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Systematics of alpha decay half-lives

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A simple semiempirical relation for the prediction of the partial alpha half-life of the favored transition in even-even nuclides has been successfully derived. Correction factors for the closed shells, $N=126$ or $Z=82$, were also obtained. Hindrance factors for odd-A and odd-odd nuclides were obtained from the relation for even-even nuclides and discussed. Alpha partial half-lives of nuclides of transactinides and superheavy elements have been predicted for the ground-to-ground state favored transitions.

I. INTRODUCTION

Alpha decay is historically the first observed radioactivity, and it conveys some important nuclear information such as nuclear mass and structure. In the heavy element region, in particular, many nuclides have been discovered and characterized by observing alpha decay as alpha particles can be detected with high sensitivity and resolution. Accordingly, a better prediction of the alpha decay half-life is always in demand for the discovery of new elements and isotopes.

The first recognition of a systematic trend in the alpha decay constant was reported by Geiger and Nuttal.[1] They found a linear relationship between the logarithm of the decay constants and the logarithm of the ranges of alpha particles emitted from nuclides belonging to a given natural radioactive decay series. Gamov[2] and Gurney and Condon,[3] independently, have elucidated the relationship between the alpha decay energy and its half-life by a quantum mechanical effect of the preformed alpha-particle tunneling through a barrier created by the alpha particle and the residual nucleus. Recently, Poenaru and Ivascu have reviewed[4,5] and reevaluated some typical empirical formulas presented in the past by Froman,[6] Wapstra *et al.*,[7] Taagepera and Nurmia,[8] Keller and Munzel,[9] Viola and Seaborg,[10] and Horshoj *et al.*[11] However, most of these formulas are valid only within a limited region or predictions are not quite satisfactory. Poenaru and Ivascu[4] have proposed a new empirical formula which can well reproduce observed data but contains complicated corrections for the neutron and proton numbers and the magic numbers. In this work, we have aimed to derive an empirical equation which is simpler in form, easier to see the physical meanings, and applicable not only to heavy elements but also to lanthanide nuclides with high accuracy.

II. DERIVATION OF EMPIRICAL RELATION FOR EVEN-EVEN NUCLIDES

The alpha decay constant, λ, can be expressed by the equation,

$$\lambda=\omega P \ , \tag{1}$$

where ω is the frequency with which an α particle exists at the barrier, or the α-particle formation factor. P is the probability of transmission through the barrier.

If we consider α decay as a fission-like process,[5,12] the barrier shape for α decay can be depicted as shown in Fig. 1.[5] (It is to be noted that the discussion below has little to do with the model of a fission-like process.) The WKB (Wentzel-Kramers-Brillouin) approximation of the penetrability, P, leads to

$$P \simeq \exp\left\{-\frac{2}{\hbar}\int_{R_0}^{R_2}\sqrt{2\mu[V(r)-Q_\alpha]}\right\}dr \ , \tag{2}$$

where

$$\mu=M_\alpha M_d/(M_\alpha+M_d)$$

and the subscripts α and d denote the α particle and the residual nuclides, respectively (see Fig. 1 for R_0 and R_2). Q_α is the measured alpha-particle decay energy, corrected for the recoil energy given to the heavy residual nuclide, and for the electron screening:[13]

$$Q_\alpha=E_\alpha A_p/A_d+\Delta E_{sc} \ , \quad \Delta E_{sc}=6.5\times10^{-5}\times Z^{1.4}\ \text{MeV} \ , \tag{3}$$

where A_p and Z refer to the mass number and the atomic number of the parent nuclide, respectively. A_d is the mass number of the residual nuclide. The centrifugal potential is neglected because only s-wave alpha-particle

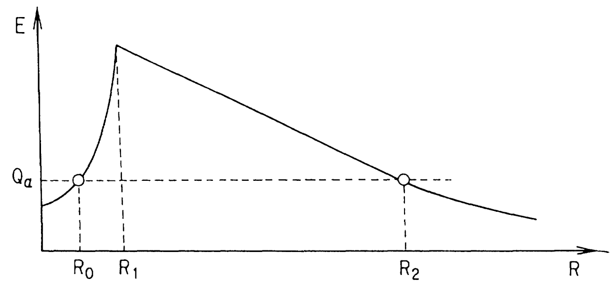


FIG. 1. Schematic drawing of the barrier shape for α decay.

($l=0$) emission is considered in this work. The potential $V(r)$ is divided into two regions. It may be approximated by the Coulomb barrier for $r \geq R_1$, and expressed as $V_i(r)$ for $r < R_1$. The integral in the exponent can be divided into two terms:

$$\int_{R_0}^{R_1} \sqrt{2\mu[V_i(r)-Q_\alpha]}dr + \int_{R_1}^{R_2} \left[2\mu \left[\frac{2Z_d e^2}{r} - Q_\alpha\right]\right]^{1/2} dr \ . \quad (4)$$

The second term mainly contributes to the penetrability P. It can be solved exactly in an analytical form and the penetrability due to the second term P_2 is expressed as

$$P_2 \simeq \exp\left\{-\frac{8e^2}{\hbar}\sqrt{2m}\,Z_d \left[\frac{A_d}{A_p Q_\alpha}\right]^{1/2} [\arccos\sqrt{X} - \sqrt{X(1-X)}]\right\} , \quad (5)$$

where

$$X = R_1/R_2 = r_0(A_d^{1/3}+4^{1/3}) \times \frac{Q_\alpha}{2Z_d e^2} ,$$

and m is the mass of one atomic mass unit, and r_0 is taken to be 1.2249 fm.[14]

The relation between the half-life $T_{1/2}$ (sec) and the Q_α (MeV) value can be expressed by the following equation where the penetrability due to the first term in Eq. (4) is denoted by P_1:

$$\log T = 1.098\,57 \times Z_d \left\{ \left[\frac{A_d}{A_p Q_\alpha}\right]^{1/2} \times [\arccos\sqrt{X} - \sqrt{X(1-X)}] \right\} + C , \quad (6)$$

where

$$C = \log_{10}(\ln 2/\omega) - \log_{10} P_1 \ .$$

C contains the frequency factor and the effect of the potential, $V_i(r)$. In order to examine the dependence of P_1 on X, the C value [the logarithm of the observed α-decay partial half-life minus the first term on the right hand side of Eq. (6)] for even-even nuclides of Th, Ra, Rn, and Po is plotted vs X in Fig. 2. Nuclides with $N \leq 126$ are not included in the figure to avoid the strong influence of the neutron shell ($N=126$, see Fig. 3) which is treated later. Figure 2 shows that the C value for each element is almost constant of X within the region of $X=0.15-0.34$ although slight deviations are observed for nuclides with $N=128$ and 130. This means that the X dependence of the decay probability is approximately treated by the

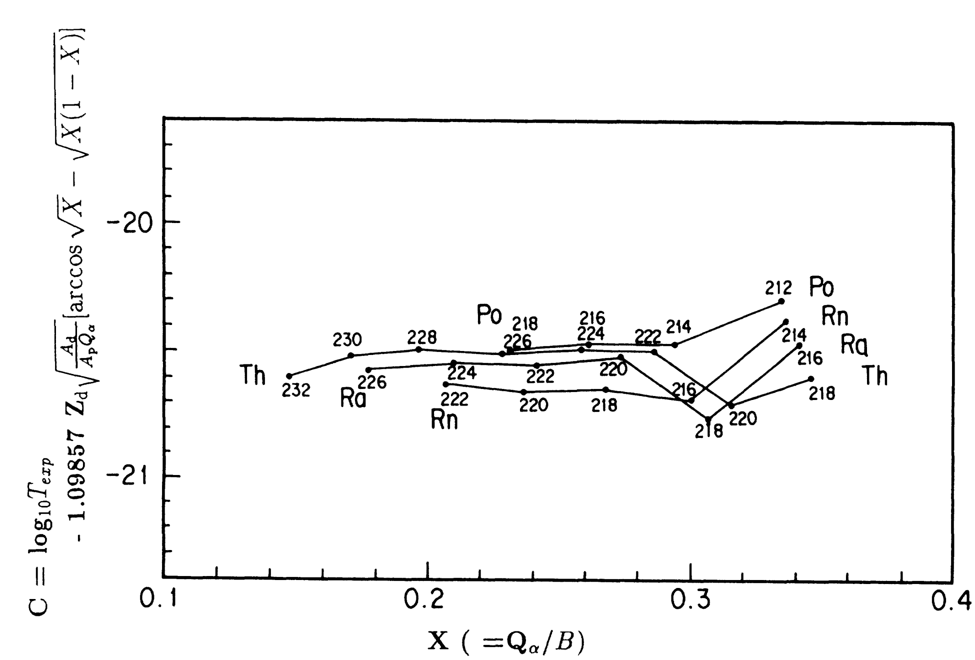


FIG. 2. The C value, the logarithm of the observed α decay partial half-life minus the first term on the right hand side of Eq. (6), is plotted for even-even nuclides of Th, Ra, Rn, and Po vs X. See text for the explanation of C and X.

functional form of the first term of Eq. (6) and that the effect of the inner barrier is minor. However, the C value clearly shows a Z dependence. From the smoothly varying feature of C values with Z, it is hard to describe that the Z dependence is caused by the nature of the orbital of the last two paired protons. This Z dependence may rather originate from the penetrability through the inner barrier P_1 and from the Z dependence of R_1 as expected if an attractive nuclear potential is taken into account, or a slight difference of the real outer barrier from an ideal Coulomb potential. There are two ways to take this Z dependence into consideration. One is to add another Z-dependent term in Eq. (6) as Horshoj *et al.* showed,[11] and the other is to find an appropriate function of Z as the coefficient outside the braces of the first term. In this work, the latter approach is taken.

In order to seek the Z dependence of the first term in Eq. (6), the alpha partial half-lives are plotted versus the quantity within the braces of the first term,

$$\left[\frac{A_d}{A_p Q_\alpha}\right]^{1/2} \times [\arccos\sqrt{X} - \sqrt{X(1-X)}] ,$$

for the $l=0$ transitions of even-even nuclides. The results are shown in Figs. 3 and 4, which are similar to Rasmussen's plot[12] of $\log_{10} T_{1/2}$ vs $Q_\alpha^{-1/2}$. Figure 3 indicates that parent nuclides with neutron numbers smaller than 126 should be treated differently for elements $Z=84$ through 90. In Fig. 4, the nuclides with neutron numbers smaller than 126 and $Z=84$ through 90, are excluded to observe the overall systematic trend. It is interesting to note that the data points in Fig. 4 fall on a straight line fairly well for each element from Gd to Fm and the slope gradually increases as the atomic number of the parent nuclide becomes larger. The slope is evaluated by least-squares fitting for each element for which more than two data points are available, with an assumption of a constant frequency, ω, for all nuclides. For the frequency, we have used the value recommended by Poenaru *et al.*;[4] i.e., $\log_{10}(\ln 2/\omega) = -20.446$. The results of fitting are drawn by solid lines in Fig. 4. The slopes thus deduced are plotted in Fig. 5 as a function of the atomic number of the parent nuclide Z. The points for $Z=82, 80, 78$ are slightly larger than those expected from the smooth variation from $Z=62$ through 100. It may be reasonable to consider that the deviation is caused by the effect of the proton shell $Z=82$. Figure 5 shows that Eq. (6) should be rewritten in the form,

$$\log_{10} T = A(Z) \times \left[\frac{A_d}{A_p Q_\alpha}\right]^{1/2} \times [\arccos\sqrt{X} - \sqrt{X(1-x)}] - 20.446. \quad (7)$$

The function $A(Z)$ can be empirically determined by seeking for an appropriate functional form that gives the best fit to the points in Fig. 5. After several trials, the following form has been found best:

$$A(Z) = 1.40 \times Z + 1710/Z - 47.7 , \quad (8)$$

in which the points for $Z=82-78$ were excluded because they seemed to be influenced by the closed shell effect.

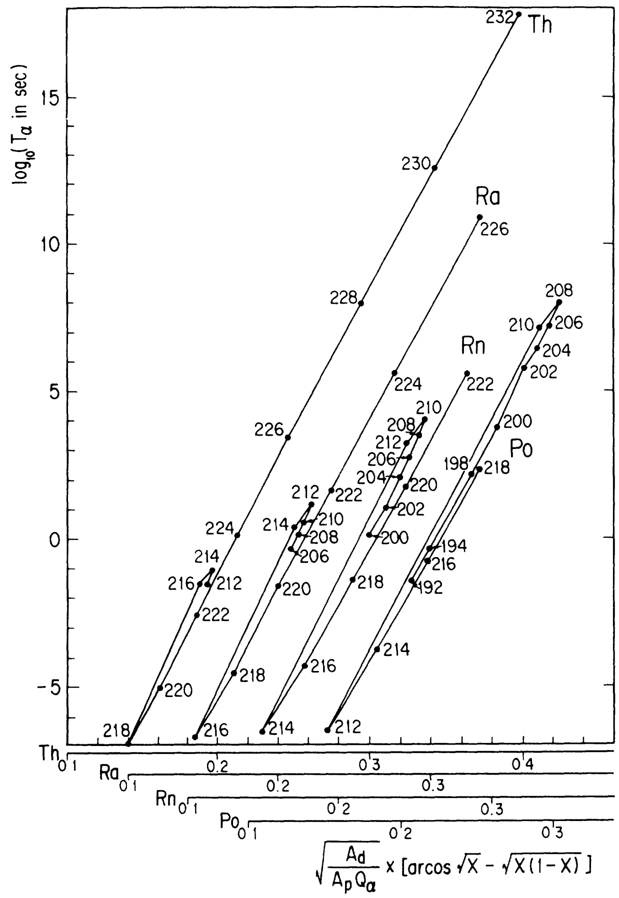


FIG. 3. The logarithm of the observed alpha partial half-life versus the X-dependent term in Eq. (5) for even-even nuclides of Th, Ra, Rn, and Po. Those with $N \leq 126$ are seen to behave differently, probably due to the effect of the neutron shell.

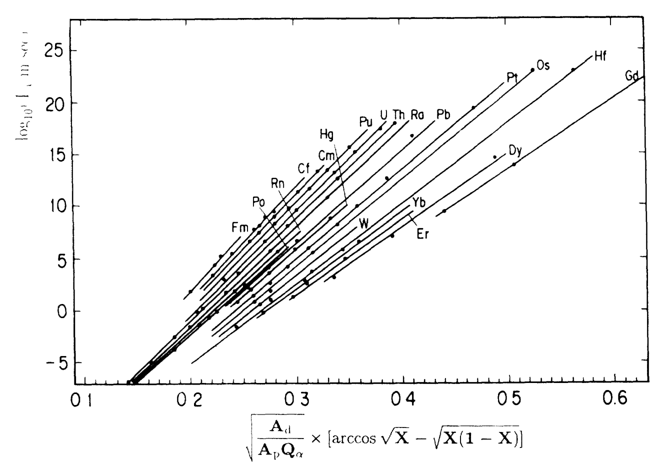


FIG. 4. The same as Fig. 3 for all the even-even nuclides with $Z=64-100$. Those apparently affected by the $N=126$ shell (see Fig. 3) are omitted. The solid line shows the result of least-squares fitting for each element.

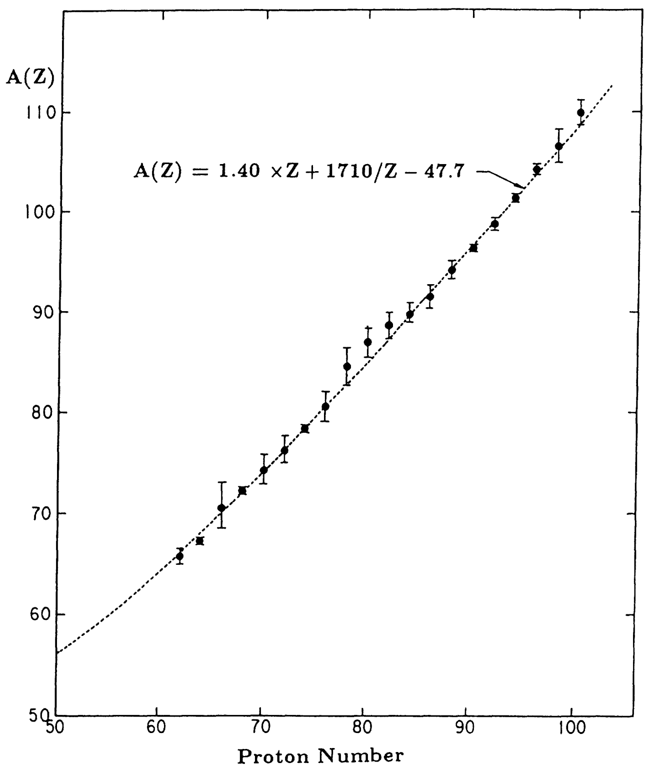


FIG. 5. The slope obtained in Fig. 4 is plotted vs Z. The dashed line shows the function, $A(Z)=1.40Z+1710/Z-47.7$, which gives the best fit to all the points excluding those for $Z=78$, 80, and 82.

The agreement between the half-lives predicted by the empirical equations (7) and (8) with the experimental data is shown in Fig. 6 where $\log_{10}(T_{\text{cal}}/T_{\text{exp}})$ is plotted versus the neutron number of the parent nuclides. A large discrepancy is observed for the nuclides with $78 \le Z \le 90$, $100 \le N \le 126$, as pointed out above. The discrepancy of those nuclides is enlarged in Fig. 7 in terms of the hindrance factor defined as $T_{\text{exp}}/T_{\text{cal}}$ and in $\log_{10}(T_{\text{exp}}/T_{\text{cal}})$. The discrepancy is apparently caused by the closed shells of $N=126$ and $Z=82$, and some systematic variation can be observed as a function of Z and A except for the different behavior of Pb isotopes. The shell effect on the formation factor ω can be approximately taken into account by the following purely empirical formula:

$$[1.94-0.020(82-Z)-0.070(126-N)] \quad \text{for } 78 \le Z < 82, 100 \le N < 126 , \tag{9}$$

$$[1.42-0.105(Z-82)-0.067(126-N)] \quad \text{for } 82 < Z \le 90, 110 \le N \le 126 . \tag{10}$$

The proposed new equation applicable for a wide region of even-even nuclides is

$$\log_{10} T = A(Z) \times \left[\frac{A_d}{A_p Q_\alpha} \right]^{1/2} \times [\arccos\sqrt{X} - \sqrt{X(1-X)}] - 20.446 + C(Z,N) , \tag{11}$$

where $C(Z,N)=0$ for the ordinary region outside closed shells,

$$C(Z,N)=[1.94-0.020(82-Z)-0.070(126-N)] \quad \text{for } 78 \le Z < 82, 100 \le N < 126 ,$$

$$C(Z,N)=[1.42-0.105(Z-82)-0.067(126-N)] \quad \text{for } 82 < Z \le 90, 110 \le N \le 126 .$$

III. EVALUATION OF THE DERIVED EMPIRICAL RELATION FOR EVEN-EVEN NUCLIDES

In order to evaluate the proposed relation, the calculated favored alpha half-lives of even-even nuclides are compared with the experimental data as shown in Fig. 8. To aid the eye, consecutive isotopes of a given element are connected with a line segment. Calculated results agree with the experimental data in a wide region from $Z=100$ to $Z=62$. The deviation in the actinide region is small. The influence of $N=152$, which is known to form a "deformed shell," is observed for Cf and Fm, but for Cm. As the influence observed is much smaller than that of $N=126$, no correction for the effect of $N=152$ has been made. A large discrepancy exists for Pb isotopes, but no

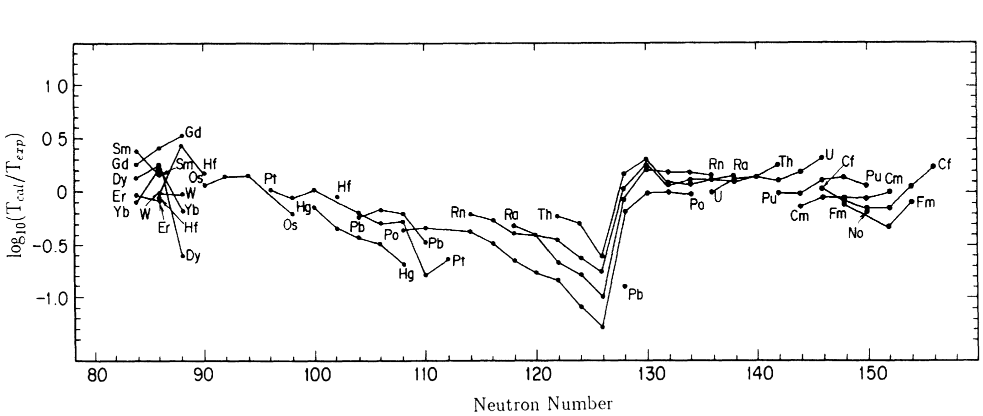


FIG. 6. Comparison of the observed half-lives with those predicted by Eqs. (7) and (8) in terms of the logarithm of the ratio, $\log_{10}(T_{\text{cal}}/T_{\text{exp}})$. No shell correction is applied.

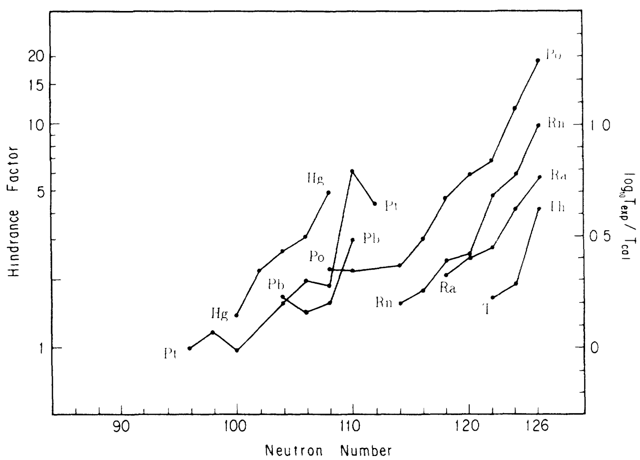


FIG. 7. A closer look at the discrepancy between the observed and the predicted half-life for the nuclides with $78 \leq Z \leq 90$, $100 \leq N \leq 126$.

fitting has been attempted to those experimental data because they are reported to have a large uncertainty.

The standard deviation (σ) of $\log_{10}(T_{exp}/T_{cal})$ was 0.13 for the nuclides in the heavy element region ($Z \geq 84$, $N \geq 128$), and $\sigma = 0.18$ for all the nuclides treated in this work ranging from $Z = 62$ to 100. Other experimentally known alpha half-lives of light element isotopes, such as Te and Xe isotopes, were predicted well by the proposed equation. One of the reasons may lie in the use of the same frequency factor, $\log(\ln 2/\omega) = -20.446$, even for those lighter nuclides.

IV. SYSTEMATICS OF FAVORED TRANSITIONS OF ODD-MASS AND ODD-ODD NUCLIDES

The favored transitions of odd-A and odd-odd nuclides were compared with the values expected from the systematics for the transitions with $l = 0$ of even-even nuclides [Eq. (7)]. Generally, favored transitions of odd-A and odd-odd nuclides with $l = 0$ are more hindered than those expected from even-even nuclides, and the hindrance factors can be estimated by comparison of the observed partial half-lives with those calculated from Eq. (11). The experimental data used in this work have already been identified and listed in the literature.[15,16] To avoid the influence of the shell effect, nuclides with $N > 126$ have been chosen. The values $\log_{10}(T_{exp}/T_{cal})$ for odd-even and even-odd nuclides are shown in Figs. 9(a) and 9(b). From the figures, it is obviously difficult to find any systematic variation of the hindrance except the fact that there are highly hindered points in the vicinity of $N = 152$ and $N = 142-146$, and also some favored points at $N = 130$. The reason for the extremely favored transition of ^{209}Ac is unknown.

With a lack of systematic trend, we have decided to take an average of those values merely for a rough prediction of α half-lives of odd-even and even-odd nuclides. The average values are

$$T_{exp}/T_{cal} = 2.00 \pm 1.28 \quad (25 \text{ nuclides}) \quad \text{for even-odd ,}$$
$$T_{exp}/T_{cal} = 2.11 \pm 1.29 \quad (28 \text{ nuclides}) \quad \text{for odd-even .} \qquad (12)$$

It is interesting to note that the average hindrance factor for odd-A nuclides is about 2.0–2.1 regardless of whether the odd nucleon is a proton or neutron. For odd-odd nuclides, the number of identified favored transitions is very limited and they are listed in Table I together with the $\log_{10}(T_{exp}/T_{cal})$ values and hindrance factors (T_{exp}/T_{cal}). The large hindrance for ^{208}At, ^{206}At, and ^{252}Es is ascribed to the neutron shell effect for $110 \leq N \leq 126$ and $N = 152$ mentioned earlier. The averaged value in T_{exp}/T_{cal} of the nuclides excluding ^{208}At ($N = 123$) and ^{206}At ($N = 121$) and their standard deviations are

$$T_{exp}/T_{cal} = 3.15 \pm 1.86 \quad (6 \text{ nuclides}) \quad \text{for odd-odd .} \qquad (13)$$

The averaged value for odd-odd nuclides is not necessarily the product of hindrance factors for even-odd and odd-even nuclides, although more data are needed to make any statement.

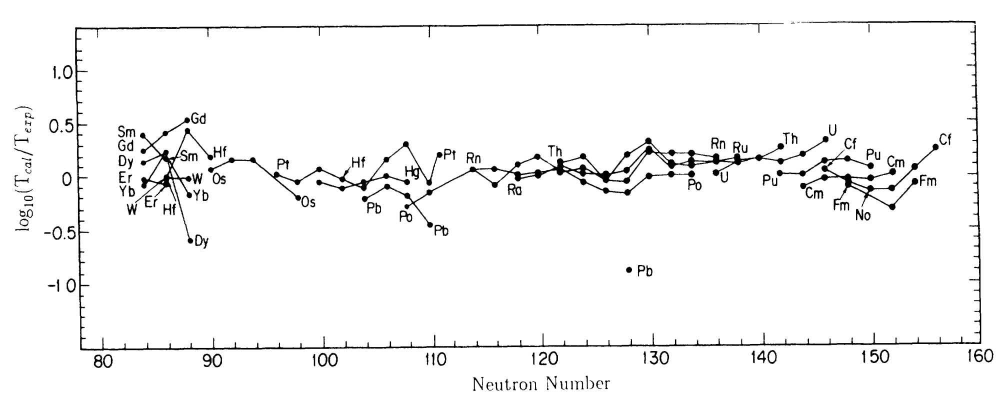


FIG. 8. Similar to Fig. 6 except that the shell correction is applied by Eqs. (9) and (10).

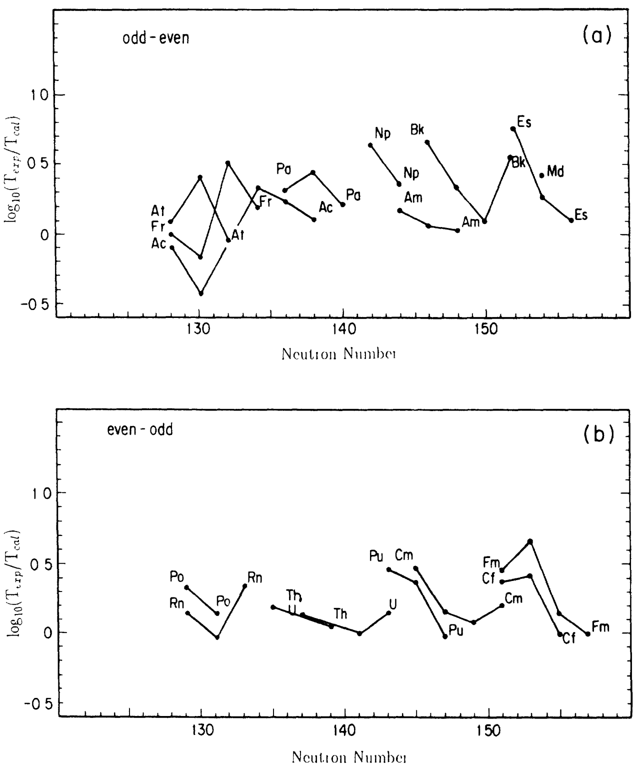


FIG. 9. The values $\log_{10}(T_{exp}/T_{cal})$ for the favored transitions of odd-even (a) and even-odd (b) nuclides.

TABLE I. Hindrance factors of the favored transition of odd-odd nuclides.

Nuclei	$\log_{10}(T_{exp}/T_{cal})$	Hindrance factor (T_{exp}/T_{cal})
^{254}Es	0.448	2.80
^{254}Es^m	0.570	3.71
^{252}Es	0.821	6.62
^{242}Am^m	0.175	1.49
^{218}At	0.372	2.36
^{216}At	0.278	1.89
^{208}At	1.236	17.22
^{206}At	0.732	5.40

V. PREDICTION OF PARTIAL HALF-LIVES FOR TRANSACTINIDES AND SUPERHEAVY ELEMENTS

Progress in the synthesis and study of heavy elements requires a continuous improvement in the theoretical prediction of the properties of those nuclides, and their identification can be best achieved by alpha-particle measurements. The predictive power of our empirical relation has been first tested by comparison with the observed alpha half-lives for nuclides with $Z \geq 100$. As the Q_α values are not definitely determined, we have used the mass table by Möller and Nix.[17] The predicted alpha half-lives for ground-to-ground state favored transitions are shown by dots and connected by line segments and compared with experimental data in Figs. 10(a)–10(j).

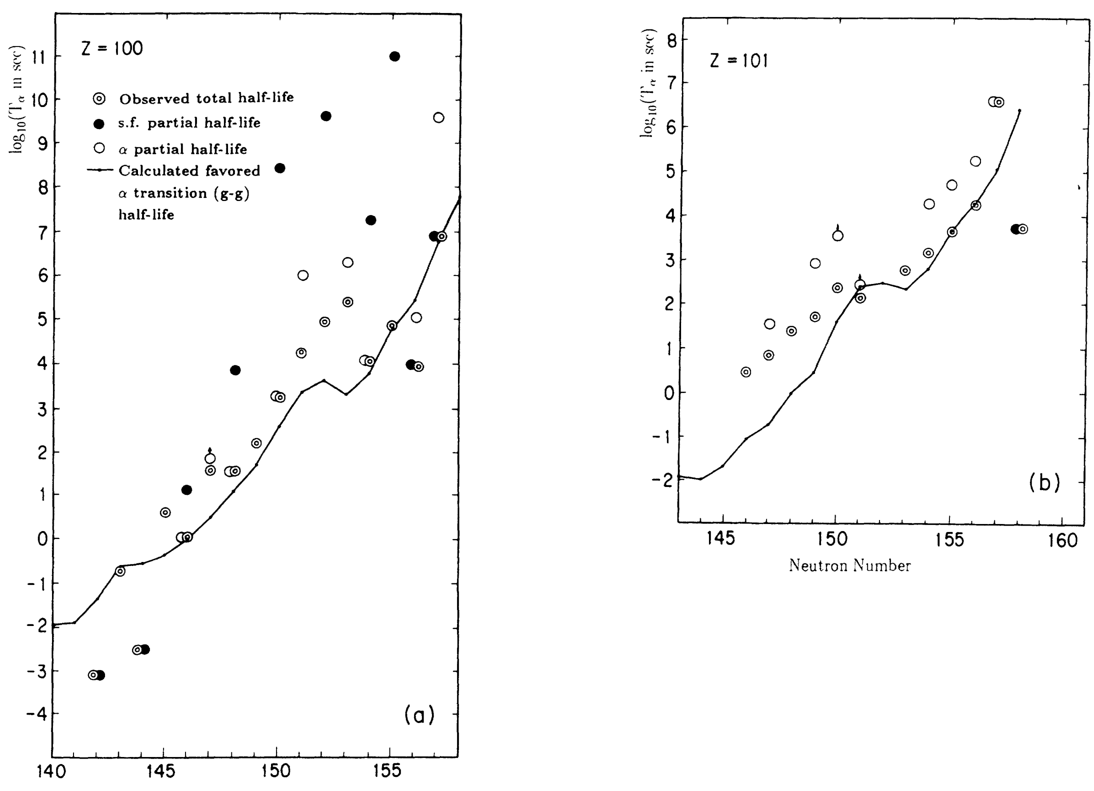


FIG. 10. (a)–(j) Plots of the predicted alpha half-lives for the ground-to-ground favored transitions for the isotopes $Z = 100$–109. Also plotted are the observed total half-lives (double circle), alpha partial half-lives (open circle)—not necessarily for favored transitions, and spontaneous fission partial half-lives (filled circles). The mass table by Möller and Nix is used for Q_α.

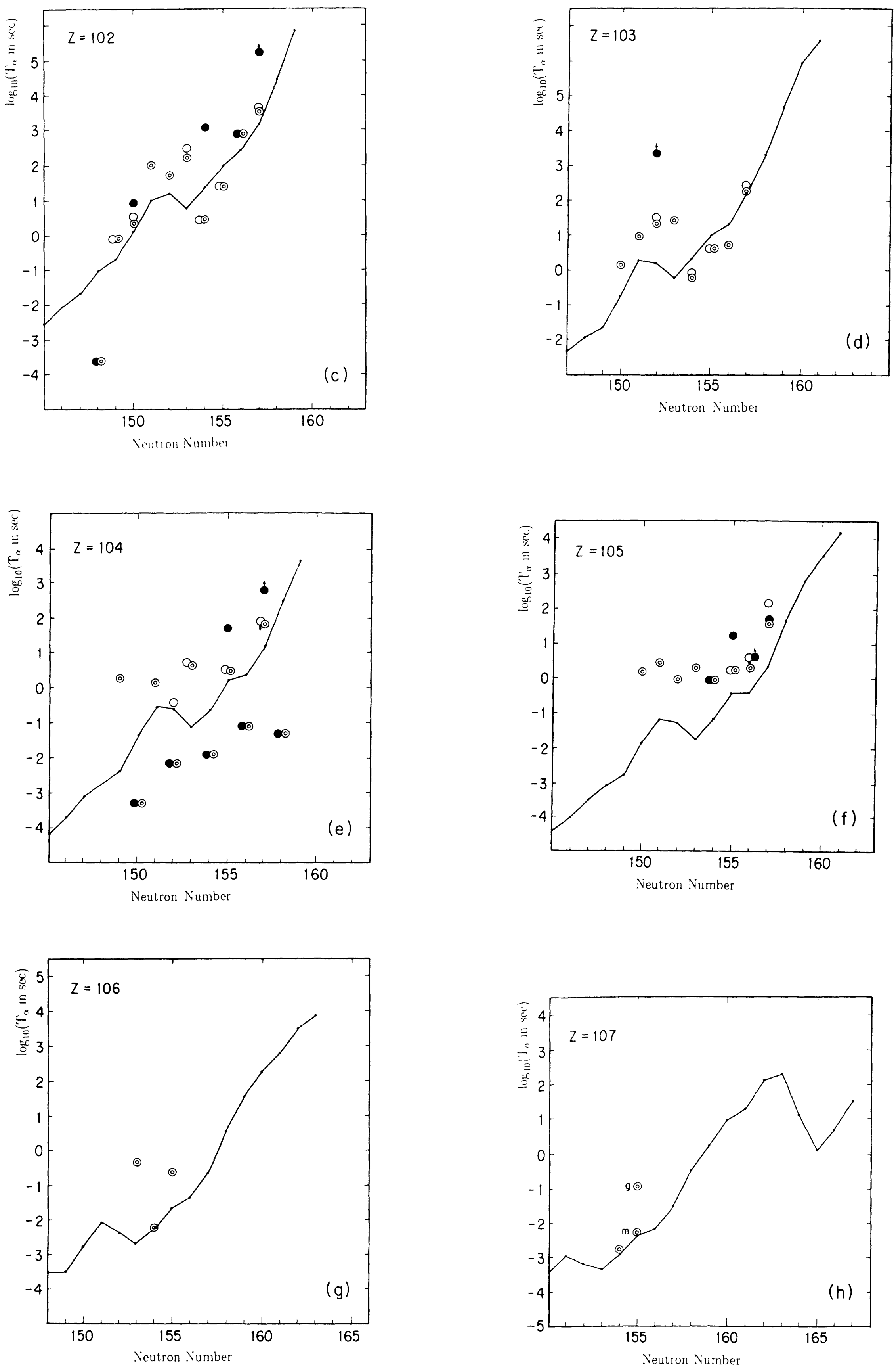
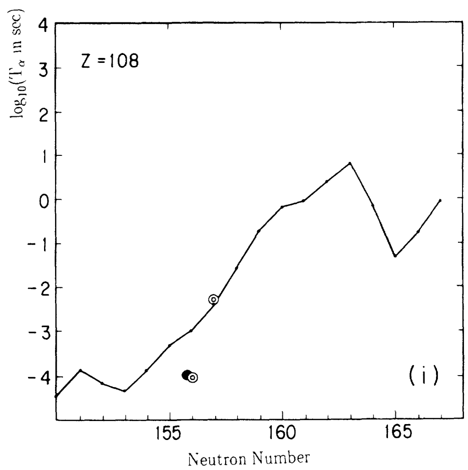


FIG. 10. (*Continued*).



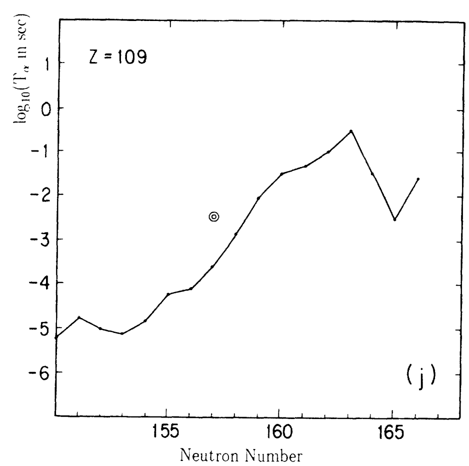


FIG. 10. (*Continued*).

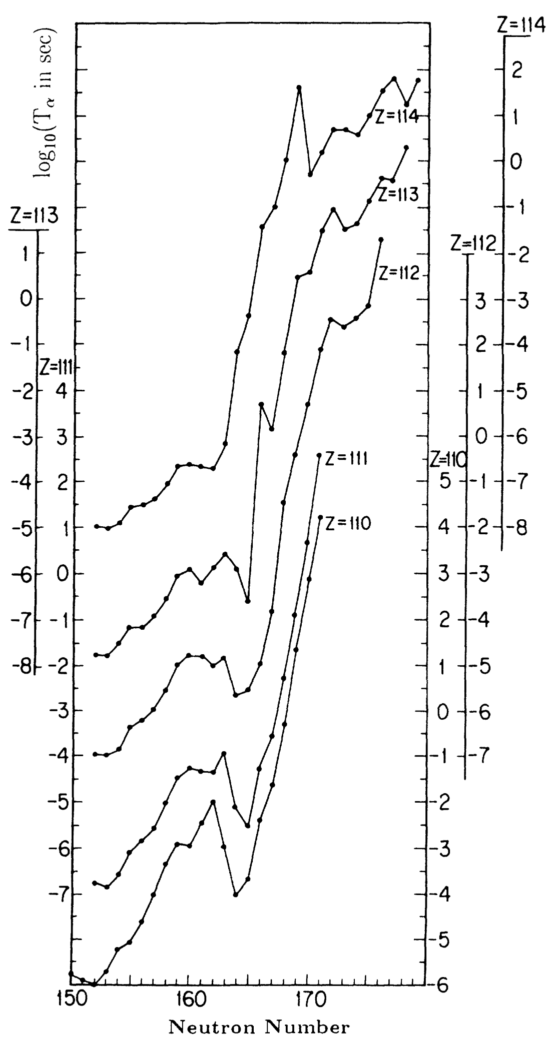


FIG. 11. Predicted alpha partial half-lives of the isotopes $Z=110-114$ for the ground-to-ground favored transitions. The mass table by Möller and Nix is used for Q_α.

The predicted values for odd-A and odd-odd nuclides are those of favored transitions. The observed total half-lives are shown by double circles. The partial alpha decay half-lives, not necessarily of favored transitions, are shown, if they are known, by open circles. Known partial spontaneous fission half-lives are designated by filled circles. For $Z=100$ through 104, alpha decay favored transitions have been observed for only eight even-even nuclides and their half-lives can be reproduced by the proposed relation within a range of $-0.25-+0.70$ in $\log_{10}(T_{\text{exp}}/T_{\text{cal}})$, its average being 0.16. It is to be noted that most of the observed total half-lives are longer than the predicted half-lives for favored ground-to-ground alpha transitions except for those of even-even nuclides for which spontaneous fission decay is known to be a dominant decay mode. It is also interesting to note that the observed half-life of ^{260}106, an even-even nuclide, is in agreement with the predicted alpha decay half-life, a fact which suggests that the spontaneous fission half-life for ^{260}106 is much longer than 6.3 ms. This supports the statement by Boning *et al.*[18] that the spontaneous fission half-life becomes longer than the α decay half-life in the $Z \geq 106$ region. But ^{264}108 has a spontaneous fission half-life magnitude shorter than the predicted alpha half-life by one order of magnitude.

The predicted α partial half-lives for $Z \geq 110$ isotopes for which no experimental data are available are shown in Fig. 11. As mentioned above, the predicted α half-lives are for the favored transitions between the ground state of the parent nuclide and the ground state of the decay product in the decay of even-even nuclides. For odd-A and odd-odd nuclides, therefore, the α decay partial half-lives are expected to be much longer due to the large hindrance factor and/or the smaller transition energy.

VI. CONCLUSION

A simple semiempirical relation for the prediction of the partial alpha half-lives of the favored transition in even-even nuclei has been successfully derived. With an incorporation of the correction factor for the closed shells, $N = 126$ or $Z = 82$, the equation can reproduce observed partial alpha half-lives for a wide range of nuclides of $Z \geq 62$ and $N \geq 84$ with a standard deviation of 0.18 in $\log_{10}(T_{exp}/T_{cal})$, and for a narrower region of nuclides of $Z \geq 84$ and $N \geq 128$ with 0.13. The averaged hindrance factors for the favored transitions in odd-A and odd-odd nuclides have been evaluated to be 2.00–2.11 and 3.15, respectively, where the hindrance factor is defined as the ratio of the observed partial half-life to that predicted by the present relation for those nuclides. The alpha partial half-lives of isotopes of transactinides and superheavy elements have been predicted for the ground-to-ground favored transitions using the Q_α values from the mass table by Moller and Nix.[17] All the observed total half-lives of heavy nuclides have been found to be longer than the predicted alpha partial half-lives except for some even-even nuclides which have very short spontaneous fission half-lives.

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