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CONSISTENCY OF NUCLEAR RADII OF EVEN-EVEN NUCLEI FROM ALPHA-DECAY THEORY

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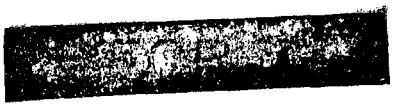

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I. Perlman and T. J. Ypsilantis

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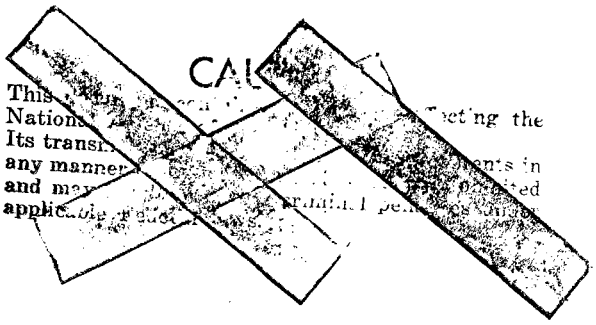

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CONSISTENCY OF NUCLEAR RADII OF EVEN-EVEN NUCLEI FROM ALPHA-DECAY THEORY

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February 27, 1950

ABSTRACT

It is shown possible to obtain a consistent function for nuclear radius if the quantitative treatment of the alpha-decay process is applied to even-even isotopes of the heavy elements. The nuclear radii so calculated for even-even isotopes of emanation, radium, thorium, uranium, plutonium, and curium, conform with the expression $r = 1.48 \cdot 10^{-13} \cdot A^{1/3}$ cm which defines the normal nuclear radii. The agreement is within 1 percent for the majority of the cases and there is reason to question the experimental data used in the calculations for at least part of those which do not show such close agreement. In cases of fine structure in alpha-decay of even-even nuclei, the radii calculated from the separate alpha-groups are compatible. The polonium isotopes and Em^{212} form a special group showing departures from normal nuclear radii explainable by consideration of shells in nuclear structure.

CONSISTENCY OF NUCLEAR RADII OF EVEN-EVEN NUCLEI FROM ALPHA-DECAY THEORY

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Recently there has been renewed interest in alpha-decay systematics and alpha-decay theory stimulated by the discovery within the past few years of many new alpha-emitters. In a recent communication[1] from this laboratory regularities in alpha-decay energies and half-lives were dealt with at some length and the correlation observed which is pertinent to the present paper is that only the even-even nuclei give evidence of quantitative agreement with present alpha-decay theory. The observation of significance was that on a decay energy vs. half-life plot even isotopes of an even element define a line which is close to the curve which is calculated by selecting a reasonable function for nuclear radius. This type of plot will be presented and discussed in the last part of this paper. The nuclear radius is the single parameter in the equation which cannot be determined experimentally with the desired precision. While the even-even nuclei showed this consistency, the nuclei with odd nucleons almost invariably departed from these curves in the direction which corresponds to prohibition of alpha-decay, that is, the half-lives were too long for the particular decay energies. The reasons for prohibition of alpha-decay in such cases have been discussed[1] and the phenomenon is mentioned here in order to dismiss these categories of nuclear type in attempting to obtain a consistent function for nuclear radius from alpha-decay theory.

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[1] I. Perlman, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 77, 26 (1950).

The present communication treats the even-even nuclides to determine their nuclear radii assuming the validity of the one-body theory for alpha-decay. The significance of the nuclear radius in alpha-decay is in determining the height and breadth of the potential barrier for a given nuclear charge. In the one-body theory, the alpha-particle being emitted is considered to be acted upon by the potential field of the product nucleus. Accordingly, the nuclear charge, barrier height, and radius employed are those of the product after alpha-emission. From the considerations of this problem to be presented here, we shall see that in most cases the simplifying assumptions inherent in the one-body theory probably do not produce inconsistencies in the calculated nuclear radii although the absolute values obtained may vary somewhat with different treatments. This follows if most parent-daughter pairs have about the same nuclear radius which indeed does seem to be the case for a wide group which we shall define as normal nuclei. The meaning of the calculated nuclear radius becomes less distinct where there is a large change in nuclear radius between parent and daughter, since in such a case the contribution of the parent nucleus is tacitly ignored and any effect it may have is attributed to the daughter. More basically, it should be realized that nuclear radii calculated from alpha-decay theory will depend upon the nuclear model employed and may be expected to differ from those obtained from other phenomena related to a different model or for which the calculated radii are not sensitive to the model selected. The objective of the present paper is to test the consistency of nuclear radii as calculated from alpha-decay data of even-even nuclei and it is probable that different treatments will be equivalent in this regard even though

the absolute values of the radii may differ somewhat. In the present study the expression used was selected because of its relative simplicity and relates the decay constant, decay energy, atomic number, and nuclear radius as follows (see Bethe[2]):

$$\log_{10}\lambda = 21.843 + 1/2 \log E + \log r + 0.217 \frac{4}{A-4} - 1.104 \frac{(Z-2)}{\left\{E\left(1+\frac{4}{A+4}\right)\right\}^{1/2}} \alpha_o + 1.104 \frac{(Z-2)}{\left\{E\left(1+\frac{4}{A-4}\right)\right\}^{1/2}} \sin \alpha_o \cos \alpha_o$$

$$\cos \alpha_o = 0.5893 \left(\frac{Er}{Z-2}\right)^{1/2}$$

The numerical coefficients were calculated to give λ in units of sec.^{-1} with E (Mev) the alpha-decay energy, r (cm.) the radius of the product nucleus, while Z and A are the charge and mass number, respectively, of the emitting nucleus. The principal approximation in this expression involves taking the coefficient, K, in the formula $\lambda = Ke^{-2C}$ simply as v/r, where v is the relative velocity of the alpha-particle. Biswas[3] has tested alpha-decay data using a similar expression and Preston[4] has developed a more rigorous expression for alpha-decay which he and Biswas and Patro[5] have used in calculating nuclear radii. The values obtained from Preston's expression appear to be a few percent larger than those from the approximate solution which we employ. Of more significance, these authors and others make no distinction between different nuclear types so that a nuclear radius so calculated is an "effective nuclear radius" which appears abnormally small when alpha-decay is forbidden for any reason.

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[2] H. A. Bethe, Revs. Mod. Phys. 9, 69 (1937), p. 161.

[3] S. Biswas, Ind. J. Phys. 23, 51 (1949).

[4] M. A. Preston, Phys. Rev. 71, 865 (1947).

[5] S. Biswas and A. P. Patro, Ind. J. Phys. 22, 539 (1948).

In the present paper, calculations of nuclear radius were made for 25 even-even nuclides and of these, three have more than one recognized alpha-group for each of which separate computations were made. With a few exceptions, which may be due to inaccuracies in available data, the nuclear radii from emanation (element 86) to curium (element 96) show remarkably good agreement with the simple expression $r = 1.48A^{1/3} \times 10^{-13}$ cm. For convenience we shall call the nuclear radius determined from this relationship the normal nuclear radius. Polonium isotopes show unmistakably lower values for nuclear radius as will be discussed below.

The selection of 1.48 as the coefficient for nuclear radius was not arrived at by statistical analysis weighting all of the available data equally as will be apparent by examining Table I in which it will be seen that the deviations to be discussed below are distributed heavily on one side. It will be seen that the principal uncertainties in the calculation of nuclear radii insofar as experimental data are concerned are inaccuracies in alpha-energies. As a result greater weight was given to the relatively few determinations made by alpha-ray spectroscopy, especially since for many of the other nuclei there is reason to suspect inaccuracies, and for some it is known that the probable error is in the right direction.

Table I shows the even-even nuclides from emanation to curium with the alpha-energies and half-lives used in calculating the nuclear radii. The last two columns list the calculated nuclear radii and the deviations of these radii from the normal values (given from the expression $r = 1.48A^{1/3} \times 10^{-13}$ cm). In examining the results, one should first of all consider the possible errors in the two experimentally determined parameters, the decay constant, and decay energy. The calculated value for the nuclear radius is fairly insensitive to variation in decay constant,

TABLE I. Calculated nuclear radii of even-even nuclides.

Nuclide	α-energy	Abundance of alpha-group	Half-life	Nuclear radius* r	% Deviation of r from normal
Cm^{242}	6.18		150 days	9.14	- 0.4
Cm^{240}	6.37		30 days	9.02	- 1.4
Pu^{238}	5.59		92 yrs.	9.06	- 0.7
Pu^{236}	5.85		2.7 yrs.	9.10	0.0
U^{238}	4.25		4.51×10^{9} yrs.	9.41	+ 3.2
U^{234}	4.84		2.35×10^{5} yrs.	9.22	+ 1.7
U^{232}	5.40		70 yrs.	9.14	+ 1.1
U^{230}	5.96		20.8 days	9.15	+ 1.5
Th^{232}	4.05		1.39×10^{10} yrs.	9.41	+ 4.0
Th^{230}	4.76	0.80	1.0×10^{5} yrs.	9.09	+ 0.9
	4.69	0.20	4.0×10^{5} yrs.	9.04	+ 0.4
Th^{228}	5.52	0.72	2.64 yrs.	8.97	- 0.2
	5.43	0.28	6.79 yrs.	9.03	+ 0.5
Th^{226}	6.41		30.9 min.	9.09	+ 1.4
Ra^{226}	4.88	0.96	1700 yrs.	8.98	+ 0.2
	4.70	0.04	4.0×10^{4} yrs.	8.92	- 0.4
Ra^{224}	5.78		3.64 days	8.97	+ 0.4
Ra^{222}	6.63		38 sec.	8.98	+ 0.9
Em^{222}	5.59		3.83 days	8.95	+ 0.4
Em^{220}	6.39		54.5 sec.	8.94	+ 0.7
Em^{218}	7.25		0.019 sec.	9.03	+ 2.0

*The nuclear radius is that of the α-decay daughter calculated using the measured half-life and α-energy and is expressed in units of 10^{-13} cm.

for example, a 10 percent error in the half-life determination would only change the radius by about 1/4 percent. However, an error of only 0.02 Mev in decay-energy will change the radius by 1 percent, and it may be remarked that the energies of most of the nuclides shown in Table I are possibly in error by this amount and some by more.

It should be pointed out that the details of nuclear binding for this broad region are not sufficiently well known to give an a priori reason why all nuclear radii in this region should not deviate from the normal by more than some arbitrary degree such as 1 percent. Nevertheless, because of the agreement in so many cases, it is worthwhile examining the experimental data for those few cases which deviate by significantly more than 1 percent.

The first such example from Table I is that of U^{238} whose radius calculated from its measured energy and decay constant is 3 percent too high. In making this calculation, the alpha-particle energy selected was 4.18 Mev (decay energy 4.25 Mev) measured by Clark, Spencer-Palmer, and Woodward[6] using an ionization chamber coupled to a pulse height discriminator. To eliminate the 3 percent discrepancy in radius, it would be necessary to increase the particle energy only to 4.23 Mev. It is undoubtedly worth re-examining the alpha-energy for U^{238}, particularly since older values are higher than the one we have chosen; see for example, 4.21 Mev by Schintlmeister and Lintner[7] and 4.23 Mev by Sizoo and Wytzes.[8]

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[6] F. L. Clark, H. J. Spencer-Palmer, and R. N. Woodward, British Atomic Energy Project Report BR 522 (Oct. 1944).

[7] J. Schintlmeister and K. Lintner, Sitzber. Akad. Wiss. Wien, Abt. IIa, 148, 279 (1939).

[8] G. J. Sizoo and S. A. Wytzes, Physica 4, 791 (1937).

The calculated radius for Th^{232} is 4 percent high, and here even more than was the case with U^{238} there may be reason to question the measured alpha-energy before ascribing a real difference from the normal nuclear radius. In arriving at the nuclear radius, the alpha-particle energy selected was 3.98 Mev[9] while values as high as 4.20 Mev[10] have been reported. The value which would be required to eliminate the discrepancy in radius is 4.06 Mev. While the two cases cited represent the greatest differences between calculated and normal nuclear radii, there are others which will probably be changed by redetermination of alpha-energies.

The few examples of alpha-particle fine structure in even-even nuclides are of special interest since the correlations[1] showed that each of the alpha-groups appeared with its partial half-life in conformity with its decay energy. The quantitative agreement may be tested by the calculation of the nuclear radius since that calculated for one group should agree with that calculated from the other and both should agree with the normal relationship for an even-even nucleus. It is found that within experimental uncertainty these conditions do apply to even-even nuclei. Another way of stating this effect is that each alpha-group decays in an allowed manner as though it came from a separate even-even nucleus. In contrast, it has been shown[1] that wherever fine structure appears in nuclei with an odd nucleon, the ground state transition is highly forbidden and shorter range groups become progressively less prohibited.

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[9] F. L. Clark, H. J. Spencer-Palmer, and R. N. Woodward, British Atomic Energy Project Report BR 584 (Mar. 1945).

[10] J. Schintlmeister, Sitzber. Akad. Wiss. Wien, Abt. IIa, 146, 371 (1937).

Table II shows data on three cases of well defined alpha-particle fine structure in even-even nuclei. For ionium the first value for the energy which is listed[11] is that of measurements which did not separately measure the different groups and resulted in something approaching a weighted mean. The next set of data[12] comes from a more recent determination in which the fine structure was determined. The differences in calculated nuclear radii illustrate the extreme sensitivity to alpha-energy and show that in this case, the calculated nuclear radius is in better agreement with the normal using the refined measurements in which fine structure was separated. In the case of radiothorium, the same situation is found as that discussed for ionium.

The third example shown in Table II is that of radium in which uncertainty in experimental data was not in the energy of the short-range group but in its abundance. The energy of the ground state transition is known accurately as 4.877 Mev and the nuclear radius calculated from this is in good agreement with the normal. There is agreement among different investigators that the short-range group has a lower energy by 180-190 Kev than the ground state transition. However, one published measurement of the abundance of this group gives $\sim$ 10 percent,[13] while another gives 1.8 percent resulting respectively in a positive and negative deviation from normal nuclear radius as shown in Table II. Recently, in this laboratory,[14] the abundance of this group has been redetermined as 4.3 percent which can be seen to give good agreement between calculated nuclear radius and the normal.

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[11] H. Geiger, Zeits. f. Physik 8X, 45 (1922).

[12] S. Rosenblum, M. Valadares, and J. Vial, Compt. rend. 227, 1088 (1948).

[13] S. Rosenblum, M. Valadares, and M. Perey, Compt. rend. 228, 385 (1949).

[14] S. Rosenblum, Nucleonics 4, No. 3, 38 (1949).

TABLE II. Nuclear radii in cases of α-decay fine structure.

Nuclide Z^A	Decay energies and references	Abundance of group	Partial alpha half-life	Calc. radius for $(Z-2)^{A-4}$	Deviation from "normal"
Th^{230}(Io)	4.74[11]		8.0×10^4 yrs.	9.23	+ 2.4
	Gp0 4.76[12]	0.80	1.0×10^5 yrs.	9.09	+ 0.9
	GpI 4.69[12]	0.20	4.0×10^5 yrs.	9.04	+ 0.4
Th^{228}(RdTh)	5.48[8,9]		1.90 yrs.	9.21	+ 2.5
	Gp0 5.52[13]	0.72	2.64 yrs.	8.97	- 0.2
	GpI 5.43[13]	0.28	6.79 yrs.	9.03	+ 0.5
Ra^{226}(Ra)	4.88		1622 yrs.	9.01	+ 0.6
	Gp0 4.88[14]	0.96	1700 yrs.	8.98	+ 0.2
	GpI 4.70[14]	0.04[15]	4.0×10^4 yrs.	8.92	- 0.4
	GpI 4.70	0.10[14]	1.6×10^4 yrs.	9.08	+ 1.3
	GpI 4.70	0.018[16]	9.0×10^4 yrs.	8.78	- 2.0

The above examples were selected for discussion because of their classical importance and because some gave calculated radii with the greatest differences from the normal. One could discuss in turn all of the other nuclides shown in Table I in order to examine differences in calculated radii from the normal in terms of uncertainty in experimental data. For such a discussion to be profitable, more precise energy values should be available since many of the values are not known to better than 20-30 Kev and this

[15] A. Ghiorso, private communication.

[16] W. Y. Chang, Phys. Rev. 70, 632 (1946).

uncertainty virtually encompasses the spread of deviations. It may also be noted that certain even-even nuclides such as Pu^{232}, Pu^{234}, and U^{228} were not included in these calculations. This was done principally because of the uncertainty in alpha-decay half-life for these electron capture unstable nuclides.

The even-even polonium isotopes are treated separately in Table III because it appears clear that these belong to a family in which some members have deviations of radii from the normal which are real. It is seen that from Po^{218} to Po^{208} there is a progressive shrinkage in nuclear radius which shows greatest increments between Po^{214} and Po^{212} and between Po^{212} and Po^{210}.

TABLE III. Calculated nuclear radii for polonium isotopes.

Nuclide	α-energy	Half-life	Nuclear radius r	% Deviation of r from normal
Po^{218}	6.11	3.05 min.	8.80	- 0.6
Po^{216}	6.89	0.158 sec.	8.73	- 1.1
Po^{214}	7.83	1.5×10^{-4} sec.	8.67	- 1.5
Po^{212}	8.95	3.0×10^{-7} sec.	8.45	- 3.6
Po^{210}	5.40	138 days	8.04	- 8.0
Po^{208}	5.24	3.0 yrs.	7.90	- 9.3
Em^{212}	6.29	23 min.	8.36	- 4.7

It has been suggested above that in cases in which there may be a discontinuity in nuclear radius between parent and daughter, the meaning of radii calculated by the one-body theory is indistinct. The calculated deviation from the normal may be that of the parent, daughter, or some hybrid depending upon the contribution of each form to the effective

potential barrier. Perhaps some qualitative deductions on the contribution of parent and daughter can be made from an examination of Table III, bearing in mind the discontinuity in nuclear binding which occurs at neutron number 126 and proton number 82. The nucleus which contains both of these numbers is Pb^{208}. It would seem that the effect of 82 protons (in the daughter nucleus) on nuclear stability begins to disappear if there is a large neutron excess since the calculated radii for Po^{218} and Po^{216} show scarcely significant departures from the normal. Either this is so or the daughter nuclear radius is non-operative in determining the potential barrier. For Po^{212} the observed negative departure could be explained by the effect of the daughter nucleus, Pb^{208}, on the potential barrier. The further shrinkage shown for Po^{210} must bring in the contribution of the parent which in this case has 126 neutrons. We might then picture the effective potential barrier as some hybrid dependent upon both the parent and daughter nuclei. The similar large departure from the normal for Po^{208} simply means that a discontinuity in nuclear binding at a closed shell occurs only above the closed shell, and that binding energies of nucleons in the vicinity below are as great or greater as that which completes the shell.

There is one other known even-even nuclide, Em^{212}, in which the neutron number 126 appears and should affect the nuclear radius. The facts of its low energy and measurable half-life alone attest to the correctness of this view as has already been discussed.[1,17] The present calculation shows a negative departure of nuclear radius of 4.7 percent.

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[17] A. Ghiorso, W. W. Meinke, and G. T. Seaborg, Phys. Rev. 76, 1414 (1949).

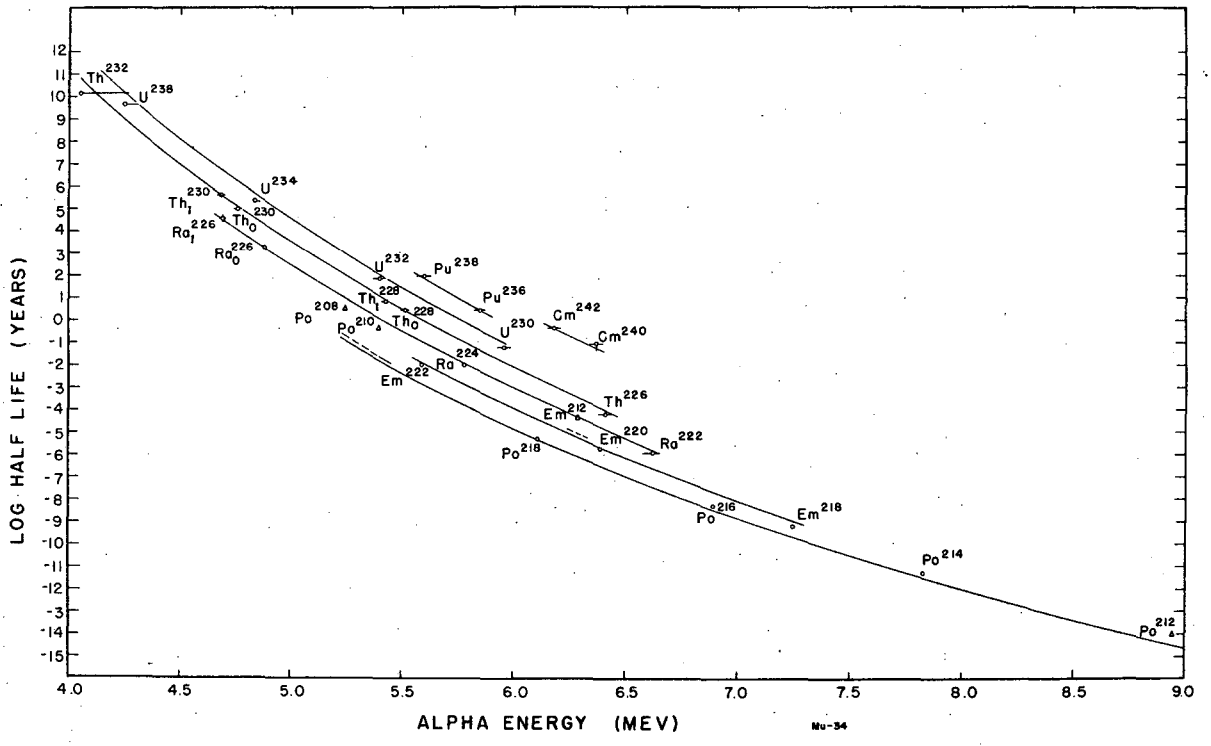
One of the graphical methods for comparing the variables which describe the alpha-decay process consists of a plot of half-life or decay constant against the decay energy in which the variable Z is eliminated as an arbitrary variable by joining points of constant Z.[1,3] This results in a family of curves which for the even-even nuclides are sufficiently regular[1] that it may be deduced that the position of a point is either insensitive to nuclear radius or nuclear radii vary in a regular fashion as has already been assumed. It is possible to justify comparison of such empirical curves with theoretical curves defined by assuming values for the nuclear radius. In defining normal nuclear radii as a simple function of the mass number and realizing that for normal nuclei the decay energy varies regularly with mass number for each element,[1] it is seen that single curves of the type shown in Figure 1 will result.

Figure 1 shows experimental points in comparison with theoretical curves which were constructed in the following manner. For each measured even-even nuclide or alpha-group, the half-life was calculated by using its measured energy and assigning it a normal nuclear radius. Such points define the curves of Fig. 1; the excellent agreement of most of the experimental data indicates the validity of the assumptions and the theory. The agreement in the case of fine structure is worthy of special note. As already mentioned, a point below the curve such as is the case for U^{238} may mean that the measured energy is in error or that its nuclear radius is greater than normal. Its half-life could hardly be sufficiently in error to bring about the observed disagreement. A point above the curve may mean an experimental error in the opposite direction, an abnormally low nuclear radius, or, as is the case for odd nuclei, the alpha-decay may be forbidden for reasons peculiar to such nuclei.

Fig. 1. Calculated curves and experimental points for the half-life vs. decay-energy relationship.

o Alpha-emitters which may have normal nuclear radii.

Δ Alpha-emitters with abnormally small nuclear radii.


LOG HALF LIFE (YEARS)
ALPHA ENERGY (MEV)
Mu-34
Th232
U238
U234
Th1230
Th0230
Ra1226
Ra0226
U232
Pu238
Th1228
Th0228
Pu236
U230
Cm242
Cm240
Po208
Po210
Ra224
Em222
Th226
Em212
Em220
Ra222
Po218
Po216
Em218
Po214
Po212

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Special mention should be made of the polonium curve since this illustrates the difficulties in presenting data on this type of plot for nuclei in which the nuclear radii are obviously not normal. The portion of the curve between the positions of Po^{218} and Po^{212} is normal and the half-lives taken from the curve may be thought of as those which each isotope would have if its decay-energy were as measured, but its nuclear radius were normal. This would mean in the case of Po^{212}(ThC') that its short half-life of 0.3 microsecond would actually be less than 0.1 microsecond if it were normal. Returning to the polonium curve of Fig. 1 in the energy region below Po^{218}, this portion of the curve now applies to isotopes of higher mass numbers (at present not known) and those of low mass number such as Po^{210} and is therefore not a single curve. By estimating the alpha-energy of Po^{220} as 5.3 Mev, it is possible to calculate its half-life and extend the normal curve shown as the solid line in Fig. 1. However, because of the great difference in mass number, the normal curve for Po^{208} and Po^{210} would lie somewhat higher and is indicated as a segment of broken line. This segment represents the hypothetical half-life versus decay-energy curve for polonium isotopes in this mass-number range if they could have both normal nuclear radii and the measured energies. Its only significance is that it should be the baseline for comparing half-lives in ascertaining the effect of nuclear radius on prohibition of alpha-decay. It will be noted that the one other even-even nuclide in this category, Em^{212}, should be treated similarly.

We wish to thank Dr. Kenneth Street for valuable discussions and suggestions concerning this paper. This paper is based on work performed under the auspices of the U. S. Atomic Energy Commission.