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Spontaneous fission was first observed by Flerov and Petrzhak[1)] in 1940 following a suggestion by Bohr and Wheeler.[2)] By dint of great effort these pioneering experimenters were able to show that U^{238} fissioned spontaneously with a half-life of the order of 10^{16} years, about a factor of 10^6 times faster than predicted. In the next ten years the half-lives of other nuclides for this process were determined by various groups but not enough information was accumulated to indicate any specific type of correlation as being correct.

Measurements of spontaneous fission half-lives in the transuranium region have in the last few years increased in number so markedly that several sorts of systematization have become possible. Seaborg[3)] and Whitehouse and Galbraith[4)] made the initial steps in this direction when it was pointed out that in the case of even-even nuclides the half-life for spontaneous fission seems to decrease with an exponential dependence on the value of Z^2/A while nuclides with an odd number of nucleons decay by this process at a much slower rate. With the limited amount of data available at this time a plot of the logarithm of the partial spontaneous fission half-life against Z^2/A resulted in a fairly straight line. It was also noted[3)] that this line when extrapolated to the region of instantaneous rate (that is, half-life of the order of 10^{-20} second) gives a value of about 47 for Z^2/A, which corresponds with the predicted limiting value of Z^2/A. In a later paper Ghiorso et al.[5)] pointed out that certain even-even nuclides (U^{234}, U^{232}, and possibly Th^{230}) exhibited substantial deviations in the direction of rates slower than predicted by the Z^2/A line.

Following these communications Kramish[6)] published a correlation of the ratio of spontaneous fission to alpha half-lives versus Z^2/A. This plot was based on the thesis that spontaneous fission and alpha decay could be regarded as closely related but competitive decay processes for heavy nuclei. The nature of this competition was demonstrated by connecting consecutive alpha decay products familywise and it was thus shown that the spontaneous fission mode of decay becomes more prominent as Z^2/A increases.

As more data were accumulated on the spontaneous fission rates of other heavy nuclides it became evident that although the parameter Z^2/A accounted in this manner broadly for the variation in half-life over the range of Z values, for a given value of Z, this parameter did not account for the variation of half-life with A. In 1954 Huizenga[7] pointed out that for a given value of Z the half-life goes through a maximum as A varies and on the basis of the data for isotopes of uranium it was postulated that the maximum in the spontaneous fission half-lives of even-even nuclides was analogous to the corresponding variation in beta stability. It was further suggested that the shorter spontaneous fission half-lives beyond the maximum are possibly a result of the greater deformation of the larger A nuclides; an ellipsoidal deformation presumably might increase the decay rate due to the improvement in penetration through the thinner barrier.[8] Later in the same year Studier and Huizenga[9] revived the Kramish plot of the ratio of the half-lives for spontaneous fission and alpha decay versus Z^2/A except that instead of connecting consecutive alpha decay products they were able to show a more consistent relationship by correlating nuclides differing by two Z units and six A units.

It is the purpose of this paper to call attention to a new parameter which possibly has a very pronounced influence on the variation of spontaneous fission half-lives with change in A. The effect observed is that the spontaneous fission half-lives for those even-neutron isotopes of elements 98 and 100 which have more than 152 neutrons are found to progressively decrease at a rate much faster than observed before. It has been shown that there is very good evidence from alpha decay data for a subshell at 152 neutrons.[10] At the time this paper was written the evidence for such a subshell was based entirely on the variation of alpha energies for the even-neutron isotopes of californium. Recently the nuclides Fm^{250} [11] (fermium, element 100) and Fm^{252} [11] have been produced in this laboratory and their alpha energies when compared with that for Fm^{254} show a similar variation. The basis for a subshell at 152 neutrons is thus seen to be rather firm. This possible correlation of the abrupt change in spontaneous fission half-lives with this subshell shows promise as an empirical method of predicting the spontaneous fission properties of unknown isotopes.

The principal new data that leads one to postulate this type of correlation consists of the recent measurement of the spontaneous fission half-lives of Cf^{254} and Fm^{256}. The isotope Cf^{254} has been isolated[12] by means of the electron-capture branching of the 1.5-day beta emitter, E^{254m} (einsteinium, element 99). A californium isotope was detected which decayed by spontaneous fission with a half-life of 85 $\pm$ 15 days; no alpha particle branching was observed but this is not surprising since one would expect an alpha half-life for this nuclide of 10^2 years. The isotope Fm^{256} has been manufactured in two ways: (1) as the electron-capture daughter of Mv^{256} [13] (mendelevium, element 101) and (2) from neutron bombardments[14] of E^{255} via a short-lived E^{256} beta emitter. In both types of experiments a 3 to 4 hour spontaneous fission emitter was isolated as an isotope of element 100. Again no alpha branching was observed since the predicted alpha half-life of Fm^{256} is about 10 days.

Other very recent measurements tend to support the postulation of a subshell influence on the spontaneous fission rates. The half-life for spontaneous fission of Pu^{244} has been found to be $2.5 \pm 0.7 \times 10^{10}$ years,[15] a value which is only three times shorter than that for Pu^{242}. Thus over the entire region of beta stability the half-lives for spontaneous fission of the plutonium isotope vary by a factor of a few hundred; the variation for californium isotopes on the other hand is about 10^5 without including the value for the unknown heaviest beta stable member, Cf^{256}. Cm^{246} has been shown[16)] to have a spontaneous fission half-life of 3×10^7 years, a value consistent with the above observations.

Clearly we have not yet proved our hypothesis of the 152 neutron correlation. Much more proof will be at hand, however, if the spontaneous fission rates for Fm^{252} and Fm^{250} can be measured and shown to have the predicted values. It does seem obvious that some more powerful effect is influencing the spontaneous fission half-lives than has been proposed so far. Figure 1 is a plot of the spontaneous fission half-lives versus neutron number and contains in a simple form all the data known at this time. The predicted lines for the unknown heavier even Z elements are, of course, drawn with the 152 neutron prejudice clearly delineated. Included on this plot will be found the spontaneous fission half-lives for six odd nucleon nuclides. The value for U^{235} is very questionable but the others, Pu^{239}, Bk^{249}, Cf^{249}, E^{253}, E^{254}, and Fm^{255} are felt to be reasonably accurate. The "hindrance factors" for these nuclides when compared to their maximum rates as determined by the even-even lines and their hypothetical odd Z intermediates vary between 10^3 and 10^6. The most highly hindered isotope is the recently discovered long-lived isomer of E^{254} [12)] which has both an odd number of protons and an odd number of neutrons. We have not yet observed any clear systematic variation in the degree of hindrance which can be correlated with 152 neutrons, but this might well be due to insufficient data.

On Fig. 1 we have also indicated the manner in which the alpha half-lives vary in this region. Examination of the trends indicates that spontaneous fission will probably not become competitive with alpha decay even at a few atomic number units above 100 except for those isotopes beyond the subshell at 152 neutrons. This is the case because the alpha decay rates increase as fast as the spontaneous fission rates at the higher Z values. However, beyond 152 neutrons spontaneous fission seems to increase so markedly that at 156 neutrons it becomes the chief mode of decay. This has been observed to date in californium and fermium.

For the sake of completeness we have included the two other well-known plots. Figure 2, spontaneous fission half-life versus Z^2/A, demonstrates very well that one line cannot encompass all the data adequately. On the other hand, if one connects nuclides differing by two Z units and six A units there is apparent a closer correlation with the notable exceptions of Th^{232}, Cf^{254}, and Fm^{256}. Figure 3 is the Kramish-Studier, Huizenga-type of plot. Here, again, one sees notable exceptions (Th^{232}, Cf^{248}, Cf^{254}, and Fm^{256}) whose spontaneous fission half-lives do not correspond with the "jump of six" lines. Perhaps the limited success of this type of correlation is merely reflecting the fact that one is dividing the slope of one straight line by that of another (see Fig. 1, dotted lines showing approximate alpha half-life variation) and that where it fails it does so because the spontaneous fission half-life probably changes for a reason that has little to do with a change in alpha half-life.

Table I includes the data and references on spontaneous fission half-lives. Those isotopes which have so far been measured only as a rather low limit are not included.

Table I

Spontaneous Fission Half-Lives

Isotope	$T_{1/2}$(SF)(Yrs)	Ref.	Isotope	$T_{1/2}$(SF)(Yrs)	Ref.
Th^{230}	$\geq 1.5 \times 10^{7}$	18	Cm^{244}	1.4×10^{7}	5
Th^{232}	1.4×10^{18}	18	Cm^{246}	3×10^{7}	16
U^{232}	$8 \pm {}^{5.5}_{3} \times 10^{13}$	32	Bk^{249}	6×10^{8}	31
U^{234}	1.6×10^{17}	5	Cf^{246}	2.1×10^{3}	24
U^{235}	1.8×10^{17}(?)	18	Cf^{248}	7×10^{3}	25
U^{236}	2×10^{16}	19	Cf^{249}	1.5×10^{9}	31
U^{238}	8.0×10^{15}	18	Cf^{250}	1.5×10^{4}	28, 29, 30
Pu^{236}	3.5×10^{9}	5	Cf^{252}	66	28, 29
Pu^{238}	4.9×10^{10}	20	Cf^{254}	0.2	16, 12
Pu^{239}	5.5×10^{15}	18	E^{253}	3×10^{5}	26, 31
Pu^{240}	1.2×10^{11}	21	E^{254}	1.5×10^{5}	31
Pu^{242}	$7.25 \pm 0.3 \times 10^{10}$	16, 17	Fm^{254}	0.5	26, 27
Pu^{244}	$2.5 \pm 0.7 \times 10^{10}$	15	Fm^{255}	20	31
Cm^{240}	1.9×10^{6}	5	Fm^{256}	3×10^{-4}	13, 14
Cm^{242}	7.2×10^{6}	22, 23			

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FIGURE CAPTIONS

Fig. 1. Spontaneous fission half-life versus neutron number. Dotted lines indicate the experimentally observed alpha half-life variation except in the cases for elements 102 and 104.

Fig. 2. Spontaneous fission half-life versus Z^2/A.

Fig. 3. The ratio of spontaneous fission half-life to alpha half-life versus Z^2/A.