PHYSICAL REVIEW C VOLUME 31, NUMBER 5 MAY 1985

Radioactive decay of ^{232}U by ^{24}Ne emission

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(Received 26 December 1984)

Using polyethylene terephthalate track-recording films, which are sensitive only to particles with $Z > 6$, we have detected for the first time the exotic radioactive decay mode in which an energetic ^{24}Ne nucleus is emitted. For ^{232}U the branching ratio for ^{24}Ne relative to alpha decay is $(2.0 \pm 0.5) \times 10^{-12}$. We point out that the spontaneous fission rates reported for ^{232}U and several other nuclides based on early measurements of energetic pulses in ionization chambers may actually have been rates for emission of ^{24}Ne or other heavy clusters.

The discovery by Rose and Jones[1] of the spontaneous emission of ^{14}C from ^{223}Ra has confirmed the prediction by Sandulescu *et al.*[2] of rare modes of radioactive decay intermediate between alpha decay and fission. Gales *et al.*,[3] Alexandrov *et al.*,[4] and Price *et al.*[5] have confirmed the measurement by Rose and Jones, and Price *et al.* have discovered two additional cases of ^{14}C emission—from ^{222}Ra and ^{224}Ra. Poenaru *et al.*[6] and Shi and Swiatecki,[7] using semiempirical potentials for the separation of two spherical nuclei with unequal masses, have calculated that the rare decay mode, termed superasymmetric fission, involves emission of a specific nuclide with a very high Q value, leading to a tightly bound daughter nucleus with a branching ratio usually less than 10^{-10} relative to alpha decay. For elements heavier than radium the most likely nuclide is predicted by both groups to be heavier than ^{14}C, such that in most cases the daughter is close to the doubly magic ^{208}Pb. Because of the extreme sensitivity of calculated decay rates to the shape and magnitude of the superasymmetric fission barrier, advances in our understanding of the process will necessitate the discovery of additional decay modes involving emission of particles over a wide range of masses. In this Rapid Communication we report the discovery of ^{24}Ne decay of ^{232}U.

In their study of ^{14}C emission from various radium isotopes, Price *et al.*[4] used polycarbonate detectors,[8] which are sensitive to particles with $Z > 2$, to measure the charge and range of carbon ions at alpha particle backgrounds up to 6×10^{10} cm^{-2}. The maximum tolerable alpha dose was determined by the overlap of short tracks of carbon and oxygen recoils produced in elastic collisions with alpha particles. In the present study we used a much less sensitive track-recording polyethylene terephthalate film called Cronar, which records tracks of ions only with $Z > 6$. From our present experience, in which we exposed Cronar to 7×10^{10} alphas/cm^2 (Fig. 1), we estimate that the limiting alpha particle dose for this plastic is $\sim 2 \times 10^{11}$ cm^{-2}. With the requirement that at least ten tracks of an ion $Z > 6$ be recorded, and with a hemispherical array of Cronar at a distance of 7.5 cm, this dose permits a limiting branching ratio of $\sim 10^{-14}$ to be detected.

Guided by predictions[6,7] of a favorable branching ratio for ^{24}Ne decay, we obtained a 0.5 ± 0.05 millicurie source of ^{232}U, prepared by Isotope Products on a 2.5 cm disk covered by a 100 μg/cm^2 gold film to prevent the escape of recoil nuclei. We exposed a hemispherical array of Cronar film for one month to the ^{232}U source inside a vacuum chamber at a pressure of 0.01 torr. During this time the activity of the daughter ^{228}Th slowly growing into the freshly prepared ^{232}U source was negligible.

We etched the Cronar for eight hours at 70 °C in 6.25 normal NaOH solution, scanned 200 cm^2 of surface in transmitted light (Fig. 1), and located 24 etched tracks of ^{24}Ne ions. In an additional 50 cm^2 of Cronar that had been covered with a 15 μm absorber film to slow down Ne ions, we found seven tracks of ^{24}Ne. In the uncovered Cronar we also found a small background of spontaneous fission tracks (~ 20 cm^2) whose origin we traced to the use by Isotope Products of a ^{252}Cf-contaminated evaporator to prepare the gold film on the ^{232}U source. The fission events were easily distinguished from ^{24}Ne by their short range and much higher ionization rate.

Because of the optical nonuniformity of Cronar, we made silicone replicas[8] of the surface and measured the diameters and lengths of the tracks on the replicas instead of in the Cronar. The charge and range of an energetic ion are determined from the diameter and length of the etched track.[8] Figure 2 compares our data for the ^{24}Ne tracks with calibrations in which we irradiated Cronar with ^{20}Ne ions at the Lawrence Berkeley Laboratory Superhilac and with ^{18}O ions

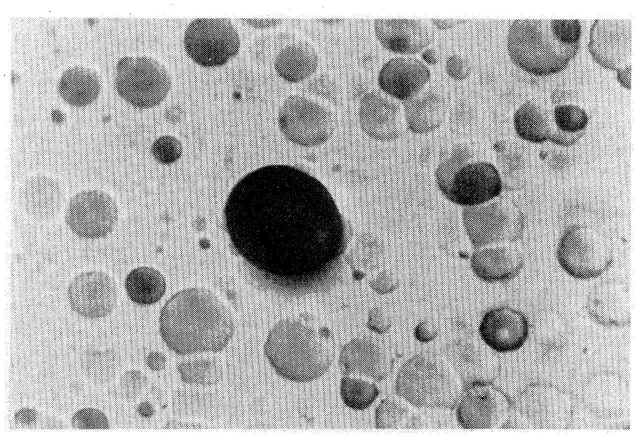

FIG. 1. Photomicrograph showing one etch pit due to a 56 MeV ^{24}Ne ion striking a Cronar detector nearly head on. About 3×10^6 alpha particles passed through this field of view.

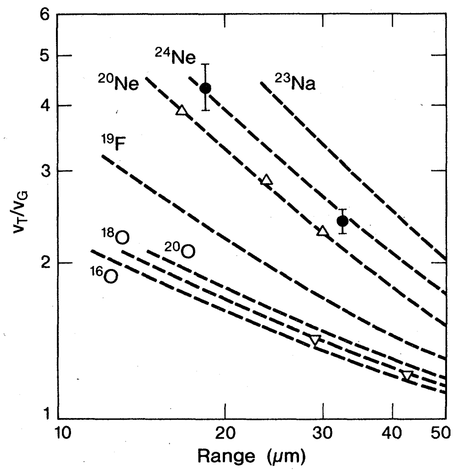

FIG. 2. Comparison of average signal of ^{24}Ne nuclei (●) emitted from ^{232}U with calibrations (dashed lines) obtained with ^{18}O (▽) and ^{20}Ne (Δ) ions at Lawrence Berkeley Laboratory accelerators. Ratio of etching rate along track to general etching rate v_T/v_G, is plotted as a function of residual range.

at the 88-inch cyclotron. The dashed lines for ions other than ^{20}Ne and ^{18}O are based on a track formation model in which the ratio of track etch rate v_T, to general etch rate v_G, is a function of restricted energy loss rate. The errors for the calibrations are smaller than the data points. The 1 σ errors in v_T/v_G for the ^{24}Ne ions are indicated by bars. The distribution of measured ranges for the 24 events in the uncovered Cronar is compared in Fig. 3 with ranges calculated for several Ne isotopes based on Q values and assuming ground states for parent, daughter, and emitted ion. Not only does ^{24}Ne give the best fit for both v_T/v_G and range; the Q values for all other Ne isotopes are much smaller than for ^{24}Ne and lead to barrier penetration factors that are more than 10 orders of magnitude smaller than for ^{24}Ne.

Table I summarizes our results. The mean energy computed from the measured mean range and a range-energy relation agrees with the calculated ^{24}Ne kinetic energy to within 0.6 out of 55.85 MeV. The measured branching ratio for ^{24}Ne emission relative to alpha decay $(2.0 \pm 0.5) \times 10^{-12}$, is quite close to that predicted by Poenaru and coworkers[6,9] The stated error includes both statistical error and uncertainty in source strength.

One can view the superasymmetric fission process in two

TABLE I. Measurements of ^{24}Ne decay of ^{232}U

Events detected	31
Q (MeV)	62.3
Calc. range in Cronar (μm)	33.2
Measured mean range (μm)	32.8 ± 0.23
Branching ratio, $\lambda(^{24}Ne)/\lambda(\alpha)$	
Measured	$(2.0 \pm 0.5) \times 10^{-12}$
Calculated (Ref. 7)	4.87×10^{-11}
Calculated (Ref. 6)	1.58×10^{-12}
Calculated (Ref. 9)	6.7×10^{-12}
Measured $\lambda_{SF}/\lambda_\alpha$ (Ref. 11)	1.2×10^{-12}

quite different ways. If it is viewed as the emission of a preformed cluster that escapes from the parent nucleus after many assaults on the barrier, one is faced with the problem of calculating the preformation probability as a function of cluster size. Poenaru *et al.*[6,9] and Shi and Swiatecki[7] view the new decay process as a type of fission in which the deformation parameter is the distance R between centers of two overlapping spheres. Shi and Swiatecki use a proximity potential and improved values of nuclear radii to calculate ratios of penetrabilities. Poenaru *et al.* derive an analytic expression for half-life based on a quadratic fit of barrier height to R in the region of overlap of the two spheres and on use of Coulomb plus centrifugal potential in the nonoverlap region. Requiring that their relation fit the extensive data on alpha decay half-lives as well as the ^{14}C decay of ^{223}Ra gave them an empirical expression for assault frequency in terms of zero-point vibrational energy: $E_v = 0.13 A_2$ MeV,[6] or more recently,[9]

$$E_v = Q[0.056 + 0.039 \exp(4 - A_2)/2.5] \text{ MeV},$$

where A_2 = mass of emitted particle. Both groups have pointed out that a more refined theory must replace the two-sphere approximation with a more complex set of deformation variables that would allow the disintegrating system to seek out a more favorable path in configuration space. The agreement to within about an order of magnitude with the ^{14}C data[1,3-5] and with our ^{24}Ne result suggests that, for Z_2 up to 10, the separating fragments are probably not deformed far from spheres during barrier penetration.

To test the models further will require measurements of branching ratios for nuclei emitting particles heavier than ^{24}Ne. Poenaru *et al.* have suggested several candidates with branching ratios greater than 10^{-13} for emission of particles with $Z > 10$. We want to point out that indirect evidence for superasymmetric fission already exists in the form of data on spontaneous fission half-lives published mainly in the 1950s when ionization chambers were used to associate fission activity with very energetic pulses. In a number of cases, especially those of importance to reactor physics, mass distributions have subsequently been measured, and neutrons emitted concurrently with fission have been detected. However, there are eight cases tabulated by Vandenbosch and Huizenga[10] in which the branching ratio for spontaneous fission relative to alpha decay is less than 10^{-10} and mass distributions have not been measured. To examine whether it is possible that the energetic pulses detected

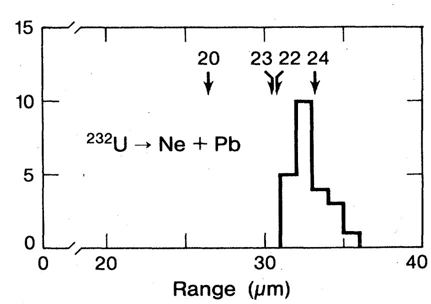

FIG. 3. Measured range distribution for ^{24}Ne nuclei emitted from ^{232}U, compared with ranges expected for various Ne isotopes calculated from Q values.

TABLE II. Half-life predicted for heavy ion emission compared with measured spontaneous fission half-life.

Decay mode	Predicted τ_x (yr)	Measured τ_{SF} (yr)	Ref.
$^{232}U \rightarrow {}^{24}Ne$	1×10^{13}	6×10^{13}	11
$^{232}Th \rightarrow {}^{26}Ne$	1×10^{22}	$\geqslant 1\times10^{21}$	12
$^{231}Pa \rightarrow {}^{24}Ne$	4×10^{14}	$\geqslant 1.1\times10^{16}$	13
$^{230}Th \rightarrow {}^{24}Ne$	3×10^{17}	$\geqslant 1.5\times10^{17}$	13
$^{233}U \rightarrow {}^{25}Ne$	9×10^{15}	1.2×10^{17}	14
$^{234}U \rightarrow {}^{28}Mg$	2×10^{17}	1.6×10^{16}	15
$^{237}Np \rightarrow {}^{30}Mg$	3×10^{18}	$\geqslant 1\times10^{18}$	16
$^{241}Am \rightarrow {}^{34}Si$	2×10^{15}	2.3×10^{14}	17

in the decay of these nuclides might have been due to emission of an energetic ion such as ^{24}Ne instead of normal fission, in Table II we compare the half-life for the most favorable mode of superasymmetric fission, calculated using the expression of Poenaru *et al.*,[9] with the spontaneous fission half-life reported in the literature. The numbers agree in each case within one order of magnitude, which is at least as good as the agreement of the data with the best fission theories.[10] In fact, our measured value for the half-life of ^{232}U for ^{24}Ne decay agrees closely with the reported value for spontaneous fission measured in 1951 by Jaffey and Hirsch.[11] Because of the presence of the spontaneously fissioning contaminant in the gold cover, we cannot yet prove that the branching ratio for spontaneous fission of ^{232}U is lower than 10^{-12}; we can only show that the events observed by Jaffey and Hirsch are consistent with our observed ^{24}Ne emission rate. The other results in Table II, if interpreted as detection of energetic heavy ions instead of conventional fission, can be used to predict superasymmetric fission half-lives. We are initiating searches with Cronar detectors for superasymmetric fission of several of these nuclides.

We are indebted to Albert Ghiorso for the use of his facilities and to A. Ghiorso, W. J. Swiatecki, and E. Segré for many helpful discussions. We thank the staff of the Superhilac and of the 88-inch cyclotron for their assistance. This work was supported by the Department of Energy.

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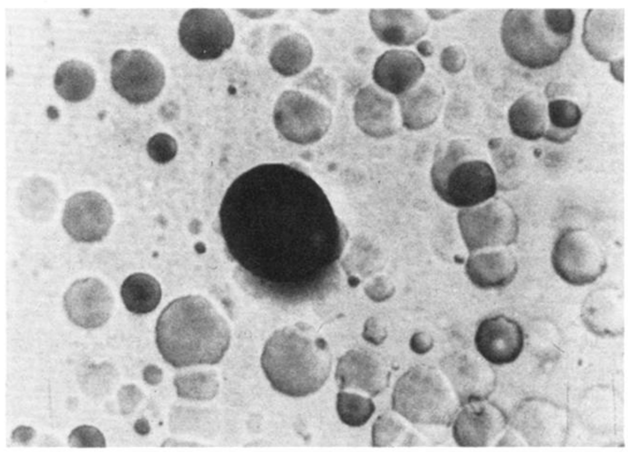

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