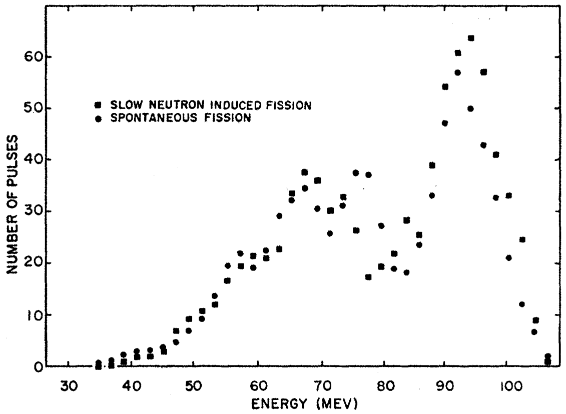

FIG. 1. Histograms of the ionization produced by spontaneous fission pulses of Pu^{240} and by slow-neutron fission pulses of Pu^{239}.

fission pulses. However, an absolute determination of the energy of the fission fragments was not needed since a comparison was made with fragments produced by slow neutrons. The recording was done by feeding the output of the amplifier to an oscilloscope and photographing the screen.

In order that drifts in gain of the amplifying equipment could be detected and corrected for, a series of pulses from a standard pulse generator was impressed on the film at the beginning and end of each run. For this calibration the pulse generator was connected directly to the screen electrode of the chamber and, by capacitance coupling to the collecting electrode, the pulse was fed through the entire apparatus.

Figure 1 presents a histogram of the spontaneous-fission pulses recorded. For comparison the pulse-height distribution of slow-neutron-induced fissions is shown on the same graph. The induced-fission curve was obtained with the same apparatus and sample. The slow neutrons were obtained from a radium-plus-beryllium source with water as the slowing-down medium.

The abscissa scale is a scale of deflections of the oscilloscope, in arbitrary units. We assume that the energy of the fragment is proportional to the organization produced. Under this assumption we have calibrated the abscissa scale in such a way that the energy corresponding to the light fragment peak is 93 Mev as measured by Deutsch.[1] In the ordinates we have the number of pulses giving an ionization between definite limits.

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Spontaneous Fission Systematics

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Previous correlations of spontaneous fission half-lives *vs* Z^2/A predict that the half-lives of even-even isotopes increase with A. A deviation from the above correlation is discussed, and it is shown that the spontaneous fission half-lives of the even-even isotopes go through a maximum with increasing A. The shorter spontaneous fission half-lives beyond the maximum may be a result of the greater deformations of the larger-A nuclides. Some comments are made on fission thresholds.

THE most successful correlations of spontaneous fission half-lives to date are those of Seaborg[1] and Whitehouse and Galbraith.[2] These authors plotted the logarithm of the spontaneous fission half-lives of several nuclides as a function of the parameter Z^2/A, and observed that the logarithm of the spontaneous fission half-lives of even-even nuclides decrease with increasing Z^2/A values in a linear manner. Whitehouse and Galbraith point out that the exception of U^{234} is so striking that the question of experimental error cannot be dismissed.[2] Ghiorso *et al.*[3] remeasured the spontaneous fission half-life of U^{234} and confirmed the earlier results.

The purpose of this note is to call attention to a consistent deviation from the Z^2/A relationship suggested by the above authors,[1,2] and in addition to point out a new relation, namely, that each group of even-even isotopes exhibits a maximum stability toward spontaneous fission. The latter phenomenon suggests the possibility of obtaining information on nuclear deformation and nuclear configuration from spontaneous fission half-lives.

At constant Z, the Z^2/A parameter predicts that the spontaneous fission half-lives of even-even nuclides increase with increasing values of A. For example, at constant Z an increase of two mass units in the heavy region of the periodic table should produce a tenfold increase in spontaneous fission half-life. This can be readily calculated using the slope of the spontaneous

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[1] G. T. Seaborg, Phys. Rev. **85**, 157 (1952).

[2] W. J. Whitehouse and W. Galbraith, Nature **169**, 494 (1952).

[3] Ghiorso, Higgins, Larsh, Seaborg, and Thompson, Phys. Rev. **87**, 163 (1952).

fission half-life *vs* Z^2/A line of Seaborg plotted in Fig. 1. The predicted spontaneous fission half-life of U^{238} is one hundred times longer than that of U^{234}. The experimental half-life of U^{238} is shorter than that of U^{234} by a factor of two. The spontaneous fission half-lives of the even-A uranium isotopes therefore do not obey the $(1/A)$ parameter.

Previously reported spontaneous fission rates of the plutonium and curium nuclides have not included the neutron excess isotopes since they are more difficult to prepare. Uranium data give one the first indication of the effect of neutron excess on spontaneous-fission rate. From Fig. 1, it can be seen that the uranium data suggest, at constant Z, a maximum in the spontaneous fission half-lives of even-even nuclides analogous to beta stability.

The latter model of spontaneous fission predicts that the half-lives of the neutron-excess even-A plutonium isotopes will not continue to increase. Assuming the spontaneous fission half-life of Pu^{242} to be equal or less than that of Pu^{240}, calculations showed the possibility of detecting a spontaneous fission contribution from Pu^{242} in a plutonium sample containing a small amount of this isotope. The measured spontaneous fission half-life of Pu^{242} was 7×10^{10} years,[4] a value twofold less than that of Pu^{240}.[5] These data show that the even-A plutonium isotopes deviate from the Z^2/A parameter and have a maximum stability for spontaneous fission analogous to the behavior of the even-A uranium isotopes. The maximum stability can be seen in Fig. 2 where the logarithm of the spontaneous fission half-lives of the even-A isotopes of even Z elements are plotted as a function of A (Z/N, A-$2Z$, N, etc., are just as effective as A).

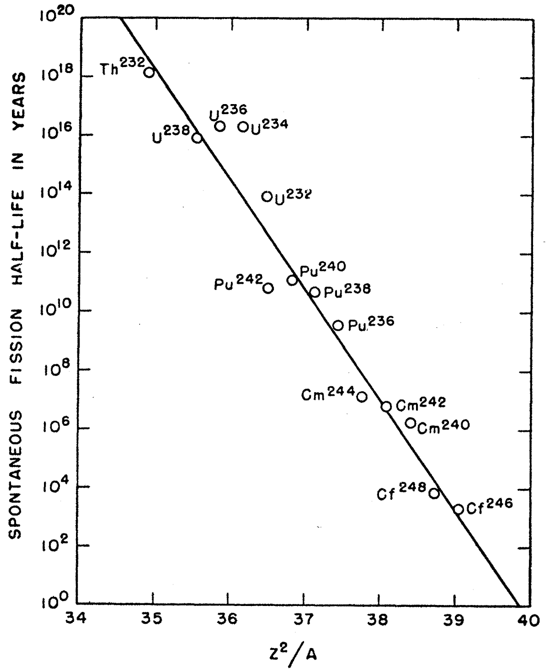

FIG. 1. Plot of spontaneous fission rates as a function of Z^2/A. Spontaneous fission half-lives and references to the data are summarized in the chapter referred to in reference 12 of this paper.

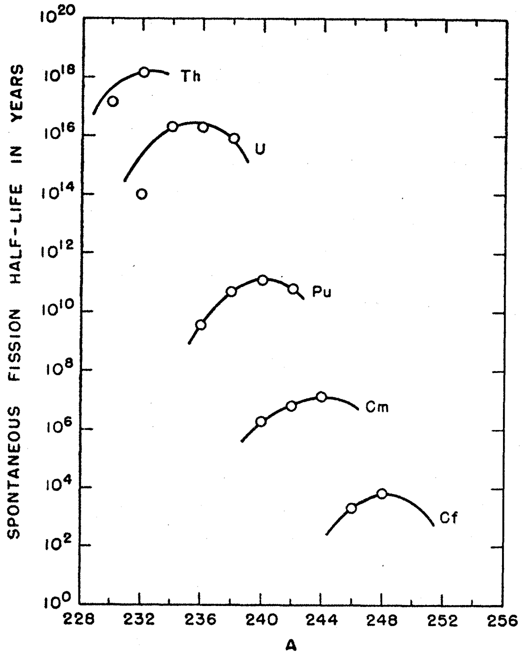

FIG. 2. Plot of spontaneous fission rates as a function of A and Z. Spontaneous fission half-lives and references to the data are summarized in the chapter referred to in reference 12 of this paper.

It is apparent from the above discussion that in predicting the spontaneous fission half-lives of even-even nuclides an error of several factors of ten can be introduced by extrapolating with the Z^2/A parameter. The larger even-A isotopes become more unstable and their spontaneous fission half-lives become shorter. This effect can be seen in Fig. 2.

Bohr and Wheeler[6] in a natural extension of the well-known theory of alpha decay calculated the probability per unit time for a spontaneous fission event. Flügge[7] and Turner[8] have also attempted to evaluate spontaneous fission half-lives with slight modifications of the Bohr-Wheeler formula, but as Segrè[9] points out, neither was successful. Frankel and Metropolis[10] have derived an equation of the following form for the spontaneous-fission half-life in seconds

$$T=10^{-21+k\Delta E},$$

where ΔE is the energy deficit at the saddle point in Mev and k is a constant for a particular nuclide.

Some information on fission thresholds has been

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[4] M. H. Studier and A. Hirsch (private communication).

[5] Chamberlain, Farwell, and Segrè, this issue [Phys. Rev. **93**, 156 (1954)].

[6] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[7] S. Flügge, Z. Physik **121**, 294 (1943).

[8] L. Turner, Revs. Modern Phys. **17**, 292 (1945).

[9] E. Segrè, Phys. Rev. **86**, 21 (1952).

[10] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).

obtained from experiments involving a fast fission process, i.e., one in which fission takes place in times comparable to those involved in gamma and neutron emission. Thus, for fission to occur it must compete favorably with other nuclear processes. It is interesting to note that measured photofission thresholds[11] and those calculated[12] from other nuclear data indicate that fission thresholds are relatively constant in the heavy element region. A correlation of thermal-neutron fission cross sections also supports the idea that fission thresholds in the heavy-element region are approximately constant.[13]

The induced-fission data are therefore at variance with the large variation in fission thresholds calculated by Frankel and Metropolis (see Figs. 2b and 3 in reference 10). If one assumes that the general form of the Frankel and Metropolis equation for spontaneous fission half-lives is correct, some limits on the variation of fission thresholds can be calculated employing experimental spontaneous fission half-lives. Such calculations give $k\Delta E$ values of 46.7 and 35.4 for Th^{232} and Cm^{242}, respectively. If k were constant over this region, ΔE would change by 24 percent (if $\Delta E = 5.4$ Mev for Th^{232}, then $\Delta E = 4.1$ for Cm^{242}; these two nuclides represent the range over which information is available for photofission and neutron fission). The value of k decreases[10] in going from Th^{232} to Cm^{242}, so that the actual change in ΔE may be small enough to be within the experimental error of the experiments mentioned above. The longer spontaneous fission half-lives of the odd isotopes can possibly be explained by a small increase in their fission threshold. An increase in $k\Delta E$ of 10 percent (change in $\Delta E \leq 0.5$ Mev) in going from Pu^{238} to Pu^{239} would account for the increase in spontaneous fission half-life and not be inconsistent with other data. Thus, within experimental error the variations in fission thresholds calculated from spontaneous fission data are possibly in agreement with neutron fission and photofission thresholds.

The collective model[14,15] predicts that the energy of the first excited state of even-even nuclei depends principally on the equilibrium value of the nuclear distortion. The large changes in the energies of the first excited states at major shell closures have been explained in terms of nuclear distortion. [16] The energy of the first excited state of even-even nuclei in the region thorium to curium is not decreasing very fast, and thus it is assumed that the change in deformation is not large in this region. At constant Z, the Coulomb energy decreases with increase in radius (at constant Z, the radius may not increase proportionally to $A^{\frac{1}{3}}$). The spontaneous fission half-lives of the even-even isotopes, however, do not continue to increase with A. The shorter spontaneous fission half-lives beyond the maximum are possibly a result of the greater deformations of the larger-A nuclides. The ellipsoidal deformation may increase the decay rate due to the improvement in penetration through the thinner barrier.[15] Therefore spontaneous fission half-lives may be a sensitive measure of the deformation of a nucleus and thus yield some information on nuclear configuration.

It would be interesting to have data on the relative nuclear deformations of even and odd nuclei, since the possibility exists for explaining the slower spontaneous fission rates of the odd nuclei in terms of their smaller deformation. Other factors, of course, such as the mixing of prolate and oblate deformations of the nucleus may also cause the spontaneous fission process to be inhibited.

It is a pleasure to acknowledge Dr. W. M. Manning for his interest and comments regarding this work.

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[11] Koch, McElhinney, and Gasteiger, Phys. Rev. **77**, 329 (1950).

[12] J. R. Huizenga, unpublished data discussed in Chapter 20 of *The Transuranium Elements: Survey* (McGraw-Hill Book Company, Inc., New York, 1954), National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV.

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[14] A. Bohr, Kgl. Danske Vibenskab. Selskab Mat.-fys. Medd. **26**, No. 14 (1952).

[15] D. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).

[16] K. W. Ford, Phys. Rev. **90**, 29 (1953).