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## Uranium mineralization and granite magmatism in the British Isles

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[Plate 1]

Uranium mineralization associated with granites in the Caledonian and Hercynian provinces of the British Isles is shown to be genetically related to the uranium content and distribution, age, and structural setting of these granites.

The uranium content of whole rock samples, analysed by the delayed neutron method, is used to demonstrate that mineralization is associated with intrusive complexes with a high mean content of uranium which also exhibit a high concentration of incompatible elements, low K/Rb ratio, low total Sr, low initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio and high geothermal gradient. The standard deviation for uranium is greater where such intrusives are mineralized but the mean value is relatively unaltered. Therefore mineralization is the result of uranium redistribution, and does not involve further introduction of uranium.

Fission track studies indicate that the high 'background' uranium content of granites, away from mineralization, is due to the occurrence of uranium in resistate primary phases such as zircon. Uranium is released by the dissolution of these resistate minerals. The processes of greisenization and tourmalinization which have been shown by oxygen isotope studies to involve massive interaction with meteoric water all extract uranium, which is redeposited down the *PT* gradient as 'primary' uraniferous ore minerals in vein-type mineralization.

It is suggested, therefore, that mineralization involves leaching of granite magma enriched in metals and fluorine by water of meteoric origin containing dissolved carbonate. The breakdown of primary zircon is attributed to a phase of short duration of high temperature interaction of granite magma with meteoric water, and uranium mineralization is thought to have occurred at this time. However, the high concentration of uranium, thorium and potassium of the 'background' granite which produce hot rock districts may cause redistribution of uranium by hydrothermal mineralization during periods of high average heat flow from the mantle (as in the Tertiary of southwest England) or during dyke emplacement. An extensive system of channels for heating and circulating water is necessary for this system to function, and faults in granite would be particularly favourable.

The regional trend of uranium and incompatible elements shown by late Caledonian (Devonian) and Hercynian granites in Britain is related to dehydration reactions during subduction of oceanic crust. The importance of phlogopite breakdown in accounting for the characteristics of uraniferous granites is discussed in relation to magma generation, with the use of closed and open system models with partial fusion of ocean crust or upper mantle. Uranium enrichment by scavenging of subcontinental lithosphere is considered important, but late stage assimilation of uranium from higher levels in the crust is relatively insignificant.

The applications of the models for uranium mineralization to exploration at a regional and local scale are discussed.

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## 1. INTRODUCTION

In recent years, models for ore genesis have been developed on the basis of plate tectonic theories for several types of mineralization associated with acid igneous rocks, which include porphyry copper-molybdenum deposits and tin-tungsten-fluorite mineralization (Mitchell & Garson 1972; Sillitoe 1972, 1974). This type of study has not been extended to uranium mineralization although the association with granites is very strong. A large number of detailed studies on uranium mineralization, particularly in the Hercynian province of Central Europe and France, have shown that uranium enrichment is associated with 'two mica granites'. Characteristically such intrusives contain unusually high concentrations of uranium thought to be predominantly in uraninite, tend toward soda enrichment and are accompanied by aureoles of tin-tungsten veins. Uranium mineralization is generally 50 Ma younger (250 Ma) than related granite intrusives (300 Ma) and occurs in veins or fractures within a few hundred metres of the weathering surface (Gangloff 1970; Geffroy 1971; Sarcia *et al.* 1958).

The genesis of 'two mica' granites is attributed to palingenesis/anatexis in areas of thickened sial in geosynclines by continental authors such as Tischendorf (1973) and Moreau (1976) who distinguish them from calc-alkaline granites which are attributed to melting beneath destructive continental plate margins (Moreau 1976). A model for uranium mineralization in the two mica granites known as '*per descensum*', whereby primary uraninite is oxidized on weathering to give highly soluble uranyl complexes which are carried down by percolating groundwaters to form pitchblende in veins, is favoured by some authors (Moreau 1976; Barbier 1974), although others advocate a hydrothermal magmatic model (Gangloff 1970; Geffroy 1971). Moreau (1976) has suggested that mineralizing fluids originate along with the two mica granite magma from crustal anatexis. Several difficulties are encountered in applying these concepts to uraniferous granites in a more general way. For example, the Hercynian granites occur in areas of high temperature – low pressure metamorphism (Lorenz & Nicholls 1976; Lorenz 1976; Zwart 1969) indicating an unusually high geothermal gradient in a thin crust, and outside the Hercynian province there is little association between areas of thickened sialic crust in 'geosynclines' and tin, tungsten, fluorine and uranium mineralization.

In the British Isles, late Precambrian, Caledonian, Hercynian and Tertiary granites occur and it is therefore possible to study the association between uranium and granite in several tectonic settings. The data of Simpson, Plant & Cope (1976) and those of Brown & Hennessy (1978) reveal variations in uranium concentration related to differences in tectonics, structural setting and age distribution within each province. In the present paper the uranium contents of granites from five tectonic settings are discussed and the origin of the granites is considered in the light of isotopic geochemical and geophysical evidence. The detailed distribution of uranium within selected granites has also been investigated in an attempt to evaluate the rôle of magmatic and postmagmatic processes in determining its distribution. The relative ages and structural relations of the granites studied are as follows:

(1) Late Precambrian or Caledonian precursor intrusive suites of Southern Britain whose origin is thought to be related to subduction of oceanic lithosphere at an Andean-type destructive plate margin.

(2) Caledonian granites generated during an early Palaeozoic orogenic episode in Northern Scotland by partial melting of existing sialic material.

(3) Caledonian granites of Lower Devonian age with a dominantly sub-crustal origin as in (1).

(4) Hercynian granites emplaced in conditions of high geothermal gradients in a continent (microplate) – continent collision setting.

(5) Tertiary granites of the Western British igneous province thought to be related to a constructive plate margin initiated beneath continental crust.

## 2. ANALYTICAL METHODS FOR URANIUM

### (a) *Delayed neutron and fission track methods*

Whole rock analyses by the delayed neutron method (Amiel 1962) provide accurate and precise values for uranium concentration and the Lexan plastic fission track (L.p.f.t.) method (Price & Walker 1963; Kleeman & Lovering 1967; Bowie, Simpson & Rice 1973) enables its distribution to be established. Reliable textural and mineralogical information concerning the distribution of uranium is valuable since reduced ( $U^{4+}$ ) species are relatively insoluble whereas oxidized ( $U^{6+}$ ) species are soluble, mobile, and readily reprecipitated in secondary mineral phases.

In general, the textural relations indicate the relative order of events and the mineralogical phases associated with uranium indicate the geological conditions under which introduction or removal of uranium occurred. The fission track method is specific for uranium and can be quantified by the selection of suitable standards and thermal neutron doses. For petrographic study, the sectioned material is irradiated with a relatively large integrated dose of about  $5 \times 10^{16}$  thermal neutrons  $cm^{-2}$  in order to distinguish uranium-bearing from uranium-free mineral phases. Under these conditions the uranium-bearing minerals produce saturated track distributions in the detector and uranium present in associated minerals is detectable above 0.05 parts/ $10^6$ .

In the present work it was observed that rocks with uranium secondary minerals such as zippelite, have more than 200 parts uranium/ $10^6$  and where primary uranium minerals occur the whole rock uranium content is greater than 2000 parts/ $10^6$ . Uraninite has been recorded elsewhere as a discrete phase in granite (Coppens & Mayanda 1969; Barbier 1972; Snetsinger & Polkowski 1977) from samples which were probably mineralized. The most uraniferous non-mineralized granite recorded in this study contains 67 parts U/ $10^6$  and most of the uranium is held cryptically in uraniferous zircon.

### (b) *Gamma ray spectrometry*

Gamma ray spectrometry was used to determine the distribution of uranium in the Loch Doon granite. The method is based on the measurement of the  $^{214}Bi$  gamma peak which is separated from its  $^{238}U$  parent isotope by several intermediate daughters, some of which are soluble in surface weathering. In order to avoid problems resulting from secular disequilibrium, the gamma spectrometric results are calibrated by using the delayed neutron method.

## 3. GRANITES IN THE BRITISH ISLES, URANIUM CONTENTS AND MINERALIZATION

### (a) *Caledonian granites*

These intrusions were divided by Read (1961) into two groups: the older granites and the newer, post tectonic granites consisting of a suite of forceful intrusions, and a later suite of

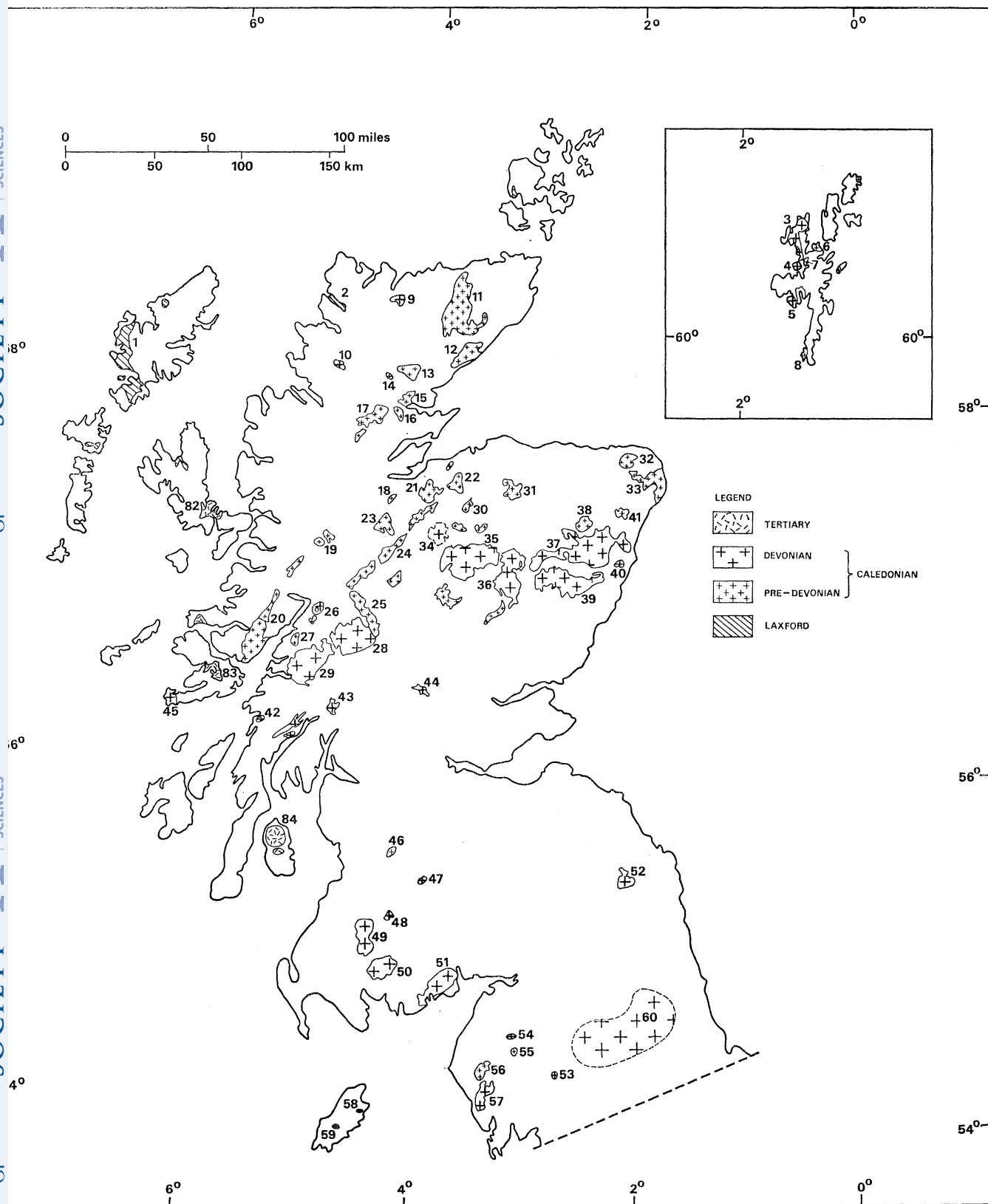


FIGURE 1 *a*. Granites in Scotland and northern England; for key see p. 390.

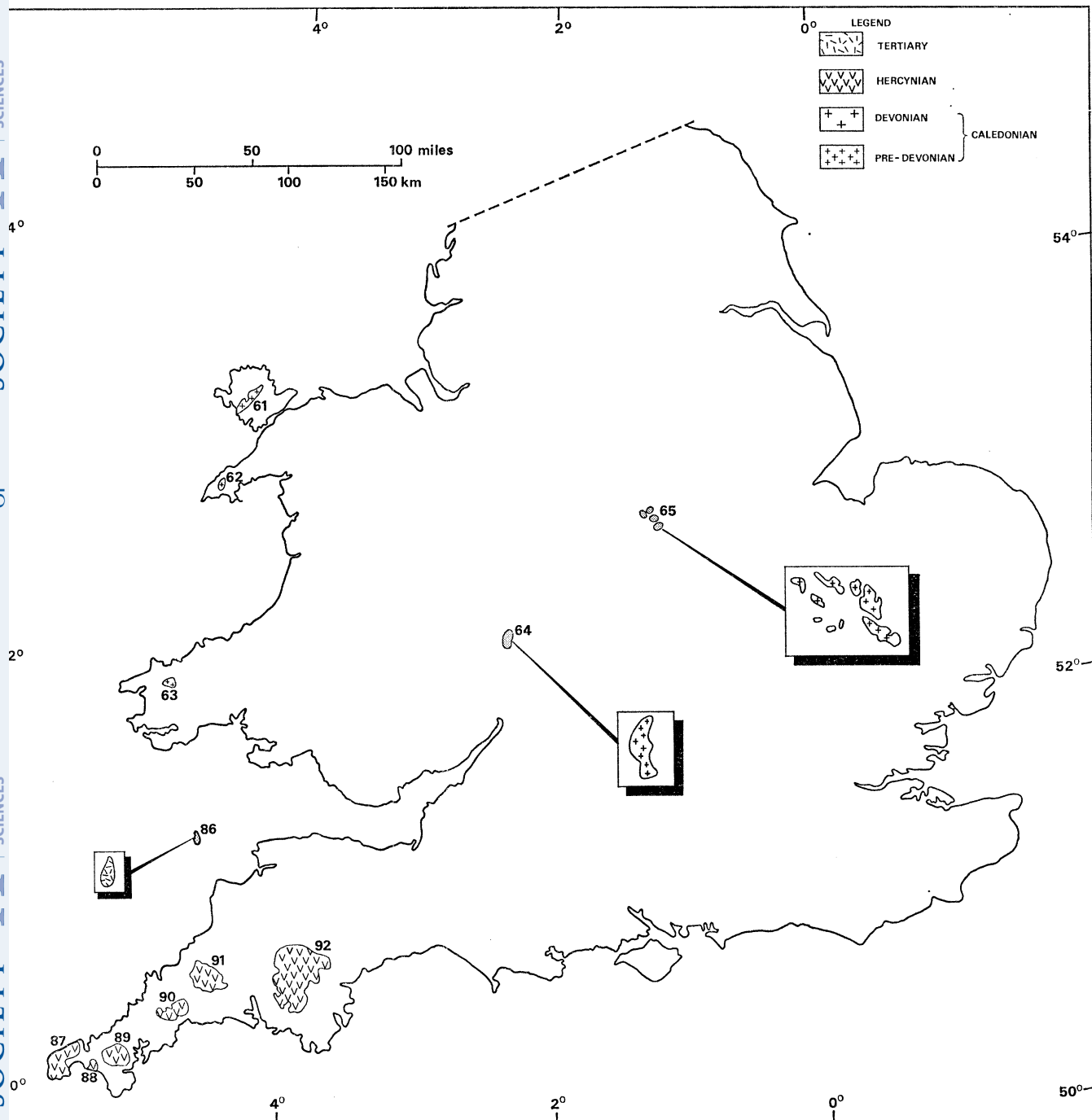
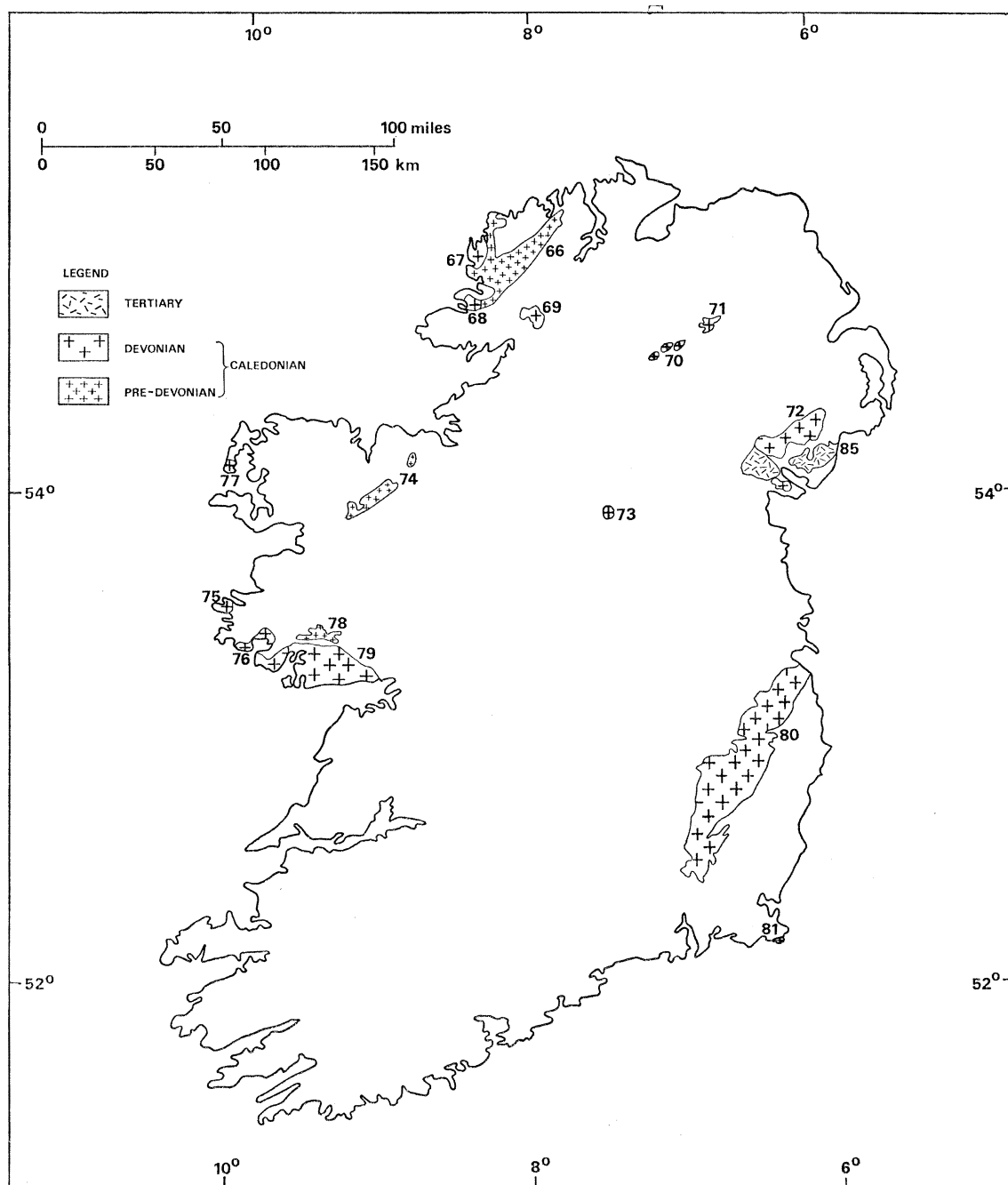


FIGURE 1 *b*. Granites in part of England and Wales; for key see p. 390.



FIGURE 1 *c*. Granites in Ireland; for description see below.

Key to figure 1 (a)–(c): 1, Lewis; 2, Laxford; 3, North Maben; 4, Muckle Roe; 5, Sandsting; 6, Graven; 7, Brae; 8, Spiggie; 9, Ben Loyal; 10, Borrolan; 11, Halladale; 12, Helmsdale; 13, Lairg Rogart; 14, Grudie; 15, Migdale; 16, Fearn; 17, Carn Chuinneag; 18, Abriachan; 19, Cluanie; 20, Strontian; 21, Moy; 22, Ardlach; 23, Foyers; 24, Strathspey; 25, Strath Ossian; 26, Ben Nevis (complex); 27, Ballachulish; 28, Moor of Rannoch; 29, Etive; 30, Grantown; 31, Ben Rinnes; 32, Strichen; 33, Peterhead; 34, Monadhliath; 35, Cairngorm; 36, Lochnagar; 37, Hill of Fare; 38, Bennachie; 39, Mount Battock; 40, Auchlee; 41, Ardlethen; 42, Kilmelford; 43, Garabal Hill; 44, Comrie; 45, Ross of Mull; 46, Distinkhorn; 47, Spango Water; 48, Cairnsmore of Carsphairn; 49, Loch Doon; 50, Cairnsmore of Fleet; 51, Criffel–Dalbeattie; 52, Cheviot; 53, Shap; 54, Skiddaw; 55, Threlkeld; 56, Ennerdale; 57, Eskdale; 58, Dhoon; 59, Foxdale; 60, Weardale; 61, Coedana; 62, Sarn; 63, South Wales Dimetian; 64, Malvernian; 65, Mt Sorrel and Charnian Diorites; 66, Donegal; 67, The Rosses; 68, Ardara; 69, Blue Stack Mountains; 70, Omagh; 71, Slieve Gullion; 72, Newry; 73, Bellanagh; 74, Ox Mountains; 75, Omev; 76, Roundstone; 77, Inish; 78, Oughterard; 79, Galway; 80, Leinster; 81, Carnsore; 82, Skye; 83, Mull; 84, Arran; 85, Mourne Mountains; 86, Lundy; 87, Land's End; 88, Tregonning–Godolphin; 89, Carnmenellis; 90, St Austell; 91, Bodmin; 92, Dartmoor.

permitted granites. Watson & Plant (1979, this volume) have suggested that in northern Scotland the principal geochemical and geophysical distinction is between older granites and newer forceful granites on the one hand, and the newer permitted granites on the other (figure 1). In this study granites have been allocated to the Pre-Devonian and to the Devonian on the basis of reliable Rb-Sr dates wherever possible (see also Brown & Hennessy 1978), the division being placed at about 410 Ma. Where Rb/Sr data are lacking, intrusions are classified on the basis of structural, geophysical and petrographic criteria. An important feature of the British Caledonides is the contrast in metamorphic style between northern Scotland and regions to the south. In the northern province, magmas were emplaced in Moinian or Dalradian metasediments of greenschist or higher metamorphic grade over a presumed northerly dipping subduction zone whereas, in the southern province, magmas were emplaced into slightly

TABLE 1. URANIUM CONTENTS (parts/10<sup>6</sup>) OF GRANITES IN THE BRITISH ISLES

age	intrusion	no. of samples	method of analysis	references	mean	s.e.	range	s.d.	log <sub>10</sub> (mean)	log <sub>10</sub> (s.d.)
Caledonian pre-Devonian	Foyers	65	d.n.m.	1	1.49	0.07	3.50-0.50	0.54	1.41	1.41
	Skene Complex†	32	d.n.m.	2, 3	2.89	0.22	6.60-1.50	1.22	2.69	1.48
	Threlkeld	4	d.n.m.	3	3.16	0.62	4.63-1.89	1.08	2.95	1.48
	Peterhead	10	d.n.m.	3, 4	3.97	0.48	6.40-1.40	1.73	3.63	1.62
	Ballachulish	10	d.n.m.	3	4.76	0.63	9.12-2.82	1.87	4.46	1.45
	Helmsdale	29	d.n.m.	2	7.54	0.67	18.1-3.60	3.61	6.92	1.55
	Strontian	17	d.n.m.	3	8.72	1.00	15.5-2.4	4.14	7.59	1.74
Devonian	Leinster	42	d.n.m.	3	3.70	0.29	9.19-1.24	1.83	3.31	1.59
	Ardara	9	d.n.m.	3	4.56	0.60	8.0-2.0	1.70	4.27	1.51
	Fleet	29	d.n.m.	3	4.87	0.37	9.74-2.14	1.97	4.57	1.48
	Loch Doon	36	d.n.m.	3	5.04	0.37	12.0-0.20	2.22	4.36	1.95
	Balmoral	11	d.n.m.	3	5.23	1.01	12.1-2.42	3.20	4.79	1.70
	Rosses	4	d.n.m.	3	5.75	1.42	12.0-1.0	2.43	3.98	2.95
	Skiddaw	6	d.n.m.	3	8.70	2.19	15.54-1.15	4.90	6.61	2.57
	Shap	17	d.n.m.	3	9.68	0.74	16.94-5.90	2.96	9.33	1.38
	Cairngorm	24	d.n.m.	5	12.66	1.22	32.9-6.3	5.98	11.75	1.48
	Weardale	6	d.n.m.	3	12.89	1.65	20.05-8.24	3.69	12.30	1.35
	Etive	3	d.n.m.	3	17.29	5.55	24.0-6.28	7.85	14.80	2.09
Hercynian	Dartmoor	56	d.n.m.	2	11.33	0.81	35.5-3.2	6.08	10.00	1.62
	Carn Brea	34	d.n.m.	5	13.86	0.89	32.2-6.5	5.18	13.18	1.41
	Land's End	52	d.n.m.	5	16.62	1.04	38.2-2.6	7.49	14.45	1.78
	St Austell	9	d.n.m.	5	24.16	5.89	66.6-9.50	17.67	20.40	1.82
	St Austell (all samples)	30	d.n.m.	5	16.93	2.66	66.6-0.60	14.95	10.96	2.95
	Cligga	14	d.n.m.	5	24.98	2.12	35.8-8.0	7.92	23.4	1.48
Tertiary	Skye (1)	9	γ-ray	6	1.1	0.08	1.3-0.5	0.23	1.26	1.35
	Skye (2)	13	d.n.m.	7	1.43	0.11	2.14-1.08	0.39		
	Lundy	2	γ-ray	6	8.2	0.2	8.4-8.0	0.2	—	—
	Mourne Mts	10	γ-ray	6	11.2	1.55	21.4-5.2	4.65	10.23	1.55

† All of the specimens, except one, are from the pre-Lower Devonian intrusive phase. The Hill of Fare granite mapped by Bisset (1933, p. 83) consists of an 'earlier Caledonian' Skene Complex of dominantly grey granites and a 'later Caledonian' permitted red granite which forms the Hill of Fare. One sample of this granite was obtained from Craigton Quarry and contains the highest value (6.6 parts U/10<sup>6</sup>) obtained from all the Hill of Fare granites studied by Bowie *et al.* (1973).

References: 1, Simpson, Plant & Williams (1977); 2, Simpson, Plant & Cope (1976); 3, Brown, Cassidy & Hennessy (1977); 4, Bowie, Simpson & Rice (1973); 5, Simpson, Plant & Green (1977); 6, Tammemagi (1976); 7, Moorbath & Welke (1969).



metamorphosed Lower Palaeozoic sequences of the southern Uplands, Lake District, Wales and southern Ireland. A southerly dipping subduction zone under the southern Caledonides is inferred (see, for example, Phillips *et al.* 1976) and a major structural break between the two provinces has been identified seismically at Moho depths (Bamford & Prodehl 1977). This has been correlated with a continent suturing event as the Proto-Atlantic or Iapetus ocean closed (Dewey 1974; Phillips *et al.* 1976). Palaeomagnetic data suggest little post-Ordovician closure (Morris 1976) so that this suturing event apparently preceded newer granite formation.

(i) *Pre-Devonian granites*

Early granites range from *ca.* 750–560 Ma but there are few available data concerning their uranium content. The existing data, however, suggest that uranium (table 1) and thorium are not enriched.

In the northern province background uranium values occur in association with early pegmatites and gneisses of late Precambrian age (*ca.* 750 Ma), older granites such as Carn Chuinneag (*ca.* 560 Ma), vein complexes such as Strath Halladale and newer forceful granites such as Foyers (Watson & Plant 1979, this volume). None of these intrusions are associated with aeromagnetic or gravity anomalies (M. Lee, personal communication to J.P.). This major Pre-Devonian suite contains intrusions ranging from adamellite to gabbro, the older granites have diffuse contacts and are largely gneissose to migmatitic while the newer forceful granites show evidence of intrusion, often with marginal hybridization. Hence, the Pre-Devonian granites of the northern province show characteristics of derivation by crustal ultrametamorphism and partial melting and merge with the background geophysically and geochemically. It is probable that they were emplaced during and following the climax of metamorphism about 500 Ma ago (Brown & Hughes 1973). The newer forceful granites have probably risen further from the site of partial melting.

(ii) *Lower Devonian granites*

The Lower Devonian (permitted) intrusions are post-tectonic, generally undeformed and unmetamorphosed, and comprise ring complexes beneath andesitic volcanoes (e.g. Cheviot, Ben Nevis) and other high level Caledonian intrusions. Their discordant margins and metamorphic aureoles contrast with the Pre-Devonian granites of northern Scotland and all of the Lower Devonian granites have intense negative Bouguer anomalies (e.g. Weardale (Bott 1966) and Cairngorm (R. Hipkin, personal communication to G.C.B.)) which are coupled, in the northern province, with strong aeromagnetic anomalies. The Lower Devonian intrusions are therefore of batholithic proportions. According to gravity data these plutons, which correspond to average granodiorite composition, contrast with higher density basement at depth. In northern Scotland this basement is undoubtedly Lewisian, mainly refractory granulites, and a similar basement may extend to the suture (Bamford & Prodehl 1977); further south the basement is deeply buried and of unknown type. A crustal magma source cannot be definitely excluded in the southern province although initial strontium isotope ratios for lower Devonian granites (table 2) suggest that their source is deeper. The granites were emplaced after closure of the Iapetus suture and are not therefore of normal Andean type. Lower Devonian granites occurring on both sides of the Caledonian suture zone tend to show high uranium concentrations. In the Lake District, significant uranium anomalies occur (e.g. Shap, Skiddaw) and the potassic intrusives contrast with the uranium poor sodic Caradocian granites mentioned earlier.

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In southern Ireland little uranium enrichment has been noted in the poorly exposed Leinster granite (Bruck & O'Connor 1977; Brown, Cassidy & Hennessy 1977).

In the northern province the Cairngorm-Aberdeen line of granites are anomalously enriched in uranium and other incompatible elements (table 1: figure 2). Indeed, these granites are among the most alkalic Caledonian intrusives and contain rare minerals such as beryl and topaz in addition to high levels of incompatible elements (Watson & Plant 1979, this volume;

TABLE 2. INITIAL STRONTIUM ISOTOPE RATIOS ( $^{87}\text{Sr}/^{86}\text{Sr}$ ) FOR GRANITES IN THE BRITISH ISLES

(See Brown & Hennessy (1978) for Caledonian references supplemented from Bell (1968), Moorbath & Shackleton (1966), Cribb (1975) and Gale & Wadge (personal communication). St Austell data from Harding & Hawkes (1971) and Tertiary data from Dodson & Long (1962), Moorbath & Bell (1965) and summary by O'Connor (1976).)

*Caledonian*: units as defined in text

Northern Province		Southern Province	
		pre-Lower Devonian	
Donegal	0.708	Oughterard	0.706
Glencroft-Ardlethan	0.714	Ox Mountains	0.706-0.707
Strichen	0.717	Carnsore	0.709
Kennethmont	0.714	Threlkeld	0.706
Aberchirder	0.716	Coedana	0.702
Peterhead	0.717	Charnian	0.706-0.707
Carn Chuinneag	0.710		
Dunfally Hill	0.718		
Glen Clova	0.729		
		Lower Devonian	
Lochnagar	0.707	Omey	0.706
Garabal Hill	0.705	Inish	0.704
		Roundstone	0.705
		Galway	0.705
		Leinster	0.704-0.708
		Newry	0.706
		Weardale	0.706
		Shap	0.708
		<i>Hercynian</i>	
		St Austell	0.706
		<i>Tertiary</i>	
		Skye	0.708-0.714
		Lundy	0.734
		Mull (Granophyres)	0.709-0.710
		Arran	0.717

Institute of Geological Sciences 1977). The Helmsdale granite (table 1; figure 2) should also be mentioned here since high levels of uranium and fluorite are associated with this granite and the overlying Ousdale Arkose (Gallagher *et al.* 1971; Bowie, Simpson & Rice, 1973) and the small Abriachan granite which is also associated with U-F mineralization and fenitization. Both are thought to be among the latest of the forceful granites and are cut by the Helmsdale and Great Glen fault systems respectively. The Strontian granite has a similar tectonic setting. None of the three granites is associated directly with either strong gravity or aeromagnetic anomalies.

Simpson, Plant & Cope (1976) have argued that the mineralization at Helmsdale is fault controlled and post-dates the granite, and Watson & Plant (1979, this volume) suggest that it

occurred after unroofing of the pluton but contemporaneously with emplacement of the permitted granites. A marked aeromagnetic anomaly offshore at Helmsdale is interpreted as a buried Devonian granite.

Anomalous enrichments of uranium also characterize intrusions near the southern uplands suture zone (Loch Doon: table 1 and figure 2). Uranium mineralization occurs in SE–NW trending veins in the aureole of the Criffell granodiorite and in a shatter zone within the pluton (Miller & Taylor 1966; Gallagher *et al.* 1971; Bowie, Simpson & Rice, 1973).

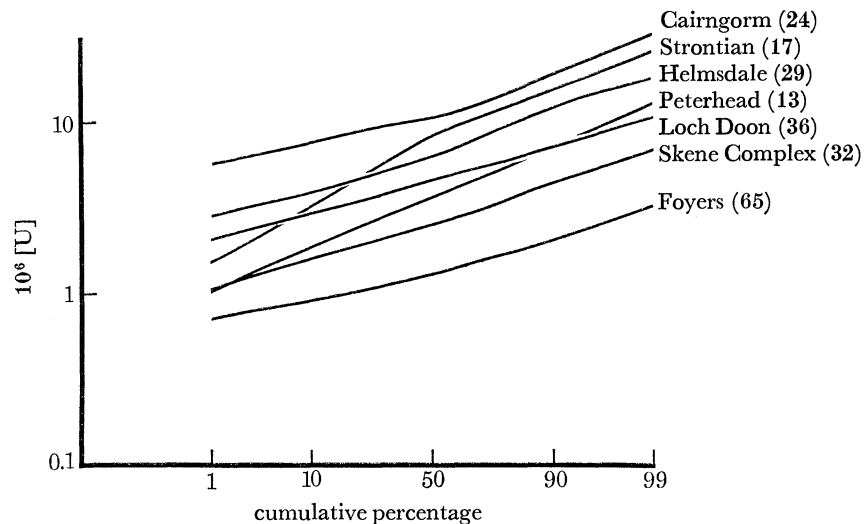


FIGURE 2. Uranium content (parts/10<sup>6</sup>) of selected Caledonian granites determined by the delayed neutron method and studied by the fission track method. Numbers of specimens analysed are given in parentheses.

#### (b) Hercynian granites

The Permian post-tectonic two mica granites of southwest England and their associated tin–tungsten–fluorite uranium mineralization are characteristic of the Hercynian province which extends through the Erzgebirge to central Europe. A model for the granites and metalliferous mineralization must be generally applicable to the European Hercynian province.

Several plate tectonic models have been used to interpret the geology and mineralization of southwest England (Badham & Halls 1975; Bromley 1975; Mitchell 1974; Riding 1974). A collision hypothesis has been particularly favoured as an explanation of the tin mineralization (Mitchell & Garson 1976), whereby sediments deposited on oceanic crust which normally carries only minor amounts of tin were under-thrust by continental sial from which granite and tin are thought to be derived at depth. Mitchell (1974) drew an analogy between the high potassium southwest England granites and those of the late Tertiary Malarkachung granite of the Himalayas. Himalayan style tectonics are notably lacking in southwest England, however, and the scarcity of large nappes and the lack of areas of strong post collision uplift arising from one continental plate overriding another have been noted by Lorenz (1976). Regional negative Bouguer anomalies other than that associated with the granite are also lacking (Institute of Geological Sciences 1976).

The model used in this paper is based on those of Dewey *et al.* (1973), Badham & Halls (1975) and Badham (1976). During the Lower Palaeozoic, southwest England was part of the stabilized South Europe plate. Subduction of Tethyan oceanic crust probably began in the Lower Devonian following closure of the 'Caledonian seaway' (Badham & Halls 1975).

Various major strike slip faults were initiated at this time which divided the south Europe Brioverian–Cadomian craton into a series of independent microplates. As the ocean between the North America – Europe plate and the South Europe plate was consumed along a southerly dipping Benioff zone the microplates were rotated and displaced, perhaps partly as a result of a component of lateral movement between the two major plates (Badham & Halls 1975; Badham 1976).

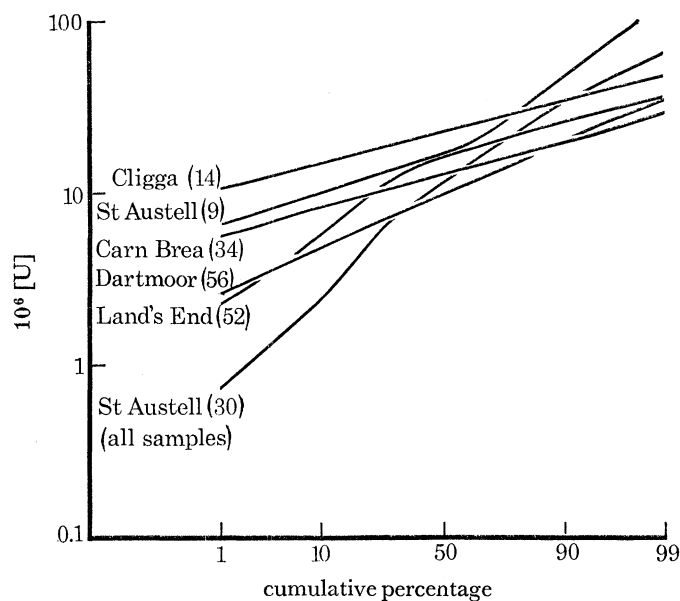


FIGURE 3. Uranium content of selected Hercynian granites determined by the delayed neutron method and studied by the fission track method. Values for Cligga are for greisen veins only. Numbers of specimens analysed are given in parentheses.

The microplates are thought to have impacted with the North American craton, an event which had a similar style and timing from Cornwall to Spain and Bohemia and was followed by emplacement of closely similar granites and their associated mineralization. The general dependence of the distribution of the late Variscan tin–tungsten (uranium) bearing granites on Precambrian structures of the European Variscides has been indicated by Zoubek (1977).

The southwest England granites intrude a sequence of volcanics, grits and conglomerates which had previously been metamorphosed under low grade greenschist facies. The plutons, which include Land's End, Tregonning-Godolphin, Carnmenellis, St Austell, Bodmin and Dartmoor, are thought to represent the exposed cupolas of a batholith which trends WSW and extends for over 250 km of southwest England (Bott *et al.* 1958; Edmonds *et al.* 1975; Beer, Burley & Tombs 1975; Tombs 1977). The commonest lithology is a coarse biotite adamellite passing into biotite–muscovite adamellite and enriched in several incompatible elements including B, Li and F (Exley & Stone 1964). K feldspar megacrysts are widely distributed, and two of the plutons, St Austell and Tregonning-Godolphin, contain lithium mica granite and fluorite granite.

Mean uranium contents for the granite plutons in this study (table 1 and figure 3) range between 11.3 and 24.2 parts/ $10^6$ . The overall mean for the Cornubian batholith is 14.0 parts/ $10^6$ . These values are high compared with published values of the average concentration of uranium in granite (Peterman 1963).

The mineralization is spatially associated with the granites (Dines 1956; Dangerfield & Hawkes 1969) and includes tin and copper with arsenic, tungsten, molybdenum and fluorite in east–west veins and cobalt–nickel–uranium mineralization in younger north–south veins. Uranium mineralization is sporadic and pitchblende is found with secondary minerals such as autunite, torbernite and zippeite in lenses principally at the intersections of cross fractures (Dines 1956). At least three periods of uranium mineralization have been proposed on the basis of the U–Pb method: at 290 Ma, contemporaneous with emplacement of the granites; at 225 Ma; and at 50 Ma, coinciding with emplacement of the Tertiary Lundy Island granite (Darnley *et al.* 1965). Although interpretation of the isotopic data is controversial, field and other evidence indicates several phases of introduction of mineralization and there is some indication that this continued after unroofing of the granites in Permian times (Dangerfield & Hawkes 1969).

(c) *Tertiary granites*

Tertiary intrusive activity in NW Britain was initiated in the preliminary stages of sea floor spreading as America rifted from Europe, starting some 70 Ma ago, and took place within the outer margin of sialic continental crust. Centres of activity developed where pre-existing northeast–southwest faults intersect the north–south rift. Extrusive and minor intrusive magmas associated with these centres are of olivine tholeiite and nepheline normative compositions which require a mantle depth of some 80 km for their generation (Thompson *et al.* 1972). Two principal possibilities for the production of granitic liquids in the Tertiary have been considered: fractional crystallization of large volumes of basic magma, or partial melting of sialic continental crust due to high heat flow from the mantle (Gass & Thorpe 1976). It is important to determine the source of the magma since this has implications for the genesis of uranium associated with the granites. The Tertiary granites have strong positive Bouguer anomalies which, coupled with strong aeromagnetic anomalies, indicate the presence of dense, magnetic gabbro cylinders penetrating from mantle depths and underlying a thin granitic layer (Bott & Tuson 1973; Brown & Mussett 1976). Estimates by Bott & Tuson (1973) show that only 5% of the total igneous mass in Skye is granitic.

Uranium data are available for the Skye, Mourne Mountains and Lundy granites (table 1; Tammemagi 1976; Moorbath & Welke 1969). Mineralogically and geochemically the rocks range from granite (s.s.) to alkali granite with the Mournes tending more strongly towards the latter (Meighan & Gamble 1974). The Mourne granite is enriched in uranium compared with the Lundy granite where the uranium content is background (Devonian slates?) and Skye where the uranium content is very low. Rare earth data for Skye and the Mournes (Meighan & Gamble 1974; Thorpe, Potts & Sarre 1977) suggest that both acid magma suites originated by fractional crystallization of basic magma which might have been contaminated by crustal material.

The high level Tertiary granites have uranium concentrations between those of the older Scottish granites and the Cairngorm granite of Northern Scotland. The extent to which this is a primary feature or is related to removal of uranium in hydrothermal solutions is discussed in more detail later in the paper.



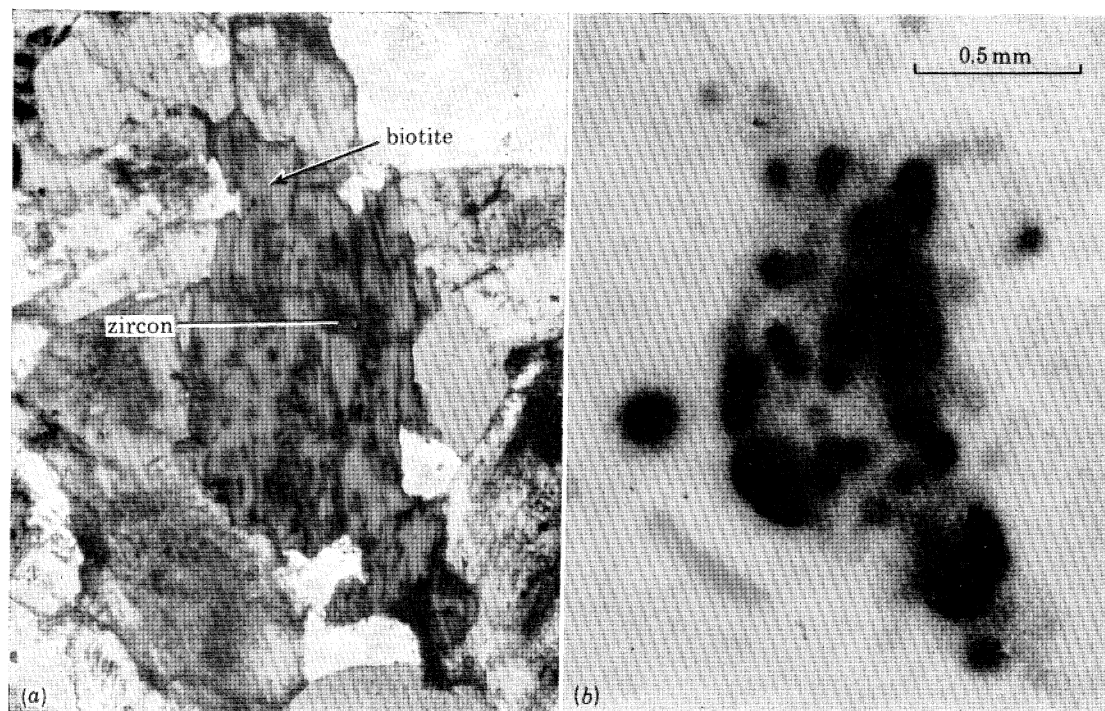


FIGURE 4. Uranium in zircon in primary biotite from Cairngorm. (a) Polished thin section; (b) lexan print showing fission track distribution.

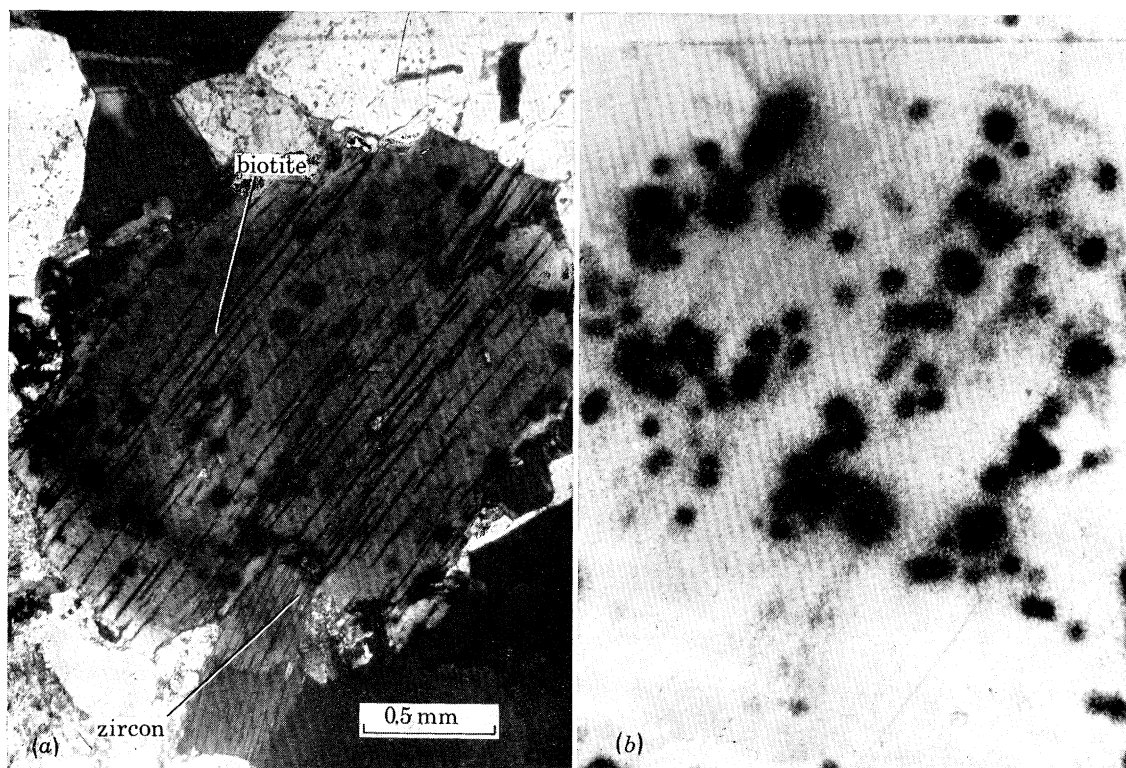


FIGURE 7. Uranium in zircon in primary biotite from Southwest England (Dartmoor). (a) Polished thin section; (b) lexan print showing fission track distribution.

(Facing p. 397)



(d) *Mineralogical evidence for uranium concentration and redistribution*(i) *Evidence for the derivation of uranium from the lower crust/upper mantle*

Evidence collected from three separate localities suggests that uranium in some British granites is derived from the lower crust/upper mantle.

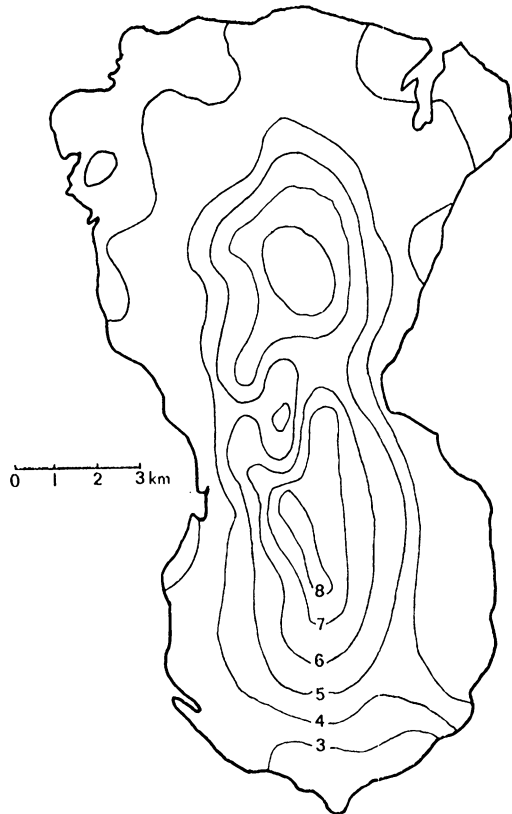


FIGURE 5

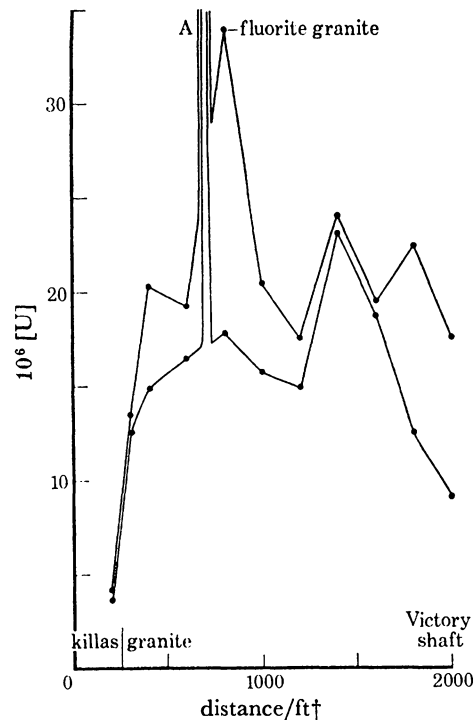


FIGURE 6

FIGURE 5. Uranium distribution in Loch Doon granite determined by gamma ray spectrometry and calibrated by the delayed neutron method. Uranium contoured in parts/ $10^6$ .

FIGURE 6. Uranium content of whole rock samples from underground traverse in Geevor tin mine, 14 level. The uranium-rich vein at A contained 2187 parts U/ $10^6$ . † 1 ft = 0.3048 m.

Geochemical maps show that uranium and beryllium are anomalously enriched in the Cairngorm granite relative to the metamorphic country rock and to other granites in the region (Institute of Geological Sciences 1977). Follow-up rock sampling, whole rock analysis for uranium, and fission track study show that in the typical medium to coarse grained Cairngorm granite (mean 12.7 parts U/ $10^6$ ) uranium is held in zircon in primary biotite (figure 4). A minor separate fine-grained granitic phase, which outcrops along the Lairig Ghru and on the plateau immediately to the northeast, contains a much lower mean value of 3.3 parts U/ $10^6$  which is similar to that obtained from the metamorphic country rock.

At Loch Doon, uranium determined by gamma ray spectrometry (figure 5), calibrated by the delayed neutron method and plotted against the differentiation index (Brown, Cassidy & Hennessy 1977), shows that uranium increases with magmatic differentiation. Fission track

study shows that uranium is concentrated in zircon in primary biotite in the uranium-rich core of the pluton (about 8 parts/ $10^6$ ) but is present mainly in apatite in the uranium poor marginal phase (about 3 parts/ $10^6$ ). In both areas the uranium is held in primary magmatic minerals.

At Geevor mine in the Lands End granite and South Crofty mine in the Carn Brea granite, underground sampling traverses across the killas/granite contact, show that the granite is enriched in uranium, up to the sharp contact (figure 6). The uranium content falls steeply in

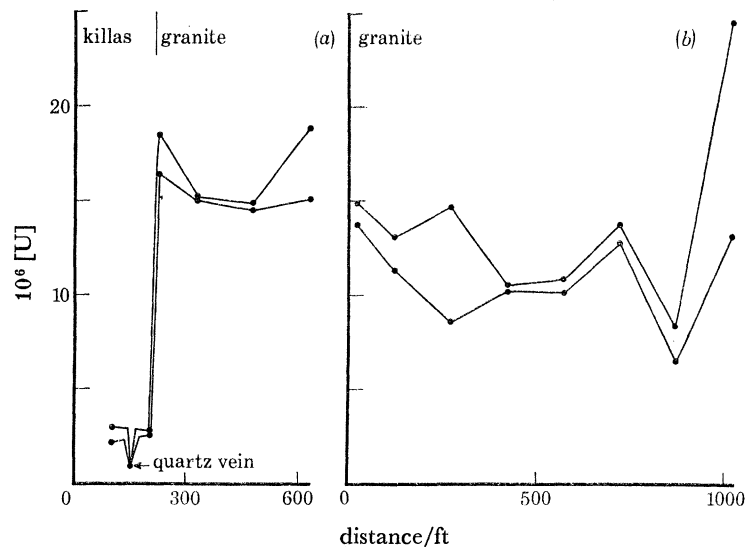


FIGURE 8. Uranium content of whole rock samples from two underground traverses in South Crofty tin mine. (a) Rosskear crosscut north; (b) Cooks 340 main crosscut north.

the killas to uniformly low values (less than 3 parts/ $10^6$ ). There is here no geochemical evidence for mobility of uranium across the contact. The uranium in the granite is held in zircon in primary biotite. This is also a feature of uranium distribution in Dartmoor (figure 7) studied by Simpson, Plant & Cope (1976). The mineralogical evidence suggests therefore that the high uranium levels of the granites studied are a primary magmatic feature. This is accounted for by derivation from a lower crustal/upper mantle source region and not by contamination from sediments intruded by the granites since high initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios would also occur. Furthermore, the uranium-rich granites such as the Cairngorm Massif and the Cornubian batholith appear to have been intruded rapidly along deep faults during brittle fracture late in the orogenic cycle further suggesting that the uranium comes from depth.

#### (ii) *Uranium redistribution processes*

A mineralogical investigation of processes involved in the remobilization of uranium leading to mineralization and depletion has been undertaken in the Cornubian batholith. The highest value for mineralized granite (66.6 parts U/ $10^6$ ; table 1) is from a coarsely megacrystic rock from Goonbarrow china clay pit in the St Austell granite which contains abundant outcrops of megacrystic and 'feather textured' granite with potash feldspar xenocrysts. Biotite containing uraniferous zircon forms radiating sheaves of crystals, suggesting that uranium was enriched to this extent late in the magmatic cycle. The occurrence of uraniferous megacrystic granite in

the Goonbarrow pit is therefore thought to provide evidence of the interaction of volatiles and water with hot granite, and the later formation of kaolinized granite also at this site suggests throughput of volatiles at several stages during cooling of the magma. Sites such as Goonbarrow are closely related to the 'emanative centres' discussed by Dines (1933).

Fresh, unmineralized and homogeneous granite samples from underground traverses in Geevor (figure 6) and South Crofty mines (figure 8) were studied. The specimens were generally paired to provide estimates for within and between-site differences. Local occurrences of vein type uranium mineralization are discussed later in the text.

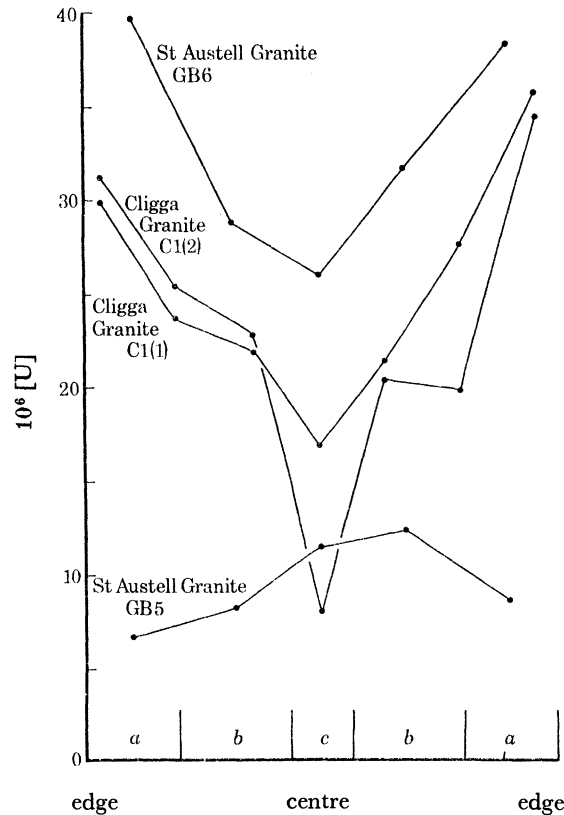


FIGURE 9. Uranium distribution in greisen veins from the Cornubian batholith. Overall width approx. 25 cm; a, kaolinized granite; b, greisenized granite; c, quartz-tourmaline vein.

Two of the traverses (Geevor and South Crofty, Roskear crosscut north) are sited in granite cut by tin veins near to the killas/granite contact. The third is sited (in South Crofty, Cooks, 340 main crosscut north) in granite, away from known mineralization and the killas/granite contact. This traverse has the lowest uranium content although it is anomalously high when compared with the unmineralized Dartmoor granite (table 1) (Simpson *et al.* 1976). The traverse in Geevor mine where discrete mineralized veins occur has the highest uranium values for unmineralized granite, and the Roskear traverse has intermediate values. However, in all three traverses, the uranium is held in zircon in primary biotite. This study suggests that the distribution of uranium and tin mineralization is associated with late magmatic or deuteric enrichment of uranium in the region subsequently cut by vein mineralization.

(iii) *Greisen veins*

Two greisen veins from Goonbarrow china clay pit in the St Austell granite, and one from Cligga Head, have been studied (figure 9). All have a central quartz–tourmaline vein system with a marginal greisen consisting essentially of quartz and white mica, passing outwards into granite unaffected by the greisenizing fluids, but which is partly kaolinized. These features indicate that the greisenized granite is fresher and more resistant to subsequent kaolinization than altered granite. It should therefore be possible to study the primary distribution of uranium within the greisen vein system even though the greisen veins cut granite which has generally been greatly altered and kaolinized.

The greisen veins were sliced symmetrically about the centre of the vein, usually along the well defined boundaries between the principal portions of the vein systems described above. The separate portions were analysed for uranium content, and, in the case of the Cligga vein, sampled and analysed in duplicate.

Two distinct uranium distributions are apparent (figure 9). The variation of the first type (no. 6 from Goonbarrow and nos 1(1) and 1(2) from Cligga) involves relative depletion in the central portion of the vein; uranium content increases progressively towards the margin of the vein which is partially kaolinized. Fission track analysis indicates that uraniferous zircon is present in normal amounts in the kaolinized margin, undisturbed by greisenizing or kaolinization, and occurs to a lesser extent in biotite altered to white mica in the outer edge of the greisenized zone. In the centre of the vein, zircon is completely replaced and the uranium normally held in this phase is dispersed. Secondary redeposition of uranium is observed in hairline cracks which parallel and cut the earlier tourmaline veins. This suggests that uranium is released into solution and remobilized with the progressive dissolution of zircon during the greisen vein formation. In the waning stages and at lower temperatures, uranium is redeposited in this portion of the vein system, overgrowing and cutting the higher temperature low-uranium mineral assemblages. This suggests that the vein system is a pathway for the upward migration of uraniferous solutions. The china clay pit at Goonbarrow and other vertical exposures of granite such as Cligga Head exhibit abundant parallel subvertical sheeted greisen veins which are shown in this study to have remobilized uranium held in zircon and probably represent an important phase in the development of uraniferous hydrothermal systems in this batholith.

In the second type of variation (no. 5 from Goonbarrow; figure 9), the uranium content at the vein margin is lower than at the centre. Depletion of uranium in the centre of the vein is attributed, from the textural studies, to the combined effect of greisen and tourmaline formation as before. Relict haloes are clearly observed in tourmaline which has replaced primary biotite and zircon with consequent loss of uranium. The lower uranium values on the vein margin are accounted for by a pervasive general alteration of the biotite to tourmaline before kaolinization, with resultant loss of uranium from zircon. In this case only a few large zircon crystals survive, in tourmaline formed at the expense of biotite.

In disaggregated and kaolinized granite from the Godolphin granite which contains only 8 parts U/10<sup>6</sup> and Bostraze china clay pit in Lands End granite with only 5.4 parts U/10<sup>6</sup>, biotite is now absent, but small secondary tourmaline grains are common both here and in other kaolinized samples. This suggests that granite, before general kaolinization, was hydrothermally altered by deeply penetrating boron metasomatism which replaced zircon and

biotite with tourmaline, thus releasing uranium. The few remaining zircons that have not been replaced by tourmaline are stable, although the granite is now completely kaolinized. The extent to which this type of extensive tourmalinization occurred before kaolinization is not known but the close correspondence in the samples studied, combined with the generally low levels of uranium observed both in this study and previously by Badham *et al.* (1976) suggest that a close link exists between these two processes. Kaolinized zones are therefore likely to represent areas of general upward remobilization of uranium through hydrothermal systems following the greisenizing event. Secondary redeposition of uranium in close cutting chloritic veinlets is observed (GB 6; figure 9), indicating that minor amounts of uranium may be redeposited in kaolinite.

Boron metasomatism is also observed in the Wheal Remfrey breccia pipe in the St Austell granite. This is an explosion breccia containing a mixed population of rounded to angular fragments of granite and killas in a tourmaline rich matrix. Uranium contents for the breccia range between 1.4 and 11.7 parts U/10<sup>6</sup> suggesting a loss of uranium by boron metasomatism. Rocks that are partly or completely haematitized are commonly encountered, representing vein systems in granite traversed by oxidizing solutions and these samples are all relatively depleted in uranium compared with whole rock values.

#### 4. ISOTOPIC AND OTHER GEOCHEMICAL EVIDENCE ON THE SOURCE OF URANIFEROUS GRANITES IN THE BRITISH ISLES

It has been demonstrated that uranium enrichment in British granites has a primary magmatic origin and it is therefore important to deduce the source of the associated magmas. Few lead isotope determinations have been reported but strontium isotopic data are more generally available (table 2).

Initial <sup>87</sup>Sr/<sup>86</sup>Sr ratios ( $r_i$ ) are thought to distinguish between magmas derived from sources rich in radioactive <sup>87</sup>Rb such as the continental crust ( $r_i$  usually  $\geq 0.710$ ) and <sup>87</sup>Rb-poor sources such as the mantle ( $r_i$  usually 0.704) (Faure & Powell 1972). However, oxygen isotopic studies have introduced doubts about the interpretation of the strontium isotopic data with the use of a simple closed system model and the possibility of contamination of mantle-derived Tertiary granites by mature crustal strontium within hydrothermal systems has been widely discussed (e.g. by Taylor & Forester 1971; Walsh *et al.* 1978).

Other doubts concerning the significance of high initial strontium ratios have been raised: Bowden, Lameyre & Vidal (1977) showed that in peralkaline complexes of Nigeria, high  $r_i$  values cannot unequivocally be attributed to crustal melting particularly in granites with evidence of volatile activity. High values could result from internal strontium isotope homogenization or Rb metasomatism shortly after emplacement (Moorbath 1965). Bailey & Macdonald (1975) have independently provided evidence for the migration of Rb as a volatile phase correlated with F levels but unrelated to feldspar fractionation.  $\delta^{18}\text{O}$  values provide evidence of processes involving epizonal waters while correlations of Rb/Sr ratios with initial strontium ratios provide evidence of rehomogenization or redistribution processes. Such evidence is not available systematically for British granites.

Lower Devonian granites in the northern province, which are the only Caledonian intrusions to show uranium enrichment (table 2), have unambiguous mantle-type values of  $r_i$ . A tendency for  $r_i$  to increase with time is not detected, and a reverse, *cooling* trend was deduced from a



comparison of ages and  $r_1$  by Brown & Hennessy (1978). This contrast with modern Cordilleran granites of the Andes (McNutt *et al.* 1975) gives support to the post-closure emplacement model suggested earlier. Tensional rebound would promote the escape of magma to high crustal levels and Read's (1961) concept of late, permitted granites is appropriate in this context. The low initial ratios show that crustal scavenging during ascent of the magma is unlikely to have resulted in the acquisition of significant uranium. The older granites of the southern province also have mantle-type ratios. The higher values of the pre-Lower Devonian group of the northern province are consistent with the more intense tectonism and are thought to reflect crustal melting.

TABLE 3. K/Rb RATIOS AND TOTAL Sr CONTENTS OF GRANITES IN THE BRITISH ISLES  
(r, rock; s.s., stream sediment.)

	K/Rb	total Sr
Caledonian		
Foyers (s.s.)	203–267	492–633
Loch Doon (r)	181	234
Cairngorm (s.s.)	112	65
Hercynian (r)	90	50

Isotopic studies in the Cornubian batholith are represented by a single value for  $r_1$  (Harding & Hawkes 1971) supported by a series of D/H and  $^{18}\text{O}/^{16}\text{O}$  measurements (Sheppard 1977). The St Austell granite has a low value of  $r_1$  (0.706) which is much lower than the crust into which it is emplaced (0.720; Floyd 1972) and predominantly mantle derivation would account for this; the correlation of U rich granites and low  $r_1$  values also holds for the Hercynian.  $\delta^{18}\text{O}$  values (10.8–13.2‰; Sheppard 1977) are above the normal range for magmatic water as in most muscovite-bearing granites (Taylor 1977). Later greisenization produces  $^{18}\text{O}$  depletion generally attributed to meteoric hydrothermal circulations.

In the Tertiary province,  $r_1$  in the low uranium granites of Skye may have been raised by meteoric circulation indicated by  $^{18}\text{O}$  depletion (Taylor & Forester 1971). Uranium-rich intrusions elsewhere in the Tertiary province are unlikely to be altered in this manner and  $r_1$  may give more definitive information on the granites. The situation in the Cornubian batholith suggests that pervasive alteration of granite by circulating hydrothermal solutions does not eradicate the primary levels of uranium and the general uranium levels in Tertiary granites may therefore be primary.

Relatively few geochemical data on British granites are available (table 3). The Cornubian batholith is characterized by low total Sr, low K/Rb ratios (Harding & Hawkes 1971) with high levels of incompatible elements (Sn, U, Li, Rb, Be, B, F) (Floyd 1972; Hall 1973). Regional geochemical maps over northern Scotland indicate that the pre-Devonian granites are characterized by high levels of total strontium, and levels of incompatible elements and K/Rb ratios comparable to background for the region, the principal exception of the area covered to date being Cairngorm which has characteristics similar to unmineralized Cornubian granites (table 3).



## 5. GEOTHERMAL GRADIENTS

Geothermal gradients in the crust are dependent on many factors and in a static system (where magma is immobile and heat transfer is by conduction alone), the principal controls are the temperature of the base of the conducting layer and the heat production and thermal conduction of this layer. Many authors, especially Albarede (1975), have commented on the existence of a linear relation between heat production and heat flow for continental provinces. The heat flow for zero production from radioactive decay in the layer is due entirely to conduction from the base of the layer but, where upper crustal heat sources contribute to the heat flow, geothermal gradients increase within a few kilometres of the surface. Thus, an average gradient of less than 10 °C/km for the continental lithosphere is greatly enhanced in static upper crust which has high heat production and can reach 40–50 °C/km without hydrothermal convection as at Los Alamos (E.R.D.A. 1977).

In Britain, Garnish (1976) has summarized the available thermal data and although the mean, 'background' geothermal gradient is only 25 °C/km, high values are associated with rising hot water in certain deep sedimentary basins and with hot rock within the granite areas of Cornwall and Durham (Weardale) which are also notable areas of metalliferous mineralization. These hot rock provinces correlate with high uranium concentrations (table 1). No systematic study of geothermal heat production has been published for Britain and there are major gaps in recorded data but this correlation noted between hot granites and uranium concentration could probably provide a reliable test for identifying hot rock provinces.

For example, in the Cornubian province where uranium levels in bedrock are exceptionally high, Tammemagi & Wheildon (1974) have reported observations of heat flow, ranging up to three times the national average at 0.18 W m<sup>-2</sup>, and geothermal gradients between 40 and 50 °C/km: these are equilibrium gradients, due to conduction alone. The hydrothermal convective systems which were responsible for secondary mineralization would be difficult to initiate even in such hot rock regions where heat is supplied only by conduction through solid rock. High temperature hydrothermal processes would be initiated in the post-magmatic period when the silicate liquid-solid interface, at about 700 °C, was within a few kilometres of the surface. Low temperature hydrothermal systems could be maintained and fluid flow focused into faults and fractures within and above the granite roof, above local geothermal anomalies. In Caledonian granites, such hydrothermal systems were apparently short-lived and little evidence of post-magmatic uranium redistribution is found (see primary distribution of uranium in Loch Doon, figure 5). However, in Hercynian granites, heat producing elements appear to have been concentrated around zones which acted as fluid channels. In contrast to U-rich Caledonian and Hercynian granites, geophysical evidence indicates that Tertiary granites overlie U-poor basic intrusive 'cylinders' and therefore any high heat productivities measured at the surface cannot be extrapolated to depth. Hence prolonged hydrothermal convection with consequent uranium redistribution is not favoured.

## 6. DISCUSSION

In Britain the highest concentrations of uranium in granites occur in the intrusions of south-west England, and the late permitted granites of the Caledonian. These intrusions are characterized by low initial <sup>87</sup>Sr/<sup>86</sup>Sr ratios, high δ<sup>18</sup>O values, low K/Rb ratios and high levels of

incompatible elements including Sn, F, Li, Be, Th and Pb. Fission track and other geochemical studies indicate that the high background concentrations of uranium away from mineralization are related to uranium occurring as resistate primary phases. Within individual granite plutons, uranium concentration increases with an increase in fluorite and topaz as accessory minerals (St Austell, Cairngorm) but high level assimilation of country rocks does not appear to have increased the concentration of uranium or other incompatible elements (Loch Doon).

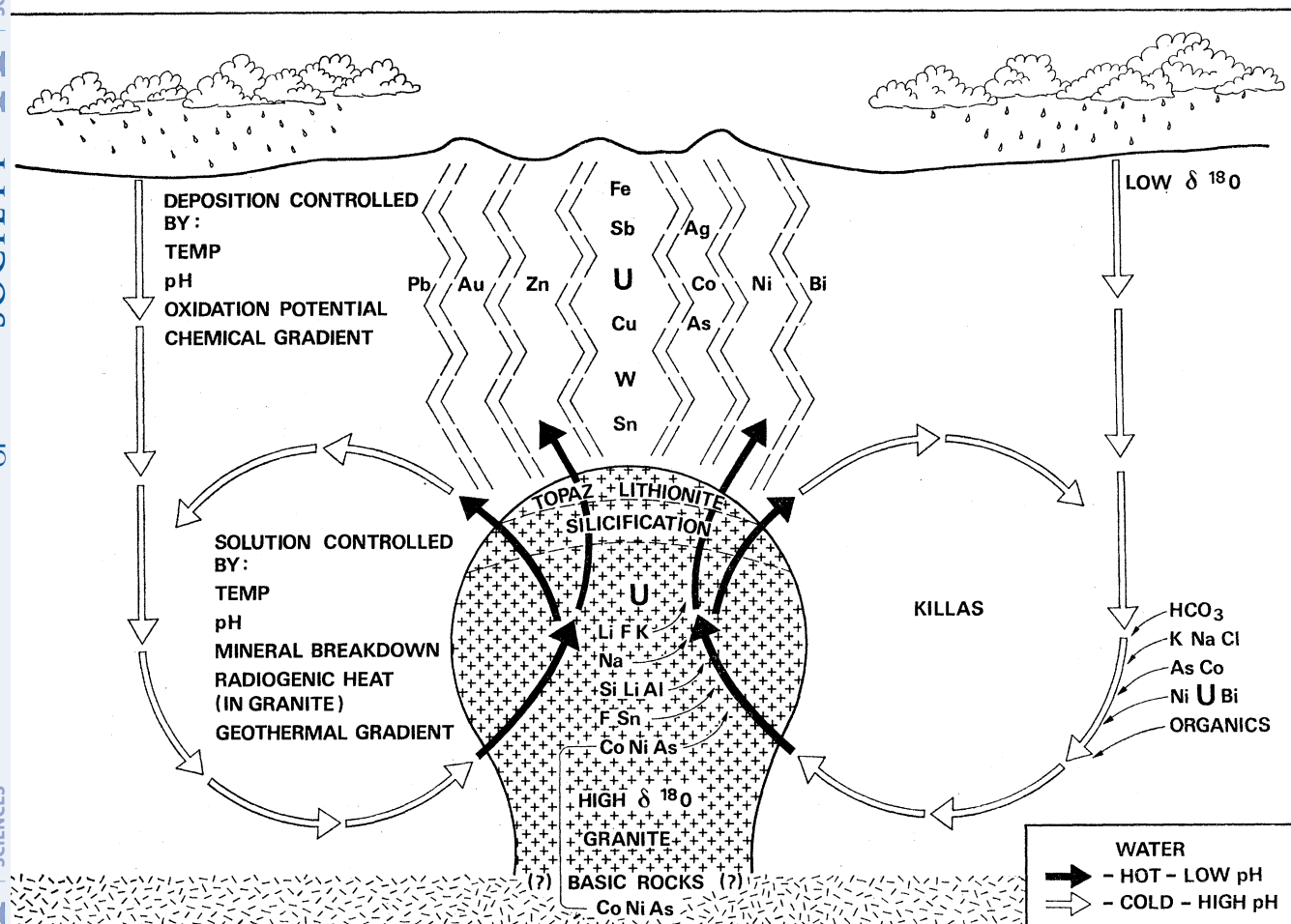


FIGURE 10. Model for the interaction between granite magma enriched in metals, fluorine and chlorine and meteoric hydrothermal convective systems to produce vein type mineralization.

Mineralization and greisenization are shown by D/H and  $^{18}\text{O}/^{16}\text{O}$  (Sheppard 1977) studies to involve dominantly meteoric water which probably circulated in a convecting hydrothermal system. Field, metallogenic and U–Pb data suggest that several episodes followed emplacement of hot magma and continued after unroofing of the granite. Textural evidence strongly suggests that mineralization involved progressive breakdown of silicates and resistate accessory minerals followed by redistribution of U, Zr, Sn, rare earths, Be and other elements to form a near-surface system of ore veins and lodes. The critical basic requirement for the leaching of tin by hydrothermal fluids has been shown to be a minimum activity of fluorine in mineralizing solutions (Barsukov 1957; Beus & Sobolev 1964; Tischendorf 1973). Tin carried principally in dark micas is released when the later are sericitized and chloritized and Tischendorf has

calculated that 150 g Sn, 3 kg Li and 8600 g F are released per tonne of sericitized dark mica of the main intrusive phase of the western Erzgebirge pluton. The presence of acid-forming volatiles (F, Cl,  $\text{BO}_3^-$ ), of which F is the most important, is the principal control on the solubility of tin (Barsukov & Kuril'chikova 1966). A scheme proposed for post magmatic tin-fluorite mineralization by residual granite fluids (Beus & Sobolev 1964) can be used as a basis for considering the relation between U and Sn mineralization in 'two mica granites' to be due to interaction of meteoric hydrothermal water with granite. This scheme involves heating and acidification of alkaline surface meteoric water as a result of dissolution of acid-forming volatiles such as  $\text{F}^-$  from granite magma. The acid solutions extract tin from mica which is subsequently deposited as cassiterite, F being precipitated as topaz and fluorite. The uranyl ion forms several complexes with  $\text{F}^-$ ;  $(\text{UO}_2\text{F}_6)^{4-}$  is stable up to pH 6.7 at 25 °C and uranous ions form a soluble fluoride complex, possibly  $(\text{UF}_2)^{2+}$ , which is stable up to pH 4 (Rich, Petersen & Holland, 1977). Thus uranium fluoride complexes can exist in acid solutions irrespective of the oxidation potential of the mineralizing solution. Dissolution of uranium from primary minerals in a hydrothermal system associated with tin and fluorite would be consistent with textural information.

This system is thought to be generally comparable with those described by Ellis & Mahon (1964) from active volcanic areas where high-temperature low-pressure steam contains high fluoride derived from hydrogen fluoride. With a decrease in temperature the acid sulphate-fluoride system evolves into acid sulphate waters containing carbon dioxide. Postmagmatic circulation would probably involve neutral chloride waters with sodium and potassium and containing arsenic, boric acid, sulphate, bicarbonate, ammonia, bromide, fluoride, lithium rubidium and caesium. Fluid inclusion studies of pitchblende mineralization from the Hercynian province of France and Europe suggests that pitchblende is generally deposited at temperatures below 150 °C from  $\text{CO}_2$ -bearing aqueous fluids of low salinity. Thus uranium is thought to be maintained in solution as soluble carbonate complexes and to be deposited only at the cool surface part of the convecting hydrothermal systems.

A comparison of the sodium, potassium and hydrogen ion concentrations in deep waters from hydrothermal areas (Brown & Ellis 1970) with experimental data (Ellis & Mahon 1967) reveals that high temperature (200–350 °C) water compositions within many rock types correspond to the equilibrium assemblage quartz, potassium, feldspar, albite, muscovite (Ellis 1973), providing further evidence that many of the characteristics of 'two mica granites' are the result of interaction of biotite adamellite with epizonal meteoric waters.

Thus a model (figure 10) involving deep circulation of meteoric water which is initially alkaline but becomes strongly acidic by interaction with hot granite magma containing acidic ions, particularly fluorine, is favoured for the main mineralization event. During hydrothermal circulation, primary minerals are dissolved and uranium ions from accessory minerals are released into solution and subsequently redeposited in near surface low temperature veins. Further uranium mineralization events could have resulted from the circulation of warm fluorine and/or bicarbonate bearing solutions.

Although some uranium, fluorine and tin may be derived from the aureole, the evidence from Loch Doon and southwest England respectively suggests that the principal source of these metals is from hot granite magma and subsequently from granite maintained at a high temperature by the local geothermal gradient reflecting the local concentration of uranium, potassium and thorium.

Mineralization is most likely to occur when a hot granite magma is emplaced into wet sediments and a convective hydrothermal system established. However, conductive sources of heat later than the granite could continue to drive such a system and could therefore also bring about mineralization. This could explain the continuation of mineralization in several episodes which may continue after unroofing of granites particularly at times of higher mantle convective heat flow such as the Tertiary. The local geothermal anomaly related to concentration of heat producing elements would be enhanced at such times. The model developed here depends on high primary concentrations of tin, uranium and other elements in granites and the problem of determining the fundamental controls of mineralization is that of identifying the conditions necessary to generate granite magmas with high levels of volatiles and incompatible elements.

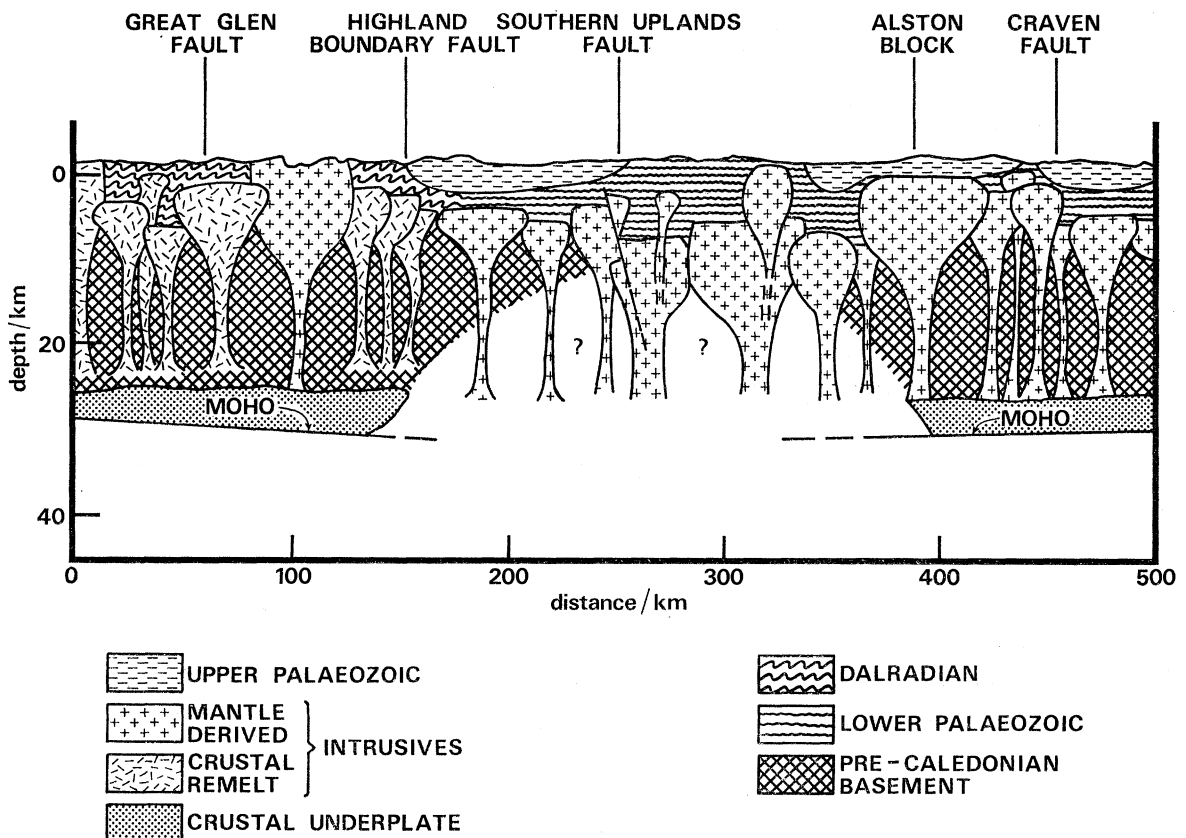


FIGURE 11. Suggested relation between granites and the deep structure of Northern Britain.

#### *Genesis of uraniumiferous granites*

It has been argued earlier that uranium enriched granites of the Hercynian and Caledonian provinces were derived from sub-crustal levels at active or recently active destructive plate margins. In the Caledonides the magmatism associated with uranium occurred during the declining phases of modified Andean type plate margins.

In the Hercynides, magmatism associated with uranium and tin occurred after collision and rotation between the microplates of South Europe and the North America–Europe plate. In the Caledonian, Hercynian and Tertiary provinces of Britain magmatism associated with



uranium enrichment may therefore have occurred in a tensional tectonic setting in which Precambrian crust was disrupted (figure 11).

The consanguinity of granites with extrusive andesite suites such as at the modern plate margin of the Andes is beyond doubt (Brown 1977) and it appears that progressive metamorphism of subducted ocean crust provides an important source of volatiles and molten magma, which may ascend, scavenge and partially melt both the overlying mantle wedge (Best 1975) and, to a progressively greater extent, the lower crust (Brown & Hennessy 1978). Modern destructive plate margin magmas show a potassium enrichment trend with decreasing K/Rb ratios from margin to interior (Nielson & Stoiber 1973), and in the same direction Sillitoe (1974) found a zonation of mineralization from iron through copper–molybdenum and copper–lead–zinc to tin–tungsten associations. Many of the high uranium British granites have the geochemical characteristics of intrusions remote from marginal areas and are of mantle initiation to judge from their low initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, and their uranium is a primary magmatic component.

In order to account for the characteristics identified for uraniferous granites it is necessary to consider genetic theories for high K – low K/Rb, Sn–W-rich and U-rich granites, all of which have received independent attention. Closed system differentiation models for combining U, F and Sn enrichments in granites were considered and rejected by Simpson *et al.* (1976). Our present starting point is Bailey & MacDonald's (1975) model of a magma chamber open at depth to the ingress of volatiles rich in halogen with metallic elements forming preferred complexes with either F (Zr, Rb) or Cl (Nb, Yt and Zn). They suggested an origin for the volatiles by partial melting of phlogopite or Ti-rich clinopyroxene which are increasingly recognized as significant upper mantle phases. For example, Beswick (1976) assumed that almost all upper mantle K and Rb are concentrated in phlogopite which is able to release volatile fluids over a wide stability range owing to ionic substitution. In fact the high-pressure stability of phlogopite is known to increase markedly in cool environments (Flower 1971), such as a subduction zone, and the substitution of F for OH groups in the structure also increases phlogopite stability (Wones 1967). Some support for the influence of phlogopite in plate margin magmas comes from the study of distribution coefficients between melt and residuum during partial melting of basic to ultrabasic components. Where dehydration contributes to the melting process, K/Rb ratios in the melt are several orders of magnitude higher (*ca.* 1000) than under the influence of phlogopite (K/Rb *ca.* 200; Griffin & Murthy 1969). Thus Beswick (1976) maintains that the destructive margin magma trends towards K enrichment and falling K/Rb reflect not only a progressive decrease in the volume of melt but also a transition from amphibole to phlogopite as the main volatile bearing phase in the melting zone. A second alternative for producing progressive K-enrichment in subduction zone magmas, that of buffering by sanidine in eclogite (Marsh & Carmichael 1974), seems less satisfactory in view of the inevitable hydrothermal metamorphism of subducted oceanic lithosphere leading to the hydrous phases whose subsequent breakdown is required for the initiation of melting. A third possibility for K-enrichment was proposed by Best (1975) in which hydrous fluids, initiated by dehydration of subducted ocean crust, scavenge the overlying mantle wedge for incompatible elements: the degree of enrichment is proportional to the thickness of mantle scavenged, thus increasing from the margin – the so-called K–h trend.

Of the three K-enrichment processes that we have mentioned, those of Beswick (1976), which also accounts for the K–Rb trend, and Best (1975) seem most plausible but require further

constraints. Systematic *uranium* variations in plate magmas have received little attention but a report of E–W traverses over central Andean volcanics by Zentilli & Dostal (1977) showed that uranium and potassium do not vary sympathetically in the manner expected of a common source. U values increase from about 1 to above 3 parts/10<sup>6</sup> and then fall to below 2 parts/10<sup>6</sup> on average as K increases progressively. Zentilli & Dostal concluded that these enrichments over the axial region of the Andes, where crustal thickness is greatest, suggest a lower crustal contamination source for uranium. It would be instructive to follow initial strontium isotope ratios through the same region and also to analyse *intrusive* rocks which might show similar variation in uranium content. A further point that we consider important is that crustal thickening in the axial region is not satisfied by extrusion and intrusion alone but some degree of underplating of subcontinental lithosphere is inevitable as ascending cool hydrous magmas encounter cold crust (Brown 1973). This new underplated crust could be more easily scavenged for U than old U-poor refractory granulite basement. Accumulation of volatile and metal enriched juvenile crust beneath refractory basement would be favoured during the compressive phase of orogenesis.

It should be noted that an alternative hypothesis for the origin of subcontinental lithosphere has been proposed (Brooks *et al.* 1976) whereby lithosphere accreted from rising blobs from non-depleted mesosphere is reactivated to give magmas during tectonism such as subduction of rifting. A kinematic model for the mantle in which relatively uniform and non-radiogenic asthenosphere is penetrated by and mixed with plumes derived from an isolated (1500–2000 Ma) and chemically heterogeneous mesosphere has also been proposed by Brooks *et al.* (1976). Evidence for the derivation of incompatible elements including tin from the lower mantle has been provided from the Azores hot spot (Dmitriev *et al.* 1971) and from Iceland (Jankovic 1972). However, evidence which suggests that at least some U and K may be derived from subducted ocean floor is provided by the work of Hart (1969) and Maccougall (1977) who show, from the levels of U and K and the K/U ratio, that low temperature weathered basalt provides the major ocean sink for these two elements. Oceanic sediments contribute no U and K since, according to Fyfe (1979, this volume), they are not subducted.

## 7. CONCLUSIONS

In this paper we have suggested that uranium-enriched granite magmas of the British Caledonian and Hercynian provinces are derived from subcontinental lithosphere underplated onto pre-existing Precambrian basement; release of this magma is favoured by the tensional régime following the end of subduction at destructive margins. Magmas generated by anatexis within the continental sedimentary pile are not associated with uranium enrichment or mineralization (see Watson & Plant 1979, this volume). The source of the underplated magmas is not clear and a contribution from an undepleted mesospheric source may be involved, but derivation from subducted oceanic lithosphere controlled by phlogopite dehydration is favoured. The concentration of Sn and U and the low K/Rb ratios in the granite magmas are attributed to scavenging during the ascent of fluorine-rich volatiles after the breakdown of phlogopite. This occurs in the range 1000–1200 °C (Kushiro 1969) and at a maximum thermal gradient of 10 °C/km (Oxburgh & Turcotte 1970) implies a minimum distance of 100 km from the margin for a 45° subduction zone.

In the crust, mineralization occurs by the interaction of epizonal meteoric water with



uranium and volatile (fluorine)-rich magma; petrographical changes also occur to produce large quantities of 'two mica' granite. The high concentrations of uranium, thorium and potassium which produce hot rock regions may maintain hydrothermal mineralizing systems whereby groundwaters leach metals and volatiles from granite after emplacement and unroofing particularly during periods of higher average heat flow from the mantle such as the Tertiary. An extensive system of channels for heating and circulating water is required and faults in the granite would be particularly favourable.

This model for uranium mineralization associated with granites has considerable application to exploration at a regional and a local scale. For example, with the use of regional geochemical maps, subcrustal subduction-related granites can be readily distinguished from barren granites by their contents of uranium, incompatible elements, or K/Rb, Rb/Sr ratios or total Sr contents. Alternatively, analysis of fresh granite samples can distinguish between relatively barren crustal granites and subcrustal granites which may be associated with mineralization. Preliminary data from Scotland (Rachel Williams, personal communication to J.P.) suggest that the simple and inexpensive determination of fluorine in water by using a specific ion electrode could be used by exploration field workers to distinguish these two types of granite.

At a more local level, whole rock uranium analysis or gamma spectrometry may provide genetic information. Where this is complemented by mineralogical studies with the use of fission track methods for uranium, it should be possible to identify from mineralogical changes the precise position of the sample in respect to the hydrothermal system. In this way a high degree of control of drilling for mineralization is possible.

The complex interrelation between ancient geotherms, modern geothermal systems, and uranium enrichment and mineralization is apparent. The most significant features of the model for uranium mineralization associated with granites are, first, that it explains the association of mineralization with tectonism and magmatism over a long period of time in the same area, and secondly, why in an area containing uraniferous granite, local geothermal anomalies may be capable of maintaining hydrothermal convective systems which can produce mineralization long after initial emplacement of the magma.

This paper forms part of a programme of work on studies of uranium occurrence and distribution in granites in the British Isles initiated and directed by Dr S. H. U. Bowie, F.R.S., when chief geochemist of the Institute of Geological Sciences, London. G. C. Brown wishes to thank R. S. Thorpe for helpful discussions and for unpublished data on the earliest Caledonian magmatism in Southern Britain. P. R. Simpson thanks K. Gilbert, Geevor Mine, C. Bristow, C. Gronow and I. Wilson of English China Clays Lovering Pochin and J. Lewis, South Crofty Mine for their cooperation in facilitating sample collection at the company properties. P. M. Green is thanked for data processing and R. Mogdridge for assistance with the reference list. The paper is published by permission of the Director, Institute of Geological Sciences, London.

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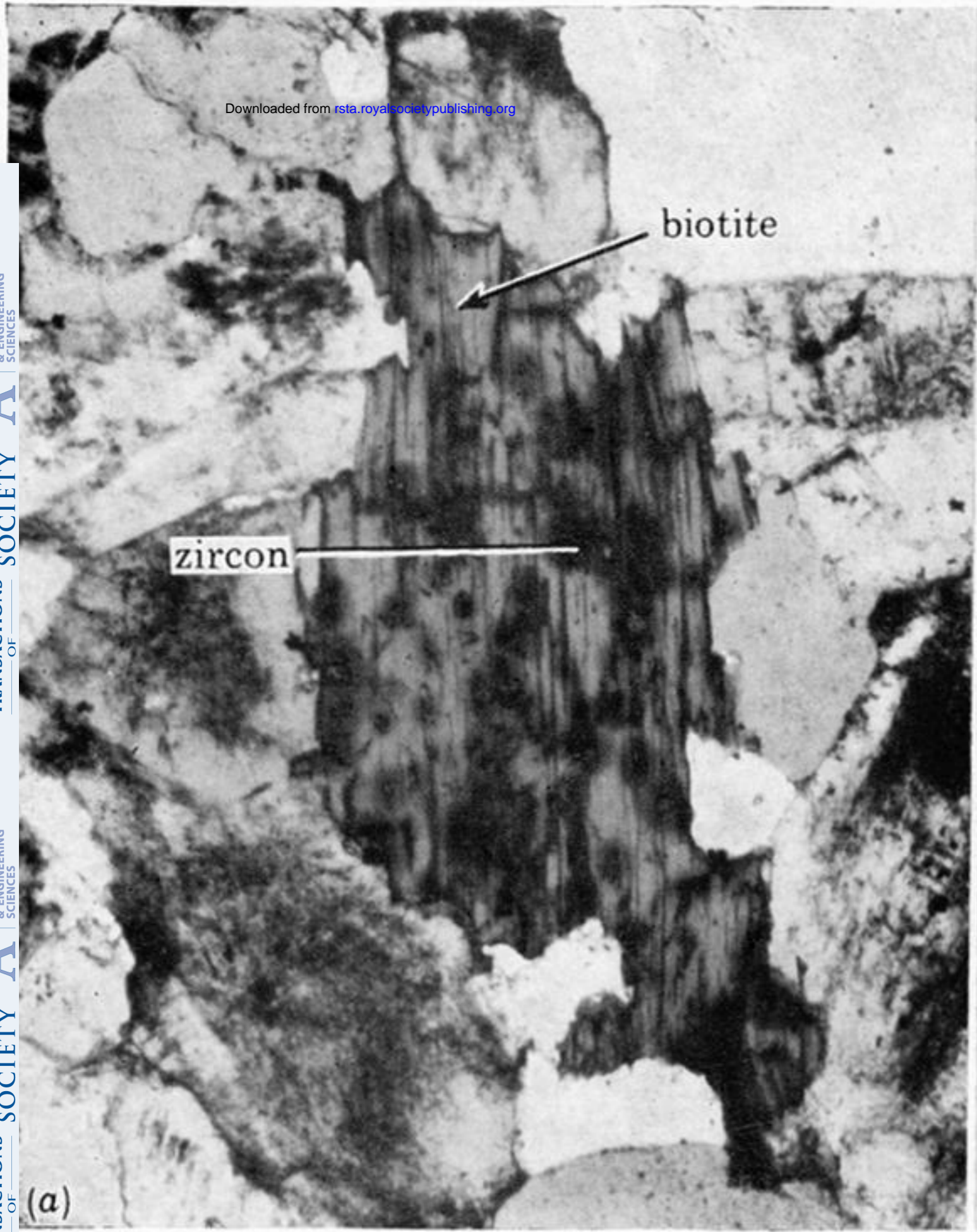


FIGURE 4. Uranium in zircon in primary biotite from Cairngorm. (a) Polished thin section; (b) lexan print showing fission track distribution.



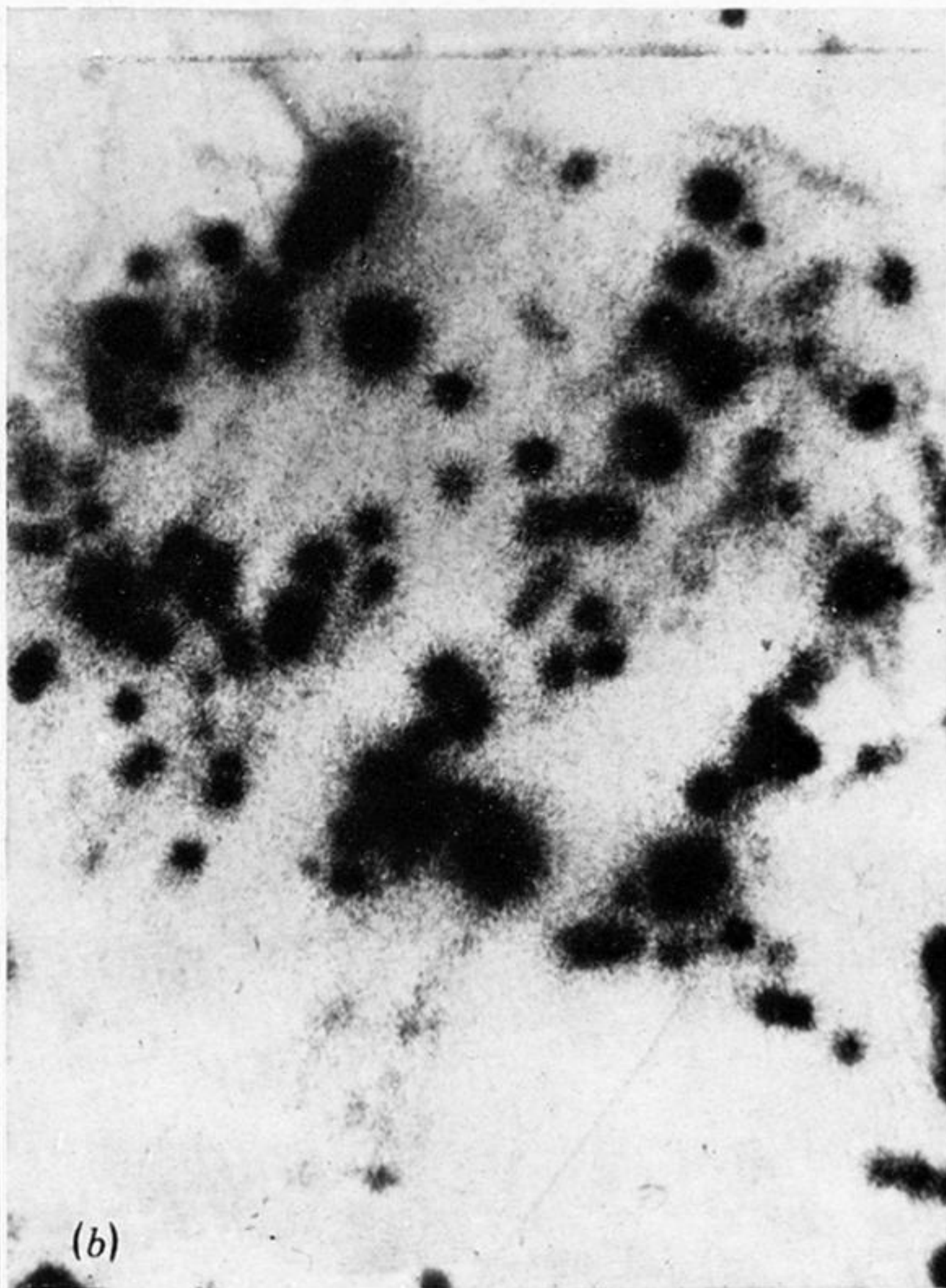
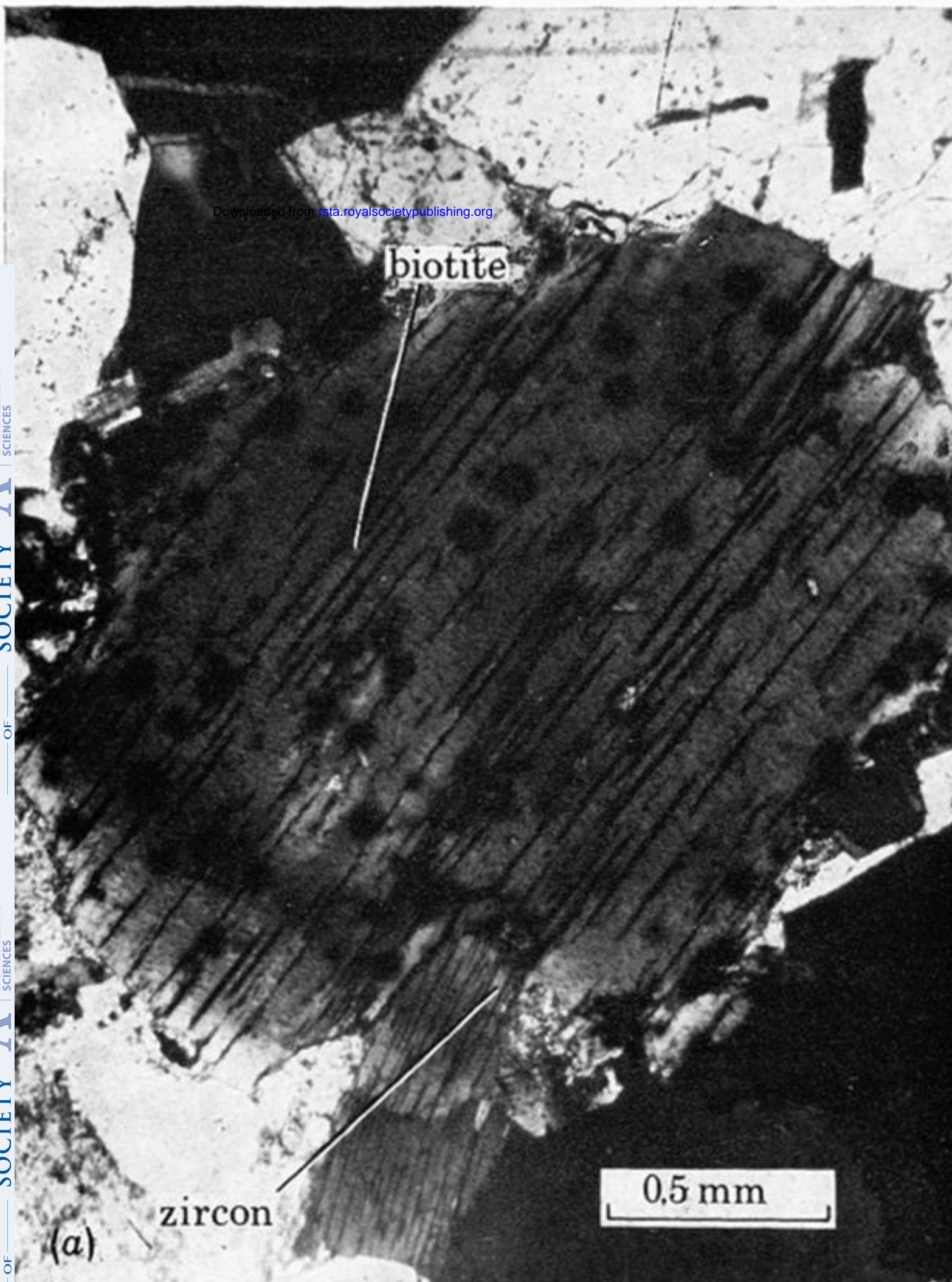


FIGURE 7. Uranium in zircon in primary biotite from Southwest England (Dartmoor). (a) Polished thin section; (b) lexan print showing fission track distribution.