Discovery of Radioactive Decay of ^{222}Ra and ^{224}Ra by ^{14}C Emission

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Using the ISOLDE on-line isotope separator at CERN to produce sources of ^{221}Fr, ^{221}Ra, ^{222}Ra, ^{223}Ra, and ^{224}Ra, and using polycarbonate track-recording films sensitive to energetic carbon nuclei but not to alpha particles, we have discovered two new cases of the rare ^{14}C decay mode—in ^{222}Ra and ^{224}Ra. Our results for branching ratios, B, relative to alpha decay are for ^{221}Fr and ^{221}Ra, $B < 4.4\times10^{-12}$; for ^{222}Ra, $B = (3.7 \pm 0.6)\times10^{-10}$; for ^{223}Ra, $B = (6.1 \pm 1.0)\times10^{-10}$; for ^{224}Ra, $B = (4.3 \pm 1.2)\times10^{-11}$.

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Rose and Jones[1] recently reported the discovery of a new mode of radioactive decay in which a ^{223}Ra nucleus emits a 29.9-MeV ^{14}C nucleus with a branching ratio relative to alpha decay of $(8.5 \pm 2.5)\times10^{-10}$. They observed 19 counts in 383 days. Using a magnetic spectrometer to reject the enormous background of alpha particles, Gales *et al.*[2] confirmed the discovery. Both groups use silicon ΔE and total E detectors to measure charge and energy. Although an examination of Q values and ratios of penetration factors for heavy-particle emission relative to alpha emission indicates that ^{223}Ra is the most favorable case, there are a number of other nuclides with $Z \geqslant 87$ for which modes of radioactive decay by emission of particles heavier than alpha particles might also be detectable. Study of the systematics of such exotic decay modes might tell us about the validity of intranucleus clustering concepts and provide us with improved knowledge of internucleus potentials and extremely asymmetric fission phenomena.

We produced radioisotopes in spallation reactions by bombarding a 55 g/cm^2 thorium target (in the form of ThC_2 powder) with 2.6 μA of 600 MeV protons from the CERN synchrocyclotron. The temperature of the target was held at $\sim 2100\,°C$ so that most of the elements produced in nuclear reactions diffused out of the target. Radium and francium nuclides were selectively ionized in a high-temperature surface-ionization source.[3] The beam extracted from the ion source was separated into its constituent atomic masses by the ISOLDE electromagnetic isotope separator. The 0.2 to 1.3 nA 60 keV beams of the masses 221, 222, 223, and 224 were implanted in the bases of four cylindrical aluminum cups. We deployed polycarbonate track-recording films at the tops and sides of the cups to record tracks of particles with $Z \geqslant 5$ emitted in vacuum from the sources during and after their deposition. Such plastic track detectors[4] are ideally suited for the selective detection and identification of a few highly ionizing particles in a huge background of less highly ionizing particles. Figure 1 is a sketch of a cup showing the beam entry hole and the locations of the plastic detectors and the implanted atoms.

We determined the number of collected atoms in two ways: (1) by integration of a Faraday cup reading of the beam intensity, which we corrected for the atoms lost on the entrance aperature of the aluminum cup, as determined by beta counting; (2) by gamma-ray counting of the samples by means of an intensity-calibrated Ge(Li) spectrometer. The two methods agreed within 10%. For each cup about half of the atoms deposited were radium and half were francium with the same mass number. For $A = 221$ both Fr and Ra are alpha emitters, whereas for $A = 222$, 223, and 224, all the Fr beta decays to an alpha-emitting Ra isotope. In each cup contamination from atoms with neighboring mass numbers was less than 10^{-3}.

The polycarbonate detectors consisted of a 125 μm Tuffak sheet covered with a 10 μm Makrofol sheet. We found it unnecessary to use tracks in the Makrofol to assist in the identification. We etched the Tuffak 8 h at 70 °C in 6.25N NaOH solution and scanned an area corresponding to a solid angle of ~ 2 sr. All of

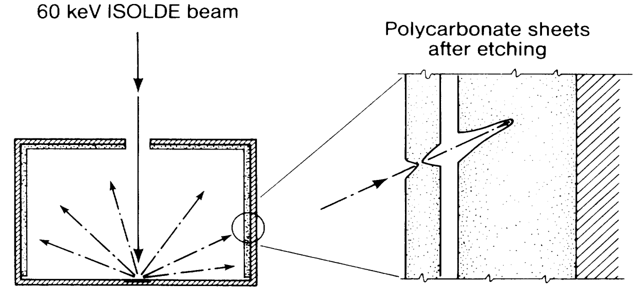

FIG. 1. Radium isotope collector cup, lined with polycarbonate sheets for detection of energetic particles with $Z > 5$ emitted in radioactive decay.

the carbon tracks were on trajectories that pointed back to the position of the radioactive source at the center of the base of each cup.

We used measurements of etched-track diameters and lengths to measure charge and range, respectively. We based the charge identifications on calibrations in which plastic films were irradiated both with high densities of alpha particles and with beams of boron, carbon, and neon ions at the Lawrence Berkeley Laboratory Superhilac. Figure 2 compares track etch rate as a function of residual range for carbon nuclei emitted from ^{223}Ra with the results of the Superhilac calibrations shown as dashed lines. The magnitudes of the lines at B, C, and Ne were determined to within 4%; the lines at N, O, and F are interpolations based on a restricted energy-loss model of track production. At the highest alpha-particle doses such as were received by the detectors for the sources with $A = 221$ and 224, the positions of the charge calibration lines were slightly lower than in Fig. 2. The positions of the data for ^{14}C nuclei emitted from the source with $A = 224$ were consistent with the lowered position of the carbon calibration line. No ^{14}C events were detected from the mass 221 source.

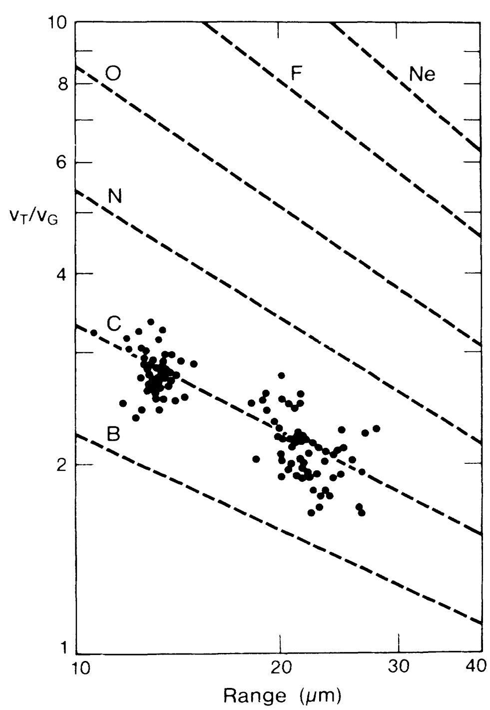

FIG. 2. Comparison of signal of ^{14}C nuclei emitted from ^{223}Ra with calibrations (dashed lines) obtained with heavy ions at Lawrence Berkeley Laboratory Superhilac. Ratio of etching rate along track to general etching rate, v_T/v_G, is plotted as a function of residual range. Measurement at two different points along the trajectory are made for each ^{14}C nucleus.

Figure 3 compares histograms of measured ranges with expected values calculated from a range-energy table. In determining ranges we took into account the portion of the range lost due to implantation in the aluminium base and due to penetration through the Makrofol layer and the portion of Tuffak etched away. The carbon tracks were readily detectable above the background, which consisted of a high density of short, barely detectable tracks of carbon and oxygen recoil atoms produced in elastic collisions of alpha particles in the plastic. Fortunately, in each case the alpha energies were sufficiently small that the maximum recoil ranges were much shorter than the ranges of the ^{14}C nuclei.

Table I summarizes our results. Although we did not directly determine the mass number of the emitted carbon nuclei, the measured mean ranges agree extremely well with the ranges calculated from the Q values for ^{14}C, and are inconsistent with ranges calcu-

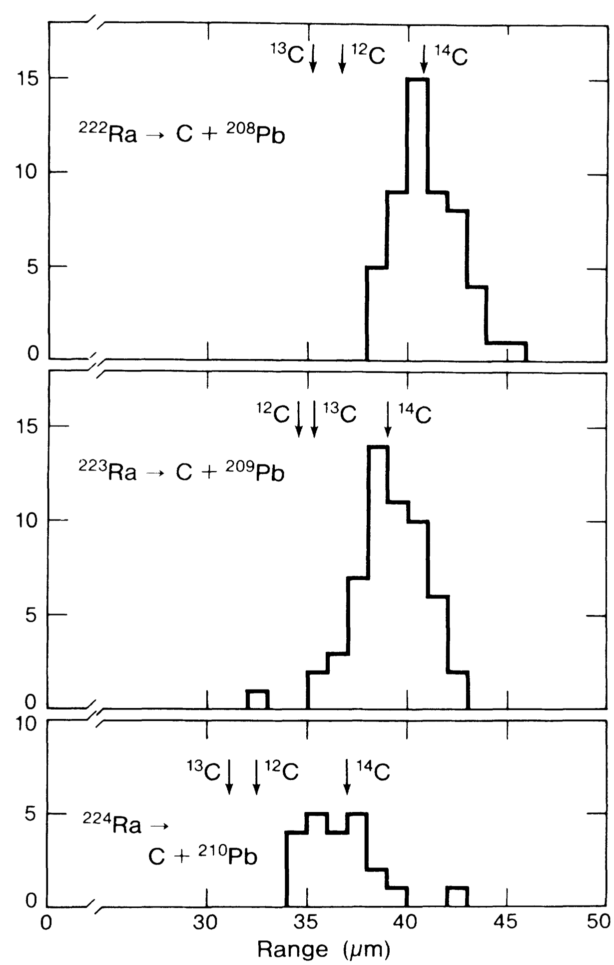

FIG. 3. Measured range distributions for ^{14}C nuclei emitted from ^{222}Ra, ^{223}Ra, and ^{224}Ra, compared with ranges calculated from Q values.

TABLE I. ^{14}C decay of radium isotopes.

Z	A	Q (MeV)	No. of Ra atoms	Obs. decays	Mean range (μm) calc.	Mean range (μm) meas.	Measured B $\equiv \lambda(^{14}C)/\lambda(\alpha)$	Ratio of Gamow factors[a]	Calc. B (Ref. 5)	Calc. B (Ref. 6)
87	221	31.26	2.6×10^{12}	0	38.2	...	$< 4.4\times10^{-12}$(90%C.L.)	8.7×10^{-13}	8.0×10^{-12}	1.4×10^{-14}
88	221	32.39	2.6×10^{12}	0	40.0	...	$< 4.4\times10^{-12}$(90%C.L.)	6.5×10^{-13}	8.2×10^{-12}	8.0×10^{-13}
88	222	33.05	7.8×10^{11}	52	41.1	41.0 ± 0.2	$(3.7\pm0.6)\times10^{-10}$	7.5×10^{-11}	1.7×10^{-9}	2.5×10^{-12}
88	223	31.87	4.7×10^{11}	56	39.2	39.2 ± 0.2	$(6.1\pm1.0)\times10^{-10}$	$\equiv 6.1\times10^{-10}$	6.9×10^{-9}	2.5×10^{-9}
88	224	30.53	2.4×10^{12}	22	37.1	36.5 ± 0.4	$(4.3\pm1.2)\times10^{-11}$	1.4×10^{-11}	6.1×10^{-11}	1.3×10^{-12}

[a]For square-well potential and $R = r_0(A_d^{1/3} + A_x^{1/3})$; $r_0 = 0.98$ fm.

lated from Q values for ^{13}C and ^{12}C. The corresponding mean energies computed from the range-energy relation agree with the expected ^{14}C kinetic energies to within 0.4 MeV, which is as good as was attained in Ref. 2.

Column 8 of the table lists our measured branching ratios for ^{14}C decay relative to alpha decay. The errors take into account the uncertainty in the number of collected Ra atoms as well as the statistical error. The branching ratio for ^{223}Ra agrees well with the results of Rose and Jones[1] and Gales *et al.*[2] The branching ratios for ^{222}Ra and ^{224}Ra are lower by factors of about 2 and 10 than that for ^{223}Ra.

Column 9 gives the ratio of Gamow penetration factors for ^{14}C emission relative to alpha emission in the approximation of a Coulomb potential plus square-well nuclear potential with a width $R = r_0(A_1^{1/3} + A_2^{1/3})$, where A_1 and A_2 are the mass numbers of the daughter and emitted particle. Choice of a value of r_0 in the customary interval 1.1 to 1.4 fm leads to values of B several orders of magnitude too high.[1] We chose the value $r_0 = 0.98$ fm simply because it led to a branching ratio for ^{14}C decay relative to alpha decay that agreed with the measured ratio, without the need for a preformation factor. Such a small value of r_0 is qualitatively within the spirit of the discussion by Shi and Swiatecki[5] of the appropriate value of nuclear radius to use in a barrier-penetration calculation. They have shown that more realistic choices of the nuclear potential and nuclear radii lead to branching ratios in order-of-magnitude agreement with our data, without the need for any adjustable parameter. Column 10 shows their values of B.[5] Column 11 shows the values of B independently estimated by Poenaru *et al.*,[6] using an extension of the fission theory of alpha decay. Their values of B fall off much more rapidly on either side of ^{223}Ra than do those in columns 9 and 10.

It is interesting to note that nuclides in the vicinity of radium are believed to have a stable octupole (pear-shaped) deformation in the ground state,[7-9] from which one can depict the ^{14}C as coming from the small end of the pear and the doubly magic ^{208}Pb or nearly magic ^{209}Pb or ^{210}Pb as coming from the approximately spherical large end of the pear.

It should be possible to use the track-etching technique to find additional cases of exotic radioactivity, including emission of particles heavier than ^{14}C. By using less sensitive plastics such as polyesters[3] one may be able to observe branching ratios as low as or even lower than 10^{-13} for emission of particles with $Z \geqslant 8$. With knowledge of the dependence of the branching ratio on both the charge of the emitted particle and the Q values, one should be able to distinguish between the concepts of barrier penetration by a heavy cluster and of superasymmetric fission, as discussed in Refs. 5 and 6.

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