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Heavy-fragment radioactivity of ^{238}Pu: Si and Mg emission

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In addition to its two known hadronic decay modes—alpha decay (half-life 88 yr) and spontaneous fission (5×10^{10} yr)—we have found that ^{238}Pu also decays by emission of a Mg nucleus ($\sim2\times10^{18}$ yr) and by emission of a Si nucleus ($\sim6\times10^{17}$ yr). This extends to four (the other two being ^{14}C emission and Ne emission) the number of known modes of decay by emission of a fragment of mass between that of an alpha particle and that resulting from fission. The branching ratios relative to alpha decay are the lowest yet observed: $\sim6\times10^{-17}$ for Mg emission and 1.4×10^{-16} for Si emission.

Heavy-fragment radioactivity is the two-body decay, by barrier penetration, of a heavy nucleus into a fragment with $Z>2$ and a nearly magic residual nucleus.[1] Four nuclides (^{222}Ra, ^{223}Ra, ^{224}Ra, and ^{226}Ra) are known[2–4] to decay by emission of ^{14}C; four nuclides (^{230}Th, ^{231}Pa, ^{232}U, and ^{233}U) are known[5–8] to emit Ne but no other intermediate-mass species; and ^{234}U is known[9] to emit both Ne and Mg (as well as alpha particles and binary fission). The rough rule of thumb that the most favorable parent for emission of a particular fragment is one leading to a heavy daughter close to doubly magic ^{208}Pb, dictates that emitters of Si and heavier fragments will be transuranic nuclides. Until now, attempts to find nuclides that emit Si have concentrated on ^{241}Am, with negative results.[4,8,10,11] Other nuclides, for which the branching ratio for emission of Si relative to alpha decay may be large enough to warrant a search, include ^{237}Np, ^{238}Pu, ^{240}Pu, ^{240}Cm, and ^{242}Cm.[12–15]

The experimental difficulty in searching for decay modes involving emission of Si or heavier fragments is twofold: According to most models[13–15] the branching ratio relative to alpha decay is expected to be below 10^{-15}, and the branching ratio relative to spontaneous fission decreases with Z^2/A and is expected to be below $\sim10^{-7}$ for most even-even nuclides. To identify one $\sim$80-MeV fragment with $Z=14$ out of $>10^7$ fission fragments with similar energy is a formidable experimental challenge.

We decided to search for heavy-fragment emission from ^{238}Pu, due to the possibility that the results would help to discriminate among competing models of heavy-fragment radioactivity. Three of the models predict both Mg and Si emission from ^{238}Pu with comparable probability,[13,15,17] whereas other models predict that Mg (Refs. 13 and 18) or Si (Ref. 14) will dominate. Furthermore, odd-A hindrance factors $>10^2$ are typical in heavy-fragment radioactivity,[1] so that an even-even nuclide is a better prospect than an odd-A one. To deal with the problem of the high expected background of spontaneous fission, we exploited the fact that the range of the monoenergetic Si is nearly twice as great as the range of fission fragments. Our approach was to cover the detectors with an Al foil just thick enough to stop fission fragments, and to establish with accelerator calibrations the optimal procedure for identifying Si and other heavy fragments that would penetrate the Al with reduced energy.

Branching ratios predicted for Mg and Si emission relative to alpha decay for ^{238}Pu range from $\sim10^{-15}$ to $\sim10^{-18}$ for the more successful models. To deal with the enormous backgrounds of alpha particles and fission fragments, we used LG750 phosphate glass detectors, which easily distinguish Si ions, Mg ions, and fission fragments, and are not sensitive to ions lighter than Ne.[11] Tests with various thicknesses of Al showed that a 13-μm Al foil on each glass plate is just thick enough to stop fission fragments while allowing Mg and Si ions with the expected energy to penetrate the foil and emerge with an energy of $\sim$1 MeV/nucleon. Range-energy calculations showed that ions of 67-MeV ^{30}Mg, 67.32-MeV ^{28}Mg, and 78.95-MeV ^{32}Si penetrating the Al foil would have a residual range in LG750 glass of 7.9 to 9.6 μm when account is taken of self-absorption in the source (0.22 mg/cm^2) and a distribution of pathlengths in the Al foil corresponding to zenith angles from 0 to 25°. These energies and isotopes are expected on models that assume two-body decay with both fragments in the ground state. The range of al-

pha particles from ^{238}Pu is calculated to be ~ 2 μm shorter. Moreover, alpha particles do not show up directly as tracks; instead, at a sufficiently high dose, they alter the response of the glass to heavy particles that do produce tracks. In Ref. 11 it was shown that the general etching rate of LG750 increases and the ratio v_T/v_G of track etch rate to general etch rate decreases as the alpha particle fluence increases above about 10^{14}/cm^2, and that this can be taken into account by a calibration. By lining the inside surface of a large, evacuated, hemispherical dome with glass plates and putting the ^{238}Pu source at the focal point, we were able to collect tracks at nearly normal incidence (zenith angle $\leq 25°$) over a large enough detector area to reach a branching ratio below 10^{-16} while limiting the alpha-particle fluence to $\sim 10^{14}$/cm^2.

We prepared the source from an 80-mg sample of ^{238}Pu oxide of 98.3% isotopic purity that had been made several years ago. It was necessary to separate the ^{234}U daughter, which would otherwise contribute a background of Ne and Mg events.[9] We dissolved the sample in a mixture of HNO_3 and HF, converted it to the chloride by repeatedly evaporating the solution to dryness upon addition of HCl, dissolved the product in 9M HCl, and loaded it on an anion-exchange column. After washing the activity with more 9M HCl, we eluted plutonium from the column with a warm 5% solution of HI in 10M HCl. We evaporated the column eluant to dryness, picked the activity up in 8M HNO_3, and loaded it on a second anion-exchange column. We washed the column with 8M HNO_3, then with 4M HNO_3, and then eluted the plutonium with 1M HCl. We sampled the column eluant and did a radiochemical assay which showed that the mass of ^{234}U remaining in the sample was less than 0.02% of the ^{238}Pu mass. We evaporated the column eluant to dryness, picked the activity up in 8M HNO_3, and repeated the second anion-exchange step to further reduce impurities in the sample.

To prepare the source we dissolved the dried ^{238}Pu sample in 0.5M HCl and used the following technique to electrodeposit the activity from an isopropanol solution onto a 1.5-mm-thick nickel disk. We clamped a Teflon chimney onto the face of the disk and filled the resulting reservoir with 100 ml of isopropanol and 7 ml of the Pu stock solution. The shape of the chimney defined the source area as a circle 8 cm in diameter. We suspended a large-area platinum electrode ~ 1 cm above the nickel surface and applied a bias of ~ -20 V. The current through the solution was ~ -150 mA. We mixed the solution and rotated the cell constantly during the 3 h that the bias was on. Layers of ^{238}Pu were applied to the disk in two separate plating procedures, after which we heated the source for several hours at ~ 200 °C to remove residual isopropanol from the deposited plutonium salt.

We determined the source thickness by placing a track detector directly over the deposit for a short time both before and after the 172-day exposure of the detectors in the actual experiment. The density of fission tracks defines the thickness of the source, given the spontaneous fission half-life[16] of 5×10^{10} yr. The activity was lower by 25% after the exposure, and the source showed evidence of blistering; under the intense alpha-particle bombardment the source was gradually destroying itself. We used the average of the two determinations to arrive at a total deposit of 10.5 mg, or a thickness of 0.21 mg/cm^2 of ^{238}Pu.

We mounted the source facing upwards in the bottom of a vacuum chamber and then placed the hemispherical dome of Al-foil-covered track detectors over the source. The 9.5-cm inner radius of the dome was constrained by the size of the glove box door. The circular base of the dome was propped ~ 2 cm above the plane of the surface of the source. The vacuum chamber was kept at a pressure of less than 10 Torr throughout the exposure. Based on the alpha half-life of 87.7 yr, we calculated that the total number of alphas that impinged on the detectors during the 172-day exposure was 3.6×10^{16}, the alpha fluence was 1.02×10^{14}cm^2, and the fission fragment fluence was 3.5×10^5/cm^2. The amount of ^{234}U in the source due to decay of ^{238}Pu increased from 0.5 μg/cm^2 at the start of the exposure to 1.3 μg/cm^2 at the end. From its known branching ratio for Mg emission, we estimate that only 0.01 of a Mg event would have been produced from the ^{234}U.

At the end of 172 days we removed the dome from over the source. We passed each track-detector plate from the glove box individually, stripped off the Al foil, and cleaned the glass by soaking the pieces in 1M HCl and rinsing with ethanol. The contamination of the Al foils was typically $\sim 10^3$ alpha dis/min/cm^2 due to self-transfer of the ^{238}Pu in the vacuum chamber. Prior to washing, the glass plates were contaminated on the front and back sides equally with $\leq 10^2$ alpha dis/min/cm^2, probably caused by handling prior to passing them from the glove box.

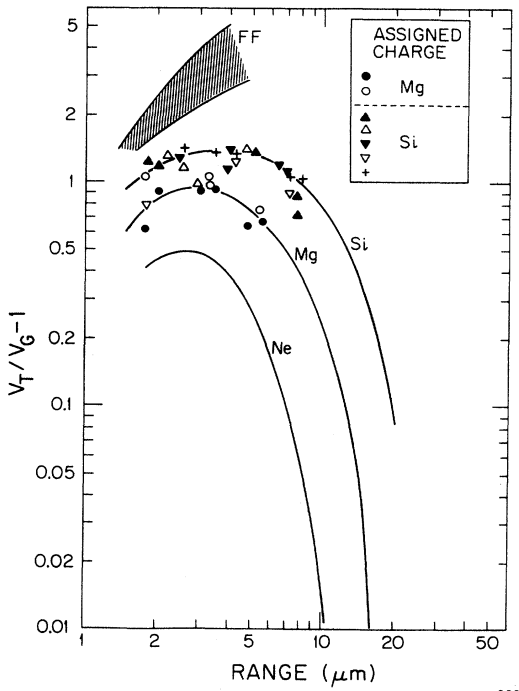

FIG. 1. Identification of fragments emitted from ^{238}Pu as Mg, Si, and fissions (FF). Each symbol designates a specific event. The curves are based on calibrations obtained with ^{28}Si, ^{24}Mg, and ^{20}Ne ions at Lawrence Berkeley Laboratory SuperHilac.

Figure 1 contains both calibrations and data. The three calibration curves were obtained by irradiating LG750 plates at the Lawrence Berkeley Laboratory SuperHilac with ^{20}Ne, ^{24}Mg, and ^{28}Si ions and then exposing the plates to doses of ^{238}Pu alpha particles ranging from 1.6 to $3.5\times10^{14}/\text{cm}^2$. To establish the response curves as a function of range of Mg and Si ions, we did a series of six etches in 48% HF acid at 22 °C, increasing in time from 8 to 60 min, which removed increasing amounts of glass. After each etch we made cellulose acetate replicas of the glass surfaces so that the profiles of the conical etchpits could be measured at several residual ranges. The response curves were insensitive to alpha fluences in the interval 1.6 to $2.5\times10^{14}/\text{cm}^2$.

The cross-hatched region in Fig. 1 labeled FF (for fission fragments) was obtained by a self-calibration during the actual search. This came about because of occasional localized regions in the Al foil, typically about one region a few hundred μm in diameter every ~30 cm^2, that were thin enough to allow fission fragments from the Pu to penetrate to the glass. The finite width of the cross-hatched region reflects the broad mass and charge distribution of the fragments.

The total usable detector area (after breaking one plate and contaminating another with Pu) was 352 cm^2. We first etched the glass plates for a time such as to remove 3.7 μm of glass, which represents a compromise between the desirability of etching for a long time such as to produce large, easily detectable etchpits, and the desirability of etching for a short time so as to obtain the most accurate information about ionization rate and range of Mg and Si ions with very short range. After scanning and locating a number of candidates for heavy-fragment decays, we made acetate replicas for detailed measurements. To scan 352 cm^2 of glass manually at the high magnification necessary to detect a small number of tracks less than ~9 μm long, viewed approximately head on, would have been extraordinarily difficult and time consuming, and the efficiency might well vary from day to day due to human factors. Therefore, we used an automated microscopic scanning system which appears to have an efficiency of unity, based on a comparison with the result of a manual scan of one plate of area 36 cm^2.

An etched glass plate is viewed by transmitted light with a 25× microscope objective (field of view 320×250 μm) on a stage that is moved automatically in a raster-scan pattern. A Cohu CCD camera sends digitized images of the fields of view to a Datacube Image Processor, which performs a threshold operation to select dark pixels. The X,Y positions of these pixels are then sent to a SUN Microsystems workstation, which stores those clusters of contiguous pixels that lie within a predetermined size interval and have a somewhat circular shape. The sequence of stage movement, digitization, image processing, and clustering takes ~0.8 sec/field of view. The typical density of candidate events is $\sim10^2/\text{cm}^2$ for a reasonably clean and scratch-free etched surface. At the end of a scan the system cycles through all the events at a controllable rate (typically ~0.2 candidate event/sec) for visual inspection. This inspection serves to select *bona fide* etched tracks from flaws that pass the crude acceptance criteria of size and circularity. The overall procedure is at least a factor of 10 faster than scanning the entire glass surface area manually.

After locating events by this technique we made measurements on acetate replicas, etched the plates a second time to increase the thickness removed from 3.7 to 4.2 μm, and made new replicas and new measurements. On the first replica the etchpits had pointed tips; on the second replica the etchpits had rounded tips indicating that the end of the range had been reached. For each event we were able to measure the quantity $s=v_T/v_G$ at up to three different residual ranges for each of the two etch times, using the expressions for track geometry given in Ref. 1. A measurement of the etchpit mouth diameter gave s at the largest range; a measurement of the diameter of the tip of the etchpit gave s at the smallest range; and a measurement of the average etchpit cone angle gave s at intermediate range. Figure 1 shows the results of these measurements of etchpit geometry. In order to expand the scale at values v_T/v_G close to 1, we plotted $\log_{10}(v_T/v_G-1)$ instead of $\log_{10}(v_T/v_G)$. One can see three groupings of data at average ranges of about 2, 5, and 8 μm. We assigned a charge parameter to each data point, based on its position with respect to the calibration curves, and took the average value of the charge parameter for each of the seven events. Five of the events had an average charge parameter Z' close to 14, corresponding to Si, and two had Z' close to 12, corresponding to Mg. The background radiation damage precluded isotopic resolution, so that only Z, not A, could be determined.

Figure 2 shows the etchable ranges of the seven events. The bracket above the histogram indicates the residual range in the glass expected when finite source thickness and zenith angle distribution in the Al foils are taken into account. A possible shift of ~1 μm between the calculated and measured range distribution suggests that the last 1 μm of range may be unetchable, due to the drop in ionization rate near the end of range. Alternatively, a 1-μm range deficit might correspond to a kinetic energy deficit of less than 5% due to fragment excitation, which would be much less than the 13% to 14% seen in fission.

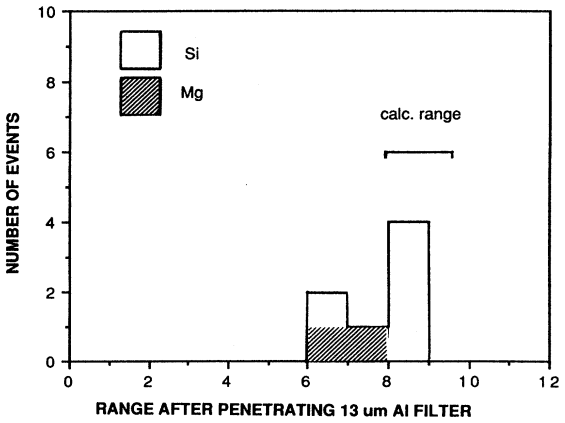

FIG. 2. Measured residual ranges of Mg and Si fragments emitted from a 0.21 mg/cm^2 ^{238}Pu source after passing through a 3.5 mg/cm^2 Al foil.

TABLE I. Decay Modes of ^{238}Pu and branching ratio (B) relative to alpha decay.

Emitted fragment	^{4}He	Sp. fission	^{28}Mg+^{30}Mg	^{32}Si
No. of events	...	...	2	5
Partial half-life (yr)	87.7	5×10^{10} [a]	$\sim1.6\times10^{18}$ [b]	$\sim6\times10^{17}$ [b]
Measured $\log_{10}B$	$\equiv0$	-8.8 [a]	-16.25 ± 0.25 [b]	-15.86 ± 0.16 [b]
Calculated $\log_{10}B$				
Poenaru *et al.* (Ref. 15)			-16.0	-15.5
Poenaru *et al.* (Ref. 19)			-16.4	-16.6
Price *et al.* (Ref. 3)			-15.9	-16.0
Shi and Swiatecki (Ref. 13)			-15.2	-18.5
Pik-Pichak (Ref. 14)			<-17.1	-17.1
Blendowske and Walliser (Ref. 17)			-16.4	-16.3
Buck and Merchant (Ref. 18)			-17.7	-18.9

[a]Reference 16. [b]This work.

Table I summarizes our results and compares the measured branching ratios with those predicted by various models. Despite the limited statistics, it seems clear that the results are in comparable accord with the predictions of the simple square-well plus Coulomb potential model,[3] with the unified model of Poenaru *et al.*,[15] and with the cluster model of Blendowske and Walliser,[17] regarding both the magnitudes of the branching ratios for Mg and Si emission and the relative rates of Mg and Si emission. The unified model of Shi and Swiatecki[13] predicts far too low a rate of Si emission. Both the unified model of Pik-Pichak[14] and the cluster model of Buck and Merchant[18] predict too low a rate of both Mg and Si emission. It seems rather surprising that the simplest possible model[3] of penetration through a barrier consisting of a square-well plus Coulomb potential, with a radius parameter chosen in 1985 to be $r_0=0.98$ fm in order to fit ^{14}C data, gives at least as good a fit to all data on the subsequently studied Ne, Mg, and Si decays as is achieved by the more sophisticated unified and cluster models.

The success of the range filter technique for eliminating the fission background in the case of ^{238}Pu should spur the investigation of heavy fragment radioactivity in a number of other spontaneously fissioning transuranic nuclides.

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