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ACTIVATION ENERGY FOR FISSION

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August 29, 1952

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ABSTRACT

The experimentally determined exponential dependence of spontaneous fission rate on Z^2/A has been used to derive an expression for the dependence of the fission activation energy on Z^2/A. This expression has been used to calculate the activation energy for slow neutron induced fission and photofission. The correlation with the experimental data on these types of fission seems to be quite good.

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The rate of spontaneous fission for even-even nuclides has a simple exponential dependence[1-3] on Z^2/A and a plot of the logarithm of the "half-life" for this process vs. Z^2/A (Fig. 1 of references 1 and 3) yields the relationship

$$T = 10^{-21} \times 10^{178 - 3.75\, Z^2/A} \text{ sec} . \qquad (1)$$

It is the purpose of this note to point out how this information can be related to the activation energy for fission and hence be correlated with the known information on slow neutron and photofission of heavy nuclides.

The barrier penetration probability for spontaneous fission has been shown to have the general form $10^{-k\Delta E}$ where ΔE is the energy deficit at the saddle point[4,5] and in particular Frankel and Metropolis[5] have derived for the liquid drop model the relationship

$$T = 10^{-21} \times 10^{7.85\Delta E} \text{ sec} \qquad (2)$$

where ΔE is in Mev. On the assumption that their treatment for the rate process is essentially correct so that the general form of (2) is valid, even though the calculation of ΔE is not, as evidenced from the failure to account for experimental spontaneous fission rates,[1] we can relate (1) and (2) and obtain

$$T = 10^{-21} \times 10^{7.85\,(22.7 - 0.477\, Z^2/A)} \text{ sec} \qquad (3)$$

$$\text{where } \Delta E = (22.7 - 0.477\, Z^2/A) \text{ Mev.} \qquad (4)$$

However, (2) actually applies only to U^{238} ($Z^2/A = 35.56$) of different degrees of excitation, and extension to different values of Z^2/A leads to a somewhat more complicated expression. When this is related to (1) we find that ΔE can be approximately represented over a limited range of Z^2/A by

$$\Delta E = (19.0 - 0.36\, Z^2/A) \text{ Mev.} \qquad (5)$$

When ΔE is calculated using (5) and compared with the binding energy (NBE) of the added neutron[6] for each of the nuclides whose slow neutron fission cross sections[7] or upper limits are known, remarkable agreement is observed as shown in Table I. Something approaching a quantitative correlation is attained if the individual values of NBE-ΔE are compared with the corresponding values of σ_f/σ_r (σ_f = fission, σ_r = n,γ cross section for slow neutrons) for each nuclide; since the probability for gamma emission might be approximately the same for all these nuclides, the ratio σ_f/σ_r may give a good measure of the relative probability for fission[8] and hence can be used to better advantage for comparative purposes than σ_f alone. Such a plot is shown in Fig. 1 where the available points, perhaps fortuitously, are rather well represented by a straight line with some exceptions discussed below.

Perhaps all of the presently available data on three types of fission (spontaneous, slow neutron, photo) are consistent with the view that an odd nucleon has the effect of slowing the fission process

Table I

Correlation of Slow Neutron Fissionability with Potential Barrier to Fission and Corresponding Neutron Binding Energy

Nuclide	ΔE (Mev)	NBE[a] (Mev)	NBE[a]-ΔE (Mev)	Slow Neutron Fissionability[b]
Ra^{226}	6.7	4.9	-1.8	-
Ra^{228}	6.8	4.6	-2.2	-
Ac^{227}	6.5	5.1	-1.4	-
Th^{227}	6.2	7.0	0.8	+
Th^{228}	6.3	5.4	-0.9	-
Th^{229}	6.3	6.7	0.4	+
Th^{230}	6.4	4.6	-1.8	-
Th^{232}	6.5	4.8	-1.7	-
Th^{234}	6.6	4.7	-1.9	-
Pa^{230}	6.1	6.9	0.8	+
Pa^{231}	6.1	5.4	-0.7	-
Pa^{232}	6.2	6.7	0.5	+
Pa^{233}	6.3	5.1	-1.2	-
U^{232}	5.9	5.9	0.0	+
U^{233}	6.0	6.7	0.7	+
U^{234}	6.0	5.4	-0.6	-
U^{235}	6.1	6.5	0.4	+
U^{238}	6.2	4.8	-1.4	-
Np^{234}	5.7	7.1	1.4	+
Np^{237}	5.9	5.2	-0.7	-
Np^{238}	6.0	6.4	0.4	+
Np^{239}	6.0	5.0	-1.0	-

continued

Nuclide	ΔE (Mev)	NBE[a] (Mev)	NBE[a]-ΔE (Mev)	Slow Neutron Fissionability[b]
Pu^{238}	5.7	5.7	0.0	+
Pu^{239}	5.8	6.5	0.7	+
Pu^{241}	5.9	6.2	0.3	+
Am^{241}	5.5	5.3	-0.2	-
Am^{242m}	5.6	6.5	0.9	+
Am^{242}	5.6	6.5	0.9	+
Am^{243}	5.7	5.2	-0.5	?
Cm^{242}	5.4	5.9	0.5	?

[a] neutron binding energy for nuclide with mass number A + 1 from ref. 6

[b] + denotes σ_f greater than about 1 barn,

- denotes σ_f less than about 1 barn (possibly due to larger radii than corresponding even-even nuclides giving smaller values of Z^2/r^3 which Z^2/A is meant to represent). In the spontaneous fission case nuclides with odd nucleons have rates up to some 10^3 times slower than corresponding even-even nuclides (Fig. 1 in reference 1). In the case of photofission[9] odd nucleon nuclides like U^{233}, U^{235}, and Pu^{239} have thresholds 0.4 - 0.5 Mev nearer the top of the barrier than U^{238} and Th^{232} corresponding to rates some $10^2 - 10^3$ times slower than those of U^{238} or Th^{232}, indicating that the slowing effect of an odd nucleon is operative even at excitation to near the top of the barrier. However, the probability for gamma re-emission may be less for the even-even nuclides, due to a larger level spacing, which means that fission is relatively favored and would occur at lower excitation relative to the barrier height; thus the odd nucleon may slow the photofission process in a manner analogous to the effect in spontaneous fission or due to its effect on level spacing and therefore on the probability for competitive gamma emission. If we apply these considerations to slow neutron fission, we must think in terms of the intermediate fissioning nucleus which is formed upon capture of the neutron. Thus it would be interesting to see if even-odd nuclides (even protons, odd neutrons), where the intermediate fissioning nuclei are of the even-even type, undergo slow neutron induced fission with greater probability than other nuclear types at comparable excitation energy. Unfortunately there are no presently known cases for $NBE - \Delta E < 0$ but only for nuclides which are apparently excited above the barrier in the slow neutron fission process; those that form intermediates with an odd nucleon (see especially Cm^{242}) seem to be slower in undergoing this process; again either of the two mechanisms for the slowing effect of an odd nucleon may be operative. The variation in the positions of the points for the even-odd nuclides in the region above the barrier (NBE-ΔE > 0) may perhaps be explained by a small non-uniform variation in the probability for gamma emission.

The empirical relationship (1) depends, of course, upon how the line is drawn through the points representing the measured spontaneous fission rates of the even-even nuclides. In order to examine this point further, other possibilities were examined; for example, a line drawn somewhat higher with a steeper slope gives more weight to the point[3] for U^{234} and passes somewhat above such points as those for Cm^{244}, U^{238}, and Th^{232} as it might do if these latter nuclides have slightly shrunken radii due to proximi.y to closed subshells. Such considerations lead to somewhat different constants in relation (5) but do not change perceptibly the results in the correlations presented in Table I and Fig. 1.

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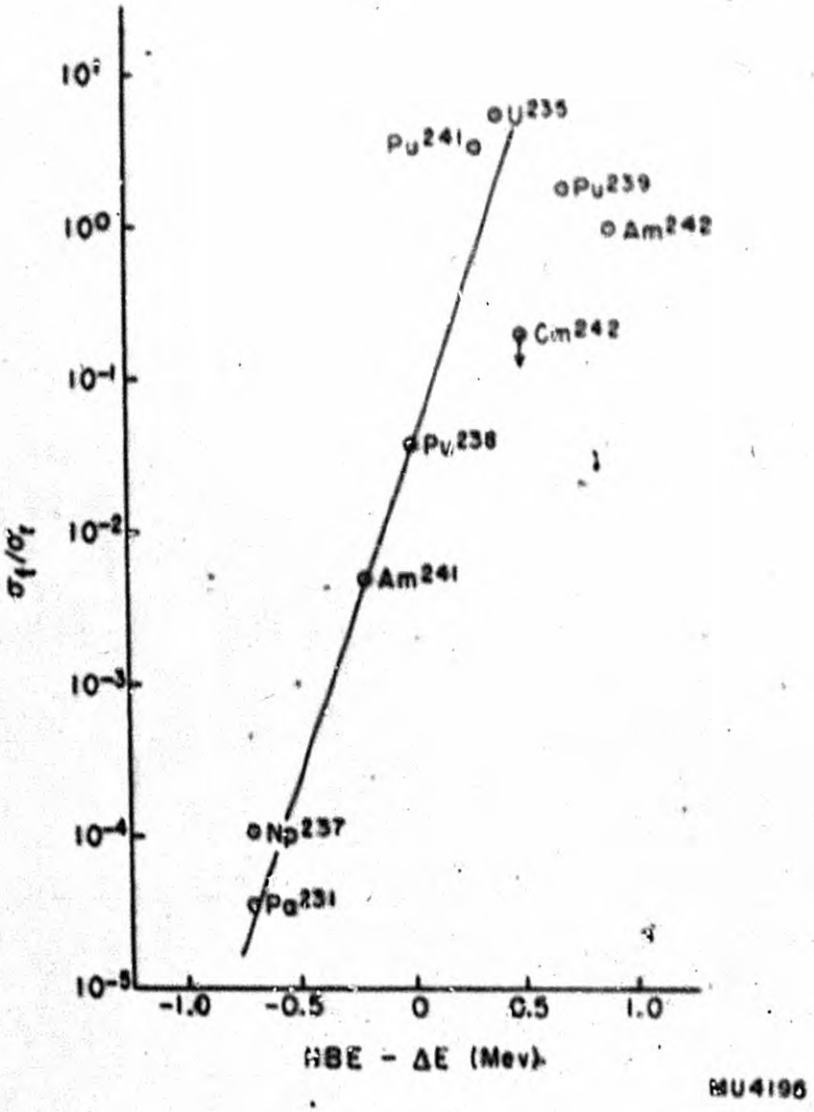

Fig. 1. Plot of comparative slow neutron fissionability. σ_f/σ_r denotes ratio slow neutron fission to n,γ cross section ($\overset{\circ}{\downarrow}$ signifies upper limit), NBE-ΔE denotes difference between neutron binding energy and potential barrier.