

Rare earth element distribution in a metasomatic zoned ultramafic pod from Fiskenaasset, West Greenland

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ABSTRACT. The distribution of *REE* in a zoned ultramafic pod formed by incomplete re-equilibration of ultrabasic and quartzofeldspathic reactant compositions has been studied. Transport of the heavy *REE* (*HREE*) as well as the light *REE* (*LREE*) over several metres has occurred during the diffusion-controlled metasomatism of the protolith mineral assemblages. The largest resultant concentration range (Eu) exceeds two orders of magnitude. In general, *REE* abundances increase towards the marginal zones, and differences between the behaviour of *LREE*, middle *REE* (*MREE*) and *HREE* subgroups are observed. *LREE* are least mobile in the aqueous transporting medium. Complexing by carbonate ligands is probably not an important factor in this system, and the final *REE* distribution is thought to be governed largely by the crystal structure of the major zonal minerals.

THE conditions that can give rise to element mobility during processes of rock alteration, metamorphism or metasomatism are of current importance since certain trace elements, including the rare earths, are sometimes used as indicators of a rock's parentage. This use is based on the assumption that the distribution of the elements, at least on the scale of a typical hand specimen, is often unaffected by such secondary processes. There is convincing evidence that in some circumstances, the rare earth elements (*REE*) have not been affected by metasomatism. For example, Muecke *et al.* (1979) report almost unchanging *REE* concentrations across a metabasite-epidosite contact in an amphibolite-facies terrain. On the other hand, significant quantities of *REE*, especially light *REE* (*LREE*), can be introduced during metasomatism (e.g. the significant increase in *LREE* abundances with degree of fenitization of a quartzite as described by Martin *et al.*, 1978), while in other cases (e.g. during boron metasomatism) *REE* may be lost (Alderton *et al.*, 1980).

Complexing has long been recognized (e.g. Mineyev, 1963) as a significant factor in *REE* transport, and the importance of pH and the presence of certain ligands (notably F^- , Cl^- , CO_3^{2-}) has become clear. Unfortunately, there is often little direct evidence of which ligand or ligands were involved in a given metasomatic process. Thus, studies of the introduction and removal of trace elements during different types of metasomatism, and of the possible roles that newly-generated minerals may play in fixing these elements, should be helpful.

The purpose of this paper is to present the results of a study of the *REE* distribution after the metasomatic development of a zoned ultramafic pod in order to give further evidence on the types of alteration that lead to element, especially *REE*, redistribution. It is intended also to show the degree of *HREE* mobility, relative to that of the *LREE*, under the prevailing conditions.

Geological setting and relationships of samples

The rocks used in this study are from a zoned ultramafic pod, 6 m in diameter, and its associated rock types set in an Archaean granodioritic to tonalitic orthogneiss complex at SW Akugdliungit, near Fiskenaasset. The following genetic model is applied throughout this paper.

Tectonic disruption of large, possibly layered ultrabasic protoliths (e.g. Friend and Hughes, 1977) gave rise to sub-spherical ultrabasic inclusions (e.g. GGU 104758) in the host quartzo-feldspathic gneisses (e.g. GGU 68511), between 60 cm and 10 m in diameter. These were subjected to two phases of fluid activity. The first resulted in widespread hydration of the ultrabasic mineral assemblage to

a low-grade association of antigorite + magnesite + iron ore + talc. Remnants of this immediate precursor to the zonal sequence are preserved as unzoned, homogeneous ultramafic masses (e.g. GGU 68549). The second phase was probably initiated during a subsequent prograde metamorphism (Fyfe *et al.*, 1978), and generated the typical zoned pods that form the subject of this investigation [samples GGU 68551, 68552/3(i)-(iv) and 68553(v)] by incomplete re-equilibration of the antigoritic assemblage with the enclosing acid country rocks. Expulsion of MgO and H₂O from the reacting pod produced an impersistent, rather diffuse shell of second-generation antigorite infiltration in the adjacent gneisses (e.g. GGU 68557).

The analysed samples may thus be divided into the groups below, all from SW Akugdlinguit, except GGU 104758, which was necessarily collected away from the region of later intense metasomatism, at Marrardlugtoq.

Parts of the following discussion require a knowledge of the position of the original discontinuity between ultrabasic pod and country rock, with respect to the zonal sequence. Unequivocal statement of its present locus is impossible with the available data since this particular system lacks elemental 'inert markers' (Brady, 1977). However, Fowler *et al.* (1981) suggested on a morphological basis that the discontinuity is probably now represented between the hornblende and chlorite zones. Recent experimental work (Koons, 1981), in addition to studies of other natural occurrences (Koons, 1981; Sanford, 1982), substantiate this inference that the chlorite zone was derived from the granodioritic mass. This interpretation is therefore retained here.

It is not the purpose of the paper to discuss at length the theoretical aspects of zonal metasomatism, since these have received detailed treatment in numerous recent publications (e.g. Brady, 1977; Frantz and Mao, 1976, 1979; Joesten, 1977; Sanford, 1982; Weare *et al.*, 1976). Rather, it is intended to study in particular the behaviour of the *REE* group during zone formation, and thereby to

provide data of the type considered to be important by Sanford (1982). The reader is referred to Fowler *et al.* (1981) for further details of the genetic history, mineralogy, and geochemistry of the pod.

Analytical techniques.

Samples were analysed for eleven *REE*, U and Th by instrumental neutron activation analysis using an intrinsic Ge detector coupled to a Nuclear Data 6620 analysing system. 150–200 mg of sample together with multi-element standards were irradiated in a thermal neutron flux of about 10^{12} n cm⁻² s⁻¹ for 24 hours. Details of the procedure are given in Henderson and Williams (1981), but because of the low levels of La in the samples, this element was determined additionally using a Ge(Li) detector for the 487.0 and 1596.2 keV peaks of ¹⁴⁰La. U was determined by means of the 106.1 keV peak from ²³⁹Np, with 1 µg of U included in the multi-element standard.

The results are given in Table I, together with values for USGS standard rock BCR-1. Correspondence between these values and the 'usable values' of Abbey (1980) are good (Table I). At concentration levels around 1–2 times chondritic abundance (i.e. as in the majority of these samples), the error is estimated at better than ± 10% for La, Ce, Sm, Eu, Tb, Yb, and Lu, and 10–20% for Gd, Ho, Tm, and Nd. H₂O and CO₂ were determined with a C–H–N analyser, using the technique described by Din and Jones (1978).

Results and discussion

Chondrite-normalized *REE* plots reveal variations between the zones in the ultramafic pod itself (fig. 1a), and between the pod and the other samples studied (fig. 1b). The homogeneous ultramafic rock (GGU 68549), which represents the products of the first metasomatic episode, shows an overall enrichment of all the *REE* when compared with its unaltered dunite precursor (fig. 1b), together with some fractionation since the *HREE* are enriched over the *LREE* by a factor of 2. Thus, some bulk movement of the *REE* was apparently associated with the initial phase of fluid activity which hydrated the original ultrabasic bodies to the low-grade assemblages observed. The resultant

(i) Original ultrabasic dunite	GGU 104758	from Marrardlugtoq	
(ii) Unzoned, homogeneous ultramafic	GGU 68549	both from the locality	} Reactants
Unaltered gneiss	GGU 58511	of the zoned pod	
(iii) Central zone	GGU 68551	all from 6 m diameter zoned pod	} Products
Talc zone	GGU 68552/3(i)		
Inner tremolite	GGU 68552/3(ii)		
Outer tremolite	GGU 68552/3(iii)		
Hornblende	GGU 68552/3(iv)		
Chlorite	GGU 68553(v)		
Altered gneiss	GGU 68557	from immediately adjacent to pod	

(Samples 68552/3(i)-(iv) are averages of samples 68552(i)-(iv) and 68553(i)-(iv) quoted in Fowler *et al.* (1981)).

Table I. Analytical Results

Element (ppm)	GGU 68551	GGU 68553(i)	GGU 68553(ii)	GGU 68553(iii)	GGU 68553(iv)	GGU 68553(v)	GGU 68549	GGU 104758	GGU 68557	GGU 68511	BCR-1 (this work)	BCR-1 (Abbey 1980)
	Central zone	Talc zone	Tremolite zone (Inner) (Outer)		Hornblende zone	Chlorite zone	Altered unzoned pod	Unaltered pod	Altered gneiss	Country rock		
	[core.....rim]											
La	0.45	0.40	0.43	0.82	2.0	2.0	0.70	0.40	24.9	11.3	26.2	27
Ce	< 0.8	0.83	0.83	2.3	7.4	8.2	2.3	1.4	47.5	29.4	53.2	53
Nd	< 1.0	< 1.0	< 1.0	1.8	8.2	9.95	1.2	0.83	9.57	7.60	27.0	26?
Sm	0.06	0.07	0.18	0.75	2.94	3.70	0.33	0.16	0.79	1.25	6.70	6.5
Eu	0.007	0.02	0.05	0.16	0.61	1.16	0.08	0.03	0.30	0.42	2.01	2.0?
Gd	0.1	< 0.3	< 0.3	0.51	2.2	3.3	< 0.3	< 0.3	0.74	< 0.8	6.9	6.6?
Tb	0.02	0.01	0.04	0.09	0.40	0.57	0.03	< 0.02	0.08	0.14	1.12	1.0
Ho	< 0.1	< 0.1	< 0.1	0.1	0.44	0.84	< 0.1	< 0.1	< 0.1	< 0.2	1.4	1.2?
Tm	0.02	0.02	0.02	0.07	0.18	0.32	0.02	0.02	0.02	0.07	0.53	0.6?
Yb	0.13	0.05	0.19	0.35	1.20	1.61	0.14	0.04	0.06	0.40	3.33	3.4
Lu	0.02	< 0.02	0.03	0.05	0.16	0.20	0.02	< 0.01	< 0.01	0.05	0.49	0.5?
Th	< 0.1	< 0.1	< 0.1	< 0.1	0.14	1.12	0.14	< 0.1	3.11	5.5	6.2	6.1
U	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.21	0.1	< 0.1	< 0.1	0.5	1.78	1.7
wt% H ₂ O+	5.61	3.46	3.72	3.09	6.28	12.7	11.6	0.36	5.18	0.66	-	-
wt% CO ₂	2.17	0.29	0.20	0.13	0.13	0.35	0.61	0.07	< 0.02	0.35	-	-

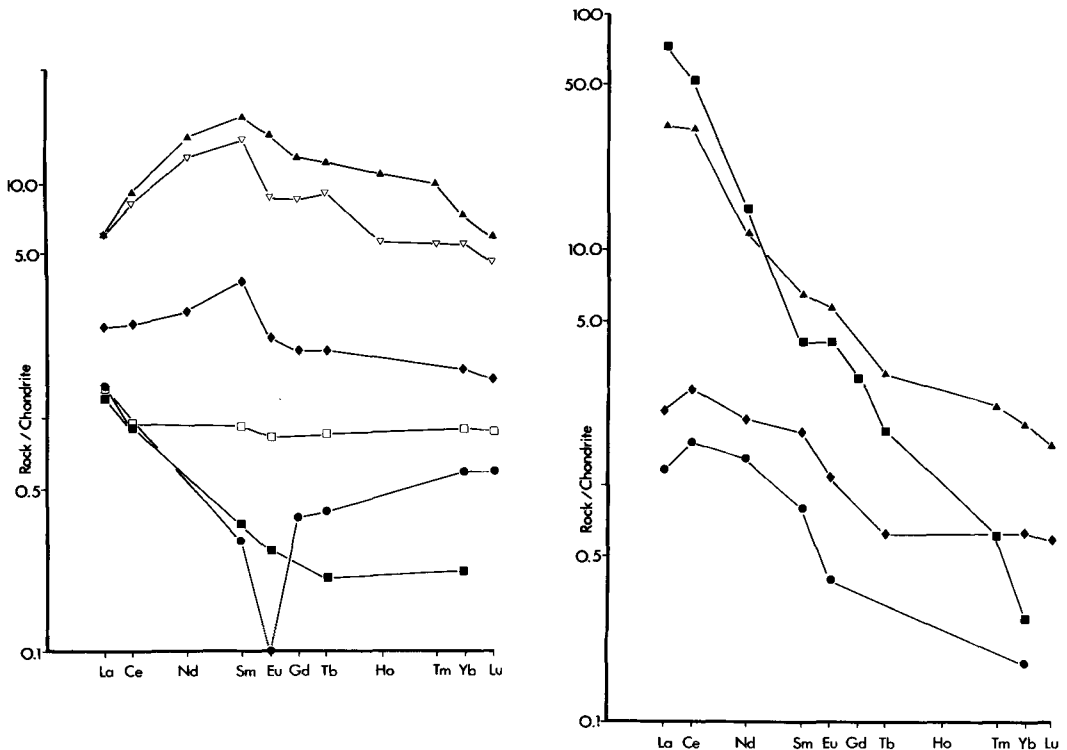


FIG. 1. (a, left) Chondrite-normalized (values from Wakita *et al.*, 1971) REE plots of samples from the zonal sequence: filled triangles = chlorite zone; inverted triangles = hornblende zone; diamond = outer tremolite zone; open square = inner tremolite zone; filled circle = central zone; filled square = talc zone. (b, right) Chondrite-normalized plots of remaining samples: triangles = unaltered gneiss (GGU 68511); squares = altered gneiss (GGU 68557); diamonds = unzoned homogeneous ultramafic (GGU 68549); circles = unaltered dunite (GGU 104758).

REE pattern is slightly *LREE* enriched ($(La/Yb)_{cn} = 3.22$), has a flat *HREE* section and no significant Eu anomaly. Although the gneissose reactant is also *LREE* enriched (to a greater degree, $(La/Yb)_{cn} = 18.3$), the pattern is more fractionated and has a steep *HREE* trend.

Zonal sequence. The *REE* distributions in the reaction products (i.e. the mineral zones) produce a series of patterns with regularly varying shape and abundance (fig. 1a). This implies substantial *REE* mobility during the metasomatic event, which resulted in increasing *REE* concentrations towards the perimeter of the pod. Some degree of inter-element fractionation is also apparent. *MREE* exhibit a greater interzone concentration range than either the *LREE* or *HREE*, with the maximum variation shown by Eu (greater than two orders of magnitude). It is also apparent that the volume within which *REE* movement occurred was not limited to the zonal sequence itself, since the sample of patchily developed altered gneiss (GGU 68557) adjacent to the pod has a highly fractionated *REE* pattern ($(La/Yb)_{cn} > 270$), compared with that of the unaltered gneiss ($(La/Yb)_{cn} = 18.3$).

In discussing the movement of a coherent group of trace elements, such as the *REE*, during an alteration event, it is convenient to normalize to an assumed precursor in order to facilitate a quantitative assessment of concentration changes. For the reasons outlined above, the chlorite zone has been normalized to the values for the unaltered granodioritic gneiss; all other zones have been normalized to the homogeneous ultramafic rock. The assumption is also made that the *REE* were uniformly distributed in the precursor rocks.

With these assumptions, precursor-normalized values for the *REE* were plotted against atomic number (fig. 2). The plots show that substantial *REE* redistribution has occurred within the zonal sequence, particularly of the *MREE* sub-group. Variable depletion in the inner zones is complemented by enrichment in the outer tremolite and hornblende zones. Similarly, the development of a slight positive Eu anomaly in the talc and inner tremolite zones may be partly balanced by larger, negative anomalies in the less voluminous outer tremolite and hornblende zones. A very large, negative Eu anomaly is shown by the central zone. The pattern for the chlorite zone has similar *HREE* and *MREE* characteristics to the amphibole zones, but shows a rapid decrease in those elements lighter than Sm. La and Ce are depleted by factors of 16 and 3.6 respectively relative to the precursor values.

A plot of representative members of the *LREE* (La), *MREE* (Sm), and *HREE* (Yb) against distance

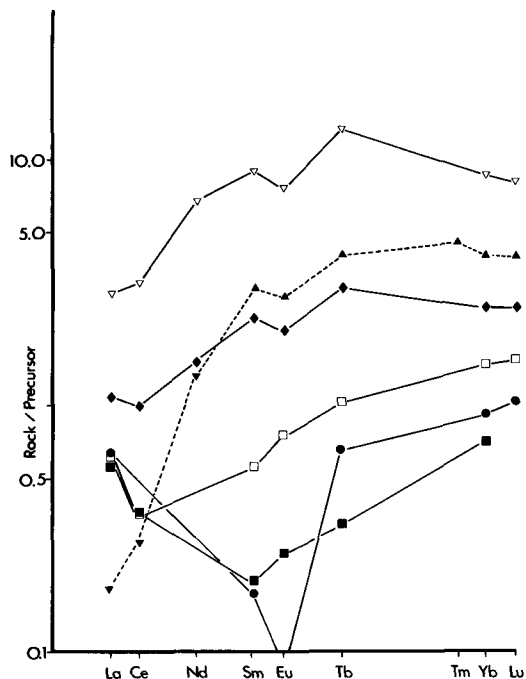


FIG. 2. Precursor-normalized *REE* plots of samples from the zonal sequence, all normalized to GGU 68549 (except chlorite zone, which is normalized to GGU 68511, on the assumption that it was derived from the granodiorite). Symbols as in fig. 1a.

(fig. 3) gives an indication of the intra-group variation as concentration profiles. Thus, La varies little in the three inner zones, increases slowly to about three times its assumed original concentration in the outer zones, then rises sharply into the altered gneiss only to fall again to the concentration of the unaltered gneiss. On the other hand, Sm rapidly increases throughout the entire zonal sequence (by a factor of about 50), reaching nine times its original abundance in the hornblende zone. It continues to increase in the chlorite zone, but falls in the altered gneiss to a value lower than that of the unaltered country rock. Yb initially decreases from the central to the talc zones, and then behaves in a similar manner to Sm.

Mass balance. Several recent studies have considered the importance of volume change in the assessment of mass balance in general (Rubie, 1982) and in zonal metasomatism in particular (Sanford, 1982). Moreover, the latter author points out that significant volume change may occur with only minor variation in linear dimension. Thus, volume changes may be difficult to quantify using linear measurements, but in this particular study no contemporaneous deformation structures were

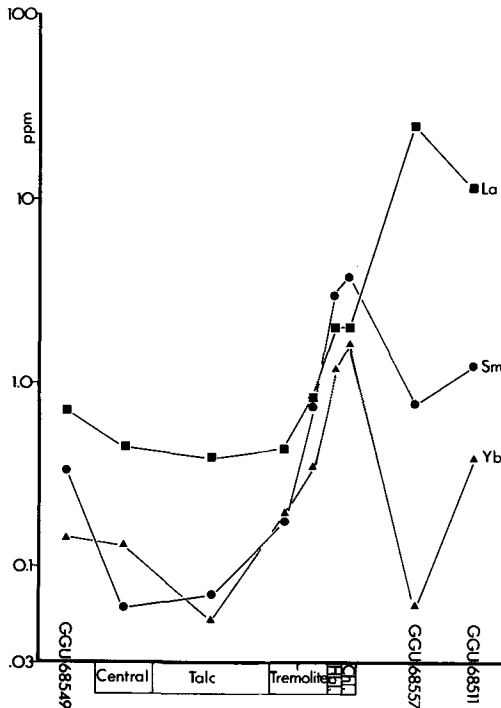


FIG. 3. Element abundance variation diagram for La, Sm, and Yb. (GGU 68549 = ultramafic precursor; GGU 68511 = gneissose precursor; GGU 68557 = altered gneiss; Hbl. = hornblende zone, Chl. = chlorite zone.)

observed in either the pod or the adjacent country rock. Bearing these points in mind, the calculations in Table II are on a constant volume basis. In addition, field observations of the pod showed some zone boundaries to be diffuse and zone widths to vary; the calculations here are based on average zone widths. The mass balance calculations in Table II therefore should be considered only as

an indication of trends. They do nevertheless substantiate the differences between the REE subgroups discussed above, and imply that Sm and Yb exhibited greater degrees of mobility than did La. It is appropriate also to note that although volume change alone may cause some variation in element abundance, it is not capable of interelement fractionation such as that described here. All REE apparently suffered some loss from the pod, indicating some interaction with the surrounding country rock. This information, in conjunction with the profiles in fig. 3, suggests that La was deposited in the altered gneiss surrounding the pod (cf. Allen and Pennell, 1978; and discussion below). However, both Sm and Yb are depleted in the altered gneiss, indicating that mobility of the MREE and HREE may have continued—indeed, some leaching may have occurred—until deposition at a greater distance from the boundary of the pod.

REE mobility and fixation. Some migration of the REE has clearly occurred during the metasomatic event which produced the zonal sequence. Evidence from comparable sequences described by Allen and Pennell (1978) and Sharpe (1980) suggests that the REE redistribution due to metasomatic events of this type may be similar. With REE data limited to La and Ce, Sharpe (1980) found that those elements were more abundant in amphibole and chlorite zones than in the others. Allen and Pennell (1978), in describing a zonal sequence produced from serpentinite blocks within a hornblende diorite country rock (Trinity Co., California), found that the LREE were lost from the precursor serpentinite, and apparently deposited in a reaction zone around the inclusion. They also found that Sm concentration when plotted against distance, had a sigmoidal pattern similar to that shown in fig. 3. In addition, the chondrite-normalized Eu anomaly changes from negative within the zonal sequence to positive outside it.

TABLE II. Mass balance calculations for La, Sm, and Yb in the zonal sequence

Zone	Width (cm)	% mass*	% loss (-) or gain (+)		
			La	Sm	Yb
Central	84.5	2.2	-0.79	-1.79	-0.3
Talc	200	83.0	-35.6	-65.4	-53.3
Inner Tremolite	7	6.5	-2.51	-2.99	+2.26
Outer Tremolite	7	6.8	+1.15	+8.59	+10.1
Hornblende	1	1.0	+1.84	+7.85	+7.51
Total	299.5	—	-34.1	-43.5	-23.1

* Assumes no volume differences between original body and the zones, and $\rho = 2.5$ throughout, density differences being insignificant compared to other uncertainties.

A preliminary assessment of the relative importance of some of the factors involved in *REE* mobility and fixation may be attempted here. The transporting medium was aqueous, as is indicated by the hydrous mineral assemblage and whole-rock H_2O^+ values (Table I). Carbonate ligands were probably not of significance in *REE* transport as CO_2 contents (Table I) are low and do not correlate with *REE* distribution, and calcite is observed as a trace mineral only in the inner zones. The presence of large Eu anomalies in some of the zones suggests that the oxygen fugacity was sufficiently low to maintain a significant proportion of Eu in the divalent state. Other physico-chemical parameters, including the role of other possible anions (e.g. F^- , Cl^-), are difficult to define because of the complex and changeable nature of the fluid, as reflected in the major and trace element variation profiles presented by Fowler *et al.* (1981).

The presence of *REE*-rich trace or minor minerals may influence the overall *REE* pattern of the rock samples, and for this reason a detailed microscope examination of the minor minerals was undertaken. Sphene was the only observed minor mineral that is capable of accommodating large concentrations of the *REE* (e.g. Staatz *et al.*, 1977). In the pod, it was observed only in the hornblende and chlorite zones where it is developed in grains that are rarely larger than 10 μm across, and at an estimated modal proportion of less than 0.05%. Several grains of sphene from the hornblende zone were analysed by wavelength-dispersive electron probe microanalysis for some of the *REE*. The recorded concentration levels are close to the detection limit and are therefore subject to a large error, but consistent values were obtained for La (0.02%), Ce (0.17%), Gd (0.05%), Dy (0.07%), and Er (0.04%). These data and the estimated modal proportion of sphene, indicate that only 10–15% of the *REE* in the hornblende and chlorite zones is located in this accessory mineral. Thus it is considered that the *REE* patterns observed for the zonal sequence are dominated by those of the major mineral assemblages.

Since the sizes of the available structural sites in talc are too small to allow easy incorporation of any of the *REE*, their observed depletion in the central and talc zones may be readily explained (fig. 2). Interestingly, the depletion is most pronounced for those elements with intermediate ionic radii, suggesting an additional control on the substitution reactions involved. The fact that the central zone is in general less depleted than the talc zone may simply reflect the presence of more relict ultramafic minerals in the former. In amphiboles, Ca^{2+} occupies the approximately 9-fold coordinated M_4 site. Its ionic radius under such conditions is similar

to some of the trivalent *REE*, notably Nd^{3+} and Sm^{3+} (Shannon, 1976). Thus, preferential incorporation of these ions in the tremolite and hornblende zones might be one of the more important factors that have led to their convex-upward patterns (figs. 1 and 2). In chlorite, on the other hand, the *REE* are most likely to enter one of the octahedral sites, where the *HREE* and *MREE* (6-fold coordinated $Yb^{3+} = 0.868 \text{ \AA}$) are better suited to substitute for Fe^{2+} (0.78 \AA) than are the larger *LREE* ($La^{3+} = 1.03 \text{ \AA}$). Hence, in comparison with its country-rock predecessor, the chlorite zone is *HREE*-enriched and substantially *LREE*-depleted (La, Ce).

Thus, within the zonal sequence, crystal-chemical differences between the precursor assemblage and the zonal minerals are a major controlling factor on the final *REE* patterns.

Sphene was also observed in the altered gneiss around the pod, but at a much lower modal abundance than in the hornblende and chlorite zones. Its very low modal abundance and small size precluded a reliable analysis by microprobe. If the *REE* contents of this sphene are similar to those of the sphene in the pod then this mineral has not significantly affected the overall *REE* distribution in the gneiss. However, the major minerals—quartz, feldspar, and stringers of secondary antigorite—cannot be readily correlated with the observed *LREE*-enriched pattern, and it has not been possible to define the major controlling factor(s) on the *REE* pattern of the altered gneiss.

Conclusions

The *REE* were mobilized during metasomatic development of the zoned ultramafic pod. Varying proportions of all the *REE* were apparently lost during zone generation, but the *MREE* and *HREE* showed a greater mobility than did the *LREE*. This is in marked contrast to many other examples of *REE* mobility (notably Martin *et al.*, 1978) where *LREE* have been shown to be the most mobile. The fluid responsible was aqueous, probably not CO_2 -rich, and of relatively low oxygen fugacity. The resultant *REE* distribution may have been governed largely by the structural characteristics of the major zonal minerals in the pod.

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