APPLICATION OF GAMOW'S THEORY OF α-EMISSION TO (4n+1) RADIOACTIVE SERIES.*

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ABSTRACT. Gamow's theory of α-emission is applied to neptunium (4n+1) series. The values of the assumed "nuclear radius" r_0 are found to vary irregularly as in actinium (4n+3) series. A large drop of r_0 occurs in $_{83}Bi^{213}$ similar to C-products of U, Th and Ac series A complete calculation of r_0 values for all the members of U, Th and Ac series is also included with their extension to transuranic region using latest experimental values. It has been discussed that the existing theories of α-emission with angular momentum are inadequate in explaining these irregular variations of r_0, specially in the odd radioactive series. It appears that the nuclear charge Z has something to do with the irregularities of r_0.

INTRODUCTION

Gamow's theory of leakage of α-particles through a potential barrier has been applied to three radioactive series known so long. The relation between the disintegration constant λ and decay energy E contains a term r_0 which is referred as "nuclear radius" on the assumption of simplified potential field. This denotes the distance from the centre of the nucleus to the point where the inverse square law of repulsion suddenly changes to a force of attraction as assumed by Gamow. In reality, the fall of potential near the nuclear boundary cannot be so abrupt ; but a calculation of r_0 from the experimentally determined values of λ and E are made to see whether these are consistent. The values of r_0 are in general agreement with the liquid drop model of a nucleus ($r_0 = R.\ A^{\frac{1}{3}}$) for U, and Th series and less satisfactory for Ac series. But abnormally low values of r_0 are obtained for all the C-products. Since Gamow's work r_0 values have been calculated by Bethe (1937) and by Preston (1946, 1947) for U, Th and Ac series. Recently the missing (4n+1) radioactive series has been identified by two groups of investigators (Hagemann *et al*, 1947 and English *et al*, 1947). Further data have been reported by Seaborg (1948). With these values of E and λ it is worthwhile to observe the consistency of r_0 values for this series. The present work is undertaken with this end in view.

Method of Calculation

The value of r_0 is calculated with experimental values of decay constant λ and disintegration energy E. Various forms of the relations used by different

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investigators have been referred in a previous paper. Rigorous calculation of transparency factor (Saha, 1944) and Laue's (1929) semi-classical arguments yield the relation

$$\lambda = \frac{v}{r_0} e^{-2K} \qquad \dots (1)$$

where v = velocity of α-particle relative to the product nucleus.

r_0 = radius of the product nucleus.

$$2k = \frac{16\pi e^2(Z-2)}{hv}(u_0 - \sin u_0 \cos u_0).$$

$$u_0 = \cos^{-1}\left[\frac{r_0}{R}\right]^{\frac{1}{2}} = \cos^{-1}\left[\frac{mv^2 r_0}{4e^2(Z-2)}\right]^{\frac{1}{2}}.$$

Preston (1946, 1947) uses more complex expressions, deduced from complex eigen-function,

$$\lambda = \frac{2v}{r_0}\frac{\mu^2 \tan u_0}{\mu^2 + \tan^2 u_0} e^{-2K} \qquad \dots (2a)$$

$$\mu = -\tan u_0 \tan(\mu k r_0) \qquad \dots (2b)$$

where $\mu = (1 - U/E_\alpha)^{\frac{1}{2}}$

$$k = \frac{2\pi mv}{h}.$$

These equations are very sensitive to the small variation in the exponential term, so the additional factor $\frac{\mu^2 \tan u_0}{\mu^2 + \tan^2 u_0}$ is not of much consequence. The values of r_0 calculated from relation (1) are given in Tables I and II. Preston (1946) remarks that the additional term in (2a) gives a refinement in the value of r_0. The method adopted for solution of (2a) and (2b) is, however, not referred. For the comparison of the values of, obtained from the two methods, the latter equations are also used in this work. Solutions of (2a) and (2b) for r_0 and μ are done here graphically by assuming a new variable $y = \mu k r_0$. Two explicit relations of y and u_0 are used to determine their values graphically. These come out as

$$y_1 = \frac{\pm(kR)\sin u_0 \cos u_0}{\left[\frac{2v}{\lambda R}\tan u_0 (1 + \tan^2 u_0)\, e^{-2K} - 1\right]^{\frac{1}{2}}}$$

$$y_2 = \frac{\mp 1}{\left[\frac{2v}{\lambda R}\tan u_0 (1 + \tan^2 u_0)\, e^{-2K} - 1\right]^{\frac{1}{2}}}$$

From the value of u_0, r_0 is calculated, and from r_0 and y, μ is calculated which in turn gives the value of U.

TABLE I

r_0 values for (4n + 1) Neptunium radioactive series.

Nuclei	$E\alpha$ in (Mev)	$v \times 10^{-9}$ cm/sec	λ sec^{-1}	From relation (1)		From relation (2)	
				$r_0 \times 10^{13}$ cm.	$R \times 10^{13}$ cm.	$r_0 \times 10^{13}$ cm	$R \times 10^{13}$ cm.
$_{95}Am^{241} \rightarrow {}_{93}Np^{237}$	5.50	1.661	4.40×10^{-11}	8.93	1.44	9.78	1.58
$_{93}Np^{237} \rightarrow {}_{91}Pa^{233}$	4.73	1.540	9.78×10^{-15}	9.09	1.48	9.79	1.59
$_{92}U^{233} \rightarrow {}_{90}Th^{229}$	4.825	1.556	1.37×10^{-13}	9.06	1.48	9.65	1.58
$_{90}Th^{229} \rightarrow {}_{88}Ra^{225}$	4.85	1.561	3.18×10^{-12}	9.03	1.52	9.67	1.59
$_{89}Ac^{225} \rightarrow {}_{87}Fr^{221}$	5.801	1.708	8.00×10^{-7}	8.68	1.44	9.23	1.53
$_{87}Fr^{221} \rightarrow {}_{85}At^{217}$	6.31	1.782	2.31×10^{-3}	8.67	1.41	9.34	1.54
$_{85}At^{217} \rightarrow {}_{83}Bi^{213}$	7.023	1.880	33	8.91	1.49	9.60	1.61
$_{83}Bi^{231} \rightarrow {}_{81}Tl^{209}$	5.86	1.718	3.15×10^{-6}	7.13	1.20	7.43	1.25
$_{84}Po^{213} \rightarrow {}_{82}Pb^{209}$	8.336	2.049	1.52×10^{-5}	8.40	1.42	9.10	1.53

TABLE II

r_0 values for Th, U and Ac -series.

(4n) Th-series.

Nuclei	$E\alpha$ in (Mev)	$v \times 10^{-9}$ cm/sec.	λ sec^{-1}	From relation (1)		From relation (2)	
				$r_0 \times 10^{13}$ cm.	$R \times 10^{13}$ cm.	$r_0 \times 10^{13}$ cm.	$R \times 10^{13}$ cm.
$_{96}Cm^{240} \rightarrow$	—	--	2.68×10^{-7}	—	—	—	—
$_{90}Th^{232} \rightarrow {}_{88}MsTh1^{228}$	3.92	1.400	1.58×10^{-18}	9.90	1.62	10.01	1.64
$_{90}RdTh^{228} \rightarrow {}_{88}ThX^{224}$	5.420	1.656	9.33×10^{-9} *	9.03	1.49	9.33	1.54
$_{88}ThX^{224} \rightarrow {}_{86}Tn^{220}$	5.681	1.690	2.2×10^{-6}	8.87	1.47	9.29	1.54
$_{86}Tn^{220} \rightarrow {}_{84}ThA^{216}$	6.282	1.778	1.27×10^{-2}	8.91	1.49	9.28	1.55
$_{84}ThA^{216} \rightarrow {}_{82}ThB^{212}$	6.774	1.847	4.39	8.69	1.46	9.12	1.53
$_{83}ThC^{212} \rightarrow {}_{81}ThC''^{208}$	6.054	1.746	1.75×10^{-5} *	7.07	1.19	7.57	1.28
$_{84}ThC'^{212} \rightarrow {}_{82}ThD^{208}$	8.776	2.102	2.31×10^{6}	8.65	1.46	9.15	1.54

TABLE II (contd.)

(4n + 2) U-Series

Nuclei	$E\alpha$ in (Mev)	$v\times10^{-9}$ cm/sec.	λ sec^{-1}	From relation (1)		From relation (2)	
				$r_0\times10^{13}$ cm	$R\times10^{13}$ cm.	$r_0\times10^{13}$ cm.	$R\times10^{13}$ cm.
$_{94}Pu^{238}\rightarrow{}_{92}UII^{234}$	5.496	1.669	4.39×10^{-10}	9.54	1.55	9.69	1.57
$_{92}U^{238}\rightarrow{}_{90}UX_1^{234}$	4.20	1.452	4.87×10^{-18}	9.27	1.46	9.37	1.52
$_{92}UII^{234}\rightarrow{}_{90}Io^{230}$	4.76	1.537	8.17×10^{-14}	9.38	1.53	9.26	1.51
$_{90}Io^{230}\rightarrow{}_{88}Ra^{226}$	4.66	1.530	2.65×10^{-13}	9.20	1.51	9.26	1.52
$_{88}Ra^{226}\rightarrow{}_{86}Rn^{222}$	4.791	1.552	1.35×10^{-11}*	9.03	1.49	9.29	1.53
$_{86}Rn^{222}\rightarrow{}_{84}RaA^{218}$	5.486	1.661	2.10×10^{-6}	8.96	1.49	9.28	1.54
$_{84}RaA^{218}\rightarrow{}_{82}RaB^{214}$	5.998	1.738	3.77×10^{-3}	8.80	1.47	9.14	1.53
$_{83}RaC^{214}\rightarrow{}_{81}RaC''^{210}$	5.502	1.664	1.06×10^{-7}*	7.30	1.27	7.80	1.31
$_{84}RaC'^{214}\rightarrow{}_{82}RaD^{210}$	7.680	1.966	4.62×10^{3}	8.74	1.47	9.34	1.57
$_{83}RaE^{210}\rightarrow{}_{81}Tl^{206}$	4.87	1.556	1.60×10^{-13}	6.50	1.10	6.63	1.12
$_{84}RaF^{210}\rightarrow{}_{82}RaG^{206}$	5.303	1.634	5.89×10^{-8}	8.04	1.36	8.27	1.40

TABLE II (contd.)

(4n + 3) Ac-series.

Nuclei	$E\alpha$ in (Mev)	$v\times10^{-9}$ cm/sec.	λ sec^{-1}	From relation (1)		From relation (2)	
				$r_0\times10^{13}$	$R\times10^{13}$	$r_0\times10^{13}$	$R\times10^{13}$
$_{94}Pu^{239}\rightarrow{}_{92}U^{235}$	5.137	1.605	9.28×10^{-12}	9.52	1.51	10.13	1.64
$_{92}U^{235}\rightarrow{}_{90}UY^{231}$	4.36	1.479	3.19×10^{-17}	8.97	1.46	9.55	1.56
$_{91}Pa^{231}\rightarrow{}_{89}Ac^{227}$	5.01	1.586	5.59×10^{-13} *	8.38	1.36	8.18	1.34
$_{90}RdAc^{227}\rightarrow{}_{88}AcX^{223}$	6.049	1.743	1.02×10^{-7} *	7.78	1.28	8.56	1.41
$_{88}AcX^{223}\rightarrow{}_{86}An^{219}$	5.719	1.695	2.86×10^{-7} *	8.08	1.34	8.70	1.44
$_{86}An^{219}\rightarrow{}_{84}AcA^{215}$	6.824	1.854	0.124 *	8.20	1.37	8.90	1.45
$_{84}AcA^{215}\rightarrow{}_{82}AcB^{211}$	7.365	1.925	3.79×10^{2}	8.65	1.45	8.99	1.51
$_{83}AcC^{211}\rightarrow{}_{81}AcC''^{207}$	6.619	1.825	4.78×10^{-3} *	7.65	1.29	7.90	1.33
$_{84}AcC'^{211}\rightarrow{}_{82}AcD^{207}$	7.434	1.934	1.39×10^{2}	8.20	1.39	8.85	1.50

*. Values of $\lambda\alpha_0$ (partial decay constant for o-group α-particles)

DISCUSSION

A study of the r_0 values for (4n + 1) neptunium radio-active series reveals the following features:

Firstly: The values of r_0 do not vary in a regular way as required by the rule $r_0 = R.A^{\frac{1}{3}}$. Such anomaly in r_0 value is predominant in actinium series. In fact the two odd series (4n + 1) and (4n + 3), behave in an irregular way as regards the r_0 values. On the other hand, two even series Th (4n) and U (4n + 2) show nearly regular variation of r_0 values with the exception of C-products. Calculation of r_0 are given by Preston (1946, 1947) for well-known members of U, Th and Ac series. Recently these known series are extended to the transuranic region and some of the experimental data have been revised. So a complete calculation of r_0 values for all the members of U, Th, and Ac series are also included with the latest experimental data. These are given in Table II. The values for (4n + 1) series together with those of U, Th and Ac series are given graphically in Fig. 1. The r_0

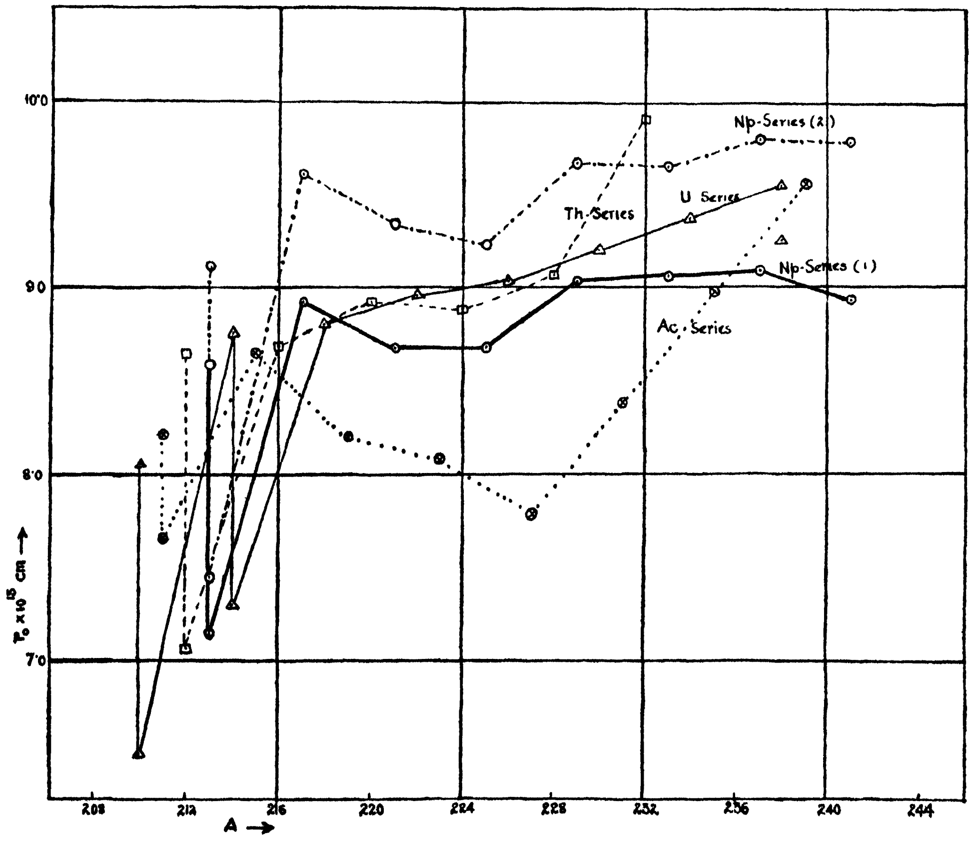

FIG. 1

values of Np series calculated according to both the relations (1) and (2) are plotted on curves. These show that values obtained by relation (2) are higher than the other values by a nearly constant quantity. The plotted values of U, Th and Ac series are those calculated from relation (1). Ac series shows a sharp regular fall to $RdAc^{227}$ and then a rise in r_0 up to AcA. But in Np series the irregularities are not so wide.

Secondly: The value of r_0 for ${}_{83}Bi^{213} \rightarrow {}_{81}Tl^{209}$ is abnormally low. Bi^{213} is the corresponding C-product of neptunium series. It has been observed that in U, Th, and Ac series there occurs an abrupt fall in the value of r_0 in $C \rightarrow C''$ disintegrations. The value of r_0 again assumes normal magnitude in $C' \rightarrow D$ disintegrations. Similar phenomena occur also in the (4n+1) radioactive series. It is interesting to note that ${}_{83}RaE^{210}$ which has been recently observed to be α-active (Broda and Feather, 1947) exhibits an abnormally low value of r_0 in ${}_{83}RaE^{210} \rightarrow {}_{81}Tl^{206}$ disintegration. Thus all the α-active isotopes of ${}_{83}Bi$ show abnormal value of "nuclear radius."

The drop in the values of r_0 for the C-products and the members of the Ac series have been attributed by Gamow (1937) as due to emission of α-particles with angular momentum different from zero ($l \neq 0$). The effective radius in such a case, as deduced by Gamow (1937) is supposed to follow the relation:

$$r_{eff} = r_0 - \frac{h^2}{4\pi^2 m e^2 (z-2)} \, l(l+1)$$

Allotments of l-values to different α-ray lines are rather arbitrarily made to fit the experimental data. No quantitative treatment on the above line has been found to be satisfactory.

Emission of α-particle with angular momentum $l \neq 0$ has been treated by Preston (1947). Calculations are made by him with the complicated relations for a few α-disintegrations having excited states. The method of calculation is very round about and l values are chosen arbitrarily to give a more or less consistent value of r_0 for different excited states for ThC−ThC″ and a few others. On the whole, the problem of emission of α-particle with $l \neq 0$ is at present far from satisfactory.

As in the Ac series, the irregularities in r_0 for the Np series is probably due to emission of α-particles with angular momentum different from zero. The experimental observations on (4n+1) series are rather preliminary. Further investigations are sure to reveal complex α-spectra in many members of (4n+1) radioactive series. A detailed experimental observations are required before any theoretical treatment is attempted. Since the irregularities in r_0 are found with the nuclei with odd mass number and in Bi which is the first member in the even series having odd atomic number it is plausible that the even-odd property of a nucleus affects the α-emission process to a large extent.

In this connection it may be mentioned that in U and Th series r_0 varies more or less regularly. However, the value of $R = r_0 A^{-\frac{1}{3}}$ is seen to be not a constant but increases from lightest member to heaviest one in the series to the extent of about 15%. The rule $r_0 = RA^{\frac{1}{3}}$ cannot be expected to hold accurately over the whole radioactive series because an increase in Z increases the Coulomb repulsion which tends to decrease the nuclear density, and increase the nuclear radius. The simple rule would hold if the nuclear binding energy contained the only term $E = \alpha A$. But E is given by the expression

$$E = \alpha A - \beta \frac{I^2}{A} - \gamma A^{\frac{2}{3}} - \delta \frac{Z^2}{A^{\frac{1}{3}}}$$

Hence with increasing Z, R the average distance between the nucleons should drift to a higher value. This is actually observed in the value of R calculated from r_0. Present (1940) proposed the following formula for nuclear radius after employing the corrections for $N \neq Z$, surface tension and Coulomb repulsion.

$$r_0 = R.A^{\frac{1}{3}} = R^* A^{\frac{1}{3}} \left[1 + 0.8 \frac{I^2}{A^2} - 0.3 A^{-\frac{1}{3}} + 0.01 \frac{Z^2}{A^{\frac{4}{3}}} \right]$$

In this relation R^* in place of R should be constant for all members in the series. For two extreme members of U series, R varies from 1.55 to 1.36 from $Pu^{238} \rightarrow U\,II^{234}$ to $RaF^{210} \rightarrow RaG^{206}$. With above relation, R^* comes out as 1.48 and 1.35. In case of Th series R for $Th^{232} \rightarrow MsThI^{228}$ is 1.62 and for $ThC'^{212} \rightarrow ThD^{208}$ 1.46. The corresponding values of R^* are 1.55 and 1.40. Thus the proposed relation is far from satisfactory. Although from the very definition of r_0, the relation between r_0 and actual nuclear radius is rather vague, the above relation cannot account for the variation of R along the radioactive series quantitatively.

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