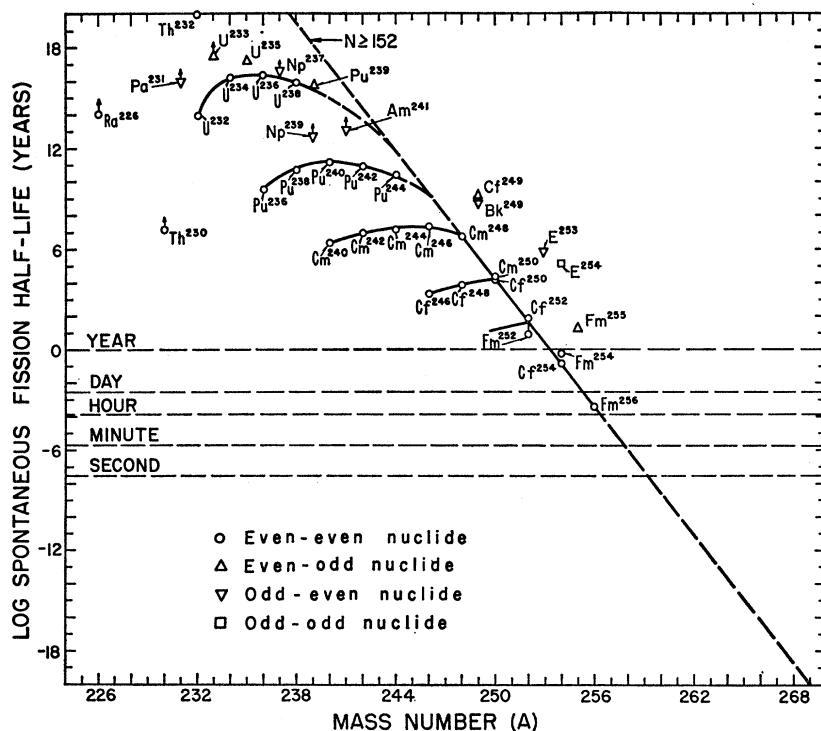


FIG. 2. Spontaneous fission half-lives as a function of mass number. (From Foreman and Seaborg.⁵)

⁷ F. Hoyle and W. A. Fowler, *Astrophys. J.* **132**, 565 (1960).

⁸ J. Wheeler, as quoted in reference 7.

TABLE I. Time-integrated fluxes.

Energy range (Mev)	Flux (neutrons/cm ²)	Problem
0.0 - 0.01	1.20×10^{24}	A, C
0.0 - 0.01	4.81×10^{24}	B, D
0.01- 0.05	7.14×10^{21}	All
0.05- 0.10	1.04×10^{22}	All
0.10- 0.20	1.23×10^{22}	All
0.20- 0.50	1.85×10^{22}	All
0.5 - 1.5	1.71×10^{22}	All
1.5 - 3.0	1.90×10^{22}	All
3.0 - 5.0	5.05×10^{21}	All
5.0 - 7.0	6.41×10^{21}	All
7.0 -14.1	4.16×10^{22}	All

energy intervals, and time steps are selected so that the maximum $dN_{z,n}/N_{z,n}$ for each time cycle is between 5% and 10%. Neutron flux, $\phi(E,t)$, is specified at ten times (with linear interpolation between) for each of the ten energy intervals.

Integration of the system of equations was performed with an IBM-7090 at the Lawrence Radiation Laboratory, Livermore, California.

IV. DATA

Nuclei considered are indicated in Fig. 3. The following data are required for each nucleus: neutron flux in each of ten energy intervals; beta decay constant; alpha decay constant; spontaneous fission decay constant; and $\sigma_{n,\gamma}$, $\sigma_{n,2n}$, and $\sigma_{n,f}$ for each of ten energy intervals. Wherever experimental values are not available, the data are estimated as follows:

1. *Neutron Fluxes.* On the basis of a series of Monte Carlo calculations on "slowing-down fluxes" in burning deuterium, which were carried out by Davis,⁹ it was concluded that for neutron energies greater than 10 kev integrated flux depends only on efficiency of burn. Therefore, the only parameter available to fit the Mike yield curve is the amount of "thermal" flux

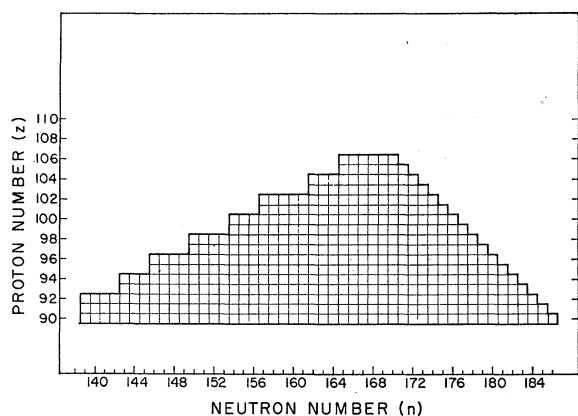


FIG. 3. Elements considered in the calculation.

⁹ D. Davis, Monte Carlo Calculations, Lawrence Radiation Laboratory, University of California, Livermore, 1960 (unpublished).

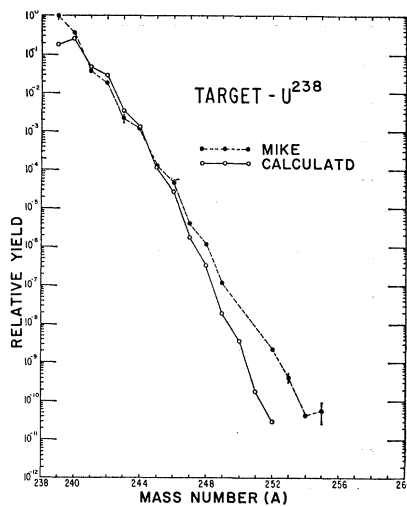


FIG. 4. Comparison of Mike heavy-element yields with the theoretical yields obtained assuming a thermal flux of 1.20×10^{24} neutrons/cm² and spontaneous fission half-lives as predicted by Dorn.⁴

(0-10 kev). This integrated thermal flux is a function of both the number of neutrons produced and of the time the target material is exposed to this flux. Since total exposure times are short compared with possible beta-decay lifetimes, it is immaterial for the calculation whether we assumed a high neutron density for a short time or a low density for a long time. Therefore, we tabulate only the time-integrated fluxes; these are given in Table I.

2. *Beta Decay.* Lifetimes associated with this decay mode are obtained from the following approximate expression for allowed beta decay:

$$\log_{10} ft_{1/2} |M|^2 = K, \quad (2)$$

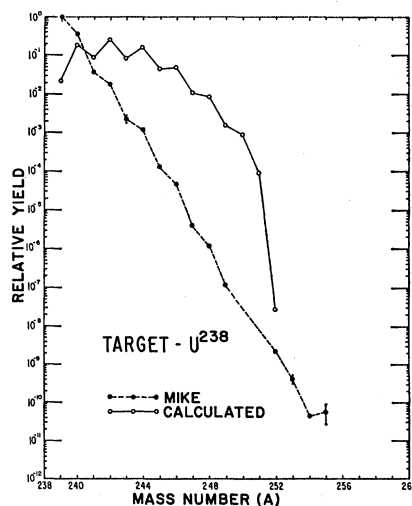


FIG. 5. Comparison of Mike heavy-element yields with the theoretical yields obtained assuming a thermal flux of 4.81×10^{24} neutrons/cm² and spontaneous fission half-lives as predicted by Dorn.⁴

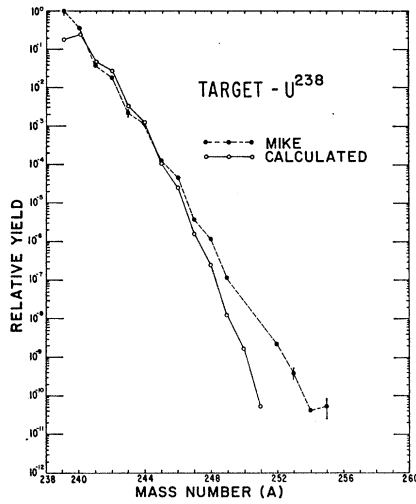


FIG. 6. Comparison of Mike heavy-element yields with the theoretical yields obtained assuming a thermal flux of 1.20×10^{24} neutrons/cm² and spontaneous fission half-lives as predicted by Dorn⁴ with the condition that $\tau_{\frac{1}{2}}$ (spontaneous fission) $\geq \tau_{\frac{1}{2}}$ (β^- decay).

where f is the Fermi function as tabulated by Trigg,¹⁰ $t_{\frac{1}{2}}$ is the partial half-life for beta decay, $|M|^2$ (here assumed to be 10^{-2}) is the square of the nuclear matrix element connecting the initial and final states, and the constant K is estimated to be 4.2 from calculations carried out by the author.¹¹ Beta-decay energies, used in the determination of f , are obtained from Cameron's¹² tabulation.

3. *Alpha Decay.* Alpha decay partial half-lives are estimated using the expression derived by Gallagher and Rasmussen¹³:

$$\log_{10} t_{\frac{1}{2}} = A_z Q_{eff}^{-1} + B_z + \log_{10} F, \quad (3)$$

where A_z and B_z are semiempirical constants defined in their paper, Q_{eff} is the total energy available for alpha decay plus the orbital electron screening correction, and F is the hindrance factor. In the present work, F is assumed to be zero for even-even nuclei and 10 for all others.

4. *Spontaneous Fission.* Spontaneous fission half-lives are calculated according to the expression given in the author's previous paper⁴:

$$\log_{10} \left\{ \begin{array}{l} T_{\frac{1}{2}ee} \\ T_{\frac{1}{2} \text{ odd} A} \\ T_{\frac{1}{2}oo} \end{array} \right\} = \left\{ \begin{array}{l} -30.06 \\ -23.46 \\ -18.56 \end{array} \right\} - 7.8\theta + 0.35\theta^2 + 0.037\theta^3 + 1389(z^3/A) - (4-\theta)\delta m, \quad (4)$$

where $\theta = (z^2/A) - 37.5$ and δm is the difference in Mev between the semi-empirical ground-state mass of a nucleus as given by Cameron¹² and the smooth mass

¹⁰ G. L. Trigg, Washington University, 1950 (unpublished).

¹¹ D. W. Dorn, thesis, Purdue University, 1959 (unpublished).

¹² A. G. W. Cameron, Atomic Energy of Canada Limited, Chalk River Report CRP-690, 1957 (unpublished).

¹³ C. J. Gallagher and J. O. Rasmussen, J. Inorg. & Nuclear Chem. **3**, 333 (1957).

surface as quoted by Swiatecki³:

$$M = 1000A - 8.3557A + 19.120A^{\frac{2}{3}} + 0.76279(z^2/A^{\frac{1}{3}}) + 25.44(n-z)^2/A + 0.420(n-z). \quad (5)$$

5. $\sigma_{n,\gamma}$. Cross sections for (n,γ) reactions are estimated by using U^{235} and U^{238} as standards for odd and even n nuclei, respectively. The expressions used are:

$$[\sigma_{n,\gamma}(E)]_{z,n} = \frac{B_{z,n}}{B_{92,143}} [\sigma_{n,\gamma}(E)]_{92,143} \quad (n \text{ odd}), \quad (6a)$$

$$= \frac{B_{z,n}}{B_{92,146}} [\sigma_{n,\gamma}(E)]_{92,146}, \quad (n \text{ even}), \quad (6b)$$

where $B_{z,n}$ is the binding energy of the last neutron in a nucleus of z protons and n neutrons. Binding energies used are from Cameron's tabulation.¹²

6. $\sigma_{n,2n}$. Thresholds for $(n,2n)$ reactions are assumed to be 0.8 Mev above the binding energy of the last neutron in the target nucleus. Cross sections for this reaction are taken to be zero below and 0.7 b above the threshold. These assumptions represent a reasonable fit to available data.

7. $\sigma_{n,f}$. Neutron-induced fission thresholds are determined from Swiatecki's expression,¹⁴ which has been modified to give the threshold energies directly.

$$(E_{th})_{z,n} \left\{ \begin{array}{l} ee \\ oo \end{array} \right\} = \left\{ \begin{array}{l} 0.5746 \\ 1.6946 \\ 2.8146 \end{array} \right\} + 0.00395 \left[50.13 - \frac{z^2}{A} \right]^3 - 0.166 \left(40.2 - \frac{z^2}{A} \right) - \delta m_{z,n} - B_{z,n+1}, \quad (7)$$

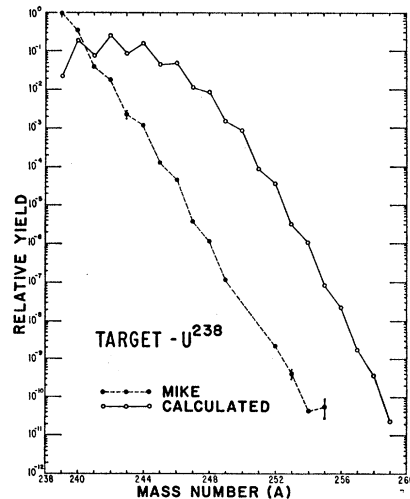


FIG. 7. Comparison of Mike heavy-element yields with the theoretical yields obtained assuming a thermal flux of 4.81×10^{24} neutrons/cm² and spontaneous fission half-lives as predicted by Dorn⁴ with the condition that $\tau_{\frac{1}{2}}$ (spontaneous fission) $\geq \tau_{\frac{1}{2}}$ (β^- decay).

¹⁴ W. J. Swiatecki, Phys. Rev. **101**, 97 (1956).

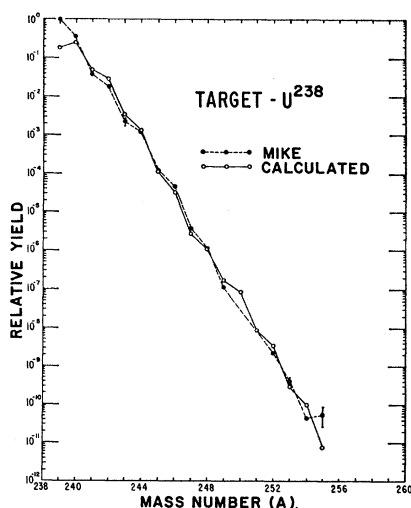


FIG. 8. Comparison of Mike heavy-element yields with the theoretical yields obtained assuming two flux regions (99.99% of the target at 1.21×10^{24} neutrons/cm² thermal flux and 0.01% at 4.81×10^{24} neutrons/cm²) and spontaneous fission half-lives as predicted by Dorn⁴ with the condition that $\tau_{\frac{1}{2}}$ (spontaneous fission) $\geq \tau_{\frac{1}{2}}$ (β^- decay).

where δm is the same mass difference used in Eq. (4), and $B_{z,n+1}$ is the binding energy of the last neutron in a nucleus of z protons and $n+1$ neutrons. The fission cross section is assumed to be zero below threshold; 0.5 and 1.5 b for even n and odd n nuclei, respectively, above E_{th} ; and 1.0 and 2.0 b, respectively, above the $(n,n'f)$ threshold (here assumed to be located at $B_{z,n+1} + 0.8$ Mev).

V. RESULTS

Figures 4 and 5 show a comparison of problems *A* and *B* with the Mike yield data. Values quoted for Mike yields as well as for the calculations are taken at the end of the thermonuclear reaction. Of particular interest is the fact that, when spontaneous fission half-lives

determined by Swiatecki extrapolation procedures^{3,4} are used, the calculations predict no isotopes beyond $A=252$. If, however, the beta-decay half-life of each element is used as a lower limit for its spontaneous fission half-life, the results shown in Figs. 6 and 7 (problems *C* and *D*) are obtained. Note also that when the thermal flux is increased by a factor of four, the shape of the yield curve changes drastically and a maximum develops around $A=242$.

Figure 8 is a comparison of Mike data with a composite of problems *C* and *D* in which it is assumed that 99.99% of the target material sees the flux of problem *C*, and 0.01% sees the flux of problem *D*.

The disagreement at mass 239 may be due to experimental difficulties (based on a discussion with G. H. Higgins of this laboratory). It should be noted that Cameron has also analyzed the yield curve.¹⁵ He postulated only one flux region but had to assume increasing capture cross sections.

VI. SUMMARY AND CONCLUSIONS

To fit the Mike yield curve, we must assume at least two flux regions: 99.99% of the U^{238} at 1.21×10^{24} neutrons/cm² thermal flux, and 0.01% at 4.81×10^{24} neutrons/cm². Other choices of fluxes and flux zones are possible, and in fact are probably indicated. However, the choice of two flux regions seems to be the simplest one which will still fit the data. In addition, we must also assume that spontaneous fission cannot occur faster than beta decay for a given nucleus (Figs. 4, 5, and 8). Both Foreman⁶ and Johansson⁷ predict lifetimes which are consistent with the proper yields. Dorn's extrapolation⁴ is incorrect for nuclei far from the beta stability curve on the neutron-rich side. Thus, Mike results enable us to eliminate the empirical extrapolation. Conclusions regarding relative validity of the other two approaches must be postponed until more information is available for higher z elements.

¹⁵ A. G. W. Cameron, Can. J. Phys. **37**, 322 (1959).