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SEARCH FOR SUPERHEAVY ELEMENTS AMONG FOSSIL FISSION TRACKS IN ZIRCON*

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ABSTRACT

Fossil tracks with long ranges were found in some crystals of natural zircons. Their length is 120 to 200% of that of uranium fission tracks in zircon. A few of the developed fossil tracks are also 3 to 4 times as wide as fission tracks from uranium (observed in the same crystal). The long and/or fat tracks could be explained as having resulted from fission of several superheavy elements from the regions of predicted closed shells (around elements 114, 126, and 154). All of these could be present in zircons.

The data reported herein support the earlier observations, where fat tracks (3 to 4 times as wide as those from neighboring uranium fission events) were detected by the mylar foil technique in a high-temperature fraction of HfO_2 chlorination.

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INTRODUCTION AND REVIEW

1. Theoretical Predictions and Prospects for SHE in the Universe

Superheavy elements (SHE) can exist in nature or could be prepared in accelerators and survive long enough to be detected only if they contain isotopes with closed nuclear shells. Such closed shells would cause nuclear stability in these elements in comparison to neighboring elements or isotopes which do not have closed shells and are unstable towards nuclear fission. The greatest stability results if both proton and neutron shells are closed, i.e., the nucleus is doubly-magic. Such doubly-magic shells were suggested in 1957 when an element $^{310}126$ with closed shells of N = 184 neutrons and Z = 126 protons was postulated (1) and used in calculating nuclear masses related to deformation (2). The next doubly-magic nucleus beyond the presently known elements, i.e., beyond ^{208}Pb, was suggested in 1967 to be at Z = 114, N = 184 (3).

Since 1968, a number of theoretical papers were published describing the predicted half-lives of elements in the Z = 126 to 110 regions (4,5,6,7,8,9,10,11,12). Energy levels of neutrons and protons calculated with three sets of parameters were given (13,14). In all cases it appears that the neutron levels form gaps which may indicate the existence of closed shells at N = 126, 184, 228, 308 and 406. The proton levels would thus form gaps for closed shells at Z = 82, 114, and 164. One set of parameters used in (13) and all three sets of parameters utilized in (14) indicate the possible existence of an additional closed shell at Z = 154. Another set of parameters (13,14) indicates a gap at Z = 204 (i.e., the closed shell at Z = 164 plus a gap after the full shells $1k_{17/2} + 1g_{13/2} + 2g_{7/2}$, containing altogether 40 protons).

Summarizing these various theoretical predictions of doubly-magic nuclei, it can be concluded that regions of stability are indicated; they are centered around elements 114, 126, 154, 164, and 204.

The longest lifetime predicted for a nucleus in the element 114 region could survive in nature, i.e., $^{294}110$ has a predicted half-life of 10^{10} years for spontaneous fission (SF) and 10^{8} years for alpha-decay and is beta-stable (5). The nucleus $^{293}110$ should have half-lives about 10 times longer than $^{294}110$ for both modes of decay. Half-lives for $^{294}110$ of comparable or even somewhat larger magnitude were given (15). Similar half-lives were predicted (14) for the element 154 region (of the order of $> 10^{10}$ years for SF and 10^{7} years for alpha-decay).

From the results of the various calculations cited above, it can be concluded with some degree of optimism that several regions of SHE could exist (around Z = 114, 126, 154, 164, and perhaps 204), and that some isotopes might have possibly survived in nature since the last nucleosynthesis. Thus, if the theoretical predictions of islands of stability and long half-lives of some isotopes are accepted, then a search for SHE in nature is justified.

According to present astrophysical theories, only elements with $A < 7$ were formed shortly after the Big Bang (16). Elements with $A > 7$ were formed by thermonuclear synthesis inside the stars or by supernovae explosions. In the case of our solar system, these reactions resulted in a mass abundance of elements, as, for example, given in (17) for the s-, p-, and r-processes.

The abundance curve is almost level in the high mass region (with peaks at closed shells). An extrapolation of the abundance curve resulting from the r-process beyond A = 238 to the region of the predicted additional closed shells (around Z = 114, 126, etc.) produces abundances not unlike those of uranium, depending on the slope which is used. The abundances will, of course, be zero in the regions of spontaneous fission instability, but in the vicinity of doubly-magic nuclei it could follow the general trend of the r-process. Abundances in the 114, 126, etc., regions might be 10^{-4} of that of uranium. The existence of U and Th in our solar system is proof that the r-process, or a similar rapid neutron absorption process occurring in neutron stars (18), did take place in our galaxy.

Calculations of production ratios for the isotopes ^{129}I, ^{244}Pu, ^{232}Th, ^{238}U and ^{235}U were used to estimate the time scales for the last r-process, the average time of nucleosynthesis, and the time between the end of nucleosynthesis and solidification (19, 20, 17). These calculations indicate that the last time superheavy elements could have been formed in our solar system or near part of our galaxy by the r-process or in a supernova explosion was 5.5 to 6.9 x 10^9 years ago. If their half-lives were at least 3 x 10^8 years, they should still be observable.

There are two differing points of view concerning the probability of SHE-production in the r-process. Some theoreticians conclude that the element 114 region cannot be reached because spontaneous fission would terminate the r-process (21). Others calculated that SHE can be formed by the r-process (22, 23) or in supernovae (24).

The composition of a neutron star was described in recent reviews (25,26). It might have a radius of about 10 km and a density of 10^{11} g/cm^3 on the surface and 10^{14} g/cm^3 in the center. Its approximately

1 km thick crust would contain neutrons mixed with nuclei, protons and electrons. Elements up to Z = 90 should be present in the crust. Most of them should have closed shells and be very neutron-rich. For example, lead (Z = 82) and uranium (Z = 92) could be bound with 164, 184, or 228 neutrons, and element 114 could be bound with an N = 228 or 306 closed shell. If the neutron star were to collapse further into a black hole, or if it were to collide with a normal star, part of the neutron star's crust could be either ejected or stripped off, thus bringing possible superheavy elements back into interstellar space (or very heavy, neutron-rich isotopes such as ^{320}U could be transformed into SHE by a chain of beta-decays). It was predicted (18) that, if a neutron star and a black hole were to collide, the neutron star would have been tidally disrupted before the actual collision. This would cause ejection of a substantial part of its mass into interstellar space. Elements up to Z = 118 would survive such an event, and element 126 could also be formed. As early as 1949 the process of creation of heavy elements by "fission of neutron star" into droplets of nuclear matter was described (27).

Most recently, r-process calculations were presented which showed that masses A > 260 could be formed and possibly also elements in the Z = 114 region (28). It is known that a supernova explosion could create a powerful α-n reaction in its He-layer. A large neutron flux would be created during $\sim$ 1 sec which could produce masses up to A = 340 in substantial abundance (29). The conclusion was (29) that SHE formation is possible if such nuclei could survive during fission-beta decay competition of the A = 340 nuclei.

Summarizing the astrophysical citations, there is some chance that superheavy elements are being created in the universe, either through the r-process or in supernovae or neutron star explosions. Some of these SHE might have survived the last 6 to 7 x 10^9 years.

2. Some Experimental Results

A spontaneous fission activity was observed in several fractions isolated from technical HfO_2 by thermochromatographic separation of chlorides (30). A hypothesis was postulated which explained the decaying and growing activities as belonging to some natural decay chain of superheavy elements (analogous to the ^{238}U decay chain, with some branching into spontaneous fission).

The fractions analyzed contained, besides some regular (decaying) fission activity, also an activity resulting in fission fragments capable of penetrating at least 26 μm of mylar foil and producing unusually "fat" tracks. The latter were interpreted as resulting from fission fragments with $54 < Z < 72$, having energies $150 < E < 250$ MeV per fragment. The fat tracks were attributed to the fission of elements 126 and/or 154. Fission fragments from element 126 could have a total energy of $E_t = 293$ MeV (31), whereas fission of element 154 should have $350 < E_t < 450$ MeV. Therefore the light single fragments from fission of elements 126 and 154 should have $E_{126} < 180$ MeV and $E_{154} < 220$ MeV, respectively.

In another experiment, sources of radiogenic lead totaling 3600 cm^2 were exposed underground (100 mwe) to 13 μm thick mylar foils (32). This lead was primarily composed of the isotopes ^{206}Pb, ^{207}Pb and ^{210}Pb from the decay chains of ^{238}U and ^{235}U in pitchblende, with a small amount of natural lead also present from the uranium ores.

Alpha-spectroscopic measurements of this radiogenic lead showed no traces of either the ^{226}Ra or the ^{228}Th decay chains. After a three-year exposure, the mylar foils were developed. They contained fission tracks with a range 19% longer than tracks produced by uranium fission fragments. Ordinary lead, which was simultaneously exposed to mylar foils under the same conditions, yielded a background effect of only 3% of the effect from radiogenic lead. These long-range tracks can be explained as resulting from the spontaneous fission of natural, superheavy elements in the suggested element 114 stability region, which were co-precipitated with the radiogenic lead.

The postulated natural decay chain of superheavy elements was assumed to begin in the $Z = 126$ region and decay, at least partially, by an alpha-decay chain into the $Z = 114$ region (30,32). The further assumption was made that elements with $110 < Z < 114$ should at least partially decay by spontaneous fission with a total energy of 215 to 235 MeV (as theoretically predicted (31)), producing fission fragments detectable on mylar foils. Another assumption was that some amount of element 124 (eka-uranium) should always be present along with natural uranium, its chemical homolog. Thus, if element 124 is present in uranium ores, then element 114 (eka-lead) should chemically follow radiogenic lead or radiolead as waste material during industrial isolation from uranium ores.

SEARCH FOR SHE IN ZIRCONS

1. Reasons for a Search in Zircons

The results disclosed in (30,32) indicate the possible existence of some superheavy elements in the Z = 154 and/or 126 regions (in hafnium) and in the Z = 114 region of stability (in radiogenic lead). The fact that the HfO_2 contained a spontaneously-fissioning activity which resulted in fat tracks led to the conclusion that the original mineral from which the HfO_2 was isolated should be thoroughly investigated. This mineral was Australian zircon mined from beach sands (alluvial deposits). It originally formed and crystallized together with granite. Many Australian zircons came from granite deposits 2.5×10^9 years old, which were not altered during this time interval. Zircon crystals are hard, highly weather-resistant, and they are transparent. Thus, investigation of this mineral offers the chance for a search for SHE islands of stability in a material which is old enough and unaltered to allow observation of the old footprints left in the sand since solidification of the surface of the earth.

Since concentration of superheavy elements in nature is low (otherwise they would have already been isolated in macroscopic amounts), the most sensitive methods, using the longest possible experimental exposure to yield the best statistics, must be enlisted for this search. Since fractions isolated from HfO_2 produced unusual fat tracks in mylar, similar "fat" tracks may be present as fossil tracks in zircon because the HfO_2 was originally isolated from this mineral. Thus, it was concluded that the most sensitive method available for a search for superheavy elements would be a measurement and energy calibration of fossil tracks in natural zircon crystals. Here are some of the other reasons for the choice of this mineral:

Predicted elements 126, 154 and 204 are eka-Pu, eka-eka-Hf (eka-Rf) and eka-154 (eka-eka-Rf). All three are expected to be carried well in zircon;

The total kinetic energy, E_k, of nuclear fission in zircon is represented by the sum of the track length of both fragments. It is possible that tracks from SHE are longer than those from uranium and could thus be easily distinguished. The normal uranium tracks could serve as internal calibration;

Concentrations of SHE as low as 10^{-23} to 10^{-26} g/g could be measured with a 10^8 year exposure.

2. Experimental Background

Tracks of uranium fission fragments were first developed with hydrofluoric acid (HF) in 1962 (33). Similar tracks were developed in zircon by heating zircon crystals in boiling 80% phosphoric acid, H_3PO_4 (at about 500°C) for one minute (34). Zircon is extremely resistant to track fading; fission tracks do not disappear until after about 100 min. at 700°C (35). By comparison, fission tracks in mica fade fast above 500°C, whereas in zircon they show no change below 570°C. Tracks developed in mica appear as etched cylindrical or rectangular holes, whereas tracks in zircons appear as cylindrical or rectangular crystals of phosphate glass which formed inside the zircon crystals. The fission fragments leave about 50 Å thick channels in the crystal, as first observed with the aid of an electron microscope (36). Etching of tracks in mica with HF enlarged these thin channels to a width of about 1 μm. Etching with

H_3PO_4 attacked the zircon and formed dark, cylindrical tracks which are visible with the optical microscope. The fission fragment range is represented by these tracks, provided that the energy loss (dE/dx) in mica or zircon is above some critical value $(dE/dx)_{crit}$. For mica and zircon $(dE/dx)_{crit}$ was reported to be 13 and 19 $MeV/mg/cm^2$, respectively (37). The track below $(dE/dx)_{crit}$ was not developed and thus cannot be seen as a track. This was demonstrated with Cf-fission fragments impinging on a mica surface in vacuum. The etched tracks were measured to have a maximum length of about 12.5 μm, whereas the calculated length of the light Cf-fission fragment was 15 μm (38). The tracks developed in zircon and described in (34) were not statistically evaluated as far as length is concerned, but a photograph showed them to be about 7 to 8 μm long. These tracks were used only to date the zircon. It was found that the "fission track age" of some large zircon crystals was 0.7 to 1.4×10^8 years (i.e., the zircon was probably below 570°C for the past $\sim 10^8$ years. The real age of many zircons, as measured by the $^{206}Pb/^{208}Pb$ ratio is much greater, often 1 to 1.7×10^9 years (39).

3. Calculated Length of Fission Tracks in Zircon

The length of tracks in many small zircon crystals of Australian origin was measured. The results were used to establish the total kinetic energy E_K of the fission fragments (in a crude pulse height analysis) and thus determine the possible presence of SHE in zircons. The experimentally measured E_K for ^{235}U+n fission and ^{252}Cf-fission (40) was used to calculate the range of fission fragments in zircon. The first step in this procedure involved calculating $(dE/dx)_{ZrSiO_4}$ from (dE/dx) data given separately for Zr, Si and O (41). The sum rule (composing $\left[dE/dx\right]_{ZrSiO_4}$ from the $\left[dE/dx\right]$ values of its components according to their molar fractions) was used for several energies between 1.5 MeV/AMU to 0.1 MeV/AMU. It was found that for the fission of ^{235}U+n into ^{97}Sr + ^{141}Xe or into ^{119}Pd + ^{119}Pd fission fragments, $(dE/dx)_{ZrSiO_4}$ is approximated well by $(dE/dx)_{Ar}$ over the whole energy range. Thus, the Ar-ranges (41) were used to approximate $ZrSiO_4$ for any other fission fragment. Four cases of ^{235}U+n fission were calculated by this method. The following values for E_K from (40) were used: 204 MeV for $E_{K,max}$,172 MeV for the average kinetic energy $\overline{E}_K$ in asymmetric as well as symmetric fission, and 138 MeV for $E_{K,min}$. The calculated ranges of both fragments are given in Table I. Both fragments were assumed to have full range in zircon, with one of them ending close to the surface of the crystal (< 1 μm deep) so it could be developed by H_3PO_4. Another way for such a full-range, two-fragment track to develop is via a short track which penetrates the crystal surface and also intersects the full track inside the crystal, thus providing access of H_3PO_4 to the track. The E_K values for the fragment pairs in columns 1 and 2 are given in column 3. The calculated ranges based on the use of Ar-ranges for each stopped fragment and after summing their ranges are given in column 5. The kinetic energy for each ion was obtained from E_K by dividing it in inverse proportion to the fragment mass (conservation of momentum). The relative number of fragment cases (from 40) are given in the last column in order to demonstrate the statistical impact of the track lengths.

The calculated range for each fragment (from 41) was reduced by the range below $(dE/dx)_{crit}$ where the track is not detectable. A value

TABLE I

CALCULATED TRACK LENGTH OF TWO-FRAGMENT EVENTS
FROM ^{235}U+n FISSION IN ZIRCON (ρ = 4.67 g/cm^3)

Fragment Light $^{A}Z_{,L}$	Fragment Heavy $^{A}Z_{,H}$	Total Kinetic Energy, E_K MeV	Approximate Neutron Multiplicity ν	Two-Track Length in Zircon μm	Relative Number of Events
$^{104}Mo_{42}$	$^{132}Sn_{50}$	204	0	11.65	0.7
$^{97}Sr_{38}$	$^{141}Xe_{54}$	172	2.5	10.21	100
$^{119}Pd_{46}$	$^{119}Pd_{46}$	172	2.5	9.50	0.14
$^{88}Se_{34}$	$^{148}Ce_{58}$	138	5	8.93	0.7

of $(dE/dx)_{crit}$ = 13 MeV/mg/cm^2, as given for mica (38) was used. The quoted (38) value for zircon, $(dE/dx)_{crit}$ = 19 MeV/mg/cm^2, is too low; it results in a reduced range which is less than the shortest range, ∿ 10 μm, which later was observed experimentally. The calculated two-track length was expressed in μm of zircon after subtracting the ranges below $(dE/dx)_{crit}$, namely, R_{crit} ∿ 1 to 1.5 mg/cm^2, and using 4.67 g/cm^3 for the density of zircon. The data presented in Table I are also fully applicable to spontaneous ^{238}U-fission, since kinematic energy measurements (42) produce the same E_K-curves for spontaneous fission of ^{238}U and for fission of ^{235}U+n.

Column 4 lists an estimate of the neutron multiplicity ν for each fission event. An average ν = 2.5 was used for E_K = 172 MeV. The lowest possible ν = 0 was used for the highest energy (204 MeV), and ν = 5 was listed for the lowest energy (corresponding to the highest fission multiplicity detected for ^{235}U+n). The ^{132}Sn nucleus is doubly-magic (N = 82, Z = 50) and will be spherical, nondeformed (resulting in E_K = 204 MeV). Additionally, the $^{104}Mo_{42}$ nucleus will have low deformation. Thus, all the energy from the mass defect of the fission fragments will be liberated as kinetic energy. The fragments in lines 2 and 3 are moderately deformed and in the last line they are strongly deformed, emitting five neutrons. From Table I it can be concluded that the experimentally observable range of ^{235}U+n fission fragments will be between 8.9 and 11.7 μm and should not exceed 12 μm.

Experimentally observable ranges were calculated in the same manner for fission fragments of SHE in zircon. The results are summarized in Table II. The first two columns give Z and A of probable fissioning

TABLE II

CALCULATED TRACK LENGTH OF TWO-FRAGMENT EVENTS FROM FISSION OF SHE IN ZIRCON (ρ = 4.67 g/cm^3)

Fissioning Nucleus Z	Fissioning Nucleus A	Assumed Neutron Multiplicity ν	Equation (1)[43] Total Kinetic Energy E_K MeV	Equation (1)[43] Calculated Total Track Length in Zircon μm	Equation (2)[44] Total Kinetic Energy E_K MeV	Equation (2)[44] Calculated Total Track Length in Zircon μm
108	286	10 def	210.7 [a]	11.2	224.2 [a]	11.9
108	286	2 sph	274.7 [b]	14.2	288.2 [b]	14.9
120	342	10 def	245.0 [a]	11.9	262.7 [a]	12.6
120	342	2 sph	309.0 [b]	14.8	326.7 [b]	15.5
148	450	12 def	340.2 [a]	14.0	369.2 [a]	14.9
148	450	2 sph	420.2 [b]	16.3	449.2 [b]	17.2
196	594	14 def	543.8 [a]	18.6	597.2 [a]	19.8
196	594	4 sph	623.8 [b]	20.6	677.2 [b]	21.7

[a] $E_K = E_{Kdef}$ was calculated for deformed fission fragments using Eqs. (1) and (2) and a high $\nu = \nu_{def}$ was assumed.

[b] $E_K = E_{Ksph}$ was calculated for spherical fragments from the total fission energy $E_t = (E_{Kdef} + 8\,\nu_{def})$ MeV $= (E_{Ksph} + 8\,\nu_{sph})$ MeV and low $\nu = \nu_{sph}$ was assumed.

SHE (fission was assumed to be preceded by three alpha-decays from the doubly-magic numbers $^{298}114$, $^{354}126$ and $^{462}154$, and by four alpha-decays from doubly-magic $^{610}204$). The third column gives assumed neutron multiplicities for two cases of fission for each nucleus: (a) highly-excited fragments (ν = 10, 12, and 14) from a highly-deformed nucleus, and (b) less excited spherical fragments with closed shells (ν = 2 or 4) for a spherical nucleus.

Column 4 gives predicted values for the total kinetic energy, using a semi-empirical equation given in (43):

$$E_K = 0.119Z^2/A^{1/3}. \tag{1}$$

Similarly, column 6 shows calculated values for the expected total kinetic energy using a more recent equation (44):

$$E_K = 0.13323Z^2/A^{1/3} - 16.64 \text{ MeV}. \tag{2}$$

The sum of the ranges calculated for both fission fragments using E_K from Eqs. 1 and 2 is given in columns 5 and 7 (the ranges falling below $(dE/dx)_{crit}$ = 13 MeV/mg/cm^2 were subtracted). The kinetic energy values denoted by superscript [a] (i.e., $E_K = E_{Kdef}$) were calculated with Eqs. 1 and 2, respectively, and a high neutron multiplicity $\nu = \nu_{def}$ (Column 3) was assumed. The kinetic energy values denoted by superscript [b] (i.e., $E_K = E_{Ksph}$) were obtained from the total fission energy, $E_t = (E_{Kdef} + 8\nu_{def})$ MeV = $(E_{Ksph} + 8\nu_{sph})$ MeV. E_{Kdef} and the values for ν_{def} and ν_{sph} from Column 3 were used. This model assumes an equal E_t yield for both deformed and spherical fission fragments and also a total energy expenditure of 8 MeV per evaporated neutron.

The conclusion from Table II is that if there are any fission fragment tracks in zircon exceeding 12 μm in length, particularly in the 14 to 16 μm range, they could be due to fission fragments from SHE.

RESULTS OF TRACK MEASUREMENTS

1. Tracks Developed with Phosphoric Acid

Fossil fission tracks were developed in Australian zircons with the aid of hot phosphoric acid (80% H_3PO_4 for 30 minutes at 470-490°C). The majority of the tracks were developed well, but some were still underdeveloped with a thickness of only 0.05 μm, and others were overdeveloped, with wide, often crooked, appearance. Each crystal appears to have a unique optional development time, which may be dependent on its radiation damage due to the presence of U or Th. This fact was accounted for by grouping and comparing only data from crystals which contain tracks of approximately the same diameter (thin tracks in all crystals in one group, or single crystals containing thin and wide tracks together). Crystals with tracks which were obviously overdeveloped or crystals which contained so many tracks that analysis was not possible were excluded. The crystals were observed in oil immersion with the optical microscope (1000X magnification). Length and depth of tracks were measured with a precision of 0.5 μm (±0.5 μm reading error for depth). The width of the tracks varied between 0.05 and 2.0 μm. The track diameter or width was measured with approximately a 50% error at 0.1 μm, a 25% error at 0.3 μm and a 10% error at 1.0 to 2.0 μm. After measuring apparent length ℓ and depth d of each track, the real length L was calculated by Pythagorean rule, i.e., $L = (\ell^2 + d^2)^{1/2}$.

The distribution of the longest tracks was established by measuring only the two or three longest tracks in each of 178 crystals. The results are plotted in the histograms in Fig. 1. There were 34 crystals which had no visible tracks; they are registered at the zero μm track length position. A few crystals had tracks in the 5 to 6 μm range, which crossed the surface of the otherwise clean crystal (without full tracks). Some of the observed crystals had tracks in the 7 to 8 μm range from fission events below the crystal surface. The majority of the crystals had tracks in the 9 to 11 μm region. Their range corresponds to that of ^{238}U-fission fragments (see Table I). Based on the relative number of expected events as given in Table I, a small number (0.7%) of U-fission fragments could produce tracks up to 12 μm long. Figure 1a clearly shows a significant number of tracks which have a range in excess of 12 μm. Finally, there were 15 crystals (8.4% of the total of 178), each of which had at least one long track in the region between 12 to 16.5 μm, i.e., in the SHE-region. The track density was typically from 1 to 5 per 100 μm^2 (= 10^{-6} cm^2) in the 144 crystals which showed visible tracks.

Some of the crystals (22 of 178) contained a few fat tracks. Their diameter or width was usually at least three times that of the average thin tracks in the same crystal. These wide tracks are plotted in the histogram of Fig. 1b. Four of 22 tracks, or 18%, are in the range above 12 μm. By comparison, only 15 of 144 thin tracks, or 10%, exceed a range of 12 μm. However, statistically the excess of apparent SHE-tracks (> 12 μm) is approximately the same for both fat and thin diameters.

During this systematic investigation of 178 crystals, some crystals were observed to have only short tracks, usually < 10 to 11 μm in length.

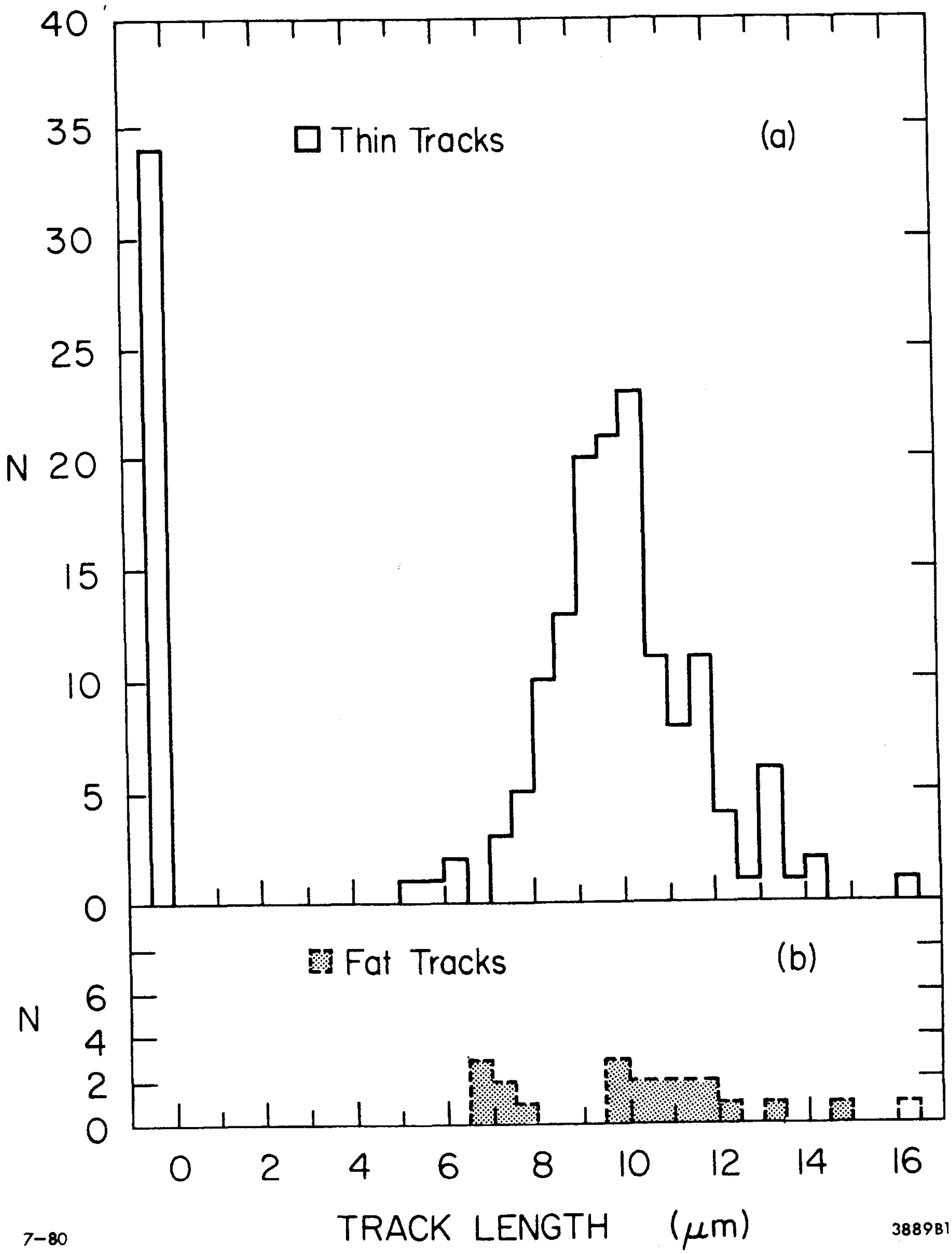

Fig. 1 Longest tracks as found in each of 178 scanned crystals: a) histogram of narrow or thin tracks; b) histogram of fat tracks ($\geq$ 3 times as wide as thin tracks).

Other crystals had significantly longer tracks, a few of them up to 14 to 15 μm long. Both the "short-track" crystals and the "long-track" crystals contained tracks of about the same diameter (typically ∿ 0.3 μm). This is an indication that both types of crystals were developed under similar conditions. Track distributions in two such crystals are plotted in Fig. 2; neither crystal had any fat tracks. The short-track crystal had only tracks with a range of up to 10 μm length. This is in precise agreement with the range of U-fission fragments displayed in Table I. The long-track crystal contained a substantial number of tracks having a length in excess of 11 μm. The distribution also shows a sharp drop-off below 9.5 μm. About 30% of the tracks in this crystal were clearly in the range of SHE tracks as calculated and presented for the Z = 114 to 126 region in Table II. Even the presence of some amount of element 154 was not excluded. About 60% of the tracks could clearly be assigned to U-fission fragments.

In a further investigation, 2000 crystals were scanned under the view field of the microscope, and 29 unusual crystals were chosen for further analysis. Each of these 29 contained either only short tracks, only longer tracks, or fat tracks together with thin tracks.

Six of the 29 selected crystals had only thin short tracks; the results of these are plotted in Fig. 3. None of the tracks exceeded 10.5 μm and seem to precisely fit the calculated values for uranium (see Table I).

The measurements of 12 other crystals (of the 29) containing somewhat longer tracks are shown in the histograms of Fig. 4. Approximately 30% exceed the range of U-fission fragments and also the 10.5 μm upper limit of the longest tracks given in Fig. 3. A substantial number of

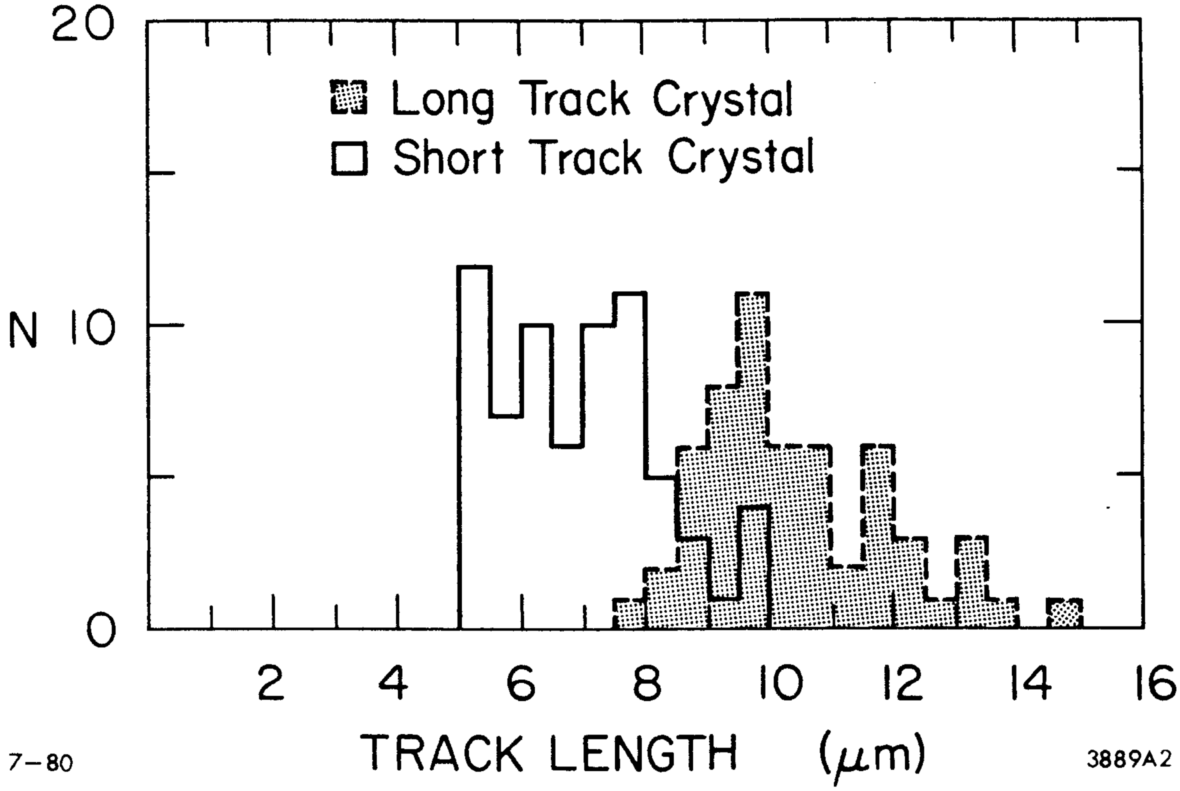

Fig. 2 Histograms of track length distribution in a typical long-track and short-track crystal.

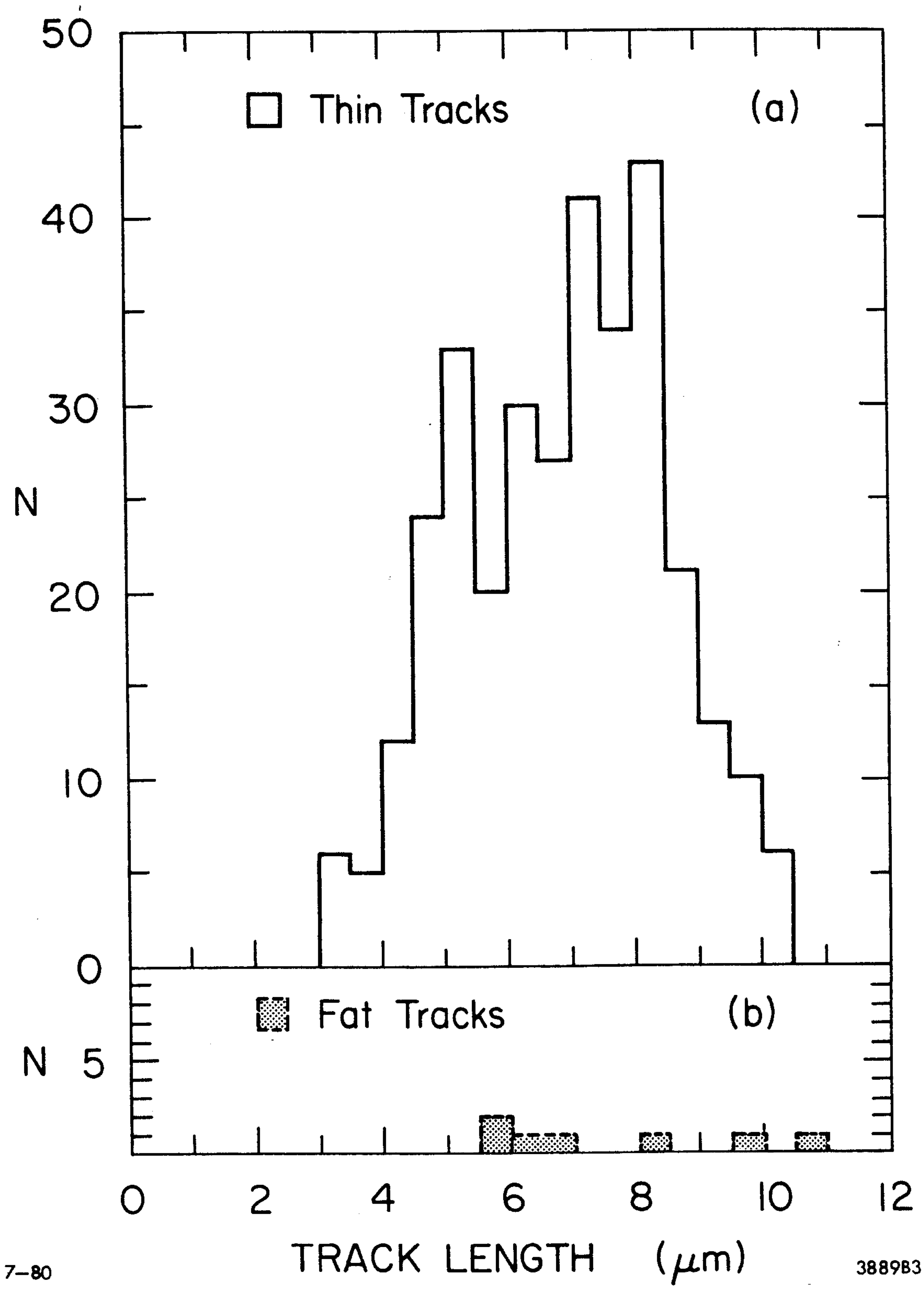

Fig. 3 Histograms of all tracks combined from six short-track crystals.

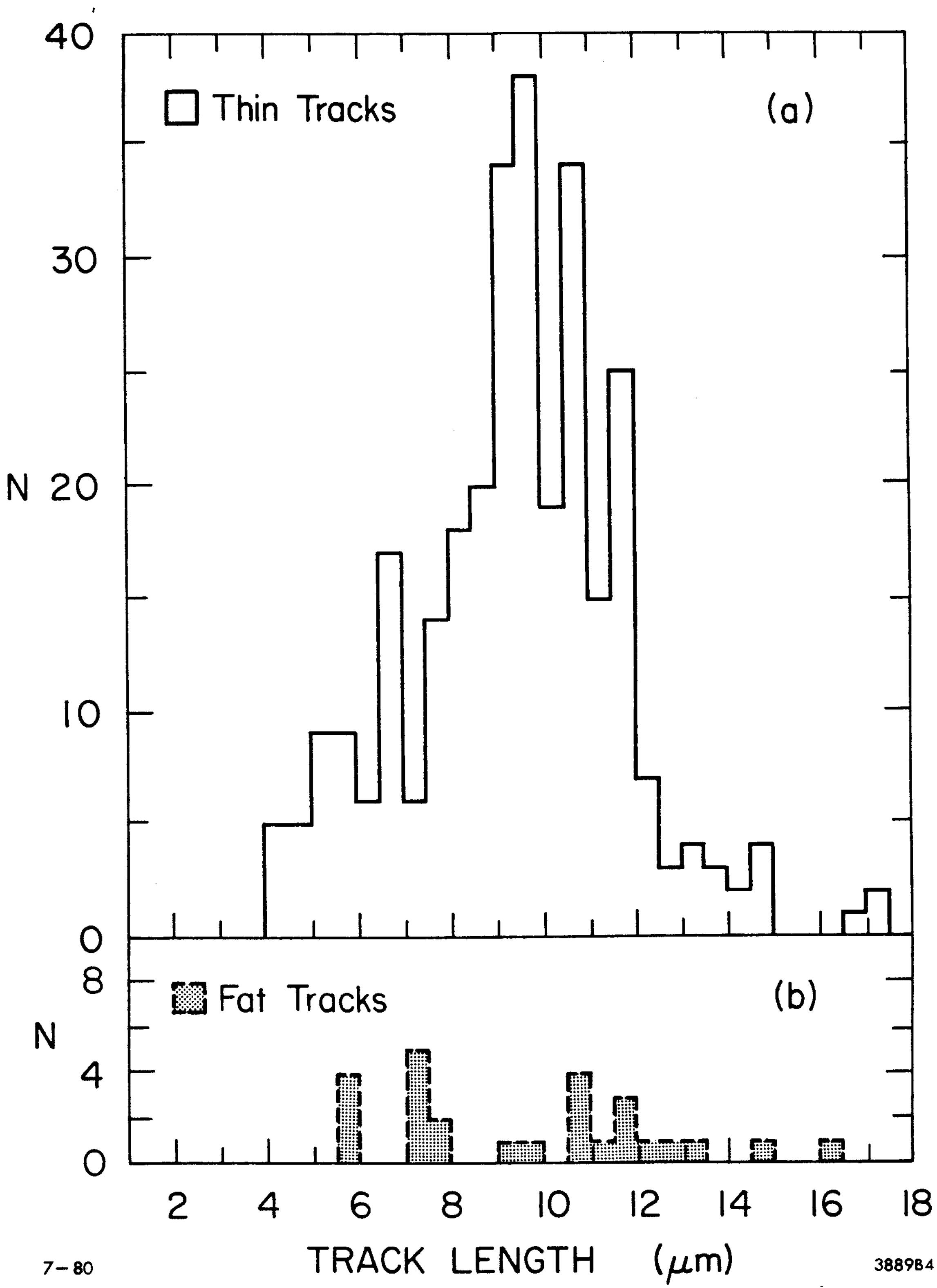

Fig. 4   Histograms of all tracks combined from twelve long-track crystals.

tracks fell into the 12 μm interval; they, as well as most of the even longer tracks, could possibly be the result of SHE-fission from the element 114 region. The longest tracks (∿ 10%) could indicate fission from the 126 and 154 region.

The longest of the fat tracks in Fig. 4b has a range of 16.5 μm and could correspond to fission fragments of elements from the 154 region. These fat tracks were three times as wide as the thin tracks and were probably produced by very heavy fission fragments.

2. Relationship of Track Diameter and Ionization Loss

The relationship between track diameter D(μm) and ionization loss dE/dx of fission fragments (ions) in zircon is not yet precisely known. The accepted empirical equation for the etching velocity or leaching rate V_t(μm/hr) in lexan is (45)

$$V_t = K_1 D = B\left[I(\beta, Z^*)\right]^{1.8} = B\left[K_2 \frac{dE}{dx}(\beta, Z^*)\right]^{1.8} \qquad (3)$$

where B, K_1 and K_2 are proportionality constants, $\beta = v/c$ = ion velocity, c = velocity of light, and Z^* = effective charge of the ion with Z and A. In the absence of specific experimental data, this equation was used to describe H_3PO_4-etching of zircon. It was assumed that Eq. (3) is generally valid for the exponent 1.8, but with different constants B and K_2 than for lexan. Thus, for zircon, Eq. (3) can be rewritten as

$$V_t = K_{1,Zr} D = \frac{dR}{dt} = B_{Zr} I^{1.8} = B_{Zr} K_{2,Zr} \left(\frac{dE}{dx}\right)_{ion}^{1.8} \qquad (4)$$

where V_t = dR/dt is the etching velocity or rate of growth of the radius inside the track; it is perpendicular to the track length. Thus, the diameters of any two tracks developed together in the same crystal can be related to the respective ionization losses of the ions which formed them by

$$D_1/D_2 = (dE/dx)^{1.8}_{av,1} / (dE/dx)^{1.8}_{av,2} \qquad (5)$$

where $(dE/dx)_{av}$ is the average ionization loss over the full length of the track. For each comparison of fat and thin tracks the average range of 10.2 μm for U-fission (as given in Table I for ^{97}Sr and ^{141}Xe) was assigned to the thin, short track. Thus, dividing the total energy $E_{t,U}$ = 172 MeV by the range of 10.2 μm yields the average ionization loss $\rho(dE/dx)_{av,92}$ = 16.86 MeV/μm. Only an average value $(\rho dE/dx)_{av}$ from all the energy deposited is reproduced as track through etching.

The calculated total kinetic energies and track lengths, as given in Table II, were processed in this manner to yield the average ionization loss values as given in Table III. Also presented in Table III are the relative track diameters normalized to asymmetric uranium fission. It can be seen that fragments from element 108 should generate tracks comparable in diameter to uranium tracks or perhaps 25% wider, whereas element 120 should produce tracks ∿ 1.5 times as wide, element 148 ∿ twice as wide, and only fission of element 196 can result in tracks perhaps 3 times as wide as those from U-fission. A few tracks were observed which were 4 times as wide as tracks from U-fission (see Fig. 6). If these tracks are real, it suggests that for zircon etched with H_3PO_4 the exponent in Eq. (5) should perhaps be larger than 1.8 (for example, 2.5). The precise exponent could experimentally be determined in a calibration using heavy ion beams from an accelerator.

Fat, medium and thin tracks, as measured in two crystals (A, B) are presented in the histograms of Fig. 5. It is interesting to note that the

TABLE III

AVERAGE $(dE/dx)_{av}$ FOR URANIUM AND SHE FISSION FRAGMENTS
AND CORRESPONDING RELATIVE TRACK DIAMETERS

Fissioning Nucleus		Equation (1)			Equation (2)		
Z	A	Total Kinetic Energy, E_K MeV	$\left(\frac{dE}{dx}\right)_{av}$ MeV/μm	Relative Diameter of Track	Total Kinetic Energy, E_K MeV	$\left(\frac{dE}{dx}\right)_{av}$ MeV/μm	Relative Diameter of Track
92	236	172 asym	16.85	1.0	172 asym	18.11	1.14
92	236	204	17.51	1.07			
92	236	138	15.45	0.86			
108	286	210.7	18.81	1.22	224.2	18.84	1.22
108	286	274.7	19.35	1.28	288.2	19.34	1.28
120	345	245.0	20.59	1.43	262.7	20.85	1.47
120	345	309.0	20.88	1.47	326.7	21.08	1.50
148	450	340.2	24.30	1.93	369.2	24.78	2.00
148	450	420.2	25.78	2.15	449.2	26.12	2.20
196	594	543.8	29.24	2.70	597.2	30.16	2.85
196	594	623.8	30.28	2.83	677.2	31.21	3.03

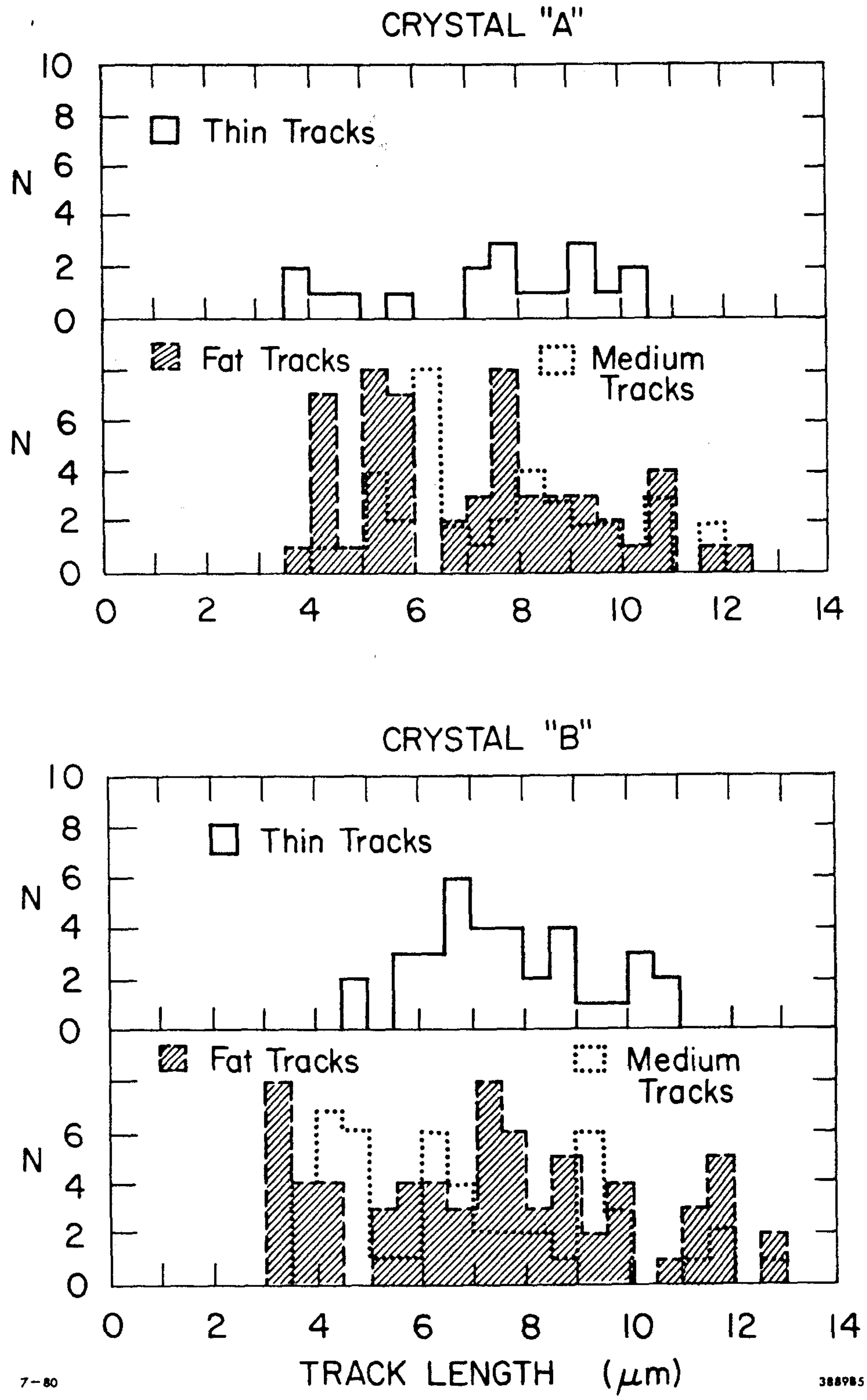

Fig. 5 Histograms of crystals "A" and "B" containing thin, medium and fat tracks.

thin tracks range only up to $\sim$ 11 μm. This is well within the range of U-fission fragments. The fat tracks are 3 to 4 times as wide as the thin tracks, as shown in Fig. 6; the medium tracks have diameters which are in between those of the fat and the thin tracks. The fragments that produced both the fat and medium tracks have a noticeably longer range (up to 13 μm) than those which produced the thin tracks.

An even greater difference exists between the fat and thin tracks in crystal C, as shown in Fig. 7. This crystal has fat and long tracks (up to 14 μm) close to its surface, together with a number of thin tracks developed around the fat tracks (with a maximum range of $\sim$ 9.5 μm).

All these three crystals (A, B, C) contain thin tracks corresponding exactly to tracks from uranium fission fragments right next to longer (13 to 14 μm), fat tracks. The ranges of the fat tracks match those expected for fragments from the element 126-154 region (see Table II). Their diameters more closely resemble those calculated for the element 204 region. As pointed out above, the exponent of 1.8 in Eq. (5) might be low for zircon (H_3PO_4), and the tracks could still be the result of fission from the element 154 region of stability.

Analysis of the data presented in Figs. 5-7 must include an important observation: a few of the thin tracks are only 3.5 to 6 μm long. These tracks penetrate the surface of the crystal. They were produced by a nucleus fissioning either at or close to the crystal surface. Thus only one fragment produced the track and the other (or others) has escaped. Since such "half-tracks" developed only into thin tracks despite good coverage by the etching fluid, they must represent U-fission fragments. The other short but fat tracks in Fig. 5 might be incomplete or "half" SHE-fission tracks.

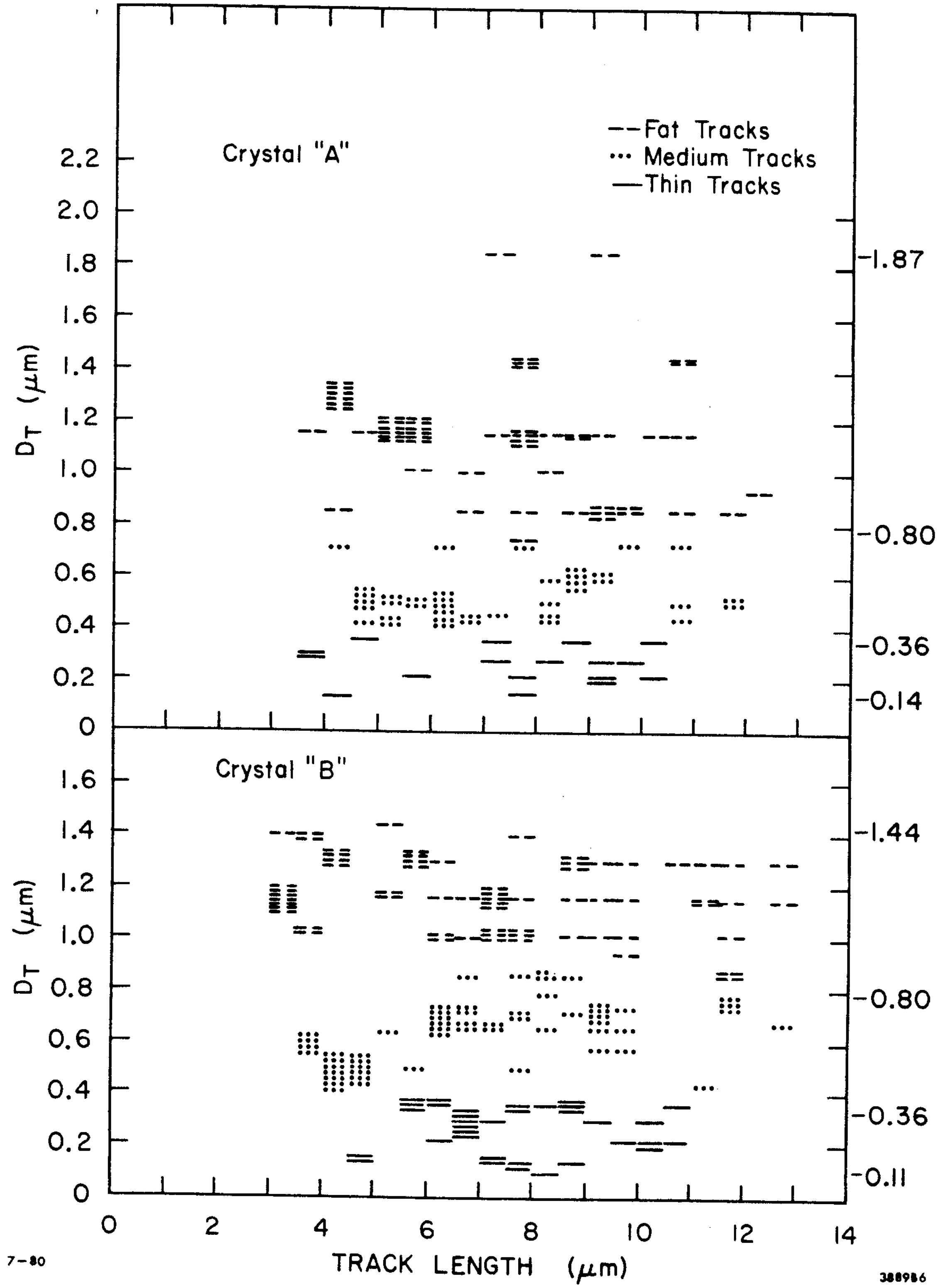

Fig. 6 Measured diameter or width of tracks, D_T, versus measured length of the fat, medium and thin tracks as found in crystals "A" and "B" (from Fig. 5).

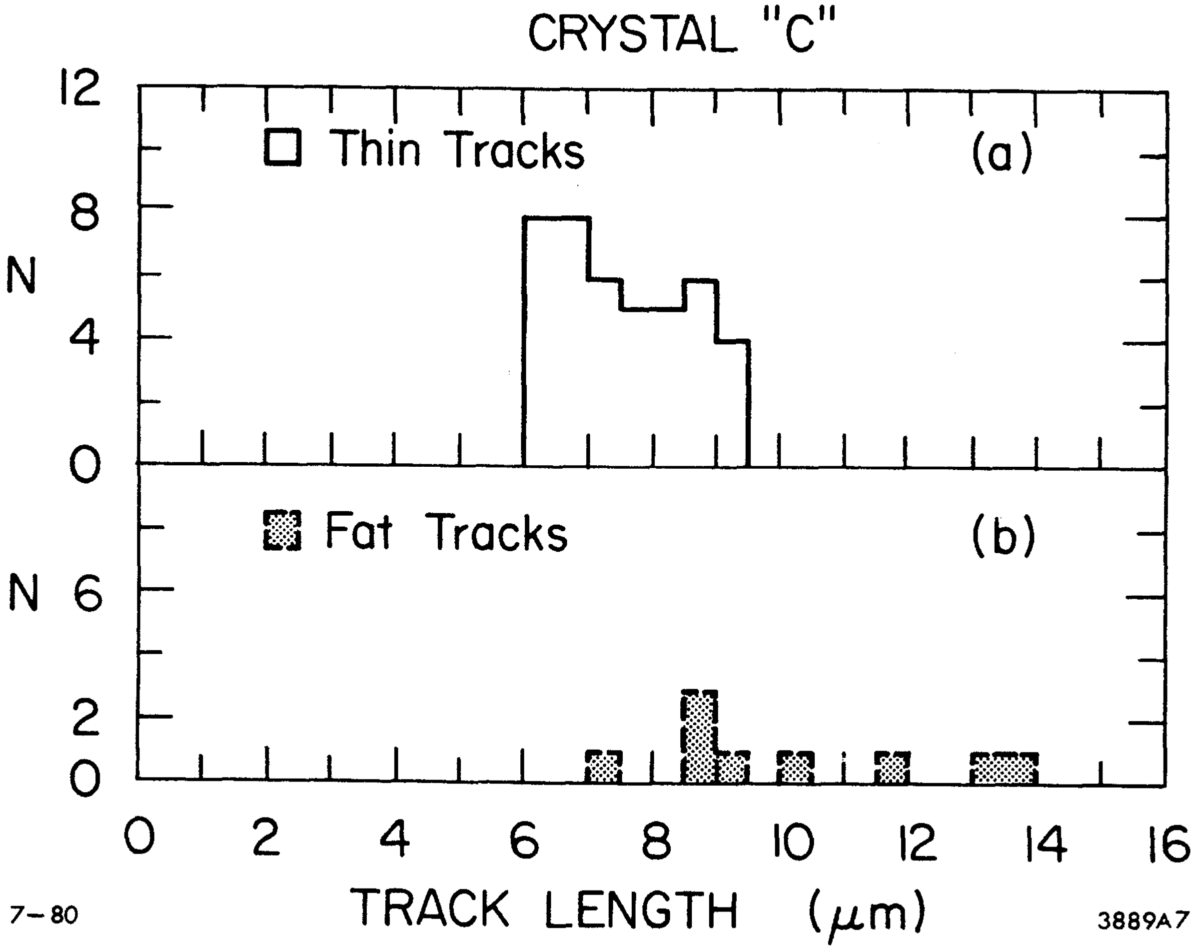

Fig. 7 Histograms of detected number of tracks versus track length in crystal "C"; a) thin tracks; b) fat tracks.

A plausible explanation of the fat tracks in crystals A, B, and C is fission fragments of SHE, similar to the fragments which created the fat tracks in mylar foils (Figs. 2 and 3 in Ref. 30). The data on fat tracks reported herein have better statistics than those reported in (30). Fat tracks of full length (13-14 μm) were observed with good statistics in several crystals (70 fat + 50 medium tracks, for example, in one crystal).

A combined histogram of tracks from 8 crystals containing both long, thin and long, fat tracks is shown in Fig. 8. As in previous histograms, a substantial fraction of the tracks end around the uranium range (10.5 μm). Some of the long tracks reach beyond 20 μm could be explained as fission fragments from the Z = 204 region (see Table III). Among all the tracks observed and catalogued, a total of seven tracks had a length between 17 and 21.5 μm: two thin tracks in Fig. 4a and two fat and three thin tracks in Fig. 8. The long, thin tracks could be explained as deeply buried fat tracks which ended below the crystal surface and were thus only partially developed. No clear (planar) triple tracks were observed.

A brief statistical analysis of occurrence of different types of crystals was performed on about 1000 crystals subsequent to the systematic analysis of the 29 unusual crystals. Also estimated was the length of exposure of the zircon to uranium. The average projected track density, as measured in 100 crystals, was found to be 4.82×10^6 tracks/cm^2. Based on these results, the spontaneous fission rate of uranium, and the known value of the uranium concentration in zircon (measured by fission track counting with the mylar foil technique and found to be 1×10^{-4} g/g), the "fission track age" of this Australian zircon was found to be $(1.4 \pm 0.7) \times 10^8$ years. This was thus the time when the temperature of the zircon

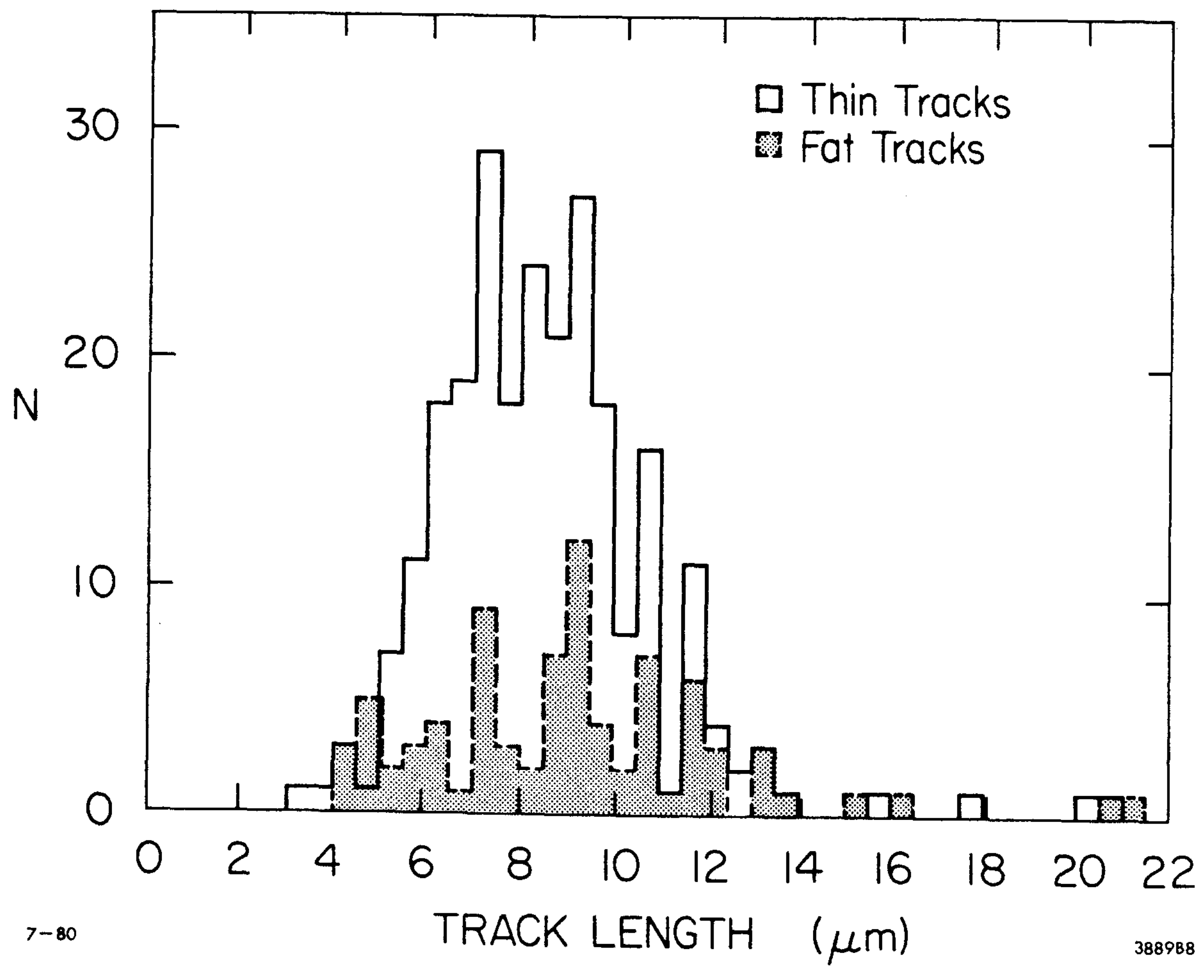

Fig. 8 Combined histograms of eight crystals containing both long thin as well as long fat tracks.

dropped the last time below 570°C, and after which the tracks were preserved. This is in good agreement with (34), which gives a fission track age in several zircons between 0.7 and 1.4 x 10^8 years. Some preliminary conclusions can be drawn from the data presented so far and the subsequent statistical analysis of occurrence:

(a) Approximately 2 fissions/(kg x day) of possible SHE from the element 114 and/or 126 region were observed, i.e., ten percent of the 178 crystals which were analyzed contained 1 to 3 tracks in the length interval of 10 to 13.0 μm;

(b) Approximately 0.7 fission/(kg x day) which generated fat tracks in the crystals were observed, i.e., three percent of the crystals in the analysis contained fat tracks. Of these, about 30% had a length up to 13-14 μm. This value is in good agreement with 0.8 fat track fissions/ (kg x day) as found in technical HfO_2 prepared from Australian zircon (30). This fat track fission activity could be indicative of the existence of the 154 and/or 126 region of SHE;

(c) Approximately 0.35 fissions/(kg x day) were found which created long tracks, i.e., one percent of the crystals included in the occurrence analysis contained up to 30% of long tracks with lengths up to 17 μm. This effect could correspond to SHE-fission from the element 126 and/or 154 region;

(d) Approximately 10^{-3} fissions/(kg x day) were observed which generated very long tracks. For an average of 4.8 x 10^5 tracks in a view field, there were seven tracks with lengths from 17 to 21.5 μm. If real, this effect could be due to fission of nuclei in the Z = 204 region. Another possible explanation is given in the next section.

Recent neutron multiplicity data (46) provide an interesting parallel compared to the effect from (a) above, i.e., 2 fissions/(kg x day) of possible SHE from the element 114 and/or 126 region. In that investigation several monazites showed a small effect, corresponding to an apparent half-life of 1.5 to 2.1 x 10^{21} years for a neutron multiplicity of ν = 6 to 10. This effect was presented only as an upper limit, since the statistical error in the counting corrections was approximately half of this magnitude. However, this effect corresponds to a fission rate of 2.4 to 3.2 fissions/(kg x day). This is almost exactly the same fission rate as that observed in this investigation for 10 to 13 μm-long tracks in zircon (2 fissions/(kg x day), which could be assigned to the element 114 to 126 region. Thus the results in (46) could be an indication of the possible presence of SHE in monazites with a neutron multiplicity of ν = 6 to 10.

If the apparent half-life of SHE concentrations in monazites ($T_{1/2}/C$ = 1.5 to 2.1 x 10^{21} years) is compared to that found in the Allende and other meteorites (47) ($T_{1/2}/C$ = 3.3 x 10^{22} to 3.3 x 10^{23} years), it is evident that the effect in meteorites is only 0.67 to 6.7% of that in monazites. This corresponds to a SHE-fission effect of 6.3 x 10^{-3} events with ν > 3 or 4 per kg-day of meteorite as observed in (47). It appears that further work in meteoric material is not promising.

It is noteworthy to mention that the largest effect in monazites (13 events with ν = 5 neutrons in 29.3 days) was measured in a monazite of Australian origin. The long-track results in (a), (b) and (c) above were observed in Australian zircon. The latter is usually mechanically separated from the same beach sand deposits as the Australian monazite.

3. Observation of "Old Tracks" Without H_3PO_4 Development

Approximately 1% of the zircon crystals contained already before development with H_3PO_4 a number of thin as well as some very fat, often transparent cylindrical tracks which were named "old tracks." They were observed to be mostly cylindrical voids located deep inside the crystal. Heating to 500°C without phosphoric acid did not result in further development. However, after the zircon was raised to 500°C with H_3PO_4, new tracks appeared, often starting as thin "needles" from some of the wide "old" tracks. Many of the "old tracks" were similar in length to the newly-developed tracks, but a few of them were very long. Old tracks from three crystals (D,E,F) are shown in the histograms of Fig. 9. The majority of old tracks in crystal F end at 11-12 μm, but a substantial fraction has a length of up to 20 μm with a very few extending clear out into the range of 50 μm.

If these tracks are just air or gas voids, it is difficult to explain why they "froze" into thin, long cylindrical shape during solidification. A gas bubble of this shape in molten zircon would have quickly shrunk into a spherical geometry. One explanation for these old tracks might be that they are the result of flying fission fragments which evaporated part of the zircon. This would have had to occur at a time soon after crystallization (assumed to have taken place 1.5 to 3 x 10^9 years ago), when the temperature was just below the melting point and the crystals were still partially plastic. If this was so, then the voids are indeed old fission tracks in the crystals. Tracks with a length up to 12 μm could represent fission fragments of, for example, ^{244}Pu. Longer tracks in the 16-μm range could represent fission of element 154, and tracks

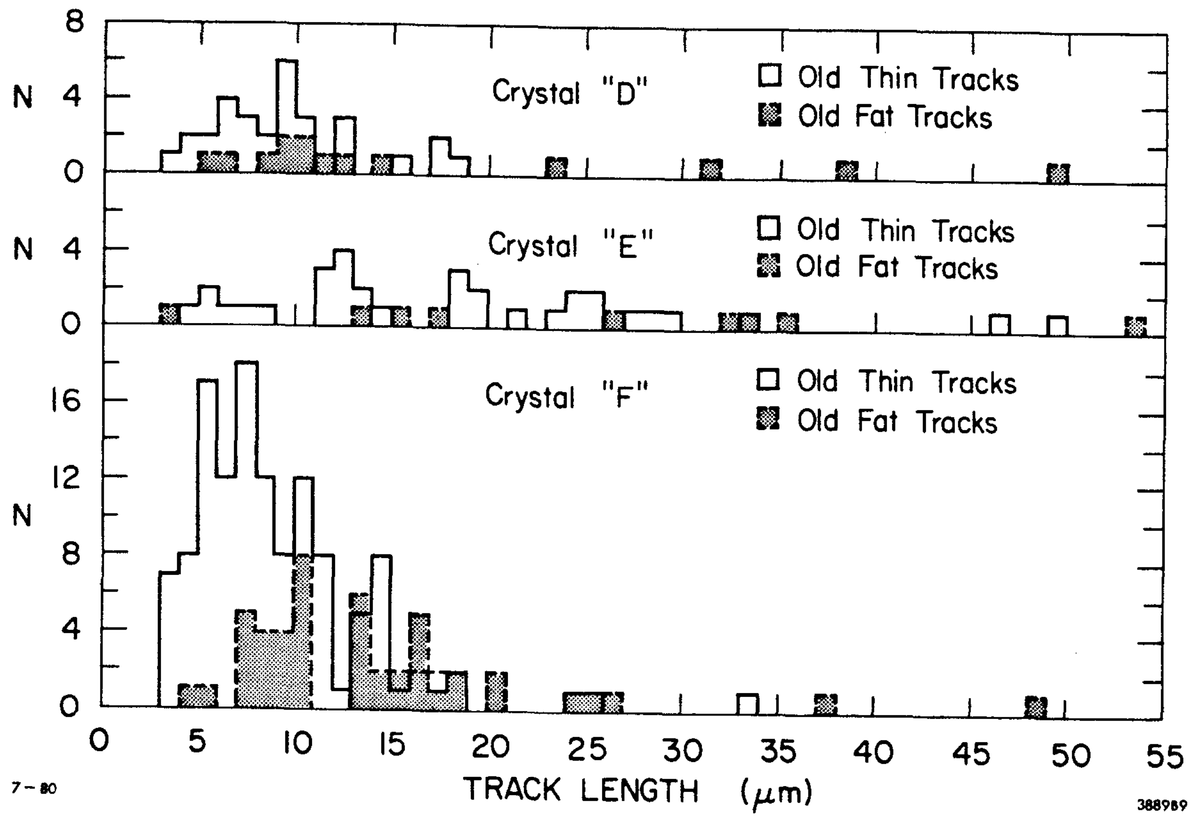

Fig. 9 Histograms of "old tracks" in crystals (D, E, F) not developed with H_3PO_4.

in the 22-μm range might be indicative of fission fragments from the element 204 region. Some SHE could have been present at the time of crystallization of the granite, about 1.5 to 3 x 10^9 years ago. Those tracks which are still longer ($\sim$ 25 μm) might be explained as cracks along the crystallographic planes along which gas collected at the time of crystallization. If all the very long "old tracks" represented fission events, then superheavy elements in the Z = 300 to 400 region would have had to be present at the time of the last zircon solidification. This is not plausible since such SHE are expected to fission into four fragments, whereas the long old tracks are simple cylinders indicative of binary fission only.

Finally, Fig. 10 shows histograms of new tracks developed during the course of this investigation in two crystals (G, H) containing already "old tracks." The new tracks have a length of about 10.5 μm and correspond to the range of U-fission fragments; they were all thin. Some of the old tracks are quite long, up to 41 μm. They were often fat (3-4 μm in width), or they were at least .25 μm in diameter (thin old tracks). The fact that some of the old tracks are also thin and appear much like new tracks might explain the few (four) very long tracks in Fig. 8. It is possible that these long tracks (> 20 μm) were really thin old tracks which penetrated the surface of the crystal. This is unusual for old tracks, which were usually observed deep inside the crystals. All long tracks in Figs. 1 to 8 penetrated the crystal surface (a criterion for new tracks). Another criterion for new tracks was dark color and narrow width. New fat tracks, such as given in Fig. 5, had the shape of an etched rectangular column, whereas the old fat tracks appeared only as simple circular cylinders, often transparent.

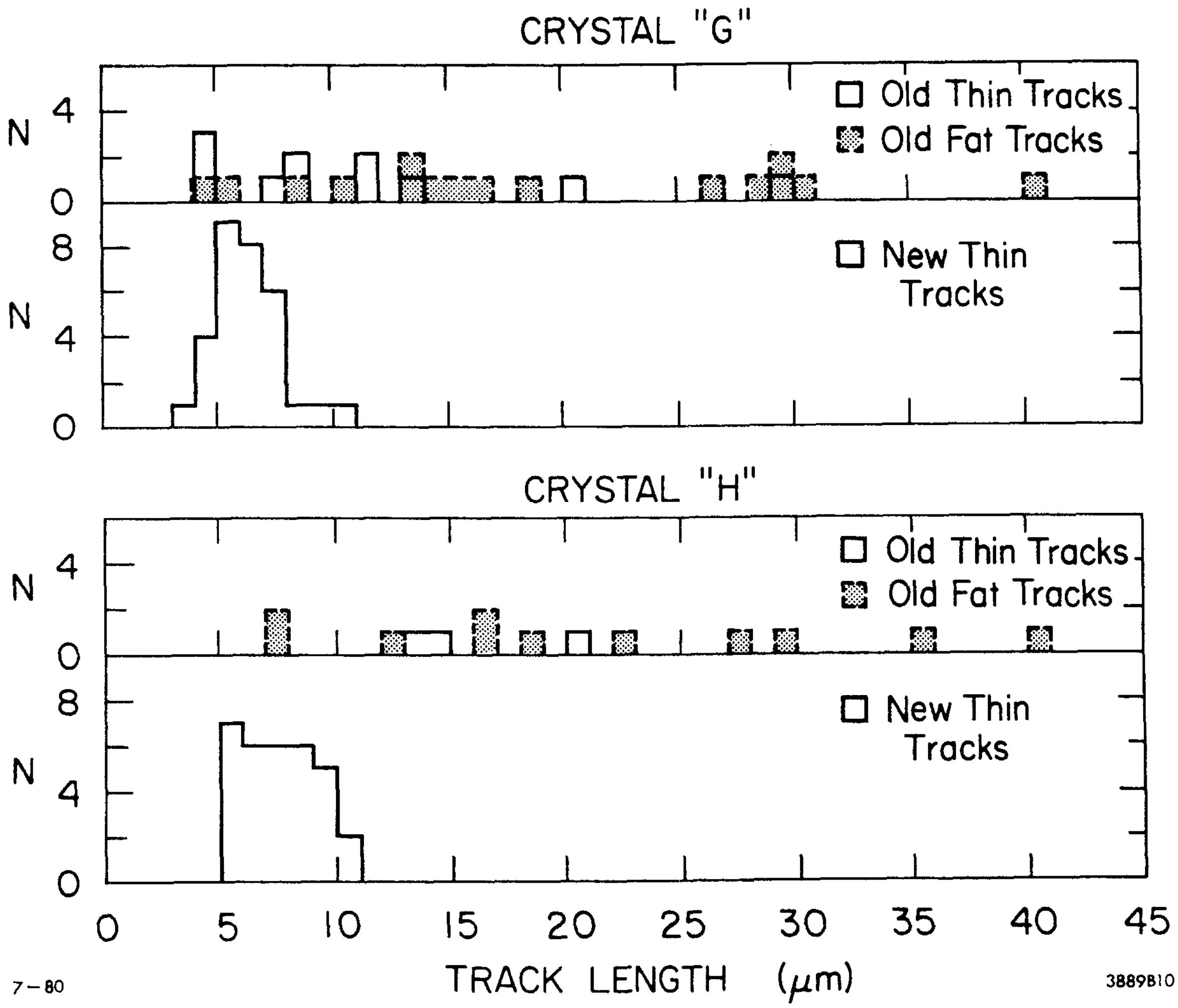

Fig. 10 Combined histograms of "old tracks" together with new tracks developed with H_3PO_4 in two zircon crystals (G and H).

PRESENT CONCLUSIONS

The theoretical literature was referenced for predictions of superheavy elements and probable half-lives. The results of various calculations led to the optimistic conclusion that several islands of stability of superheavy elements might exist, around Z = 114, 126, 154, 164, and perhaps even near Z = 204. While half-life predictions vary by many orders of magnitude, several of them concluded that some isotopes might exhibit long enough half-lives to have survived in nature since the last nucleosynthesis.

The literature was also searched for prospects of superheavy element production in the universe in general and in our solar system or near part of our galaxy in specific. The various processes of nucleosynthesis were mentioned with respect to their potential contribution to the production of superheavy elements. The astrophysical data indicate that there is a finite chance that superheavy elements were, and are, being created in the universe. The r-process or supernovae and neutron star explosions appear to be likely mechanisms and environments for synthesis of such heavy nuclei.

Some pertinent experimental results from the investigation of HfO_2 and radiogenic lead were cited. These chemicals produced long-range and fat (only HfO_2) tracks in mylar foils. The latter were interpreted as being the result of fragments from fission of natural, superheavy elements in the Z = 126 and/or 154 and 114 regions of stability, respectively. The HfO_2 was isolated from zircon minerals. They also contain uranium, from which the radiogenic lead was produced by an alpha-decay chain. Moreover, predicted elements with Z = 126, 154 and 204 are expected to be carried well in zircon. Thus, a thorough investigation of zircons was thought to be justified.

A very sensitive method to detect the existence of spontaneously-fissioning elements in very small abundances is the search for fossil tracks in ancient natural zircon crystals. This method readily offers an exposure time of more than 10^8 years, and concentrations of 10^{-23} to 10^{-26} g/g could be measured. Zircon crystals of Australian origin were etched with phosphoric acid and then analyzed under the optical microscope for the presence of fossil fission tracks. Since uranium is present in zircons at concentrations of 10^{-4} g/g, its fission tracks are a natural, built-in calibrator. In addition to regular uranium fission tracks with a range of up to 10.5 μm in zircon, there were also fossil tracks found with long ranges, 120 to 200% of those of uranium fission tracks. Some of the tracks were also 3 to 4 times as wide as uranium fission tracks detected in the same crystal. These extra-long and/or wide tracks could be explained as the result of spontaneously-fissioning superheavy elements from the predicted regions of stability (around Z = 114, 126, and 154).

Based on the known fission half-life of uranium and its concentration in zircon, the age of the zircons since they were last below 570°C was calculated to be $1.4 \pm 0.7 \times 10^8$ years. The approximate effect of possible superheavy element fissions from the element 114 and/or 126 regions was 2 fissions/(kg x day). The possible effect from the 154 and/or 126 region was found to be 0.7 fissions/(kg x day), which is in good agreement with the effect of 0.8 fissions/(kg x day) found earlier in technical HfO_2 from the same mineral by the mylar foil technique. A small effect ($\sim 10^{-3}$ fissions/(kg x day)) of very long tracks (17-21.5 μm) was also detected. If true, this might be indicative of fission from the element 204 region, perhaps now practically extinct.

The effect of 2 fissions/(kg x day) in zircon, which could be assigned to the element 114 and/or 126 region, closely matches the fission effect recently observed by neutron multiplicity in monazites (46). Several monazites, including one of Australian origin which was separated from the same beach sands as the zircons in this investigation, showed a neutron multiplicity up to $\nu = 10$ (corrected for counter detection efficiency). The effect corresponds to an apparent half-life of 1.5 to 2.1 x 10^{21} years and a fission effect of 2.4 to 3.2 fissions/(kg x day) as an upper limit.

Approximately 1% of the zircon crystals contained some visible tracks already before H_3PO_4 development. A few of these were also of long range and large width. They could also be explained as fission of ^{244}Pu and of superheavy elements present at solidification of zircon.

The relationship between the track diameter and the ionization loss, as well as the energy-range dependence of uranium fission fragments in zircon, is not yet precisely known. Thus, dE/dx in $ZrSiO_4$ was calculated from dE/dx data given separately for Zr, Si and O. All the track data must be viewed in light of this uncertainty. One of the next steps in the quest of superheavy elements should thus be the precise calibration of fossil fission tracks in zircon with regard to experimental length and track diameter, using ions of known energy and measuring their (dE/dx), track length and track diameter. Krypton, xenon and lead ions, and perhaps hafnium or ytterbium ions, exposed to zircon crystals which already contain some probable superheavy element fission tracks, should answer many of the outstanding questions. Further work is in progress.

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