agreement with the bubble-chamber data of Blobel *et al.*[8] at 12 and 24 GeV/c for π^+, π^-, and p. Furthermore, our data fit well into the global plots of Cronin *et al.*[1]

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Search for Superheavy-Element Decay in Samples of Madagascar Monazite*

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Two samples of Madagascar monazite from the same geological formation as the biotite studied by Gentry *et al.* were examined by using a neutron multiplicity counter capable of detecting binary or ternary spontaneous fission decay in any element. No events characteristic of spontaneous fission decay of superheavy elements were found. Derived limits indicate that if superheavy elements were present, then their spontaneous fission half-lives must be extremely long or their concentrations extremely small.

A recent communication reported[1] x-ray energy spectra of monazite inclusions in biotite mica subjected to low-energy proton bombardment. Spectral analyses were interpreted as evidence for at least three superheavy elements with most probable atomic numbers of 116, 124, and 126. These results indicate unusual stability, because the existence of primordial superheavy elements in nature implies half-lives $\gtrsim 10^8$ years. Such long-lived superheavy elements might be expected to decay by α-particle emission or by spontaneous fission.[2] Since all heavy nuclei are energetically unstable with respect to binary fission, it is expected that long-lived α-decay chains beginning with a superheavy element would eventually terminate with a spontaneous-fission step.

The mass of primordial superheavy elements present in the giant halo inclusions was estimated to be as high as several hundred picograms.[1] If we assume, conservatively, an average value of only 100 pg in a 2-μg inclusion, the mass ratio of superheavy element is 5×10^{-5}, or 50 ppm. This concentration is so large that, even if bulk monazite deposits near the sites of the giant halo monazite inclusions were to contain somewhat lower concentrations of superheavy elements, it should be possible to observe spontaneous fission events from decay of the superheavy elements, or from daughter nuclei in radioactive equilibrium. Neutron-multiplicity counting is capable of detecting spontaneous-fission decay of any element, either in the binary- or ternary-fission mode, with high sensitivity and low background.

The average total number of neutrons per fission $\bar{\nu}$ is determined primarily by the average total excitation energy and by the neutron separation energies of the fission fragments. Most calculations predict an increase in $\bar{\nu}$ with increasing Z for binary fission.[2,3] For spontaneous binary-fission decay of superheavy elements, the num-

ber of emitted neutrons $\bar{\nu}$ is expected to be 8–10, significantly higher than for any known actinide. Thus a high value of $\bar{\nu}$ may be taken as unambiguous evidence for decay of a superheavy element, but must be combined with other techniques, such as chemical separation, to establish the atomic number. A recent calculation by Kolb[4] indicates that ternary fission decay for $Z = 114$ would lead to high total kinetic energies, low excitation energies, and $\bar{\nu} \simeq 2$ to 4. We have previously examined a large number of natural samples at our laboratory[5,6] for indications of spontaneous fission by neutron-multiplicity counting, all with negative results.

The two samples studied here were from the Chaines Anosyennes in the extreme southeastern part of the island of Madagascar, Malagasy Republic.[7] These samples were of particular interest because they were taken from the same granitic-charnockitic sill as the biotite which contained the giant halos studied by Gentry *et al.*[1] Although we did not look for giant halos in these samples, geochemical studies have shown that element distributions are similar for bulk monazite and for monazite inclusions in biotite. Sample D6356 was taken near the Maloto River, southeast of Haut Mandrare. When received it was a rock of irregular shape, coarsely crystalline, and dark in color. It consisted of monazite with hypersthene and cordierite. The second sample of monazite was D7187, from an area near Ampasimena. This sample was texturally similar to the first and consisted of monazite and apatite in pegmatite.

The monazite samples were measured in an improved version of our neutron multiplicity counter,[8] which contains thirty ^{3}He detectors in a cylindrical array, embedded in a paraffin and polyethylene matrix surrounding a sample chamber 10 cm in diameter and 30 cm long. This present configuration has a single-neutron counting efficiency of[9] 0.47, as determined with a ^{252}Cf source ($\bar{\nu} = 3.73$) in a gated mode with a time window of 600 μs to permit thermalization and absorption of neutrons. The counter was provided with a scintillation-type anticoincidence mantle to suppress recording of neutron bursts from muon reactions in structural materials or samples. Approximately 250 muon events per second were observed in the mantle, leading to ~15% dead time.

Sample D7187 was counted as one piece, after which it was ground up and homogenized for analysis. The other sample (D6356) was ground prior to analysis and neutron counting. Uranium and thorium concentrations were determined by nondestructive γ-ray spectrometric analysis; the results showed 0.348 g U and 8.16 g Th in D7187, and 0.265 g U and 3.63 g Th in D6356. Cerium was determined chemically, while the other rare earths were analyzed by mass spectrometry, based on the cerium concentration as a standard. Counting times were 12.85 days live time (15.40 days clock time) for sample D6356 and 15.64 days live time (17.98 days clock time) for D7187.

The empty-chamber background rates, in events per day for higher multiplicities, are approximately 20 of multiplicity two, 1 of multiplicity three, 0.1 of multiplicity four, and 0.03 of multiplicity five or greater. In most monazite samples the largest contribution to multiplicities in the range of about 2 to 5 results from the spontaneous fission of ^{238}U in natural uranium; about 97% of the corrections were due to this source. Thorium and rare earths cause some neutron bursts due to reactions with muons. Some neu-

TABLE I. Summary of neutron multiplicity counting data on Madagascar monazites.

Multiplicity	Total counts	Counter background	Corrections	Net counts	Net counts per live day
		Sample D6356, 349 g			
2	539 ± 23.2	190.7 ± 8.3	352.7 ± 6.3	− 4.4 ± 24.8	− 0.34 ± 1.9
3	87 ± 9.3	13.7 ± 2.2	71.8 ± 2.1	+ 1.5 ± 9.8	+ 0.12 ± 0.8
4	6 ± 2.4	1.4 ± 0.7	9.8 ± 0.9	− 5.3 ± 2.7	− 0.41 ± 0.2
≥5	0	0.36 ± 0.36	2.1 ± 0.5	− 2.5 ± 0.6	− 0.19 ± 0.05
		Sample D7187, 232 g			
2	884 ± 27.7	304.6 ± 26.3	564.5 ± 12.6	+ 14.9 ± 40.2	+ 0.95 ± 2.6
3	133 ± 11.5	20.5 ± 6.8	113.7 ± 4.4	− 1.2 ± 14.1	− 0.08 ± 0.9
4	19 ± 4.3	1.6 ± 1.6	15.5 ± 1.9	1.94 ± 5.0	+ 0.12 ± 0.32
≥ 5	3 1.7	0	3.7 ± 0.9	− 0.7 ± 2.0	− 0.04 ± 0.13

trons are absorbed in monazite by highly absorbing rare earths, particularly Sm and Gd. The latter two corrections are opposite in sign and about equal in magnitude.

Correction factors were determined for each of the above effects in a series of separate experiments by use of methods to be described elsewhere.[10] A summary of the observations and corrections applied are listed in Table I. The discrete multiplicities through 7 and all ≥ 8 were recorded, but because of the small number of events with high multiplicity we have shown a single entry for those ≥ 5. Because single-neutron events are produced in large numbers by several processes [e.g., (γ, n), (α, n)], it is not useful to evaluate multiplicities of one in terms of fission.

The errors shown in Table I are standard deviations which result from propagating all counting errors. Uncertainties of analysis for uranium, thorium, and absorbing rare earths do not contribute significantly to the final errors. It is evident that neither sample produced a significant net surplus of multiple neutron events. Therefore, the maximum net number of events for each multiplicity was taken as twice the standard error. Maximum spontaneous-fission disintegration rates for the samples, D_{max}, for $\bar{\nu}$ of three and ten were obtained by fitting to the maximum event distribution the expected distribution $P(n)$ of observed events[3] with neutron multiplicity n

$$P(n) = \sum_{\nu=n}^{\nu_{max}} p(\nu) \binom{\nu}{n} \epsilon^n (1-\epsilon)^{\nu-n}.$$

The term $\binom{\nu}{n}$ is the usual binomial coefficient $\nu!/[n!(\nu-n)!]$, ϵ is the single-neutron efficiency, and $p(\nu)$ is the probability of emitting ν neutrons per fission, assumed to be a Gaussian distribution with width parameters σ_ν taken[3] as 1.15 for $\bar{\nu}=3$, and as 1.71 for $\bar{\nu}=10$. For both samples, if $\bar{\nu}$ were ten we find the value of D_{max} to be 0.43 disintegrations/day; if $\bar{\nu}$ were three, D_{max} would be 15 disintegrations/day.

Methods which make use of radioactivity can determine the value of the constant relating the half-life and the concentration of a radionuclide. Although the work reported here did not succeed in detecting spontaneous fission decay in the two monazite samples, the upper limits obtained place important constraints on the interpretations of the data of Gentry *et al.*[1] In calculating chemical concentrations we have assumed a superheavy element with $A = 354$: The evidence discussed by Gentry *et al.* was strongest for element 126; moreover, a long-lived isotope of element 126 requires a neutron number of at least 222 to be β stable,[11] and 228 is favored.[11-13]

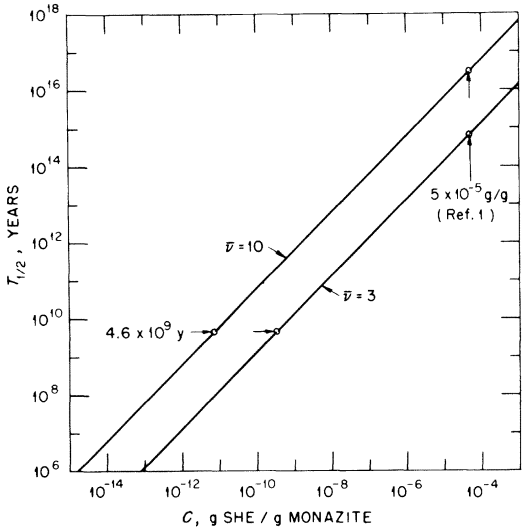

FIG. 1. Detection limits for the monazite samples, for $\bar{\nu}$ values of 3 and 10. For a given value of C, the plot gives a minimum value for $T_{1/2}$. For a given $T_{1/2}$, a concentration greater than the corresponding value of C would be detected.

It is convenient to summarize our limits in the form $T_{1/2}/C \geq k_\nu$. Here, $T_{1/2}$ is the half-life of the superheavy element; C is the mass ratio, expressed as grams of superheavy element per gram of sample; and k_ν is a constant for spontaneous fission with average neutron emission $\bar{\nu}$.

Because the results of the present experiments must be compared with evidence for primordial superheavy elements said to be present in inclusions of pure monazite,[1] we prefer to compute limits for $T_{1/2}/C$ on the basis of the monazite present, rather than on total sample weight. To derive these more conservative limits we estimated the actual monazite present from the chemical composition, assuming that all rare earths and thorium in the samples were present as monazite. In this way we estimated 49 g of monazite for D6356 and 103 g for D7187. The mean values of k_ν now become $k_3 = 1.6 \times 10^{19}$ years and $k_{10} = 6.1 \times 10^{20}$ years.

These final limits are displayed graphically in Fig. 1. We include our detection sensitivity for $\bar{\nu}=3$ because of the suggestion that ternary fission may be a favored decay mode for some superheavy elements[4]; however, the limit applies equally well to detection of spontaneous binary-fission decay of transplutonium nuclides with $\bar{\nu}$

$\simeq 3$.

A few numerical examples will illustrate the sensitivity of these measurements. If the half-life of element 126 is assumed to be equal to the age of the earth, 4.6×10^{9} y, then our lower limits of detection would be mass ratios $C \geq 7.5\times10^{-12}$ g/g for $\bar{\nu}=10$ and $C \geq 2.9\times10^{-10}$ g/g for $\bar{\nu}=3$. For the same half-life of 4.6×10^{9} y and a mass ratio $C \simeq 5\times10^{-5}$ g/g as in the experiments of Gentry *et al.*,[1] our 103-g sample of monazite would have produced about 4×10^{6} disintegrations/day, which is more than 10^{5} times our detection limit for $\bar{\nu}=3$ and $\sim10^{7}$ times our detection limit for $\bar{\nu}=10$.

Our failure to detect any neutron activity other than that due to impurities permits us to establish a minimum value for the half-life of a superheavy element, if the mass ratio C is known. As indicated above, the mass ratio derived from the data of Ref. 1 is about 5×10^{-5}. For the same concentration in our monazite samples, $T_{1/2} \geq 3.0\times10^{16}$ years for $\bar{\nu}=10$, or $T_{1/2} \geq 8.0\times10^{14}$ years for $\bar{\nu}=3$.

Gentry *et al.*[1] reported that superheavy elements were found only in monazite inclusions of giant halos. In his study of giant halos in Madagascar biotite, Gentry[11] found that halos with diameters >80 μm comprised $\sim7\%$ of those measured. Chemical analyses do not show large differences between the compositions of monazites in biotite and in bulk monazite from pegmatites. Even if superheavy elements were inhomogeneously distributed among monazite inclusions, the dilution of superheavy-element concentration in bulk monazite suggested by Ref. 11 does not significantly alter our conclusions.

The minimum half-lives calculated above are much longer than the diverse results of recent theoretical calculations[12-15] for the nuclei around $Z=126$. Wong[12] has calculated that $^{354}[126]$ in a spherical configuration would have a half-life of only 0.03 year, and that toroidal or bubble shapes might be required to achieve half-lives of $\sim10^{8}$ years. A calculation by Petrovich *et al.*[13] was different in detail and was able to demonstrate qualitatively that the half-life for spontaneous fission decay of $^{354}126$ might be long ($>10^{9}$ years); however, the α-decay half-life, and thus the total half-life for disappearance of the nuclide, was less than 100 years. A macroscopic-microscopic calculation by Möller and Nix[14] found $^{354}[126]$ to be β stable, but the spontaneous-fission half-life only 18 years. Long half-lives of at least 10^{8} years were calculated by Möller and Nix only for nuclei in the island of stability below $Z \simeq 120$. Andersson *et al.*[15] employed a readjustment in the nuclear surface diffuseness to produce a proton energy gap at $Z=126$, but their neglect of a large macroscopic restoring force against changes in surface diffuseness suggests that their half-lives should be regarded as upper limits.[14]

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