by Kienle.[30] This spectrum exhibits very broad absorption lines which are due in part, to the linewidth of the target. Assuming that the structure was due to magnetic hyperfine interaction with a small admixture of quadrupole splitting, a magnetic energy splitting of $(7.8\pm0.4)\times10^{-6}$ eV and a quadrupole-interaction energy of $(0.9\pm0.4)\times10^{-6}$ eV were obtained. Using the effective magnetic field in erbium of $H=7.46\times10^6$ Oe,[31] we obtain a value of g_R of 0.33 ± 0.02. This result agrees within the experimental error with the value of 0.31 predicted by Nilsson and Prior.[32] Further work with a target having a narrower linewidth should permit a more accurate determination of the values of g_R for all the even-even isotopes of erbium.[33]

[30] P. Kienle, Rev. Mod. Phys. **36**, 372 (1964).
[31] H. Dobler, G. Petrich, S. Hüfner, P. Kienle, W. Wiedemann, and H. Eicher, Phys. Letters **10**, 319 (1964).
[32] S. G. Nilsson and O. Prior, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **32**, No. 16 (1961).
[33] After the publication of the present result by J. Eck, Y. K. Lee, E. T. Ritter, R. R. Stevens, Jr., and J. C. Walker [Phys. Rev. Letters **17**, 120 (1966)] there was another experiment by E. Münk, D. Quitman, and S. Hüfner [Z. Naturforsch **21**, 847 (1966)] who reported $g_R=0.331\pm0.010$ for Er^{168}, in good agreement with the present results.

V. CONCLUSIONS

Mössbauer studies were carried out following Coulomb excitation in Gd^{155}, Gd^{156}, Gd^{158}, Gd^{160}, Dy^{164}, and Er^{168}. For the last three isotopes there are no suitable radioactive parents, and the present work is the first observation of Mössbauer effect in these nuclides. The systematic variation of the quadrupole moments in Gd^{156}, Gd^{158}, and Gd^{160} was observed by using Coulomb excitation to populate the first excited states of these nuclei, and subsequently observing the Mössbauer effect from these levels. The ratios of quadrupole moments obtained agreed within the experimental uncertainty with the corresponding ratios obtained from Coulomb-excitation cross-section data, and thus these results provide additional confirmation of the collective theory of deformed nuclei.

The results of these experiments demonstrate the feasibility of carrying out systematic studies of nuclear properties using the Mössbauer effect following Coulomb excitation, and further demonstrate that the highly deformed nuclei in the rare earths are quite accessible to detailed study by this technique.

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Fission and the Synthesis of Heavy Nuclei by Rapid Neutron Capture*

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The role of fission is examined in the synthesis of heavy nuclei by multiple capture of neutrons in thermonuclear explosions. We begin by reviewing evidence from the recent Tweed and Cyclamen experiments indicating that neutron-induced fission is a serious source of depletion in neutron capture chains which start from targets of ^{242}Pu and ^{243}Am. An analysis of Tweed abundances (Sec. 2) is made to obtain capture-to-fission ratios for the odd-A plutonium isotopes through $A=253$. We next use the liquid-drop model of Myers and Swiatecki plus empirical shell corrections and pairing energies, in order to correlate and predict spontaneous fission lifetimes (Sec. 3) and fission barriers (Sec. 4). For nuclei having $Z\leq101$ and $N\leq157$, we extrapolate the shell correction, assuming it to be a function of N plus a function of Z, and thus obtain neutron binding energies, fission barriers, and spontaneous fission lifetimes for neutron-rich heavy nuclei (Sec. 6). Capture-to-fission ratios are estimated for many of these nuclei in Sec. 7, and qualitative agreement is found with laboratory and Tweed results. In Sec. 8, the extrapolation is continued out to $N=159$ and $Z=104$. We conclude that by using the liquid-drop model plus semiempirical shell corrections, one can obtain capture-to-fission ratios and spontaneous fission half-lives which are usefully accurate. However, for predicting properties of nuclei having $Z>104$, $N\gtrsim159$, one needs, in this formalism, an accurate way of predicting shell corrections or nuclear masses.

1. INTRODUCTION

We consider the synthesis of heavy nuclei by the multiple capture of neutrons in nuclear explosions. In debris from the first experiment of this type, namely the Mike thermonuclear event of November 1952, nuclei through mass 255 were detected,[1] starting from a target of ^{238}U. In two more recent and more readily interpretable experiments, the Par[2] and Barbel[3] underground explosions of October 1964, nuclides through mass 257 were identified, again starting from a target of ^{238}U. A theoretical interpretation of the abundances of the various neutron capture products

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] H. Diamond *et al.*, Phys. Rev. **19**, 2000 (1960).
[2] D. W. Dorn and R. W. Hoff, Phys. Rev. Letters **14**, 440 (1965).
[3] Los Alamos Radiochemistry Group, Phys. Rev. Letters **14**, 962 (1965).

has been given[4] and will hereafter be referred to as I. In considering the abundances, it is well to recall that radiochemical analysis of the debris does not begin for a day or more after the explosion. Therefore, the neutron-rich nuclides which were formed in the original neutron capture chains will have undergone β decay and will be identified in the debris at higher Z than 92. For example, mass-257 nuclides are identified as ^{257}Fm. In I it was concluded that those nuclides in the debris having $A \lesssim 248$ were formed as uranium nuclides by multiple neutron capture on ^{238}U, while those with $A \gtrsim 250$ were primarily produced by neutron capture in an odd-Z chain. The possibilities which were considered were protactinium and neptunium capture chains starting from (n,p) or (d,n) reactions on ^{238}U. Although very little of the odd-Z material is formed ($\lesssim 10^{-3}$ relative to the uranium), it will become dominant after ten or so captures because of the systematically larger capture cross sections of odd-Z nuclides. The Np capture chain was favored because of its higher computed neutron-capture cross sections.

In I, it was assumed that the neutrons which were being captured (and which were taken to have energies of the order of 10 keV) could not induce fission. It is a major purpose of this paper to investigate how the situation is changed when the possibility of neutron-induced fission is taken into account.

If one ignores fission, it is at once apparent that targets of higher mass than 238 should be used in order to increase the atomic weights of nuclides which can be recovered in detectable amounts from the debris. Even considering fission, it was hoped that after one had added a few neutrons to a target nucleus such as ^{243}Am, the neutron-binding energy would have fallen enough to make fission a weak competitor to capture. Some recent experiments have, however, indicated that neutron-induced fission is a very serious source of depletion in the capture chain when one starts from targets of ^{242}Pu or ^{243}Am and probably also in Np capture chains.

First of all, there was LRL's Tweed experiment[5] in which ^{242}Pu was used as the target. The observed abundances fell more rapidly with A than had been observed for Par[2] and Barbel,[3] but the interpretation was obscured by uncertainty as to the neutron exposure which was achieved. Therefore, the experiment was more or less repeated as the Vulcan[6] event but with a ^{238}U target. The Vulcan abundances as a function of A were nearly identical to those found in Par, thereby indicating that very nearly the same neutron exposures were achieved in Par, Vulcan, and Tweed, and also Barbel. The disappointing abundances in Tweed as compared to Par, Vulcan, or Barbel may then be attributable to competition from fission when one starts from a ^{242}Pu target. Such an interpretation of Tweed has been suggested by Ingley.[5] We have independently made an analysis of Tweed which is given in Sec. 2, where we conclude that Pu most likely fissioned for all odd-A target nuclides through at least $A=253$ and that the Np capture chain starting from ^{242}Pu$(n,p)^{242}$Np also suffered from fission competition. Any Am chain, starting, for example, from ^{242}Pu(d,n)-^{243}Am is believed to have been severely depleted by fission as discussed in the next paragraph.

In the Cyclamen experiment,[7] a composite target containing both ^{238}U and ^{243}Am was used. From the observed abundances at low A (especially masses 245 and 246, recovered as Pu isotopes), one can deduce the neutron exposure received by the uranium, or, if not the absolute exposure, at least that relative to other events. It was concluded[7] that the Cyclamen exposure exceeded that achieved in Par and Barbel by about 70%. However, it also appeared that all of the observed abundances through $A=257$ could be explained without any contribution from the ^{243}Am. (While there is some evidence that not all the ^{243}Am received the full neutron exposure, it seems unlikely that all the ^{243}Am escaped the full exposure.) The absence of any apparent contribution to mass 257 from an americium capture chain suggests that this capture chain was strongly depleted by fission. More specifically, the results would be consistent with a fission depletion of the americium capture chain by a factor (at $A=257$) of 10^7 or greater as compared to a chain without fission. Some of the depletion (about a factor 100) had been anticipated from exposure to fast neutrons. The remaining factor of 10^5 suggests that the even A americium target nuclei remain fissionable to quite large A.

If one accepts the Tweed and Cyclamen results as implying fissility of neutron-rich neptunium, plutonium, and americium nuclei, and if one further assumes that capture chains in neutron-rich nuclei of higher atomic number will suffer even more severely from neutron-induced fission, then one concludes that the use of neutron capture targets with Z higher than 92 will not be a fruitful approach to follow in creating new and heavier elements or nuclides. In addition, there is some evidence from Cyclamen to suggest that spontaneous fission half-lives of high-A capture products may be so short as to make difficult their identification in explosion debris. In particular, no nuclides of $A \geq 258$ were observed in the debris although any simple extrapolation of the abundances as a function of A would predict that detectable quantities had been made, certainly through $A=259$ and perhaps through $A=261$. No mass-256 nuclides were found either, but this might be expected because of loss by spontaneous fission at ^{256}Fm with a half-life of 2.7 h. More likely, the mass-256 β-decay chain was terminated by spontaneous fission

[4] G. I. Bell, Phys. Rev. **139**, B1207 (1965).

[5] J. Ingley, Bull. Am. Phys. Soc. **11**, 655 (1966). Results are given in Table II.

[6] Experiment by LRL, Livermore. Preliminary results by LRL Livermore, LRL Berkeley, ANL, and LASL communicated by R. W. Hoff.

[7] Los Alamos Progress Report, 1966 (unpublished).

at ^{256}Cf (see Sec. 8). Similarly, identification of ^{258}Fm was not anticipated because of its probable very short spontaneous fission half-life,[8] or because of the spontaneous fission of ^{258}Cf. The absence of detectable amounts of ^{259}Fm or ^{259}Md in the debris could be explained if the spontaneous fission half-life of ^{259}Fm is $\lesssim 5$ h, though this is by no means the only possible explanation for its absence.[9] These results raise the general question as to whether spontaneous fission half-lives may be so short for higher mass products ($A=261$, 263, 265, $\cdots$) as to make them undetectable.

In this paper we wish to consider whether the neutron-induced and spontaneous fission of heavy nuclei can be understood from a uniform point of view. We are, of course, particularly interested in neutron-rich nuclei.

In Sec. 2 the results of the Tweed experiment are analyzed in some detail, primarily in order to obtain estimates of capture-to-fission ratios for the heavier plutonium isotopes.

In Sec. 3, we present a correlation of known spontaneous fission half-lives with the predictions of a liquid-drop model plus empirical shell corrections, while in Sec. 4 fission barriers are considered on the same basis. The recent liquid-drop model of Myers and Swiatecki[10] is used together with an empirical shell correction. Extrapolations of the shell correction are made in Sec. 5 assuming that the neutron and proton shell corrections can be made independently. The resulting fission barriers and neutron-binding energies are given in Sec. 6, and the capture-to-fission ratios in Sec. 7. In the concluding Sec. 8 comparisons are made with experiment, and extrapolations are made to $Z=102$ and 104.

In general, the results which we obtain by using the liquid-drop model plus shell corrections seem to be consistent with all the experimental data. Our predicted capture-to-fission ratios for plutonium and americium isotopes are consistent with the results of the Tweed and Cyclamen experiments. We do not, however, attempt to predict the properties of nuclei having neutron or proton numbers higher than those observed, that is, $Z>104$, $N\gtrsim 159$. In order to make such predictions meaningful a reliable method of predicting shell corrections would be required.

2. INTERPRETATION OF TWEED ABUNDANCES

Let us begin by assuming that the Tweed target, ^{242}Pu, received the same neutron exposure as did the Vulcan target, ^{238}U. Furthermore, because of close agreement between Vulcan and Par mass-abundance data, we assume that the targets had the same neutron exposure in all the experiments, Par, Vulcan, Tweed, and Barbel. According to the model used to interpret Par and Barbel in I, this is an exposure of 7 neutrons/b for the neutrons at a nominal 20-keV energy.

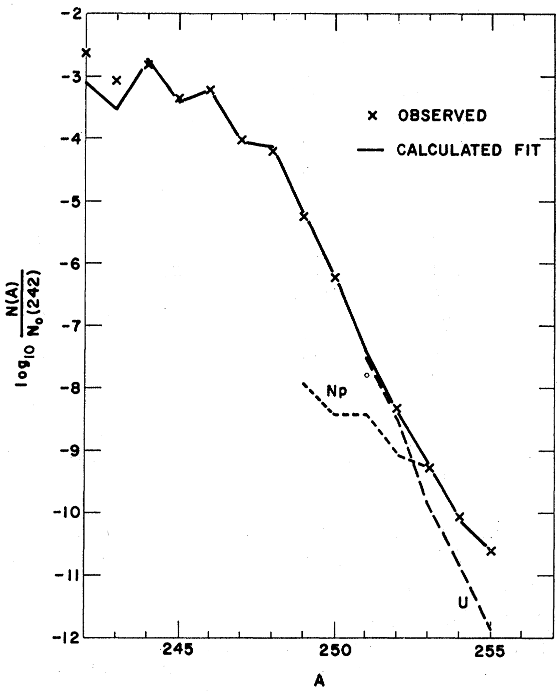

FIG. 1. Abundance versus atomic number for Tweed. The data which are plotted are given in Table II.

Disappointing abundances of heavy nuclides in Tweed, as compared to Par (and Barbel), would then seem to imply unexpectedly low-capture cross sections or serious competition from fission in capture chains starting from a ^{242}Pu target. If the problem were simply low-capture cross sections, then the Pu capture cross sections would have to be lower than those calculated as in I by, on the average, a factor of two with further superimposed variations. Because we are not aware of any evidence for rapid variations in strength functions, Γ_γ, or in other model parameters in this region of the chart of the nuclides, we regard such low Pu cross sections as very unlikely. It seems rather that competition from fission is the more likely explanation, and we have proceeded with an interpretation on that assumption.

A glance at the Tweed data, Fig. 1, shows that for $A\leq 249$, the odd-even alternations are such as would be expected in an even-Z capture chain; that is, the even-A nuclides are relatively more abundant. On the other hand, the masses 253, 254, and 255 look as if they were formed in an odd-Z capture chain, presumably Am or Np. If one deduces from the Cyclamen results that Am is quantitatively destroyed by fission, then a Np capture chain would seem the likely source of 253, 254, and 255.

[8] Combined Radiochemistry Group, Phys. Rev. **148**, 1192 (1966).

[9] Los Alamos Radiochemistry Group, Los Alamos Scientific Laboratory Report No. LA-DC-8103 (revised), 1966 (unpublished).

[10] W. D. Myers and W. J. Swiatecki, University of California Lawrence Radiation Laboratory Report No. UCRL-17070 (unpublished). Earlier versions with different shell corrections and liquid-drop parameters were given in Nucl. Phys. **81**, 1 (1966) and University of California Radiation Laboratory Report No. UCRL-11980, 1965 (unpublished).

TABLE I. Plutonium cross sections (barns).

A	$\sigma_{n,\gamma}^0$ (no fission)[a]	$\sigma_{n,\gamma}$ (fission)[b]	$\sigma_{n,\gamma}+\sigma_{n,f}$	$\alpha=(\sigma_{n,\gamma}/\sigma_{n,f})$
242	0.63	0.63		
243	1.25	0.54	2.35	0.30
244	0.44	0.44		
245	1.04	0.31	2.55	0.14
246	0.32	0.32		
247	0.82	0.33	2.25	0.17
248	0.23	0.23		
249	0.64	0.06	2.91	0.02
250	0.165	0.165		
251	0.50	0.07	(2.54)	0.03
252	0.115	0.115		
253	0.35	0.11	(2.0)	0.06
254	0.09			

[a] Computed as in I.
[b] Computed from Eq. (2) and to match Tweed data. No fission assumed for even A. For 251 and 253, the values of $\sigma_{n,\gamma}+\sigma_{n,f}$ are slightly smaller than given by Eq. (2).

Using the Np capture cross sections given in I and an exposure of 7 b^{-1}, a good fit is found to these three points for a Np chain starting at $A=242$ and having an abundance of 5×10^{-8} times the initial ^{242}Pu. We will discuss this abundance later on. However, it is to be noted that such a chain would contribute about 20% of the observed 252 abundance, and negligibly to all points for $A\leq250$.

Hence all the data for $A\leq252$ are to be explained as coming from a Pu capture chain. If one now allows each Pu isotope to fission, there are then two parameters, $\sigma_{n,\gamma}$ and $\sigma_{n,f}$, to determine for each isotope and thus there is no unique way to determine them from one datum (abundance) per isotope. We have, therefore, proceeded as follows: We assume that the even-even Pu target nuclides do not fission upon absorbing a neutron, and we calculate their capture cross sections as in I. For an odd-A Pu target nuclide we write

$$\sigma_{n,c}=\sigma_{n,n}+\sigma_{n,\gamma}+\sigma_{n,f}, \qquad (1)$$

where these quantities are the cross sections for, respectively, compound nucleus formation, compound elastic scattering, radiative capture, and fission. We ignore the slight possibility of inelastic neutron scattering. In the calculations reported in I, $\sigma_{n,c}$ was taken to be a constant (3.2 b) and $\sigma_{n,\gamma}$ was calculated assuming $\sigma_{n,f}=0$. In principle one could estimate $\sigma_{n,f}$ by assuming a spin of the target nucleus, a position of the fission barrier with respect to the neutron-binding energy, and a spectrum of transition states, but such complications seem hardly warranted. Instead we have assumed that when fission is allowed, the ratio $\sigma_{n,\gamma}/\sigma_{n,n}$ retains the same value as computed without fission. In other words, we assume that fission is equally effective in competing with scattering and with capture. In our interpretation, it will turn out that $(\sigma_{n,f}+\sigma_{n,\gamma})$ is large compared to $\sigma_{n,n}$ so that the results are insensitive to our assumption about $\sigma_{n,n}$. (We have also noted in calculations of capture-to-fission ratios, using single-level formulas for a single spin state, that the ratio $\sigma_{n,n}/\sigma_{n,\gamma}$ increased by only about 20% as $\sigma_{n,\gamma}/\sigma_{n,f}$ varied from around 1.0 to 0.1.)

Letting $\sigma_{n,\gamma}/\sigma_{n,n}$ have the same value as computed without fission, namely, $\sigma_{n,\gamma}^0/\sigma_{n,n}^0$, we see that

$$\sigma_{n,\gamma}=\sigma_{n,c}\Big/\left(1+\frac{1}{\alpha}+\frac{\sigma_{n,n}^0}{\sigma_{n,\gamma}^0}\right) =\sigma_{n,\gamma}^0\Big/\left[1+\frac{1}{\alpha}\left(1+\frac{\sigma_{n,n}^0}{\sigma_{n,\gamma}^0}\right)^{-1}\right], \qquad (2)$$

where α is the capture-to-fission ratio, $\sigma_{n,\gamma}/\sigma_{n,f}$. Thus using this equation, we can compute $\sigma_{n,\gamma}$ for any postulated α. Calculating $\sigma_{n,\gamma}^0$ and $\sigma_{n,n}^0$ as in I, we have then only a single parameter for each of the odd-A Pu isotopes. After a trial we were able to fit most of the observed abundances within quoted uncertainties using the cross sections in Table I.

The comparison with observed abundances is given in Table II for a plutonium capture chain starting from 6.5% of the initial ^{242}Pu, $N_0(242)$, plus a neptunium capture chain starting from $^{242}\text{Np}=5\times10^{-8}\ N_0(242)$.

The results indicate that Pu is fissioning seriously at all odd-A target isotopes through 253. In particular, ^{249}Pu and ^{251}Pu seem to have very low capture-to-fission ratios, and this may continue at higher isotopes. The Np does not seem to be undergoing significant fission for masses around 254.

The abundances of the postulated Pu and Np capture chains merit discussion. From Table I it will be noted that we have not made any effort to fit the data at $A=242$ or 243. At face value, our Pu chain would imply that 6.5% of the Pu survived fast fission and participated in the slow-neutron capture and fission. This is about five times as much ^{242}Pu as one would expect to survive fast fission in DT burning to 100% efficiency and even so there is much more 242 observed

TABLE II. Tweed abundances.

A	Observed[a]	$N(A)/N_0(242)$ Calc.[b] (Pu chain)	Calc.[c] (Np chain)	Calc. total
242	2.5×10^{-3}	7.8×10^{-4}		7.8×10^{-4}
243	8.8×10^{-4}	2.9×10^{-4}		2.9×10^{-4}
244	1.6×10^{-3}	1.9×10^{-3}		1.9×10^{-3}
245	4.5×10^{-4}	3.9×10^{-4}		3.9×10^{-4}
246	6.0×10^{-4}	6.0×10^{-4}		6.0×10^{-4}
247	9.5×10^{-5}	8.8×10^{-5}		8.8×10^{-5}
248	6.5×10^{-5}	7.7×10^{-5}		7.7×10^{-5}
249	5.6×10^{-6}	5.7×10^{-6}		5.7×10^{-6}
250	6.0×10^{-7}	6.1×10^{-7}		6.1×10^{-7}
251		3.4×10^{-8}	3.8×10^{-9}	3.8×10^{-8}
252	5.2×10^{-9}	3.4×10^{-9}	8.4×10^{-10}	4.2×10^{-9}
253	5.3×10^{-10}	1.4×10^{-10}	5.0×10^{-10}	6.4×10^{-10}
254	8.7×10^{-11}	1.4×10^{-11}	1.8×10^{-11}	8.2×10^{-11}
255	2.5×10^{-11}	1.3×10^{-12}	2.6×10^{-11}	2.7×10^{-11}

[a] Data from LRL Livermore, LRL Berkeley, ANL, and LASL Radiochemistry Groups; communicated by R. W. Hoff.
[b] Pu chain, with abundance 0.065 $N_0(242)$, cross sections from Table I, and exposure = 7 neutrons/b.
[c] Np chain starting from ^{242}Np at 5×10^{-8} $N_0(242)$ with $\sigma_{n,\gamma}$ from I, and exposure = 7 neutrons/b.

than calculated. The $(n,2n)$ and $(n,3n)$ reactions would not seem to help the situation since these lead to nuclei ^{241}Pu and ^{240}Pu, which are at least as fissionable as ^{242}Pu. While the 6.5% value could be somewhat reduced by increasing α for ^{243}Pu, we feel that the data suggest that a significant fraction of the ^{242}Pu target did not receive the full neutron exposure and/or that other sources of Pu isotopes were present.

The Np chain abundance of 5×10^{-8} $N_0(242)$ is substantially less than would have been predicted in I. For a ^{242}Pu$(n,p)^{242}$Np cross section of 1 mb, one would anticipate (I) the formation of about 10^{-3} ^{242}Np$/N_0(^{242}$Pu) and if 1% of this survived fast fission then a Np chain of about 10^{-5} $N_0(242)$ would be expected. While the (n,p) cross section of ^{242}Pu may not be as large as 1 mb and ^{242}Np may suffer somewhat more from fast fission, nevertheless the low abundances deduced for the Np chain suggest that it was depleted by a factor of about 100 in getting from ^{242}Np to ^{253}Np. If such fission competition is present in the Np isotopes, then when one uses a uranium target it may be that, contrary to our interpretation in I, Pa is more important than Np as an odd-Z capture chain.

It is worth bearing in mind that these conclusions as to the seriousness of fission competition depend on the Tweed exposure being near to that achieved in Vulcan. If, for example, the Tweed exposure were half that of Vulcan, there would be no evidence for serious fission competition in Tweed. We shall, however, proceed under the assumption that Tweed performed as described above.

3. SPONTANEOUS FISSION HALF-LIVES

At the present time there does not exist any generally accepted method for predicting either the spontaneous fission half-life of a nucleus or the energy barrier against neutron-induced fission. Various forms of the liquid-drop model[11–13] have been used with some success in correlating spontaneous fission half-lives of observed nuclei, but most unfortunately for predictive purposes, the departures of actual nuclear energies from those predicted by the liquid-drop model are of decisive importance. Other calculations have been made of half-lives based on single-particle energy levels in a deformed potential[14,15] and have shown encouraging agreement with experiment. However, at present there seem to be enough uncertainties in these calculations, such as the precise positions of energy levels, whether K is a good quantum number from equilibrium deformation to transition deformation, and the significance of the pairing interaction during deformation that for the present studies we have simply used a liquid-drop model with empirical shell corrections.

Swiatecki showed in 1955[11] that there is a good correlation between spontaneous fission half-lives and the liquid-drop fissility parameter, Z^2/A, provided that a correction is made for the deviation of the actual ground-state mass of a nucleus from the mass predicted by the liquid-drop model. Thus, Swiatecki considered $\log_{10}\tau_{1/2}+k\delta M$ as a function of Z^2/A, where $\tau_{1/2}$ is the spontaneous fission half-life, δM is the experimental mass minus the liqud-drop mass (as given by Green's[16] 1954 mass formula), and for a good correlation k was chosen to be $k=5-(Z^2/A-37.5)$, for δM in mMU and $\tau_{1/2}$ in sec. Slight modifications in this correlation have been suggested by Dorn,[12] and more recently Viola and Wilkins[13] have employed considerably more elaborate variations.

We have based our considerations on the recent deformed liquid-drop model with shell corrections, of Myers and Swiatecki.[10] Here the volume and surface energies are both taken to include a symmetry-dependent factor proportional to $[1-C((N-Z)/A)^2]$ so that the liquid-drop fissility parameter is now proportional to $Z^2/A[1-C((N-Z)/A)^2]$. While in principle one might prefer a dependence on symmetry, $(N-Z)/A$, in the nuclear surface energy which is different from the symmetry dependence in the volume energy, in practice the coefficient in the surface energy cannot be precisely determined[17] from empirical mass data. Furthermore, recent studies[18] of nuclear matter indicate that the assumption of equal symmetry dependences for surface and volume energies is a reasonable one. At one time we had hoped that the correlation of spontaneous fission half-lives might enable us to draw conclusions regarding the magnitude of C in the surface energy but, as will be seen, this is not the case.

At any rate, we have taken for our fissility parameter the quantity $X=Z^2/A[1-C((N-Z)/A)^2]$, where the value $C=1.7826$ was found by Myers and Swiatecki to give a good fit to empirical nuclear masses. For the moment we regard C as an adjustable parameter. We have then considered $\log_{10}\tau_{1/2}+k\delta M$ as a function of X, where δM is now the experimental mass minus the Myers and Swiatecki spherical liquid-drop mass. (We use units of MeV for δM and years for $\tau_{1/2}$ in this paper.) The factor k is treated as an adjustable constant which *a priori*[10,13] we may expect to have a value around 7.0 MeV^{-1}.

Spontaneous fission half-lives have been taken from the compilation of Hyde,[19] and experimental masses

[11] W. J. Swiatecki, Phys. Rev. **100**, 937 (1955).
[12] D. W. Dorn, Phys. Rev. **121**, 1740 (1961).
[13] V. E. Viola, Jr. and B. D. Wilkins, Nucl. Phys. **82**, 65 (1966).
[14] S. A. E. Johansson, Nucl. Phys. **12**, 449 (1959) and University of California Radiation Laboratory Report No. UCRL-10474 1962 (unpublished).
[15] J. R. Primak, Phys. Rev. Letters **17**, 539 (1966) and undergraduate thesis, Princeton University, 1966 (unpublished).
[16] A. E. S. Green, Phys. Rev. **95**, 1006 (1954).
[17] R. Brandt, F. G. Werner, M. Wakano, R. Fuller, and J. A. Wheeler, in *Proceedings of the International Conference on Nuclidic Masses*, edited by A. E. Duckworth (University of Toronto Press, Toronto, Canada, 1960).
[18] H. A. Bethe (private communication).
[19] E. K. Hyde, *Nuclear Properties of the Heavy Elements, III, Fission Phenomena* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Table 1.7.

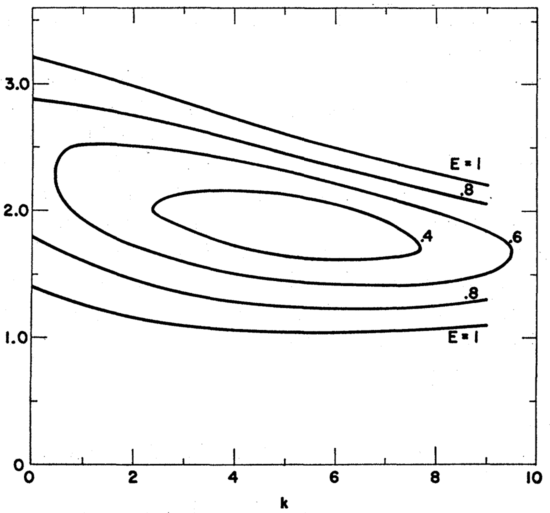

FIG. 2. Error in predicted spontaneous fission half-lives. Contours of the rms difference E between logarithms$_{10}$ of the predicted and experimental half-lives for the even-even nuclei are shown as a function of C (the coefficient of the symmetry dependence in the surface energy) and k (the coefficient connecting energy and half-life).

from the 1964 mass table of Mattauch[20] *et al.* We have made a slight adjustment of mass (+0.10 MeV) for ^{244}Pu and α related parents, based on a computation of Q_α (4.66 MeV) for ^{244}Pu.[21] We have also included ^{256}Fm ($\log_{10}\tau_{1/2}=-3.5$; mass excess = 85.395 MeV)[22] and ^{257}Fm ($\log_{10}\tau_{1/2}\simeq 2.0$; mass excess $\simeq$88.316 MeV).[22] Liquid-drop masses were taken from a listing kindly furnished us by Myers and Asaro.[23] The data are shown in Table III.

For twenty even-even nuclei having known $\tau_{1/2}$ and δM, we have considered $\log_{10}\tau_{1/2}+k\delta M$ as a third-order polynomial in X with coefficients to be determined by least squares. The root-mean-square error in the fit to the data has been considered as a function of C and k with results as shown in Fig. 2. Because of uncertainties in $\tau_{1/2}$ and δM, we regard all fits within the error contour $\simeq$0.4 as more or less equally good. We note that good fits can be obtained for values of C in a rather limited range, approximately $C=1.9\pm0.3$. On the other hand, acceptable values of k are found over quite a wide range, including at least $3\lesssim k\lesssim 7$ MeV^{-1}.

It might be thought that our finding of a limited range of acceptable C values, $\sim$1.9, is evidence for the existence and magnitude of the symmetry dependence of the surface energy. However, it will be recalled that in the liquid-drop model,[10] $C=1.7826$, and it turns out that we are to some extent just recovering a similar value. To show this, we computed a set of δM values with Green's[16] liquid-drop model in which $C=0$, and we found that for $k\gtrsim 5$ MeV^{-1}, the best correlation of spontaneous fission half-lives then occurs with $C=0$. This correlation has an rms error in $\log_{10}\tau_{1/2}$, $E=0.56$, which is not as good as that found with $k=3$, $C=1.0$ ($E=0.43$) nor as good as using the Myers-Swiatecki correlation ($E_{\min}\simeq 0.30$). Nevertheless, we cannot say that spontaneous fission half-lives provide any strong evidence for the symmetry dependence of the surface energy, and in what follows we will simply use the Myers-Swiatecki value of $C=1.7826$.

Our best correlation, found for $k=5.0$ MeV^{-1}, and giving an rms error (E) of $E=0.28$ for $\log_{10}\tau_{1/2}$ of twenty even-even nuclei, was

$$\log_{10}\tau_{1/2}+5\delta M=F_5(Y)=9.031-8.099Y+0.442Y^2+0.0052Y^3 \quad (3a)$$

with

$$Y=\frac{Z^2}{A[1-1.7826((N-Z)/A)^2]}-40.0. \quad (4)$$

We have also used the best correlation with $k=7.0$ MeV^{-1},

$$\log_{10}\tau_{1/2}+7\delta M=F_7(Y)=8.090-9.3614Y+0.140Y^2+0.1654Y^3 \quad (3b)$$

which leads to an rms error of $E=0.44$. In Table III, the half-lives obtained from these equations are compared with experiment.

Half-lives and δM are known for eight odd-A nuclei and these nuclei have systematically longer half-lives than would be predicted by Eqs. (3). A separate correlation may be made for these nuclei, but if one leaves out ^{235}U (which from our point of view has a peculiarly short half-life) then almost as good a correlation can be obtained by simply adding a constant, $\delta\log\tau$, to Eq. (3a) or (3b). The constant is $\delta\log\tau=4.34$ for Eq. (3a) and this yields $E=0.70$ for seven odd-A nuclei, as compared with $E=0.51$ for the best fit to the odd-A nuclei alone. For $k=7$, $\delta\log\tau=4.22$, yielding $E=1.2$.

The spontaneous fission half-life is not known for any odd-odd nucleus. For ^{254}Es, $\tau_{1/2}>2\times10^7 y$,[24] which is nearly a factor 10^8 larger than would be predicted for an even-even nucleus having the same value of Y. One might expect that the half-life of an odd-odd nucleus could be estimated from Eq. (3a) with $2\delta\tau$ added to the right-hand side and this is evidently consistent with the known limit for ^{254}Es.

The physical basis for a correlation such as we have used is presumably that the liquid-drop model gives a fairly good representation for deformations near the fission barrier. This is in agreement with the mass formula of Myers and Swiatecki[10] wherein it is assumed that departures from the liquid-drop model (shell effects) diminish with increasing deformation. In the

[20] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **65**, 1 (1965).
[21] E. K. Hyde, I. Perlman, and G. T. Seaborg, *Nuclear Properties of the Heavy Elements, I* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Eq. (4.12).
[22] T. Sikkeland, A. Ghiorso, R. Latimer, and A. E. Larsh, Phys. Rev. **140**, B277 (1965).
[23] Data in the listing were computed by W. D. Myers using the methods and parameters of Ref. 10.
[24] P. R. Fields *et al.*, Nucl. Phys. **A96**, 440 (1967).

context of a shell model, the single-particle energies are clustered most strongly, because of degeneracies, for spherical nuclei; with increasing deformation, the single-particle energies become more evenly spaced, leading to a loss of shell effects. In addition, if the nuclear pairing energy increases with increasing deformation,[25] as might be expected if pairing is an effect associated with the nuclear surface, then this will lead to a wider distribution[26] of partially occupied levels about the Fermi energy for the highly deformed nuclei, and hence to a smearing of shell effects. Thus, there are reasons for hoping that a liquid-drop model may be increasingly accurate for large deformations and that after correcting the ground-state energy for shell effects, (i.e., by δM) the liquid-drop model might then be used for estimating the height of the fission barrier. A possible expectation is sketched in Fig. 3.

Knowing the height of the barrier, one could hope for a 1-to-1 correspondence with half-life to complete the correlation. For example, with a barrier of parabolic shape having E_f the energy at barrier peak and E the ground-state energy (so that $E_f - E$ is the energy re-

TABLE III. Spontaneous fission results.

Z	A	N	δM[a] (MeV)	$\log_{10}\tau_{1/2}$(sf)(years) Exp.[b]	Calc. Eq. (3a)[c]	Calc. Eq. (3b)[c]
92	232	140	−0.170	13.9	14.07	14.01
	234	142	−0.349	16.2	16.00	16.39
	236	144	−0.111	16.3	15.80	15.78
	238	146	0.003	15.77	16.16	15.97
94	236	142	−0.741	9.54	9.40	9.37
	238	144	−0.885	10.68	11.09	11.53
	240	146	−0.667	11.1	10.93	11.09
	242	148	−0.505	10.83	11.00	10.98
	244	150	−0.227	10.4(10.8)[d]	10.44	10.00
96	240	144	−1.401	6.28	5.93	5.94
	242	146	−1.424	6.86	6.94	7.15
	244	148	−1.213	7.15	6.74	6.70
	246	150	−1.169	7.30	7.34	7.36
	248	152	−0.914	6.66	6.84	6.50
98	246	148	−1.930	3.32	3.39	3.80
	248	150	−1.919	3.85	4.11	4.56
	250	152	−1.812	4.24	4.32	4.62
	252	154	−1.162	1.92	1.79	0.87
100	254	154	−1.893	− 0.26	− 0.72	− 0.24
	256	156	−1.297	− 3.51	− 3.07	− 3.85
92	235	143	−0.285	17.26	(20.53)	(20.72)
94	239	145	−1.006	15.74	16.51	17.15
95	241	146	−1.183	14.36	13.86	14.19
97	249	152	−1.353	8.78	9.79	9.51
98	249	151	−2.105	9.18	9.77	10.49
99	253	154	−1.532	5.47	4.81	4.22
100	255	155	−1.867	4.08	3.82	4.08
100	257	157	−1.197	2.00	1.09	− 0.16
99	254	155	−1.36	> 7.3	8.6	7.6

[a] Experimental mass (see Ref. 20) minus liquid-drop mass (see Ref. 10) with minor changes as noted in text.
[b] Values from Ref. (19) with ^{257}Fm from (22) and (23), and ^{254}Es from Ref. (24).
[c] With $\delta \log\tau$ added for odd-A nuclei and $2\delta \log\tau$ for the odd-odd nucleus, ^{254}Es.
[d] Recent determination by P. R. Fields *et al.*, Nature **212**, 131 (1966).

[25] H. C. Britt, W. R. Gibbs, J. J. Griffin, and R. H. Stokes, Phys. Rev. **139**, B354 (1965).

[26] For example, S. G. Nilsson and O. Prior, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **32**, No. 16 (1960).

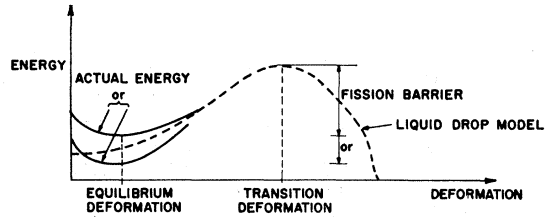

FIG. 3. An energy diagram of this sort may underlie the correlation of spontaneous fission half-lives. The deviations of the actual energy from the liquid-drop energy are assumed to be most important for small deformations, so that once one has corrected the ground state for shell effects the height of the barrier may be taken from a liquid-drop model.

quired to excite the nucleus from its ground state to an energy at which the barrier no longer needs to be penetrated), then[27]

$$\tau_{\rm sf} \simeq \tau_0(E) e^{2\pi(E_f - E)/\hbar\omega}, \qquad (5)$$

where $\hbar\omega$ is the characteristic oscillator energy and $\tau_0(E)$ is the mean life for the nucleus in the absence of barrier penetration.[27a] We expect[27,28]

$$\tau_0(E) = \frac{h}{\Gamma_f(E)} \simeq \frac{h}{D(E)}, \qquad (6)$$

where D is the spacing of levels having the appropriate quantum numbers in the initial nucleus. For levels near the ground state, $D \sim 1$ MeV and $\tau_0 \sim 10^{-20}$ sec.

From Eq. (5) we have

$$\log_{10}\tau_{1/2} = \log_{10}(0.7\tau_0) + \frac{2\pi}{\hbar\omega}(E_f - E) \times \frac{1}{2.3}, \qquad (7)$$

a form similar to Eq. (3a). If we take $E = E_{\rm LQD} + \delta M$ and $E_f = E_{f,\rm LQD}$, where the subscript LQD means calculated by liquid-drop model, then equating $\log_{10}\tau_{1/2}$ from Eqs. (3a) and (7), and equating coefficients of δM, we find

$$\log_{10}(0.7\tau_0) + \frac{0.87\pi}{\hbar\omega}(E_{f,\rm LQD} - E_{\rm LQD}) = F_5(Y), \qquad (8)$$

and $0.87\pi/\hbar\omega = 5$ MeV^{-1} or $\hbar\omega = 0.55$ MeV. Alternatively for $k = 7$ MeV^{-1}, $\hbar\omega = 0.39$ MeV. These values of $\hbar\omega$ are not in disagreement with other estimates of the same quantities, based on the rate of opening of fission channels. However, the smaller value is definitely in better agreement with correlations between barriers

[27] J. A. Wheeler, in *Fast-Neutron Physics*, edited by J. B. Marion and J. F. Fowler (Interscience Publishers, Inc., New York, 1963), Ch. V. 5.

[27a] *Note added in proof.* A nice discussion of this and more accurate expressions has just been given by J. R. Nix [Ann. Phys. (N. Y.) **41**, 52 (1967)]. Predictions of $\hbar\omega$ and correlations with experiment are also given.

[28] J. E. Lynn, in *Proceedings of the International Conference on Study of Nuclear Structure with Neutrons, Antwerp, 1965*, edited by M. Neve De Mevegnies *et al.* (North Holland Publishing Company, Amsterdam, 1966).

TABLE IV. Fission barriers.

Measure-ment	Comp. nucleus	Threshold[a] (MeV)	S_n[b]	Barriers (MeV) B(exp)	B_0[c]	B_{calc}[d]
n,f	^{233}Th	1.4	4.96	6.36	5.76	6.74
	^{232}Pa	0.5	5.52	6.02	4.82	6.04
	^{235}U	0.5	5.27	5.77	5.17	5.8
	^{237}U	1.0	5.30	6.30	5.70	6.0
	^{239}U	1.4	4.78	6.18	5.58	5.8
	^{238}Np	0.6	5.43	6.03	4.83	5.7
	^{239}Pu	0.4	5.62	6.02	5.42	5.3
	^{241}Pu	0.6	5.41	6.01	5.41	5.3
	^{243}Pu	0.7	5.05	5.75	5.15	5.15
	^{242}Am	0.9	5.47	6.37	5.17	5.1
	^{244}Am	0.9	5.29	6.19	4.99	4.9
d,pf	^{234}U	−1.1	6.78	5.68	5.68	5.8
	^{236}U	−0.3	6.47	6.17	6.17	5.7
	^{240}Pu	−1.2	6.46	5.26	5.26	5.1
γ,f	^{232}Th			5.9[e]	5.9	6.6
	^{233}U			5.5[e]	4.9	5.8
	^{238}U			5.8[e]	5.8	5.8
	^{237}Np			5.5[e]	4.9	5.4
	^{239}Pu			5.5[e]	4.9	5.3
	^{241}Am			6.0[e]	5.4	5.0

[a] n,f threshold from Ref. 32; d,pf from Ref. 31.
[b] Neutron separation energies from Ref. 20.
[c] Experimental barrier corrected for pairing by −0.6 and −1.2 MeV for odd-A and odd-odd nuclei, respectively.
[d] Calculated barrier is the saddle mass of Ref. 10 (a liquid-drop barrier) minus δM (actual ground-state mass minus liquid-drop mass).
[e] Data from Ref. 33.

and fission half-lives.[10,13] Of course, there is no reason for the barrier shape to be parabolic over a large range of deformations so we need not take the above numbers as more than indicative of the real situation.

It remains to discuss why the half-lives are systematically longer for nuclei with unpaired nucleons. This effect may be partly due to a larger pairing energy for transition-state nuclei than for ground-state nuclei. An increase in pairing energy of about 0.6 MeV has recently been suggested[25] and this would evidently explain most of the deduced effect. On the other hand, if one takes the single-particle calculations for fission seriously and assumes that K (the projection of the spin on the nuclear symmetry axis) is conserved during deformation, then the presence of one or more unpaired nucleons would increase the fission barrier. Thus, one has at least two possible reasons for the increased fission half-lives. Additional effects, associated with nuclear superfluidity have also been invoked for the increased half-lives of odd-A nuclei.[29]

Evidently in order to use the correlation in Eq. (3a) or (3b) for predicting half-lives, one must either know or estimate δM. We will return to this problem in Sec. 5. Conversely, if one knows $\tau_{1/2}$ then deductions may be made concerning δM, and we will try such an approach in Sec. 8.

4. BARRIERS AGAINST NEUTRON-INDUCED FISSION

Myers and Swiatecki[10] have already noted that their (revised) mass formula gives fission barriers which are in rather good agreement with experiment. To compute the barrier they use a liquid-drop value minus δM, where, as before, δM is the actual ground-state mass minus the liquid-drop value.

We have reexamined the comparison between these predicted barriers and experimental barriers. Unfortunately the experimental barriers are generally rather uncertain. There are first of all barriers deduced from positive-energy thresholds in neutron-induced fission. Here all of the compound nuclei involved have unpaired nucleons, and therefore there are uncertainties in which spins and parities are involved in fission with the lowest barrier. In some cases, the apparent thresholds may be due to requirements for neutrons of nonzero angular momentum to reach the lowest transition states and in every case there are presumably single-particle effects at the barrier which cannot be included in our method of prediction. Other thresholds have been deduced from (d,pf) reactions[30,31] where presumably the lowest transition state is 0^+. Even here the threshold seems uncertain[31] by perhaps a few hundred keV. Finally for the (γ,f) thresholds there is substantial difficulty in relating the observations to any physically clear barrier. Barriers have frequently been deduced from spontaneous fission half-lives,[10,13] but inasmuch as we have already considered the half-lives, we shall not include them again here.

The barriers which have been considered are shown in Table IV.[32,33] We have considered not only the experimental barrier, B, but also a barrier, B_0, corrected for a pairing energy which was assumed to be larger at the transition state than at the ground state by $\Delta=0.6$ MeV.[25] We have normalized B_0 to a value for even-even nuclei and have thus taken $B_0=B-n\Delta$, where n is the number of unpaired nucleons in the compound nucleus. Both B and B_0 are to be compared with the calculated barrier of Myers and Swiatecki, which it will be recalled includes an experimental δM.

From Table IV we observe that there is qualitative agreement between the calculated barriers and either B or B_0. Quantitatively the agreement is not so impressive and, in particular, it is not apparent whether the pairing correction helps or hinders the agreement. Except for the lighter nuclei, which are not of much interest here, we regard the pairing correction as generally helpful as well as physically plausible, and have therefore included it in the prediction of barriers.

We have, therefore, estimated fission barriers by taking the Myers-Swiatecki liquid-drop value, sub-

[29] M. G. Urin and D. F. Zaretsky, Nucl. Phys. 75, 101 (1966).

[30] J. A. Northrop, R. H. Stokes, and K. Boyer, Phys. Rev. 115, 1277 (1959).

[31] L. N. Usachev, V. A. Pavlinchuk, and N. S. Rabotnov, Zh. Eksperim. i Teor. Fiz. 44, 1950 (1963) [English transl.: Soviet Phys.—JETP 17, 1312 (1963)]; H. J. Specht, J. S. Fraser, and J. C. D. Milton, Phys. Rev. Letters 17, 1187 (1966); and see also Ref. 25.

[32] D. J. Hughes and R. B. Schwartz, *Brookhaven National Laboratory Report No. BNL-325* (U. S. Government Printing and Publishing Office, Washington, D.C., 1958), 2nd ed.; J. R. Stehn *et al.*, *ibid.* Suppl. 2.

[33] I. Halpern, Ann. Rev. Nucl. Sci. 9, 259 (1959).

TABLE V. Shell corrections, δM.[a]

N\Z	Th 90	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101
140	0.283	0.256	−0.170	−0.428	−0.477							
141	0.361	0.108	−0.468	−0.653	−0.732	−1.283±1						
142	0.483	0.147	−0.349	−0.519	−0.741	−1.200	−1.209					
143	0.444	0.150	−0.285	−0.651	−0.991	−1.366±1	−1.402±1.1					
144	0.574	0.496	−0.111	−0.542	−0.885	−1.183	−1.401	−1.586±1				
145		0.855	−0.330	−0.677	−1.006	−1.446±1	−1.708	−1.967±1	−1.931±1.1			
146		0.695	0.003	−0.266	−0.667	−1.183	−1.424	−1.551	−1.789	−1.917[b]±1		
147		(0.62)	0.063	−0.346	−0.830	−1.204	−1.435	−1.715±1	−1.999	−2.457[b]±1		
148		(0.84)	0.275	−0.100	−0.505	−0.793	−1.213	−1.614	−1.930	−1.911[b]	−2.132[b]	
149		(0.78)	(0.21)	0.029±1	−0.542	−0.869	−1.498	−1.744±1	−2.045±1	−2.391±1	−2.083[b]±3	
150		(1.01)	(0.44)	(0.26)	−0.227	−0.592	−1.169	−1.543	−1.919	−2.265	−2.356[b]	−2.485[b]±1.4
151		(0.94)	(0.37)	(0.19)	−0.154±1	−0.593	−1.206±1	−1.722	−2.105	−2.382±1.4	−2.884[b]±1	−2.995[b]±1.4
152		(1.21)	(0.64)	(0.46)	−0.051	−0.144±1.4	−0.914	−1.353	−1.812	−2.177	−2.442	−2.873[b]±1
153		(1.31)	(0.74)	(0.56)	(0.05)	(−0.04)	−0.807	−1.130	−1.872±1	−2.040	−2.536	−2.675[b]±1.7
154		(1.95)	(1.38)	(1.20)	(0.69)	(0.60)	(−0.17)	−0.431±1.4	−1.162	−1.532	−1.893	−2.569
155		(2.08)	(1.51)	(1.33)	(0.82)	(0.73)	(−0.04)	(−0.30)	−0.945	−1.364	−1.867±1	(−2.44)
156		(2.65)	(2.08)	(1.90)	(1.39)	(1.30)	(0.53)	(0.27)	(−0.381)	(−0.80)	−1.297	(−1.87)
157		(2.75)	(2.18)	(2.00)	(1.49)	(1.40)	(0.63)	(0.37)	(−0.28)	(−0.70)	−1.197	(−1.77)

[a] Values of δM as in Table III, with extrapolation (in brackets) to higher N. Extrapolation assumed δM to be the sum of a function of N and a function of Z.
[b] Value not used to determine extrapolation.

tracting δM, and adding $0.6n$, where $n=0, 1, 2$ for even-even, odd-A and odd-odd nuclei, respectively. This procedure has been used to obtain barriers for all the heavy nuclei which have measured thermal neutron capture and/or fission cross sections.[34] (The results are indicated in Table VI.) By comparison of the barrier with the neutron binding energies we have estimated whether the nuclei will fission upon absorption of a thermal neutron, and compared the prediction with experiment. In general, the agreement between prediction and experiment is quite encouraging, although there are a number of instances in which an adjustment of the calculated barrier by a few hundred keV would seem to give the observed capture-to-fission ratios much more naturally (see Sec. 7).

Once again, in order to predict fission barriers one must know δM, the shell correction.

5. EXTRAPOLATION OF δM, THE SHELL CORRECTION

Values of the shell correction, δM, which is the experimental nuclear mass minus the Myers and Swiatecki spherical liquid-drop mass, are given in Fig. 4 and Table V for known heavy nuclei having more than 139 neutrons. In Fig. 4, it is observed that δM systematically decreases with increasing Z. This important effect, which makes the increase in fissionability much slower with Z than predicted by liquid-drop models, is presumably due to the clustering of single-particle levels as described by Myers and Swiatecki[10] and is largely reproduced in their calculated shell corrections.

It may also be observed that δM seems to increase rather gradually with neutron number (for constant Z) up to $N=153$, after which it increases quite rapidly. This important effect may lead to an increase of fissionability with increasing neutron number (and constant Z), contrary to the predictions of a liquid-drop model. It is presumably due to the details of shell or single-particle effects, such as a 152 neutron gap, but is not included in the shell corrections of Myers and Swiatecki. One may hope to be able to calculate these effects on δM, following a method recently proposed by Strutinskii.[35] In this method, the difference is found between

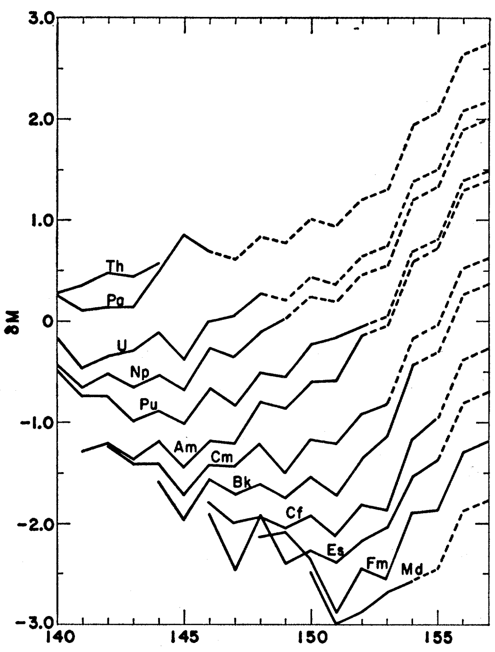

FIG. 4. The shell correction δM, in MeV. Solid lines connect experimental values of δM (experimental mass minus liquid-drop mass); dashed lines connect extrapolations based on the assumption that δM is a function of neutron number N plus a function of proton number Z, as explained in text.

[34] Reference 19, Table 1.6.

[35] V. M. Strutinskii, Yadern. Fiz. 3, 614 (1966) [English transl.: Soviet J. Nucl. Phys. 3, 449 (1966)]; Nucl. Phys. A95, 420 (1967).

TABLE VI. Neutron-binding energies S_n and fission barriers B. Both S_n and B are in MeV. Neutron binding energies S_n are experimental values from Ref. 20 where available; otherwise they are calculated from δM values in Table V and liquid-drop masses in Ref. 10. "f" denotes in an (A,Z) entry that experimentally the target nucleus $(A-1,Z)$ is fissionable upon absorbing a thermal neutron, i.e., $\alpha \lesssim 1.0$ as given in Ref. 34. "x" denotes that the target nucleus does not fission upon absorbing a thermal neutron, "?" indicates incomplete data, and a blank indicates no data.

A	Th(90)			Pa(91)			U(92)			Np(93)			Pu(94)			Am(95)		
Compound	S_n		B	S_n		B	S_n		B	S_n		B	S_n		B	S_n		B
228	7.1	f	6.5															
229	5.4	x	7.1															
230	6.7	f?	6.6															
231	5.1	x	7.2	6.7	f	6.4	5.9	?	5.9									
232	6.4		6.6	5.5	x	7.2	7.2	f	5.4									
233	5.0		7.3	6.7	f?	6.9	5.9	f?	6.4									
234	6.1		6.7	5.1		7.4	6.8	f	5.8									
235				6.1	?	6.5	5.3	x	6.4	7.0	f	5.9						
236				4.5		6.8	6.5	f	5.7	5.7		6.7						
237				6.4		6.5	5.3	x	6.6	6.7	f	6.0	6.0	f	5.8			
238				4.7		7.3	6.1		5.8	5.4	x	6.9	6.9	f	5.1			
239				5.7		6.5	4.8	x	6.4	6.2	f	5.9	5.6	x	5.9			
240				4.5		7.3	5.9		5.7	5.1		6.7	6.4	f	5.1			
241				5.5		6.5	4.7		6.4	6.1		5.9	5.4	x	5.9			
242				4.2		7.2	5.7		5.7	4.7		6.5	6.2	f	5.0	5.5	x	6.3
243				5.2		6.4	4.5		6.4	5.9		5.7	5.0	x	5.7	6.3	f	5.3
244				3.9		7.0	5.4		5.6	4.6		6.4	6.0		4.9	5.3	x	6.1
245				4.6		5.8	4.0		6.2	5.6		5.6	4.7		5.5	6.2	f	5.2
246				3.6		6.4	4.8		5.0	4.2		6.2	6.0		4.9	5.0		5.9
247				4.5		5.3	3.8		5.5	5.0		5.0	4.4		5.4	5.8		4.9
248				3.4		5.8	4.6		4.4	4.0		5.5	5.2		4.2	4.6		5.5
249							3.6		5.0	4.8		4.4	4.2		4.8	5.4		4.3
250										3.8		5.0	5.0		3.7	4.4		4.8
251													4.0		4.2	5.2		3.7
252																4.2		4.3
	Cm(96)			Bk(97)			Cf(98)			Es(99)			Fm(100)			Md(101)		
243	5.7	x?	5.4															
244	6.7	f	4.6															
245	5.7	x	5.5															
246	6.4	f	4.7															
247	5.2	x	5.4	6.7		5.1												
248	6.2	f?	4.5	5.5		6.0	7.0		4.4	6.5		5.6						
249	4.8		5.1	6.3		5.0	5.8		5.4	7.2		5.0						
250	5.6		3.9	4.9		5.5	6.5	f	4.4	5.9		5.7	7.8		4.1			
251	4.6		4.4	5.7		4.2	5.4	?	5.1	6.8		5.0	6.5		5.2			
252	5.4		3.3	4.8		4.7	5.9	f	3.9	5.4		5.5	6.8		4.3	6.7		5.6
253	4.4		3.8	5.6		3.6	4.9		4.3	6.3		4.4	5.9		5.0	7.3		5.0
254				4.6		4.2	5.8		3.2	5.1		4.9	6.3		3.8	5.7		5.4
255							4.8		3.7	6.0	f	3.8	5.5		4.4	7.4		4.7
256										5.0		4.3	6.2		3.3	5.5		5.2
257													5.2		3.8	6.4		4.1
258																5.4		4.7

the sum of the single-particle energies in a deformed shell model with a residual (pairing) interaction and the energy of a corresponding liquid drop. This difference should be just an estimate of δM which includes all the shell effects. It would appear that some such calculations of δM may have to be made before one can have confidence in any extensive extrapolation of δM, or indeed any extrapolation involving neutron or proton numbers which have not been observed.

However, for obtaining estimates of δM for nuclei which are (1) not far removed in A or N from nuclei with known δM and (2) have values of Z and N which are included in the known nuclei we have followed Cameron[36] in assuming that δM is the sum of a neutron plus a proton shell correction. Some confidence in such an assumption can be gained from Fig. 4 in which the curves for constant Z are seen to be roughly parallel to each other. Some of the more pronounced deviations from parallelism are associated with nuclei having large uncertainties in their experimental masses.

In Strutinskii's calculation,[35] the neutron and proton shell corrections are calculated independently and then added to determine the equilibrium deformation as that deformation which gives minimum energy. The neutron and proton shell corrections to the energy of the ground state are therefore independent of one another except insofar as each shell correction plays a role in determining the equilibrium deformation at which both shell corrections are to be computed. We may thus expect that as long as we consider nuclei which have roughly the same deformation, the neutron and proton shell corrections will be nearly independent of one another. The known heavy nuclei in Table V have similar deforma-

[36] A. G. W. Cameron, Can. J. Phys. 35, 1021 (1957); A. G. W. Cameron and R. M. Elkin, *ibid.* 43, 1288 (1965); and Goddard Institute for Space Studies Report, 1965 (unpublished).

TABLE VII. Predicted spontaneous fission half-lives. Entries are $\log_{10}\tau_{sf}$(years), calculated from Eq. (3a) with δM from Table V. $\delta \log_{10} \tau$ has been added to the result for odd-A and $2\delta \log_{10} \tau$ for odd-odd nuclei with $\delta \log_{10} \tau = 4.34$.

$N \backslash Z$	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101
140	21.0	14.1									
141	26.6	20.4									
142	22.5	16.0	16.7								
143	27.3	20.5	22.1	15.5							
144	21.8	15.8	17.7	11.1							
145	24.8	21.7	23.4	16.5							
146	21.7	16.2	17.5	10.9							
147	26.8	20.6	22.6	16.6	18.7	11.8					
148	21.8	15.7	17.5	11.0	12.7	6.7					
149	26.9	20.8	21.6	15.9	17.8	12.9			11.6		
150	21.9	15.8	16.5	10.4	12.5	7.3	10.0	4.1	7.1	0.3	
151	27.0	21.0	21.6	14.8	17.2	12.2	15.6	9.8	12.3	7.6	9.8
152	21.6	15.6	16.3	10.3	11.2	6.8	9.8	4.4	7.3	1.4	5.3
153	25.7	19.8	20.6	14.5	15.3	11.0	13.3	9.3	11.3	6.5	9.0
154	18.5	12.6	13.4	7.4	8.0	3.8	5.9	1.8	4.8	−0.7	4.4
155	22.6	16.7	17.5	11.4	12.0	7.9	10.0	5.4	8.6	3.8	8.3
156	15.8	9.8	10.6	4.7	5.1	1.0	3.2	−1.4	1.7	−3.1	1.4
157	19.9	14.0	14.9	8.9	9.4	5.3	7.3	2.8	6.0	1.1	5.5

tions and are thus expected to have approximately additive shell corrections. As one adds more neutrons or protons, he may refer to some energy-level diagram of the Nilsson type in order to estimate how many nucleons he may add before the equilibrium deformation is much changed. From a recent diagram of this type,[37] we estimate that adding neutrons up to $N \lesssim 160$ and protons to $Z \lesssim 104$ will not lead to substantial changes in deformation, while for larger N and/or Z the nuclei may become considerably more spherical. This very qualitative conclusion gives us some courage to extrapolate δM.

Assuming that the neutron and proton shell corrections are additive we have extrapolated δM to neutron-rich nuclei having $91 \leq Z \leq 101$ and $N \leq 157$. Neutron number 157 is the highest for which an accurate mass is known (^{257}Fm). These extrapolations are included in Fig. 4 and Table V.

It may be observed in Fig. 4 that there is a tendency for the extrapolated values to cluster in the pairs (U,Np), (Pu,Am), and to a lesser extent (Cm,Bk). Since such clustering is not seen in the experimental values of δM, we presume that it is not a real effect but is a coincidence due to errors in the experimental masses for the heaviest isotopes from which the extrapolations started. In particular the heaviest listed[20] nuclei of Np, Am, and Bk (^{242}Np,^{247}Am,^{251}Bk) have large uncertainties in their experimental masses; if their masses were decreased by about [0.15 MeV, 0.25 MeV, 0.2 MeV, respectively] the clustering would be removed. It is possible that the correspondingly adjusted extrapolated δM values for Np, Am, and Bk would be more accurate than those shown in Fig. 4. However, it will be seen that such small adjustments do not have an important bearing on our results. The sawtooth structures in Fig. 4 presumably indicate that the neutron-pairing energy has been taken to be too large by roughly 0.2 MeV.

6. FISSIONABILITY OF NEUTRON-RICH NUCLEI

Now that we have estimated some values of δM for neutron-rich nuclei, we may combine these with the liquid-drop barriers of Myers and Swiatecki, as discussed in Sec. 4, to obtain estimated fission barriers. By comparing these with the corresponding neutron-binding energies, we may estimate whether the various nuclei are fissionable upon absorption of slow neutrons. The comparison between fission barriers and neutron-binding energies is given in Table VI.

From the results in Table VI, we may draw the qualitative conclusion that for any given Z, the difference between the neutron binding energy and the fission threshold does not change much when one adds a pair of neutrons. More specifically we conclude that: (1) None of the listed Pa nuclei are fissionable upon absorption of slow neutrons; (2) none of the listed even-A uranium target nuclei are fissionable while the odd-A uranium target nuclei are marginally nonfissionable; (3) the odd-A neptunium target nuclei are nonfissionable while the even-A neptunium nuclei may or may not fission; (4) the even-A plutonium nuclei are nonfissionable while those of odd A will fission; and (5) odd-A americium nuclei will not fission but all those of even A will fission. All these conclusions are deduced for nuclei having $N \leq 157$. The marked increases in δM at $N=154$ and $N=156$ are in addition associated with increases in fissionability. In the next section we will estimate capture-to-fission ratios for some of the nuclei of interest.

It will be recalled that we have included in our calculated barriers a pairing energy increase of 0.6 MeV for odd-A compound nuclei and 1.2 MeV for odd-odd

[37] C. Gustafson, I. L. Lamm, B. Nilsson, and S. G. Nilsson, paper presented at Conference on Why and How Should We Study Nuclides Far Off the Stability Line?, Lysekil, Sweden, August 1966 (unpublished).

TABLE VIII. Number of open channels N versus excitation energy. In these estimates, explained more fully in the text, E^x is the excitation energy (in MeV) above the fission barrier and N is a rough average over spin and parity of the number of open channels of fixed spin and parity.

A. Even-even compound nuclei	
$0 \leq E^x < 0.6$	$N = \frac{1}{2}$
$0.6 \leq E^x < 1.0$	1
$1.0 \leq E^x < 1.6$	2
B. Odd-A compound nuclei	
$0 \leq E^x < 0.4$	$N = \frac{1}{2}$
$0.4 \leq E^x < 0.7$	1
$0.7 \leq E^x < 1.0$	2
$1.0 \leq E^x < 1.3$	3
$1.3 \leq E^x < 1.6$	4
C. Odd-odd compound nuclei	
$0 \leq E^x < 0.3$	$N = \frac{1}{2}$
$0.3 \leq E^x < 0.5$	1
$0.5 \leq E^x < 0.7$	2
$0.7 \leq E^x < 0.9$	3
$0.9 \leq E^x < 1.0$	4

compound nuclei. If such an allowance were not included, the main effect for our purposes would be to increase the fissionability of the even-A Np nuclei, making them more unambiguously fissionable, and to also increase the fissionability of Am nuclei.

With the extrapolated values of δM we may also compute spontaneous fission lifetimes and the results, using Eq. (3a), are listed in Table VII. As expected from δM, there is a substantial drop in τ_{sf}, as one adds neutrons with $N > 153$. Of the half-lives shown in Table VII, only that of ^{256}Fm is short compared to the times required for recovery and processing of debris. For nuclei with $N \geq 158$ and/or $Z \geq 102$, half-lives may be still shorter and we will return to this aspect in Sec. 8.

A few of the calculated spontaneous fission half-lives in Table VII may be compared with experimental values which were not listed in Table III, either because the δM values were unknown or τ_{sf} was not available when the correlation was made. These experimental values of $\log_{10}\tau_{1/2}$(sf) are: ^{254}Cf(−0.8),[19] ^{250}Cm(4.0),[38] ^{249}Cf(10.8),[38] ^{255}Es(3.4),[38] and ^{252}Fm(2.1).[38] From Table VIII, it can be seen that we have underestimated all these half-lives; by far the worst disagreement is for ^{255}Es, where our underestimate is by a factor of about 50, otherwise we are within a factor of 10.

7. CAPTURE-TO-FISSION RATIOS

Estimates of neutron capture and fission cross sections may be made from the statistical model once the capture and fission level widths, Γ_γ and Γ_f, are known for the appropriate states of the compound nucleus. For Γ_γ, which varies only slightly between heavy nuclei, we may use the values given in I. The fission width is considerably more complicated. For states of a compound nucleus having given spin and parity, one may estimate[27,28] the average fission width

$$\langle \Gamma_f{}^{J\pi} \rangle = \frac{D^{J\pi}}{2\pi} N^{J\pi}, \tag{9}$$

where $D^{J\pi}$ is the average level spacing, which may, for example, be taken from Eq. (10) of I, and $N^{J\pi}$ is the number of open channels. The number of open channels may in turn be estimated if one postulates some spectrum of transition states. Unfortunately the spectrum of transition states is largely unknown. Therefore, we have been content with a very schematic description of $N^{J\pi}$. It may be noted that the number of open channels will determine not only the average fission width $\langle \Gamma_f{}^{J\pi} \rangle$, but also the distribution of fission widths about the average. Thus, for N open channels we expect[39,40] the fission widths to have a χ^2 distribution with N degrees of freedom; with $x = \Gamma_f / \langle \Gamma_f \rangle$ and $\rho = 2N$

$$P(x)dx = \Gamma(\rho)(\rho x)^{\rho-1} e^{-\rho x} \rho dx. \tag{10}$$

Estimates of $N^{J\pi}$ were made as follows, and are summarized in Table VIII. The values of N are supposed to represent in a qualitative sense an average over spin and parity of the number of open channels. For comparison with the results of nuclear explosions, we are interested in fission by both s- and p-wave neutrons[4] so that both parities and several spins of the compound system will be involved. For even-even compound nuclei we referred to the spectrum of transition states given by Lynn.[28] A particular vibrational state plus its associated rotational band counts as an open channel only for states of one parity. Therefore, N in Table VIII is roughly half the number of vibrational states lying below the available excitation energy. By $N = \frac{1}{2}$ we mean one channel, half open, so that for this case N equal $\frac{1}{2}$ in Eq. (9) but N equals unity in Eq. (10).

For odd-A compound nuclei, we expect that the density of intrinsic transition states will be about the same as the density of single-particle states for near equilibrium deformation, namely about 5 MeV^{-1}. Each of these intrinsic states may be combined with collective states and a rough counting led to the enumeration in Table VIII. For odd-odd compound nuclei, the situation is much the same except that we may expect the density of the intrinsic two quasiparticle states to increase linearly with energy, roughly as $25E$, per MeV.

Using Table VIII to obtain the number of open channels, $\langle \Gamma_f \rangle$ is at once found from Eq. (9). The ratio of capture-to-fission cross sections, α, is not, however, simply equal to $\Gamma_\gamma / \langle \Gamma_f \rangle$ because the fission widths vary widely from resonance to resonance while Γ_γ is nearly constant. The result is that α may be substantially

[38] A. M. Friedman, in Proceedings of the International Symposium on Transplutonium Elements, Oak Ridge, Tennessee, November 1966 (unpublished), communicated by J. D. Knight.

[39] C. F. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).
[40] J. D. Garrison, Ann. Phys. (N.Y.) **30**, 269 (1964).

larger than $\Gamma_\gamma/\langle\Gamma_f\rangle$. If we assume that the cross sections are given by a sum of single-level Breit-Wigner terms and moreover neglect Γ_n compared to $\Gamma_\gamma+\Gamma_f$, then α is readily calculable[41] and the results are shown in Fig. 5. If we had not neglected Γ_n, α would have been smaller (approaching $\Gamma_\gamma/\langle\Gamma_f\rangle$ in the limit of large Γ_n), while by neglecting interference between levels we have probably underestimated α.[42,43] At any rate we have used Fig. 5 to estimate capture-to-fission ratios for a variety of nuclei.

It may be worthwhile to recapitulate the method of calculation. First, for a compound nucleus, the neutron binding energy, S_n, and fission barrier, B, are taken from Table VI and the available excitation energy is then $E^x=S_n-B$. N is then taken from Table VIII, while Γ_γ and D are taken from Eq. (10) and Fig. 4 of I. For simplicity, we assume in computing D that for target nuclei which are even-even, odd, or odd-odd, the spins of the compound nuclei are $\frac{1}{2}$, 3, and $\frac{5}{2}$, respectively. $\langle\Gamma_f\rangle$ is next calculated from Eq. (9), and α is then obtained from Fig. 5.

Results, for a variety of fissile nuclei, are given in Table IX. Several features are worth commenting upon. First of all we may compare some of the estimated capture-to-fission ratios with experiment. Capture-to-fission ratios have been measured[32,44] as a function of incident neutron energy for ^{233}U, ^{235}U, and ^{239}Pu, and if we compare our estimates of α with the measured values for neutron energies around 10–20 keV, we find fair agreement as shown in Table IX. At lower neutron energies α is, for ^{239}Pu, substantially larger than our estimate, but this is presumably because ^{239}Pu is a $\frac{1}{2}^+$ target and the 1^+ fission channel has an unusually high barrier.[28,42]

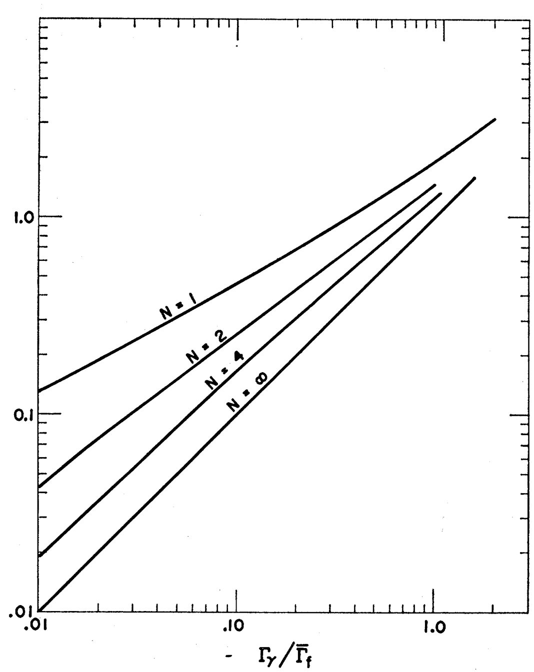

FIG. 5. Capture-to-fission ratios assuming $\Gamma_n \ll \Gamma$. The ratio α of the average capture cross section to the average fission cross section is shown as a function of the ratio of capture to average fission width for a sequence of isolated Breit-Wigner levels. N is the number of open fission channels and the distribution of fission widths is χ^2 with N degrees of freedom.

TABLE IX. Capture-to-fission ratios, α.

Target nucleus		D^a (eV)	No. of open channels, N	α(calc)	α(exp)
U	233	1.6	2	0.2	0.12[b]
	235	2.2	1	0.4	0.35[b]
	237	3.7	$\frac{1}{2}$	0.4	
Np	234	0.6	3	0.4	
	236	0.8	2	0.3	
	238	1.4	$\frac{1}{2}$	1.0	
	240	1.6	$\frac{1}{2}$	0.9	
	242	2.0	$\frac{1}{2}$	0.8	
Pu	239	2.5	2	0.2	0.35[b]
	241	3.1	2	0.1	$\sim$0.3[c]
	243	4.0	2	0.1	0.3[d]
	245	3.8	2	0.1	0.14[d]
	247	11	2	0.04	0.17[d]
	249	14	2	0.03	0.02[d]
Am	242	1.2	3	0.2	$\sim$1.0[c]
	244	1.3	3	0.2	
	246	2.2	2	0.2	
	248	3.6	3	0.05	
	250	4.7	4	0.02	
Cm	243	1.6	>2	$\lesssim$0.2	$\sim$0.5[c]
	245	2.3	$\gtrsim 2$	$\lesssim$0.2	$\sim$0.07[c]
	247	2.9	$\gtrsim 2$	$\lesssim$0.1	$\sim$1.0[c]
	249	6.1	$\gtrsim 2$	$\lesssim$0.06	
	251	8.0	$\gtrsim 2$	$\lesssim$0.05	
Bk-	250	2.4	4	0.04	
	252	2.7	$\gtrsim 4$	$\lesssim$0.03	
Cf	249	1.9	$\gtrsim 2$	$\lesssim$0.2	$\sim$0.2[c]
	251	4.2	$\gtrsim 2$	$\lesssim$0.1	$\sim$1.0[c]
	253	4.7	$\gtrsim 2$	$\lesssim$0.08	
Es	252	1.1	$\gtrsim 4$	$\lesssim$0.1	
	254	1.6	$\gtrsim 4$	$\lesssim$0.07	$\sim$0.02[c]
	255	2.7	3	0.04	
Fm	254	6.2	2	0.07	
	255	2.7	$\gtrsim 2$(4)	$\lesssim$0.14(0.04)	
	256	9.2	4	0.01	

[a] D is the level spacing in the compound nucleus at an excitation energy equal to the neutron-binding energy.
[b] 20-keV value from Ref. 23.
[c] Thermal-neutron value from Ref. 34.
[d] Estimated from Tweed, as in Table I.

Capture-to-fission ratios are known[34] for other nuclei exposed to thermal neutrons. These values are very uncertain in many cases, including ^{242m}Am and ^{254}Es. Moreover, they may represent the ratio of capture to fission widths in only one or a few nearby dominant resonances and thus not be typical of the results averaged over many resonances. The quoted values for ^{247}Cm and ^{251}Cf seem peculiarly large compared with our estimates, but it is difficult to draw any general conclusions from comparison with the rather uncertain α values for thermal neutrons.

For heavy plutonium nuclei, we may compare our estimates of α with the values obtained from the analyses

[41] S. Oleksa, J. Nucl. Energy: Pt. A 5, 16 (1957).
[42] G. I. Bell, Rev. Mod. Phys. 39, 59 (1967).
[43] J. D. Garrison, paper presented at ANS Topical Meeting on Reactor Physics in the Thermal and Resonance Regions, San Diego, California, February 1966 (unpublished).
[44] G. de Saussure *et al.*, Nucl. Sci. Eng. 23, 45 (1965).

of Tweed, reported in Table I. In general the agreement is encouraging, though we would seem to have underestimated α for ^{247}Pu.

As is evident from Eq. (9), the fission width may increase either because the level spacing, D, increases or because the number of open channels, N, increases. In Table IX it can be seen that sometimes the increase in the level spacing is the more important effect. In particular, for the sequence of Pu nuclei the number of channels remains unchanged while α decreases by a factor of seven; most of this change is simply caused by the increase in D, although a minor part is caused by a 35% decrease in Γ_γ.

For those cases in which the excitation energy, E^x, lies outside the range considered in Table VIII, we have simply used the maximum number of channels in Table VIII and indicated that the value of α so obtained is likely to be an upper limit.

While the estimates of α which are given in Table IX are clearly quite rough, as is indicated by quoting them to only one significant figure, we suggest that the predicted trends are more or less correct. In particular, it appears that the capture-to-fission ratios of Pu and Am nuclei may well decrease substantially as one adds neutrons in a capture chain. We do not regard our estimates of fission barriers for neutron-rich U and Np nuclei to be accurate enough to enable us to draw conclusions concerning their fissionability.

8. CONCLUSIONS AND EXTRAPOLATIONS

The general purpose of the foregoing exercises was to understand the role of fission in heavy element formation by rapid multiple neutron capture. We started out with the notion that the Tweed and Cyclamen experiments indicated that nuclei of plutonium and americium retained or increased their fissility as one added pairs of neutrons. These notions have been confirmed by the theoretical estimates of capture-to-fission ratios given in Table IX. Moreover, it does not appear that any escape from the disastrous fission competition will be found by going to target nuclei with $Z>95$. Rather, one is inclined to ask whether fission competition may not be already depleting the capture chains in U and Np. We do not feel that our calculations are accurate enough to answer this question.

It remains to speculate as to whether the spontaneous fission lifetimes of nuclei in the β-decay chains having $A>257$ may be so short as to render them undetectable by current techniques. The retentive reader will recall that we required the mass of a nucleus in order to predict its spontaneous fission lifetime, and that we had no method for predicting masses for nuclei having more than 157 neutrons. Now there is some negative information on nuclei with more than 157 neutrons: (1) From a failure to detect spontaneous fission of ^{258}Fm, formed by ^{257}Fm$(n,\gamma)^{258}$Fm, it has been concluded[8,45] that τ_{sf} is less than or of the order of seconds for ^{258}Fm. (2) The failure to detect mass 259 in Cyclamen debris could be understood[9] if $\tau_{sf}\lesssim 5$ h for ^{259}Fm, though this is by no means a necessary conclusion. (3) The mass 257 nuclei recovered from explosions as ^{257}Fm were presumably formed[4] as ^{257}Pa or ^{257}Np and hence with 266 or 264 neutrons. From the recovery of mass 257 and lighter nuclei in roughly expected amounts, we may conclude that neither neutron-induced fission in the capture chain, nor spontaneous fission in the β-decay chain have introduced any drastic depletion. These results place some constraints on allowable nuclear masses, or δM values. However, as previously noted, it may be that ^{257}Pa, for example, is much less deformed than ^{266}Fm so that they do not have nearly the same neutron shell correction. In general we have found the constraints to be so gross as to be not very useful.

We have attempted to employ this negative information as follows. First of all, for any spontaneous fission half-life we may calculate δM from Eq. (3a), and we have done this assuming half-lives of 1 sec and 5 h for ^{258}Fm and ^{259}Fm, respectively. The corresponding values of δM turn out to be -0.3 MeV and -0.22 MeV and by referring to Fig. 3 or Table V we see that these values are larger than those for ^{256}Fm and ^{257}Fm, respectively, by about $+1.0$ MeV. This would indicate that the trend in $\delta M(N)$ which was seen in Fig. 4 is not only continuing but is even accelerating somewhat. Since these deduced values of δM continue a trend and are not far from zero, they seem quite reasonable. From them and our previous method of calculation, we have deduced that the spontaneous fission half-life of ^{256}Cf is about 30 sec. For ^{259}Md and ^{260}Md we find spontaneous fission half-lives of about 10 h and 15 y, respectively. We have also estimated the capture to fission ratio of ^{257}Fm, finding an excitation energy of 3.2 MeV above the fission barrier. For four open channels this leads to $\alpha\simeq 0.02$ and it would seem likely that N is larger and α smaller.

In order for β stable nuclei having 161, 163, $\cdots$ neutrons to live longer than a few hours, it appears that the $\delta M(N)$ curves in Fig. 3 must at least level off after $N=159$. When and whether this happens would seem to be answerable only by experiment or perhaps by a detailed calculation.

Recent progress[46,47] in determining the decay properties of isotopes of element 102 makes it possible to extend our predictions of spontaneous fission lifetimes to these nuclei. Happily the workers at Berkeley and Dubna now seem to agree on the α-decay energies and half-lives for masses 252 through 256. The results are given in Table X.[48] Q_α was taken from recent Dubna[46] and Berkeley[47] reports, and from this we obtained the

[45] E. K. Hulet (private communication and Ref. 22).

[46] G. N. Flerov, in Proceedings of the Dubna Conference on Heavy-Ion Physics, October 1966, communicated by G. A. Cowan (unpublished).

[47] A. Ghiorso, T. Sikkeland, and M. J. Nurmia, Phys. Rev. Letters **18**, 401 (1967).

[48] G. N. Flerov *et al.*, At. Energ. (USSR) **17**, 310 (1964).

nuclear masses for $^{252}102$ through $^{257}102$. For the odd-A nuclides, it was assumed that the α decay proceeded to the daughter ground state. If the decay is primarily to excited states, then the odd-A masses may be slightly larger than indicated. Judging from the δM values the odd-A masses have not been much underestimated but perhaps they should be larger by a few tenths of a MeV. The resulting shell corrections, δM, were extrapolated from $^{256}102$ out to $N=159$, $A=261$, as before and τ_{sf} was then obtained as in Sec. 3. The predicted spontaneous fission half-life for $^{256}102$ is about 1200 sec, in reasonable agreement with the observed half-life of 1500 sec,[48] while the predicted half-life of $^{258}102$ is 2 sec which may be compared with Ghiorso's estimate[47] of much less than a second. Note, however, that for $^{256}102$ we are using an experimental shell correction, not an extrapolated one.

Flerov *et al.*[48] have reported a spontaneous fission half-life of 0.3 sec for $^{260}104$. From this half-life we have deduced the shell correction, extrapolated the shell correction, and computed spontaneous fission half-lives for other isotopes of element 104. These results are also shown in Table X.

TABLE X. Properties of elements 102 and 104.

Z	A	Q_α[a] (MeV)	M[b] (MeV)	δM[c] (MeV)	$\log_{10}\tau_{1/2(sf)}$[d] (years)
102	252	8.54	82.77	−2.47	− 4.0
	253	≥8.14	84.28	−2.68	1.6
	254	8.25	84.71	−2.70	− 2.4
	255	≥8.21	86.25	−3.12	4.3
	256	8.55	87.88	−2.20	− 4.4
	257	≥8.41	89.94	−2.33	1.0
	258		91.46	−1.50	− 7.2
	259		93.99	−1.40	− 3.1
	260		96.08	−0.50	−11.6
	261		98.84	−0.40	− 7.5
104	256		93.47	−3.66	− 1.8
	257		95.14	−3.56	2.3
	258		96.11	−2.92	− 5.0
	259		98.05	−2.79	− 1.0
	260		99.19	−2.22	− 8.0
	261		101.34	−2.12	− 3.9
	262		103.05	−1.22	−12.5
	263		105.43	−1.12	− 8.5

[a] Data from Refs. 46 and 47.
[b] First six entries were calculated from mass excesses in Ref. 20 and Q_α; remainder obtained from δM and liquid-drop masses from Ref. 10.
[c] First six entries from M minus liquid-drop mass from Ref. 10; remainder extrapolated except for $^{260}104$ which was deduced from τ_{sf}.
[d] Values from Eq. (3a) and δM, except for $^{260}104$ which is from Ref. 48.

If we are concerned with the production and detection of heavier nuclei by rapid neutron capture, the most important question would seem to be: What are the spontaneous fission half-lives of nuclides with 161, 163, etc. neutrons? We have not attempted to answer this question. This is because we have no reliable method for extrapolating shell corrections out to these neutron numbers. Various predictions have, however, been made of nuclear masses for these neutron numbers, and it is of some interest to see what one would deduce from these.

First of all the calculated shell corrections of Myers and Swiatecki[10] are, in the region of $N\simeq160$, uniformly increasing with N, by about 0.1 MeV per neutron added. For ^{216}Fm, their calculated δM is -1.92 MeV, which *per se* would give $\tau_{sf}\sim10^6$ years. Thus, one may say that these shell corrections give long spontaneous fission lifetimes, but if one extrapolates from the δM which we deduced for ^{259}Fm (by assuming $\tau_{sf}=5$ h), using a slope of 0.1 MeV/neutron then τ_{sf} will decrease by about a factor two per added neutron pair.

More impressive agreement with experiment is obtained using the semiempirical extrapolations of nuclear masses by Viola and Seaborg.[49] In particular, we note that their predicted nuclear masses for elements 102 and 104 are in rather good agreement with our values given in Table X. This would suggest that their extrapolation to high proton numbers is going very well out to $Z=104$. Their extrapolation to high neutron numbers gives, in general, values of δM which are substantially larger than ours and values which increase strongly (by ~0.5 MeV per neutron) with N. For ^{261}Fm and ^{263}Fm, we would obtain $\delta M=+0.7$ MeV and $+2.5$ MeV, respectively, which would lead to exceedingly short spontaneous fission lifetimes (~1 sec and 10^{-8} sec). It may be noted, however, that Viola and Wilkins[13] have used these masses to find much longer half-lives.

Cameron has employed a shell correction in just the same manner as we have used for our extrapolation, i.e., a function of N plus a function of Z. In a 1965 report[36] he used a shell correction which was assumed to be independent of N for $156\leq N\leq175$ and which decreased with Z by about 0.6 MeV per added proton for $Z\sim100$. Extrapolating from the heavy fermium isotopes with these slopes, we would find fission half-lives which were roughly independent of N and Z.

We thus see that by using published extrapolations of nuclear masses or shell corrections we are able to obtain a wide range of possible half-lives for nuclei with $N\gtrsim160$. It seems to us that the extrapolations must be made from a detailed physical model before one can have any confidence in them. Moreover, there remains uncertainty as to why and whether fission half-lives can really be predicted from a knowledge of shell corrections alone.

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