ISSN 0029 - 3865

CBPF-NF-015/86

RADIOACTIVE DECAY OF RADIUM AND RADON ISOTOPES BY ^{14}C EMISSION

by

H.G. de Carvalho[†], J.B. Martins and O.A.P Tavares

Centro Brasileiro de Pesquisas Físicas - CBPF/CNPq
Rua Dr. Xavier Sigaud, 150
22290 - Rio de Janeiro, RJ - Brasil

[†] On leave of absence from the Brazilian Comissão Nacional de Energia Nuclear-CNEN, Rio de Janeiro.

Estimates are given for the 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 the naturally occurring radon isotopes are ^{14}C emitters like their parents radium isotopes whose activities by emission of ^{14}C nuclei have been recently 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^{13} yr. For radium and radon isotopes the minimum half-life is obtained when the double-magic ^{208}Pb and the semi-magic ^{206}Hg are the daughter products, respectively, of these new radioactive decay modes.

Key-words: Radioactivity; ^{14}C-decay; Radium isotopes; Radon isotopes; Half-life predictions; Branching ratio; WKB method.

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 early in 1975-1977 in which both experimental and calculated results were given[1-4]. In fact, it was in 1974 during the course of an experiment aimed to re-determine 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 a perfect identification of the charge, mass and energy of recorded ions visualized as short-range nuclear-tracks in the emulsion, it was concluded that these tracks originated by spontaneous nuclear disintegrations from the ^{238}U isotope, a case of emission of large nucleon-clusters of intermediate masses in the region from neon to nickel. The half-life for such a decay process was estimated to be $(2\pm1)\times10^{15}$ 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 for 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 mass number ranging from 20 to 70, whose half-lives were about 10^{15} to 10^{18} yr, in agreement with the experimental observation. Shell effects were clearly manifested, since the calculations indicated the processes involving magic number either for the emitted fragment or for the daughter product as the most probable emission modes[2,3,5].

The surprising results mentioned above were promptly recognised by Săndulescu and Greiner[6] as a case of very large asymmetry in the mass distribution of the fragments of fissile nuclei generated by shell effects of one of the fragments close to doubly magic nuclei[6,7]. Later, more refined and extensive calculations were performed by Săndulescu *et al.*[8] for heavy nuclear clusters emission by penetration through nuclear plus Coulomb potential barriers. The conclusion was drawn that the conditions are the most favorable for spontaneous emission of clusters such as ^{24}Ne and ^{28}Mg from the Th isotopes, ^{32}Si and ^{34}Si from the U isotopes, ^{48}Ar from Pu and Cm isotopes, and ^{48}Ca from the Cf, Fm and No isotopes. Therefore, heavy nuclei 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 the alpha-decay as a fission process of superasymmetry[9-11].

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[12] from the University of Oxford who reported the 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.*[13], and soon after by Gales *et al.*[14] and Price *et al.*[15]. The decay by emission of ^{14}C of the ^{222}Ra and ^{224}Ra isotopes was also discovered by Price *et al.*[15] with half-lives of $(3.3\pm0.5)\times 10^3$yr and $(2.3\pm0.7)\times10^8$ yr, respectively.

The well-established new type of radioactive decay[16] 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.*[17] and Greiner *et al.*[18] 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 ($T_{1/2} = 2.5\times10^{18}$ yr), ^{34}Si from ^{238}U ($T_{1/2} = 1.6\times10^{20}$ yr), and ^{46}Ar from ^{252}Cf ($T_{1/2} = 4.0\times10^{18}$ yr). Also, Shi and Swiatecki[19,20] 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 can account for the recently observed branching ratios between alpha-particle and ^{14}C emissions from radium isotopes and can be used to estimate penetrability ratios for a number of such new decay modes involving heavier fragments like O, Ne and Mg.

The advance in treating theoretically exotic radioactive decays of heavy nuclei involving the emission of complex nuclear fragments such as C, O, Ne, Mg and Si with good predictions for the half-life of these decays has motivated several experimental groups to search for and to identify new types of radioactive decay modes. Barwick *et al.*[21] have detected for the first time the decay mode in which an energetic ^{24}Ne nucleus is emitted from ^{232}U with a half-life of $T_{1/2} = (7.0\pm1.8)\times10^{13}$ yr. Hourani *et al.*[22] give the first experimental evidence for the radioactive decay of ^{226}Ra by ^{14}C emission having obtained a half-life for this process of

$T_{1/2} = (5.0\pm2.5)\times10^{13}$ yr. These authors also confirmed the decay of ^{222}Ra by ^{14}C emission previously reported by Price *et al.*[15], and an upper limit of 3×10^{-12} for the branching ratio relative to alpha-decay was found for the emission of ^{34}Si from ^{241}Am. Researchers at Dubna have also contributed to new findings such as the emission of ^{24}Ne from ^{231}Pa with a half-life of $T_{1/2} = 5\times10^{15}$ yr[23], and the discovery that ^{233}U is a Ne emitter[24] with a half-life of $T_{1/2} = (2.2\pm0.8)\times10^{17}$ yr. Kutschera *et al.*[25] designed a detailed experiment to measure the energy and mass of the 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 the previous results from other laboratories[12-15]. Very recently, Tretyakova *et al.*[26] have reported the radioactive decay of ^{230}Th by ^{24}Ne nuclei emission with a half-life of $(1.3\pm0.3)\times10^{17}$ yr. For decays by heavy cluster emission from ^{237}Np and ^{241}Am upper limits for the branching ratio relative to alpha emission were set at about 4×10^{-14} and 3×10^{-15}, respectively. All these experimental data are summarised in Table I and results compared to estimates obtained from the superasymmetric fission models by Poenaru *et al.*[17,27] and by Shi and Swiatecki[20]. In addition, during 1984 and 1985 a number of important papers have been published. The reader is referred to publications by Poenaru *et al.*[27-30] which give a detailed description of the new phenomenon.

In the present work we describe a very simple formalism we used to interpret the events recorded on emulsion plates which we reported as a new type of radioactivity exhibit by ^{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[31] could be extended to decays by emission of fragments heavier than alpha particles. 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 new possible cases of radioactive decay of nuclei by ^{14}C emission. We will focus attention on the cases of 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 much as their parents radium isotopes as emitters of ^{14}C as we shall see in the next sections.

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

Early in 1975 we had 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]. Later, 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 the fragment (Z_1,A_1) with the formation of the 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

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

where the M's represent the atomic mass (expressed in u) whose values are taken from the 1983 Atomic Mass Evaluation by Wapstra and Audi[32] when available, or estimated otherwise from systematics.

ii) The entire positive Q-value is taken as 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 considered 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, $\lambda = \lambda_0 P$, for emission of a heavy cluster ($Z_1 > 2$, $A_1 > 4$) is calculated as the product of a frequency factor, λ_0, of the order of magnitude of collective oscillations (10^{21}-10^{22} 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 due to 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 in the attempts to penetrate the barrier, according to which the fragments are thought of as moving back and forth with a frequency given by

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

where v_1+v_2 is the relative velocity, C_1 and C_2 are the "central" radii (see below) of the fragments, 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) The penetrabilities for heavy-particle emission modes through the Coulomb potential barrier are calculated in the same way as for an alpha-particle emission[31]. It is used throughout the one--dimensional WKB approximation, where the classical turning points are chosen as the touching distance and the distance between fragments at which the energy available for relative motion equals to the potential Coulomb energy. Accordingly, we have

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

where $V(r) = Z_1Z_2e^2/r$ is the potential energy which depends on the distance r 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 ones, 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 to the half-density radius, i.e. the distance where the nuclear charge density has dropped to half its central value[33]. The central radius

C, the "equivalent sharp radius" R, and the "equivalent root-mean-square radius" Q are related to a good approximation by[34]

$$Q = R\left(1 + \frac{5}{2}\frac{b^2}{R^2}\right) \tag{4}$$

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

where $b = 1$ fm is the nuclear "surface width"[33]. Root-mean-square radii have been calculated by using the expression[35]

$$Q = 1.15A^{1/3} + 1.80A^{-1/3} - 1.20A^{-1} \quad \text{fm} \tag{6}$$

following the "extended-liquid-drop" model by Myers and Schmidt[36]. These values are then combined with Eqs. (4) and (5) to give the central radii.

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

$$T_{1/2} = 3.16cV\times10^{\frac{G}{\ln 10} - 30} \quad , \tag{7}$$

where

$$G = 0.62994397Z_1Z_2FV \tag{8}$$

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

$$F = \arctan(d/c-1)^{1/2} - [c/d-(c/d)^2]^{1/2} \quad . \tag{10}$$

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

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

$$\partial \log T_{1/2}/\partial Q \approx -0.137 Z_1 Z_2 \{\arctan[d/c-1]^{1/2} + [c/d(1-c/d)]^{1/2}\}/Q \qquad (11)$$

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

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

$$\Delta \log T_{1/2} = |\partial \log T_{1/2}/\partial Q|\,|\Delta Q| + |\partial \log T_{1/2}/\partial c|\,|\Delta c| \qquad (13)$$

gives, in orders 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 $\partial \log T_{1/2}/\partial c = 4.97$. If uncertainties such as $\Delta Q \approx 0.5$ MeV and $\Delta c \approx 0.1$ fm are allowed for the Q-value and the touching distance, respectively, we obtain a maximum uncertainty of $\sim$ 1.5 orders of magnitude for the half-life. In general, Eq. (7) gives calculated half-lives within 1-2 orders of magnitude. This corresponds to an accuracy of some 3-4% in the *G* factor.

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

Before entering the 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 estimated values by other authors. These data are presented in Table II and Fig. 1. As can be seen, the present estimates are in quite good agreement with those of Poenaru *et al.*[17,27], and in general they differ only by about one order of magnitude from Shi and Swiatecki's[20] estimates (the only exception is in the case of ^{222}Ra for which the half-life-value by Shi and Swiatecki is about two orders of magnitude lower than ours, but the former agrees quite well with the experimental value as it is shown in Fig. 1). Quite good agreement is also noted by comparing Shi and Swiatecki's estimates with the experimental results for ^{224}Ra and ^{226}Ra isotopes, while for ^{223}Ra a difference of about one order of magnitude is noted. The small deviations of the calculated from the experimental half-lives as well as from each other, however, are of less importance since both the experimental and calculated values are affected by large uncertainties. Fig. 1 shows clearly the shell effect exhibited in the case of ^{222}Ra for which the double magic ^{208}Pb is produced in the disintegration by emission of ^{14}C giving, therefore, the lowest half-life 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 Section 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.*[17] obtained on the basis of a superasymmetric fission model. Again, good agreement is observed between the results from the two extreme models. In the case of ^{222}Rn some 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 two orders of magnitude. Even so, the data displayed in Table III are sufficient to conclude that the radon isotopes belonging to the naturally occurring radioactive series are the most active towards emission of ^{14}C with half-lives in the range 10^{11}-10^{13} yr. Again, shell effects are clearly manifested since ^{220}Rn, whose daughter product is the semi-magic ^{206}Hg in the decay by emission of ^{14}C, has shown to have 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. From data of

Table III it is seen 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 ($\sim 5\times10^{-16}$) of ^{14}C emission relative to alpha emission.

V. CONCLUSIONS

During 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 in the framework of cluster emission from heavy nuclei in a similar way to the case of 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 shown to be very 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 the radon isotopes of the naturally occurring radioactive series are also ^{14}C emitters with predicted half-lives in the range 10^{11}-10^{13} yr, and that ^{220}Rn is the most active towards emission of ^{14}C among radon isotopes. These new modes of radioactive decay can be understood 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 semi-magic ^{206}Hg (in the case of radon isotopes) are the daughter product nuclei in the disintegration processes. The extent of the calculation method of 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 referred in the introductory part of this work.

ACKNOWLEDGEMENTS

The authors would like to thank L.T. Auler and A.G. da Silva for many valuable discussions. The partial support by the Brazilian Comissão Nacional de Energia Nuclear-CNEN, Rio de Janeiro, is gratefully acknowledged.

TABLE I - Observed radioactive decay of nuclei by emission of particles heavier than alpha particles.

		Mode of decay			Experimental Results				Calculated values	
Parent nucleus	Alpha-decay half-life $T^{\alpha}_{1/2}$ (yr)	Particle emitted	Daughter product	Q-value (MeV)	Branching ratio relative to alpha-decay, B	Partial half-life for decay, $T_{1/2}$(yr)	Detection method	Ref.	Branching ratio, B (Ref.27)	Ratio of penetrabilities (Ref.20)
^{221}Ra	8.9×10^{-7}	^{14}C	^{207}Pb	32.4	$< 4.4\times10^{-12}$	$> 2.0\times10^{5}$	a	15	4.4×10^{-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	15	1.0×10^{-11}	1.7×10^{-9}
					$(3.1\pm1.0)\times10^{-10}$	$(3.9\pm1.3)\times10^{3}$	b	22		
^{223}Ra	3.1×10^{-2}	^{14}C	^{209}Pb	31.9	$(8.5\pm2.5)\times10^{-10}$	$(3.6\pm1.1)\times10^{7}$	c	12	2.0×10^{-9}	6.9×10^{-9}
					$(7.6\pm3.0)\times10^{-10}$	$(4.1\pm1.6)\times10^{7}$	c	13		
					$(5.5\pm2.0)\times10^{-10}$	$(5.6\pm2.0)\times10^{7}$	b	14		
					$(6.1\pm1.0)\times10^{-10}$	$(5.1\pm0.8)\times10^{7}$	a	15		
					$(4.7\pm1.3)\times10^{-10}$	$(6.6\pm1.8)\times10^{7}$	d	25		
^{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	15	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	22	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	26	3.2×10^{-13}	—
^{231}Pa	3.3×10^{4}	^{24}Ne	^{207}Tl	60.4	6×10^{-12}	5×10^{15}	e	23	1.0×10^{-10}	9.4×10^{-12}
^{232}U	70	^{24}Ne	^{208}Pb	62.3	$(1.00\pm0.25)\times10^{-12}$	$(7.0\pm1.8)\times10^{13}$	e	21	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	24	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	2.8×10^{-11}[g]	—
^{237}Np	2.1×10^{6}	^{30}Mg	^{207}Tl	75.0	$< 4\times10^{-14}$	$> 5.4\times10^{19}$	e	26	2.5×10^{-12}[h]	—
^{241}Am	4.3×10^{2}	^{34}Si	^{207}Tl	94.2	$< 3\times10^{-12}$	$> 1.4\times10^{14}$	b	22	4.0×10^{-13}[i]	5×10^{-15}[i]
					$< 3\times10^{-15}$	$> 1.4\times10^{17}$	e	26		

[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.

[h] Quoted in Ref. 29.

[i] Quoted in Ref. 22.

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

Mass number A	Q-value (MeV)	Experimental half-life $T^C_{1/2}$(yr)	Ref.	Calculated half-life, $T^C_{1/2}$(yr)			
				Ref. 17	Ref. 27	Ref. 20	This work
220	31.0	—	—	7.3×10^{8}	1.3×10^{9}	—	6.3×10^{8}
221	32.4	$>2.0\times10^{5}$	15	1.1×10^{6}	2.0×10^{6}	1.1×10^{5}[a]	1.5×10^{6}
222	33.1	$(3.2\pm0.5)\times10^{3}$	15	2.4×10^{5}	1.3×10^{5}	7.2×10^{2}	8.1×10^{4}
		$(3.9\pm1.3)\times10^{3}$	22				
223	31.9	$(3.6\pm1.1)\times10^{7}$	12	2.5×10^{7}	3.2×10^{7}	4.5×10^{6}	1.1×10^{7}
		$(4.1\pm1.6)\times10^{7}$	13				
		$(5.6\pm2.0)\times10^{7}$	14				
		$(5.1\pm0.8)\times10^{7}$	15				
		$(6.6\pm1.8)\times10^{7}$	25				
224	30.5	$(2.3\pm0.6)\times10^{8}$	15	4.5×10^{9}	8.0×10^{9}	1.6×10^{8}	3.9×10^{9}
225	29.5	—	—	—	1.3×10^{12}	—	6.9×10^{11}
226	28.2	$(5.0\pm2.5)\times10^{13}$	22	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 $T^C_{1/2} = T^{\alpha}_{1/2}/B$, where $T^{\alpha}_{1/2}$ 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. 20.

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)		Branching ratio relative to alpha decay, B	
			Ref. 17	This work	Ref. 17	This work
217	1.7×10^{-11}	25.9	—	2.8×10^{18}	—	6.1×10^{-30}
218	1.1×10^{-9}	26.9	—	8.3×10^{15}	—	1.3×10^{-25}
219	1.3×10^{-7}	28.1	6.3×10^{12}	9.3×10^{12}	2.1×10^{-20}	1.4×10^{-20}
220	1.8×10^{-6}	28.5	2.9×10^{11}	9.7×10^{11}	6.3×10^{-18}	1.8×10^{-18}
221	2.2×10^{-4}	27.7	—	9.2×10^{13}	—	2.4×10^{-18}
222	1.0×10^{-2}	27.9	—	2.0×10^{13}	—	5.0×10^{-16}
223	—	25.9	—	1.4×10^{18}	—	—

Figure Captions

FIG. 1 - ^{14}C-decay half-life plotted against mass number of radium isotopes. Experimental points: ● , mean value from Refs. 12-15,25; ○ , Ref. 15; ■ , mean value from Refs. 15,22; △ , Ref. 22. Estimated values are joined by lines: -------, Poenaru *et al.*[27]; -.-.-.-, Shi and Swiatecki[20] (see note on Table II); ———, present estimates.

FIG. 2 - ^{14}C-decay half-life plotted against mass number of radon isotopes. Estimates of the present work are joined by solid lines, and the results by Poenaru *et al.*[17] by a dashed line.

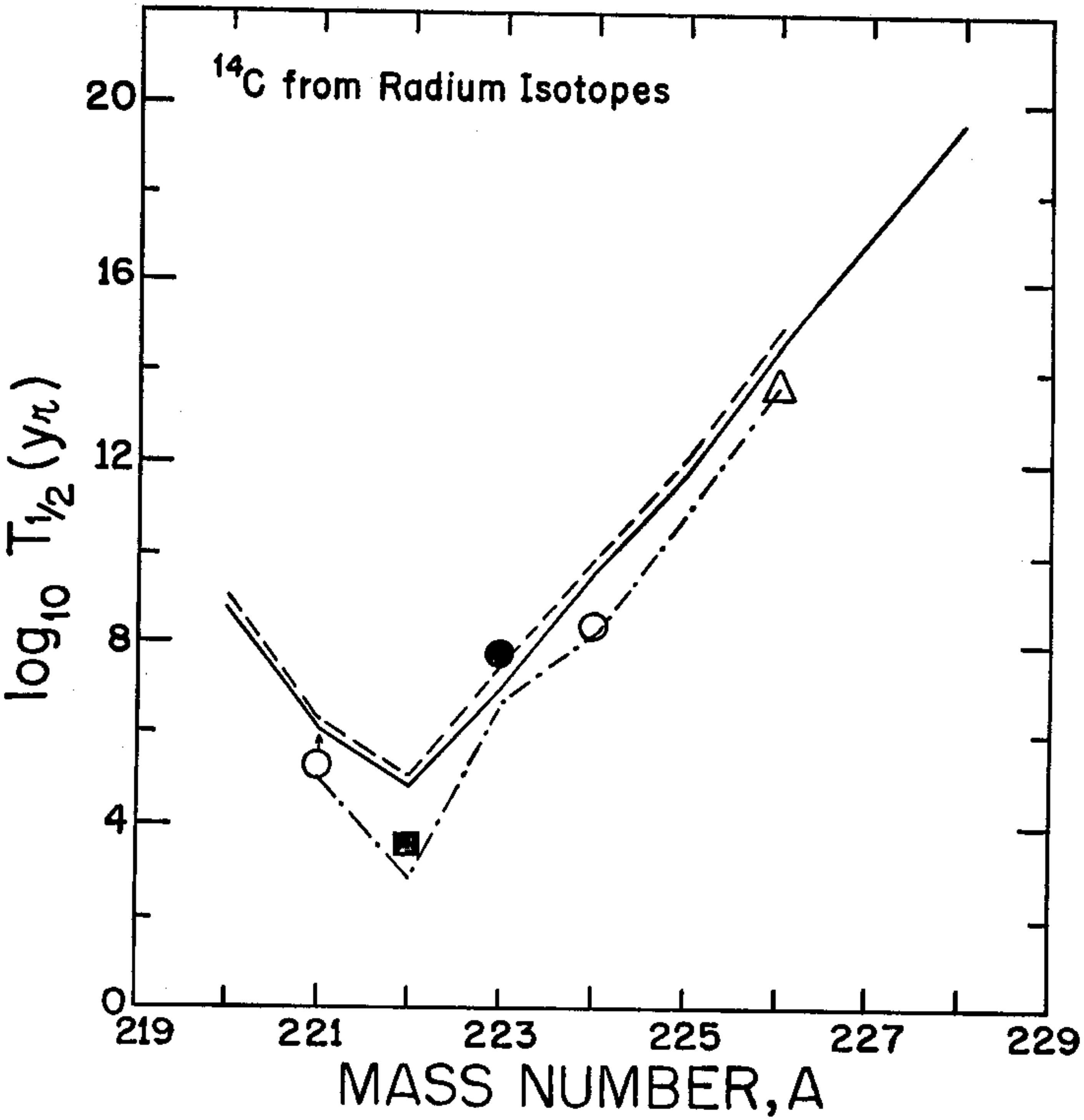
^{14}C from Radium Isotopes
$\log_{10} T_{1/2}$ (yr)
0
4
8
12
16
20
219
221
223
225
227
229
MASS NUMBER, A

FIG. 1

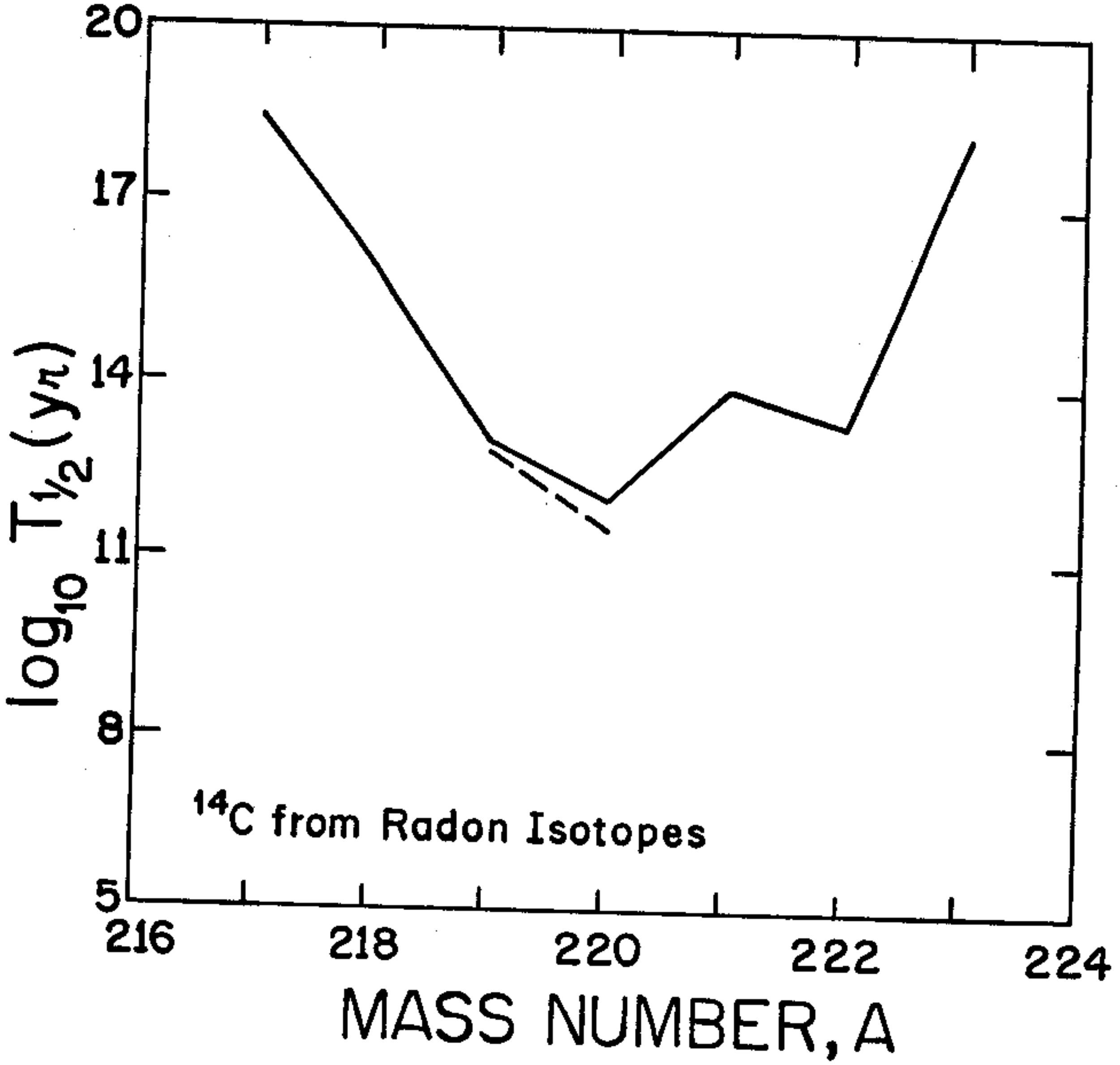
20
17
14
11
8
5
216
218
220
222
224
$\log_{10} T_{1/2}$ (yr)
MASS NUMBER, A
^{14}C from Radon Isotopes

FIG. 2

References, Footnotes and Comments

[†] On leave of absence from the Brazilian Comissão Nacional de Energia Nuclear-CNEN, Rio de Janeiro.

1. H.G. de Carvalho, J.B. Martins, I.O. de Souza and O.A.P. Tavares: Emissão Espontânea de Íons Pesados pelo Urânio. Communicated to the Brazilian Academy of Sciences on September 23 , 1975; An. Acad. brasil. Ciênc. 47(3/4), 567(1975).

2. I.O. de Souza: Meia-Vida de Fissão Espontânea do Urânio-238 e uma Provável Nova Forma de Radioatividade. MS Thesis presented at the Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro, September 1975.

3. H.G. de Carvalho, J.B. Martins, I.O. de Souza and O.A.P. Tavares: Spontaneous Emission of Heavy-Ions from Uranium. An.Acad. brasil. Ciênc. 48(2),205(1976); also as Report A0013/76, Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro,May 1976.

4. H.G. de Carvalho, J.B. Martins and O.A.P. Tavares: Experimental Evidences of Spontaneous and Low-Energy Induced Emission of Heavy Ions from Even-Even Heavy Nuclei. Report A0025/77 , Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro, June 1977 (unpublished).

5. O.A.P. Tavares: Alta Assimetria de Massa na Desintegração Espontânea e Induzida de Núcleos Pesados. Doctoral Thesis presented at the Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro, December 1978.

6. A. Săndulescu and W. Greiner: Mass asymmetry in fission, fusion and mass transfer due to the fragmentation in valleys . J. Phys. G: Nucl. Phys. 3(8),L189(1977).

7. A. Săndulescu and W. Greiner (private communication, July 1978); A. Sandulescu, H.J. Lustig, J. Hahn and W. Greiner: High mass-asymmetry distributions of fissioning nuclei. J. Phys. G : Nucl. Phys. 4(11),L279(1978).

8. A. Săndulescu, D.N. Poenaru and W. Greiner: New type of decay of heavy nuclei intermediate between fission and α decay.Fiz. Elem. Chastits At. Yadra 11,1334(1980) [Sov. J. Part. Nucl.

11(6),528(1980)]; see also references quoted therein.

9. D.N. Poenaru, M. Ivascu and A. Săndulescu: Alpha decay as a fission-like process. J. Phys. G: Nucl. Phys. 5(10), L169 (1979).

10. D.N.Poenaru, M. Ivascu and A. Săndulescu: La radioactivité alpha étudié par les méthodes de la fission. J. Physique Lett. 40(18),L465(1979).

11. D.N. Poenaru and M. Ivascu: Estimation of the alpha decay half-lives. J. Physique 44,791(1983).

12. H.J. Rose and G.A. Jones: A new kind of natural radioactivity. Nature 307,245(1984).

13. D.V. Aleksandrov, A.F. Belyatskiĭ, Yu. A. Glukhov, E. Yu. Nikol'skiĭ, B.G. Novatskiĭ, A.A. Ogloblin, and D.N. Stepanov : Observation of the spontaneous emission of ^{14}C nuclei from ^{223}Ra. Pis'ma Zh. Eksp. Teor. Fiz. 40(4),152(1984)[JETP Lett. 40(4),909(1984)].

14. S. Gales, E. Hourani, M. Hussonnois, J.P. Schapira, L. Stab, and M. Vergnes: Exotic Nuclear Decay of ^{223}Ra by Emission of ^{14}C Nuclei. Phys. Rev. Lett. 53(8),759(1984).

15. P.B. Price, J.D. Stevenson, S.W. Barwick and H.L. Ravn: Discovery of Radioactive Decay of ^{222}Ra and ^{224}Ra by ^{14}C Emission. Phys. Rev. Lett. 54(4),297(1985).

16. A. Săndulescu, D.N. Poenaru, W. Greiner, and J.H. Hamilton : Comment on "Exotic Nuclear Decay of ^{223}Ra by Emission of ^{14}C Nuclei". Phys. Rev. Lett. 54(5),490(1985).

17. D.N. Poenaru, M. Ivascu, A. Săndulescu and W. Greiner: Spontaneous emission of heavy clusters. J. Phys. G: Nucl. Phys. 10,L183(1984).

18. W. Greiner, M. Ivascu, D.N. Poenaru, and A. Săndulescu: On Exotic Nuclear Decay of ^{223}Ra by Emission of ^{14}C Nuclei. Z. Phys. A320,347(1985).

19. Yi-Jin Shi and W.J. Swiatecki: Theoretical Estimates of the Rates of Radioactive Decay of Radium Isotopes by ^{14}C Emission. Phys. Rev. Lett. 54(4),300(1985).

20. Yi-Jin Shi and W.J. Swiatecki: Estimates of Radioactive Decay by the Emission of Nuclei Heavier than α-Particles. Nucl. Phys. A438,450(1985).

21. S.W. Barwick, P.B. Price, and J.D. Stevenson: Discovery of Radioactive Decay of ^{232}U by ^{24}Ne Emission. Phys. Rev. C31 (5),1984(1985).

22. E. Hourani, M. Hussonnois, L. Stab, L. Brillard, S. Gales , and J.P. Schapira: Evidence for the Radioactive Decay of ^{226}Ra by ^{14}C Emission. Phys. Letters 160B(6),375(1985).

23. A. Săndulescu, Yu. S. Zamyatnin, I.A. Lebedev, B.F. Myasoedov, S.P. Tretyakova, and D. Hasegan: Ne Emission by Spontaneous Decay of ^{231}Pa. JINR Rapid Communications, No.5-84(1984).

24. S.P. Tretyakova, A. Săndulescu, Yu. S. Zamyatnin *et al.*: Ne Emission by Spontaneous Decay of ^{233}U. JINR Rapid Comunnications No. 7-85(1985).

25. W. Kutschera, I. Ahmad, S.G. Armato III, A.M. Friedman, J.E. Gindler, W. Henning, T. Ishii, M. Paul, and K.E. Rehm: Spontaneous ^{14}C emission from ^{223}Ra. Phys.Rev. C32,2036(1985).

26. S.P. Tretyakova, A. Săndulescu, V.L. Micheev, D. Hasegan, I. A. Lebedev, Yu. S. Zamyatnin, Yu. S. Korotkin, and B.F. Myasoedov: On the Spontaneous Emission of Clusters by the ^{230}Th, ^{237}Np and ^{241}Am Nuclei. JINR Rapid Communications No. 13-85 (1985).

27. D.N. Poenaru, M. Ivascu, A. Săndulescu, and W. Greiner: Atomic Nuclei Decay Modes by Spontaneous Emission of Heavy Ions. Phys. Rev. C32(2),572(1985); also as JINR Dubna Report E4-84 -811(1984).

28. D.N. Poenaru, M. Ivascu, A. Săndulescu, and W. Greiner: New natural radioactivities by emission of heavy ions. Central Institute of Physics, Bucharest, Report NP-41(1985).

29. D.N. Poenaru, M. Ivascu, A. Săndulescu, and W. Greiner: Decay Modes of Nuclei. Central Institute of Physics, Bucharest, Report NP-46(1985).

30. D.N. Poenaru, W. Greiner, M. Ivascu, and A. Săndulescu: Heavy cluster decay of trans-zirconium "stable" nuclides. Phys.Rev. C32,2198(1985).

31. See, for instance, P. Marmier and E. Sheldon: *Physics of Nuclei and Particles* (Academic Press, Inc., New York, 1969) , vol. I, p. 298.

32. A.H. Wapstra and G. Audi: The 1983 Atomic Mass Evaluation . Nucl. Phys. A432,1(1985).

33. W.D. Myers: Geometric Properties of Leptodermous Distributions with Applications to Nuclei. Nucl. Phys. A204,465(1973).

34. G. Süssmann: Description of the Nuclear Surface by Moments . Z. Physik A274,145(1975).

35. To save original notation, we retained the symbol Q to denote the root-mean-square radius. This, of course, should not be confused with the Q-value introduced in the assumption i) and used throughout.

36. W.D. Myers and K. Schmidt: An Update on Droplet-Model Charge Distributions. Nucl. Phys. A410,61(1983); see also B.A.Brown, C.R. Bronk, and P.E. Hodgson: Systematics of nuclear RMS charge radii. J. Phys. G: Nucl. Phys. 10,1683(1984).