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Radioactive decay of ^{234}U via Ne and Mg emission

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We have identified three modes of radioactive decay of ^{234}U by barrier penetration (in addition to alpha decay): emission of a Ne ion, emission of a Mg ion, and spontaneous fission. Lifetimes are $(3.7^{+1.2}_{-0.9})\times10^{17}$, $(1.1^{+1.3}_{-0.6})\times10^{18}$, and $(1.9\pm0.1)\times10^{16}$ yr, respectively. This is the first example of a nuclide with four hadronic decay modes.

Heavy fragment radioactivity—the two-body decay of a heavy nucleus into a fragment with $Z>2$ and a nearly magic residual nucleus by a barrier penetration process —has been firmly established experimentally as a rare alternative to alpha decay.[1-11] Four nuclides (^{222}Ra, ^{223}Ra, ^{224}Ra, and ^{226}Ra) are known to decay by emission of ^{14}C and four nuclides (^{230}Th, ^{231}Pa, ^{232}U, and ^{233}U) are known to emit an isotope of Ne. Until now no nuclide has been found to emit ions heavier than Ne.

Theoretical models fall into two categories. (1) Cluster models[12,13] regard the fragment as having a certain probability of forming out of a cluster of nucleons inside the parent nucleus and of assaulting the barrier with a characteristic frequency. In one of these models only ^{14}C emission was considered; in the other an unrealistically simple square-well nuclear potential was used.[13] (2) Unified models[14-17] view alpha decay, heavy fragment radioactivity, and spontaneous fission as similar processes in which the parent nucleus spontaneously deforms through a shape barrier that includes both Coulomb and nuclear forces. They differ considerably in details: Two of them correct for nuclear ground-state deformations;[15,16] one evaluates the mass coefficient by a hydrodynamic model;[16] and one treats tunneling in internal shape degrees of freedom as a superfluid pairing phenomenon.[17] None of the models agrees with available data to within an order of magnitude,[18] and none consistently disagrees so badly with data as to be ruled out. Experimental studies of heavy fragment radioactivities over a wider range of fragment charges are needed. In this paper we report the observation of both Ne and Mg emission from ^{234}U, with comparable branching ratios.

We were guided in our choice of ^{234}U by the prediction of the unified model of Poenaru *et al.*[14] that both Ne and Mg are emitted with branching ratios of $\sim10^{-11}$ to 10^{-12} relative to alpha decay, and by the availability of large amounts of adequately pure material.

We obtained 150 mg of ^{234}U with an isotopic purity of 99.84%. To remove any contaminating spontaneous fission activities we performed the following chemical procedure. The uranium oxide sample was dissolved in aqua regia, then precipitated with NH_4OH. This precipitate was dissolved in HCl and the solution was passed through a column of Dowex 1×8 anion exchange resin. The column was washed with solutions of HCl containing HI, 9*M* HCl, and 8*M* HNO_3 before the absorbed uranium was eluted with dilute HCl. Sources were prepared on aluminum by electrodeposition of the uranium from an isopropanol solution of the nitrate, followed by heating to convert the deposited uranium salt to the oxide. The only activities we were able to observe in the final samples by alpha-particle spectrometry were ^{234}U and ^{232}U (2.0×10^{-3}% of the uranium mass).

We prepared three 39.5 cm^2 U_3O_8 sources having ^{234}U masses of 36.0, 43.9, and 42.9 mg. Three polished plates of PSK-50 phosphate glass (Schott Glass Technologies) of area 25 cm^2 were kept nearly in contact with the sources (separated by 1-mm-thick washers) in air for times of 68, 99, and 111 d. After exposure we reduced the level of radioactivity of the glass detectors by rinsing them in dilute HCl.

A previous study[19] of the sensitivities of a number of phosphate glass track detectors to various heavy ions had shown that type PSK-50, when etched in nitric acid, is ideal for the study of Ne, Mg, and Si ions with energies ~2 MeV/u. Figure 1 shows curves resulting from a calibration of PSK-50 glass at the Lawrence Berkeley Laboratory SuperHilac, using beams of ^{20}Ne, ^{24}Mg, and ^{28}Si ions. Curves for nuclides from ^{19}F to ^{28}Si were calculated from the calibration data using a restricted energy loss model for track formation. The sensitivity, s, defined as the dimensionless ratio of track etching rate to general etching rate, is a measure of the half cone angle, ϕ, of the etchpit: $s=1/\sin\phi$. As Fig. 1 shows, at a given residual range s increases strongly with Z and weakly with A of the ion. The background alpha particle fluence, $\sim10^{12}/cm^2$ in our experiment, is at least two orders of magnitude lower than the fluence at which the calibration in Fig. 1 is affected.

Our procedure in searching for various decay modes was as follows. We first etched a PSK-50 plate in 70% HNO_3 at 65 °C for 2.5 min, scanned the plate at 320× in transmitted light to locate the fully developed etched tracks of spontaneous fission, measured the lengths and mouth diameters of these tracks, and calculated the ranges and dimensionless track etch rates for the fission fragments as described below. Next we etched the plate in the same solution for an additional 14 min, rescanned it

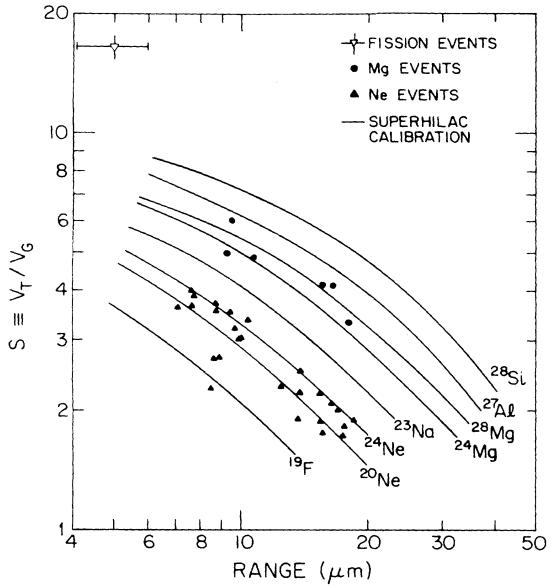

FIG. 1. Identification of ions emitted from ^{234}U as Ne and Mg. The curves are based on calibrations obtained with ^{28}Si, ^{24}Mg, and ^{20}Ne ions at Lawrence Berkeley Laboratory SuperHilac.

to locate the Mg ion tracks, which had reached the end of their range and developed rounded tips, and measured these tracks. Finally, we etched for an additional 5 min and rescanned to locate and measure the Ne ion tracks. After each stage of etching we made cellulose acetate replicas of the glass surfaces and peeled off the replicas. Measurements of track length and radius of the tip were best made by viewing a replica in reflected light. To make a complete determination of the dimensions of an etched track, from which to identify the particle's charge and energy, we used a combination of measurements in reflected and transmitted light on both the etched track in glass and the replica. We measured fission events on only one plate, the Mg ion tracks on two plates, and the Ne ion tracks on all three plates.

Figure 2 shows photomicrographs of a fission track, a Mg track, and a Ne track in a PSK-50 plate after an etching time of 20 min. PSK-50 is far too insensitive to record tracks of alpha particles; the lightest particle capable of producing an etchable track is $Z \approx 8$. One can see that the background of shallow etch pits resulting from recoil atoms struck by alpha particles is insignificant.

Measurements of the track geometry (length, minor axis at the etchpit mouth, radius of the tip of the etchpit, zenith angle) provide a value of s near the etchpit mouth and a separate value of s averaged along the entire etchpit length, as described in Ref. 20. Thus, each of the Ne and Mg events is characterized in Fig. 1 by two data points that serve to identify it. (The fission tracks have values of $s \approx 16$ that lie well above the calibration line for Si in Fig. 1; all of these events lie within the error limits for a single point.) The data of Ne and Mg provide an accurate measurement of Z and a rough estimate of A. From the experimental spread of values of s and R shown on Fig. 1, and from the calibration data, we estimate that the standard deviations for charge and mass are $\sigma_Z = 0.13$ and $\sigma_A = 1$ u.

Figure 3 shows the distribution of ranges as determined by etchpit lengths. The arrows indicate the ranges in PSK-50 glass calculated from the kinetic energy distribution for spontaneous fission of ^{234}U and from the Q values for emission of ^{28}Mg, ^{24}Ne, and ^{26}Ne, taking into account energy loss in the U_3O_8 source, assuming emission from the center of the source. (The ions ^{28}Mg, ^{24}Ne, and ^{26}Ne have relatively high Q values and are the most likely to be emitted, according to unified models.[14])

Based on the evidence in Figs. 1 and 3, we conclude that we have established the occurrence of decays by spontaneous fission, Mg emission, and Ne emission in a source consisting of 93% ^{234}U and 7% ^{232}U by activity. Our technique is not able to distinguish between the two predicted isotopes of Ne. To calculate half-life, we must evaluate the detection efficiency of PSK-50 as a function of zenith

TABLE I. Decay modes of ^{234}U.

Emitted ion	Partial half-life (yr) Prior work	This work	Predicted	No. of events	Background due to ^{232}U
4.86 MeV ^{4}He	2.45×10^5	...			
Spont. fission	$(2\pm1)\times10^{16}$ [a] $(1.4\pm0.1)\times10^{16}$ [c]	$(1.9\pm0.1)\times10^{16}$	$\sim10^{18}$ [b]	590	$\lesssim3$
52.8 MeV ^{24}Ne + 52.9 MeV ^{26}Ne	...	$(3.7^{+1.2}_{0.9})\times10^{17}$	1.8×10^{17} [d] 2.5×10^{18} [b] 1.6×10^{19} [e]	14	2.5
65.3 MeV ^{28}Mg	...	$(1.1^{+1.3}_{0.6})\times10^{18}$	1.6×10^{16} [d] 2×10^{18} [b] $>2\times10^{19}$ [e]	3	<0.18

[a]Reference 22.
[b]Reference 24.
[c]Reference 23.
[d]Reference 14.
[e]Reference 16.

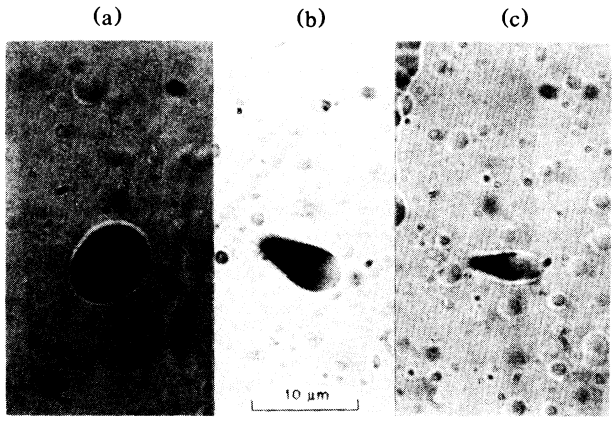

FIG. 2. Photomicrographs showing etch pits due to (a) spontaneous fission, (b) Mg emission, and (c) Ne emission from ^{234}U source.

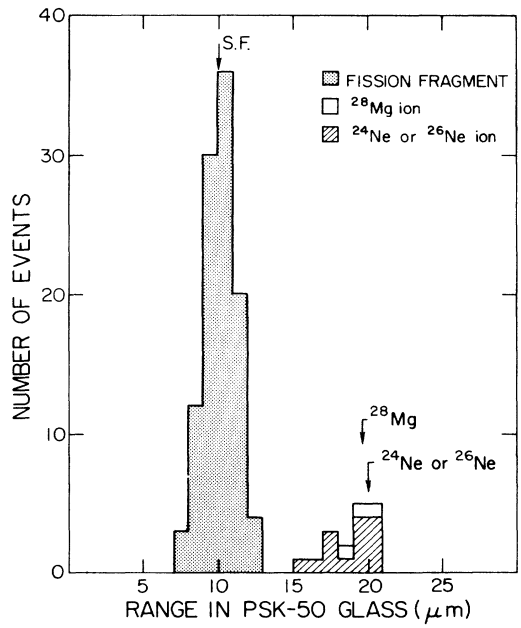

FIG. 3. Measured range distributions for fission fragments, Ne ions, and Mg ions emitted by ^{234}U source.

angle of entry for each type of ion and for fission fragments. Our measurements show that the number of Ne events per unit solid angle is uniform from 0° to about 40°, dropping rapidly to zero at larger angles, whereas the fission events have a uniform distribution out to a zenith angle of about 80°. This leads to an efficiency of 12% of 4π for Ne and of 83% of 4π for fission events (taking into account that either fission fragment can be detected). With only 3 Mg events we cannot determine detection efficiency directly from an angular distribution. Since all 3 Mg events had a zenith angle less than 40°, we assumed the same efficiency as for Ne, 12% of 4π.

Using the known spontaneous fission half-life of ^{232}U (Ref. 21), the recently measured Ne-emission half-life of ^{232}U (Ref. 7), and the absence of any Mg decays out of 24 Ne decays of ^{232}U (Ref. 7), we calculate that the ^{232}U impurity contributed $\sim$3 out of the 590 observed fission events, $\sim$2.5 out of the 14 Ne events, and less than 0.18 out of the 3 Mg events. Thus, the great majority of all three types of decay events comes from ^{234}U.

Table I summarizes the results of our measurements of half-lives for three decay modes of ^{234}U, corrected for contributions by ^{232}U. We have also listed results of previous measurements of alpha decay half-life and spontaneous fission half-life. The predicted Ne half-lives are for emission of all isotopes of Ne. Our result for the spontaneous fission half-life is about 30% larger than the most recent measurement,[23] which may be due to a slight overestimate of the maximum zenith angle for detection of fission fragments in PSK-50 glass. It is interesting that fission, Ne emission, and Mg emission have half-lives within a factor 10^2 of each other and some 11 to 13 orders of magnitude longer than the alpha-decay half-life. This is roughly consistent with predictions by Poenaru and coworkers.[14,24] Our observed ratio of partial half-lives, $\tau_{Mg}/\tau_{Ne} = 3.0 \pm 2$, is larger than the value 0.09 predicted in an early version of their unified model[14] and comparable to the value 0.8 predicted in a more recent version[24] which had been modified with parameters introduced to improve the fit to data available up to 1986.

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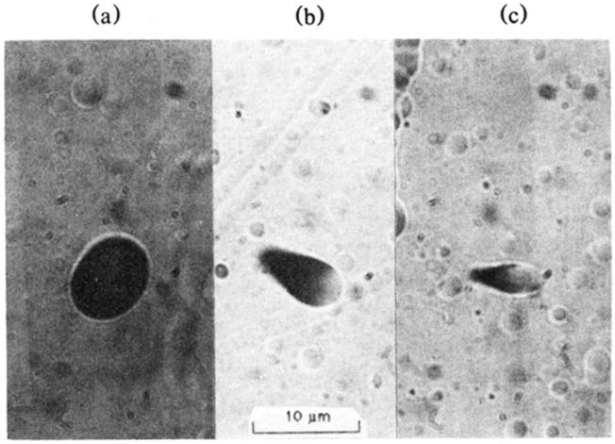

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