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Uranium and selected trace elements in granites from the Caledonides of East Greenland

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ABSTRACT. The Caledonian fold belt of East Greenland contains calc-alkaline granite (*sensu lato*) intrusions with ages ranging from *c.*2000 Ma to *c.*350 Ma. The Proterozoic granites have low U contents and the pre-Devonian Caledonian granites contents of U corresponding to the clarke value for U in granites. Some aspects of the geochemistry of U are discussed using U-K/Rb, U-Sr, U-Zr, and U-Th diagrams. Secondary enrichment and mineralization occurs in fractured and hydrothermally altered granites and rhyolites situated in or near a major NNE fault zone. The U is associated with iron oxides or hydrocarbons. It is suggested that the source of the mineralization was Devonian acid magma, which also acted as a heat source for circulating hydrothermal fluids.

THE content and distribution of U in granites from the Caledonides of Scotland, Ireland, and the eastern United States of America have recently been studied in relation to the geochemistry and tectonic setting of the granites, and to the occurrence of U mineralization (e.g. Simpson *et al.*, 1979; Rogers *et al.*, 1978). This paper presents the results of preliminary studies of granites of the East Greenland Caledonian fold belt, which can be viewed as a northwards continuation of the British Caledonides.

Geological setting

The East Greenland Caledonian fold belt extends from latitudes 70 to 82° N, and occupies the land between the inland ice and the coast (fig. 1). The fold belt includes Archaean and early Proterozoic gneiss complexes, as well as middle and late Proterozoic sedimentary sequences which are variously affected by Caledonian deformation, metamorphism, and plutonism. The western Caledonian marginal thrust approximates to the boundary of the inland ice and exposures of the foreland are rare. Continental sediments of Devonian to lower Permian age overlie the folded rocks and Upper Permian and Mesozoic marine sediments are preserved in the coastal zone from 72 to 76° N.

The pattern of Caledonian deformation, metamorphism and plutonism in East Greenland (Henriksen and Higgins, 1976) has much in common with the British Caledonides as indicated by Higgins and Phillips (1979). Major structural features of the British Caledonides, such as the Moine thrust and the Great Glen Fault, have their counterparts in East Greenland, in the marginal thrust zone to the west and the major NNE fault zone (fig. 1) respectively.

All the granitic plutons within the fold belt were erroneously considered by Haller (1970, 1971) to be Caledonian. Haller's interpretation was revised later (Higgins, 1976, Henriksen and Higgins, 1976) in the light of age dates based on whole-rock Rb-Sr and zircon U-Pb methods, which showed a number of granitic bodies to be early or middle Proterozoic. Further work is required to establish the full extent of Caledonian plutonic activity. In fig. 1 the granite intrusions are classified according to Hansen *et al.* (1978) into Caledonian and Caledonian or older.

Among the Caledonian granitic plutons, Higgins and Phillips (1979) tentatively distinguished an early granite suite (*c.*475 Ma) (Steiger *et al.*, 1979) and later granites (*c.*450 Ma). Many of the late leucocratic granites (440-5 Ma; Rex *et al.*, 1976) were emplaced along or near the contact between the late Proterozoic sediments and crystalline complexes (fig. 1).

Devonian high-level granites and acid volcanic rocks occur in Hudson Land and neighbouring islands to the south. Absolute ages are not available but the age of the magmatism determined stratigraphically spans the upper Middle Devonian (Givetian) to Upper Devonian (Fammenian) (Alexander-Marrack and Friend, 1976). The Devonian magmatic province is situated close to the major Caledonian NNE fault zone which was active several times in post-Caledonian time (Haller, 1971; Surlyk, 1978).

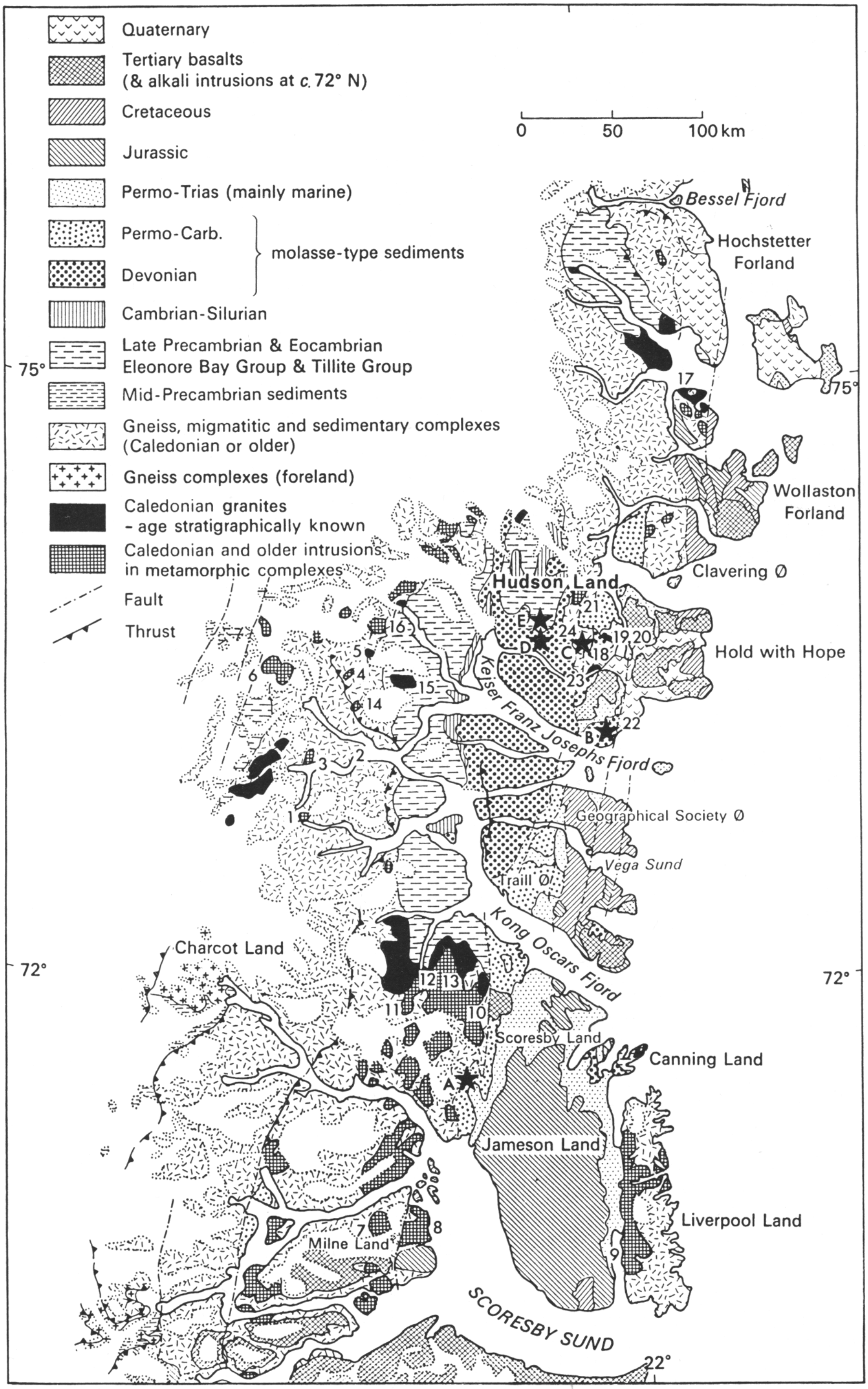

FIG. 1. Geological map of central and northern East Greenland based on Haller (1970), Henriksen and Higgins (1976), and Hansen *et al.* (1978), with location of the analysed granites of Table I. Stars mark locations of U mineralization.

Geochemistry

The granitic plutons within the fold belt are calc-alkaline, and comprise monzonite, grandiorite, and granite. Associated gabbroic or alkaline intrusions are rare. Granites from 24 intrusions situated between 70 and 76° N have been analysed for U by delayed neutron analysis (DNA) and for K and trace elements by energy-dispersive isotope-excited X-ray fluorescence (EDX) (Kunzendorf, 1979).

The concentrations of K, Rb, Sr, Zr, U, and K/Rb and Rb/Sr ratios in granites with $SiO_2 > 68\%$ are listed in Table I (analyses for Rb are too high

TABLE I. *Concentrations of selected elements and elemental ratios in granites ($SiO_2 > 68\%$) from the Caledonian fold belt in East Greenland*

Loc.	Age	n	K (%)	Rb (ppm)	Sr (ppm)	Zr (ppm)	U (ppm)	K/Rb	Rb/Sr
1	1975 Ma	15	3.39	180	260	80	1.6	190	0.7
2	?1800 Ma	4	3.85	160	460	120	1.1	248	0.4
3	1950 Ma	9	3.52	230	120	50	2.3	156	2.1
4	1080 Ma	4	3.46	450	30	30	5.7	77	17.7
5	1000 Ma	5	4.10	220	140	60	2.4	186	1.6
6	650 Ma	9	3.11	190	410	n.d.	0.9	168	0.5
7	Cal.	6	3.75	280	470	330	3.8	137	0.6
8	373 Ma	3	3.64	470	130	140	6.2	79	3.6
9	Cal.	9	2.57	170	560	210	1.0	148	0.3
10	Cal.	4	4.05	480	140	130	3.2	89	3.7
11	Cal.	17	3.98	460	370	190	4.2	91	1.4
12	Cal.	11	4.28	470	880	350	5.8	94	0.6
13	Cal.	5	3.11	350	280	200	4.6	89	1.4
		5	3.56	370	170	240	3.1	95	2.4
14	433 Ma	12	3.90	240	110	n.d.	3.6	164	2.3
15	377 Ma	5	3.86	300	250	230	2.0	129	1.2
16	550 Ma	4	3.98	290	110	70	2.7	141	2.5
17	Cal.	4	4.65	400	125	120	4.2	117	3.2
Hudson Land granites									
18	Cal.	12	4.12	520	60	70	9.7	84	9.6
		7	3.70	330	210	90	5.8	113	1.7
19	Cal.	5	4.82	350	80	30	17.7	139	4.5
20	Cal.	3	4.77	300	340	260	7.1	161	0.9
21	Cal.	4	3.59	440	290	290	4.3	83	1.5
	Cal.	5	3.88	420	150	120	6.7	96	2.9
22	Dev.	3	5.55	410	80	290	14.8	136	5.2
23	Dev.	3	5.49	560	30	180	5.8	100	19
24	Dev.	7	3.90	870	10	80	16.0	46	82
	Dev.	3	5.12	530	40	110	7.7	96	15
Acid volcanics (geometric means of analyses from the entire province)									
	Dev.	122	4.74	460	40	140	6.0	110	12

Loc. = Location of granitic bodies as shown in fig. 1. The radiometric ages given are based on radiometric age dating by D. C. Rex and co-workers (Rex *et al.*, 1976, Rex and Gledhill in press) (Loc. nos. 1-6, 14-16) and by Hansen and Tembush (1979) (Loc. no. 8). Other age estimates are from Henriksen and Higgins (1976). Cal. = Caledonian. Dev. = Devonian. n = number of samples. n.d. = not detected. The samples were analysed at Risø National Laboratory, Roskilde, Denmark by isotope excited energy dispersive X-ray fluorescence (Kunzendorf, 1979) for K, Rb, Sr, Zr, and by delayed neutron analysis for U. The values for Rb are systematically 50% higher than values found by Rex at Leeds University, whereas Sr values agree well between the two laboratories.

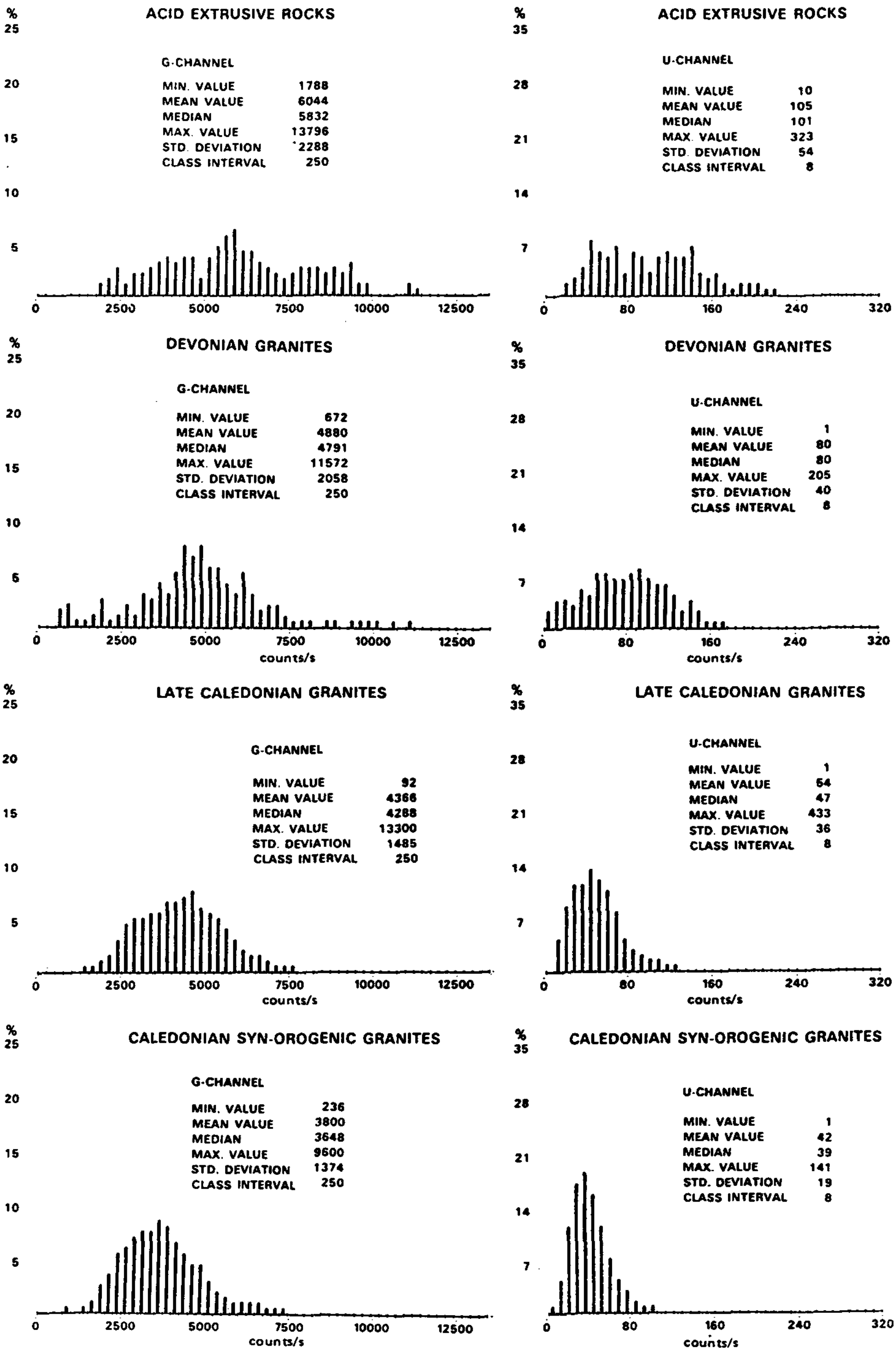

FIG. 2. The radioactivity of granites in the East Greenland fold belt as illustrated by histograms of count-rates obtained in the U-channel of an airborne γ-ray spectrometer. (From Nielsen and Steenfelt, 1977.)

as explained in the text of Table I). The radiometric age, where available, or the stratigraphically determined or assumed age is also indicated.

The variation of U relative to other element contents and ratios is illustrated in figs. 3-7. For comparison the position of three Scottish Caledonian granites, the Foyers, Helmsdale, and Cairngorm intrusions (data from Plant *et al.*, 1980) are shown on some of the diagrams. The Cairngorm (and to a lesser extent the Helmsdale granite) was intruded at the end of the Caledonian event in Scotland, probably in the Lower Devonian.

The Proterozoic granites with one exception (loc. no. 4) have low contents of U (av. 1.7 ppm U) compared with average granite (4.8 ppm U; Taylor, 1964). They have high K/Rb values (fig. 3), medium Sr (fig. 4), and medium to low Zr (fig. 5) as compared with later granites. U is negatively correlated with K/Rb, Sr, and Zr.

Most of the granites of known or assumed Caledonian age have moderate contents of U which have a negative correlation with K/Rb ratios (fig. 3), but there is no correlation between U and Sr (fig. 4) or Zr (fig. 5). The concentration ranges of U, K, Rb, Sr, and Zr found in East Greenland are comparable with those found in British Caledonian granites (Simpson *et al.*, 1979; Plant *et al.*, 1980) and indicated by the means of the Foyers and the Cairngorm granites.

The Devonian granites of Greenland have slightly higher U concentrations than most of the earlier Caledonian and older granites. The highest U contents are found in the granites and acid volcanics of Hudson Land situated in or near the major NNE fault zone.

The pattern of increasing U in the granites with decreasing age was also recognized in an air-borne reconnaissance gamma spectrometry survey (Nielsen and Steenfelt, 1977). The count-rate frequency distribution for the granites in the survey area (72-76°) is shown in fig. 2. The term syn-orogenic refers to the classification by Haller (1971) and includes many granites which were later recognized as Proterozoic. The Devonian granites are characterized by a higher mean count rate and a skewness towards high values when compared with other granites. The air-borne survey also demonstrates the higher radioelement content of the Devonian acid volcanic rocks. Nielsen and Steenfelt (1977) considered the Devonian granites as potential source rocks for U mineralization.

In Hudson Land the Caledonian granites show decreasing Sr and Zr with increasing U (figs. 4 and 5), while the Devonian granites in the same area have low Sr, variable Zr, and no apparent correlation between the two elements and U.

Further data on the Hudson Land granites and rhyolites are shown in figs. 6 and 8, in which all samples are plotted, in contrast to figs. 4 and 5 where the averages of Table I are used. Some of the granites and some of the acid volcanics in fig. 6 plot in the same field as granites outside Hudson Land (fig. 3), whereas a large number of points are displaced towards U enrichment. Some of the U-enriched granite samples, including the mean of the Helmsdale granite, has moderate K/Rb ratios (between 70 and 150) and another group (mainly Devonian granite) has low K/Rb ratios (< 70) and plot close to the mean of the Cairngorm granite, on an extension of the trend of the granites in fig. 3.

U is plotted against Th in fig. 7. Both elements were determined by γ-ray spectrometry. The Th/U ratios show a considerable scatter although many samples plot within a range of Th/U of 2-6, which is commonly recorded in granites (e.g. Bailey, 1979; Moreau, 1976; Rogers *et al.*, 1978; Stuckless *et al.*, 1977; Taylor, 1964). Several samples show U enrichment relative to Th (Th/U < 2). The samples marked a, b, c, d also show high U contents relative to the K/Rb ratio (fig. 6).

The Devonian granites are characterized by a high Th content, compared with the Caledonian granites (in Hudson Land) and with the Helmsdale and Cairngorm granites. The Th content is relatively constant whereas U varies. A Th/U ratio greater than 6 may indicate loss of U.

Recent loss, or gain, of U in rocks can be indicated by plotting γ-spectrometrically determined U (eq. U) against chemically determined U (U_{DNA}) (fig. 8). Ratios of eq. U/U_{DNA} greater than 1 indicate recent U loss (while daughter products from the radioactive decay of U have remained).

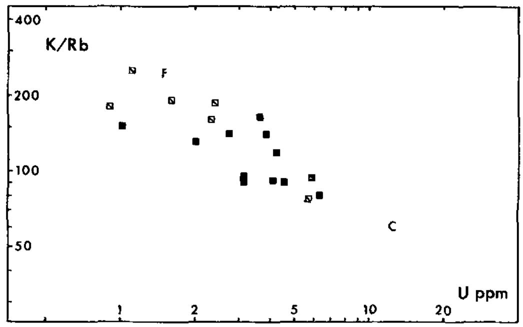

FIG. 3. K/Rb ratios against U concentrations in granites from the East Greenland fold belt outside Hudson Land. ▨ Proterozoic granites. ■ granites of Caledonian or older age. F = Mean of Foyers granite, Scotland; C = Mean of Cairngorm granite, Scotland (Plant *et al.*, 1980). Rb values for the Scottish granites are multiplied by 1.5 for comparison with Rb values used here.

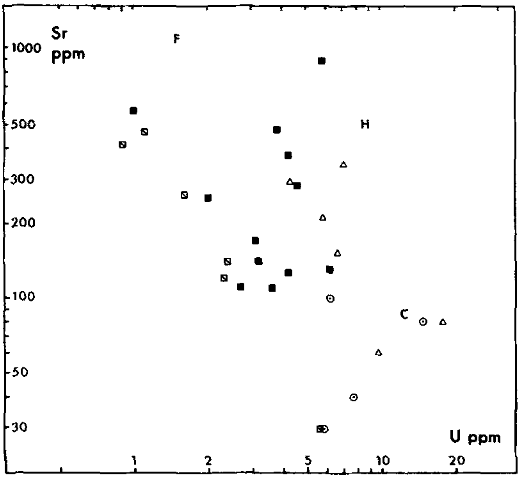

FIG. 4. Sr concentration against U concentration in granites from the East Greenland fold belt. All granites of Table I. △ Caledonian granites in Hudson Land. ⊙ Devonian granites. Other symbols as in fig. 3.

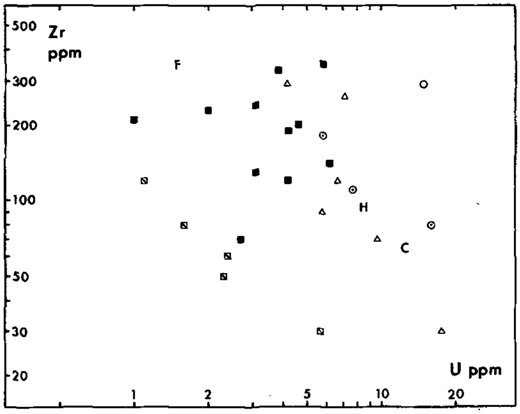

FIG. 5. Zr concentration against U concentration in granites from the East Greenland fold belt. All granites of Table I. Symbols as in fig. 4.

In fig. 8 the Devonian granites plot close to the line of radioactive equilibrium (eq. $U/U_{DNA} = 1$) indicating that they have suffered no recent U loss. In contrast most of the Caledonian granites appear to have lost 10–30% (maximum 50%) of their U, probably during weathering. Samples with significant recent U loss (solid symbols) have low Th/U ratios (solid symbols in fig. 7) indicating that the excess U (relative to Th) in the granite occurs in compounds that are readily leached.

Discussion

Uranium enrichment in the granites. U and Rb behave incompatibly in granitic melts, so that where U concentration is governed by magmatic processes Rb and U would be expected to show a positive correlation, as is the case (fig. 3) for most of the Caledonian granites.

In fig. 6 which presents data for granites and acid volcanics in Hudson Land, two kinds of U enrichment are observed. In some of the Devonian granites and acid volcanics, as well as in the Cairngorm granite of Scotland high U is correlated with low K/Rb and the U enrichment is therefore thought to be magmatic. In other granite samples there is an excess of U relative to K/Rb (fig. 6) and also relative to Th (fig. 7), suggesting that U has been added to the granite post-magmatically.

The regional geology of Hudson Land has yet to be fully described. Some geological and petrographical aspects of the Devonian magmatic province are treated by Noe-Nygaard (1937), Graeter (1957), Bütler (1954), and Alexander-Marrack and Friend (1976) but little information is available on the geochemistry and origin of the magmas. Analyses of granites and rhyolites in connection with this work (Steenfelt, unpublished data) indicate the presence of calc-alkaline to mildly alkaline magmas. Many of the samples have high levels of K, Rb, Y, Nb, U, Th, and low contents of Ca, Mg, Ti, Fe, Sr, and they have low K/Rb ratios. A few determinations of Sn and Ba on one of the granite intrusions showed high Sn and low Ba contents. Analyses for F have not been made, but fluorite is abundant in the granites and rhyolites.

The Devonian magmatic rocks exhibit similar chemical characteristics to the Cairngorm granite in Scotland (Plant *et al.*, 1980) and the U-enriched granites of Sweden (Wilson and Åkerblom 1980). They also show a close chemical resemblance to granites and rhyolites of a late Palaeozoic acid magmatic province in Queensland, Australia (Bain, 1977), which has U mineralization associated with it.

In the East Greenland Devonian granites, as in the British uraniferous granites (Simpson *et al.*, 1979), the U-bearing phases, as seen in fission-track prints, are predominantly accessory minerals. Minor redistribution of U due to alteration of biotite and opaque minerals is observed in the East Greenland granites.

Three samples of Devonian granites (marked by heavy circles on figs. 6, 7, and 8) plot close together and have a normal Th/U ratio of 3.4; they show no signs of recent U loss and plot on the trend of magmatic U enrichment. They probably represent unaltered granite. Other Devonian granites have similar Th (fig. 7) but less U, and are thought to

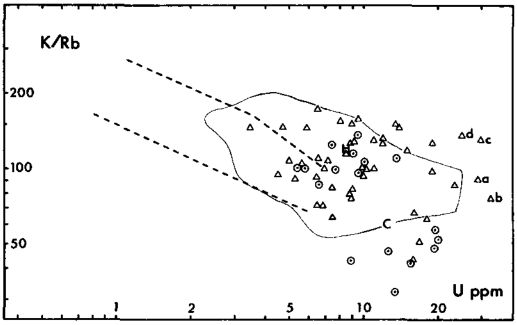

FIG. 6. K/Rb ratios against U concentrations in granites from Hudson Land, northern East Greenland. All analysed samples. △ Caledonian granites. ⊙ Devonian granites. The solid line surrounds the main field of Devonian rhyolites (Steenfelt unpublished data). H = Mean of the Helmsdale granite, Scotland (from Plant *et al.* 1980). The dashed lines outline the field of granites shown in fig. 3. Gamma spectrometer analyses for U concentrations are used where available instead of DNA analyses, because of the recent leaching of U described in the text.

have lost some of their U. This loss is not due to recent weathering (eq. U ~ U_{DNA} in fig. 8) and may have occurred at a late or early post magmatic stage.

The fault zone in Hudson Land intersects Caledonian granites as well as some of the Devonian granites and rhyolites. The rocks affected by the faulting are jointed, fractured, and often hydrothermally altered. Fluoritization and hematization are typical alteration phenomena and kaolinization is observed in some rhyolites.

Within the granite U is associated with chloritized biotite and other altered minerals, and typically with hydrated iron oxides in microfractures.

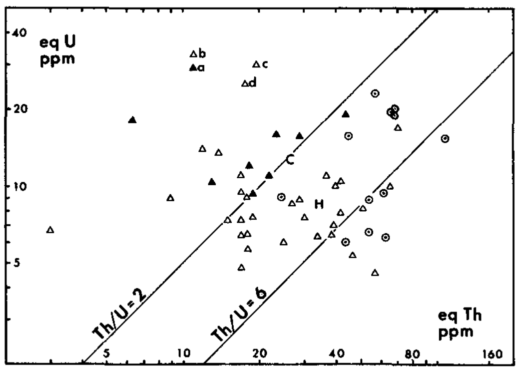

FIG. 7. Concentration of eq. U against eq. Th determined by γ-spectrometry for granites in Hudson Land, northern East Greenland. Symbols as in fig. 6. Solid and heavy symbols are explained in text.

The mineralogical distribution of the U supports chemical evidence that U was remobilized and introduced post-magmatically. Comparisons can be made with the location of U in the fracture zones of the Granite Mountains granite, Wyoming (Stuckless *et al.*, 1977), in the Helmsdale granite, Scotland (Bowie *et al.*, 1973), and in the acid granites of Shetland (Simpson *et al.*, 1976). The Helmsdale and Shetland granites are situated near the faults of the Great Glen system and Simpson *et al.* (1976) suggested hot-spring activity associated with the faulting to explain their U enrichment.

U mineralization in the fault zone. There are several examples of U mineralization along the main post-Caledonian fault zone. The mineralization is of small extent and is everywhere related to joints or fractures in the host granites and rhyolites. Table II summarizes the mineralized localities indicated on fig. 1.

The southernmost occurrence of mineralization has been known for a long time, while others were discovered during a combined geophysical and geochemical U exploration survey (Nielsen and Steenfelt, 1977; Steenfelt and Kunzendorf, 1979).

The mineralization is epigenetic and the mineralizing fluids have percolated through the joints and fractures associated with the faults. In the Arkosedal breccia U was precipitated as U oxide with fluorite (Wollenberg 1971). In the jointed granites (Arve, Ritom Sø), iron oxides caused precipitation

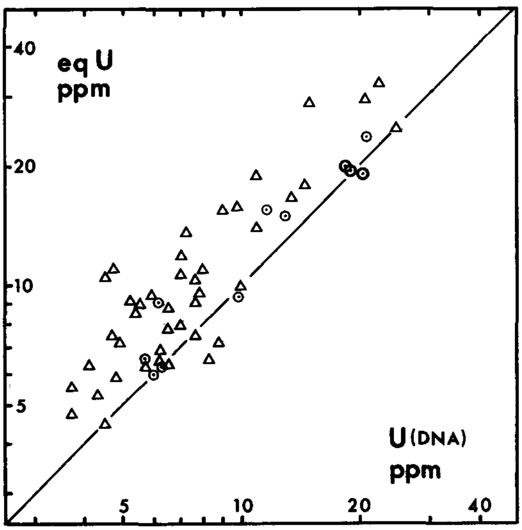

FIG. 8. Concentration of eq. U determined by gamma spectrometry against U determined by delayed neutron analysis in granites from Hudson Land, northern East Greenland. Symbols as in fig. 6. Solid and heavy symbols are explained in text.

TABLE II. *U mineralization related to the major NNE fault zone in central and northern East Greenland*

	Location	Host rock	Uranium-bearing phases	Max. U conc. of mineralized samples (ppm)	Accompanying elements	References
A	Arkosedal	Breccia in Caledonian granite	Fluorite, pigmentary material	3000	F,Ba	Nielsen and Løvborg (1976) Wollenberg (1971)
B	Kap Franklin	Devonian rhyolite	Carburan, wölsendorfite, limonite	5000	Pb,Zn	Secher *et al.* (1976) Beddoe-Stephens and Secher (1982) Kunzendorf *et al.* (1978)
C	Arve	Caledonian fractured granite	U-oxide in chlorite and limonite hematite	400	Cu,Zn	Steenfelt and Nielsen (1978) Steenfelt and Kunzendorf (1979)
D	Moskusokseland	Upper Devonian rhyolite	Pitchblende, carburan, β-uranophane	10000		Steenfelt (1976), Steenfelt and Nielsen (1978)
E	Ritom Sø	Caledonian fractured granite	Limonite, sericite	480	Cu,Pb,F	Unpublished data

of U, and in the rhyolites (Kap Franklin, Moskusokseland) U was fixed by hydrocarbons in the form of carburan (thucholite), except in one locality where pitchblende occurs in microfractures.

The distribution of U-mineralized granites in Hudson Land is indicated by the occurrence of U anomalies in a stream water geochemical survey (fig. 9*a*). This confirms that present-day leaching of the fractured granites is taking place as indicated by the disequilibrium shown in fig. 8. The stream sediments contain the same amount of U as the granites, and no significant anomalies occur (fig. 9*b*).

Analyses indicate that U is often associated with Cu, Zn, and Pb in the mineralized fractures, and the fault zone also contains quartz-fluorite veins with pyrite and sulphides of Pb, Zn, and Cu (Graeter 1957). Geochemical exploration confirmed that these elements are more abundant in the faulted area.

Watson and Plant (1979) suggested that the U and base metal mineralization associated with the Helmsdale granite in the Great Glen fault zone (Gallagher *et al.*, 1971) resulted from the interaction of circulating meteoric water in deep fault zones with magma or granite containing U and fluorite.

In East Greenland the mineralization can be explained using a similar model associated with repeated volcanic intrusions in Middle and Upper Devonian times, the faults providing channels for circulating fluids. The high Th/U ratio of some of the Devonian granite provides additional evidence of removal of U from the magma.

A similar genetic model was used by Bain (1977) to explain U mineralization associated with late

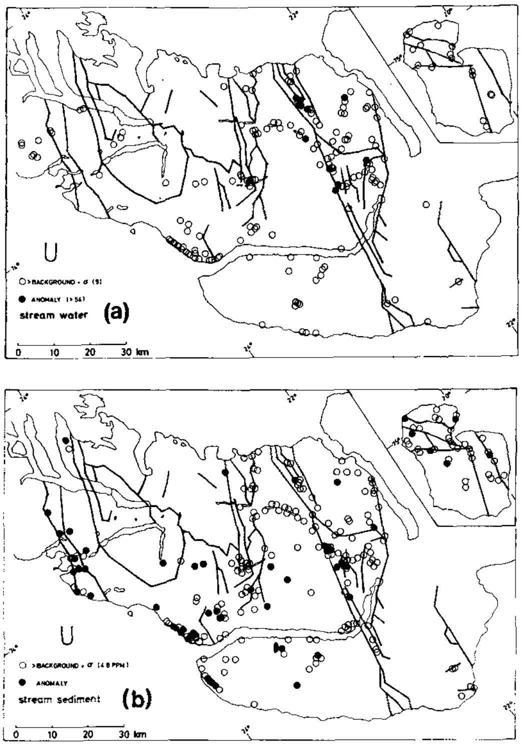

FIG. 9. High values and anomalies in the U content of drainage samples in Hudson Land and vicinity, northern East Greenland. (*a*) Stream water. The values shown express: U concentration in ppb × 10^3/conductivity in μ mhos/cm. (*b*) The −0.15 mm fraction of stream sediments. Sampling density: 1 sample/2–5 km^2. From Steenfelt and Kunzendorf (1979). The black dots indicate U enrichment in the fault zones, which are marked by heavy lines.

Palaeozoic acid magmatism in north-east Queensland, Australia. The source rocks are believed to be granites or acid volcanics with high background U content. Hydrothermal fluids rich in fluorine are suggested to have been the main transporting agent with deposition of U in breccias, joint systems and other permeable zones. The intermittent activity of fault systems over a considerable period of time is emphasized.

Summary and conclusions

The pre-Caledonian and Caledonian calc-alkaline granites have generally low to moderate concentrations of U. The post-orogenic Devonian acid magmatic rocks are characterized by a higher content of incompatible lithophile elements including U. The magma was emplaced near to or at the surface during the Middle to Upper Devonian. The Devonian magmatic province is situated in areas which were subjected to extensive post-Devonian block-faulting characterized by faulting and uplifting in the Devonian (Alexander-Marrack and Friend, 1976).

The Devonian granites and rhyolites are chemically similar to the uraniferous granites of the British Isles, and their U enrichment is considered to be magmatic. The origin of the granite magmas is not known.

U enrichment, occasionally leading to U mineralization, is encountered in the granites situated in the major NNE fault zone. U was introduced into faulted granite during a hydrothermal mineralizing event. The type of mineralization and the structural setting of the mineralized granites are equivalent to those of the mineralized granites in the Great Glen fault system of Scotland.

It is suggested that the Devonian acid magmas supplied the heat, U and F for hydrothermal fluids percolating in the joints and fractures along the faults consistent with the models of Bain (1977), Simpson *et al.* (1979), and Watson and Plant (1979). The uranium was possibly transported as fluorine complexes and subsequently precipitated in or along fractures. The uranium occurs as pigmentary oxide associated mainly with fluorite, hematite, or limonite, and additionally in carburan and uranophan. No significant U mineralization has been found in Devonian sediments surrounding the mineralized granites and acid volcanics.

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