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U, Th, and K distribution in a differentiated charnockite-granite intrusion and associated rocks from SW Sweden

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ABSTRACT. The concentrations of U, Th, and K in a charnockite-alkali granite intrusive complex from SW Sweden and associated, partly charnockitized country rocks are presented. The charnockitic components have constant low U and Th levels throughout the compositional range with a mean Th/U ratio of 1.2 to 1.5. The transition from charnockite to granite is marked by a sharp increase in Th and U concentrations with mean Th/U ratios increasing to 3.4-4.4. However, the K concentration increases relatively smoothly with increasing acidity through the compositional range studied. Late shearing of the alkali granite led to significant Th depletion with little change in the U and K concentrations. Charnockitization of the country rock granite gneisses is marked by depletion in Th and K while the U content is almost unchanged.

It is suggested that the behaviour of U and Th reflects the tendency for Th to associate with magma-dissolved water whilst U shows a greater affinity for coexisting free volatiles. In the absence of a significant free volatile phase during crystallization of this charnockite-granite suite, U was incorporated into primary mineral lattice sites whereas Th entered sites from which it could be liberated more easily.

MAGMATIC and metamorphic charnockites outcrop in the southern segment of the southwest Swedish gneiss belt and are particularly well exposed in the coastal region around Varberg, 70 km south of Gothenburg (Hubbard, 1975, 1978; Hubbard and Constable, 1980; Hubbard and Whitley, 1978, 1979). Here we report a study of the U and Th distribution in the charnockites and their associated rocks using laboratory gamma-ray spectrometry. The region is particularly suitable for an investigation of the mechanisms of crustal depletion of the radioelements by charnockite-granite plutonism and *in situ* charnockitization. The magmatic system allows the stages in charnockite-granite differentiation to be identified and the incomplete charnockitization of the granite gneisses allows comparison of parent and product.

The trend in medium-high-pressure granulite-facies metamorphic terrains is towards low Th, U, and K concentrations with high Th/U ratios (e.g. Heier and Thoresen, 1971; Sheraton *et al.*, 1973). Low concentrations of U and Th also seem typical of charnockitic rocks in comparison with similar non-charnockitic suites (Killeen and Heier, 1975*a*; Smithson and Heier, 1971; Smithson and Decker, 1973; Heier and Thoresen, 1971). These workers have suggested that the parameters which control the U and Th distribution during charnockitization are intimately related to those which determine the mineralogical distinctions between the charnockitic and non-charnockitic phases. Complexing with volatile species is therefore thought to be the method by which radioelements diffuse during differentiation. Within a normal (non-charnockitic) intermediate to acid igneous differentiation series, U and Th generally increase progressively with acidity (see Rogers and Adams, in Wederpohl, 1969). However the Th/U ratio is reported to increase, remain approximately constant or even decrease in such systems. U is generally considered more volatile and soluble than Th (and therefore more mobile); any increase in the Th/U ratio is ascribed to removal of U into later-stage volatile phases (Rogers and Ragland 1961).

Regional setting

The intermediate-acid intrusive complexes with charnockitic and non-charnockitic members in SW Sweden are associated with repeated basic to ultrabasic intrusions and followed conduits established by the earliest pulses of basic magma. Granulite facies over-prints the regional

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amphibolite-facies gneisses in some areas and the dominant granitic gneisses were partially transformed to charnockites *in situ*. The belts of rocks of mixed lithology, characterized by a high proportion of conformable mafic rocks occurring within the granite gneisses, were more completely reconstituted to produce a suite of two-pyroxene granulites.

The assemblage of basic intrusives, charnockite-granite differentiated plutons, and reconstituted gneisses is essentially anorogenic. The charnockitic components give an Rb–Sr isochron age of 1450 ± 53 Ma which has been suggested as the date of intrusion (Welin and Gorbatschev, 1978). The Varberg Charnockite–Granite Association (CGA) (Hubbard, 1975) has been interpreted as an anatectic emulsion, derived from crustal rocks as a consequence of the high heat flow and volatile influx that followed mantle-derived basic magma emplacement. The exposed rocks of the CGA are considered in terms of four groups. The marginal, medium-grained, sparsely-porphyritic, charnockite of quartz-diorite to granodiorite composition is known as the Varberg Charnockite (VC). It has disruptive, intrusive contacts with the country rocks and contains many xenoliths. The Apelviken–Getterön Charnockite (AGC) is compound unit with a matrix of similar composition and fabric to the VC, but with numerous schlieren of coarse charnockite and sub-charnockite which resemble the host rocks. The AGC also contains discrete, internally-differentiated aggregates with sharp contacts and an inverted drop form. The bulbous heads of these drops are alkali-granite which grades down into subordinate charnockitic 'tails' of granodiorite to adamellite. To the north, the AGC gives way to the Trönningenäs Charnockite (TC), a coarse-grained, charnockitic granodiorite; in turn, this passes gradationally through sub-charnockitic adamellite to the microcline–biotite–hornblende Torpa Granite (TG). The form and compositional range of the TC–TG doublet is similar to the discrete, internally-differentiated aggregates found within the AGC but on a much larger scale. The TG is a large sheet intrusion at the structural boundary between the northern and southern segments of the gneissic country rocks. Some detached granite sheets (TGS), of similar petrology and geochemistry to the TG, occur at higher levels in the northern segment gneisses. It is not known whether these are directly associated with the Varberg complex or are derived from other separate differentiated intrusions of similar type. The distribution of the CGA components, the TG sheets and the granulite overprinted zones of the southern gneisses are indicated in fig. 1.

Evolution of the charnockite–granite association

The petrological characters and field relationships of the Varberg CGA suggest that the complex represents an incomplete differentiation cycle in which the composition and availability of a volative phase played an important role. During ascent the CGA magma flowed as an emulsion in which the melt/volatile component was concentrated in the central parts of the conduit. In this central zone of enhanced mobility, (AGC) magma globules separated from the magma–crystal–volatile emulsion and amalgamated to form discrete magma systems which developed independently. Within the magma bodies there was some major-element differentiation, but the main redistribution was of water and associated trace elements, including U and Th. These accumulated at the increasingly buoyant and less viscous tops of the ascending drops. When fully developed such processes could lead to complete separation of a volatile and LIL element-enriched granite from a depleted charnockitic residue. In the CGA system this separation was prevented by crystallization, probably as the result of an abrupt change in the flow of magma from vertical to lateral at a major structural boundary in the country rocks. A similar change in intrusion mode had been established by the preceding basic and ultrabasic intrusions.

The field evidence suggests that a separate fluid phase coexisted with the magma drops. Streaming of this fluid probably produced the sub-pegmatitic, sub-charnockitic schlieren of the AGC as may be predicted for granitic magma systems with significant CO_2 (Holloway 1976). Indeed, the fluid inclusions of the quartz grains of the CGA are dominated by CO_2 (Touret, pers. comm.). In such a system fractionation of the available water will occur between the fluid and the magma. Similarly, trace elements with strong affinities for the volatile phases will fractionate. The magma segregations subsequently evolved as sub-systems closed to volatiles, as is demonstrated by the sharp contacts between hydrous granite and the enclosing anhydrous charnockite observed in the smaller aggregations of the AGC. Also there is no evidence of extensive pegmatitic developments or quartz veining associated with the intrusion of the TG (or the detached TGS). Thus, during crystallization, the magma-dissolved water was largely held in the hydrous minerals and the final rocks to crystallize were sparsely developed aplites. Thus the Th/U ratio in the granite probably corresponds closely with that at fractionation, with little, if any, late-stage removal of U or Th. However, the CO_2–H_2O fluid phase which separated from

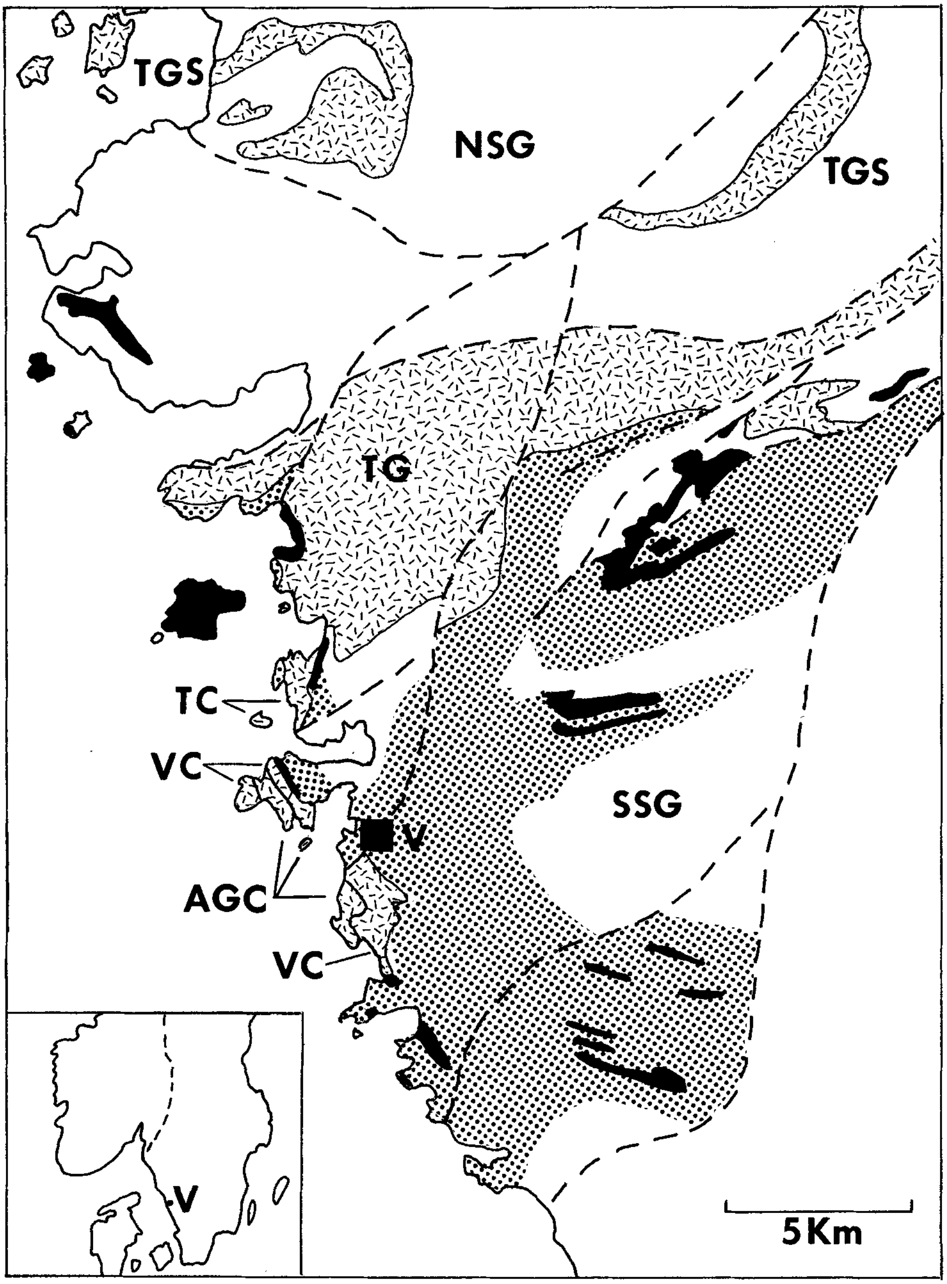

FIG. 1. Map showing the geology of the Varberg area. V = Varberg. *Solid shading*: Basic-ultrabasic suite. *Stippled area*: CGA and Torpa Granite sheets; VC = Varberg Charnockite, AGC = Apelviken-Getterön Charnockite, TC = Trönningenäs Charnockite, TG = Torpa Granite, TGS = Torpa Granite Sheets. *Unshaded land area*: Amphibolite-facies gneisses; NSG = Northern Segment Gneisses, SSG = Southern Segment Gneisses. *Dotted area*: Partial or complete granulite facies overprint on the amphibolite-facies gneisses.

the granite magma may have partially escaped to higher levels carrying a proportion of the trace elements.

TABLE I. *Th, U, K, and SiO_2 concentrations and Th/U, $(Th/K) \times 10^4$, and $(U/K) \times 10^4$ ratios for the rock units of the CGA*

	Th ppm	U ppm	K%	SiO_2%	Th/U	(Th/K) $\times 10^4$	(U/K) $\times 10^4$
VC							
HV 75/47	0.7	0.6	2.87	54.85	1.2	0.2	0.2
HV 75/20	0.7	0.8	3.78	60.20	0.9	0.2	0.2
HV 79/13	0.9	0.6	2.71	55.92	1.5	0.3	0.2
HV 79/14	2.0	1.5	2.67	55.42	1.4	0.7	0.6
Mean	1.1	0.9	3.01	56.60	1.2	0.4	0.3
AGC							
HV 75/29	2.4	2.2	3.57	56.88	1.1	0.7	0.6
HV 75/48	2.3	2.7	3.93	63.96	0.8	0.6	0.7
HV 75/45	1.5	1.1	3.87	65.52	1.4	0.4	0.3
HV 165	1.2	0.2	3.72	65.26	5.1	0.3	0.1
HV 75/49	2.7	1.9	3.73	61.14	1.4	0.7	0.5
HV 75/46	0.9	0.5	3.61	62.54	1.6	0.2	0.1
HV 75/25	1.4	1.0	3.96	63.21	1.4	0.4	0.3
HV 75/26	1.0	1.8	3.55	59.89	0.5	0.3	0.5
HV 75/27	2.5	4.3	4.08	62.49	0.6	0.6	1.1
Mean	1.8	1.7	3.78	62.32	1.5	0.5	0.5
TC							
HV 75/40	1.5	1.2	3.77	64.07	1.3	0.4	0.3
HV 75/44	1.6	2.5	4.08	63.47	0.6	0.4	0.6
HV 75/41	2.1	2.0	3.76	63.93	1.1	0.6	0.5
HV 75/42	1.6	1.7	3.91	66.82	1.0	0.4	0.4
HV 79/6	1.8	1.2	3.64	66.08	1.5	0.5	0.3
HV 79/5	2.3	1.8	3.54	64.78	1.3	0.6	0.5
HV 75/43	2.7	2.0	4.40	62.48	1.4	0.6	0.5
Mean	1.9	1.8	3.87	64.51	1.2	0.5	0.5
TG							
HV 93	15.2	2.3	5.03	67.70	6.6	2.9	0.5
HV 79/1	13.6	4.4	3.82	68.44	3.1	3.6	1.2
HV 79/7	12.2	4.2	3.93	68.23	2.9	3.1	1.1
HV 79/11	3.6	1.1	3.33	62.88	3.3	1.1	0.3
JC 77/51	19.9	4.0	3.97	71.14	5.0	5.0	1.0
Mean*	15.2	3.7	4.19	68.88	4.4	3.7	1.0
TGS							
JC 77/17	27.0	6.5	4.83	73.67	4.2	5.6	1.3
JC 77/23	24.5	7.7	4.77	71.68	3.2	5.1	1.6
JC 77/40	15.5	4.1	4.11	66.31	3.8	3.8	1.0
JC 77/41	13.4	6.2	4.09	70.19	2.2	3.3	1.5
Mean	20.1	6.1	4.45	70.46	3.4	4.4	1.3

VC = Varberg Charnockite; AGC = Apelviken-Getterön Charnockite; TC = Trönningenäs Charnockite; TG = Torpa Granite; TGS = Torpa Granite Sheets.

* Mean excluding sample HV 79/11.

Analytical methods

U and Th were determined by laboratory gamma-ray spectrometry following procedures described by Adams and Gasparini (1970), Raade (1973), and Killeen and Heier (1975*d*). A lead-shielded 7.6 × 7.6 cm Na (Tl) detector with two single-channel analysers was used to measure energy intensities at 2.615 Mev (Tl^{208}) for Th and 1.76 Mev (Bi^{214}) for U. Corrections were made for any peak drift that occurred. The radioelement concentration of artificial standards (monazite and uraninite mixed with dunite) were determined by gamma-ray spectrometry at the University of London Reactor Centre. Sample weights ranged from 780 to 860 g and count times varied from 400 to 999 min. A precision of ±4% for Th and ±7% for U was achieved down to concentrations of 1.5 ppm Th and 0.5 ppm U. INAA check analyses at the Scottish Universities Research and Reactor Centre, East Kilbride, indicated viable accuracy for both elements. K and SiO_2 were determined by standard XRF procedures.

Results and discussion

U and Th in magmatic charnockites. In comparison with non-charnockitic granodioritic rocks the Varberg charnockites show a strong relative depletion in Th and, to a lesser extent, U (Table I). Clark *et al.* (1966) gave average values of 9.3 ppm Th and 2.6 ppm U for granodiorites and 8.5 ppm Th and 2.0 ppm U for diorites/quartz diorites. However, the potassium concentrations of the Varberg area charnockites are considerably higher than the averages for granodiorites (2.55%) and quartz diorites (1.18%) presented by Nockolds (1954). Both Th and U increase only very slightly through the range of the charnockitic rocks although there is an abrupt increase in the Th and U concentrations of granitic end-members. In contrast, K exhibits a much more uniform increase throughout the whole compositional range of the CGA (fig. 2). The charnockitic units have mean Th/U ratios of 1.2 to 1.5 while the granite units have mean Th/U ratios of 3.4 and 4.4.

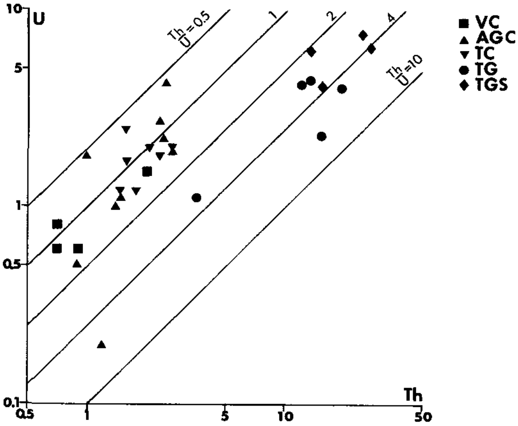

FIG. 2. Th (ppm) *v.* U (ppm) plot for the samples from the CGA.

In the charnockitic components of the complex the Th and U concentrations and the Th/U, Th/K, and U/K ratios are all constant and uniformly low, although the VC and AGC units are thought to be in part recrystallized restite while the TC rocks are interpreted as having crystallized from volatile-depleted magma. The data suggest that the levels of U and Th in plutonic charnockites are a function of their mineralogy. The U and Th values are low in comparison with rocks of similar bulk composition but with non-charnockitic mineralogies, e.g. the Idaho Batholith (Larsen and Gottfried, 1960). The low values in the plutonic charnockites suggest efficient scavenging of trace elements by volatiles during the differentiation process. There is no evidence for U-Th enrichment in the fluid-modified zones of the AGC which show the results of enhanced water availability only in the crystallization of hydrous mafic minerals and the coarsened fabric. In contrast, there is a marked concentration of U and Th (and other LIL elements) in the magma segregations; in particular, their volatile-enriched zones. Th seems particularly concentrated in the magma during fractionation.

U and Th in the alkali granites. Heier and Rogers (1963) presented average values of 17.36 ppm Th with 3.47% K and 4.75 ppm U with 3.79% K for granitic rocks while Clark *et al.* (1966) gave average values of 20.0 ppm Th, 4.7 ppm U, and 4.26% K for alkali granites. The U and Th concentrations for the TG and TGS compare quite closely with the average values for granite and alkali granite respectively but their K concentrations are higher. Preferential concentration of Th with the magma volatiles again is suggested by comparison with the levels retained in the charnockites. In the absence of data on the original levels of U and Th in the parent magma or the proportion of these elements associated with the CO_2-H_2O fluid phase no calculation is possible. But the retention of relatively high levels of U in the TG and TGS agrees with the field evidence of volatile retention in crystallizing magma aggregates. (One sample of the TG unit (HV 79/11) gives anomalously low radioelement and SiO_2 results compared with the rest of the unit: the reasons for this are uncertain.)

U and Th in the country rocks. Average values of radioelements for the Varberg country rocks are presented in Table II. The U values are relatively high but transformation of the granite gneisses to charnockite was marked by depletions in Th, K, and SiO_2 and a reduction in the Th/U ratio.

Taken together with the reduced K and Si levels in charnockite, the Th and U data suggest that a process of partial melting may have been involved in charnockitization. There is, however, no field evidence of anatexis. Moreover, chondrite-normalized *REE* plots (Hubbard and Whitley, 1979) show parallel patterns with no evidence of the fractionation which would be expected if partial melting had been involved. This evidence therefore suggests a weak hydrothermal mechanism of depletion rather than anatexis. The high mobility of Th and Rb (which is also depleted in this complex—Hubbard, 1978) as compared with the other trace elements considered is probably related to the sites in which they were held before reworking. The release of K, Si, and Rb may indicate the instability of K-feldspar, biotite mica, and quartz. The Th in the granite gneisses may be largely held in dislocations in the K-feldspars and biotite whilst a large proportion of the U and the *REE* are contained, and retained during the charnockitization, in more refractory minerals.

U and Th during deformation. Locally, the Torpa Granite has been deformed and recrystallized during post-crystallization 'hot' shearing. Table II shows that while there has been a substantial loss of Th as a result of the shearing, the U content is unchanged with insignificant loss of SiO_2 and K. The shearing caused recrystallization of the quartzofeldspathic components and it is likely that there was a considerable flow of water through the shear zones. These data provide further evidence that much of the Th was weakly bound in the granite by adsorption and/or inclusions in minerals whilst a greater proportion of the U was held in mineral sites which persisted through shearing and recrystallization. Thus, although the depletion process was hydrothermal flushing, Th was mobilized in preference to U.

Comparative mobility of U and Th. In both of the types of metamorphic transformation studied, Th appears to have been more mobile than U. This is believed to reflect the differing nature and stabilities of U and Th sites in the affected mineral assemblages and the incompleteness of the mineral reconstitution achieved in the metamorphism. However, the data from the volatile-closed CGA

TABLE II. *Mean Th, U, K, and SiO_2 concentrations and Th/U, (Th/K)×10^4, and (U/K)×10^4 ratios for sheared and unsheared Torpa Granite and some of the CGA country rocks*

Th ppm	U ppm	K%	SiO_2%	Th/U	(Th/K) ×10^4	(U/K) ×10^4
Torpa Granite (n = 4)						
15.2	3.7	4.19	68.88	4.4	3.7	1.0
Sheared Torpa Granite (n = 3)						
5.8	3.5	4.06	66.24	1.8	1.5	0.9
Granite Gneiss (n = 4)						
6.7	1.4	4.49	75.60	4.8	1.6	0.3
Charnockitic Granite Gneiss (n = 8)						
3.8	2.0	3.44	67.49	2.0	1.2	0.6
Granulite (n = 3)						
2.6	1.4	2.49	65.80	2.1	1.1	0.7

developments also suggest that there can be differential accumulation of Th as compared with U in siliceous melts without there having been volatile-borne U loss. Thus Th, which is markedly less soluble than U in hydrothermal waters, shows an apparent preference for magma-dissolved water. In a hydrous silicate melt, polymerization of the anionic framework is inhibited by the weakening effect of the hydroxyl ions on the Si-O (and Al-O) bonds. Thus it is possible that the tetravalent Th might associate with the anionic magma component by the formation of transitory complexes by coordination with oxygen. The control would be a high hydroxyl ion concentration and therefore the Th would migrate and redistribute with the hydroxyl in the magma. On final crystallization the Th would largely be retained by adsorption and inclusion in such late phases as K-feldspar, quartz, and biotite.

Comparisons with other areas. Killeen and Heier (1975 *a*, *b*, *c*, and *d*) have published a series of papers reporting their extensive studies of the radioelement distribution in the granitic rocks of southern Norway. The rocks of this region are commonly linked with those of southwest Sweden in a Sveconorwegian province. Charnockitic rocks associated with non-charnockitic granites occur in the Farsund area of the Norwegian sub-province and were examined by Killeen and Heier (1975*a*). The Farsund charnockite is less depleted in Th than the Varberg rocks with a Th/U ratio of 4 compared with a mean Th/U ratio of 1.2 to 1.5 for the charnockitic members of the CGA. Killeen and Heier noted that the dark Farsundite (charnockite) is the only granitic body with a $(\mathrm{Th/K}) \times 10^4$ ratio less than 1.0 of the twenty-two Precambrian granites they investigated in S. Norway. The same ratio for the Varberg charnockites is 0.2 to 0.7 (mean 0.5). On the basis of the radioelement evidence, Killeen and Heier concluded that the hornblende-granite 'light farsundite' is not comagmatic with the charnockitic 'dark farsundite'. This interpretation seems mainly based on the abruptness of the changes in the radioelement contents and ratios and the drop in K in passing from dark to light farsundites. The evidence presented above from the Varberg CGA suggests this interpretation may be ill-founded.

The granites of the inland belt of the Norwegian complex give a wide range of radioelement concentrations with increases in Th and U apparently related to increases in the extent of granitization. Those rocks granitized in both the Svecofennian and Sveconorwegian orogenies give the highest values (Killeen and Heier, 1975*b*).

The suite of granites in a northwest-trending belt crossing the Permian Oslo Province and including the Bohus granite of SW Sweden show extremely high Th and U concentrations compared with crustal averages. Killeen and Heier (1975*c*) believed that the U values were modified by the effect of the Permian activity. In their discussion of the remobilization of U, Killeen and Heir suggested that a $(\mathrm{U/K}) \times 10^4$ ratio of *c*. 1.2 is a stable ratio and any increase in U which increases this ratio represents U which is easily lost. In the modified rocks of the Varberg area, in which Th was mobilized rather than U, the mean $(\mathrm{U/K}) \times 10^4$ ratios are well below this value (TG = 1.0; sheared TG = 0.9; granite gneiss = 0.3; charnockitized granite gneiss = 0.6). Using the evidence from Varberg of Th mobility, the distribution indicated by the trend surface analyses for the Iddefjord granite, presented by Killeen and Heier, might reflect Th and K (and by association H_2O) enrichment westward rather than U migration eastward.

Conclusions

In charnockite-granite differentiated intrusions, developed during the reworking of metamorphic rocks, U and Th are associated with the volatile-enriched granite differentiates whereas only low concentrations of these elements are retained in the charnockitic residues. The controlling factor in the redistribution is the volatile concentration. In the presence of CO_2 a fluid and a magma phase may segregate with trace elements fractionating between the two. Th is thought to have been associated with magma-dissolved volatiles and with anionic magma polymers: this may account for differences in the behaviour of U and Th in volatile-enriched melts. Much of the Th in the crystalline product of such melts can be released readily when shearing and recrystallization affect the late crystallizing minerals with loosely-held Th. U is therefore less easily released to migrating fluids during metamorphism and this may result in the unusual higher depletion of Th rather than of U during the restricted reworking.

Crustal reworking in the Varberg area is restricted and both magmatic and metamorphic processes have been active. But neither process has run to completion because of the lack of penetrative deformation during the thermal and volatile influx period. Although we cannot be sure that the mechanisms dominant in the localized anorogenic environment of the Varberg area would be those of syntectonic conditions, the restriction on their full development allows comparison of source, intermediate and product rocks without uncertain long-distance extrapolation and interpolation.

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