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DEUTERIC ALTERATION AND
URANIUM MINERALIZATION PROCESSES
IN LEUCOGRANITE INTRUSIONS FROM THE
NAMAQUALAND METAMORPHIC COMPLEX

L.J. ROBB and A.E. SCHOCH

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UNIVERSITY OF THE WITWATERSRAND
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ABSTRACT

Studies of the moderate- to high-level, leucogranite intrusions in the Namaqualand Metamorphic Complex (circa 1100 Ma), indicate a link between a pervasive deutereric alteration and the concentration of uranium. Quantitative fission track studies indicate that unusually high contents of uranium are associated with the products of deutereric alteration, suggesting efficient flushing of a uranium-charged fluid phase through the system.

Fluid inclusion data indicate distinct populations of brine, pure water and CO₂. Unmixing of the brine -CO₂ probably occurred at temperatures corresponding to the onset of deutereric alteration, and may have been responsible for the precipitation of uranium from the fluid phase. Fluid entrapment conditions of 0,6-1,2 kb and 170-340°C indicate a prolonged circulation of fluids, during the latter stages of which secondary redistribution of uranium occurred.

Uranium mineralization in the leucogranites is considered to be a function of a closed-system alteration process and the retention of uranium *in situ*, rather than the outgassing of the fluid phase and the precipitation of uranium in extraneous vein, breccia or pegmatite systems.

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I. INTRODUCTION

Exploration for high tonnage - low grade uranium deposits associated with granitoids in the circa 1100 Ma Namaqualand Metamorphic Complex has resulted in the delineation of numerous, late stage, leucogranite intrusions characterized by anomalous uranium concentrations and visible secondary mineralization at surface. Two of these intrusions, the Manus and Windmill bodies, occurring on the farm Nooitgedacht some 30 km southeast of Springbok (Fig. 1), have been investigated in order to assess the distribution of uranium and the mechanisms of its concentration. An attempt is made here to present an explanation of the mineralization processes in the light of petrographic, geochemical, fission track micro-mapping and fluid inclusion data. Although superficially similar in some respects to the "alaskites" of the Rössing uranium mine, in Namibia, the present study shows the sub-economic Namaqualand leucogranites to be significantly different in terms of the relationships between petrogenesis and uranium mineralization processes.

The Namaqualand Metamorphic Complex in the Springbok region comprises a sequence of highly deformed metavolcano-sedimentary units which have been intruded by syn-tectonic, potassic granitoids. These assemblages have been subjected to a low-pressure granulite grade metamorphism (Clifford *et al.*, 1975; Clifford *et al.*, 1981). Late to post-tectonic granitoids, which include the Concordia granite and the leucogranites of the Kweekfontein suite, were subsequently emplaced into a variety of the pre-existing rock-types. Magmatism in the region was terminated by the intrusion of small norite, diorite and anorthosite bodies with which is associated the copper mineralization for which the region is otherwise well-known.

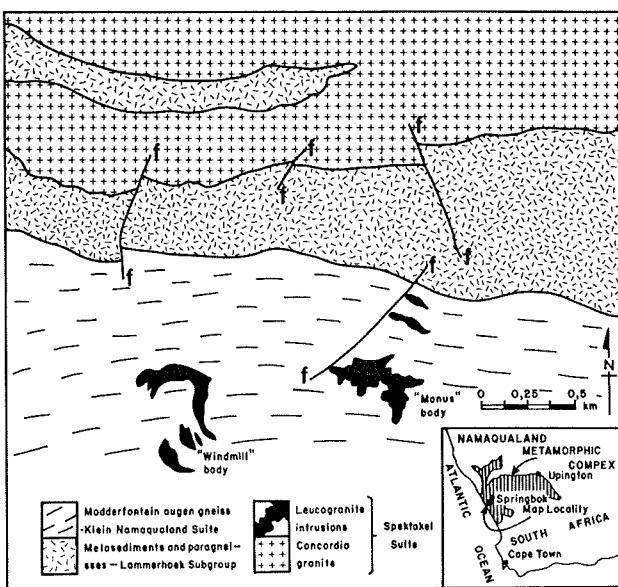


Figure 1 : Locality and generalized geological setting of the leucogranite intrusions.

The area studied here is underlain by the Modderfontein augen gneiss and a metavolcano-sedimentary package known as the Lammerhoek sub-group (Fig. 1). The above units are intruded in the study area by the extensive, shallow-dipping Concordia granite sheet. Geographically distinct from the Concordia granite and intrusive into the Modderfontein augen gneiss, occur the two leucogranite intrusions studied here. These bodies form small, irregular, often dyke-like intrusions which truncate the prominent, dominantly linear, east-west trending fabric in the augen gneiss unit (Fig. 2). The contacts are sharp and abrupt, although partially digested xenoliths of the host rock do occur within the leucogranite.

The two leucogranite intrusions are tentatively correlated with the numerous other late-stage, post-tectonic, leucogranites in the region, known as the Kweekfontein suite. There is a possibility that the Kweekfontein suite is genetically linked (at least with respect to the nature of the source rock) to the Concordia granite as the latter is chemically very similar and, in places, also characterized by anomalous uranium contents. The nature, and probably also the timing, of emplacement of the two granitic suites are, however, quite different and it is also apparent from petrological considerations that the Kweekfontein leucogranites do not represent a simple, late-stage, differentiate of the Concordia granite (Robb, 1985).

II. PETROGRAPHIC AND GEOCHEMICAL CHARACTERISTICS

A. Petrography

The leucogranites of the Windmill and Manus bodies are typically massive, medium- to coarse-grained and hypidiomorphic; associated pegmatitic and aplitic phases are rare although occasional coarser-grained miarolite-like patches may occur. Quartz, microcline and pervasively sericitized plagioclase form the dominant mineral constituents with occasional granophytic intergrowths occurring. The only ferromagnesian mineral is an altered biotite which, in most cases, has reverted to an assemblage comprising chlorite, epidote, magnetite and ilmenite. The abundance of ferromagnesium minerals is generally low (<3%) although certain

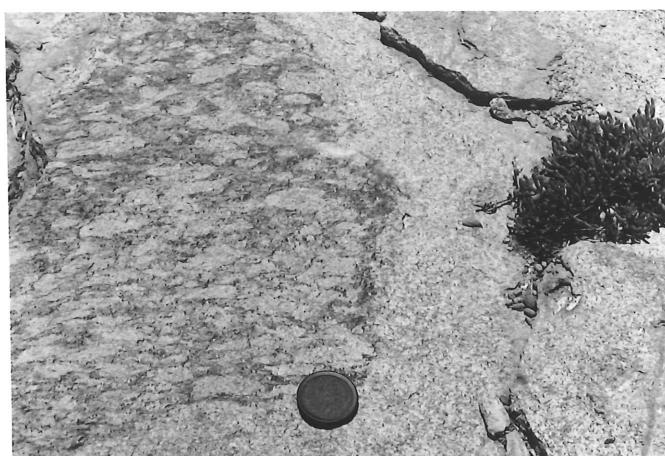


Figure 2 : Abrupt contact between the "Windmill" leucogranite body and the coarse-grained, foliated Modderfontein augen gneiss.

samples contain zones of enhanced biotite-chlorite-epidote-magnetite concentration. The principal primary opaque oxide in the leucogranite is magnetite which is usually somewhat martitized along grain boundaries and microfractures. Ilmenite occurs as exsolution laths within magnetite or as irregular granular exsolution products associated with a sphene-like phase or anatase. Secondary magnetite overgrowths, probably related to deuterian alteration of the leucogranites, are occasionally seen rimming the granular ilmenite and its associated alteration product.

The occurrence of accessory mineral phases varies markedly from one sample to another. Certain samples are dominated by zircon and, to a lesser extent, apatite, whereas others contain numerous small grains of allanite as well as other hydrated silico-phosphates of calcium, thorium, uranium and the light rare-earth elements (i.e. cheralite, britholite and ningyoite). Another minor mineral phase, detected in only a few samples, is purple fluorite, which is invariably seen intimately associated with chlorite and epidote. The presence of this mineral, which probably formed on the release of fluorine during alteration of biotite to chlorite (Bailey, 1977), implies a fairly shallow level of leucogranite emplacement. Although depth of intrusion cannot be quantified by the presence of fluorite, its association with differentiated granite phases, and its precipitation from vapour phases in the roof zones and margins of granite bodies, argue in favour of moderate- to high-level emplacement.

Accessory minerals in which uranium and thorium occur as major stoichiometric constituents are rare in the Windmill and Manus bodies. Occasional uraninite and uranothorite grains were reported in an unpublished report made to an interested exploration company, but positive confirmation of the existence of these phases was not obtained in the present study. Very small uranium-thorium-silicate minerals were detected with the microprobe, but these are insignificant in comparison to the accessory mineral suite described above. In weathered, near-surface samples a yellow, secondary uranium mineral is commonly observed and has been identified as beta-uranophane (Robb, 1985; Robb *et al.*, 1985).

B. Chemistry

A large number of leucogranitic samples, both from the surface and from borehole core, have been analysed for major and trace elements. A detailed account of the geochemical and petrological aspects pertaining to this data is presented in previous publications (Robb, 1985; Robb *et al.*, 1985). For the purposes of the present study, a representative subset of this data is presented in Table I below.

The leucogranites are characterized by high SiO_2 contents, typically in the range 73-75%, low $\text{Fe}_{2}\text{O}_3 + \text{MgO}$ contents generally less than 2%, and high K_2O contents generally in excess of 5.5% (Table I). $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios are almost invariably in excess of 1.5; the leucogranites are not peraluminous and generally have $\text{Al}/(\text{Na}+\text{K}+\text{Ca}/2)$ ratios of less than 1.1. They have relatively high Rb abundances (200-300 ppm), low Sr contents (typically less than 125 ppm) and concomitantly high Rb/Sr ratios of between 2-3. K/Rb ratios are low (generally less than 200) but samples do not relate to Shaw's "pegmatite-hydrothermal trend" (Shaw, 1968).

Uranium and thorium contents, and the Th/U ratios, are very variable and differ by an order of magnitude from sample to sample (Table I). Typical, fresh leucogranite samples have U contents in the range 35-150 ppm although certain samples characterized by secondary enrichment have abundances of several hundred ppm (Robb *et al.*, 1985). Thorium contents tend to fall in the same broad range as that for uranium, although those samples affected by secondary enrichment of uranium do not exhibit similar enrichments of Th. On average, the Th/U ratio approaches unity, but nevertheless varies considerably from sample to sample (Table I).

III. ALTERATION CHARACTERISTICS

A. Definition of Terms

In view of the loose terminology often applied to late-magmatic and sub-solidus alteration processes in the literature, the relevant terms used here are defined. The alteration processes characteristic of the leucogranites are envisaged as occurring during the hydrothermal stage, namely "That stage in the cooling of a magma during which the residual fluid is strongly enriched in water" (Glossary of Geology, 1980). More specifically, however, the alteration is viewed as essentially autometasomatic and, hence, *deuterian* in nature. The latter term refers specifically to reactions between primary magmatic minerals and water-rich solutions that separated from the same body of magma, and does not apply to alteration of a secondary nature which may be related to solutions of extraneous derivation (Glossary of Geology, 1957).

TABLE I

Chemical Analyses of Leucogranites and Surrounding Rocks

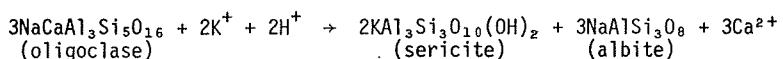
	"Windmill" body				"Manus" body		Borehole core	Augen gneiss	host-rock	
wt.%	W1	W3	W9	M1	M2a	M4	M2/3(3)	W1/2(2)	W6	W1/2(2a)
SiO ₂	74,47	74,20	72,75	73,50	68,19	75,01	74,60	73,44	73,73	72,02
TiO ₂	0,12	0,10	0,24	0,22	0,48	0,11	0,11	0,20	0,40	0,42
Al ₂ O ₃	13,49	13,56	13,44	13,93	15,04	13,58	13,63	13,52	12,60	13,01
Fe ₂ O ₃	1,08	0,97	2,41	1,83	3,34	1,24	1,17	1,84	2,76	3,12
MnO	0,01	0,03	0,03	0,03	0,11	0,02	0,04	0,03	0,04	0,08
MgO	0,30	0,10	0,20	0,20	1,00	0,20	0,30	0,30	0,70	0,70
CaO	0,64	1,18	1,51	1,21	1,40	1,14	1,08	1,34	1,22	1,36
Na ₂ O	2,30	2,34	1,58	2,66	2,61	2,58	2,56	2,54	2,19	2,09
K ₂ O	6,58	5,90	5,14	5,52	6,12	5,59	5,70	5,63	5,58	6,12
P ₂ O ₅	0,01	0,22	0,05	0,03	0,20	0,02	0,03	0,03	0,11	0,09
L.O.I.	0,38	0,67	0,62	0,56	0,93	0,53	0,39	0,61	0,63	0,73
TOTALS	99,38	99,27	98,97	99,69	99,45	100,02	99,61	99,48	99,86	99,74
ppm										
Rb	288	238	214	238	273	218	282	244	306	280
Sr	112	154	110	89	213	83	78	120	104	86
Ba	354	337	282	281	1766	404	257	267	579	604
Y	10	23	22	17	9	68	2	17	47	45
Zr	80	638	61	173	506	5	44	292	194	208
U	57	152	231	98	235	54	42	373	<10	<10
Th	43	95	113	164	<10	26	18	225	27	56

Further clarification is required with respect to alteration processes which are essentially isochemical and those in which open-system behaviour occurs. By definition, deuteritic alteration, which affects only the magmatic system from which the fluids are derived, is a process which is likely to remain essentially isochemical, particularly at higher pressures and temperatures. The process of propylitization (i.e. the alteration of biotite or hornblende to chlorite + epidote \pm calcite \pm magnetite), for example, has been shown to be an isochemical, autohydralional process that occurs in the temperature range 550–750°C (Ivanov, 1979) and, therefore, is in accordance with the definition of deuteritic alteration. In contrast, many other forms of hydrothermal alteration (e.g. silicification, sericitization, kaolinitization, alunitization, etc.) involve the migration of elements in an open system and may occur at lower temperatures than those mentioned above. Many of these processes equally affect the country rocks into which the magmatic body was emplaced and, additionally, may involve fluids of extraneous origin, either together with, or separate from, the magmatic fluids.

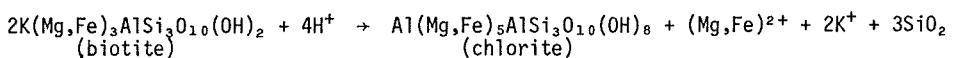
B. The Case for Deuteritic Alteration in the Leucogranites

The Windmill and Manus leucogranites consist essentially of an assemblage comprising microcline-sericite-quartz-chlorite-epidote-magnetite. The alteration processes that have resulted in this assemblage are:-

(i) sericitization of plagioclase -



(ii) propylitization of biotite -



Both reactions require the action of hydrogen ions on primary magmatic minerals. The above reactions also suggest a mutual interdependence in that K⁺ released during the breakdown of biotite is made available for sericite formation, whilst the release of Ca²⁺ during sericitization, and Fe²⁺ and Mg²⁺ during chloritization, facilitates the formation of epidote. Consequently, the alteration reactions indicate that processes could certainly have been autohydralional and, to a large extent, isochemical too.

The nature of the alteration processes in the Namaqualand leucogranites can also be assessed in terms of chemico-mineralogical diagrams (La Roche, 1964). In Fig. 3, the leucogranites are seen to occupy a restricted compositional field with little or no evidence of a trend towards the end-member compositions of any of the major minerals in the rocks. This trait militates against an open-system metasomatic overprint such as silicification, albitization, or K-feldspathization. By contrast, the compositional field exhibited by the so-called "alaskites" of the Rössing uranium deposit is considerably broader and accords with descriptions of albite and K-feldspar alteration in these rocks (Cuney, 1980). The Rössing granitoids also have slightly lower ferromagnesian contents than do the Namaqualand leucogranites (Fig. 3).

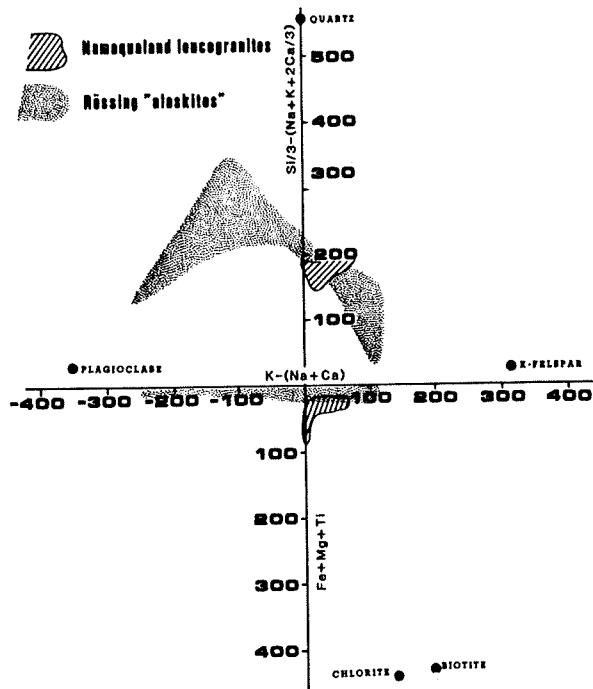


Figure 3 : Chemico-mineralogical plots (La Roche, 1964) showing the fields of the Namaqualand leucogranites (Robb, 1985) and Rössing alaskites (Cuney, 1980) in relation to actual compositions of major rock-forming minerals in the leucogranites (Robb, 1985).

Curves representing the upper stability limits of the assemblage chlorite + muscovite + quartz and also univariant curves for epidote and chlorites, are superimposed onto the phase relations for a quartz monzonite with 4 per cent H_2O in Fig. 4. It is apparent that the present alteration assemblage in the leucogranites will only have started forming under near-subsolidus conditions, probably once a separate aqueous phase had exolved from the magma. The heat-generating capacity of the leucogranites will undoubtedly have assured an efficient convective circulation of fluids such that the sericitic and propylitic reactions described above will have rapidly formed under such conditions. It is pertinent to note that if the level of leucogranite emplaced had been shallow (i.e. < 2 kb) significant reaction between primary magmatic minerals and vapour would have occurred already in the sub-liquidus crystallization interval because of the sharp deflection of the water-saturation curve at lower pressures (Fig. 4).

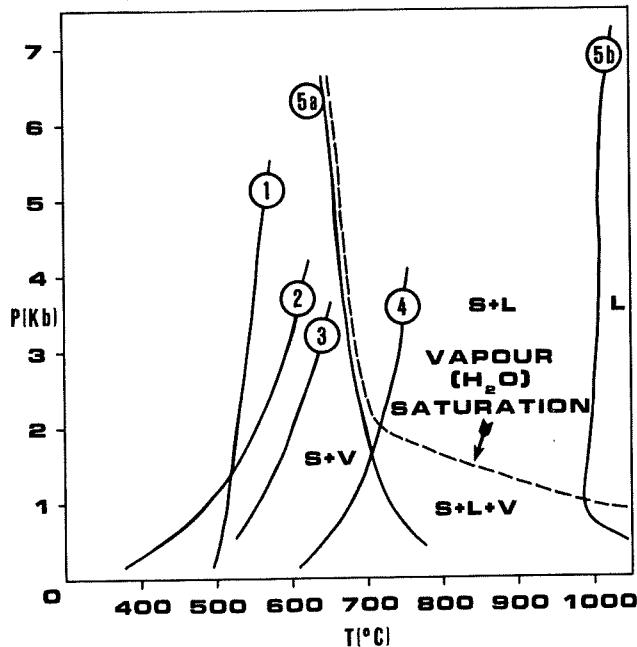


Figure 4 : Phase relationships for quartz monzonite with 4 per cent H_2O (curves 5a,5b; Whitney, 1975) in relation to the upper stability limits of various minerals and mineral assemblages relevant to the leucogranites. Curve 1 - chl + musc + qtz (Hirschberg and Winkler, 1968); curve 2 - epidote; curve 3 - Fe chlorite; curve 4 - Mg chlorite (Creasy, 1966).

In summary the alteration processes in the Namaqualand leucogranites are envisaged being of a deuteritic (*sensu stricto*) nature and, hence, essentially autohydralional and isochemical. The alteration is restricted to the leucogranites themselves and has not affected the country-rock augen gneiss. Alteration processes probably commenced at near-solidus temperatures but continued over a wide cooling interval (see later).

IV. URANIUM DISTRIBUTION

Detailed assessment of the uranium distribution in the leucogranites has been carried out by quantitative fission track micro-mapping. A full account of the techniques used and results obtained from this method is presented elsewhere (Robb *et al.*, 1985).

The leucogranite samples examined revealed three types of uranium distribution :-

(i) in primary sites represented by biotite, magnetite, zircon, allanite, apatite and other monazite-like silico-phosphates of calcium and the light rare earth elements;

(ii) in sites created during deuteritic alteration and represented by chlorite, epidote, ilmenite, magnetite and a sphene-like alteration product; and

(iii) in secondary sites, particularly in weathered surface samples, where beta-uranophane is developed or uranium is associated with ferruginous oxides/hydroxides in micro-cracks and intergranular pore-spaces.

Although a certain amount of uranium is associated with primary magmatic minerals (i.e. particularly allanite, cherlomite, britholite, zircon, apatite etc.), unusually high concentrations of the element are associated with altered biotite, chlorite, epidote and the ore minerals (Robb *et al.*, 1985). Consequently, deuteritic alteration processes are inferred to have played a significant role in the concentration of uranium in these rocks.

Figure 5 shows a photomicrograph of an altered, somewhat hydrated, in part semi-opaque, biotite grain and the corresponding fission track print. In the fresher portions of the biotite grain (where microprobe analyses still approximate biotite stoichiometry; Robb 1985) uranium concentrations are relatively low (i.e. in the range 100 - 300 ppm (Robb *et al.*, 1985) whereas in the darker, altered patches which are characterized by accumulations of Ti and Fe, contents average approximately 1500 ppm. In the leucogranites, however, most of the biotite has reverted to chlorite and epidote and in this assemblage

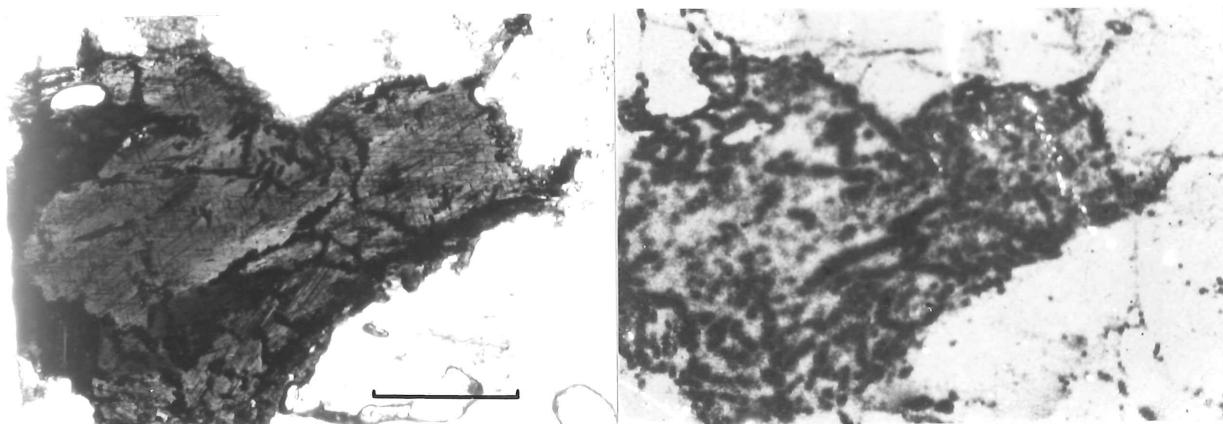


Figure 5 : Photomicrograph of a large grain of altered biotite showing opaque zones of enhanced Ti and Fe concentration, and corresponding fission track print. Transmitted light, polaris uncrossed; bar represents 1 mm; combined thermal, epithermal and fast neutron dose = $1,74 \times 10^{15} n/cm^2$

even higher concentrations of uranium may occur. Figure 6 shows three large flakes of chlorite associated with minor, interdigitated epidote and quartz, and the corresponding fission track print. In this assemblage a significant amount of uranium has been concentrated, with contents of up to 7000 ppm U having been recorded in certain patches (Robb *et al.*, 1985). In places epidote appears to contain higher concentrations of U than in adjacent chlorite, but this relationship is not consistent and the reverse may apply in certain instances. The fission track print demonstrates that U has not merely been adsorbed onto the margins of the chlorite-epidote mass, but is unevenly distributed throughout the various mineral phases. This fact, together with the high U contents associated with the alteration assemblages, points to efficient flushing of a uranium-charged fluid phase through the system and prolonged equilibration between fluid and mineral phases.

As mentioned earlier, a certain amount of uranium is associated with the opaque phases. Primary magnetite contains little uranium, but the exsolution-derived ilmenite and the sphene-like alteration product associated with it may contain localized concentrations of uranium up to 5000 ppm (Robb *et al.*, 1985). Also occasionally observed, however, are aureoles of a later magnetite phase growing around the ilmenite exsolution assemblage. This magnetite contains markedly higher uranium contents than the primary phase and may have formed during the deuteritic alteration process (Robb *et al.*, 1985).

Besides being concentrated into the products of deuteritic alteration, uranium is also distributed along grain boundaries, cracks and in intergranular pore spaces (Fig. 7). A certain amount of this uranium occurs as beta-uranophane and, since this mineral is of supergene origin (Frondel, 1958), it is likely that redistribution of uranium occurred during weathering and other surficial processes. Uranium is also associated with amorphous, Fe-rich compounds (probably limonite, goethite or Fe-Ti-Mn-oxyhydroxides) which coat many of the minerals. Concentration of uranium into these phases may also have occurred under near-surface conditions, as in the case of the beta-uranophane. Alternatively, this redistribution could have occurred during the late stages of hydrothermal alteration, a suggestion which is supported by fluid inclusion data which points to circulation and entrapment of hydrothermal fluids under conditions extending down to circa 170°C and 0,6 kb (see later).

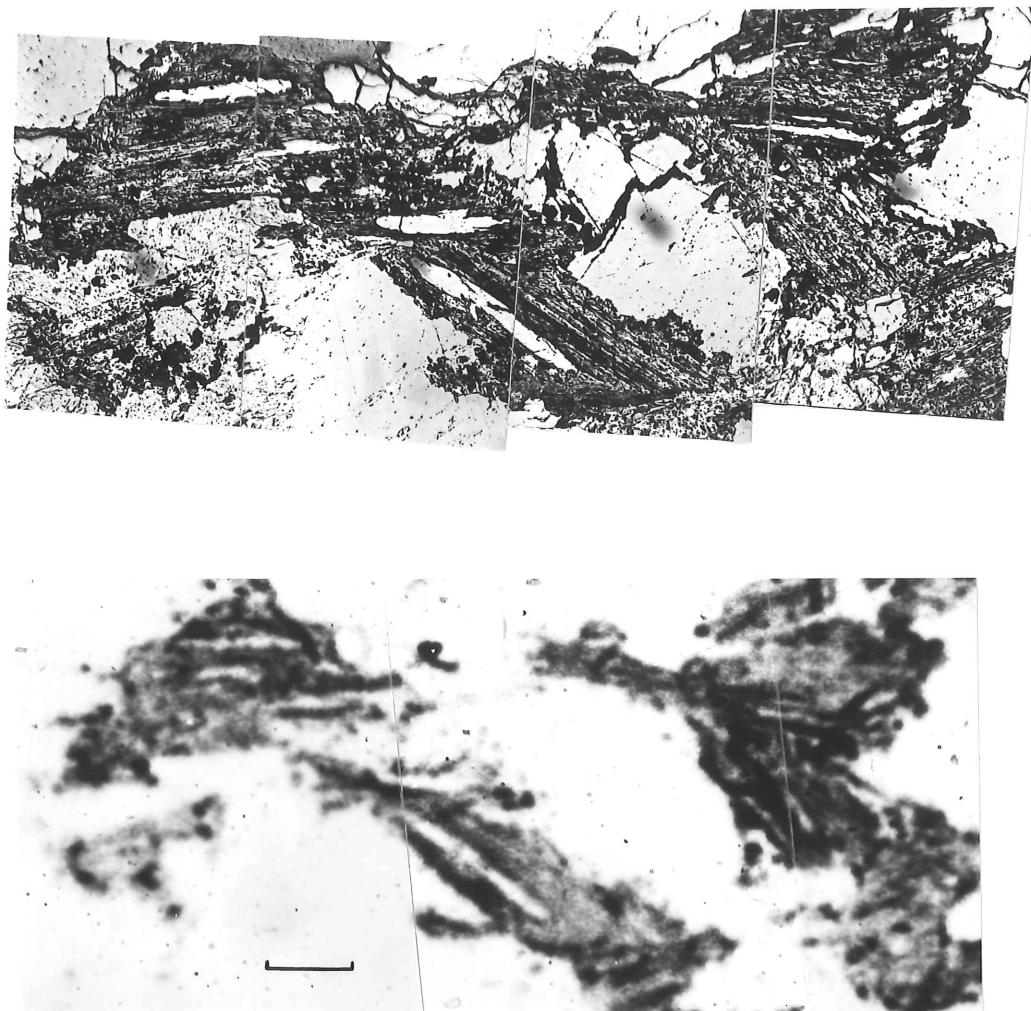


Figure 6 : Photomicrograph of chlorite grains with interdigitated epidote and quartz, and corresponding fission track print. Polars not crossed; bar represents 0,5 mm; fast neutron dose = $3,84 \times 10^{15} \text{ n/cm}^2$.

V. FLUID INCLUSION CHARACTERISTICS

Although microscopic inspection of thin sections indicated that the average size of the fluid inclusions was very small, three samples were nevertheless selected for detailed microthermometric study using two calibrated Chauxmeca stages. One of the samples represents the Manus intrusion with the other two being from the Windmill body. Large scale systematic differences in the fluid inclusion properties of the various samples were not detected and, consequently, the results are presented in accumulated form. All the measurements used in this study were duplicated within acceptable limits, namely differences of less than $0,3^\circ\text{C}$ for $T_m(\text{H}_2\text{O})$ (melting temperature), less than 5°C for $\text{Th}(\text{H}_2\text{O})$ (water homogenization), and below 1°C for $\text{Th}(\text{CO}_2)$.

The fluid inclusions encompass distinct populations of water and carbon dioxide, with an approximate XCO_2 of 0,2. The water exhibits clear bimodal properties (Fig. 8a), with approximately equal amounts of pure water ($T_m = 0,5 \pm 0,5^\circ\text{C}$) and brine ($T_m = -13,5 \pm 0,5^\circ\text{C}$). According to published freezing point curves (Collins, 1979; Potter *et al.*, 1978), the brine contains $17,5 \pm 0,5$ per cent eNaCl (NaCl equivalent). However, the entropy of the brine peak is nearly double that of the other peak, encompassing a range from $-9,5^\circ\text{C}$ to $-13,5^\circ\text{C}$, but is so asymmetrical that the latter temperature must be regarded as the turning point value. The possibility of smaller peaks cannot be regarded as statistically viable even though 289 measurements were made, but it would seem that inclusions with $T_m = -3,5 \pm 0,5^\circ\text{C}$ are better developed in the Manus sample than in the Windmill specimens.

The homogenization spectrum (Fig. 8b) defines two major isochores, namely at $\text{Th} = 165 \pm 15^\circ\text{C}$, representing a density (d) for pure water of $0,91 \pm 0,01 \text{ g/cm}^3$ (Kennedy, 1950; Fisher, 1976), and at $\text{Th} = 220 \pm 10^\circ\text{C}$, interpreted as an isochore with $d = 0,87 \pm 0,01 \text{ g/cm}^3$, for brine with 15 per cent eNaCl (Lemmlein and Klevstov, 1961). The spread of the brine homogenization values is nearly double that for the pure water, as was also reported for the melting temperatures.

The melting points of the carbon dioxide vary from $-56,6 \pm 0,2^\circ\text{C}$ to $-57,1 \pm 0,2^\circ\text{C}$, indicating a high degree of purity, with at most 0 to 2 per cent eCH₄ (Swanenberg, 1979). The homogenization spectrum is fairly complex with a bimodal tendency (Fig. 9), exhibiting a major peak at $\text{Th} = 28,0 \pm 1,5^\circ\text{C}$ which

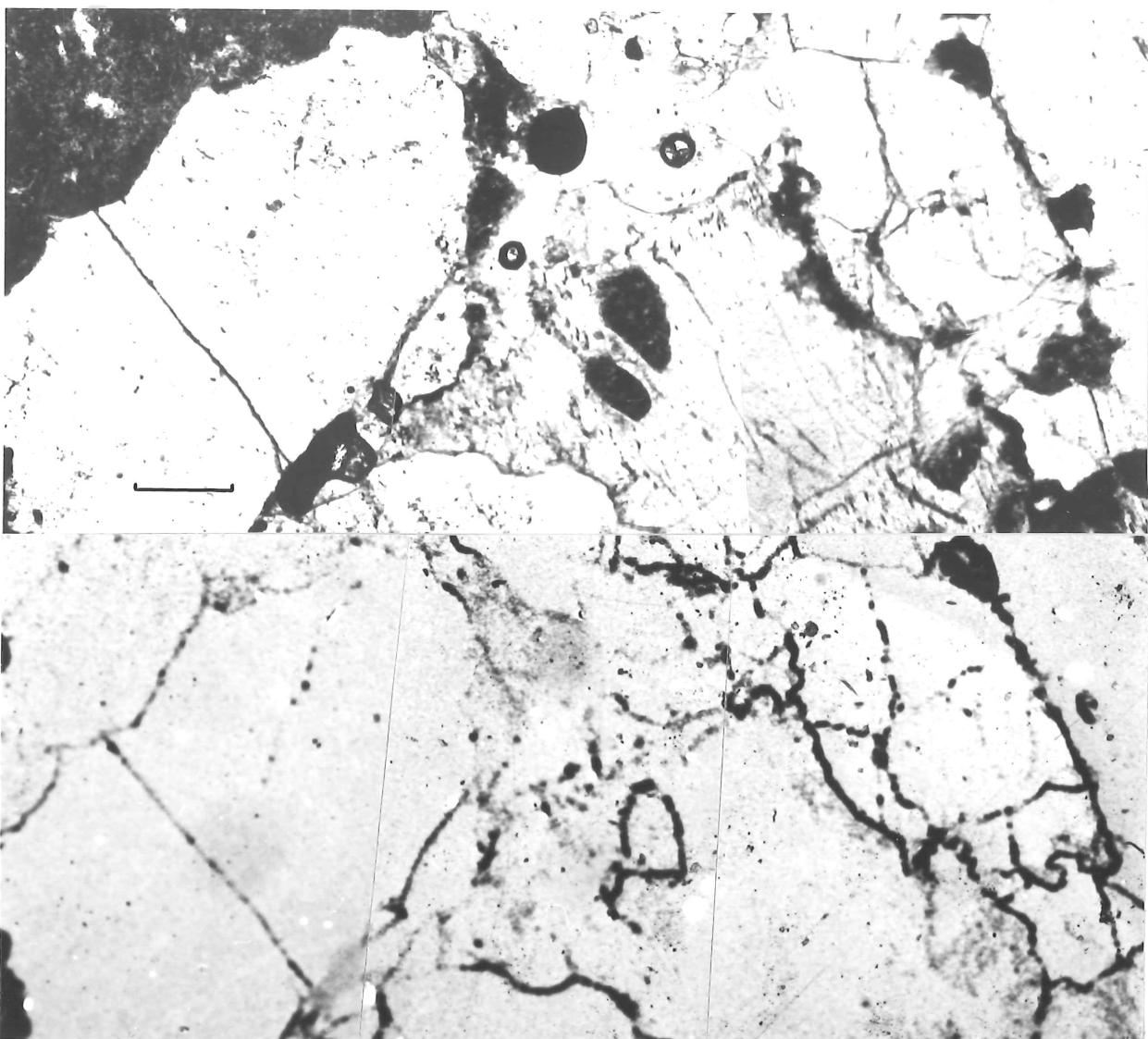


Figure 7 : Photomicrograph of quartz and felspar grains, and corresponding fission track print showing the distribution of uranium along grain boundaries and micro-cracks. Transmitted light, polars uncrossed; bar represents 0,5 mm; irradiation conditions as for Fig. 5.

represents an isochoral density of $0,66 \pm 0,04 \text{ g/cm}^3$ (Touret and Bottinga, 1979), as well as a possible peak of broader spread at $\text{Th} = 22,0 \pm 1,2^\circ\text{C}$ for $d = 0,76 \pm 0,03 \text{ g/cm}^3$. The peak-to-background ratio of a minor clustering at $\text{Th} = 15,0 \pm 1,2^\circ\text{C}$ ($d = 0,83 \text{ g/cm}^3$) is not sufficient to identify a peak. To a large degree this can indeed also be said of the second peak and, as a first approximation, the entire data set can be regarded as skewed unimodal, with a range from $15,0^\circ\text{C}$ to $30,0^\circ\text{C}$ and a turning point at $\text{Th} = 28,0 \pm 1,5^\circ\text{C}$.

The isochores determined for water, brine and carbon dioxide may be used to yield pressure and temperature conditions for the entrapment of fluids, provided that it is assumed that the fluids are consanguinous. It is not clear from the evidence in hand whether the carbon dioxide is cogenetic with the pure water, the brine or both. However, a liberal interpretation of isochoral intersections (Fig. 10) yields P-T constraints of 0,6-1,2 kb and 170-340°C for the fluid entrapment conditions.

VI. MINERALIZATION PROCESS

Any model that attempts to explain the uranium mineralization process in the Namaqualand leucogranites will require to accommodate the following observations :-

- (i) the existence of entrapped fluids which indicate the presence of water, brine and CO_2 circulating within the rock during the late-magmatic and sub-solidus cooling history of the intrusions;
- (ii) a pervasive deuteritic alteration characterized by sericitization of plagioclase, development of a propylitic assemblage comprising chlorite, epidote and magnetite, and the unmixing of magnetite-ilmenite with the formation of Fe- and Ti-rich reaction products; and
- (iii) a high proportion of uranium associated with alteration products such as chlorite, epidote, and Fe-Ti reaction products.

Chemically, the alaskite intrusions are highly fractionated, with high SiO_2 contents, high K/Na and Rb/Sr and low K/Rb ratios. It has been argued (Robb *et al.*, 1985; Robb, 1983) that such characteristics, are compatible with a magma that has undergone significant upward movement in the crust and implies, therefore,

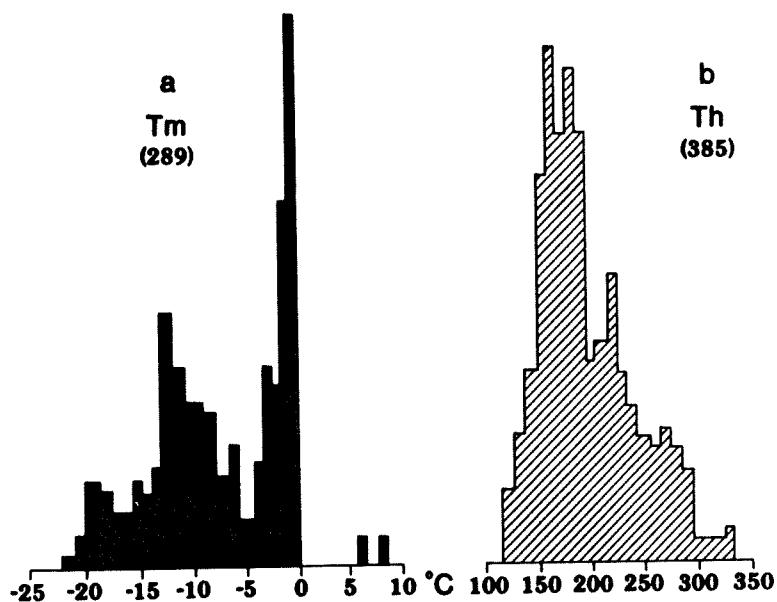


Figure 8 : Histograms of microthermometric measurements made on aqueous inclusions in quartz from the leucogranites (a. melting temperature, b. homogenisation temperatures). Number in parentheses represents number of duplicated measurements.

a fairly shallow level of emplacement. The exact level of emplacement is difficult to quantify, but will not have been too shallow as the leucogranites show neither the texture nor alteration style of "porphyry intrusions" nor the extensive granophytic crystallization of very high level (<1 kb) granite intrusions (Bussell, 1983). During ascent of the magma, crystallization of major and minor rock-forming minerals will undoubtedly have commenced with some uranium and thorium having been accommodated in the early formed

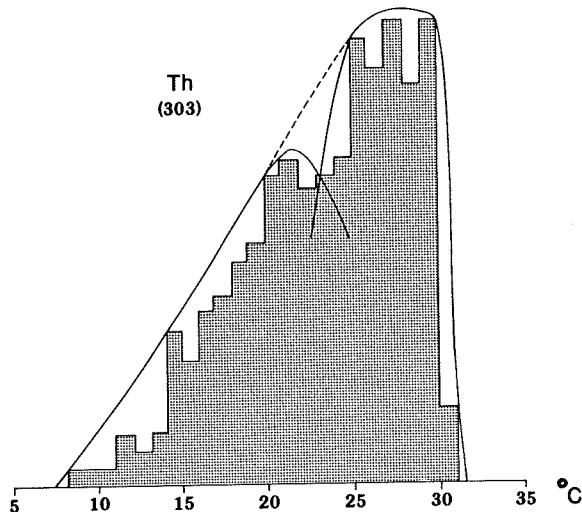


Figure 9 : Histogram of homogenization temperatures for CO_2 inclusions in quartz from the leucogranites.

accessory phases described above. During crystallization at shallow levels (i.e. 2-4 kb) it is envisaged that an attendant aqueous phase will exsolve relatively early in the crystallization interval (Fig. 4) so that equilibration between vapour and crystals occurred already in the magmatic stage. Consequently, alteration of minerals probably commenced during the late-magmatic evolution of the intrusions. However, the alteration process undoubtedly continued during the subsolidus cooling of the bodies as the dominant alteration assemblage now observed (i.e. sericite (muscovite) - chlorite) is only stable up to a maximum of about 550°C at pressures <4 kb (Fig. 4). The upper stability limits of epidote and Fe-rich chlorites (Fig. 4) also indicate that the eventual deuterian assemblage formed at subsolidus temperatures (i.e. at least less than $\approx 500^\circ\text{C}$).

Fluid inclusion characteristics indicate that the fluids circulating within the leucogranite intrusions during the deuterian alteration process comprised brine and carbon dioxide in an approximate ratio of between 4:1 and 5:3. If consanguinity of brine and CO_2 is assumed it is evident from the high NaCl content of the brine (17.5% eNaCl), that unmixing of the two phases will have occurred at relatively high temperatures (i.e. circa 500°C, Fig. 11). Consequently, CO_2 -brine unmixing will almost certainly have been broadly coincident with the deuterian alteration process and must have been complete by the time the fluids were entrapped as inclusions.

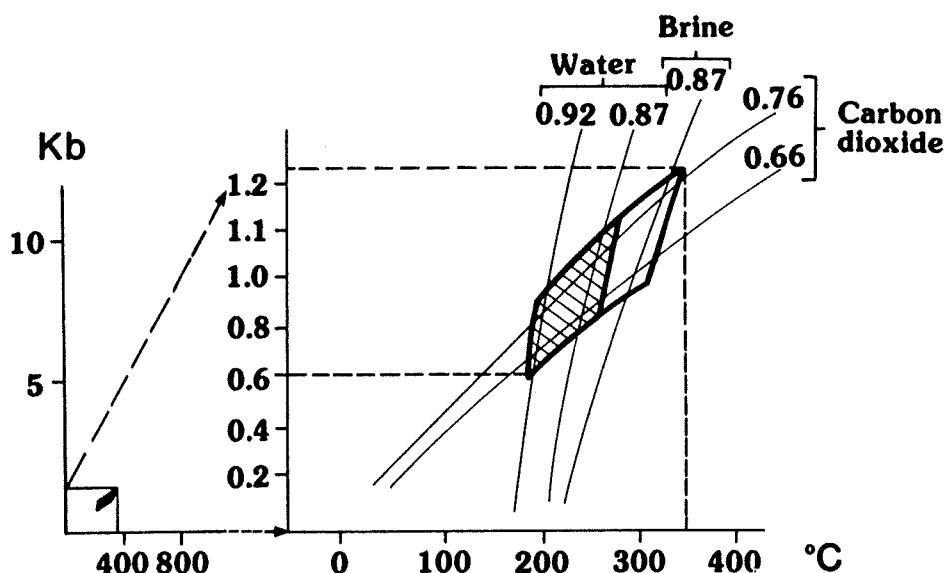


Figure 10 : The area of coincidence of isochores deduced from homogenization temperatures of pure water, brine and carbon dioxide inclusions in quartz from the leucogranites. The numbers on the isochores represent the relevant densities in g/cm³.

The fact that a high proportion of uranium is associated with the products of deuterian alteration indicates that this element was present in the fluid phase. Such an association could either have been the result of dissolution of early-formed, uranium-carrying minerals, or concentration of uranium into the fluid simply as a result of its incompatible nature. Although the presence of dissolved CO₂ in a fluid will increase the solubility of uranium, which may then exist as uranyl di- or tricarbonate ions, it is not essential for the hydrothermal transport of uranium (Poty et al., 1984). The presence of a uranium-charged

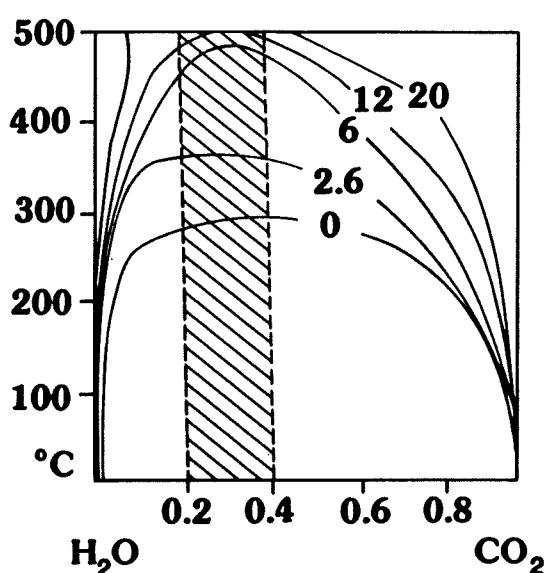


Figure 11 : The H₂O-CO₂ solvus for various amounts of dissolved NaCl. The numbers on the curves represent weight per cent NaCl. Sources :- Tödheide and Franck (1965); Hendel and Hollister (1981); Takenouchi and Kennedy (1965); Bowers and Helgeson (1983a and b)

fluid circulating within the leucogranitic bodies is most probably the cause of the anomalous concentrations of uranium in these rocks. What is less clear is the mechanism by which uranium was precipitated and retained within suitable sites represented by the deuterian alteration products. The evidence for second boiling (i.e. the relatively high temperature unmixing of CO₂-brine) perhaps points to the major mechanism whereby uranium was precipitated. However, the uncertainties in the shape of the CO₂-brine solvus and the nature and content of uranium dissolved in the circulating fluid phase means that the above suggestions must remain inferential. Additional microthermometric work is required to establish the presence of fluids inherited from the magmatic history of the leucogranite. A tentative indication of fluids more dense than those discussed above is provided by small peaks at T_m (H₂O) = -20°C (Fig. 8a), T_h (H₂O) = 270°C (Fig. 8b) and T_h (CO₂) = 15°C (Fig. 9). Although these data are statistically insignificant, they nevertheless suggest P-T conditions of entrapment of up to 2.5 kb and 500°C.

The spectrum of melting temperatures of water indicates that, besides brine, a significant component of pure H₂O is recorded within the fluid inclusions. Because of its low salinity, this water may be meteoric in origin, although it was undoubtedly heated to temperatures in excess of 165°C (Fig. 8b). Whatever the origins of this water it was probably associated with the secondary redistribution of uranium and its

concentration in ferruginous oxides/hydroxides and beta-uranophane within microcracks and intergranular pore-spaces (Fig. 7). It should be pointed out, however, that immiscibility of pure H₂O-CO₂ will occur at fairly low temperatures (i.e. ≈250°C, Fig. 11). Thus, even the secondary enrichment of uranium may have been related to fluid unmixing.

A. Comparisons with the Rössing Uranium Deposit

The leucogranites described above have many superficial similarities with the "alaskites" from the Rössing uranium mine in Namibia and it is pertinent to compare the mineralization processes in the two occurrences. Fluid inclusion studies of the Rössing alaskites (Cuney, 1980) showed that these rocks were also influenced by brine- and CO₂-rich fluids interpreted as being magmatic in origin. The Rössing brine was shown to be somewhat more saline than those characterizing the present study (i.e. about 30% equivalent NaCl) although homogenization temperatures fall in approximately the same range (i.e. 120-320°C) (Cuney, 1980). The CO₂ fluid at Rössing was less pure than that of the Namaqualand leucogranites, containing up to 8 mole per cent equivalent CH₄. At Rössing, however, a second source of CO₂ is envisaged, this being the result of decarbonation arising from the emplacement of the alaskitic magma into a series of marbles. The effect of this CO₂ was to increase the solidus temperature of the magma as well as to increase the CO₂ concentration in the vapour phase (Cuney, 1980). It is envisaged that CO₂-enriched fluids carrying significant concentrations of uranium subsequently left the immediate site of generation, migrating towards higher levels and mineralizing both the upper portions of the intrusion and the overlying metamorphic rocks. In addition, however, considerable uranium crystallized out in the form of uraninite, both during the magmatic stage and from the attendant brine-CO₂ fluid phase.

The principal difference between the Rössing alaskites and the Namaqualand leucogranites appears to be the abundance of uraninite in the former. By contrast, the present study has revealed a scarcity of this mineral in the leucogranites and a more marked concentration of uranium in the deuterian alteration assemblage. The abundance of uraninite at Rössing has been attributed to a fairly low magmatic fO₂ and a preponderance of tetravalent over hexavalent uranium (Cuney 1980). The paucity of uraninite in the Namaqualand leucogranite may indicate a significantly greater proportion of hexavalent uranium in the attendant fluid phases. Consequently uraninite crystallization will have been inhibited and a concomitantly greater proportion of uranium will have ended up in suitable sites within the deuterian alteration assemblage.

VII. CONCLUSIONS

In summary, the essential attributes of mineralization in the Namaqualand leucogranites are the prolonged reaction of rock-forming minerals with an attendant fluid phase capable of dissolving and/or carrying uranium. This uranium was precipitated into suitable sites during the unmixing of a brine-CO₂ fluid at temperatures which were broadly coincident with the onset of a pervasive deuterian alteration in these rocks. Physiochemical conditions appear to have been such that the fluid remained active *within* the system rather than removing the uranium into extraneous hydrothermal vein systems, breccias or pegmatites.

Although the present paper gives consideration to the concentration of uranium on a localized scale, the question of the regional source still remains. It should be pointed out that the entire Namaqualand region is, on average, characterized by granites and gneisses of an "enriched" character, namely, with high SiO₂ (72%), high K/Na (2,1), high Rb/Sr (1,7), low K/Rb (160) and U and Th contents exceeding average crustal values (Holland and Marais, 1983). A bulk composition such as this is unusual in granulite terranes which typically have depleted large-ion-lithophile characteristics. A recent discovery, which may have some bearing on the broad nature of the Namaqualand crust, is the existence of a globe-encircling mantle isotope anomaly centred on latitude 30°S (Hart, 1984). This long-lived anomaly, which is characterized by high ²⁰⁷Pb/²⁰⁴Pb, ²⁰⁸Pb/²⁰⁴Pb and ⁸⁷Sr/⁸⁶Sr, underlies the Namaqua metamorphic province and may have stimulated the generation of high heat production granites such as the ones described here.

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