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RARE-EARTH-RICH EUDIALYTE AND DALYITE FROM A PERALKALINE GRANITE DYKE AT STRAUMSVOLA, DRONNING MAUD LAND, ANTARCTICA

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ABSTRACT

A microgranite dyke that cuts the Straumsvola nepheline syenite complex, Antarctica, contains *REE*-rich eudialyte (*REE*$_2$O$_3$ + Y$_2$O$_3$ *ca.* 8 wt.%) and the rare mineral dalyite. The dyke rock is not particularly potassic (wt.% K/Na 0.83) compared to rocks in which dalyite has recently been discovered (lamproïtes), but it is similar in composition to the granite xenoliths of Ascension Island, in which dalyite was first recognized. Eudialyte in the dyke is richer in *REE* than that from undersaturated peralkaline rocks, but this is not true of the eudialyte from vugs in the surrounding nepheline syenite. The lack of Ca in oversaturated peralkaline magmas in general enables eudialyte to act as an effective *REE*-sink in such rocks. Although rare, eudialyte in peralkaline granites should always be *REE*-rich, whereas that in nepheline syenites need not be, as these rocks invariably are more calcic.

Keywords: eudialyte, dalyite, peralkaline microgranite, rare-earth elements, Antarctica.

SOMMAIRE

Un filon de microgranite qui recoupe le complexe de syénite néphélinique de Straumsvola, dans l'Antarctique, contient une eudialyte riche en terres rares (environ 8% en poids d'oxydes de terres rares et d'yttrium) et de la dalyite, accessoires rare. Le microgranite n'est pas particulièrement potassique (rapport pondéral K/Na 0.83) comparé aux roches lamproïtiques dans lesquelles la dalyite a été découverte récemment, mais sa composition ressemble à celle des enclaves granitiques de l'île de l'Ascension, localité type. L'eudialyte du filon est plus riche en terres rares que les exemples typiques des roches hyperalcalines sous-saturées, mais l'est moins que l'eudialyte des cavités de la syénite néphélinique encaissante. L'absence de calcium dans les magmas hyperalcaline sursaturés, en général, permet à l'eudialyte de capter les terres rares dans ces roches. Quoique rare, l'eudialyte des granites hyperalcalins serait inmanquablement enrichie en terres rares, tandis que celle des syénites néphéliniques ne le serait pas toujours, vu que ces roches sont plus calciques.

(Traduit par la Rédaction)

Mots-clés: eudialyte, dalyite, microgranite hyperalcalin, terres rares, Antarctique.

INTRODUCTION

Straumsvola is a nepheline syenite intrusive complex about 10 km in diameter on the eastern edge of the Jutulstraumen Glacier (72°S, 0°W) in Dronning Maud Land, Antarctica (Ravich & Solov'yev 1969, Allen *et al.* 1986). The nunatak Straumsvola consists of layered nepheline syenite, whereas the adjacent nunatak 5 km to the east, Tvora, consists of quartz syenite. The relationship between these two plutons is not clear, but on the basis of detailed field work, G.H. Grantham (pers. comm. 1986) suggested that they are two separate bodies. A large number of dykes cut the two plutons and the surrounding country-rocks. These dykes are extremely varied in composition and include nephelinites, pyroxenites, lamprophyres, dolerites, phonolites, and peralkaline microgranites. In one unusual dyke of peralkaline granite, the subject of this paper, rare-earth-element-rich eudialyte occurs together with the rare mineral dalyite $K_2ZrSi_6O_{15}$. This dyke crops out on the north ridge of Straumsvola, some 200 m from the summit. Dalyite has so far been described from only four localities [Ascension Island: van Tassel (1952), Sao Miguel in the Azores: Cann (1967), Sunnfjord in west Norway: Robins *et al.* (1983), and Spain (no location given): Linthout (1984)], whereas eudialyte is very rare in oversaturated rocks (3 localities are mentioned by Deer *et al.* 1986). This dyke is, therefore, extremely unusual.

THE OCCURRENCE OF EUDIALYTE AND DALYITE

The dalyite and eudialyte-bearing dyke is 1 m wide and shows pronounced banding parallel to the sides of the dyke (Fig. 1). The dyke is east–west-trending and cuts both the nepheline syenite country-rock and several dolerite dykes. Patches of pink eudialyte are clearly visible in hand specimen. The dyke is very fresh; it appears to have had little effect (either chemically or structurally) on the country rock. The banding reflects phase layering, with layers alternately rich in aegirine and in feldspar and quartz. The layering is considerably more pronounced at the margins of the dyke; the strongly acicular aegirine shows preferred orientation in the plane of the banding.

PETROGRAPHY

In thin section the dyke (ST38) is seen to consist of small euhedral laths of aegirine, up to 3 mm in length, set in a generally finer groundmass of quartz and feldspar. The feldspar shows complex subsoli-

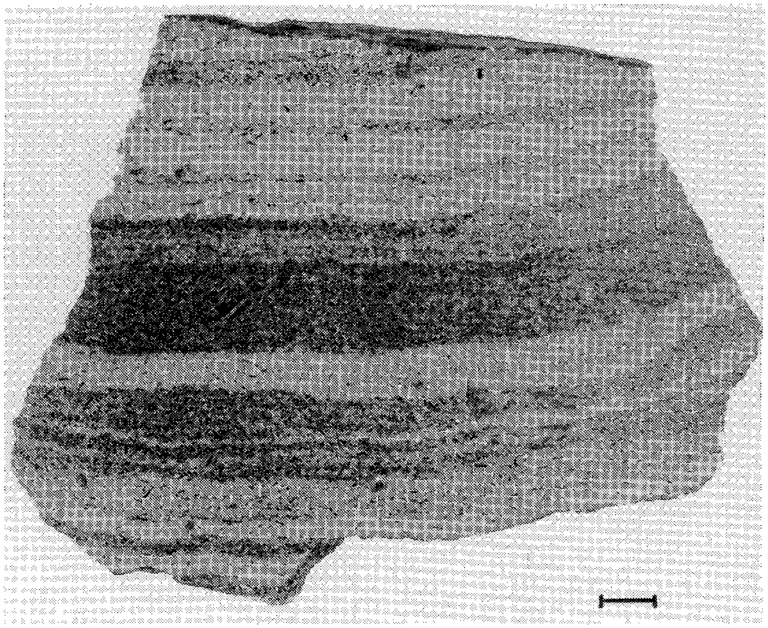

FIG. 1. Hand specimen ST37, collected from the same locality as ST38. The dark bands are relatively rich in aegirine; the light bands consist only of quartz and feldspar. The banding is vertical and parallel to the sides of the dyke. Scale bar represents 1 cm.

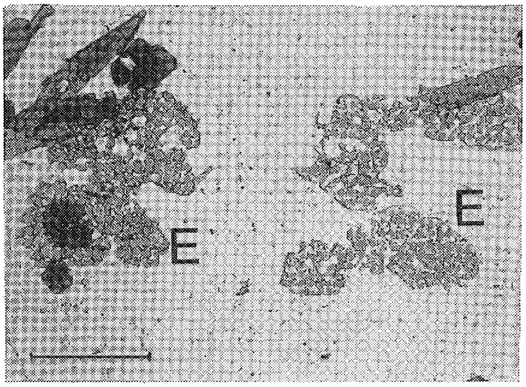

FIG. 2. Photomicrograph of ST38 showing eudialyte (E) filled with inclusions and having, in some cases, a subhedral outline. The dark mineral is aegirine. Scale bar represents 1 mm.

dus development. Some of the larger grains (up to 2 mm in diameter) have a core that exhibits faint cross-hatch twinning and a rim of exsolved albite exhibiting well-developed multiple twinning. Albite is also present as patches along grain boundaries. Well-developed cross-hatch twinning characteristic of microcline is visible on the boundaries of larger grains. Powder X-ray-diffraction patterns of the feldspar are complex but confirm the presence of two feldspar components. The 2θ values for the (060) and $(\bar{2}04)$ reflections (Wright 1968) show these two phases to be maximum microcline and low albite. Microprobe studies of the coarsely exsolved patches reveal no compositions between Or_{98} and Ab_{98}. This is in good agreement with the compositions estimated from the $(\bar{2}01)$ reflections (Or_{99} Ab_1 and Or_1Ab_{99}). The coarseness of the exsolution exhibited by the feldspar is anomalous given the lack of evidence in the rock for the existence of a fluid phase (see below). On the basis of the K/Na ratio of the whole rock, it is probable that the original feldspar was a sodic sanidine, the rock being fairly typical of hypersolvus alkali granites (Tuttle & Bowen 1958).

The only ferromagnesian mineral present is aegirine close to the end-member composition (electron-microprobe data). Fluorite, a common accessory, occurs as small interstitial grains. Zircon has not been observed. Eudialyte occurs as easily recognizable pink patches containing many inclusions, mainly of quartz (Fig. 2). These patches may attain 5 mm in diameter. Dalyite is found as small interstitial grains interspersed throughout the rock (Fig. 3).

Eudialyte is also found in vugs and is deposited along joint planes in the nepheline syenite along a 1-km segment of the summit ridge of Straumsvola, but was not found in other parts of the syenite complex.

OPTICAL MINERALOGY OF EUDIALYTE AND DALYITE

Eudialyte has very low birefringence (1st order grey) with each patch being in the same optical orientation. The eudialyte shows pink to pale yellow pleochroism, but the inclusions that pervade the mineral made determination of optic sign and $2V$ impossible. The eudialyte in two separate vugs is uniaxial positive, and thus true eudialyte rather than the optically negative variety known as eucolite (Deer *et al.* 1986). Dalyite shows low to moderate birefringence (up to 2nd order blue) and is commonly difficult to distinguish from quartz in slightly thick polished thin sections because it exhibits no cleavage. Dalyite is, however, easily recognized during probe analysis owing to its very strong blue cathodoluminescence under the electron beam. The grains in the sample studied are too small to determine optic sign and $2V$. The appearance of dalyite in thin section in ST38 is identical to that of dalyite in granite xenoliths from Ascension Island, where this mineral was first recognized.

An attempt was made to confirm the presence of dalyite by X-ray diffraction of a heavy-liquid separate (>2.75 g cm^{-3}). Reflections of quartz and feldspar in the concentrate overlap several of the strong reflections for dalyite, but those for *d* values of 3.59, 3.09 and 2.83 (van Tassel 1952) were clearly visible.

ANALYTICAL METHODS

Analyses were made using a Cameca Camebax wavelength-dispersion, 4-spectrometer microprobe operating at 25 kV, with a probe current of 40 nA. *REE* were not detected in dalyite at 25 kV and 40 nA; the compositions reported here were made at 15 kV and 20 nA to avoid loss of Na. Initial identification of the elements present in the eudialyte was made by scanning the spectrometers over their complete range. For the *REE* the resolvable peaks were scanned to determine the best analytical line and suitable positions for determination of background. The choice of analytical lines was the same as that of Exley (1980). The analysis for Y was made using the $L\alpha_1$ line and this element is used as a representative of the heavy *REE* in this paper (Eby 1975), its ionic radius being close to that of holmium. For the *REE* + Y, counting times were generally 60 s, but in eudialyte with a high abundance of *REE*, counting times of 30 s were employed to increase the speed of analysis. Na is particularly volatile in eudialyte. Counts are found to greatly decrease even after a few seconds, and consecutive analyses on the same spot reduce the amount of Na determined by up to 50%. This problem was minimized by using a counting time of 2 s for Na and K. On-line ZAF corrections were made to the raw data. Table 1 summarizes the analytical conditions used in the determination of the *REE* in eudialyte.

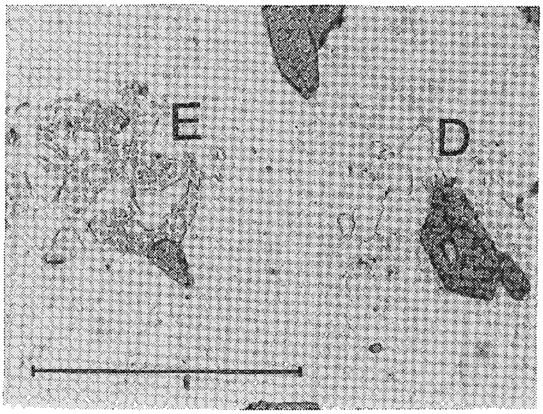

FIG. 3. Photomicrograph of ST38 showing eudialyte (E) and dalyite (D). Dalyite occurs as interstitial patches and is distinguished from quartz by its higher relief. In other areas of the slide, dalyite occurs as inclusions within eudialyte, suggesting that dalyite crystallized before eudialyte. Scale bar represents 1 mm.

The Nb$L\alpha_1$ and Gd$L\alpha_2$ peaks were found to be resolvable, but are affected by interferences from Mn$K\beta_2$ (3rd order) and Ce$L\gamma_1$, respectively. Empirical correction-factors for Nb and Gd were calculated by measuring the contribution to the analytical line by the interference for a known wt.% of Mn and Ce, respectively. Ta was also found to be present in small amounts, but quantitative analysis was impossible owing to the lack of a Ta standard. A mixture of synthetic and natural materials was used as standards, and for the *REE*, the synthetic glasses of Drake & Weill (1972) were used. The whole-rock composition in Table 2 was obtained by XRF techniques currently in use at the Department of Geochemistry, University of Cape Town. Estimates of precision and detection limits are given in Duncan *et al.* (1984) and leRoex (1985).

CHEMISTRY OF DALYITE AND EUDIALYTE

The chemical composition of eudialyte and dalyite is given in Table 3. The ideal formula of eudialyte is variously given as $Na_4(Ca,Ce)_2(Fe^{2+},Mn,Y)ZrSi_8O_{22}(OH,Cl)_2$ (Fleischer 1986) or $(Na,Ca,REE)_5(Fe^{2+},Mn)(Zr,Ti)[(Si_3O_9)_2(OH,Cl,F)]$ (Deer *et al.* 1986). About 2.7 wt.% Nb_2O_5 has been shown to be present; the niobium presumably substitutes for Zr. Even allowing for the presence of Nb and Gd, which could not be determined accurately, all the analyses show low totals (av. 96.04 wt.%). For these grains of eudialyte, assuming the formula of Deer *et al.* (1986), the 'missing' OH is about 0.8 formula units or about 1.5%; this still leaves a deficit of some 1.5% wt.%. Assuming the formula of Fleischer (1986), the 'missing' OH is 1.8 formula units or about 3 wt.%, which would be adequate to explain the low totals. In Table 4, the structural formula of the eudialyte is calculated on the basis of an expanded Fleischer formula, $(Na,K)_4(Ca,REE)_2(Fe,Mn,Y)(Zr,Nb)_{1+x}Si_8$ . . . or $A_4B_2\,CZr_{1+x}D_8$ . . . . This scheme accounts for any extra-site Zr + Nb (Giuseppetti *et al.* 1971). In both cases (Table 4, compositions 1 and 2), the sum of the cations is close to 16, assuming $15(A+B+C+D)$, which suggests that the Fleischer formula is approximately correct. In both examples of eudialyte, the number of *A*-site cations is close to 4. There appears to be a certain amount of substitution among *A,B,C*,Zr and *D* sites; Mn presumably substitutes in *B* in composition 1 and possibly for Zr in composition 2. These variations in site occupancy make the confirmation of extra-site (Zr,Nb) uncertain, though in composition 1 (ST38), (Zr,Nb) = 1.06 formula units.

TABLE 1. ANALYTICAL CONDITIONS FOR REE AND Y

Element	Analytical line	Crystal	Background position*	Detection limit(wt.%)	2 sigma
Y	$L\alpha_1$	PET	+400**	0.047	0.19
La	$L\alpha_1$	PET	+600	0.041	0.06
Ce	$L\alpha_1$	LIF	+800	0.043	0.19
Pr	$L\beta_1$	PET	+300	0.043	0.08
Nd	$L\beta_1$	LIF	+500	0.031	0.15

* For a counting time of 60 s; the peak was counted for 60 s and each background position for 30 s. ** Units are steps, where 1000 steps correspond to approximately 1° 2θ in the region of interest; step interval and 2θ do not vary linearly on the Cameca microprobe.

TABLE 2. BULK CHEMICAL COMPOSITION OF PERALKALINE GRANITE ST38

SiO_2 wt.%	71.51	La ppm	38
TiO_2	0.20	Ce	72
Al_2O_3	12.22	Nd	31
Fe_2O_3	5.13	Nb	108
MnO	0.06	Zr	1305
MgO	0.00	Y	29
CaO	0.46	Sr	23
Na_2O	6.00	Rb	144
K_2O	4.44		
P_2O_5	0.00		
LOI	0.24		
H_2O-	0.12		
TOTAL	100.38		
(Na+K)/Al	1.20		

Analyses by XRF, total normalized to 100 % (volatile free); Fe_2O_3* = total iron as Fe^{3+}
H_2O- = loss at 110°C LOI = loss at 850°C

It is evident from Table 3 that there are important differences between eudialyte in ST38 and that from the vugs in the country rock. The latter are rich in Fe and Ca and relatively poor in Mn and *REE*. These differences suggest that the eudialyte probably was not deposited during the same hydrothermal activity that produced the vugs. Given the texture of the eudialyte and the lack of alteration in the dyke, it is most likely that the eudialyte was a magmatic, albeit near-solidus phase. A possible reason why the dyke contains eudialyte that is so rich in *REE* is that the lack of Ca in the magma allowed *REE* to substitute for Ca in the structure of the eudialyte.

The dalyite in ST38 is chemically similar to that from the other localities. Table 3 gives the average of 3 dalyite compositions from ST38, the average of 3 compositions of dalyite from a granite xenolith from Ascension Island (sample H30(1), Harris *et al.* 1982) and the original composition of dalyite (van Tassel 1952). Compositions 3 and 4 are directly comparable, as they were made using the same instrument on the same day. The major difference is that Na is absent in dalyite from ST38, which consequently has higher K. The lack of Na in dalyite from ST38 is surprising, as this rock has a higher Na/K ratio than H30(1). The dalyite described by Robins *et al.* (1983) also contains very little Na, but this is consistent with the composition of the host rock, a highly potassic lamprophyre (Furnes *et al.* 1982).

PETROGENESIS

The co-occurrence of dalyite and eudialyte has been previously described in a granite xenolith from Ascension Island (Harris *et al.* 1982). Chemically, dyke sample ST38 is similar to the Ascension granite; the major mineralogical distinction is the presence of amphibole in the Ascension sample and its much coarser grain-size. The Ascension example contains abundant primary fluid-inclusions in all the minerals throughout the rock (including dalyite), which point to the water saturation of the original magma (Roedder & Coombs 1967, Harris 1986). The only fluid inclusions present in ST38 are small (<3 μm), infrequent and of indeterminate nature. They occur along fracture planes and are probably secondary. There are no hydrous phases present in the dyke; this absence suggests that it crystallized from a fairly dry melt. The presence of exsolved water is probably not, therefore, a significant factor governing the crystallization of dalyite and eudialyte in peralkaline granites.

TABLE 3. BULK CHEMICAL COMPOSITION OF EUDIALYTE AND DALYITE

	1	Range	2	Range	3	4	5
SiO_2	47.70	46.74-48.51	49.14	48.54-50.11	62.07	63.25	61.85
TiO_2	-		-		0.13	0.12	na
FeO	0.93	0.67-1.10	6.65	6.33-6.90	-	0.03	na
MnO	8.07	7.77-8.77	1.10	0.92-1.27	-	-	na
CaO	5.44	5.10-6.04	11.63	11.24-11.95	-	-	na
Na_2O	12.17	11.58-12.75	13.23	12.58-14.12	-	0.79	1.75
K_2O	0.19	0.16-0.27	0.42	0.38-0.49	15.71	14.63	14.60
ZrO_2	10.20	9.44-10.57	11.18	10.93-11.47	20.63	20.43	21.70
Nb_2O_5*	2.7		0.97		-	-	na
La_2O_3	1.58	1.46-1.85	0.22	0.19-0.26	-	-	na
Ce_2O_3	3.38	3.12-3.68	0.38	0.29-0.46	-	-	na
Pr_2O_3	0.34	0.30-0.38	-	0.04	-	-	na
Nd_2O_3	1.33	1.30-1.38	0.08	0.03-0.14	-	-	na
Gd_2O_3*	0.15		-		-	-	na
Y_2O_3	1.44	0.83-1.72	0.16	0.10-0.21	-	-	na
Cl	0.76	0.64-0.81	1.46	1.42-1.51	-	-	na
F	-		-		-	-	na
O=Cl	-0.34		-0.66				
TOTAL	96.04		95.96		98.54	99.25	100.91**

\- = not detected; na = not analyzed. ** contains 0.64 H_2O

* NB Analyses for Nb and Gd were only possible using empirical corrections (see text).

1 Average of 4 eudialyte from ST38
2 Average of 11 eudialyte from vugs ST34 and ST45
3 Dalyite from ST38 (average of 3)
4**Dalyite from Ascension (average of 3). Sample H30(1) of Harris et al. (1982)
5 Dalyite from Ascension Island (van Tassel 1952)

** Analysis made using same machine and conditions as analysis 3.

Since dalyite is commonly found as inclusions in eudialyte in ST38, it must have crystallized first. There are very few experimental data on the phase equilibria of zirconium silicates in natural systems. Caruba *et al.* (1970) synthesized dalyite from a mixture of hydrous ZrO_2, silica and either KOH or K_2CO_3, with $K_2ZrSi_6O_{15}$ stoichiometry. Wadeite was found to coexist with dalyite at higher pressures (greater than 0.2 kbar at 600°C). In natural systems, factors such as K/Na and silica activity are also likely to influence the crystallization of zirconium silicates. Dalyite can be related to wadeite by the reaction; $K_2ZrSi_6O_{15}$ (dalyite) = $K_2ZrSi_3O_9$ (wadeite) + 3 SiO_2. The absence of wadeite in ST38 is more likely to be the result of the high activity of silica than the low pressure of crystallization.

The absence of zircon must reflect its instability in peralkaline melts (Watson 1979). Table 2 shows that ST38 has a molar excess of alkalis relative to aluminum, but it is not obvious why dalyite should crystallize rather than a sodium zirconium silicate such as vlasovite $Na_2ZrSi_4O_{11}$. This mineral also occurs in the granite xenoliths from Ascension (Fleet & Cann 1967), though never in those that are dalyite-bearing (Harris *et al.* 1982). The absence of vlasovite in ST38 is surprising because the vlasovite-

TABLE 4. STRUCTURAL FORMULAE OF EUDIALYTE AND DALYITE

	1	2		3	4	5
Na	4.02	4.07	K	1.95	1.79	1.79
K	0.03	0.08	Na	-	0.15	0.32
Ca	0.99	1.98	Zr	0.98	0.95	1.01
REE	0.40	0.03				
			Si	6.03	6.05	5.95
Fe	0.14	0.88				
Mn	1.16	0.15		8.96	8.94	9.07
Y	0.14	0.01	Based on 15(0)			
Zr	0.85	0.86				
Nb	0.21	0.07				
Si	8.13	7.80				
	16.07	15.93				

1 Average of 4 eudialytes ST38
2 Average of 11 vug eudialytes
3 Dalyite ST38 (av. of 3)
4 Dalyite from Ascension (av.of 3)
5 Dalyite (van Tassel 1952)

Formula of eudialyte taken as;
$(Na,K)_4\ (Ca,REE)_2\ (Fe,Mn,Y)\ (Zr,Nb)_{1+x}\ Si_8$
or, $A_4\ B_2\ C\ Zr_{1+x}\ D_8$
Structural formulae calculated assuming (A+B+C+D) = 15

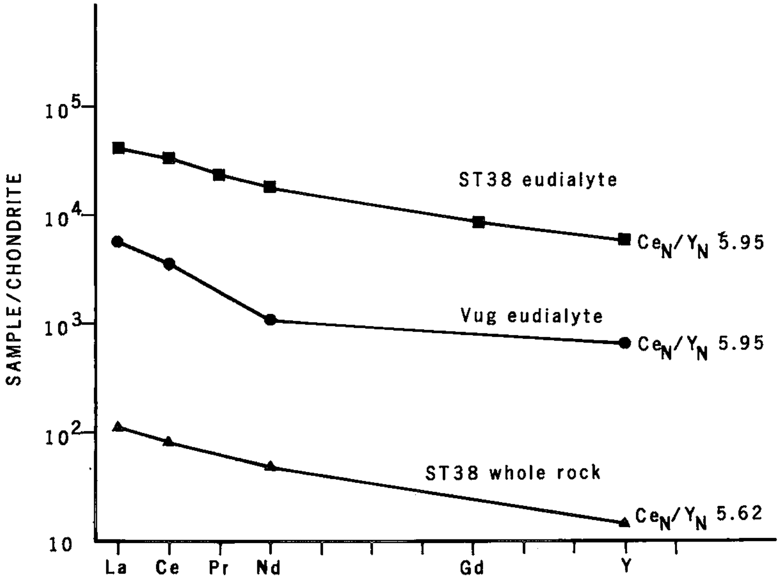

FIG. 4. Partial *REE* profile for eudialyte from dyke sample ST38, eudialyte from vugs in nepheline syenite country-rock and ST38 whole rock. The eudialyte profiles are for the averages given in Table 3. Chondrite values for normalization are taken from Nakamura (1974) and Frey *et al.* (1968).

bearing granite xenoliths from Ascension have higher K and lower Na than ST38. The established order of crystallization (dalyite followed by eudialyte) suggests that dalyite is on the liquidus for high Zr concentrations; once the Zr content of the melt is sufficiently reduced by crystallization of dalyite, eudialyte becomes the stable Zr phase.

Fryer & Edgar (1977) discussed the *REE* content of eudialyte from a range of undersaturated peralkaline rocks. They showed that eudialyte has a rather unfractionated *REE* pattern relative to the whole-rock and that the concentration of the *REE* typically increases with increasing peralkalinity of the whole rock. They observed an inverse correlation between fractionation of the *REE* pattern (*i.e.*, Ce/Y) in the eudialyte and the total *REE* enrichment in the rock. Fryer & Edgar suggested that the low values of the Ce/Y ratio are the result of complexing of the *REE* with halogens and CO_2. *HREE* complexes are more stable than *LREE* complexes, with the result that the *HREE* become enriched relative to the *LREE* in the parental magmas during protracted differentiation.

Figure 4 shows average partial *REE* profiles for eudialyte from the dyke sample ST38 and from the vugs. The whole-rock profile for ST38 is also shown. In these plots, Y is plotted in place of Ho on the basis of similarity in ionic radius, as recommended by Eby (1975). In all cases there is a slight flattening of the patterns between Nd and Y, but there is no suggestion of extreme enrichment in the *HREE* similar to that reported by Fryer & Edgar (1977), who described eudialyte with concave *REE* patterns with Ce/Yb = 1. Complexing of *REE* has been much less important during the evolution of the dyke ST38 than in the examples described by Fryer & Edgar (1977). Note that in the the two described examples of eudialyte in peralkaline granites in which the eudialyte has been analyzed for the *REE* (Ascension Island and this example), the eudialyte is enriched in *REE* over that described from undersaturated peralkaline rocks.

Marsh (1976) has shown that oversaturated peralkaline rocks often contain less than 0.15 wt.% CaO, whereas undersaturated peralkaline rocks; such as nepheline syenites always contain more than 0.5 wt.% CaO. If the efficiency of eudialyte as a *REE* sink depends on the *REE* being able to substitute for Ca in eudialyte, and therefore on the amount of Ca in the magma, then eudialyte in peralkaline granites should always be *REE*-rich. This need not necessarily be so for the undersaturated rocks; eudialyte in these rocks will not in general be *REE*-rich.

Of the other localities where dalyite has been described, the Ascension and Sao Miguel examples are very similar in paragenesis. The Sunnfjord dykes in Norway (Furnes *et al.* 1982, Robins *et al.* 1983) are very different, being poorer in Si (57 wt.% SiO_2) and very high in K (up to 12% K_2O). Linthout (1984) also reported dalyite in lamproite lavas from Spain. It would appear, then, that dalyite is stable near the liquidus for a wide range of K concentrations, in both silica-saturated and silica-undersaturated magmas.

CONCLUSIONS

1. Dalyite followed by eudialyte form a late-state association of accessory phases in a peralkaline granite dyke.
2. The eudialyte in the dyke appears to be magmatic. It is chemically very different and therefore not related to the hydrothermal eudialyte found in vugs in the surrounding nepheline syenite.
3. The eudialyte in the dyke is rich in *REE*, presumably owing to the low Ca in the granitic magma. The eudialyte in ST38 has been a very effective *REE* sink. Peralkaline granites always contain very little Ca (Marsh 1976). Eudialyte in such rocks can be expected to be *REE*-rich. This need not be the case in the more Ca-rich undersaturated rocks.
4. Dalyite will crystallize in peralkaline magmas even where the K/Na ratio is not especially high.

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