Theoretical Estimates of the Rates of Radioactive Decay of Radium Isotopes by ^{14}C Emission

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The measured branching ratios for the decays of ^{222,223,224}Ra by alpha or ^{14}C emissions can be accounted for within a factor of 10 in terms of the ratios of Gamow penetrabilities through potential-energy barriers consisting of a Coulomb repulsion, the nuclear proximity attraction, and an interpolation between the configuration of tangent fragments and the configuration of the parent nucleus.

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In this note we would like to point out that, in the spirit or Sandulescu and co-workers,[1] the branching ratios between α-particle and ^{14}C radioactivity, reported by Price *et al.*[2] (see also Rose and Jones[3] and Gales *et al.*[4]), can be interpreted with reasonable accuracy (within a power of 10, or so) by a quantum tunneling calculation, *provided* a realistic estimate of the potential-energy barrier is used.

To construct the deformation-energy barrier of a Ra nucleus disintegrating into a pair of fragments (Rn + α or Pb + ^{14}C), we added to the Coulomb repulsion between the fragments the nuclear proximity attraction of Blocki and co-workers.[5,6] After contact of the fragments, when the approximation of two spherical fragments ceases to be applicable, we used for the deformation energy a smooth, power-law interpolation between the contact configuration and the configuration of the parent nucleus (where the deformation energy is zero by definition). The explicit formula for the deformation energy $V(L)$ is thus as follows:

$$V(L) = -Q + \frac{Z_1 Z_2 e^2}{r} + V_P(z) \text{ for } L > L_c, \quad (1)$$

$$V(L) = a(L - L_0)^\nu \text{ for } L_0 < L < L_c. \quad (2)$$

In the above, L is the major axis (i.e., the overall length) of the configuration in question, L_c refers to its value at contact (equal to the sum of the fragment diameters), L_0 is the diameter of the parent nucleus, Q is the energy released in the disintegration, Z_1 and Z_2 are the atomic numbers of the fragments, r is the distance between fragment centers, and z is the distance between the near surfaces of the fragments. V_P is the proximity potential, given by

$$V_P(z) = K\Phi(z/b), \quad (3)$$

where

$$K = 4\pi \bar{R} \gamma b \quad (4)$$

and Φ is the universal nuclear proximity function of Ref. 5, to which an approximation, given in Ref. 6, reads as follows:

$$\Phi(\zeta) \approx -4.41 e^{-\zeta/0.7176} \text{ for } \zeta \geqslant 1.9475, \quad (5)$$

$$\Phi(\zeta) \approx -1.7817 + 0.9270\zeta + 0.01696\zeta^2 - 0.05148\zeta^3 \text{ for } 0 \leqslant \zeta \leqslant 1.9475. \quad (6)$$

In the above, $\zeta = z/b$, b is the width (diffuseness) of the nuclear surface ($b \approx 1$ fm), and γ is the specific nuclear surface tension, for which we used the expression given in Ref. 5:

$$\gamma = 0.9517[1 - 1.7826(N - Z/A)^2] \text{ MeV/fm}^2, \quad (7)$$

where N, Z, A are the neutron, proton, and mass numbers of the parent nucleus. The reduced radius $\bar{R}$ is given by

$$\bar{R} = C_1 C_2/(C_1 + C_2), \quad (8)$$

where C_i are the central radii of the fragments, related to the effective sharp radii R_i by

$$C \approx R - \frac{b^2}{R}. \quad (9)$$

Reference 5 gives the following semiempirical formula for R in terms of the mass number A

$$R = 1.28A^{1/3} - 0.76 + 0.8A^{-1/3}. \quad (10)$$

Disregarding ground-state deformations, the value of L_0 in Eq. (2) is given by $2C$, where C is the central radius of the parent nucleus, calculated also according to Eqs. (9) and (10).

The requirement of a smooth fit for $V(L)$ at $L = L_c$ defines the coefficients a and ν in Eq. (2) as

$$\nu = (V_c'/V_c)(L_c - L_0), \quad (11)$$

$$a = V_c/(L_c - L_0)^\nu, \quad (12)$$

where the subscript "c" refers to contact, and

$$V_c' = \left.\frac{dV}{dL}\right|_c = -\frac{Z_1 Z_2 e^2}{r_c^2} + K \frac{d}{dz}\Phi\left.\left(\frac{z}{b}\right)\right|_c = -Z_1 Z_2 e^2/r_c^2 + 0.9270K/b. \quad (13)$$

Here, r_c is the center separation at contact and 0.9270 is the dimensionless derivative of Φ at $\zeta = 0$.

The standard WKB expression for the Gamow penetrability factors was used both for the α particle and ^{14}C, and the effective mass in the disintegration degree of freedom was taken simply as the reduced mass M_r of the fragments, since most of the barrier (also in the case of ^{14}C) corresponds to separating fragments beyond scission.

The above formulae, including all relevant nuclear parameters, were taken from Refs. 4 and 5, without any adjustments. The resulting ratios of the penetrability factors for α and ^{14}C emissions from ^{222}Ra, ^{223}Ra, and ^{224}Ra were found to be 1.678×10^{-9}, 6.895×10^{-9}, and 6.150×10^{-11}, respectively. The ratios of these numbers to the measured branching ratios are 4.5, 11.3, and 1.43. Since the Gamow penetrability factors (for ^{14}C) are in the range 10^{32}–10^{38}, agreement within about a factor of 10 implies an accuracy in the estimated deformation-energy barriers of a few percent. Despite this relatively close correspondence, one should not underestimate the uncertainties of the present simple theory. In particular, the treatment of the prescission part of the barrier (for $L < L_c$) is schematic and uncertain. First, the effective mass for barrier penetration in this region is expected to be different from M_r. (In a hydrodynamical calculation, the effective mass tends to be less than M_r for small deformations. On the other hand, quantum effects tend to increase the effective mass above the hydrodynamical value, sometimes by considerable factors.) Second, the power-law interpolation [Eqs. (11) and (12)], adopted for algebraic convenience, is one of several possible recipes and has no theoretical foundation. For the alpha decay of ^{222}Ra the prescission part contributes 2.1% to the penetrability integral (which is worth 23.6 powers of ten). If the prescission part were, in fact, 50% higher (i.e., 3.15% instead of 2.1%) the alpha half-life would be increased by 0.246 powers of 10, i.e., by a factor of 1.8. In the case of ^{222}Ra decay by ^{14}C emission, the prescission contribution is 11.3% and the penetrability integral is worth 32.2 powers of 10. If the prescission contribution were 50% higher the lifetime would be increased by 1.82 powers of 10, i.e., by a factor of 66. For the emission of even heavier fragments such prescission uncertainties would become progressively greater.

The reasons why, in the present calculations, the penetrability ratios are several orders of magnitude smaller than for a pure Coulomb barrier cut off at a contact distance parametrized as $r_0(A_1^{1/3} + A_2^{1/3})$, as in Refs. 2 and 3, are the inclusion of the nuclear proximity interaction and the use of more realistic expressions for the nuclear radii [Eqs. (9) and (10)].

A fuller account of these calculations, including estimates of branching ratios for other exotic decays, will be published separately.[7]

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