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Publication Date

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October 26, 1955

Printed for the U. S. Atomic Energy Commission

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INTRODUCTION

Theoretical expressions relating alpha decay rate and energy were first derived by Gamow[1] and Gurney and Condon.[2] The relationships hold best for even-even alpha emitters. Perlman, Seaborg, and Ghiorso[3] in their alpha systematics have pointed out large and erratic hindrance in odd types. Nordstrom[4] has plotted distributions of hindrance factors in an attempt to compare "forbiddenness" in alpha decay in a manner similar to log ft values in beta decay. We have made hindrance factor plots for all observed alpha groups emitted from nuclei with greater than 128 neutrons in an attempt to establish correlations.

BASIS OF CALCULATIONS

Even-Even Nuclei - Ground State Transitions

Decay rates to ground states of even-even nuclei are correlated remarkably well by simple barrier penetration formulas with smoothly varying radius formulas. However, there are significant trends[5] as shown in a plot of "effective nuclear radius" for even-even alpha emitters. Thus, for our semi-empirical correlations of alpha decay rates we have chosen to plot decay rate trends of even-even nuclei on a type of plot which from barrier penetration theory should lead to nearly straight line plots for isotopes of a given element. The type of plot chosen sets the logarithm of the half-life against the reciprocal of the square root of the total alpha decay energy. (We have included recoil energy and the electron screening correction.)

From this relationship between log $t_{1/2}$ and $E^{-1/2}$ the "hindrance" of an alpha particle is seen as the deviation from the curves. The alpha decay energy is then taken as the independent variable, and the hindrance factor is defined as the ratio of the experimental partial alpha half-life to that predicted from the curves.

The primary problem in this correlation, therefore, was to determine the lines which would serve as the basis for the calculations. This was done by the method of weighted least squares analyses on the ground state to ground state decay of even-even nuclei. The solid lines in Fig. 1 are the result of these analyses.

The weighting consisted of giving spectroscopically determined alpha energies double weight as compared to ion chamber energy measurements. The solid triangles in Fig. 1 indicate the isotopes used in fitting the line. Solid circles indicate isotopes for which the half-lives had been estimated from previous alpha systematics[3] (i.e. Em^{216}, Ra^{220}, Th^{224}, etc.) or which appeared to deviate markedly from systematic behavior (Po^{212}, U^{228}, U^{238}) and these were not used in the least squares analyses. For the elements up to curium there was no problem in fitting the lines. For curium and higher elements the scarcity of data made the fitting somewhat arbitrary. Cm^{242} and Cm^{244} were used exclusively to set the curium line because of their well determined decay schemes, as compared with the lighter curium isotopes about which insufficient experimental data is available.

Cf^{250} and Cf^{252} were rejected in the determination of the californium line slope† because of their proximity to the closed subshell at 152 neutrons.[6] The slope for fermium was obtained by extrapolation as indicated in Fig. 2.

Alpha particle energies were obtained from the compilation of Perlman and Asaro[5] with the exception of the isotopes listed below. All partial alpha half-lives were obtained using half-lives, alpha branching ratios, and alpha particle abundances listed in the Table of Isotopes[7] except for the following: Em^{204}, Em^{206}, Reference 8; Ra^{222}, α_1, Reference 9; U^{230}, α_2, Reference 10; U^{232}, α_3, Reference 11; Np^{237}, all states, Reference 12; Pu^{238}, α_3, α_4, Reference 13; Am^{243}, α_3, α_4, Reference 14; Cm^{242}, α_3, α_4, α_5, Reference 15; Cm^{243}, α_1, Reference 16; Cm^{245}, α_0, Reference 17; Cf^{246}, α_2, α_3, Reference 18; Cf^{249}, α_0, α_1, Reference 19; Cf^{250}, α_0, α_1, Reference 20; Cf^{252}, α_0, α_1, Reference 21; E^{253}, α_0, α_1, α_2, Reference 22; E^{254}, α_0, Reference 23; Fm^{254}, α_0, α_1, α_2, Reference 24.

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†Since this paper was prepared, new data have been obtained on the lighter californium isotopes which reassign the alpha decay energy and half-life of Cf^{244} to Cf^{245} and which give the actual half-life and alpha particle energy of Cf^{244}.(25) Since these new data have been found to alter the slope of the Cf line only slightly due to the anomalous behavior of Cf^{245}, we have not recalculated the hindrance factors for the isotopes of berkelium, californium, einsteinium, and fermium which would be affected if the change were large.

It is easy to justify the above-mentioned type of plot from WKB barrier penetration formulas (the form for S-wave emission). The most important factor in these formulas is the exponential expression

$$P = \exp\left[-\frac{2}{\hbar}\int_R^{\frac{2Ze^2}{E}}\left[2M\left(\frac{2Ze^2}{r} - E\right)\right]^{1/2}dr\right] \qquad (1)$$

which is expressible in analytical form as

$$\exp\left[-\frac{2}{\hbar}(2MB)^{1/2}R\right]\left[x^{-1/2}\arccos x^{1/2} - (1-x)^{1/2}\right] \qquad (2)$$

where $x = ER/2Ze^2$, a dimensionless parameter, and $B = 2Ze^2/R$, the barrier height. If we make a Taylor series expansion of this function about $x = 0$, we obtain

$$P \approx \exp\left\{-\frac{(2M)^{1/2}B^{1/2}R}{\hbar}\left[\frac{\pi B^{1/2}}{E^{1/2}} - 4 + \frac{E}{B} - \frac{3}{4}\frac{E^2}{B^2} + \ldots\right]\right\} \qquad (3)$$

If one retains only the dominant first two terms, as has Gamow[1], then one sees that the logarithm of the penetration function is a linear function of $E^{-1/2}$.

An interesting check on simple barrier penetration theory is provided by examination of the slopes of the least squares lines of Fig. 1. The form of the first term of Equation (3) shows that the slopes of the lines of Fig. 1 should be proportional to the atomic number, Z, of the daughter nucleus to accord with simple theory. In Fig. 2 these slopes are plotted against Z. The theoretically expected behavior is well shown by the lower elements in the series, but there is a striking break beginning at the plutonium alpha emitters and then a reversal of the expected trend.

It is beyond the scope of this paper to attempt any detailed theoretical explanation of the deviations from simple theory. It should be pointed out that abnormal slopes in Fig. 1 do not necessarily mean a changed dependence of decay rate on energy, but since energy is (above 128 neutrons) a monotonic function of the neutron number for an isotopic series, an abnormal slope may reflect a change in the alpha decay rate dependent on the addition of neutron pairs. Some isotopic series pass through the region of the onset of validity of the Bohr-Mottelson[26] spheroidally deformed model and some pass through rapidly changing spheroidal deformation. Such changes must certainly influence barrier penetration, yet, interestingly enough, the elements where these changes are greatest, i.e. radium, thorium, and uranium, have the best-behaved slopes. The slopes should also be influenced by variations in the alpha formation probability entirely apart from the external barrier penetration factor.

Even-Even Nuclei - Excited State Transitions

In contrast to the regularly behaving ground state transition rates, the transitions to excited states show striking trends in rate for different nuclei.

For the elements thorium and above, alpha decay is often observed to at least three levels (0+, 2+, and 4+), identifiable as a rotational band sequence. Alpha decay to states of odd parity has been observed.[5]

To exhibit this behavior we have calculated hindrance factors to all observed states. The distribution of hindrance factors is shown in the histograms of figures 3, 4, 5, and 6. The three lines of numbers on each histogram block represents (1) element/mass number/ground state, indicated by zero if this is the ground state, (2) assigned number of excited state and energy of this state in kilo-electron volts, and (3) the value of the departure factor for this transition. Because of the increasing interest in the correlations achieved by the Bohr-Mottelson large-deformation model, we have segregated the cases in the region of nuclear rotational bands from those cases nearer the Pb^{208} closed configuration. The bold line on each histogram divides these regions, the cases below the bold line coming from the rotational band region.

For the even-even nuclides the generally increasing nature of hindrance to the higher groups is evident, and the hindrance to the 4+, 6+, and 8+ groups generally far exceeds hindrance accountable for by the addition to the barrier of the centrifugal potential associated with angular momentum of the alpha decay system.

Asaro[27] has plotted hindrance factors* against atomic number for the even parity excited state transitions. In somewhat similar manner we have plotted hindrance factors against neutron number in Fig. 7.

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*The hindrance factors of Asaro for transitions to excited states of even-even nuclei are defined slightly differently from ours. Asaro has taken as unhindered the ground state to ground state even-even alpha transitions. We have, however, calculated hindrance factors in relation to the least square curves in Fig. 1. Consequently, we may have hindrance factors for ground state transitions, whereas Asaro will not. The 0+ hindrance factors shown in Fig. 7 illustrate this and can be used to obtain Asaro type hindrance factors from the present data.

We do not expect to illustrate any trends with the 0+ hindrances, but the high hindrances of the lightest isotopes of polonium, emanation, and plutonium and the slight depression at 142 neutrons are of interest. The variation observed for the curium and californium isotopes is explained by the manner, previously mentioned, in which the lines for these elements in Fig. 1 were determined.

The dependence upon neutron number of decay to the first excited state (2+) reflects the ground state transition probability and appears relatively unhindered, although there is some increase among the heaviest isotopes.

Of very great interest is the dependence of decay to the second excited state (4+) upon N and Z. It can easily be seen from Fig. 7(c) that the hindrance factors for the second state vary with Z and pass through a maximum at Z = 96. However, also notable is the apparent neutron dependence of isotopes, as manifested by the apparent maxima in the 4+ curves of thorium and uranium. The 1- states apparently follow the behavior of the 4+ states.

The relative intensities of decay groups to the even parity states have been considered by a number of authors in connection with spheroidal alpha decay theory[28] but will not be discussed here.

Odd-Nucleon Alpha Emitters

In contrast to the regular behavior of even-even alpha decay rates, nuclei with odd nucleon numbers often exhibit erratic alpha decay rates, tending generally to be slower than comparable even-even alpha emitters. Perlman, Ghiorso, and Seaborg[3] have clearly pointed out that the hindrance must be associated with a lowered alpha formation probability rather than with the higher external barrier resulting from non-zero angular momentum.

The details regarding this hindrance are not well understood. Rasmussen[29] and Bohr, Fröman, and Mottelson[30] have selected and discussed a few special cases exhibiting little or no hindrance.

To obtain the bases for calculation of these hindrance factors for odd Z nuclei the slopes shown in Fig. 1 were interpolated to yield the intermediate odd Z slopes. For instance, the average of the slopes for elements 84 and 86 was employed as the slope for Z = 85. Since it was impossible to interpolate slopes for Z = 83 and 99, these slopes were obtained by extrapolation, as indicated by the dashed line in Fig. 2. Similarly the intercepts at $E_\alpha = \infty$ were interpolated to yield the intercepts for these nuclei.

The distributions of these hindrance factors for the various nuclear types are shown in Fig. 8 (odd-even), Fig. 9 (even-odd), and Fig. 10 (odd-odd). The histograms for odd-even and even-odd types exhibit broad distributions with the suggestion of a breakup into two groups. The main group peaks around hindrance factors of order 3, and the second and small group, around 500.

For the odd-even types in the rotational band region, most of the cases in the highest hindered group probably involve a change in parity in alpha decay. This fact is inferred from the apparent "favored" nature of some of the transitions in the same nuclei, with the levels showing the high hindrance connected to the favored levels by E1 gamma transitions, which signify a parity difference. Thus we suggest the possibility that parity change is a major hindering factor in alpha decay of odd mass nuclei. Examination of other odd-even alpha transitions possibly involving a parity change indicates that the alpha transition may generally be hindered in such cases but that the hindrance associated with the parity change becomes less at atomic numbers below 95.

For the even-odd types in the rotational band region one again observed a separate group of hindered cases for log hindrance factor from 2.0 to 3.5. Here, however, we may infer that at least two cases (in the decay of Cm^{243}) probably involve no parity change. This inference may be drawn from the probable presence of a "favored" alpha transition in Cm^{243} and the connection between favored and hindered states by M1 gamma transitions.

CONCLUSION

The data presented here represent all that are available at present on alpha decay rates. We have attempted, by presenting them in the form of hindrance factor histograms, to illustrate the distribution of the deviations from simple theory. We hope that the presentation of the data in this form can serve as a useful reference for further studies.

ACKNOWLEDGMENTS

We are indebted to Dr. Frank Asaro and Dr. Frank Stephens and to John Hummel, Albert Ghiorso, and others for valuable data in advance of publication. This study was carried out under the auspices of the U.S. Atomic Energy Commission.

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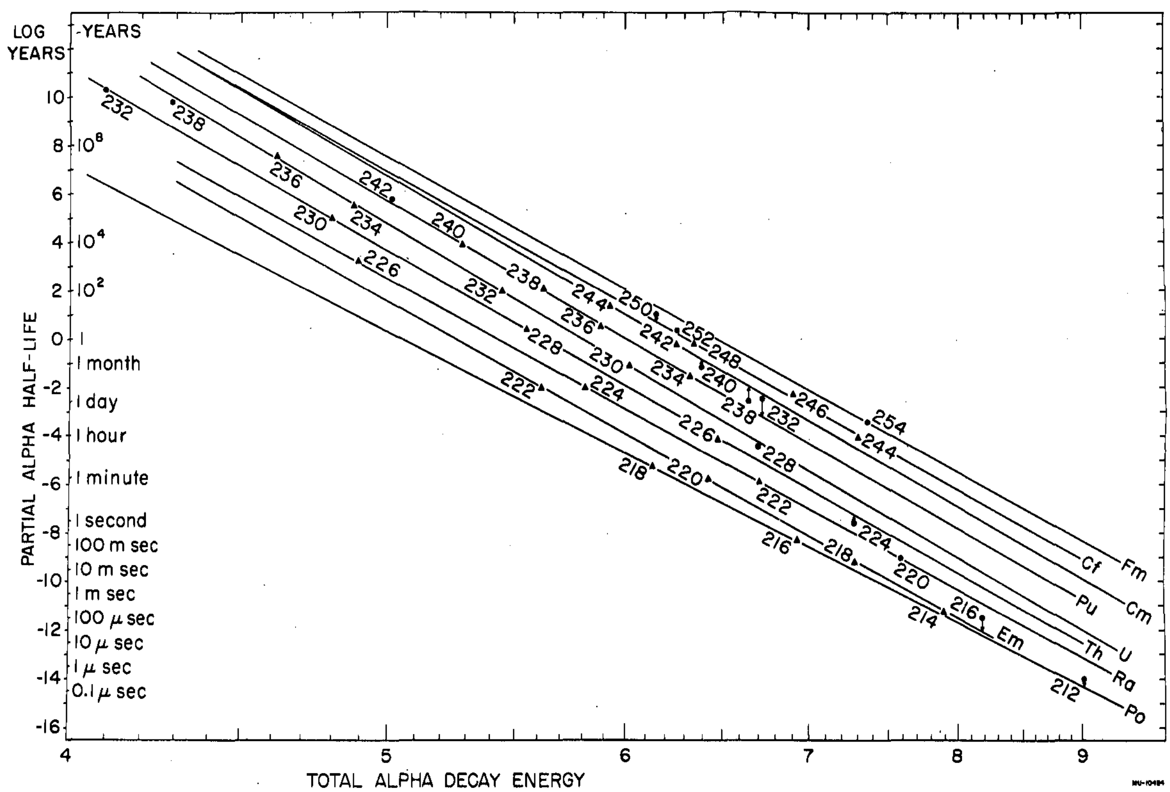

Fig. 1 Graph of log partial alpha half-life vs. the reciprocal of the total alpha decay energy (alpha particle energy plus recoil and screening corrections). Solid lines represent least square fits to data represented by solid triangles, for Z between 84 and 98. These least square fits were used to calculate hindrance factors. Solid circles represent data that were not used in the least squares analyses.

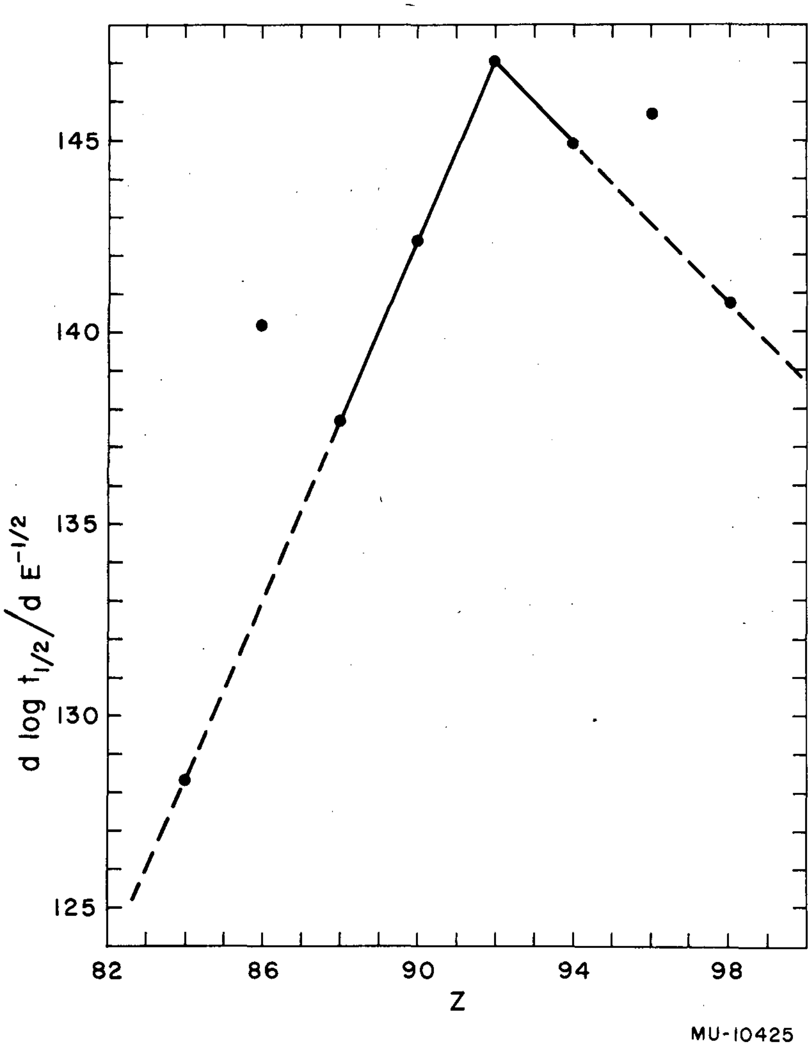

Fig. 2 Graph of $\frac{d \log t_{1/2}}{dE^{-1/2}}$ as a function of Z.

Dotted lines indicate method used to obtain slopes for elements 83, 99, and 100. Irregularities after uranium probably indicate inadequacy of simple theory.

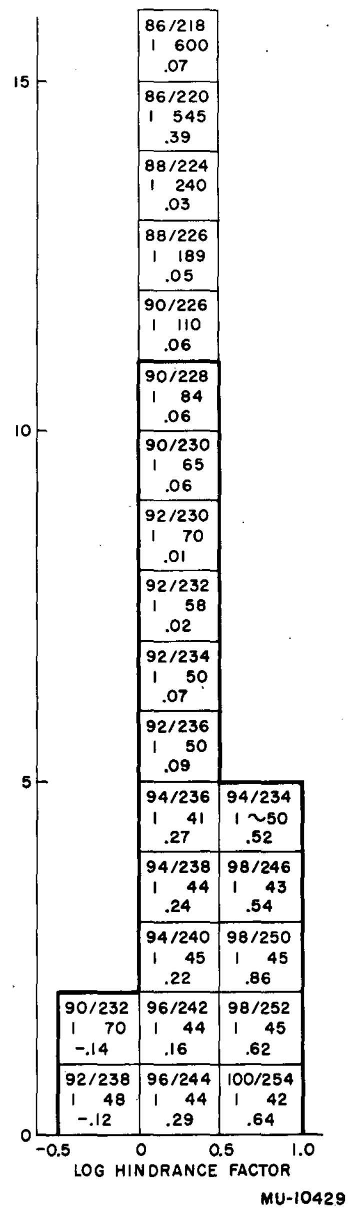

Fig. 3 Distribution of hindrance factors for alpha decay to 2+ states of even-even nuclei. Heavy line in all histograms separates rotational region from region near closed shells, where rotational model is not applicable. First line of numbers indicates element and mass assignment; second, present order of state and energy of state; third, value of log of hindrance factor.

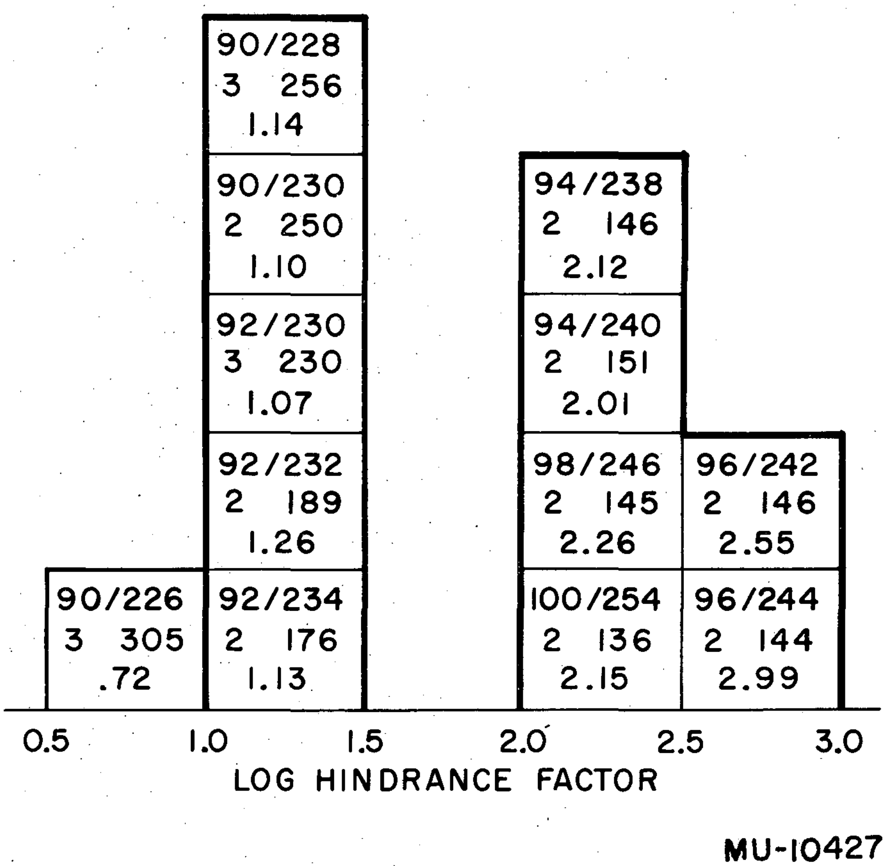

Fig. 4 Distribution of hindrance factors for alpha decay to 4+ states in even-even nuclei.

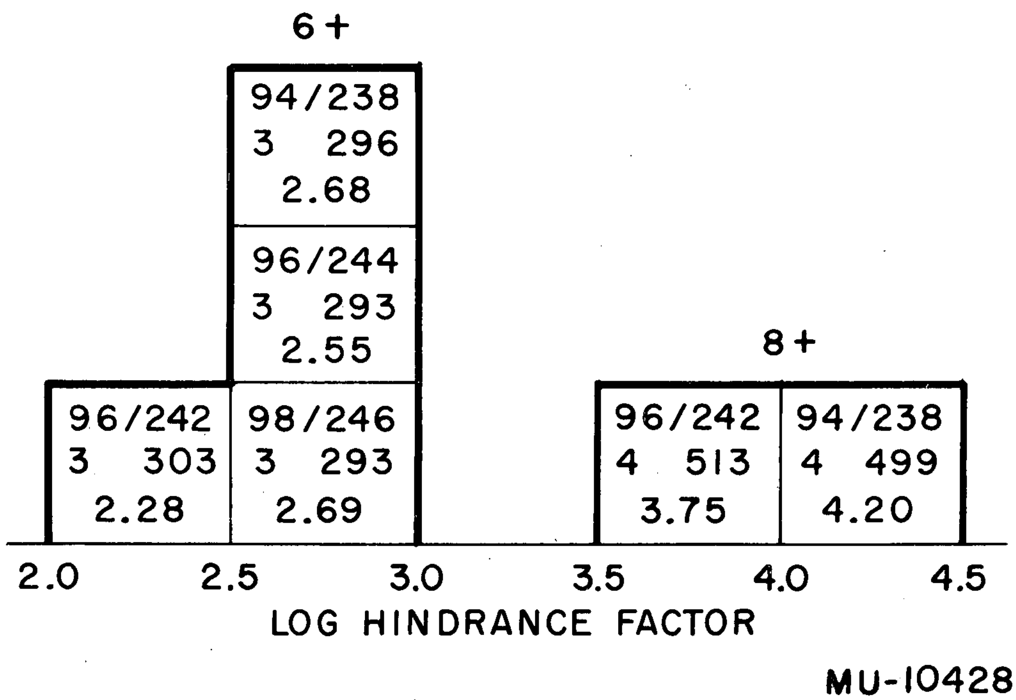

Fig. 5 Distribution of hindrance factors for alpha decay to 6+ and 8+ states in even-even nuclei.

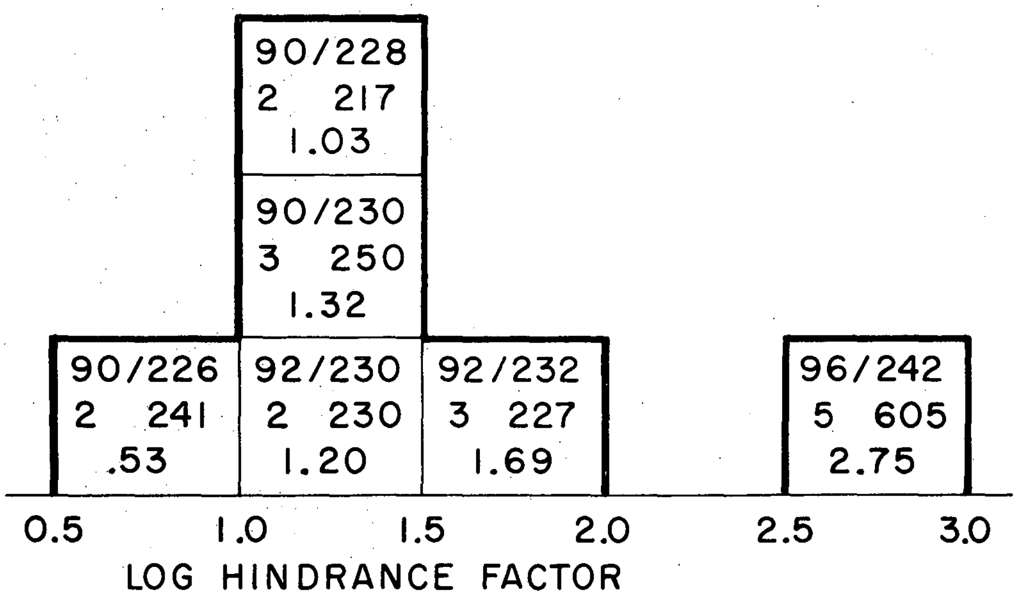

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Fig. 6 Distribution of hindrance factors for alpha decay to 1- states in even-even nuclei.

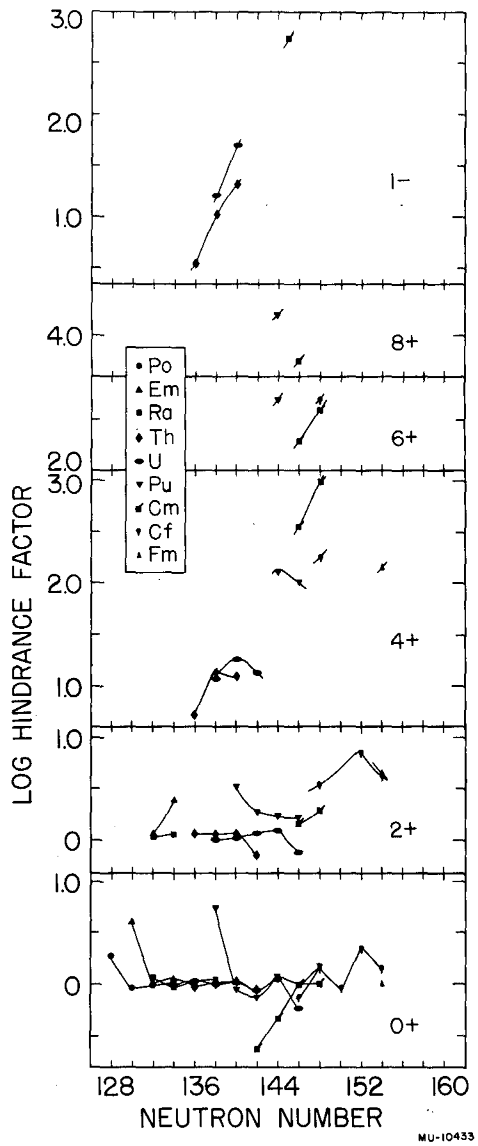

Fig. 7 Hindrance as a function of neutron number for decay to excited states in even-even nuclei. Lines connect isotopes. 0+ is included as a basis for comparison with hindrance factors of Asaro.

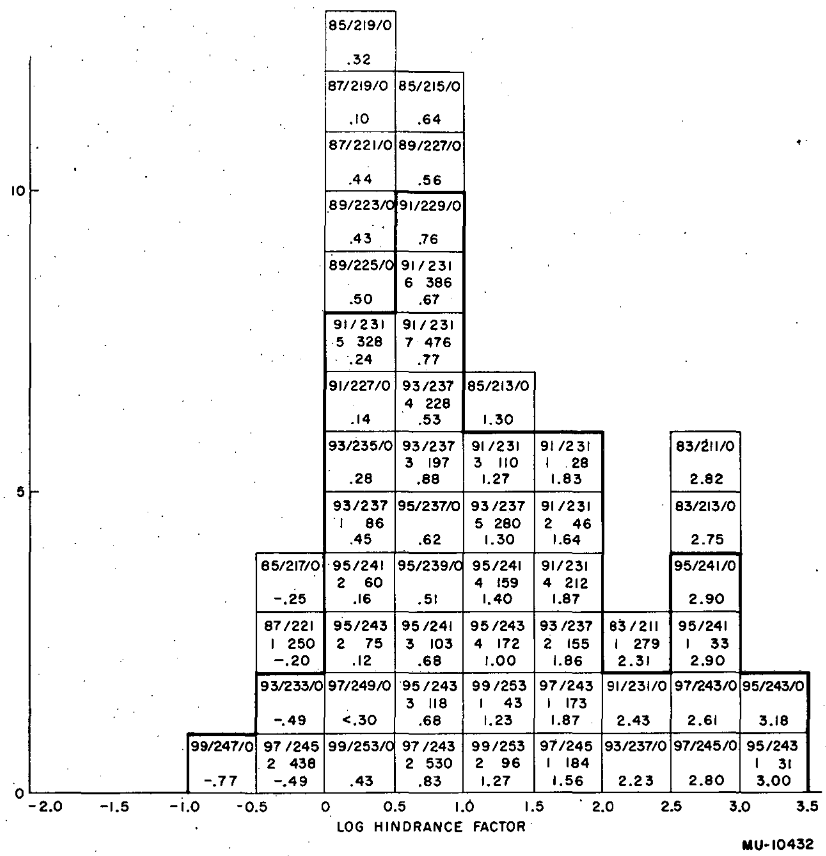

Fig. 8 Distribution of hindrance factors for alpha decay for all observed cases to states in even-odd nuclei.

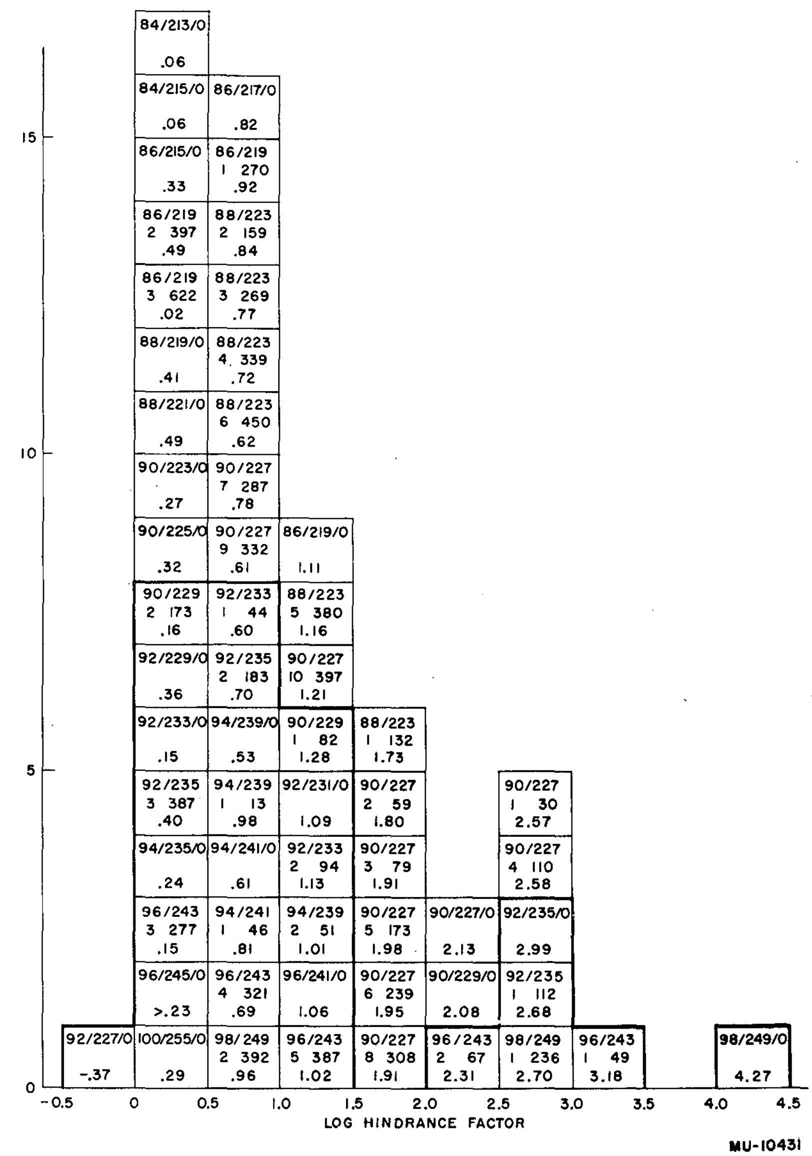

Fig. 9 Distribution of hindrance factors for alpha decay for all observed cases to states in odd-even nuclei.

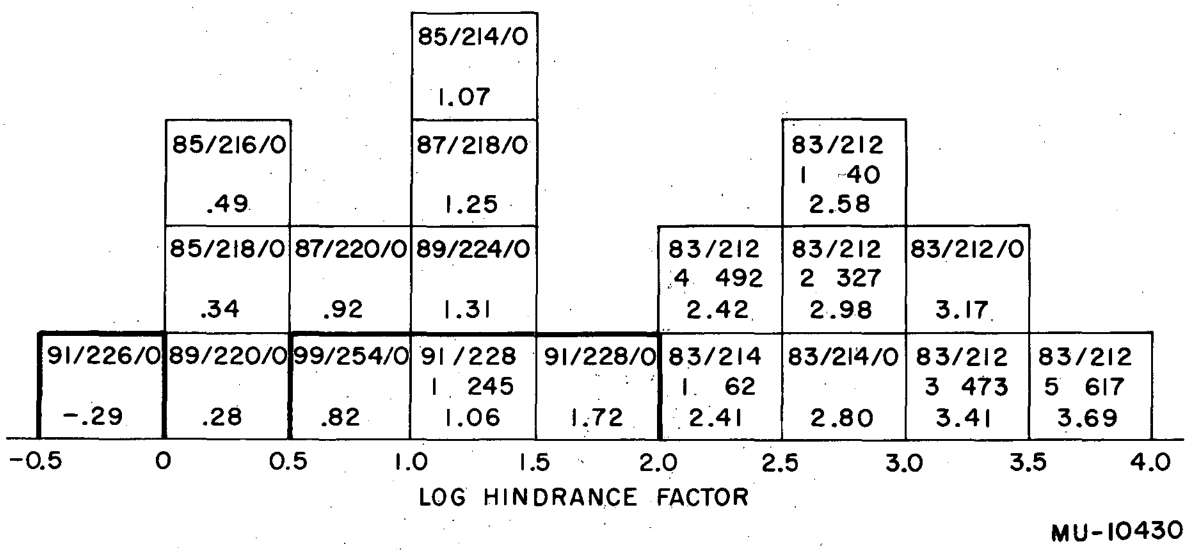

Fig. 10 Distribution of hindrance factors for alpha decay for all observed cases to states in odd-odd nuclei.