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Rare Earth Element Concentrations in a Suite of Basanitoids and Alkali Olivine Basalts from Grenada, Lesser Antilles

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Abstract. A suite of basanitoids and alkali olivine basalts from Grenada, Lesser Antilles were analyzed for rare earth elements. The REE concentrations of these rocks are characterized by a small variation in the heavy REE (7 to 9 times chondrite) and a large variation in the light REE (17 to 93 times chondrite). Among the possible mechanisms to account for the REE variations, fractional crystallization processes at low and high pressures, and partial melting processes (both batch melting and fractional melting) were examined, using the partition relationships of REE among silicate minerals and melts. It is suggested that the observed REE variations are best explained by variable degrees of batch partial melting, in which garnet is present as one of the solid phases through 2 to 17% melting of a garnet lherzolite parent rock.

Introduction

Trace element abundances in volcanic rocks are parameters sensitive to varieties in petrogenetic processes. Gast (1968) has outlined the theoretical considerations necessary for the interpretation of trace element data in terms of major processes such as partial melting and fractional crystallization. The rare earth elements (REE) are one group of elements whose variations in a given suite of volcanic rocks can be explained, at least qualitatively, by partial melting and/or fractional crystallization models (e.g. Schilling and Winchester, 1969; Zielinski and Frey, 1970; Philpotts *et al.*, 1971; O'Nions and Clarke, 1972). Fractional crystallization models based on the phenocryst assemblages and on the partition relationships of REE between phenocryst mineral and magma have been particularly successful for volcanic rocks from Gough Island (Zielinski and Frey, 1970) and for those from Reunion Island (Zielinski, 1974). Application of the partial melting models (partiticularly partial melting of garnet lherzolite) have been difficult, however, due to the uncertainty of data on the partitioning of REE between garnet and liquid. For the same reason, eclogite fractionation (O'Hara and Yoder, 1967) has also been difficult to assess. The recent experimental determination of the REE partitioning between garnet and liquid at high pressures (Shimizu and Kushiro, 1975) has made it possible to assess the petrogenetic processes at high pressures in some detail in a semi-quantitative manner.

In this article, we present REE concentrations determined on a suite of basanitoids and alkali olivine basalts from Grenada, Lesser Antilles island arc,

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and evaluate both fractional crystallization and partial melting models based on the REE partitioning among phases involved.

Samples and Procedures

The island of Grenada is located at the southern end of the Lesser Antilles island arc and is characterized by volumetrically small amounts of andesitic rocks relative to the islands to the north (Arculus, 1973; in preparation) and by occurrence of strongly silica-undersaturated lavas (Sigurdsson *et al.*, 1973). The samples selected for REE analysis represent the range of basanitoid and alkali olivine basalt compositions suggested by Arculus (1973) to have been derived by various degrees of partial melting of an upper mantle source. Major and trace element (other than REE) compositions are given in Table 1 together with a brief petrographic description. The sample localities are given in the geologic map (Fig. 1).

REE were analyzed by an isotope dilution technique (Shimizu, 1974). The REE concentrations of standard BCR-1 agree with those reported in the literature to within 5%. Total blanks of the procedure (0.2 ng for Ce and 0.01–0.06 ng for other REE) are negligible in all cases. Isotope measurements were reproducible to within 0.5%. Overall, we consider the concentrations presented here to be reliable to 5%.

Results and Discussion

REE concentrations are summarized in Table 2 and illustrated in Fig. 2. The suite of samples are characterized by very little variation in the heavy REE (Er and Yb; 7 to 9 times chondrite) and a large variation in the light REE (Ce varies by about a factor of 6). There is no detectable Eu anomaly in any of the samples studied here.

Among the possible mechanisms to account for the REE variations, fractional crystallization processes at low and high pressures and partial melting processes (batch melting and fractional melting) are examined in the following sections, based on the partition relationships of REE among phases involved.

1. Fractional Crystallization at Low Pressures. Phenocryst minerals in these rocks are olivine, clinopyroxene, spinel and plagioclase (Table 1). Because the partition coefficients of REE observed for these minerals (e.g. Schnetzler and Philpotts, 1970: $D^{Ce}_{Ol}=0.009$, $D^{Yb}_{Ol}=0.023$; $D^{Ce}_{Cpx}=0.096$, $D^{Yb}_{Cpx}=0.227$; $D^{Ce}_{Pl}=0.06$, $D^{Yb}_{Pl}=0.02$ for An_{80}; see also Grutzeck *et al.*, 1973 for D_{Cpx}) are small except for spinel, whose effect is neglected in this paper, fractional crystallization of these phases generally results in an increase of REE concentrations in the residual liquid. Significant plagioclase crystallization can be ruled out by the absence of negative Eu anomaly. A point to be noticed here is that the partition coefficients for light and heavy REE differ by a factor of about two for both olivine and clinopyroxene, so that relative fractionation of REE in the residual liquid by crystallizing olivine and clinopyroxene would be limited (*relative fractionation* refers to the difference in fractionation between heavy and light REE). Therefore, the separation of olivine and clinopyroxene and subordinate amount of plagioclase from a magma would result in an essentially parallel upward shift of the REE pattern in the residual melts in a similar way to that observed by Zielinski and Frey (1970) for volcanic rocks from Gough Island and by Zielinski (1974) for those from Reunion Island. This process is obviously incapable of producing the observed REE variations (Fig. 2) from a common parental magma.

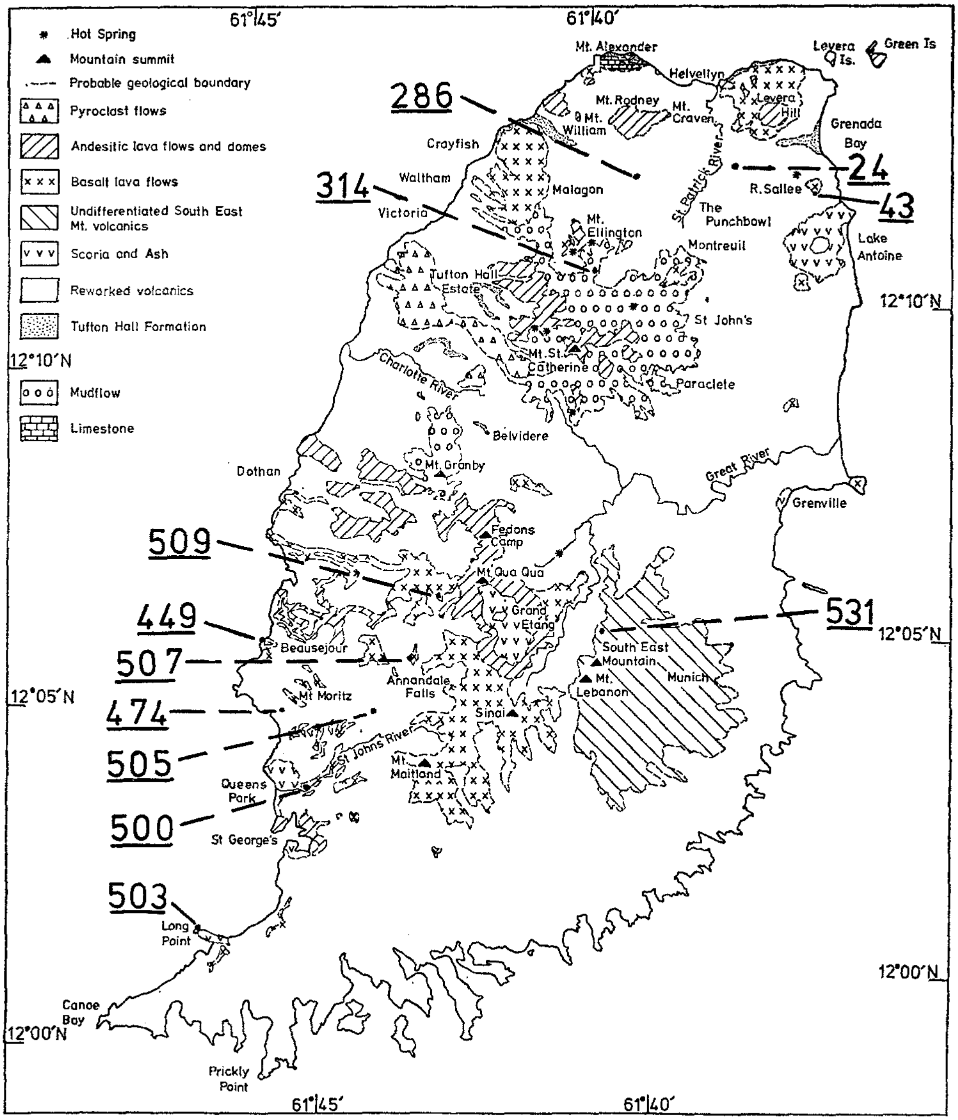

Fig. 1. Outline geologic map of Grenada with sample localities indicated by heavy numerals

2. *Fractional Crystallization at High Pressures.* If a parental magma undergoes fractional crystallization at high pressures, the effects of this process on the REE concentrations in the residual liquid may be different from those created at low pressures because of the appearance of phases stable at high pressures in the same magma. Possible high pressure phases include orthopyroxene, clinopyroxene and garnet for pressures of 20 to 35 kbar (e.g. Yoder and Tilley, 1962; Green and Ringwood, 1967; O'Hara, 1968). The fractionation of orthopyroxene has an effect

Table 1. Major and trace element analyses of Grenada basanitoids and alkali olivine basalts[a]

	474	500	24	314	507	286	505	43	449	503	531	509
SiO_2	46.50	46.45	46.95	46.22	46.46	46.82	46.39	45.98	46.74	45.60	45.14	45.07
Al_2O_3	16.31	17.27	17.60	15.65	16.45	17.62	16.66	16.55	17.62	15.91	16.08	15.49
Fe_2O_3	1.15	1.10	1.07	1.08	1.09	1.06	1.06	1.08	1.01	1.07	1.13	1.10
FeO	8.24	7.93	7.67	7.75	7.81	7.61	7.59	7.77	7.24	7.69	8.13	7.90
MgO	13.00	11.67	11.59	13.14	12.19	9.59	10.90	12.07	10.68	10.95	10.80	12.37
CaO	10.50	11.19	10.28	11.63	11.61	11.86	12.06	11.34	11.28	13.50	12.91	11.03
Na_2O	1.85	1.80	2.22	1.80	1.96	2.50	2.47	2.58	2.99	1.88	2.40	3.00
K_2O	0.33	0.48	0.59	0.48	0.40	0.71	0.53	0.49	0.42	1.01	0.92	1.46
TiO_2	0.88	0.91	0.83	0.93	0.82	0.89	1.00	0.80	0.82	0.95	1.03	1.06
P_2O_5	0.09	0.15	0.14	0.25	0.16	0.27	0.31	0.31	0.19	0.40	0.37	0.48
MnO	0.22	0.17	0.21	0.22	0.17	0.22	0.17	0.17	0.21	0.18	0.18	0.17
Or	1.93	2.82	3.45	2.81	2.34	4.18	3.11	2.86	2.45	5.94	5.40	8.47
Ab	16.45	16.05	19.71	15.04	16.35	16.40	14.93	14.26	16.93	6.65	6.42	5.27
An	34.89	37.38	35.92	32.84	34.58	34.57	32.51	31.67	32.92	31.84	30.18	24.03
Ne	0.00	0.00	0.00	0.56	0.65	3.55	4.22	5.14	5.71	6.08	8.99	12.70
Cpx	12.90	13.44	10.89	18.08	17.15	17.75	19.82	17.42	16.79	25.77	24.82	21.27
Opx	4.80	3.23	0.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ol	26.45	24.38	26.71	27.76	26.34	20.66	22.28	25.80	22.65	20.45	20.83	24.70
Mt	1.19	1.14	1.11	1.12	1.13	1.10	1.10	1.11	1.04	1.11	1.17	1.13
Il	1.21	1.26	1.14	1.28	1.13	1.23	1.38	1.10	1.13	1.32	1.42	1.45
Ap	0.19	0.31	0.29	0.52	0.33	0.56	0.64	0.64	0.39	0.83	0.77	0.99
Ba	108	114	192	454	181	474	229	177	500	345	414	448
Nb	7	3	8	12	2	14	12	2	10	9	11	14
Zr	62	64	73	104	68	102	109	68	73	123	126	212
Y	17	16	18	20	20	16	20	18	18	20	20	17
Sr	327	287	484	692	398	829	596	396	542	902	933	627
Rb	7	11	12	11	6	27	5	12	14	23	12	54
Zn	81	73	80	75	78	87	88	76	78	69	75	76
Cu	62	78	97	124	101	65	11	84	86	90	158	115
Ni	485	384	495	271	358	260	277	369	301	170	254	368

[a] The analyses are listed in order of increasing silica-undersaturation. Major and trace element analyses were completed by X-ray fluorescence (major elements in weight per cent, trace elements in ppm). Major element compositions were recalculated to 100% on a water-free basis. Normative compositions were calculated assuming an oxidation ratio of Fe_2O_3:FeO of 1:9.

Sample number, rock type and description:

474: Alkali olivine basalt. Microphenocrysts ($\leq$0.5 mm) of olivine (ol), clinopyroxene (cpx) and spinel (sp). Groundmass contains glass plus microlites of plagioclase (pl), cpx, ol and sp.

500: Alkali olivine basalt. Phenocrysts ($\leq$1.0 mm) of ol and sp. Trachytic textured groundmass contains pl, cpx, sp and ol.

24: Alkali olivine basalt. Phenocrysts ($\leq$2.0 mm) of ol, sp and sector-zoned cpx. Groundmass composed of pl, cpx, ol and sp.

314: Alkali olivine basalt. Phenocrysts ($\leq$2.0 mm) of ol, sector-zoned cpx, pl and sp in coarse-grained groundmass of same phases.

507: Alkali olivine basalt. Phenocrysts ($\leq$2.0 mm) of ol, sp and sector-zoned cpx. Groundmass contains microlites of ol, cpx, sp, pl and glass.

286: Alkali olivine basalt. Phenocrysts ($\leq$1.0 mm) of ol, cpx and sp. Groundmass consists of microlites of pl together with ol, cpx, sp and glass.

505: Basanitoid. Phenocrysts ($\leq$2.0 mm) of ol, sp and sector-zoned cpx. Fine-grained groundmass composed of pl, cpx, ol and sp.

43: Basanitoid. Phenocrysts ($\leq$2.0 mm) of ol, sp and sector-zoned cpx. Coarse-grained groundmass contains pl, ol, and sp.

449: Basanitoid. Phenocrysts ($\leq$1.0 mm) of ol, cpx and sp. One megacryst (3.0 mm) of strained ol present in section. Groundmass of pl, cpx, ol and sp.

503: Basanitoid. Phenocrysts ($\leq$2.0 mm) of ol, cpx, pl and sp. Coarse-grained groundmass composed of the same phases.

531: Basanitoid. Phenocrysts ($\leq$1.0 mm) of ol and cpx. Fine-grained groundmass composed of pl, cpx, ol and sp.

509: Basanitoid. Phenocrysts ($\leq$1.0 mm) of ol and cpx. Groundmass composed of sector-zoned cpx, ol, sp, pl and glass.

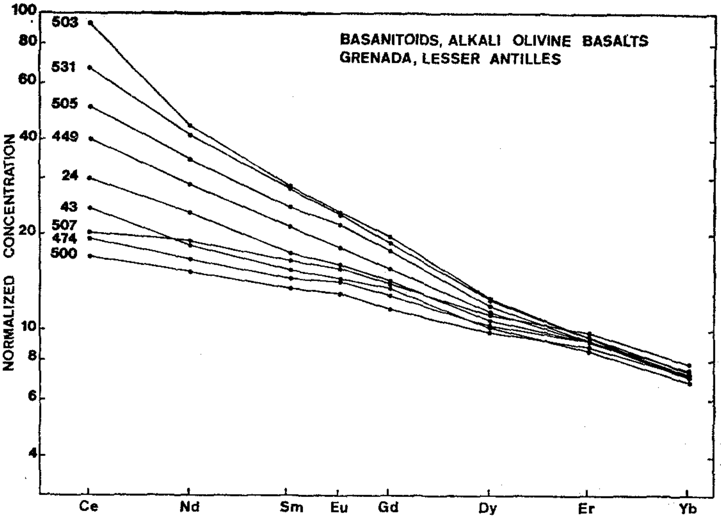

Fig. 2. Chondrite-normalized REE patterns of basanitoids and alkali olivine basalts from Grenada. Sample numbers (as in Table 2) are indicated on the left hand side. Normalizing factors are from Hubbard and Gast (1971)

Table 2. Concentrations of REE (ppm)

Sample No.	500	474	507	43	24	449	505	314	286	531	509	503
Ce	13.4	15.3	15.8	19.0	23.6	31.4	40.2	45.2	52.4	53.1	57.4	73.2
Nd	8.96	9.70	11.0	10.8	13.6	16.7	20.0	22.8	25.0	25.0	29.3	25.6
Sm	2.49	2.70	3.07	2.85	3.24	3.93	4.54	4.93	5.26	5.24	5.78	5.27
Eu	0.935	1.02	1.11	1.03	1.15	1.31	1.55	1.52	1.67	1.67	1.67	1.68
Gd	2.95	3.29	3.55	3.48	3.64	4.02	4.57	4.72	4.89	4.83	4.80	5.05
Dy	2.98	3.15	3.37	3.05	3.24	3.43	3.55	3.63	3.72	3.72	3.58	3.75
Er	1.78	1.88	1.99	1.74	1.74	1.88	1.88	1.84	1.91	1.92	1.86	1.93
Yb	1.66	1.69	1.81	1.58	1.70	1.70	1.65	1.73	1.68	1.64	1.65	1.70

similar to the phenocryst minerals mentioned above as the partition coefficients of REE between orthopyroxene and liquid are smaller than those of clinopyroxene (Schnetzler and Philpotts, 1970). However, the partition coefficients of REE between garnet and liquid (Shimizu and Kushiro, 1975; $D^{Ce} = 0.01$–0.02, $D^{Yb} = 4.0$) indicate that the separation of garnet or garnet-bearing assemblages would result in an increase of light REE and a strong depletion of heavy REE in the residual liquid and thus effectively change the slope of the REE pattern of the residual liquids.

Two different processes involving garnet crystallization were examined here. The first process consists of successive separations of garnet and one or more phases. Fig. 3A illustrates the principle of this process. Starting with the sample with the least enrichment of light REE (such as No. 500, Fig. 2), garnet is subtracted from this liquid until the residual liquid has $(Ce/Yb)_N$[1] $= 12.4$ (observed in

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[1] $(Ce/Yb)_N$ denotes concentration ratio after being normalized to chondrite.

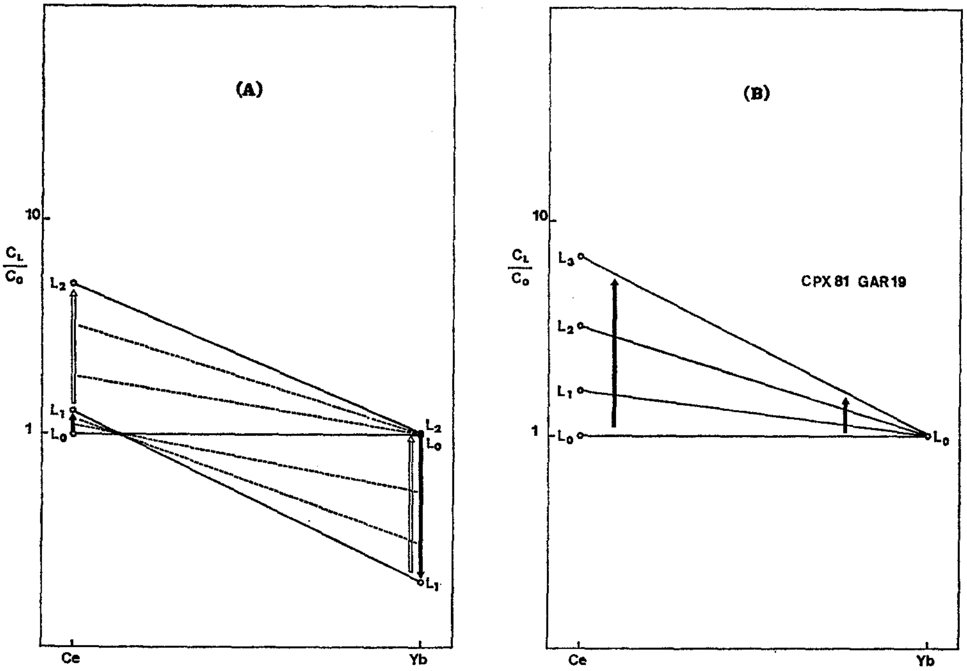

Fig. 3 A and B. Variations of the liquid REE patterns by fractionation of garnet-bearing assemblages. Enrichment relative to the initial concentrations is shown as C_l/C_0. (A) successive separations of garnet and other phases. $L_0(=C_0)$ denotes the initial liquid, L_1 the liquid after separation of garnet and L_2 the final liquid. Solid arrow from L_0 to L_1 shows the effect of garnet separation, open arrow from L_1 to L_2 the effect of other phases. (B) fractionation of eclogite (clinopyroxene 81% and garnet 19%). L_0 denotes the initial liquid, L_3 the residual liquid after 82% subtraction. L_1 and L_2 denote the residual liquids after subtracting smaller amounts of eclogite. Note that Yb stays constant for this particular eclogite fractionation

the sample most enriched in the light REE such as No. 503, Fig. 2). Then other phases are subtracted to bring the heavy REE concentrations back to the initial level. In Fig. 3A, only the enrichment relative to the initial concentrations are shown. Using the partition coefficients given above (Shimizu and Kushiro, 1975) and the Rayleigh fractionation equation, 37% of the initial liquid is subtracted as garnet, resulting in the residual liquid pattern with $(Ce/Yb)_N = 12.4$ (this liquid pattern is qualitatively shown as L1 in Fig. 3A). In order to shift this pattern back to the initial level, an additional 75% of the residual liquid must be crystallized as other phases. For samples with smaller $(Ce/Yb)_N$, smaller amounts of garnet and additional phases are to be subtracted (shown as dashed lines in Fig. 3A). The normative compositions of these rocks (Arculus, 1973) show that the rocks with small light REE enrichment (such as No. 500) have higher anorthite and olivine components than those with a large light REE enrichment (such as No. 503). If the composition of liquidus garnet could be approximated by 1:1 (by mole) mixture of normative anorthite and olivine, the normative compositions of these rocks appear to be consistent with this model. However, it would be quite fortuitous if this two-stage process resulted in almost constant heavy REE concentrations as observed in our samples.

The second process examined is "eclogite fractionation" proposed by O'Hara and Yoder (1967) and O'Hara (1968) as an effective process by which trace element concentrations can be significantly changed without greatly changing the major element composition. The proportions of garnet and and clinopyroxene of "eclogite" would be a function of pressure and temperature. In the present calculation, the proportions of garnet and clinopyroxene were selected so as to obtain the bulk partition coefficient for heavy REE (Yb) of 1. By fractionating this particular eclogite (consisting of 81% clinopyroxene and 19% garnet, using the above partition coefficients), the heavy REE concentrations in the residual liquid could be held constant, while the light REE concentrations could be successively increased. Fig. 3B shows the principle of this process. Starting again with the REE pattern of No. 500, 82% of the initial liquid is crystallized as eclogite to obtain the REE pattern of No. 503 by the Rayleigh fractionation of the eclogite. It is expected that eclogite fractionation would result in a decrease of normative albite which is potential jedeite component in clinopyroxene and of normative diopside. Although the amount of normative albite (and nepheline) is lower in No. 503 than in No. 500, the former is more enriched in normative diopside than the latter. Eclogite fractionation, therefore, seems inconsistent with the major element compositions of these rocks. If the proportions of garnet and clinopyroxene are 1:1, which appears more or less typical for eclogite inclusions in kimberlites, the situation is similar to the one mentioned previously (Fig. 3A), because the bulk partition coefficient for Yb is around 2 so that eclogite fractionation results in depletion of the heavy REE in the residual liquid. A two-stage process similar to the one mentioned above should therefore be considered and again it would be fortuitous to have almost constant heavy REE concentrations for the series of rocks as products of the two-stage process.

3. Partial Melting of Garnet Lherzolite. Two different types of partial melting processes are examined here, i.e. batch partial melting and fractional partial melting. In the batch melting, the liquid and the bulk solid are in equilibrium. The model calculations require an estimate of the proportions of phases in the initial solid and also those in equilibrium with liquid at a given degree of partial melting. Theoretically, the proportions and compositions of phases at any stage of equilibrium partial melting would be unique for a given starting material and conditions. However, no experimental data are available for the variations of phase proportions during partial melting, so that the model calculations cannot give a unique solution. The calculations were therefore made here to test whether it is possible to obtain the two extreme REE patterns observed here (such as No. 500 and No. 503) from a common source material by changing the degree of partial melting and hence changing the phase proportions of solid in equilibrium with liquid. In the calculation, the formulation of Gast (1968) was used with a modification similar to that discussed by Shaw (1970). The partition coefficients used were (for Ce and Yb): $D^{Ce}_{Cpx}=0.1$; $D^{Yb}_{Cpx}=0.23$ (Schnetzler and Philpotts, 1970; Grutzeck *et al.*, 1973), $D^{Ce}_{Gar}=0.01$; $D^{Yb}_{Gar}=4.0$ (Shimizu and Kushiro, 1975). Olivine and orthopyroxene were treated together with the partition coefficients

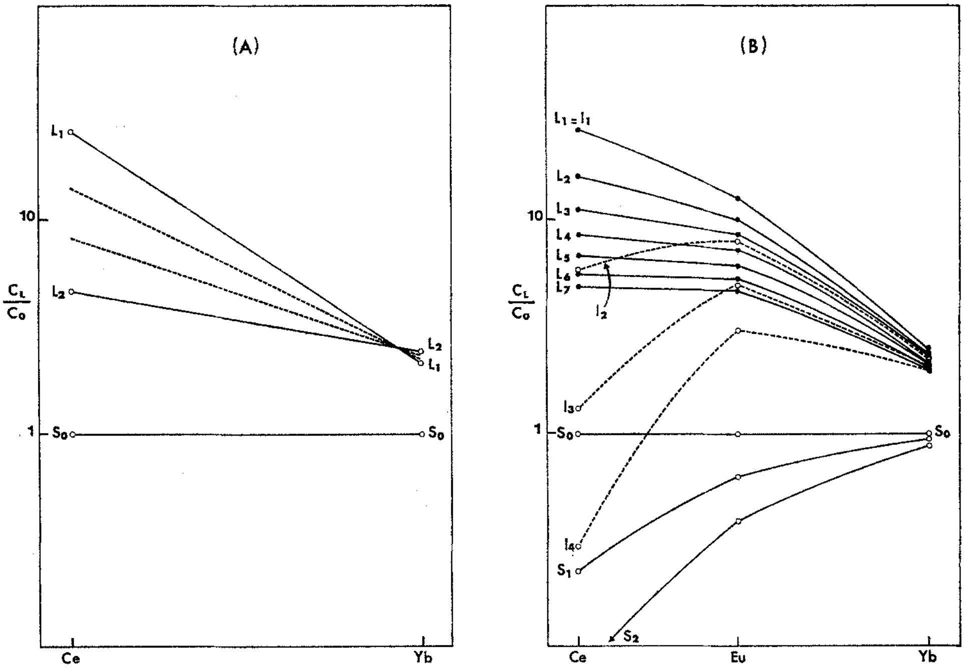

Fig. 4A and B. Variations of the liquid REE patterns by partial melting of garnet lherzolite. Relative enrichment is shown as Cl/C_0. (A) batch melting. $S_0 (= C_0)$ denotes the original solid. $L_1 = 2.2\%$ melting and $L_2 = 17\%$ melting, see text. (B) fractional melting. l_1, l_2, etc denote liquid increment of each stage. L_1, L_2, etc denote liquids in the reservoir. S_1 and S_2 are solid residua left after first and second stage melting

of 0.003 for Ce and 0.007 for Yb, based on Cpx/Opx and Cpx/Ol REE ratios reported by Philpotts *et al.* (1972). The calculated liquid must satisfy (1) $(Ce/Yb)_N$ ratios observed and (2) constant heavy REE concentrations. Fig. 4A illustrates the results of this calculation.

Starting with a garnet lherzolite with 10% garnet and 10% clinopyroxene (these proportions are typical for the sheared garnet lherzolite nodules in kimberlites from northern Lesotho, Boyd and Nixon (1972)) and with a chondritic relative REE pattern, the sample with the highest light REE enrichment can be modeled by 2.2% melting with 1% clinopyroxene, 0.5% garnet and 0.7% olivine and orthopyroxene entering the melt. On the other hand, the sample with the smallest light REE enrichment can be modeled by 17% melting with 4.8% clinopyroxene, 5.7% garnet and 6.5% olivine and orthopyroxene entering the melt. For these two liquids, the above requirements are satisfied. The phase proportions in the melt suggest that the first liquid is more enriched in normative diopside than the second, whilst the second liquid is more enriched in normative olivine than the first. These features are consistent with the normative compositions of the samples.

It is emphasized that the important factor maintaining almost constant heavy REE concentrations in the liquids generated is the presence of garnet during 2 to

17% partial melting. Melting experiments of a natural garnet lherzolite (e.g. Ito and Kennedy, 1967) show that both clinopyroxene and garnet disappear in a small temperature interval (around 50° C) above the solidus under dry conditions. If this is the typical circumstance for partial melting in nature, the Grenada magmas may have been produced in a very small temperature range (say about 30° C). Under hydrous conditions, the temperature interval in which both clinopyroxene and garnet are in equilibrium with liquid is increased to several hundred degrees (e.g. Mysen, 1973).

Fig. 4B illustrates another type of partial melting (fractional melting as described by Gast, 1968). In this process, liquid produced is immediately removed from the system and stored in the reservoir. The residue left after a stage of melting becomes a parental material for the next-stage melting. The resultant volcanic magmas erupted on the surface are considered as representatives of the reservoir at various stages. In the present case, the same starting material as the batch melting case is used and 3% melting is assumed for each increment of the liquid. As shown in Fig. 4B, the second and third stage increments are already depleted in Ce relative to Eu due to the strong depletion of light REE in the residual material as discussed by Gast (1968). However, the liquid in the reservoir is not very sensitive to the pattern of later-stage liquids because the contribution of the later-stage increment is of diminishing importance.

In order to produce the observed range of Ce (a factor of 5.5), seven successive stages were required. Compared with the batch melting model, this process encounters several minor difficulties such as: (1) the variation for Yb is larger (~20%), (2) Ce/Eu ratio of L7 is lower than that in the sample No. 500, and (3) Eu/Yb ratio of L7 is higher than No. 500. These difficulties may be removed by changing the phase proportions in the melt. In the calculation, however, constant phase proportions were assumed, which is clearly not consistent with the variations of normative compositions of the rocks. Therefore, the phase proportions should be changed in such a way that the liquid in the reservoir in the later stage has more olivine and orthopyroxene components than the earlier stage. This would be inevitable in fractional melting of a garnet lherzolite source (O'Hara, 1968). The successive modifications of the phase porportions to satisfy the normative compositions of the observed lavas then becomes incompatible with the observed REE patterns. REE concentrations (constant heavy REE abundances and variable Ce/Yb ratios) are not predicted as the melt of increasing olivine and orthopyroxene proportions in the melt.

Comparison of the calculations together with the normative composition of the rocks suggests that the observed REE variations may be best explained by various degrees of batch partial melting of a garnet lherzolite parent.

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