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ABSTRACT

Alpha decay energy data in the rare earth region are extended by means of closed decay energy cycle calculations and are then correlated for each element as a function of neutron number. The marked effect of the closed shell at 82 neutrons on the alpha decay energies is discussed. Evidence is presented for a sharp drop at 90 neutrons in the normal alpha-decay energy versus neutron number trend, as well as for a proton subshell at 64 protons. Half-lives are calculated using a formula derived from simple barrier-penetration theory. The calculated values are compared with experimental half-lives and discrepancies are discussed. Reduced level widths δ^2 are determined for rare earth alpha emitters using barrier penetration factors calculated from the real potential derived by optical model analysis of alpha elastic scattering data. The reduced widths are in turn used to propose hindrance factors for odd-nucleon alpha emitters.

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I. INTRODUCTION

From the observation of the slope of the mass-defect curve it has been noted that most isotopes whose mass numbers are greater than 150 are unstable with respect to alpha-particle emission. Radioactive decay by this means is commonly observed in both natural and artificial isotopes of elements whose atomic numbers are greater than 82. In marked contrast, alpha emission for isotopes of elements with $Z < 83$ is rather unusual. Prior to 1949 only one alpha-emitting isotope was known in the medium-heavy region, i.e. the alpha-emitting nuclide in natural samarium discovered by Hevesy and Pahl in 1932.[1] The lack of alpha-emitters in this region can be well accounted for by the fact that alpha-decay rates depend on the decay energies in a very sensitive exponential manner, as shown by the formulae developed independently by Gamow[2] and by Condon and Gurney.[3] Naturally-occurring isotopes in the medium-heavy element region might be unstable toward alpha-particle emission by as much as 2 Mev and still have half-lives too long to be readily observed.

Kohman made an analysis of alpha-particle binding energies in the same region based on the semi-empirical equation of Bohr and Wheeler and on the experimental mass-defect curve.[4] He predicted that sufficiently neutron-deficient isotopes of the medium-heavy elements might exhibit alpha decay. Such a prediction can also be made on the basis of alpha-decay energy systematics in the heavy elements where the trend (with two inversions which will be discussed later) is that of increasing alpha-decay energies with decreasing neutron numbers for a given atomic number. The electron-capture or positron decay energies also generally increase with decreasing neutron numbers. Thus, whether or not alpha emission could be detected in the medium-heavy elements as neutrons were removed before the total half-lives became inconveniently short had to be tested experimentally. In 1949 Thompson and co-workers reported positive results obtained in a survey in which targets of various elements of interest were subjected to high-energy particle bombardments.[5] They discovered alpha-radioactivity in neutron-deficient isotopes of gold and mercury and some rare-earth elements. Since that time other alpha emitters with $Z < 83$ have been reported in both naturally occurring and artificially produced isotopes. The majority of the latter were reported by Rasmussen, Thompson and Ghiorso in 1953.[6] In addition, Dunlavey and Seaborg produced Sm^{146} also in 1953,[7] while Toth and Rasmussen discovered and reported two new dysprosium alpha emitters in 1958.[8] The discovery of naturally occurring alpha emitters in the region below bismuth has been carried out chiefly by two groups (1) Kauw, Porschen and Riezler who have used nuclear emulsions to detect the alpha radioactivity[9-13] and (2) Macfarlane and Kohman who have used an ionization chamber as the detector.[14] As a result there are known at present eighteen alpha emitters with $Z < 83$ whose mass numbers are fairly certain.

The present study is limited to the rare earths because fifteen of these alpha emitters have atomic numbers between 60 (neodymium) and 66 (dysprosium). The clustering of alpha radioactivity in the 82-neutron region allows correlation of alpha decay-energy and decay-rate data in a manner similar to that used in studies of alpha systematics in the heavy elements. In this paper alpha decay data have been correlated and possible conclusions as well as predictions derived on the basis of such correlations.

II. SUMMARY OF INFORMATION

The information on rare earth alpha emitters is summarized in Table I.

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Table I

Summary of information

Alpha emitter	Total half-life	Other decay modes observed	Alpha energy (Mev)	Method of detection	Alpha half-life	Method of $T_{1/2}$ α determination	Reference
Dy^{154}	13hr±2[a]	none observed	3.35±0.05[a]	ionization chamber			8
Dy^{153}	5hr±0.5	indirect evidence for E.C. decay	3.48±0.05	ionization chamber	~13.4y	estimated from reaction yield	8,15
Dy^{152}	2.3hr±0.2	E.C. and β+	3.66±0.05	ionization chamber	~1.45y	estimated from reaction yield	6,8,15,16
Dy^{151}	19min±4	E.C. and β+	4.06±0.04	ionization chamber			6,8,16
Dy^{150}	7min±2	E.C. and β+	4.21±0.06	ionization chamber			6,8,16
Tb^{151}	19hr±1	E.C.	3.44±0.1	ionization chamber	$\sim 7.4 \times 10^{2}$y	$\frac{\text{x ray}}{\alpha}$ ratio	6,8,15
Tb^{149}	4.1hr±0.2	E.C.	3.95±0.02	alpha spectrograph	36hr±7	$\frac{\text{total}}{\alpha}$ ratio	6,16,17
Gd^{152}		natural β stable isotope	2.15±0.03	ionization chamber	$1.13 \times 10^{14} \pm 0.08$y	decay rate of known number of atoms	14
Gd^{150}	10^{5}y	β-stable	2.70±0.15	ionization chamber	$\sim 3 \times 10^{5}$y	"	6,15
Gd^{149}	9.0d±1	E.C.	3.00±0.15	ionization chamber	$\sim 4 \times 10^{3}$y	$\frac{\text{x ray}}{\alpha}$ ratio	6
Gd^{148}		β-stable(?)	3.16±0.10	ionization chamber	$\sim 1.3 \times 10^{2}$y	estimated from reaction yield	6,15
Eu^{147}	24d±2	E.C.	2.88±0.10	ionization chamber	$\sim 4.4 \times 10^{3}$y	$\frac{\text{x ray}}{\alpha}$ ratio	6,15

Table I (cont'd.)

Alpha emitter	Total half-life	Other decay modes observed	Alpha energy (Mev)	Method of detection	Alpha half-life	Method of $T_{1/2}$ α determination	Reference
Sm^{147}		natural β-stable isotope	2.18±0.02	ionization chamber	$\sim 1.3\times 10^{11}$ y	decay rate of known number of atoms	1,18,19
"		"	2.24±0.02	ionization chamber	$1.18\pm 0.05\times 10^{11}$ y	"	14
Sm^{146}		β-stable	2.55±0.05	nuclear emulsion	$\sim 5\times 10^{7}$ y	estimated from reaction yield	7
Nd^{144}		natural β-stable isotope	1.90±0.1	nuclear emulsion	$\sim 1.5\times 10^{15}$ y	decay rate of known number of atoms	20
"		natural β-stable isotope	1.84±0.02	ionization chamber	$2.36\pm 0.29\times 10^{15}$ y	"	14

[a]In recent preliminary results R. D. Macfarlane in Berkeley a long-lived dysprosium activity of about 2.9 Mev decay energy. If this newer activity indeed represents the ground state decay of Dy^{154} with the 3.35 Mev activity belonging to an isomer, then some of the discussion of this paper regarding an α-energy discontinuity between 88 and 90 neutrons would be altered.

III. ALPHA DECAY ENERGY SYSTEMATICS

Decay energy information is most conveniently summarized in a diagram where alpha-decay energies are plotted as a function of neutron (or mass) number. Compilations of similar data in the heavy elements have shown that in such a plot the points for a given element lie on an approximately straight line. As mentioned briefly in the introduction alpha-decay energies for a given Z number increase with decreasing neutron number. Discontinuities in this monotonic trend appear in two places in the heavy element region. The first, an extremely pronounced one, appears at 128 neutrons as a consequence of the closed shell at 126 neutrons. As a result of the major closed shell the maximum alpha-decay energy for a given element is reached at 128 neutrons due to the abnormally low neutron binding energies just beyond the closed shell. Alpha-decay energies are abnormally low for isotopes with 126 neutrons. The normal trend is found to resume in isotopes with less than 125 neutrons. A similar but much less pronounced effect takes place at 154 neutrons, presumably as a consequence of a neutron subshell at 152 neutrons.[21]

Information in the rare earth region ($60 \leq Z \leq 66$) is much more fragmentary, but the basic structure of increasing alpha-decay energy with decreasing neutron number is unmistakable. The influence of the closed shell at 82 neutrons is also similar to that of the shell at 126 neutrons. The maximum in alpha-decay energies is reached at 84 neutrons for a particular Z number. A discontinuity in the normal trend is presumably present at this point, since no isotopes with neutron number less than 84 have been found to emit alpha-particles despite the fact that several such isotopes with reasonable half-lives are known in this region. At a later point in this section additional evidence is presented to support the idea of a discontinuity at 84 neutrons.

The alpha-decay energy data shown in Table I can be supplemented by means of closed energy cycles. The method consists of predicting decay energies by the construction of a system of energy-balance cycles from existing alpha- and beta-energies in which an individual closed cycle is made up of two alpha- and two beta-decay energies. Then, if 3 of the 4 pieces of information that constitute a cycle are known, the fourth can be calculated. The method has been highly successful in the heavy element region.[22] The application of a similar method in the 82-neutron region is limited due to a number of reasons, the chief of which is scarcity of experimentally known alpha energies. This can be at least partially overcome through the interpolation and extrapolation of the alpha-decay energy versus neutron number family of curves. Another reason for the limited applicability of the closed-cycle method is that alpha emitters in this region decay by electron-capture, a process for which it is generally not possible to make a direct experimental measurement of the decay energy. A systematic study of closed-energy cycles in a limited region, however, has been carried out because new information does come to light. The cycles are divided into 4 sets since they connect only nuclides differing in mass number by four, and these sets are labelled, 4n, 4n+1, 4n+2, and 4n+3 respectively. The notation can be illustrated thus: "4n+2" means that the mass numbers of nuclides of this type when divided by four have a remainder of 2 and so on. The cycles are shown in Figs. 1-4. The beta-decay energies and lower limits of these numbers used in the cycles have been taken chiefly from a compilation by King,[23] and a later one by Lidofsky[24] and from the new Table of Isotopes.[25] The three exceptions are: (1) Pm^{144} E.C. decay energy > 1.8 Mev, obtained from some recent work by Toth and Nielsen;[26] (2) Eu^{146} and Gd^{146} E.C. decay energies, 3.30 and 2.15 Mev respectively, taken from a publication by Gorodinski et al.[27]; and (3) Nd^{151} Q_{β^-} = 2.4 Mev, and Sm^{155} Q_{β^-} = 1.8 Mev, taken from two recent publications of Schmid and Burson.[28,29] The Q_{eff}'s (alpha-decay energies) used throughout the cycle calculations are known as the effective decay energies and consist of the total alpha disintegration energies of the bare nuclei. These Q_{eff}'s are equal to the alpha particle energies in the laboratory system plus the recoil energy plus the orbital electron screening correction, $\Delta E_{S.C.}$, where $\Delta E_{S.C.} = 65.3 (Z+2)^{7/5} - 80 (Z+2)^{2/5}$ e.v., and is approximately equal to 20 kev for the rare earth nuclides.

Using only experimental Q_{eff}'s the following decay energies can be calculated using closed cycles:

(a) <u>4n cycle</u>	Eu^{152}	Q_{eff}	= 1.33 Mev
	Pm^{144}	Q_{eff}	> 0.52 Mev
	Pr^{144}	Q_{eff}	= 1.19 Mev
	Ce^{144}	Q_{eff}	= 0.44 Mev
(b) <u>4n+1 cycle</u>	Eu^{149}	Q_{eff}	< 3.23 Mev
	Sm^{149}	Q_{eff}	< 2.80 Mev
	Pm^{149}	Q_{eff}	< 2.44 Mev
	Nd^{149}	Q_{eff}	< 2.04 Mev
(c) <u>4n+2 cycle</u>	Eu^{150}	Q_{eff}	< 3.17 Mev
	Pm^{146}	Q_{eff}	> 1.18 Mev

(d) <u>4n+3 cycle</u>

Dy^{151}	$Q_{E.C.}$	> 1.27 Mev
Gd^{147}	Q_{eff}	> 0.11 Mev
Pm^{147}	Q_{eff}	= 1.56 Mev
Nd^{147}	Q_{eff}	= 0.93 Mev

The calculated values for Eu^{152}, Pm^{147}, Nd^{147}, Pr^{144} and Ce^{144} were added to the experimentally determined ones, bringing the total of known alpha decay energies to 20. The 20 numbers were then used to draw Fig. 5 in which they were plotted against neutron numbers. The points for particular elements have been joined in an idealized fashion by drawing straight lines through them. It is these lines that were used in estimating unknown alpha-decay energies by means of either interpolation or extrapolation. The fact that the points should in reality be joined by a series of roughly parallel zigzagging lines rather than straight lines introduces an error into any Q_{eff} that is read off these lines. However, under the circumstances (for some elements only one point is known) such straightline extrapolations and interpolations are the best that can be made.

We will now illustrate the manner in which the remainder of the cycles were constructed by two examples. In the first case 1.65 Mev is read off the praseodymium line for the Q_{eff} of Pr^{143}, and the Q_{eff}'s for Ce^{143} and Nd^{143} are calculated to be 0.81 and 0.45 Mev, respectively. The Ce^{143} value thus determined is in good agreement with the Q_{eff} estimated from Fig. 5. The second case represents a less straightforward calculation. The Q_{eff} for Pm^{146} was found to be > 1.18 Mev using the experimentally determined Sm^{146} alpha-decay energy. The lower limit agrees with the value of 1.95 Mev estimated from the promethium line. Using this approximate Q_{eff} for Pm^{146} one calculates the

Q_{β^-} for Pm^{146} to be about 1.47 Mev. In turn, using this Q_{β^-}, the Eu^{150} Q_{eff} is found to be about 2.4 Mev in good agreement with the approximate value of 2.15 Mev read off the europium curve in Fig. 5. The numbers shown in the closed cycles are labelled as to how they have been determined. Rather than go through each cycle individually, only those of special interest will be discussed.

The Q_{eff}'s for Sm^{145} and Nd^{143} have been calculated to be 1.25 and 0.45 Mev respectively. The two isotopes each have 83 neutrons and their Q_{eff}'s are to be compared with the experimental alpha-decay energies of Sm^{146} and Nd^{144}. In both cases there are decreases of approximately 1.4 Mev for the Q_{eff}'s in going from 84- to 83-neutron isotopes. This is evidence that the 82-neutron closed shell does indeed exert the same influence on alpha-decay energies as does the 126-neutron shell in the heavy elements. The Sm^{145} and Nd^{143} points have been placed in Fig. 5 and two lines drawn connecting these points to those of Sm^{146} and Nd^{144}, respectively. Similar lines have been drawn for the other elements parallel to the first two. The calculated limits for the Q_{eff}'s of Gd^{147} and Pm^{144} (also 83-neutron isotopes) of > 0.11 and > 0.51 Mev, respectively, are seen to be in agreement with the values read off the gadolinium and promethium lines. Additional evidence concerning the influence of the 82-neutron shell may be obtained in the following manner. The Q_{eff}'s for the 83-neutron isotopes Pr^{142} and Ce^{141} can be estimated to be 0.10 and -0.20 Mev, respectively. Using these Q_{eff}'s the alpha-decay energies of the 82-neutron nuclides, Nd^{142} and Pr^{141}, are found to be negative. The negative alpha-decay energies not only for the two 82-neutron isotopes but also for Ce^{141} indicate that the three nuclides are stable with respect to emission of alpha particles.

In general the closed-cycle calculations are internally consistent--indicating that the accuracy of the curves in Fig. 5 is fair considering the idealized straight-line extrapolations. Some discrepancies do arise. Beta-decay energies taken from King's compilation seem almost certainly to be in error in two instances. These are: (a) Pm^{149} $Q_{\beta-}$ 1.34 Mev, and (b) Pm^{148} $Q_{\beta-}$ 2.7 Mev. The first value is listed in King's compilation with a question-mark after it, indicating that the number is uncertain. Considering the uncertainty we should like to propose a $Q_{\beta-}$ of 0.90 Mev for Pm^{149} thus removing the inconsistency. The $Q_{\beta-}$ of Pm^{148} is employed in two cycle calculations. In one, the Q_{eff} of Eu^{152} used in Fig. 5 is calculated. In the second, using 1.25 Mev for the Pm^{148} Q_{eff} that of Sm^{148} is found to be 1.55 Mev while the number estimated from Fig. 5 is 2.0 Mev. If the Pm^{148} $Q_{\beta-}$ is decreased by say 0.25 Mev, then the calculated Sm^{148} Q_{eff} is in essential agreement with the Fig. 5 value. The Eu^{152} Q_{eff} becomes 1.58 Mev, a number that is as much above the europium line in Fig. 5 as the 1.33 Mev value is below that line. We would therefore propose that the correct Pm^{148} $Q_{\beta-}$ is less than 2.7 Mev and is closer to 2.45 Mev. Another inconsistency is apparent in the Gd^{155}, Eu^{155}, Sm^{151}, and Pm^{151} closed cycle. Here the calculated Eu^{155} Q_{eff} is approximately 0.55 Mev less than the Fig. 5 value. The Eu^{155} $Q_{\beta-}$ is probably correct. That of Pm^{151} is less certain. Perhaps the Pm^{151} $Q_{\beta-}$ is in error. A more striking inconsistency appears in connection with the following two beta-decay energies: Pm^{150} $Q_{\beta-}$ (5.0 Mev) and Pr^{146} $Q_{\beta-}$ (4.2 Mev). Because the Q_{eff} of Sm^{150} must be > the Q_{eff} of Pm^{150}, the $Q_{\beta-}$ of Pr^{146} should be > the $Q_{\beta-}$ of Pm^{150}. Another indication that the Pm^{150} $Q_{\beta-}$ is too large is found if the Q_{eff} of Eu^{154} is estimated to be 0.90 Mev. Then the calculated Gd^{154} Q_{eff} is almost 2.5 Mev larger than that expected from Fig. 5. The alpha decay energy of La^{141} has been calculated to be approximately 1.05 Mev. Starting with this point, a line may be drawn parallel to the cerium line in Fig. 5. From the lanthanum line the Q_{eff} of La^{142} can be estimated to be 0.65 Mev. Working backwards, the two beta-decay energies are found to be: (a) Pr^{146} $Q_{\beta-}$ 3.39 Mev and (b) Pm^{150} $Q_{\beta-}$ 2.49 Mev. Now using the above-estimated Eu^{154} Q_{eff} that of Gd^{154} is found to be 1.42 Mev in agreement with the value of 1.50 Mev obtained from Fig. 5.

The discrepancies in the two other instances seem to be due to rather sharp drops in the alpha-decay energy versus neutron number curves somewhere between 88 and 90 neutrons. There is experimental evidence in the case of the dysprosium alpha emitters of such an abrupt decrease. Dy^{154} has been found to have an energy of 3.46 Mev,[8] a number somewhat higher than would be expected but not in serious disagreement with the straight line drawn through the other four dysprosium decay energies. Recently Riezler and Kauw have searched for alpha activity in Dy^{156} using dysprosium enriched in Dy^{156}.[12] They report that they were unable to see any alpha tracks attributable to Dy^{156} and set a lower limit of 10^{18} years for the isotope's alpha half-life by alpha decay of energy exceeding 2.2 Mev. The expected energy for Dy^{156} according to the line in Fig. 5 should be about 2.7 Mev, so that there seems to be a sharp decrease in energy of at least half a Mev. There are rather abrupt nuclear-structure changes between 88 and 90 neutrons in neighboring rare-earth nuclides, as reflected in first-excited-state energies and spectroscopic isotope shifts of even-even nuclei and in quadrupole and magnetic moments in Eu^{151} and Eu^{153}. Perhaps the discontinuity is related to these sudden changes in isotopes having 88 and 90 neutrons. The sharp decrease could account for the following discrepancies that arise in our cycle calculations. If the Q_{eff} of Eu^{152} (89n) is taken to be 1.50 Mev, that of Sm^{152} (90n) is found to be about 0.23 Mev, while from Fig. 5 one would predict a Q_{eff} of 0.75 Mev. A similar instance exists in the cycle involving the Q_{eff}'s of Gd^{153} (89n) and Eu^{153} (90n). There the calculated Q_{eff} for Eu^{153} is 0.69 Mev while the estimated one is 1.20 Mev. One notes that using the 1.20 Mev Q_{eff} for Eu^{153} that for Sm^{153} is calculated to be 0.41 Mev, a value in good agreement with that of 0.45 Mev obtained from the samarium line. It therefore seems that the discrepancy occurs only when the two isotopes involved in the cycle happen to have 89 and 90 neutrons, respectively. This would indicate that the sharp decrease takes place between 89 and 90 neutrons and that the lines past 90 neutrons continue approximately parallel to the original slope. Since only differences between Q_{eff}'s are used, it does not matter if both of the alpha-decay energies used in a calculation have been decreased from their original values (estimated from Fig. 5) by approximately the same amount. With the available data these seem to be the only such checks on our proposal of a discontinuity.

Examining the alpha-energy trends with atomic number for 84-neutron nuclei in Fig. 5, one notes that the large break between gadolinium (64p) and terbium (65p) is outstanding, suggesting a proton subshell at 64. From the differences in alpha energies of the 84-neutron nuclei it would seem that the 65^{th} proton is less tightly bound than the general average in the region. The "break" was noted for the first time by Rasmussen, Thompson and Ghiorso[6] who proposed that a splitting between the $2d_{5/2}$ and $2d_{3/2}$ levels in the Mayer nuclear shell model could account for a slight extra stabilization of the 64-proton configuration. The single-particle states for odd-Z nuclei in the region $50 < Z < 82$ come in the following order: $g_{7/2}$, $d_{5/2}$, $h_{11/2}$, $d_{3/2}$ and $s_{1/2}$.[30] There does occur a substantial splitting between the $d_{5/2}$ and $h_{11/2}$ states which might account for a proton subshell at Z equal to 64.

In summary then, (1) the curves in Fig. 5 can be used with fair accuracy in obtaining extrapolated and interpolated alpha-decay energies at least up to 89 neutrons, (2) good evidence exists that 83- and 82-neutron isotopes have alpha-decay energies much less than those of 84-neutron nuclides for a given element, (3) some evidence is available indicating a sharp decrease in the normal alpha-decay energy versus neutron number trend in going from 89 to 90 neutrons, and (4) a proton subshell may exist at 64 protons. Table II lists beta decay energies calculated using closed cycles.

IV. ALPHA-DECAY RATE CORRELATIONS

Half-lives for alpha-decay transitions proceeding from even-even nuclei to ground levels in the daughter nuclei in the heavy element region have been found to agree well with those calculated from simple Coulombic barrier penetration theory.[31] Most decay rate correlations have been made using a semi-empirical treatment rather than employing equations that arise from the penetration theory. The treatment is based on the fact that if experimental alpha half-lives for even-even ground state transitions are plotted against $Q_{eff}^{-1/2}$ then a series of parallel straight lines are obtained, one for each element. The equation of these lines may be represented as:

$$\log t_{1/2} = \frac{A}{(Q_{eff})^{1/2}} + B.$$

Table II

Beta decay energies calculated using closed cycles

Cycle series	Isotope	Beta decay energy (Mev)	Previously known beta decay energy (Mev)
4n	Tb^{152}	> 3.52 (E.C.)	
	Eu^{148}	> 2.55 (E.C.)	> 0.56
	Pm^{148}	0.45 (E.C.)	
4n+1	Dy^{153}	1.97 (E.C.)	
	Tb^{153}	1.99 (E.C.)	
	Gd^{149}	1.28 (E.C.)	> 0.50
	Eu^{149}	0.89 (E.C.)	> 0.57
	Pm^{149}	0.90 (β-)	1.34
	Pr^{145}	1.70 (β-)	≥ 1.70
4n+2	Tb^{154}	0.16 (β-)	
	Tb^{154}	2.94 (E.C.)	
	Dy^{150}	2.65 (E.C.)	
	Tb^{150}	4.35 (E.C.)	
	Eu^{150}	1.89 (E.C.)	
	Pm^{150}	2.49 (β-)	5.0
	Pm^{146}	1.47 (β-)	> 0.7
	Pm^{146}	1.14 (E.C.)	
	Pr^{146}	3.39 (β-)	4.2
	Pm^{142}	4.24 (E.C.)	
	Pr^{142}	0.44 (E.C.)	
	La^{142}	4.24 (β-)	> 2.5
4n+3	Dy^{151}	3.16 (E.C.)	
	Tb^{151}	> 1.37 (E.C.)	
	Gd^{151}	0.38 (E.C.)	
	Gd^{147}	2.52 (E.C.)	> 0.63

The constants A and B are evaluated for even Z elements from the known experimental data, and a least squares fit to the values for A and B are obtained yielding equations for A and B, respectively, valid in the heavy element region. In this way similar lines are then drawn for odd Z elements. Transitions other than ground state transitions from even-even nuclei exhibit rates that are slower than those predicted from simple theory,[31] that is, those calculated from the above equation using the appropriate constants A and B. The slower transitions include in addition to those proceeding from odd nucleon nuclei the non-ground-state transitions of even-even nuclei. Hindrance factors are then defined for the slower transitions, taking these to be the ratios of the experimental half-lives to the half-lives calculated from the above equation.

A similar treatment in the rare earth region is not possible due to the lack of accurately known alpha half-lives. Instead we have calculated half-lives using equations directly from the barrier penetration theory and compared them with the experimentally determined ones. In the simple theory the barrier is pictured as a potential made up of a Coulombic potential, $V = \frac{2Ze^2}{r}$, with a sharp cut-off at a certain radius termed the effective nuclear radius "R". The alpha particle confined within the barrier makes frequent collisions with the wall and has a probability for penetrating the barrier. The decay constant $\lambda = fP$, where "f" is the frequency factor expressed as collisions per second with the wall, and "P" is the quantum-mechanical penetrability factor. From the WKB approximation the penetration factor is

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

where Z is the charge of the recoil nucleus and M is the reduced mass of the system. The integral has been evaluated analytically and using Bethe's notation[32] $P \approx \exp[-2g(Z,R)\,\gamma(x)]$ where $x = E/B$, B being the barrier $(\frac{2Ze^2}{R})$, $g(Z,R) = \frac{2e}{\hbar}\sqrt{MZR}$, and $\gamma(x) = x^{-1/2} \arccos(x^{1/2}) - (1-x)^{1/2}$. For the pre-exponential factor, "f", different expressions have been used by various authors. Rasmussen, Thompson and Ghiorso calculated alpha half-lives in the rare earth region using 5 different formulae for "f" and found that the calculated half-lives for particular isotopes differed very little.[6] The formulae used in the calculations presumably give the same rate dependence on decay energy. For our purposes we have chosen formula 3 in that paper which is an approximation to the decay rate formula derived by Preston.[33] The expression as derived by Preston is,

$$\lambda = \frac{2^{3/2}}{M^{1/2}R} \frac{(E-U)(B-E)^{1/2}}{(E-U)+(B-E)} P \quad (1)$$

where U is the internal potential energy of the alpha particle within the nucleus. Kaplan in his correlation of even-even alpha-emitter data[34] also used the formula and was able to show from his correlations that $E-U \approx \frac{\pi^2\hbar^2}{2MR^2}$ (11). For the heavy element alpha-emitters correlated by Kaplan E-U is about 0.52 Mev, small enough to be neglected with respect to (B-E) in the denominator of the expressed for "λ" (1). Substituting for E-U from (11), expression (1) becomes,

$$\lambda = \frac{2^{1/2}\pi^2\hbar^2}{M^{3/2}R^3(B-E)^{1/2}} P \qquad (111)$$

This then is the formula used in our calculations.

Before "λ" can be determined the effective nuclear radius, R, must be known. Using the decay rate data for Gd^{148} and Ra^{224} the R values (for the recoil nuclei Sm^{144} and Em^{220}) in the two regions were found by Rasmussen et al. to be 8.00 and 9.25 fermis respectively.[6] From these R values the constants "a" and "b" used in an effective nuclear radius formula of the type $R = (aA^{1/3} + b) \times 10^{-13}$ cm were determined. "a" and "b" were found to be 1.58 and -0.30 respectively. We have calculated the alpha half-lives of the isotopes listed in Table I using the above radius formula and expression (111) for the decay constant "λ". Table III compares the experimental half-lives with the theoretical ones. A plot of the log of the half-life versus the inverse of the square root of the decay energy is shown in Fig. 6. Both the theoretical and the experimental half-lives have been included in the diagram. As mentioned previously the points for a given element in such a plot for even-even nuclei fall on a straight line. This can be readily seen if the function γ (x) is expanded in a Taylor series about x = 0. Then the expression for P becomes,

$$\log P = \frac{(2MBR)^{1/2}}{\hbar} \left[\frac{\pi B^{1/2}}{E^{1/2}} - 4 + \frac{E}{B} - \frac{3E^2}{4B^2} + \ldots\ldots \right]$$

where it is indeed observed that the main energy dependence of log P is an inverse square root dependence.

Bearing in mind the sensitive dependence of half-lives on decay energies let us consider the results of Table III (a). Excepting for Gd^{150} the theoretical and experimental half-lives of all the even-even isotopes are in agreement within a factor of 5. Deviations of such magnitudes are not surprising if errors involved in the determinations of experimental half-lives and decay energies are considered. A change of 70 kev in a Q_{eff} is sufficient to account

Table III

Comparison of theoretical and experimental alpha half-lives

Isotope	Q_{eff} (Mev)	Theor. half-life	Exper. half-life	Ratio (Exp./Theor.)
(a) Dy^{153}	3.59	9.57 yr	13.4 yr	1.4
Dy^{152}	3.78	0.38 yr	1.45 yr	3.8
Tb^{151}	3.56	4.08 yr	$5.8x10^{2}$ yr	$1.4x10^{2}$
Tb^{149}	4.08	8.4 hr	36 hr	4.3
Gd^{152}	2.23	$2.11x10^{14}$ yr	$1.13x10^{14}$ yr	$(1.9)^{-1}$
Gd^{150}	2.80	$8.37x10^{6}$ yr	$3x10^{5}$ yr	$(2.8x10^{1})^{-1}$
Gd^{149}	3.10	$6.92x10^{3}$ yr	$4x10^{3}$ yr	$(1.7)^{-1}$
Eu^{147}	2.98	$2.09x10^{4}$ yr	$5x10^{3}$ yr	$(4.2)^{-1}$
Eu^{147}	3.06	$4.06x10^{3}$ yr	$5x10^{3}$ yr	1.2
Sm^{147}	2.26	$1.63x10^{12}$ yr	$1.3x10^{11}$ yr	$(1.25x10^{1})^{-1}$
Sm^{147}	2.32	$2.00x10^{11}$ yr	$1.18x10^{11}$ yr	$(1.7)^{-1}$
Sm^{146}	2.64	$1.78x10^{7}$ yr	$5x10^{7}$ yr	2.8
Nd^{144}	1.97	$1.37x10^{15}$ yr	$1.5x10^{15}$ yr	1.1
Nd^{144}	1.92	$1.14x10^{16}$ yr	$2.36x10^{15}$ yr	$(4.8)^{-1}$
(b) Dy^{156}	2.60	$4.40x10^{10}$ yr	$>10^{18}$ yr	
Dy^{156}	2.20	$1.16x10^{16}$ yr	$>10^{18}$ yr	
Dy^{154}	3.46	$1.04x10^{2}$ yr		
Dy^{151}	4.20	6.2 hr	>19 min	
Dy^{150}	4.35	53 min	> 7 min	
Tb^{152}	3.20	$3.95x10^{3}$ yr	$>2.2x10^{4}$ yr	

for a difference of a factor of 10 in the calculated half-life. The discrepancy in the Gd^{150} case is much greater (a factor of 28). Here again the deviation is probably due to errors in the half-life and decay energy measurements. The decay energy is correct to only ±0.15 Mev while the experimental half-life is accurate to only a factor of 2.

From heavy element data it is known that odd-nucleon alpha emitters have half-lives longer than those calculated from theory. For Dy^{153} and Gd^{149} the theoretical and experimental half-lives do not deviate by large amounts. Errors in the experimental data preclude any conclusions as to whether or not the discrepancies are significant. The Eu^{147} decay energy is based on the experimental Sm^{147} value. Using the two older decay energies of Eu^{147} and Sm^{147} the half-lives calculated are found to be greater than the experimental ones. The deviation is particularly large for Sm^{147}. A more recent measurement of the Sm^{147} decay energy has been made by Macfarlane.[14] (The remeasurement affects some of the artificial alpha emitters where Sm^{147} was used as a standard.) This number together with the consequent new Eu^{147} decay energy yields theoretical half-lives in essential agreement with the experimental ones. Larger deviations for all four of the above odd-nucleon isotopes are not excluded by the experimental data. It is not possible to state with certainty if any of the four nuclides exhibit hindered alpha decay rates. According to Table III (a) the 2 remaining odd-nucleon isotopes, Tb^{149} and Tb^{151}, seem to have hindered decays. Taking into consideration the experimental limits of error the two half-lives still appear to be greater than the calculated ones. Thus the alpha decay rates of Tb^{149} and Tb^{151} seem to be indeed hindered.

In Table III, part (b), half-lives have been calculated for Dy^{151}, Dy^{150} and Tb^{152} and are compared with the experimental half-life limits for the same nuclides. The experimental values for Dy^{151} and Dy^{150} have been taken as being greater than the total half-lives of these isotopes. The Tb^{152} limit is much more significant since it indicates that the half-life is greater than the theoretically calculated one showing that the odd-nucleon isotope has a hindered alpha-decay rate. The Tb^{152} half-life limit has been determined from an α/E.C. branching ratio limit of $< 10^{-7}$ set by Toth, Faler, and Rasmussen.[35] Theoretical alpha-decay rates have been calculated for Dy^{156} using two decay energies, 2.6 and 2.2 Mev. The first is taken from the extrapolated dysprosium line in Fig. 5, while the second is the upper limit set by Riezler and Kauw.[12] Their experimental alpha half-life limit of $> 10^{18}$ years for Dy^{156} shows that even 2.2 Mev for the Q_{eff} is still somewhat large. Thus there seems to be indeed a sharp drop from the expected decay energy of approximately 0.5 Mev at 90 neutrons at least for the dysprosium alpha emitters. The calculated Dy^{154} half-life has been included for comparison.

The Coulomb potential with a sharp cut-off at some effective nuclear radius is not a realistic nuclear potential. Hoping to obtain a more significant treatment of alpha decay data, Rasmussen has calculated barrier penetrabilities for even-even and odd-nucleon alpha emitters using a potential derived from alpha-scattering information.[36,37] The potential used was deduced by Igo from optical-model analyses of alpha-scattering data.[38] The V(r) consists of an exponential expression for the real part of the nuclear potential valid in the surface region for $|V| \lesssim 10$ Mev.

$$V(r) = -1100 \exp \left\{ - \left[\frac{r - 1.17\, A^{1/3}}{0.574} \right] \right\} \text{Mev},$$

where "r" is in fermis and "A" is the mass number. Rasmussen has calculated the penetration factors "P" using the above V(r) and then determined the reduced alpha emission widths "δ^2" from the expression: $\lambda = \frac{\delta^2 P}{h}$, where "$\lambda$" is the decay constant and "h" is Planck's constant. Hindrance factors "F" were then determined for odd-nucleon isotopes as well as for non-ground-state transitions in even-even nuclides. "F" for odd-nucleon isotopes was defined as:

$F = \frac{\delta_1^2 + \delta_2^2}{2\delta_{odd}^2}$, where "$\delta_1^2$" and "$\delta_2^2$" are reduced widths for ground-state transitions of nearest or next nearest even-even nuclei and "δ_{odd}^2" is the reduced width for the odd-nucleon isotope in question. We have taken the "δ^2" values for Sm^{146}, Gd^{148}, Tb^{149} and Tb^{151} from the two publications of Rasmussen.[36,37] In addition, employing the same IBM-650 program the "δ^2" values for Nd^{144}, Sm^{147}, Eu^{147} (using the newer experimental data for these three isotopes[14]), Gd^{150}, Gd^{152}, Dy^{152} and Dy^{153} have been calculated. The information together with hindrance factors for odd-nucleon isotopes is summarized in Table IV. Also shown in the table are the even-even isotopes used to determine "F" for any given odd-nucleon nuclide.

The reduced widths in principle should be constant for even-even (ground-state transitions) and favored alpha decay. The experimental data for Dy^{152} and Dy^{153} are so uncertain that it is difficult to state whether or not the Dy^{153} decay rate is indeed favored. The Tb^{149}, Tb^{151} and Eu^{147} decay rates are seen to be hindered even taking into account experimental limits of error. Gd^{149} is presumably favored in its alpha decay. Here the data are

Table IV

Reduced widths and hindrance factors

Nuclide	$(E_{\alpha}+E_{sc})^{*}$	Exper. $T_{1/2}\,\alpha$ (sec)	Reduced width δ^2(Mev)	Hindrance factor F	Even-even nuclides used to calculate F
Nd^{144}	1.86	7.45×10^{22}	0.223		
Sm^{146}	2.57	1.58×10^{15}	0.0152		
Sm^{147}	2.26	3.72×10^{18}	0.0750	2.03×10^{-1}	Sm^{146}
Eu^{147}	3.00	2.07×10^{11}	0.0167	3.01	Sm^{146},Gd^{148}
Gd^{148}	3.18	4.47×10^{9}	0.0852		
Gd^{149}	3.02	1.15×10^{11}	0.0954	8.20×10^{-1}	Gd^{148},Gd^{150}
Gd^{150}	2.72	2.08×10^{14}	0.0713		
Gd^{152}	2.17	3.56×10^{11}	0.0952		
Tb^{149}	3.97	1.48×10^{5}	0.0115	6.83	Gd^{148},Gd^{150}
Tb^{151}	3.42	2.40×10^{10}	6.13×10^{-4}	1.36×10^{2}	Gd^{150},Gd^{152}
Dy^{152}	3.68	1.55×10^{7}	0.0425		
Dy^{153}	3.50	1.93×10^{8}	0.0785	5.41×10^{-1}	Dy^{152}

*Does not include alpha particle recoil energy.

again uncertain enough to make any conclusion very doubtful. Sm^{147} seems to exhibit a favored alpha decay rate. This is dependent on the "δ^2" of Sm^{146} which seems to be a little low compared to the reduced widths of the other even-even nuclides. However, even if its "δ^2" is increased to a value closer in magnitude to that of the other even-even reduced widths the Sm^{147} decay rate would still tend to be favored. The Sm^{147} decay energy and experimental half-life are known sufficiently well so that its calculated "δ^2" must be quite accurate. The spins of Sm^{147} [39,40] and of its alpha-decay daughter, Nd^{143}, [41,42] have been both found to be 7/2. The wave functions of the odd nucleon in parent and daughter should be very similar thus leading to a large overlap integral between initial and final odd-nucleon wave functions. Perhaps this explanation can account for the enhanced Sm^{147} alpha-decay rate.

V. CONCLUSION

Before our knowledge of alpha systematics in the 82-neutron region can be extended any further, more data are certainly needed. Some of the already known alpha half-lives will have to be redetermined more accurately. A number of alpha emitters are known whose partial alpha half-lives have not yet been determined. New alpha emitters will have to be discovered and their decay energies and half-lives found. It is certain that alpha emitters, undiscovered as yet, do exist with measurable half-lives. Unknown alpha emitters above dysprosium must exist as indicated by the holmium alpha activity reported by Rasmussen et al.[6] and also by the following experiment carried out by the authors. Natural dysprosium containing 0.05% Dy^{156} and 0.09% Dy^{158} was bombarded with 48-Mev helium ions. Presumably erbium isotopes were produced with mass numbers 156 or greater, since (α,5n) reactions are not energetically possible at the bombarding energy, at least not with any appreciable cross sections. Chemical separations were not attempted. The samples were counted in alpha counters, and half-lives of reasonable duration (hours) were observed. These alpha emitters were either erbium or holmium isotopes, but were definitely not dysprosium nuclides, since the dysprosium daughters would have mass numbers 156 or greater and these are known not to emit alpha radiation.

REFERENCES

1. G. Hevesy and M. Pahl, Nature 130, 846 (1932).
2. G. Gamow, Z. Physik 52, 510 (1929).
3. E. U. Condon and R. W. Gurney, Phys. Rev. 33, 127 (1929).
4. T. P. Kohman, Phys. Rev. 76, 448 (1949).
5. Thompson, Ghiorso, Rasmussen, and Seaborg, Phys. Rev. 76, 1406 (1949).
6. Rasmussen, Thompson, and Ghiorso, Phys. Rev. 89, 33 (1953).
7. D. C. Dunlavey and G. T. Seaborg, Phys. Rev. 92, 206 (1953).
8. K. S. Toth and J. O. Rasmussen, Phys. Rev. 109, 121 (1958).
9. W. Porschen and W. Riezler, Z. Naturforsch. 9a, 701 (1954).
10. W. Porschen and W. Riezler, Z. Naturforsch. 11a, 143 (1956).
11. W. Riezler and G. Kauw, Z. Naturforsch. 12a, 665 (1957).
12. W Riezler and G. Kauw, Z. Naturforsch. 13a, 904 (1958).
13. W. Riezler and G. Kauw, Z. Naturforsch. 14a, 196 (1959).
14. R. D. Macfarlane, Natural Alpha Radioactivity in Medium-Heavy Elements (Thesis), Department of Chemistry, Carnegie Institute of Technology, NYO-7687, May 1959.
15. Rasmussen, Rollier, and Toth, to be published.
16. K. S. Toth and J. O. Rasmussen, J. Inorg. Nucl. Chem. 10, 198 (1959).
17. L. Winsberg, UCRL-3958, also Bull. Am. Phys. Soc. 3, 406 (1958).
18. W. P. Jesse and J. Sadauskis, Phys. Rev. 78, 1 (1950).
19. G. Beard and M. L. Wiedenbeck, Phys. Rev. 95, 1245 (1954).
20. E. C. Waldron, V. A. Schultz, and T. P. Kohman, Phys. Rev. 93, 254 (1954).
21. Ghiorso, Thompson, Higgins, Harvey and Seaborg, Phys. Rev. 95, 293 (1954).
22. Glass, Thompson, and Seaborg, J. Inorg. Nucl. Chem. 1, 3 (1955), and

22. B. M. Foreman and G. T. Seaborg, J. Inorg. Nucl. Chem. 7, 305 (1958).

23. R. W. King, Revs. Mod. Phys. 26, 327 (1954).

24. L. J. Lidofsky, Revs. Mod. Phys. 29, 773 (1957).

25. Strominger, Hollander, and Seaborg, Revs. Mod. Phys. 30, 585 (1958).

26. K. S. Toth and O. B. Nielsen, Phys. Rev. 115, 1004 (1959).

27. Gorodinski, Murin, and Pokrovsky, Izvestia Akad. Nauk S.S.S.R., Ser. Fiz. 22, 811 (1958).

28. L. C. Schmid and S. B. Burson, Phys. Rev. 115, 178 (1959).

29. L. C. Schmid and S. B. Burson, Phys. Rev. 115, 447 (1959).

30. B. R. Mottelson and S. G. Nilsson, Mat. Fys. Skr. Dan. Vid. Selsk. 1, No. 8 (1959).

31. I. Perlman and J. O. Rasmussen, Handbuch der Physik, S. Flugge, ed., Vol. 42, p. 143 (Springer-Verlag; Berlin, 1957).

32. H. A. Bethe, Revs. Mod. Phys. 9, 69 (1937).

33. M. A. Preston, Phys. Rev. 71, 865 (1947).

34. I. Kaplan, Phys. Rev. 81, 962 (1951).

35. Toth, Faler, and Rasmussen, Phys. Rev. 115, 158 (1959).

36. J. O. Rasmussen, Phys. Rev. 113, 1593 (1959).

37. J. O. Rasmussen, Phys. Rev., to be published.

38. G. Igo, Phys. Rev. Letters 1, 72 (1958).

39. G. S. Bogle and H. E. D. Scovil, Proc. Phys. Soc. (London) 65A, 360 (1952).

40. K. Murakawa, Phys. Rev. 93, 1232 (1954).

41. B. Bleaney and H. E. D. Scovil, Proc. Phys. Soc. (London) 63A, 1369 (1950).

42. K. Murakawa and J. S. Ross, Phys. Rev. 82, 967 (1951).

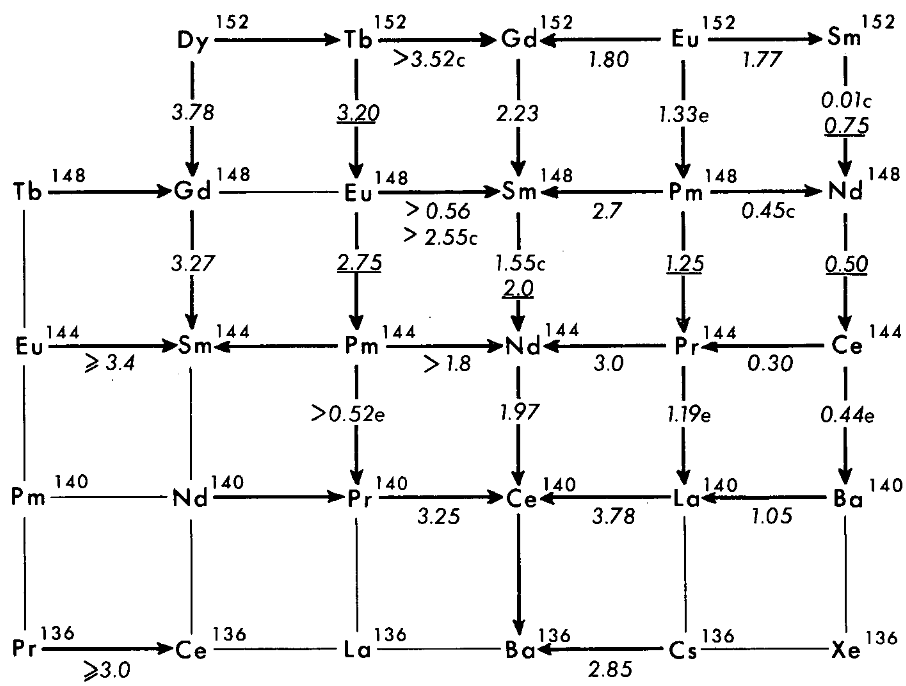

Fig. 1. Closed-decay cycle "4n".
number: indicates an experimental value.
<u>number</u>: indicates a value estimated from Fig. 5.
number e: indicates a value calculated using only experimental energies.
number c: indicates a value calculated using energies estimated from Fig. 5.

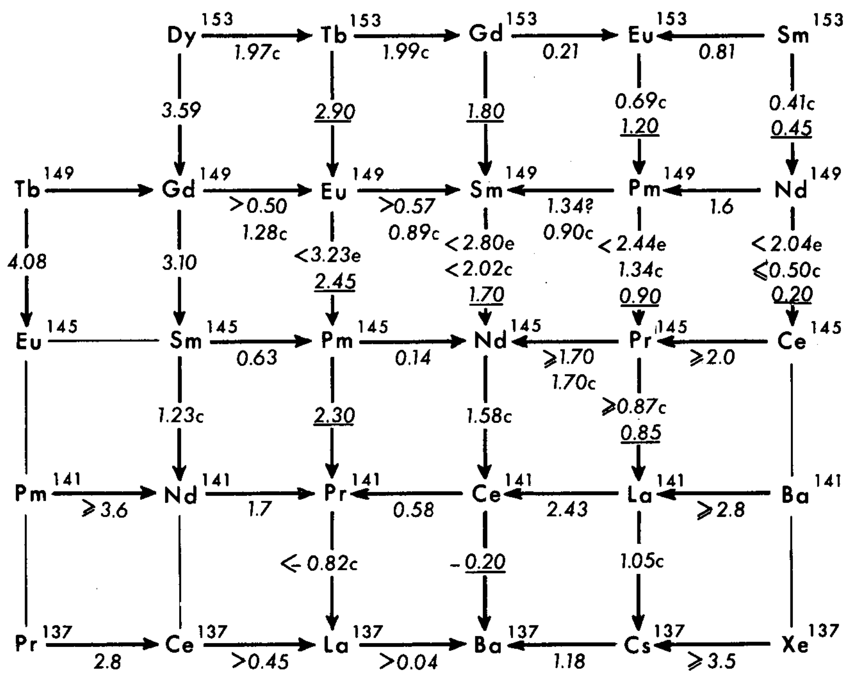

Fig. 2. Closed cycle "4n+1".
number: indicates an experimental value.
<u>number</u>: indicates a value estimated from Fig. 5.
number e: indicates a value calculated using only experimental energies.
number c: indicates a value calculated using energies estimated from Fig. 5.

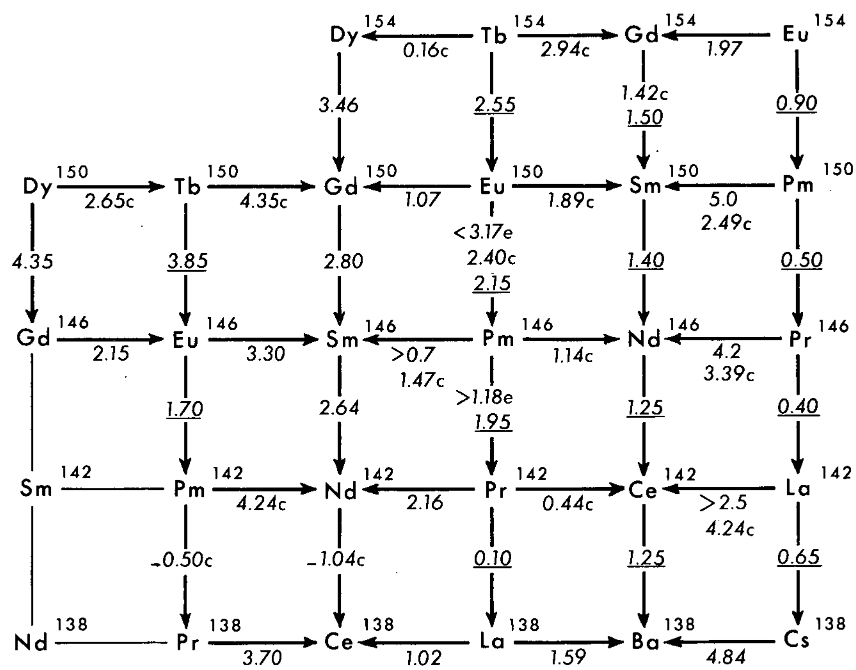

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Fig. 3. Closed-decay cycle "4n+2".
number: indicates an experimental value.
<u>number</u>: indicates a value estimated from Fig. 5.
number e: indicates a value calculated using only experimental energies.
number c: indicates a value calculated using energies estimated from Fig. 5.

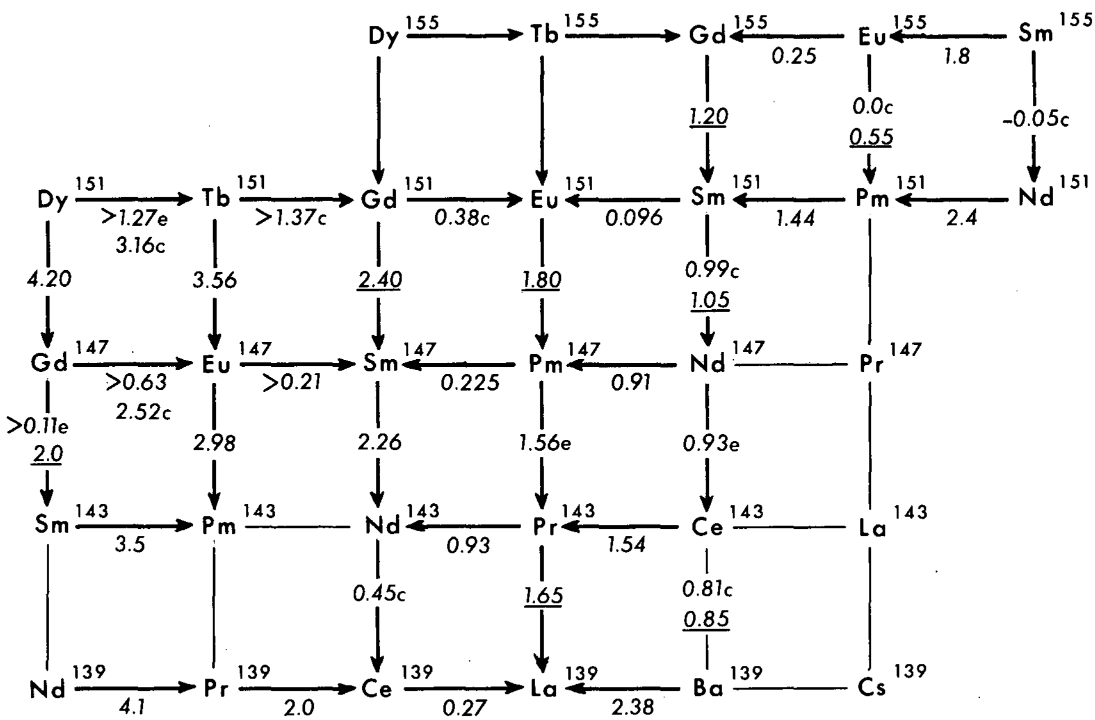

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Fig. 4. Closed-decay cycle "4n+3".
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<u>number</u>: indicates a value estimated from Fig. 5.
number e: indicates a value calculated using only experimental energies.
number c: indicates a value calculated using energies estimated from Fig. 5.

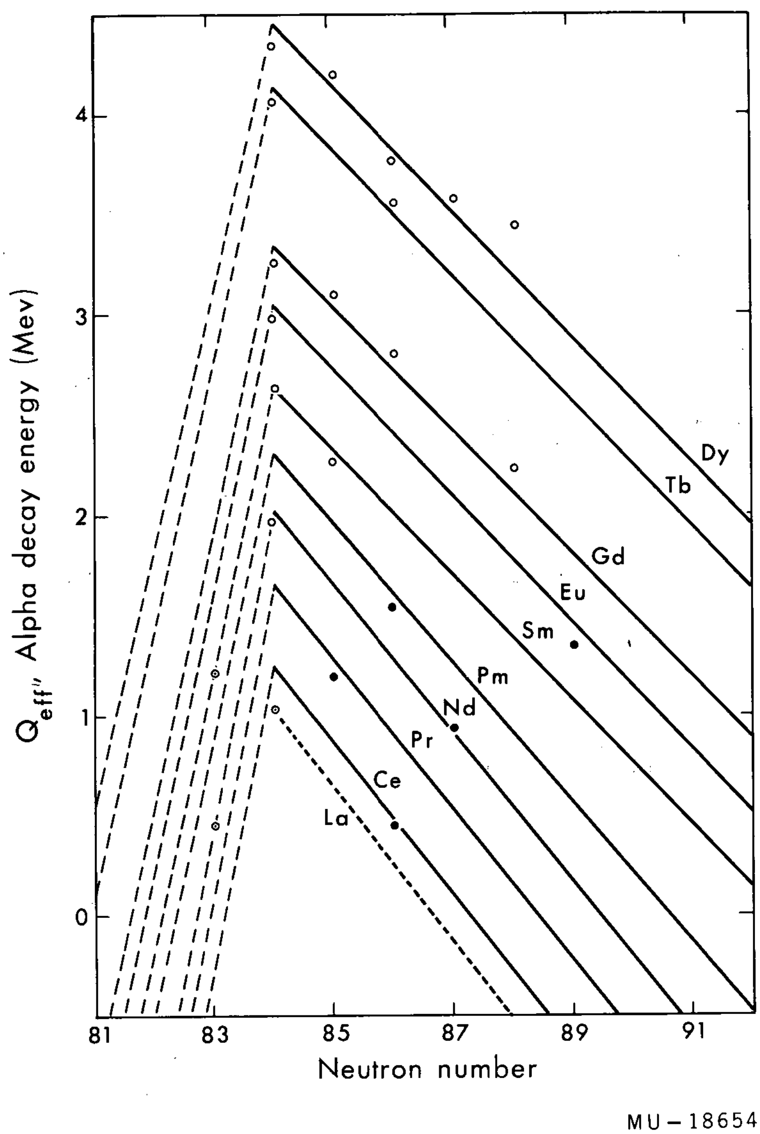

Fig. 5. Plot of Q_{eff} vs. neutron number.
o: indicates an experimental decay energy.
●: indicates a decay energy calculated from closed-decay cycles using only experimentally determined decay energies.

The Nd^{143} and Sm^{145} points have been calculated from decay cycles using Pr^{143} and Pm^{145} energies estimated from the full lines in the figure. Dashed lines have been drawn for all the elements shown in Fig. 5, with the Nd^{143} and Sm^{145} points serving as guides, to show the discontinuity occurring at 84 neutrons. The lanthanum line is drawn parallel to the cerium line and through the La^{141} point calculated from a closed-decay cycle.

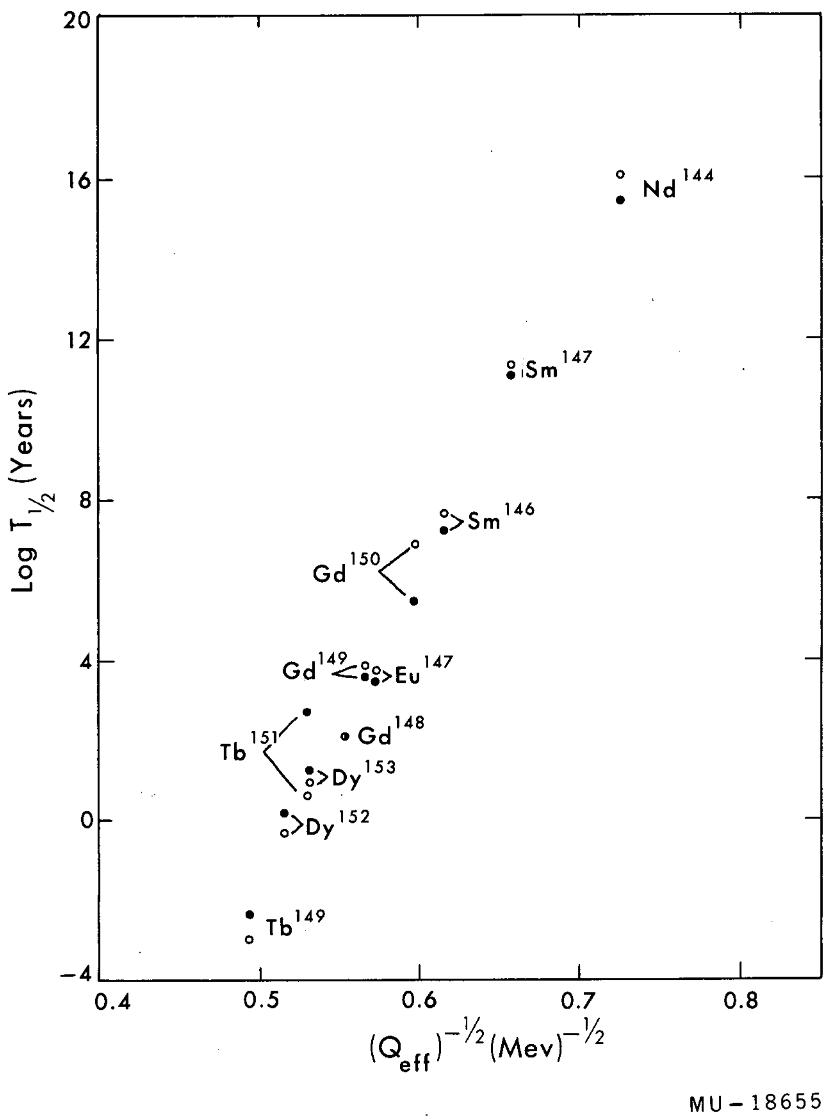

Fig. 6. Plot of $T_{1/2}$ vs. $(Q_{eff})^{-1/2}$

●: indicates an experimental half-life.
o: indicates a calculated half-life.