Radioactive decay of radium and radon isotopes by ^{14}C emission

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Estimates are given for half-lives of the radioactive decay of radium and radon isotopes by emission of ^{14}C nuclei. The classical one-dimensional WKB approximation for penetration through a pure Coulomb barrier is used in the calculations. Results indicate that naturally occurring radon isotopes are ^{14}C emitters like their parent radium isotopes, whose decay rates by emission of ^{14}C nuclei have recently been measured. The predicted half-lives for decay of ^{219}Rn, ^{220}Rn, and ^{222}Rn by emission of ^{14}C nuclei are in the range 10^{11}–10^{14} yr. For radium and radon isotopes the minimum half-life is obtained when the double-magic ^{208}Pb and the semimagic ^{206}Hg, respectively, are the daughter products of these new radioactive decay modes.

I. INTRODUCTION

The first communications reporting the existence of new modes of nuclear decay in which heavy nuclei disintegrate by emission of nuclear fragments heavier than alpha particles were presented in 1975–1977, with both experimental and calculated results given.[1–4] In fact, it was in 1974, during the course of an experiment aimed at redetermining the spontaneous fission half-life of ^{238}U, that this new type of radioactivity became evident. Although the experimental method used (uranium-loaded nuclear-track emulsion) did not allow for the unique identification of the charge, mass, and energy of the emitted ions, observed as short-range nuclear tracks in the emulsion, it was concluded that these tracks originated by spontaneous nuclear disintegrations of the ^{238}U isotope, a case of emission of large nucleon clusters of intermediate mass in the region from neon to nickel. The half-life for such a decay process was estimated as $(2\pm1)\times10^{15}$ yr.[1–3] Calculations based on the classical WKB method for penetration through a potential barrier, similar to the formalism of the alpha-decay process, were performed in order to estimate the half-lives of the new decay modes. Within the limits of the large uncertainties the method imposes, these calculations indicated the possibility of a few nuclear-fragment emission modes from ^{238}U with fragment mass numbers ranging from 20 to 70, and with half-lives from about 10^{15} to 10^{18} yr, in agreement with experimental observation. Shell effects were clearly manifested, since the calculations indicated that the processes resulting in magic numbers either for the emitted fragment or for the daughter product are the most probable emission modes.[2,3,5]

These surprising results were interpreted by Sǎndulescu and Greiner[6] as being a case of very large asymmetry in the mass distribution of fragments of fissile nuclei generated by shell effects of one or both fragments.[6,7] Later, more refined and extensive calculations were performed by Sǎndulescu *et al.*[8] for heavy nuclear-cluster emissions by penetration through nuclear plus Coulomb potential barriers. It was concluded that conditions are most favorable for spontaneous emission of such clusters as ^{24}Ne and ^{28}Mg from the Th isotopes, ^{32}Si and ^{34}Si from the U isotopes, ^{46}Ar from the Pu and Cm isotopes, and ^{48}Ca from the Cf, Fm, and No isotopes. These early predictions have been superseded by more recent calculations.[9–11] Heavy nuclei, therefore, may exhibit a new type of decay which can be interpreted either as highly mass-asymmetric fission or as emission of a heavy nuclear cluster.[8] The prediction of this new phenomenon, i.e., an intermediate type of decay between alpha emission and fission, was strongly supported by the successful description of alpha decay as a process of superasymmetric fission.[12–14]

The first experimental identification of a case of radioactive decay of heavy nuclei by the emission of nuclear fragments heavier than alpha particles was done by Rose and Jones[15] from the University of Oxford, who reported observation of the radioactive decay of ^{223}Ra by ^{14}C emission with a half-life of $T_{1/2}=(3.7\pm1.1)\times10^{7}$ yr. This result was confirmed independently by Aleksandrov *et al.*,[16] and soon after by Gales *et al.*[17] and Price *et al.*[18] Kutschera *et al.*[19] designed a detailed experiment to measure the energy and mass of carbon nuclei emitted in the decay of ^{223}Ra. The ^{14}C nature of 24 particles of 29.8 MeV emitted from ^{223}Ra was unambiguously established and a branching ratio of $(4.7\pm1.3)\times10^{-10}$ relative to alpha emission has been obtained for such a decay, thus confirming previous results from other laboratories.[15–18]

The establishment of this new type of radioactive decay[20] has encouraged researchers to perform systematic calculations in order to estimate half-lives for novel modes of radioactive decay in which heavy nuclei disintegrate by emission of intermediate-mass fragments. Poenaru *et al.*[9] and Greiner *et al.*[21] have reported half-lives for probable emission modes from a number of heavy parent nuclei, some of the most likely candidates being ^{14}C from ^{223}Ra ($T_{1/2}=2.5\times10^{7}$ yr), ^{14}C from ^{226}Ra ($T_{1/2}=2.5\times10^{14}$ yr), ^{14}C from ^{227}Ac ($T_{1/2}=5.0\times10^{15}$ yr), ^{24}Ne from ^{232}U ($T_{1/2}=6.3\times10^{13}$ yr), ^{24}Ne from ^{233}U ($T_{1/2}=2.5\times10^{16}$ yr), ^{34}Si from ^{238}U ($T_{1/2}=1.6\times10^{20}$ yr), and ^{46}Ar from ^{252}Cf ($T_{1/2}=4.0\times10^{16}$ yr). Shi and

TABLE I. Observed radioactive decay of nuclei by emission of particles heavier than alpha particles (penetr. denotes penetrabilities).

Parent nucleus	Alpha-decay half-life $T^{\alpha}_{1/2}$ (yr)	Particle emitted	Mode of decay Daughter product	Q value (MeV)	Experimental results: Branching ratio relative to alpha decay B	Experimental results: Partial half-life for decay $T_{1/2}$ (yr)	Experimental results: Detection method	Experimental results: Ref.	Calculated values: Branching ratio B (Ref. 10)	Calculated values: Ratio of penetr. (Ref. 23)
^{221}Fr	9.3×10^{-6}	^{14}C	^{207}Tl	31.3	$<4.4\times10^{-12}$	$>2.1\times10^{6}$	a	18	2.5×10^{-13}	8.0×10^{-12}
^{221}Ra	8.9×10^{-7}	^{14}C	^{207}Pb	32.4	$<4.4\times10^{-12}$	$>2.0\times10^{5}$	a	18	5.0×10^{-13}	8.2×10^{-12}
^{222}Ra	1.2×10^{-6}	^{14}C	^{208}Pb	33.1	$(3.7\pm0.6)\times10^{-10}$	$(3.2\pm0.5)\times10^{3}$	a	18	1.0×10^{-11}	1.7×10^{-9}
					$(3.1\pm1.0)\times10^{-10}$	$(3.9\pm1.3)\times10^{3}$	b	24		
^{223}Ra	3.1×10^{-2}	^{14}C	^{209}Pb	31.9	$(8.5\pm2.5)\times10^{-10}$	$(3.7\pm1.1)\times10^{7}$	c	15	2.0×10^{-9}	6.9×10^{-9}
					$(7.6\pm3.0)\times10^{-10}$	$(4.1\pm1.6)\times10^{7}$	c	16		
					$(5.5\pm2.0)\times10^{-10}$	$(5.6\pm2.0)\times10^{7}$	b	17		
					$(6.1\pm1.0)\times10^{-10}$	$(5.1\pm0.8)\times10^{7}$	a	18		
					$(4.7\pm1.3)\times10^{-10}$	$(6.6\pm1.8)\times10^{7}$	d	19		
^{224}Ra	1.0×10^{-2}	^{14}C	^{210}Pb	30.5	$(4.3\pm1.2)\times10^{-10}$	$(2.3\pm0.6)\times10^{8}$	a	18	1.3×10^{-12}	6.2×10^{-11}
^{226}Ra	1.6×10^{3}	^{14}C	^{212}Pb	28.2	$(3.2\pm1.6)\times10^{-11}$	$(5.0\pm2.5)\times10^{13}$	b	24	2.0×10^{-12}	3.1×10^{-11}
^{230}Th	7.5×10^{4}	^{24}Ne	^{206}Hg	57.8	$(5.6\pm1.0)\times10^{-13}$	$(1.3\pm0.3)\times10^{17}$	e	25	4.0×10^{-13}	
^{231}Pa	3.3×10^{4}	^{24}Ne	^{207}Tl	60.4	6×10^{-12}	5×10^{15}	e	26	1.0×10^{-10}	9.4×10^{-12}
^{232}U	70	^{24}Ne	^{208}Pb	62.3	$(2.0\pm0.5)\times10^{-12}$	$(3.5\pm0.9)\times10^{13}$	e	27	1.3×10^{-11}	4.9×10^{-11}
^{233}U	1.6×10^{5}	^{24,25}Ne	^{209,208}Pb	60.8 60.5	$(7.5\pm2.5)\times10^{-13}$	$(2.2\pm0.8)\times10^{17}$	e	28	5.0×10^{-11}	2.6×10^{-10}
^{238}U	4.5×10^{9}	not identified experimentally			$(2\pm1)\times10^{-6}$	$(2\pm1)\times10^{15}$	f	1–3	1.6×10^{-11}[g]	
^{237}Np	2.1×10^{6}	^{30}Mg	^{207}Tl	75.0	$<4\times10^{-14}$	$>5.4\times10^{19}$	e	25	2.5×10^{-12}[h]	
^{241}Am	4.3×10^{2}	^{34}Si	^{207}Tl	93.8	$<3\times10^{-12}$	$>1.4\times10^{14}$	b	24	4.0×10^{-13}[i]	5×10^{-15}[i]
					$<3\times10^{-15}$	$>1.4\times10^{17}$	e	25		

[a]Polycarbonate track-recording film.
[b]Magnetic spectrometer with a silicon surface barrier counter telescope.
[c]Solid-state counter telescope.
[d]Magnetic spectrograph with a focal plane gas ionization detector.
[e]Polyethylene terephthalate track-recording film.
[f]Uranium-loaded nuclear-track emulsion pellicle.
[g]Predicted value for the emission of ^{34}Si, the most probable emission mode of ^{238}U (see Ref. 11).
[h]Quoted in Ref. 11.
[i]Quoted in Ref. 24.

Swiatecki[22,23] also have estimated lifetimes for radioactive decay of nuclei by emission of fragments heavier than alpha particles by treating these processes as extreme cases of asymmetric spontaneous fission. They give a closed formula for the penetrability factor which accounts for the recently observed branching ratios of alpha-particle to ^{14}C emissions from radium isotopes, and it can be used to estimate penetrability ratios for a number of new decay modes involving heavier fragments such as O, Ne, and Mg.

The advance in theoretical treatment of exotic decays, with good predictions for their half-lives, has motivated several experimental groups to search for and identify new types of radioactive decay.[24–28] Table I summarizes all experimental data where results are compared to estimates obtained from the superasymmetric fission models by Poenaru *et al.*[10] and Shi and Swiatecki.[23] In addition, during the last two years a number of important papers have been published. The reader is referred to publications by Poenaru *et al.*[10,11,29–31] which give a detailed description of the new phenomenon. They report calculated half-lives which replace the previous half-life estimates.[9,21,32]

In the present work we describe a very simple formalism which we used to interpret the events, recorded on emulsion plates, which we reported as a new type of radioactivity exhibited by the ^{238}U nucleus.[2,3,5] The assumption was that the classical theory of alpha decay developed in the framework of penetration through a pure Coulomb barrier[33] could also be extended to exotic decays. With modifications introduced to take more realistic nuclear radii and Q values into account, the same method is used in the present work to estimate half-lives for possible new cases of radioactive decay of nuclei by ^{14}C emission. We will focus attention on radioactive decay of radon isotopes by emission of ^{14}C nuclei, since calculations have indicated that the naturally occurring radon isotopes are good candidates, as are their parent radium isotopes, as we shall see in the following sections.

II. DECAY OF NUCLEI BY EMISSION OF PARTICLES HEAVIER THAN ALPHA PARTICLES

Early in 1975 we developed simple systematic calculations in order to search for possible new modes of radioactive decay from ^{238}U other than alpha-particle decay.[2,3,5] Recently, we have developed a new closed formula to calculate half-lives for radioactive decay processes in the framework of emission of heavy clusters. The following are basic assumptions from which a simple formula for the half-life has been derived..

(i) A parent nucleus (Z,A) in its ground state disintegrates by emitting fragment (Z_1,A_1) with formation of a daughter product nucleus (Z_2,A_2). Z and A refer to the proton and mass numbers, respectively. The energy available in the process is given by

$$Q=[M(Z,A)-M_1(Z_1,A_1)-M_2(Z_2,A_2)] \times 931.501\text{ MeV}\ , \qquad (1)$$

where the M's represent the atomic masses (expressed in u) whose values are taken, when available, from the 1983 Atomic Mass Evaluation by Wapstra and Audi,[34] or are otherwise estimated from systematics.

(ii) The entire positive Q value is taken to be the total kinetic energy available in the decay process, i.e., $Q=E_1+E_2$ is the effective total disintegration energy of the system available for the relative-motion channel. This means that the emitted cluster and the daughter product are assumed to be produced in their ground states. Accordingly, the fragment kinetic energies are calculated as $E_1=Q-E_2$ and $E_2=Q/(1+\eta)$, where $\eta=M_2/M_1$ is the mass asymmetry.

(iii) The decay constant λ, for emission of a heavy cluster ($Z_1>2$, $A_1>4$), is calculated to be the product of a frequency factor, λ_0, of the order of magnitude of collective oscillations ($10^{21}-10^{22}\ \text{s}^{-1}$), times a penetrability factor, P, through a pure Coulomb-type potential barrier at distances equal to, or greater than, the touching distance of the fragments. Contributions to the potential barrier caused by centrifugal effects and deformations from the parent configuration to the configuration at contact are not taken into account.

(iv) The frequency factor is calculated from the relative two-body motion where the fragments are thought of as moving back and forth with a frequency given by

$$\lambda_0=\frac{v_1+v_2}{2(C_1+C_2)}=\frac{(2Q/\mu)^{1/2}}{2(C_1+C_2)}\ , \qquad (2)$$

where v_1+v_2 is the relative velocity, C_1 and C_2 are the "central" radii of the fragments (see below), and $\mu=M_1M_2/(M_1+M_2)$ is the reduced mass of the system. The frequency factor λ_0 represents the number of assaults on the barrier per unit of time. Values calculated from Eq. (2) are in close agreement with those obtained from the currently used relationship $\lambda_0=\omega/2\pi=2E_v/h$, where ω is the characteristic frequency of collective nuclear oscillations, E_v is the energy of zero-point vibrations, and h is Planck's constant.

(v) Penetrabilities for heavy-particle emission modes through the Coulomb potential barrier are calculated in the same way as in alpha-particle emission.[33] As used in the one-dimensional WKB approximation, the touching distance and the distance between fragments at which the energy available for relative motion equals the potential Coulomb energy are the classical turning points. Accordingly, we have

$$P=\exp(-G)=\exp\left[-\frac{2}{\hbar}\int_c^d\{2\mu[V(s)-Q]\}^{1/2}ds\right]\ , \qquad (3)$$

where $V(s)=Z_1Z_2e^2/s$ is the potential energy, which depends on the distance s between the separating fragments, $c=C_1+C_2$ is the inner turning point, $d=Z_1Z_2e^2/Q$ is the outer turning point, $\hbar=h/2\pi$, and e is the elementary charge. The argument G in the exponential is often called the Gamow factor for decay.

(vi) The interacting fragments are considered to be spherical, and their extension (the location of the fragment surface) is defined by the central radius C. For the commonly used Fermi nuclear-charge density distributions the central radius equals the half-density radius, i.e., the distance at which the nuclear charge density has dropped to half its central value.[35] The central radius C, the "equivalent sharp radius" R, and the "root-mean-square radius" $\langle r^2\rangle^{1/2}$ are related, to a good approximation, by[36]

$$\langle r^2\rangle^{1/2}=(\tfrac{3}{5})^{1/2}R\left[1+\frac{5}{2}\frac{b^2}{R^2}\right], \quad (4)$$

$$C=R\left[1-\frac{b^2}{R^2}\right], \quad (5)$$

where $b=1$ fm is the nuclear "surface width."[35] Root-mean-square charge radii have been calculated by using the expression

$$\langle r^2\rangle^{1/2}=(\tfrac{3}{5})^{1/2}(1.15A^{1/3}+1.80A^{-1/3}-1.20A^{-1})\ \text{fm}, \quad (6)$$

following the "extended-liquid-drop" model by Myers and Schmidt.[37,38] These values are then combined with Eqs. (4) and (5) to give the central charge radii.

From assumptions (i)—(vi) above, and by expressing masses in u, energies in MeV, lengths in fm, and time in yr, we have, for the half-life,

$$T_{1/2}=3.16cV\times 10^{G/\ln 10-30}, \quad (7)$$

where

$$G=0.629\,943\,97Z_1Z_2VF_0, \quad (8)$$

$$V=(\mu/Q)^{1/2}, \quad (9)$$

$$F_0=\arccos(c/d)^{1/2}-[c/d-(c/d)^2]^{1/2}. \quad (10)$$

Equation (7) gives absolute values for the half-life in the sense that it does not contain any adjustable parameters.

An important quantity to be discussed is the uncertainty associated with the calculated half-life. This uncertainty is caused mainly by uncertainties in both nuclear mass and radius. The former affects the Q value for decay directly, giving rise to an uncertainty ΔQ, while the latter causes the touching distance of the fragments to be affected by an amount Δc. Taking the partial derivatives of the decimal logarithm of the half-life as deduced in Eq. (7), and neglecting the contribution due to fluctuations in the reduced mass, we have

$$\frac{\partial \log T_{1/2}}{\partial Q}\approx -0.137Z_1Z_2V\{\arccos(c/d)^{1/2}+[c/d-(c/d)^2]^{1/2}\}/Q, \quad (11)$$

$$\frac{\partial \log T_{1/2}}{\partial c}\approx -0.19QV(d/c-1)^{1/2}. \quad (12)$$

These results indicate that the half-life decreases with an increase in either the Q value or the touching distance c, as consequence of a reduction in the "barrier width" in both cases. The quantity

$$\Delta \log T_{1/2}=\left|\frac{\partial \log T_{1/2}}{\partial Q}\right||\Delta Q|+\left|\frac{\partial \log T_{1/2}}{\partial c}\right||\Delta c| \quad (13)$$

gives, in order of magnitude, the maximum error of the calculated half-life. As an example, consider the decay of ^{224}Ra by emission of ^{14}C fragments with a Q value of 30.5 MeV. We have $\mu=13.128$ u, $c=8.57$ fm, and $d=23.23$ fm. Therefore, $\partial \log T_{1/2}/\partial Q=-2.03$ and $\partial \log T_{1/2}/\partial c=-4.97$. By using data in the mass table by Wapstra and Audi,[34] $\Delta Q=7.3$ keV is derived. Δc results from a combination of uncertainties in the central radii ΔC_1 and ΔC_2 of the product nuclei. We may take as an overestimate for ΔC_1 and ΔC_2 the difference between the central radius as calculated from Eqs. (4)—(6) and the value obtained by $C=1.18A^{1/3}-0.48$ fm, which is derived from data provided by electron scattering experiments.[39] In this way, for ^{14}C we have $\Delta C_1=0.40$ fm and, for ^{210}Pb, $\Delta C_2=0.08$ fm. Hence, $\Delta c=0.41$ fm. Another estimate for ΔC_1 and ΔC_2 may be obtained by taking the difference $\langle r^2\rangle_e^{1/2}-\langle r^2\rangle_c^{1/2}$ between the experimental and calculated root-mean-square charge radii[38] as an uncertainty of $\langle r^2\rangle^{1/2}$. In this case, from Eqs. (4)—(6) we obtain $\Delta C_1=0.27$ fm, $\Delta C_2=0.02$ fm, and thus $\Delta c=0.27$ fm. Therefore, the maximum uncertainty in the calculated half-life amounts to 1.4—2.0 orders of magnitude. In general, Eq. (7) gives calculated half-lives within 1—2 orders of magnitude of experimental results. This corresponds to an accuracy of some 3—5 % in the G factor.

Finally, to complete the analogy to alpha decay, let us include in the derivation of the half-life formula the centrifugal barrier associated with the rotation of the product nuclei around their common center of mass. This enhances the potential barrier an amount $V'(s)$ given by

$$V'(s)=\frac{l(l+1)\hbar^2}{2\mu s^2}, \quad (14)$$

where l is the mutual orbital angular momentum. The result consists of changing Eq. (10) to a general expression of the form

$$F_l=\frac{x^{1/2}}{2a}\ln\left[\frac{[x(x+2a-1)]^{1/2}+x+a}{(x/a)[1+(1+x/a^2)^{1/2}]^{-1}+a}\right]+\arccos\left\{\frac{1}{2}\left[1-\frac{1-1/a}{(1+x/a^2)^{1/2}}\right]\right\}^{1/2}-\left[\frac{x}{4a^2}+\frac{1}{2a}-\left[\frac{1}{2a}\right]^2\right]^{1/2}, \quad (15)$$

where

$$x=\frac{E_l}{Q},\quad E_l=\frac{20.901l(l+1)}{\mu c^2}\ \text{MeV}, \quad (16)$$

$$a=\frac{1}{2}\frac{d}{c},\quad d=\frac{Z_1Z_2e^2}{Q}, \quad (17)$$

$$e^2=1.439\,978\,4\ \text{MeV fm}.$$

The physical effect of the centrifugal barrier is to hinder the decay rate so as to cause an increase in the half-life by a factor f which is given by

TABLE II. ^{14}C decay of radium isotopes ($Z=88$).

Mass number A	Q value (MeV)	Experimental half-life $T_{1/2}^{C}$ (yr)	Ref.	Calculated half-life $T_{1/2}^{C}$ (yr) Ref. 9	Ref. 10	Ref. 23	This work
220	31.0			7.3×10^{8}	1.3×10^{9}		6.0×10^{8}
221	32.4	$>2.0\times10^{5}$	18	1.1×10^{6}	2.0×10^{6}	1.1×10^{5}[a]	1.4×10^{6}
222	33.1	$(3.2\pm0.5)\times10^{3}$	18	2.4×10^{5}	1.3×10^{5}	7.1×10^{2}	8.1×10^{4}
		$(3.9\pm1.3)\times10^{3}$	24				
223	31.9	$(3.7\pm1.1)\times10^{7}$	15	2.5×10^{7}	3.2×10^{7}	4.5×10^{6}	1.1×10^{7}
		$(4.1\pm1.6)\times10^{7}$	16				
		$(5.6\pm2.0)\times10^{7}$	17				
		$(5.1\pm0.8)\times10^{7}$	18				
		$(6.6\pm1.8)\times10^{7}$	19				
224	30.5	$(2.3\pm0.6)\times10^{8}$	18	4.5×10^{9}	8.0×10^{9}	1.6×10^{8}	3.9×10^{9}
225	29.5				1.3×10^{12}		6.1×10^{11}
226	28.2	$(5.0\pm2.5)\times10^{13}$	24	2.5×10^{14}	8.0×10^{14}	5.2×10^{13}	3.3×10^{14}
228	26.1						4.4×10^{19}

[a]The values listed in this column have been calculated by $T_{1/2}^{C}=T_{1/2}^{\alpha}/B$, where $T_{1/2}^{\alpha}$ is the experimental alpha-decay half-life and B is the branching ratio relative to alpha decay whose values were set equal to the penetrability ratios (G_α/G_C) as given in Ref. 23.

$$f=\frac{T_{1/2}(l)}{T_{1/2}(0)}=\exp[0.629\,943\,97Z_1Z_2V(F_l-F_0)]\ . \qquad (18)$$

This corresponds to a variation of $\Delta G/G=F_l/F_0-1$ in the G factor. Such results will be useful in the discussion of decays of odd-A emitters.

III. DECAY OF RADIUM ISOTOPES BY EMISSION OF ^{14}C

Before entering into discussion of the radioactive decay of radon isotopes by emission of ^{14}C nuclei, we applied the calculation method described above to obtain estimates of the half-lives of radium isotopes by emission of ^{14}C nuclei. The calculated half-lives are then compared with the measured values, as well as the values estimated by other authors. These data are presented in Table II and Fig. 1. As is seen, the present estimates are in quite good agreement with those of Poenaru *et al.*,[9,10] and, in general, they differ only by about 1 order of magnitude from the estimates of Shi and Swiatecki[23] (the only exception is in the case of ^{222}Ra, for which the half-life value given by Shi and Swiatecki is about 2 orders of magnitude lower than ours, but the former agrees quite well with the experimental value, as shown in Fig. 1). Quite good agreement is also noted in comparing the estimates of Shi and Swiatecki with experimental results for ^{224}Ra and ^{226}Ra isotopes, while for ^{223}Ra a difference of about 1 order of magnitude is noted. The small deviations between the calculated and experimental half-lives, as well as from each other, however, are of little importance since both the experimental and calculated values are affected by large uncertainties.

Let us discuss in some detail the case of the ^{223}Ra isotope. According to nuclear spin and parity conservation laws, in the transition from the ground state of ^{223}Ra ($J^\pi=\frac{1}{2}^+$) to the ground state of ^{209}Pb ($J^\pi=\frac{9}{2}^+$) through the emission of a ^{14}C nucleus ($J^\pi=0^+$), there are four units of mutual angular momentum ($l=4$). Therefore, as may be calculated using Eqs. (15)–(18), the predicted half-life under the assumption that there are no centrifugal effects ($l=0$) should in increased by a factor $f=1.66$ when such effects are to be taken into account. This correction in the calculated half-life, although not insignificant, falls, nonetheless, within the range of the measured half-life (see Table II). Besides, the G factor for decay has increased only by 0.6% and is, therefore, within the large uncertainties of the G factor itself. The reason for the

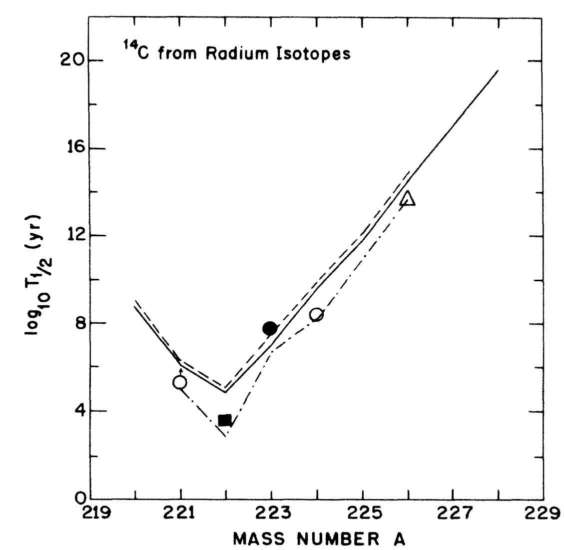

FIG. 1. ^{14}C-decay half-life plotted against mass number of radium isotopes. Experimental points: ●, mean value from Refs. 15–19; ○, Ref. 18; ■, mean value from Refs. 18 and 24; △, Ref. 24. Estimated values are joined by lines: — — —, Poenaru *et al.* (Ref. 10); —·—·—·, Shi and Swiatecki (Ref. 23) (see note on Table II); ——, present work.

TABLE III. ^{14}C decay of radon isotopes ($Z=86$).

Mass number A	Alpha-decay half-life $T^{\alpha}_{1/2}$ (yr)	Q value (MeV)	Calculated ^{14}C-decay half-life, $T^{C}_{1/2}$ (yr) Ref. 11	This work	Branching ratio relative to alpha decay, B Ref. 11	This work
217	1.7×10^{-11}	25.9	1.0×10^{19}	2.8×10^{18}	2.0×10^{-30}	6.1×10^{-30}
218	1.1×10^{-9}	26.9	2.5×10^{16}	8.3×10^{15}	5.0×10^{-26}	1.3×10^{-25}
219	1.3×10^{-7}	28.1	2.5×10^{13}	9.3×10^{12}	6.3×10^{-21}	1.4×10^{-20}
220	1.8×10^{-6}	28.5	2.5×10^{12}	9.7×10^{11}	7.9×10^{-19}	1.8×10^{-18}
221	2.2×10^{-4}	27.7	2.5×10^{14}	9.0×10^{13}	1.3×10^{-18}	2.4×10^{-18}
222	1.0×10^{-2}	27.6	3.2×10^{14}	1.2×10^{14}	3.2×10^{-17}	8.7×10^{-17}
223		25.9		1.4×10^{18}		

centrifugal barrier causing such a small effect on the half-life in the case of ^{223}Ra isotope is that the centrifugal energy at fragment contact ($E_l=0.44$ MeV) represents only 1.4% of the Q value for the decay and 0.5% of the Coulomb energy at fragment contact. This is caused by the simultaneous combination of the small value of the angular momentum and a large reduced mass of the system ($\mu\approx13$ u). Thus, for all practical purposes, centrifugal effects in decays by ^{14}C emission can be considered completely negligible. On the other hand, under the assumption that $l=0$, we may have a situation in which ^{209}Pb is produced in the 2.032-MeV excited state (the first $\frac{1}{2}^+$ state of ^{209}Pb). In this case, the available energy for the relative-motion channel would be decreased by about 2 MeV and, therefore, the half-life for such transitions would be increased by almost 4 orders of magnitude, as can be deduced by applying Eq. (11). It would, of course, be very difficult to observe such transitions. In fact, experiments[15–19] have shown that the kinetic energy of the ^{14}C nuclei produced from ^{223}Ra (29.8 MeV) is compatible with the full Q value for decay (31.9 MeV).

Figure 1 clearly shows the shell effect exhibited in the decay of ^{222}Ra to the double magic ^{208}Pb, produced in the disintegration by emission of ^{14}C, and giving, therefore, the lowest half-life value among radium isotopes. Thus we have seen that the emission of ^{14}C fragments from radium isotopes can be treated as a case of cluster emission with half-lives successfully predicted by a simple alpha-decay-like model.

IV. DECAY OF RADON ISOTOPES BY EMISSION OF ^{14}C

In a similar way we have applied the formalism developed in Sec. II to estimate the half-lives of radon isotopes by emission of ^{14}C nuclei. Results are presented in Table III and Fig. 2. In the case of ^{219}Rn and ^{220}Rn isotopes, it is possible to make a comparison with previous half-life predictions by Poenaru *et al.*,[9] obtained on the basis of a superasymmetric fission model. Again, good agreement is observed in the results from the two extreme models. In the case of ^{222}Rn a 0.5-MeV uncertainty may result in the mass evaluation of the daughter product ^{208}Hg. The same occurs for ^{223}Rn and its daughter product ^{209}Hg, since the masses of these nuclides have been evaluated by systematics, and therefore the half-life predictions may be uncertain up to about 2 orders of magnitude. Even so, the data displayed in Table III are sufficient to conclude that radon isotopes belonging to the naturally occurring radioactive series are the most active regarding emission of ^{14}C with half-lives in the range 10^{11}–10^{14} yr. Similarly, as was done in the case of ^{223}Ra, the ^{219}Rn isotope had its decay studied for some possible centrifugal effect. In this case $l=3$, $E_l=0.26$ MeV, $f=1.38$, and $\Delta G/G=0.3\%$, and the conclusions remain the same as for ^{223}Ra. Again, shell effects are clearly manifested, since ^{220}Rn, whose daughter product is the semimagic ^{206}Hg in the decay by emission of ^{14}C, has the lowest half-life (see Fig. 2). From the experimental point of view, however, the quantity of interest is the branching ratio relative to alpha emission. The data in Table III show that any attempt to detect such rare modes of decay from radon isotopes would meet serious experimental difficulties, even in the case of ^{222}Rn, which exhibits, among radon isotopes, the most favorable branching ratio ($\sim10^{-16}$) of ^{14}C emission relative to alpha emission.

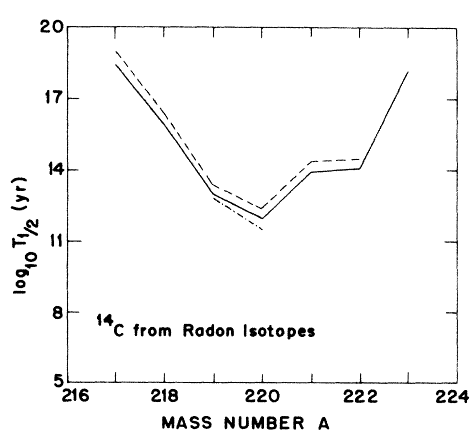

FIG. 2. ^{14}C-decay half-life plotted against mass number of radon isotopes. Estimates of the present work are joined by solid lines, previous results by Poenaru *et al.* (Ref. 9) by a dashed-dotted line, and the more recent estimates by Poenaru *et al.* (Ref. 11) by a dashed line.

V. CONCLUSIONS

In the course of the present work, the radioactive decay of radium isotopes by emission of ^{14}C nuclei has been reviewed. A closed formula for half-life predictions of such decays has been deduced within the framework of cluster emission from heavy nuclei, in a manner similar to the classical theory of alpha-particle emission. The one-dimensional WKB approximation for penetration through a pure Coulomb barrier has been assumed in the calculation. The model has proved to be adequate in reproducing, within the uncertainties the method imposes, the measured half-lives of the radioactive decay of radium isotopes by emission of ^{14}C nuclei. It has been shown that radon isotopes of the naturally occurring radioactive series are also ^{14}C emitters with predicted half-lives in the range $10^{11}-10^{14}$ yr, and that ^{220}Rn is the most active emitter of ^{14}C among radon isotopes. These new modes of radioactive decay can be recognized as a clear manifestation of the shell effects of the product nuclei, since the minimum half-life is obtained when the double-magic ^{208}Pb (in the case of radium isotopes) and the semimagic ^{206}Hg (in the case of radon isotopes) are the daughter product nuclei in the disintegration processes. The extent of the calculation method used in the present work to estimate half-lives of spontaneous emission of more complex nuclear fragments such as O, Ne, Mg, Si, Ar, and Ca from heavy nuclei (also including alpha decay and spontaneous fission as extreme modes of decay) will be discussed in a future communication. Among these cases of radioactive decay the ^{24}Ne emission from a number of heavy parent nuclei has been recently discovered, as mentioned in the Introduction.

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