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Author Wollenberg, Harold A.

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Harold A. Wollenberg

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Fission-track radiography of uranium and thorium in radioactive minerals

Harold A. Wollenberg, M.s.

Lawrence Berkeley Laboratory, University of California, Berkeley, California, U.S.A.

Synopsis

The fission-track method is a quick, relatively simple and inexpensive technique for the determination of the location and abundance of uranium and, in some cases, thorium in thin or polished sections of rocks. Thermal neutrons induce fission in 235U, and 232Th and 238U fission with fast-neutron bombardment. Therefore, the sections with appropriate track detectors are exposed, first, to thermal neutrons to induce only U to fission, and then to fast neutrons for fission of U plus Th. The detectors are etched to reveal the damaged areas (tracks) caused by passage of massively charged fission fragments. High-quality muscovite mica is the preferable track detector for minerals with U contents greater than 10-15 ppm-mainly because tracks in mica are easy to recognize. Polycarbonate plastic (Lexan or Makrofol) is preferred as a track detector for low contents of U and Th, because this plastic contains essentially no inherent U; therefore, it has no background track density. Thorium is determined successfully if the Th/U ratio in the mineral is sufficiently large. Relative errors (from counting statistics) in Th are less than 25% if Th/U is greater than 3; for ratios less than 3 the errors increase rapidly and exceed 40-50% if Th/U is less than 1.

The method was applied to the study of U and Th in granitic rocks of the Sierra Nevada batholith, California, and in strongly contrasting mineralized terrains in south and east Greenland. The intense U and Th mineralization of south Greenland is associated with lujavrites, peralkaline rocks wherein the mineral steenstrupine contributes the most to whole-rock radioactivity. Mean values of Th and U in steenstrupine are 23 000 and 6200 ppm, respectively. Adjacent grains show less radioelement variation than is seen in the granitic rocks. Lujavrites of lower radioactivity are characterized by eudialyte, whose U and Th contents are at least an order of magnitude less than those of steenstrupine. The distribution and abundance of radioelements were also investigated in monazite, thorite and pigmentary material of the lujavrites.

Fission-track examinations of thin sections of mineralized fault-zone breccia from east Greenland indicated that U is strongly associated with limonitic material in thin fluoritized veinlets, and is occasionally found concentrated near the edges of reddish opaque grains. By combination of the results of fission-track studies and an electron-microprobe analysis of the minerals, the abundance of U and Th may be correlated with that of other elements.

Determination of the location of radioactive minerals in thin sections of rock has been accomplished for many years by alpha-track autoradiography. Briefly described, *a*-particles radiating from U- and Th-bearing zones near the surface of a thin section are registered on an overlying photographic emulsion, which, when developed, locates sites of radioactivity and permits quantitative determination of their aparticle activity.^{2, 3, 9} A drawback of the technique is that even minerals with relatively high radioactivity (several hundred to a few thousand ppm uranium) require exposure times of the order of ten days to yield a statistically significant number of countable a-tracks. Further, in a collection of α -tracks it is very difficult to differentiate between those from the decay series of Th and those from the U-decay series, and it is

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also possible that there is not secular equilibrium in the U-decay series. Therefore, unless the Th/U ratios of the radioactive minerals are known through independent measurements or are assumed, *a*-particle autoradiography does not furnish independent determinations of U and Th.

The fission-track method, developed in the mid-1960s by Fleischer, Price and Walker,⁷ permits determination of the location and individual abundances of uranium and, under favourable conditions, thorium, in thin or polished rock sections after short exposures (generally a few minutes) to reactor neutrons. Over the past five years several workers have applied the method to study the uranium contents, ranging usually from tens to hundreds of ppm, in thin and polished sections containing accessory and rockforming minerals-see, for example, Kleeman and Lovering¹³ and Wollenberg and Smith.¹⁹ Uranium contents as low as a few tens of ppb have been measured in finely powdered preparations of whole-rock ultramafic samples.⁶ Much less has been accomplished in the study of the occurrence of thorium, because of limitations of the method, which are discussed later.

Fission-track techniques and their physical bases are described. Applications of these techniques are illustrated by a discussion of the distribution of uranium and thorium in contrasting intrusive occurrences. It is intended to illustrate the usefulness of the fission-track method to an understanding of the geochemistry of radioelements in mineralizations of possible future economic importance.

The fission-track method

Significant in the application of the fission phenomena to earth sciences are the facts that (1) ²³²Th and ²³⁸U fission readily under irradiation by fast neutrons; (2) ²³⁵U fissions most readily under thermal-neutron (energy < 0.025eV) irradiation; and (3) ²³⁸U fissions spontaneously (232Th, 235U and 234U also fission spontaneously, but at much slower rates than ²³⁸U). The energy threshold for fission of ²³⁸U and ²³²Th is 0.9 MeV; therefore, in rock samples thermal-neutron irradiation causes only ²³⁵U to fission. The spontaneous fission of ²³⁸U, though necessary for fission-track age determinations of a mineral, also yields a background of fission tracks in natural materials used as track detectors.

A fission track is formed when a massively charged fragment, emanating from a fissioned nucleus, enters the structure of a detector (generally a good insulating solid such as mica, plastic or glass), causing damage by .charge repulsion. The resulting damaged area is en-

hanced by etching the detector with the appropriate reagent: 40-48% hydrofluoric acid (HF) for mica or glass, a strong base (6 N KOH or NaOH) for plastic. One can then view the tracks or damage pits easily with an ordinary microscope.

The formation of tracks depends upon the specific ionization, df/dx,⁸ which determines the number of ions formed along the path of the fission fragment. $d\mathcal{J}/dx$ is a function of the effective charge of the fragment, the velocity of the fragment, the speed of light, the ionization energy of the outer electrons of the detecting material, the mass of the electron and a constant which depends on the detecting material.

For our purposes, fragments from the fission of U or Th, with atomic mass numbers ranging approximately from 90 to 140, and with average energies of 85-90 MeV, register rapidly in muscovite mica, plastic or glass. With proper etching conditions, tracks from the spontaneous fission of ²³⁸U have been observed in several rock-forming and accessory minerals.

If uranium-free detectors are used, the lower limits of detection depend mainly on the magnitude of the integrated neutron flux (regulated in most cases by the duration of the exposure). For example, Fisher⁶ used an integrated flux of $10^{16} n/cm^2$ to measure U concentrations as low as a few ppb. The uranium content and geologic age of natural mica detectors combine to furnish a background track density which, essentially, governs their detection limit—1-5 ppm with an integrated flux of $\sim 10^{13} n/cm^2$. Therefore, such detectors are applicable to neutron-induced fission of U and Th in concentrations from several tens to thousands of ppm.

Computations

The equation which governs the density of fission tracks in a detector placed directly on the flat surface of material containing U, and irradiated with neutrons, is

$$=\varphi\sigma En$$
 (1)

ġ.

ρ = where ρ is the track density, tracks/cm², ϕ is the integrated neutron flux, n/cm^2 , σ is the neutroninduced fission cross-section (~ 580 barns for thermal neutron fission of 235 U; 1 barn = 10^{-24} cm^2), E is the efficiency of detection (0.5 in the present case) and n is the number of ²³⁵U nucleii per square centimetre of thin-section surface within the effective range of fission fragments. For fragments from U and Th fission the effective range in most silicate and phosphate minerals is $\sim 3 \text{ mg/cm}^2$.

· Thus

$$n = (3 \ Ca/A) \times 10^{-3}$$
 (2)

where C is the concentration of radioelement, g/g, a is Avogadro's number and A is the atomic mass number of the radioelement.*

In practice it is not necessary to use the above equations to determine element concentrations if standards are included in the neutron exposure. The equations are most useful in estimating the neutron flux required for an irradiation.

Procedures

The basic procedure to obtain fission tracks from uranium and/or thorium is quite simple. Solidstate track detectors, usually mica or plastic, are mounted on a series of samples and standards, which are then exposed to reactor neutrons. On

Table 1 Comparison of detector materials

Natural muscovite	Polycarbonate plastic
Very durable	Surface scratches easily
Tracks easy to recognize, readily distinguished from scratches	Tracks perpendicular to surface appear as small dots, easily confused with scratches
Etching time, $\frac{1}{2}$ -4 h	Etching time, 10–15 min
Track size regulated by varying etching time without danger of appreciable surface loss	Surface removed readily as etching progresses; not possible to enlarge tracks after 10–15 min
Uranium content plus geologic age yield background tracks from spontaneous and induced fission. This governs detection limit (1–5 ppm U)	Essentially no background tracks, detection limit governed by neutron exposure
	Print of rock surface forms after etching ¹⁴

completion of the exposure, the detectors are removed and etched in the appropriate reagents. The resulting tracks are then viewed and counted with the aid of a microscope.

Choice of detectors

In choosing a fission-track detector one must weigh the advantages and disadvantages of

*The ratio of ²³⁸U to ²³⁵U, 139, is essentially constant in nature.

different materials. This discussion is limited to the two most commonly used in geologic applications: lexan polycarbonate plastic (Lexan or Makrofol) and muscovite mica; they are compared briefly in Table 1. Tracks in muscovite are long hexagons if etched for a short time, or develop into diamond shapes as etching progresses (Fig. 1(a)). Tracks appear in Lexan (Fig. 1(b)) as dark points (from fission fragments entering the

detector perpendicular to the surface) or as dark





Fig. 1 Fission-track etch pits in muscovite (a) (top) and Lexan polycarbonate (b) (bottom). Track density, $2 \cdot 25 \times 10^5 \text{ t/cm}^2$. Mica was etched in 48% HF for $\sim 1 \text{ h}$, the Lexan in $6 \times KOH$ at 60° C for 15 min

lines, the apparent lengths of which depend on the entry angle of the fragment and its starting position in the sample. Lexan's principal advantage is that it has essentially no background uranium content, whereas even the highestquality mica shows a background of tracks from spontaneous and induced fission of its ²³⁸U content. When examining minerals with U contents > 10 to 15 ppm, detector background is not a problem because track densities from the sample are considerably greater than background. If a low content of U is expected in the sample, it may be preferred to use a Lexan detector or expose a mica detector without the sample to obtain the effective background value.

Standards

To furnish track-density-element-content calibrations, and to serve as neutron flux monitors, glass standards containing known amounts of U and Th are irradiated with the samples. The neutron flux may be depressed within a stack of thin-section-detector sandwiches; therefore, standard glasses inserted at the centre and ends of the stack yield close approximations of the true calibration factors for each thin section. Soda disilicate (Na₂O.2SiO₂) is convenient as the standard glass medium, primarily because of its low melting temperature (≈ 1100 °C). The range of fission fragments in glass is roughly the same as in most minerals (approximately 3×10^{-3} g/cm²).

Irradiation

Exposure, processing and observation conditions for different uranium concentrations are summarized in Table 2. The choice of irradiation conditions depends on the element to be determined and its expected concentration. Easily observable track densities range from 10⁴ to 10⁶ tracks/cm². To determine an appropriate thermalneutron flux for a uranium irradiation, desired *

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placed as close as possible to the core of the reactor, and is encased by at least a 3-mm thickness of cadmium to permit irradiation by as 'pure' a flux of fast neutrons as possible. In the flux estimate for a thorium irradiation one must include the large contribution to the observed track density from ²³⁸U.

The estimated integrated neutron flux is then $\varphi = \rho/E(\sigma_{Th}n_{Th} + \sigma_U n_U)$ (3) where ρ is the desired track density, σ_{Th} and σ_U are the fast neutron (neutron energy ≈ 3 MeV) fission cross-sections for ²³²Th (≈ 0.3 barn) and ²³⁸U (≈ 1.1 barn), respectively, and n_{Th} and n_U are the estimated concentrations of Th and ²³⁸U nucleii.

Etching

Etching time is varied to achieve an optimum track size for a given track density. This is most important if tracks are counted from photographs of the detectors. In this respect mica has an advantage over Lexan in that track size in mica can be controlled by HF etching time; very little of the mica's surface is removed, even after etching for several hours. After 10–15 min the strong base etchant used for Lexan rapidly removes its surface, causing a continuous depletion of tracks with time—first the shallower tracks, then those of greater penetration.

Observation

To determine the location of the sites of radioelements the detector with the developed tracks can be replaced on the thin section, or matching

Table 2	Exposure,	processing	and	observation	conditions	for	various	evenly	distributed	uranium	concentrations
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U content, ppm	Neutron exposure, $n/\text{cm}^2 \times 10^{13}$	Track density, t/cm^2	Etching time, h*	Magnification	Tracks/field
50	2	1.2×10^{4}	3–4	× 36	~500
500	1	6×10^{4}	2 - 3	$\times 80$	$\sim \! 450$
5000	1	6×10^{5}	1 - 2	$\times 270$	~ 400

*Etching of high-quality muscovite mica in 40-48% HF at room temperature.

track density is combined with the estimated U concentrations of the samples in equations 1 and 2. The samples are exposed in the thermal column of the reactor (a position outside the core, surrounded by graphite to exclude fast neutrons), where the thermal/fast neutrons ratio is highest (preferably ≥ 1000).

Conversely, a fast-neutron irradiation is used for thorium. The sample-detector package is areas of sample and detector can be photographed separately and the pictures compared. The image of the rock surface on Lexan, observed by Kleeman and Lovering¹³ after intense neutron irradiations, aids in rematching plastic detectors to thin sections following etching. Otherwise, small fiducial marks on mica or plastic detectors and thin sections may be matched, or obvious track-density patterns matched with their mineral

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grain contributors. Associated track densities can be counted in the microscope, from photographs, or, if contrast and spacing permit, by machine. In the case of a thermal-neutron exposure for U, sample and standard track densities are compared directly to give the uranium content. Errors are from track-counting statistics.

Thorium determination

The use of fast neutrons permits a determination of Th content if the Th/U ratio is great enough. Since practically all Th-bearing minerals also contain some U, it is necessary, first, to determine U by a thermal-neutron exposure. Then the sample, with a new detector and U and Th standards, is exposed to fast neutrons (energies >1 MeV, usually 3–4 MeV). The resulting track density is from ²³⁸U and ²³²Th, but, knowing the U content, and having simultaneously exposed a U standard, the U contribution is determined. The contribution from Th is then the difference between the total track density and the contribution from U.

The main difficulty with this method is that the relative errors due to counting statistics are rather large; the cross-section ratio for fast neutrons favours U over Th by a factor of ~ 3 . The error in the thorium determination comes from the accumulation of counting statistics in measurements of U and Th standards, and from measurements of the samples' track densities from fast- and thermal-neutron exposures. Fig. 2 relates Th/U ratios and relative errors in Th for typical exposure and counting conditions of accessory-mineral samples. Relative errors are < 25% if Th/U is greater than ~ 3 ; for ratios < 3the errors increase rapidly, and they are considered unacceptably high (>40%) for Th/V < 1.

Results and discussion

The fission-track method has been applied to the study of the distribution of radioelements in calcalkaline granitic rocks of the Sierra Nevada batholith, California; in peralkaline rocks of the Ilímaussag intrusion, south Greenland; and in a mineralized fault zone in east Greenland. A brief review of these studies serves to demonstrate the usefulness of the method in discerning the geochemistry of radioactive mineral assemblages.

Radioactive minerals of the Sierra Nevada batholith

As part of a comprehensive investigation of the distribution of radioelements in the granitic





Fig. 2 Th/U ratio versus relative error in Th for typical exposure and counting conditions

rocks of the Sierra Nevada batholith,19 the writer determined the locations and abundances of uranium and thorium in rock and grain-mount thin sections. Photomicrographs of fission tracks from uranium registered in mica, superimposed on thin sections, are given in Fig. 3.

In the mica-rich Sierran samples U is associated mainly with biotite, occurring near the edges of grains or along dislocations within the grains. In other Sierran granitics U and Th are found primarily within the non-magnetic accessory minerals. In 11 of 13 samples, representing rocks typical of the batholith's various plutons (wholerock averages: U, 3.9; Th, 14.0 ppm), sphene, averaging 250 ppm U, contributes from 9 to 38% of the rocks' uranium (F. C. W. Dodge, U.S. Geological Survey, private communication); the abundance of sphene does not exceed 0.6%. (Sphene makes up less than 0.004% of the other two samples: thus, it is not a significant contributor to their whole-rock U.) Zircon, though less prevalent than sphene, is an important contributor to whole-rock uranium.

Only five of the samples contain appreciable thorium in sphene. Although thorium has not been investigated extensively in grain mounts of other Sierran accessory minerals, examination



Fig. 3 Fission tracks in detecting mica superimposed on uranium-bearing minerals of Sierra Nevada granodiorites: (a) tracks associated with U, rimming the end of a grain of biotite and at discontinuities within the biotite; (b) tracks associated with U (120 ppm) in a grain of sphene; (c) tracks from U (160 ppm) in sphene, encompassed by hornblende

of thin-section radiographs indicates that it is concentrated in monazite and allanite. Two

grains of allanite from a high-radioactivity granodiorite contain 8900 and 7200 ppm Th; U is, respectively, 237 and 217 ppm. In many instances uranium and thorium vary by factors of two to four between grains of the same accessory mineral within a single thin section.

Radioactive minerals of the Ilímaussaq intrusion

The radioelement and rare-element contents of the strongly differentiated Ilímaussaq alkaline intrusion^{1, 10, 11, 15, 18, 20} have been intensively studied. The intrusion (age, ca 1000 m.y.) consists of an early augite syenite followed by a sequence of peralkaline nephelene syenites.¹⁷ Lujavritic zones contain relatively high concentrations of U, Th, Zr, Nb and rare-earth elements. The highest radioelement concentrations occur in rocks of the Kvanefjeld plateau, near the intrusion's northwest border, where lujavrites are in contact with sheared cap-rock lavas. In this area steenstrupine* is the most abundant radioactive mineral; whole-rock uranium ranges from a few hundred to 800 ppm, and thorium from 200 to 2000 ppm. In the southern part of the intrusion lujavrites, rich in eudialyte[†] and containing appreciable Zr, have U and Th in the range of 30-60 ppm. Also examined by the fissiontrack technique were monazite, thorite and uranium-rich pigmentary material in lujavrite of the Kvanefjeld plateau.

Steenstrupine

Steenstrupine is characterized by a broad range in the amount of U and Th occurring, even from grain to grain, in a single thin section. This is exemplified in samples from a borehole drilled in lujavrites of the Kvanefjeld plateau. In a single thin section of rather homogeneous-appearing brown steenstrupine U ranges from 2700 to 7000 ppm and Th from 13 700 to 25 700approximately factor-of-two differences between proximate grains. There is close agreement, however, between the overall average Th/U (3.1) in steenstrupine of the thin sections of the borehole with a Th/U ratio of 2.7 in corresponding whole-rock samples. This substantiates the conclusion from observation of the thin sections that in this lujavrite the abundance of steenstrupine essentially controls whole-rock uranium and thorium. The ratios of whole-rock and steenstrupine Th and U contents indicate that this mineral makes up to 2-4% of the rock.

 $\label{eq:starses} \begin{array}{l} *Na_2Ce(Mn,Ta,Fe)H_2((Si,P)O_4)_3.\\ \dagger(Na,Ca,Fe)_6Zr(OH,Cl)(Si_3O_9)_2. \end{array}$

The distribution of U and Th within a large steenstrupine grain from a pegmatite vein is illustrated graphically in Fig. 4. The traverse shows a rather uniform distribution of uranium, whereas thorium varies by a factor of two to three. The error bars associated with the thorium points are from the counting statistics.



Fig. 4 Distribution of U and Th in profile across a large steenstrupine grain from a pegmatite

Other steenstrupine grains are distinguished by their marked zoning, which is exemplified in reddish steenstrupine (described by Buchwald and Sørensen³). Here the centres of the grains appear somewhat isotropic under crossed nicol prisms, whereas the outer areas are anisotropic (Fig. 5). Buchwald and Sørensen noted a marked difference in α -particle activity of these grains: higher activities occurred in the central portions and lower in the anisotropic borders. This difference is reflected in the U and Th contents, which are approximately twice as great in the isotropic centres as in the border areas.

The frequency distribution of radioelements can be plotted from fission-track radiography data. Fig. 6 gives histograms of U, Th and Th/U in steenstrupine, illustrating the broad variations in uranium and thorium in this mineral especially from samplings of a 160-m portion of the Kvanefjeld borehole core.

Eudialyte

There are large differences in the uranium content of eudialyte in different rock types. Uranium, averaging 209 ppm, appears rather evenly distributed in unaltered eudialyte from a borehole (Fig. 7) drilled in 'green' (aegirine-arfvedsonite) lujavrite in the southern portion of the Ilímaussaq intrusion. This contrasts with considerably lower U (50-100 ppm) in eudialyte closely associated with steenstrupine in a borehole on the Kvanefjeld plateau. The latter eudialyte has a range of U similar to the 60 ppm reported



Fig. 5 Zoned reddish steenstrupine showing fission tracks associated with U concentrated in the grain's isotropic centre (4200 ppm), and a lower U content in the rim zone (2000–3000 ppm). Field diameter, $1 \cdot 2 \text{ mm}$

by Hamilton¹⁰ in a sample from the northern portion of the intrusion. It appears that essentially all of the radioactivity in lujavrite of the southern part of the Ilímaussaq intrusion is contributed by U and Th in eudialyte. The ratio of average U contents of eudialyte to average U in corresponding whole-rock samples (35 ppm) is approximately 6—in rough agreement with an estimate by Ferguson⁵ of 10% eudialyte in green lujavrite.

Pigmentary material

Pigmentary material closely associated with arfvedsonite laths in lujavrite of the Kvanefjeld plateau (Fig. 8) contains up to 2.5% U, with little or no thorium. (This material is similar to that described by Hansen¹¹ as occurring in radioactive veins near the Ilímaussaq intrusion.) This strong contribution solely from uranium, superimposed on those of Th and U in steenstrupine, may account for the relatively low whole-rock





Fig. 6 Frequency distributions of U, Th and Th/U in steenstrupine: lined areas represent steenstrupine from a borehole on the Kvanefjeld plateau; unlined areas represent reddish and pegmatitic steenstrupine

Th/U ratio (1.54) observed in some arfvedsonite lujavrites of the Kvanefjeld.¹⁵ The apparent close association of pigmentary material with



Fig. 7 Unaltered euclidyte in aegirine-arfvedsonite lujavrite showing a rather even distribution of U (~200 ppm). Field diameter, 1.2 mm

Table 3	Comparison of	a-track	data with	radioelement contents	
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Description	$\alpha \ tracks/cm^2/h*$	U, ppm	Th, ppm	
Steenstrupine in lujavrite,				
Kvanefjeld area	1300-12 000	820-15 000	1720-40 000	
Sample 18467b-1, zoned red steens	trupine:			
anisotropic borders	3100-5300	2000-2700	7700-20 000	
isotropic centres	6200-8300	4200-7200	33 000	
			(1 measurement)	
Eudialyte in green lujavrite,				
southern area	100-640	210†		

*From Buchwald and Sørensen.³

†Mean value of borehole samples.

arfvedsonite, a mineral characterized by a predominance of Fe^{2+} over Fe^{3+} , may indicate reducing conditions in favour of the selective localization of uranium.

Thorite

Material considered likely to be thorite has been observed in veinlets of the Kvanefjeld.¹⁵ In thin

section (Fig. 9) it appears light yellowish-brown, and is associated with greenish cloudy monazite. The high thorium (28-55%) and uranium contents (2-3%) determined by fission-track radiography are consistent with those reported for thorite in reference texts.¹⁶



Fig. 8 Pigmentary material (1.4-2.6% U) in and between grains of arfvedsonite in lujavrite. Field diameter, 1.2 mm

Monazite

Monazite, which occurs generally in cloudyappearing aggregates of small elongate crystals (Fig. 10), was examined in some drill cores and surface samples of lujavrite from the Kvanefjeld plateau. Monazite associated with the thorite described above contains approximately 25% Th, and has a high Th/U ratio. Considerably lower Th contents (1-2%) were measured in monazite from medium- to coarse-grained lujavrite of the Kvanefjeld; Th/U ratios in this monazite fall in the range 9–14. These are similar to the ratios in thorite, and contrast with the considerably lower Th/U measured in steenstrupine in the same thin sections.





Fig. 9 Thorite (28-55% Th, 2-3% U) associated with greenish cloudy monazite in a veinlet in lujavrite. Large tracks are from a 16-h etch prior to the fast-neutron exposure. Field diameter, 1.2 mm

Comparison with alpha-track autoradiographic data

A comparison of uranium and thorium values with corresponding α -track densities (Table 3), reported by Buchwald and Sørensen,³ indicates relatively good concurrence. Low α -track densities of eudialyte from the green lujavrite mentioned earlier correspond to U ranging from 160 to 410 ppm. Considerably higher α -track densities, varying over nearly a factor of 10 in steenstrupine of Kvanefjeld lujavrite, match broad ranges in U and Th in thin sections from the boreholes.

There is also good agreement between the U and Th contents and the α -track data for isotropic and anisotropic zones in the red steenstrupine shown in Fig. 5. To directly compare Buchwald and Sørensen's observed α -track densities with those calculated from fission-track data the mean U and Th values for the anisotropic and isotropic zones were entered in the formula of Coppens,⁴ which relates radioelement



Fig. 10 Fission tracks from U plus Th (U ~ 1200 ppm, Th ~ 12 000 ppm) in monazite (grey) with arfvedsonite (black) in medium to coarse lujavrite. Large tracks are from a 16-h etch prior to the fast-neutron exposure. Field diameter, 1.2 mm

content with α -track density. The resulting values, 8200 and 3500 $\alpha/\text{cm}^2/\text{h}$ for isotropic and anisotropic zones, respectively, fall within the reported α -density ranges.

An east Greenland fault-zone occurrence

The distribution of uranium in thin sections from a mineralized fault zone was investigated by the fission-track method. The fault forms the boundary between crystalline rocks of the Caledonian fold belt to the west and Palaeozoic–Mesozoic sedimentary deposits of Jameson Land to the east. The geology of the area has been described in general terms by Kempter.¹² The samples from which the thin sections were prepared consist of moderately to strongly oxidized fault breccia, mineralized with varying intensity, principally by fluorite and barite. Megascopic examinations, coupled with γ -ray spectrometric determinations, indicate that the concentration of whole-rock uranium generally varies with the intensity of fluoritization.

From fission-track radiography (Fig. 11) it is apparent that U is closely, but not always directly, associated with the fluorite. U also occurs with yellowish to brownish limonite that lines fluorite veinlets, is intergranular between



Fig. 11 Strong association of U (up to 4000 ppm) with limonite (light grey) and fluorite (dark and intermediate) in fluoritized fault-zone breccia, east Greenland. Field diameter, 1.2 mm

fluorite grains, or exists separately from the veinlets. Uranium concentrations in this material range from several hundred to more than 4000 ppm. Less common associations of uranium are illustrated by very intense fission-track densities, occurring with yet unidentified translucent grains, and with U enrichment at the margin of reddish semi-opaque grains, possibly hematite (Fig. 12).

There is little consistency between whole-rock uranium contents and those of the mineralized zones in the thin sections. In this environment uranium is confined to thin veinlets, which are very unevenly distributed within the fault-zone breccia.



Fig. 12 Enrichment in U (2500 to 3000 ppm) at margin of an opaque grain (hematite?) in fluoritized fault-zone breccia, east Greenland. Field diameter, 0.88 mm

Summary and conclusions

The fission-track method is a quick, simple and inexpensive way to determine the location and abundance of uranium, and, in some cases, thorium, in uncovered thin sections. The method can be used to determine accurately U concentrations ranging from several ppb up to several per cent. The accuracy for the determination of thorium is quite poor when Th/U < 1, but improves to about 25% (relative error) when Th/U > 3.

Fission-track radiography of granitic-rock thin sections from the Sierra Nevada batholith indicates that uranium associates most strongly with biotite in rocks rich in this mica; otherwise, U occurs most frequently with the accessory mineral sphene, and Th with monazite and allanite. Such associations suggest that the uranium and thorium balance in a rock unit may be obtained by combining data from whole-rock γ -ray spectrometric analyses with data from fission-track measurements on populations of separated mineral grains.

The fission-track results and recent wholerock y-ray spectrometric measurements in the Ilímaussaq intrusion¹⁵ indicate that the abundance of steenstrupine governs whole-rock radioactivity in lujavrites of the Kvanefjeld area, and eudialyte controls radioactivity in agpaitic rocks in the remainder of the intrusion. In lujavrite large disparities were observed between radioelement contents of steenstrupine grains only a few millimetres apart. Uranium varies much less in eudialyte of the aegirine-arfvedsonite drill core from the southern portion of the intrusion. Measured uranium and thorium contents of steenstrupine agree well with α -track densities observed in autoradiographs by Buchwald and Sørensen.³

In a hydrothermally mineralized fault zone in east Greenland fission-track radiography reveals that uranium, in concentrations up to several thousand ppm, occurs closely associated with limonitic material in strongly fluoritized veinlets. Thus, the intensity of fluoritization generally controls whole-rock radioactivity.

As with almost all geochemical techniques, the fission-track method is most valuable when its results are combined with chemical data obtained by other methods, e.g. electron-microprobe examinations that disclose the distributions of rareearth elements, Zr and Ti in accessory minerals, and reveal the chemical nature of uranium-rich pigmentary material. Microprobe data would aid in identifying scattered uranium-rich minerals such as those observed in an east Greenland fault zone.

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References

1. BOHSE, H. BROOKS, C. K. and KUNZENDORF, H. Field observations on the kakortokites of the Ilímaussaq intrusion, southwest Greenland, including mapping and portable X-ray fluorescence analyses for zirconium and niobium. *Rep. geol. Surv. Grønland*, in press.

2. BOWIE, S. H. U. Nuclear emulsion techniques. In *Nuclear geology* FAUL, H. ed. (New York: Wiley, 1954), 48-64.

3. BUCHWALD, V. and SØRENSEN, H. An autoradiographic examination of rocks and minerals from the Ilímaussaq batholith, South West Greenland. *Meddr Grønland*, **162**, no. 11, 1961, 43 p.

4. COPPENS, R. Sur la mesure de la radioactivité des roches par l'émulsion photographique. *Bull. Soc. fr. Minér. Cristallogr.*, **75**, 1952, 57–8.

5. FERGUSON, J. Geology of the Ilímaussaq alkaline intrusion, South Greenland. *Meddr Grønland*, **172**, no. 4, 1964, 83 p.

6. FISHER, D. E. Homogenized fission track determination of uranium in whole rock geologic samples. *Analyt. Chem.*, **42**, 1970, 414–6.

7. FLEISCHER, R. L. PRICE, P. B. and WALKER, R. M. Tracks of charged particles in solids. *Science*, *N.Y.*, **149**, 1965, 383–93.

8. FLEISCHER, R. L. *et al.* Criterion for registration in dielectric track detectors. *Phys. Rev.*, **156**, 1967, 353–5.

9. HAMILTON, E. The distribution of radioactivity in the major rock forming minerals. *Meddr Grønland*, **162**, no. 8, 1960, 41 p.

10. HAMILTON, E. The geochemistry of the northern part of the Ilímaussaq intrusion, S.W. Greenland. *Meddr Grønland*, **162**, no. 10, 1964, 104 p.

11. HANSEN, J. A study of radioactive veins containing rare-earth minerals in the area surrounding the Ilímaussaq alkaline intrusion in South Greenland. *Meddr Grønland*, **181**, no. 8, 1968, 51 p.

12. KEMPTER, E. Die Jungpaläozoischen Sedimente von Süd Scoresby Land. *Meddr Grønland*, **164**, no. 1, 1961, 125 p.

13. KLEEMAN, J. D. and LOVERING, J. F. Uranium distribution in rocks by fission-track registration in Lexan plastic. *Science*, *N.Y.*, **156**, 1967, 512–3.

14. KLEEMAN, J. D. and LOVERING, J. F. Lexan plastic prints: how are they formed? *Radiat. Eff.*, **5**, 1970, 21–6.

15. Løvborg, L. *et al.* Field determination of uranium and thorium by-gamma-ray spectrometry, exemplified by measurements in the Ilímaussaq alkaline intrusion, South Greenland. *Econ. Geol.*, **66**, 1971, 368–84.

16. NININGER, R. D. *Minerals for atomic energy* (New York: Van Nostrand, 1954), 367 p.

17. SØRENSEN, H. On the magmatic evolution of the alkaline igneous province of South Greenland. *Rep. geol. Surv. Grønland* no. 7, 1966, 20 p.

18. SØRENSEN, H. HANSEN, J. and BONDESEN, E. Preliminary account of the geology of the Kvanefjeld area of the Ilímaussaq intrusion, South Greenland. *Rep. geol. Surv. Grønland* no. 18, 1969, 40 p.

19. WOLLENBERG, H. A. and SMITH, A. R. Radiogeologic studies in the central part of the Sierra Nevada batholith, California. *J. geophys. Res.*, **73**, 1968, 1481–95.

20. WOLLENBERG, H. A. KUNZENDORF, H. and ROSE-HANSEN, J. Isotope-excited X-ray fluorescence analyses for Nb, Zr, and La+Ce on outcrops in the Ilímaussaq intrusion, South Greenland. *Econ. Geol.*, **66**, 1971, 1048–60.

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