

## Differentiation Trend of Rare Earth Elements of the Skaergaard Intrusion

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### Skaergaard 암체의 회토류의 분화경향

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Skaergaard 암체는 광범한 *in situ* 화성분화작용을 겪은 층상관입암의 대표적인 예로 널리 알려져 있다. 따라서 이 암체는 폐쇄계에서 미량원소변이를 modeling 할 수 있는 적지가 될 것이다. 그러나, 기존의 연구에 의하면 본 암체의 전암 및 광물의 회토류는 폐쇄계로서의 예상된 경향을 보이지 않는다(Haskin and Haskin, 1968; Paster *et al.*, 1974). 발표된 분배계수, 공존광물들의 mode, 그리고 전암 및 광물의 미량원소함량을 사용한 미량원소modeling에 의하면 Skaergaard 암체의 회토류는 분화후기에 일어난 광범한 인회석 결정작용에 크게 영향을 받았음을 보여준다. 미량원소modeling은 Upper Border Series에서 간헐적으로 나타나는 인회석이 기존에 주장되어 온 바와 같은 liquidus상이 아니라 진화하는 magma의 미량원소함량에는 영향을 끼칠 수 없는 interstitial phase임을 시사한다. 분화작용 말기에 Skaergaard 마그마가 대류를 멈추거나 소규모로 대류를 할 때, 마그마 암장의 상부에 축적되는 휘발성분에 기인한 증가된  $\text{PH}_2\text{O}$ 가 인회석이 UBS에서 정출 되는 것을 방해하였을 것이다. 이와 같은 인회석의 특성을 고려해서 Skaergaard 분화작용을 modeling하면 회토류는 폐쇄계로서의 예상된 경향을 따른다. 이와 같은 결과는 최종 20% 분화작용기간 중에 양적으로 상당한 양의 마그마의 주입이나 분출을 수반하는 그 어떤 Skaergaard 암체의 분화model도 배격한다.

주요어 : 분화, 미량원소 모델링, 회토류 원소, Skaergaard

The Skaergaard intrusion is widely considered a type example of a strongly fractionated, layered intrusion that has undergone extensive *in situ* igneous differentiation. The intrusion, therefore, should be a good locality for modeling trace element variation in a closed system. Previous studies (Haskin and Haskin, 1968; Paster *et al.*, 1974), however, have suggested that the rare earth elements in whole rocks and mineral separates from the intrusion did not follow the expected trend for closed system crystallization. Trace element modeling using published distribution coefficients, modal abundances of the coexisting minerals, and the concentration of trace elements in whole rocks and mineral separates from the Skaergaard intrusion, reveals that the rare earth elements were significantly influenced by the crystallization of abundant apatite in the Layered Series during the final stages of crystallization. The results of trace element modeling also suggests that apatite, which appears sporadically in the UBS, is not a primary liquidus phase in these samples as previously suggested (Naslund, 1984) but an interstitial phase that did not directly effect trace element abundances in the evolving magma. As the Skaergaard magma ceased convection, or convected as small isolated cells during the final stages of differentiation, an elevated  $\text{P}_{\text{H}_2\text{O}}$  induced by accumulation of volatile elements near the roof of the magma chamber inhibited or delayed the precipitation of primary apatite in the UBS. If the Skaergaard differentiation is modeled assuming primary apatite crystallization in the upper part of the LS where abundant modal apatite is present, and only late stage crystallization of apatite in the UBS where apatite is less abundant, rare earth elements abundances follow a closed system variation trend. These results rule out any differentiation model for the Skaergaard intrusion that includes volumetrically significant injections or discharges of magma into or out of the chamber during the final 20% of the crystallization history.

Key words : differentiation, trace element modeling, rare earth elements, Skaergaard

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## 1. INTRODUCTION

Since its first description by Wager and Deer (1939) the Skaergaard intrusion has become one of the most intensely studied igneous bodies in the world. The intrusion can be divided into three main units: a Layered Series (LS), which accumulated on the floor of the magma chamber; a Marginal Border Series (MBS), which crystallized on the sides of the chamber; and an Upper Border Series (UBS), which crystallized under the roof of the chamber (Wager and Brown, 1967). Detailed geological descriptions of the intrusion are available in the literature (LS-McBirney (1989), MBS-Hoover (1989), and UBS-Naslund (1984)). The intrusion is widely considered the type example of a strongly fractionated, layered intrusion that has undergone extensive in situ igneous differentiation, and, therefore, it should provide one of the best examples of trace element fractionation in a closed system. Previous attempts at trace element modeling of the intrusion (Haskin and Haskin, 1968; Paster *et al.*, 1974), however, failed to demonstrate the expected trend of trace element enrichment in a closed system. This unexpected result is one of the reasons why it has been suggested that the Skaergaard intrusion was an open system during the final stages of differentiation (Hunter and Sparks, 1987, 1990; Stewart and DePaolo, 1990).

Skaergaard rocks are cumulates and, therefore, represent mixtures of minerals and melt in unknown ratios. Trace element compositions of whole rock samples are a function of the modal abundance of minerals. Some trace elements are predominately controlled by specific minerals, for example, Ni by olivine, V by magnetite, Cr by pyroxene, and P by apatite. Abrupt changes of these elements in whole rock sample during differentiation of the Skaergaard intrusion closely correspond to the appearances or disappearances of specific minerals (Jang, 1999; Jang and Naslund, 2001). The trace element compositions of individual minerals such as pyroxene and plagioclase, however, are much less subject to variations due to modal abundances in individual samples, and are more useful for modeling the trace element variation in the Skaergaard magma.

In general, most excluded trace elements appear

to follow typical Rayleigh-type trace element trends with differentiation of the Skaergaard intrusion (Jang, 1999; McBirney, in review; Tegner, in review). Significant deviations from the Rayleigh trend for the rare earth elements in both whole rock and individual mineral samples, however, have been observed at the UZa/b boundary of the Layered Series. The rare earth element content increases in whole rock samples and significantly decreases in mineral separates, such as plagioclase and pyroxene at the boundary (Haskin and Haskin, 1968; Paster *et al.*, 1974; Goles, personal communication; Jang and Naslund, 2001; McBirney, in review). The departure in the REE trend coincides with the appearance of extensive liquidus apatite. The effect of apatite crystallization on rare earth elements in liquids has been studied experimentally and the strong preference of apatite for rare earth elements is well known (Watson and Green, 1981). The UBS, which also contains apatite, however, does not show an abrupt change in REEs at this stage of differentiation. The major purposes of this paper are: 1) to model the rare earth element variation trend of the Skaergaard magma, 2) to delineate the effect of apatite on trace element trends in the LS, MBS, and UBS, and 3) to determine if trace element modeling can resolve the debate over closed system versus open system crystallization of the Skaergaard intrusion.

## 2. TRACE ELEMENT MODELING

### 2.1. Background theory

Geochemical studies of trace element variation in igneous systems have proven superior in many respects to major element studies in illuminating the changes in liquid composition that occur as the differentiation of a magma proceeds. Previous studies have shown that Henry's law can be sustained up to the region of several weight percent of trace element concentration. If liquid-crystal systems are at equilibrium, elements are partitioned among phases according to their activities in those phases. For trace elements, we normally assume Henry's law behavior,

$$a_i = h_i \cdot X_i,$$

where  $a_i$  is the activity of component  $i$ ,  $h_i$  is the Henry's law constant, and  $X_i$  is the mole fraction of

component *i*. The distribution behavior of a trace element between a melt and a coexisting crystal will be largely independent of concentration and can be described by a distribution coefficient ( $K_{Di}$ ), where:

$K_{Di}$  = concentration of element *i* in crystal / concentration of element *i* in melt.

Most geologic systems of interest are multi-phase systems, involving more than one mineral phases and a melt. Each phase will have a different distribution coefficient for a specific element. To estimate the combined effect of removing an assemblage of liquidus phases, a bulk distribution coefficient (*D*) can be calculated from the weight proportions of each mineral phase ( $W_i$ ), multiplied by the distribution coefficient ( $K_{Di}$ ) of each mineral phase:

$$D = \sum W_i K_{Di}$$

For a differentiating magma, it is necessary to derive an expression that describes the changes in the concentrations of trace elements during solidification. There are two ideal cases, perfect equilibrium crystallization and perfect fractional crystallization. They are defined as,

$$X_i^{LQ}/X_i^{int} = 1 / \{D*(1-F) + F\}$$

for perfect equilibrium crystallization, and

$$X_i^{LQ}/X_i^{int} = F^{D-1}$$

for perfect fractional crystallization,

where  $X_i^{LQ}$  is the fraction of component *i* in the remaining liquid,  $X_i^{int}$  is the fraction of component *i* in the initial magma, and *F* is the fraction of liquid remaining.

### 2.2. Procedure for calculating Skaergaard trace element trends

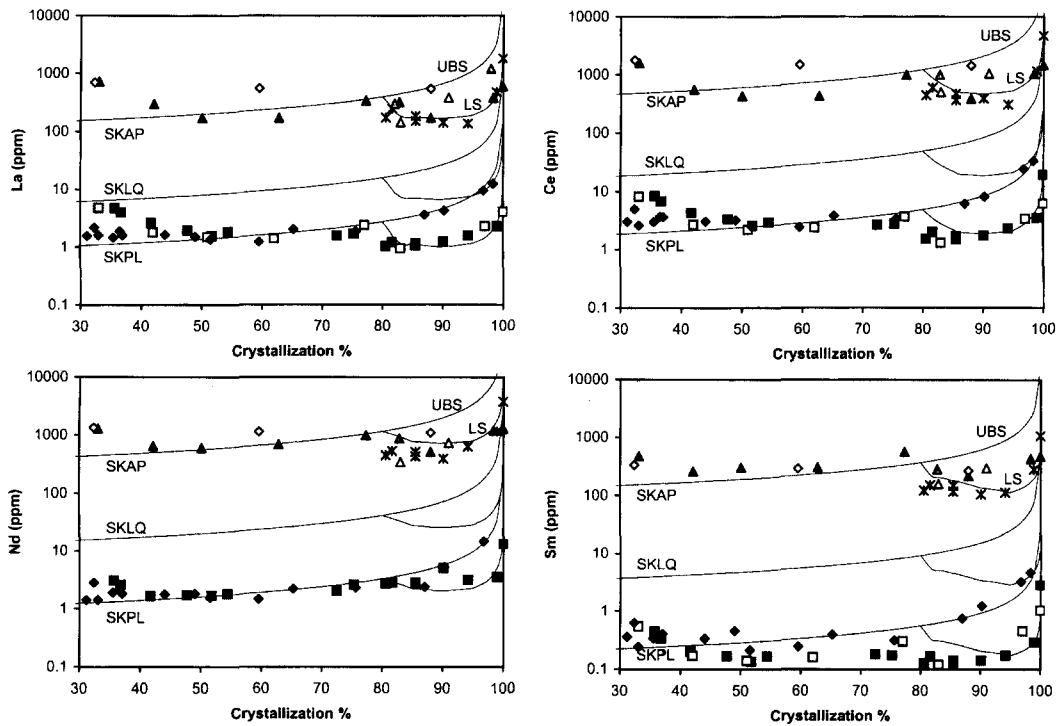
Using published  $K_{Di}$  for the rare earth elements (Arth, 1976; Pearce and Norry, 1979; Henderson, 1982; Nash and Crecraft, 1985; Rollinson, 1992; Hart and Dunn, 1993; Dunn and Sen, 1994), the concentration of trace elements in whole rock samples and mineral separates, and the modes of coexisting mineral phases in the Skaergaard intrusion (Jang, 1999; Jang and Naslund, 2001, McBir-

ney, in review), rare earth element trends in the differentiating magma, and in the crystallizing mineral phases (plagioclase and apatite) can be calculated (Fig. 1 and Table 1). There have been many studies to determine distribution coefficients in experimental charges, and in natural rocks containing phenocrysts and matrix, and as a result the range of measured distribution coefficients for some trace elements in certain minerals is pretty wide. In principle, distribution coefficients vary with compositions of the liquid and crystal, and with temperature, and pressure. Changes in pressure in the Skaergaard intrusion, however, are probably minimal during differentiation. Distribution coefficients selected for modeling were chosen from studies that were as close as possible to the Skaergaard in terms of the compositions of the liquid and crystals, and the temperature and pressure of crystallization.

The calculations were made using two assumptions. The first was that the Skaergaard intrusion did not experience any major injections and/or discharges of magma after the initial filling of the chamber. Such departures from closed system conditions could result in significant changes in the composition of the magma and the minerals crystallizing from it. A number of recent studies based on field evidence and major element mineral compositions (McBirney and Naslund, 1990; Jang and Naslund, 2001; McBirney, in review; Tegner, in review) rule out the possibility of major injections or discharges of magma from the intrusion. The other assumption is that the intrusion followed almost perfect fractional crystallization. Both of these assumptions will be tested by how well the observed mineral and whole-rock compositions fit the calculated trend.

### 3. DISTRIBUTION OF SKAERGAARD APATITE

Apatite in the Skaergaard intrusion is a distinctive minor mineral that occurs as a liquidus phase after the Skaergaard magma become saturated with  $P_2O_5$  (Wager and Brown, 1967; Watson, 1979). Skaergaard Apatite became an abundant phase in UZb of the Layered Series (LS) and in the UZb-equivalent unit of the Marginal Border Series (MBS). In the Upper Border Series (UBS), however, it appears sporadically as a coarse, abundant



**Fig. 1.** Rare earth element modeling of Skaergaard plagioclase using published mineral/magma  $K_{D_i}$ 's, assuming that the layered series magma precipitated 8% apatite from 80 to 85% crystallization, 4% apatite from 85 to 90%, 2.5% apatite from 90 to 95%, and 1.0% apatite from 95 to 100% (c.f. Table 1). UBS plagioclase does not show any apatite effect, but instead follow the calculated Rayleigh trend for apatite-free crystallization. LS plagioclase-filled squares (Jang and Naslund, 2001) and open squares (Goles, personal communication); UBS plagioclase-filled diamonds (Naslund, 1980); LS apatite-Stars (this study), filled triangles (Nash, 1976), shaded triangles (Paster *et al.*, 1974), lightly shaded triangles (Wager and Mitchell, 1951), and open triangles (McBirney, in review); UBS apatite-open diamonds (Naslund, 1980). SKLQ-calculated liquid trend; SKAP-calculated apatite trend; SKPL-calculated plagioclase trend; LS and UBS indicate the calculated LS and UBS trends after the UZa/UZb boundary (see text). Both UBS and LS plagioclase follow similar trends below the UZa/UZb boundary.

phase throughout the crystallization sequence. It has been suggested that apatite was an intermittent liquidus phase in the roof-zone of the intrusion (Naslund, 1984). Euhedral to subhedral apatite comprises up to 8% by mode of the UZb of the LS and the UZb-equivalent rocks of the MBS, and then steadily decreases in modal abundance with differentiation (Wager and Brown, 1967; Jang, 1999; Tegner, in review). Apatite, occurs as a trace accessory phase in the LS and MBS below UZb and its equivalent of the MBS, and is considered to be an interstitial phase precipitated from residual liquids trapped in these units.

Apatite has a strong preference for rare earth elements, with  $K_{D_i}$ 's ranging from 20 to over 60 (Arth, 1976; Pearce and Norry, 1979; Henderson, 1982; Nash and Crecraft, 1985; Rollinson, 1992;

Hart and Dunn, 1993; Dunn and Sen, 1994). In addition, it has a characteristic M shaped rare earth element pattern. The rare earth elements with the exception of Eu are incompatible elements in all of the major mineral phases of the Skaergaard intrusion except apatite, and as a result their abundance in both individual minerals and whole rock samples follows a typical logarithmic enrichment trend with differentiation until apatite becomes a liquidus phase. Apatite first appears as a major phase at the boundary between UZa and UZb of the LS, and at the equivalent stage in the MBS, and as a result, its influence on rare earth element abundances and patterns in both whole rock samples and mineral separates should be apparent in samples formed after this level. In Skaergaard whole rock samples above the UZa/b boundary over 90%

**Table 1.** (a) Distribution coefficients (KDi) used for REE modeling of Skaergaard plagioclase and pyroxene.

	Plagioclase	Olivine	Pyroxene	Oxides	Apatite
La	0.17	0.007	0.38	0.09	25
Ce	0.1	0.007	0.39	0.09	25
Nd	0.08	0.006	0.41	0.09	28
Sm	0.06	0.007	0.59	0.09	40

**Table 1.** (b) Bulk Distribution Coefficients (D) calculated using KDi and the mode of Skaergaard whole rock samples.

X1%	La	Ce	Nd	Sm
0-80	0.25	0.22	0.21	0.28
80-85	2.17	2.14	2.37	3.38
85-90	1.17	1.04	1.25	1.77
90-95	0.85	0.79	0.88	1.25
95-100	0.47	0.45	0.47	0.66

X1%=Crystallization percent calculated as explained in text.

**Table 1.** (c) Mode used for REE modeling of Skaergaard plagioclase and pyroxene.

X1%	Plagioclase	Olivine	Pyroxene	Oxides	Apatite	Others*
0-80	50.0	5.0	40.0	5.0		
80-85	49.0	15.0	20.0	8.0	8.0	
85-90	43.5	21.5	22.0	9.0	4.0	
90-95	43.0	14.0	36.0	4.5	2.5	
95-100	30.0	15.0	40.0	5.0	1.0	9.0

X1%=Crystallization percent calculated as explained in text.

Others\* include myrrmekite, quartz, etc.

**Table 1.** (d) Calculated REE values of apatite by inventory method.

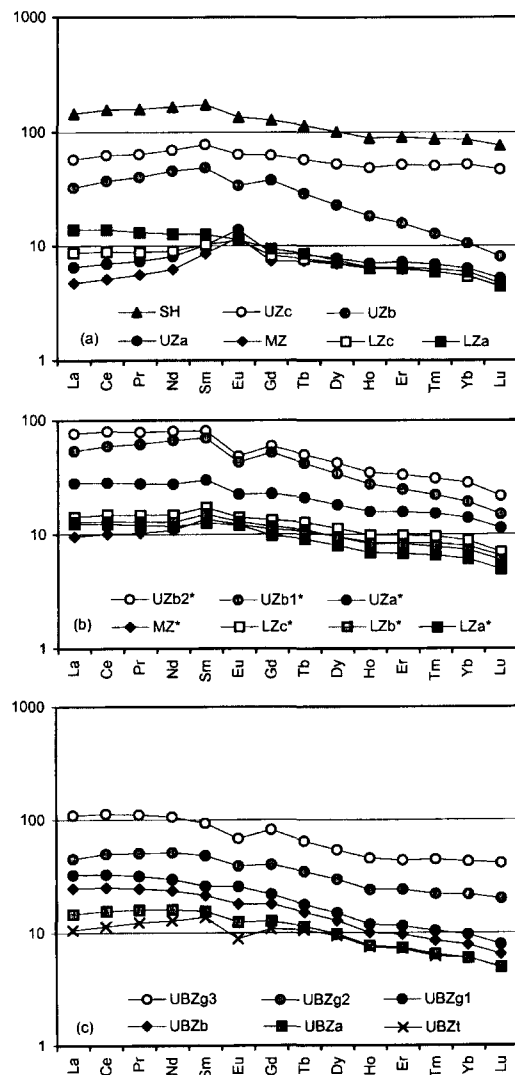
X1%	La	Ce	Nd	Sm
81	172	437	448	122
82	247	592	528	151
86m	150	359	428	117
86f	184	466	509	147
90	142	387	393	105
94	136	303	633	113
99	472	1138	1159	279
100	1813	4712	3791	1064

X1%=Calculated crystallization percent.

86 m and 86f represent mafic and felsic samples collected from the same stratigraphic horizon.

of the rare earth element content is stored in apatite, and hence the rare earth element pattern of apatite dominates over other mineral phases. As expected, the first appearance of abundant, coarse apatite in the LS coincides not only with a sudden increase in rare earth elements abundance in whole rock samples but also with a change in the rare earth element pattern to one which resembles the

characteristic M shaped pattern of apatite (Fig. 2). Similar rare earth element trends are observed in whole rock samples from the MBS. Samples from the UBS, however, do not show any abrupt changes in rare earth elements at the same level of crystallization, suggesting that apatite in the UBS is not a liquidus phase as originally suggested by Naslund (1984), but rather that UBS apatite pre-



**Fig. 2.** Chondrite normalized rare earth element patterns of whole rock samples from the LS (a), MBS (b), and UBS (c) of the Skaergaard intrusion (Jang, 1999; McBirney, in review). REE patterns of the LS significantly change at the UZa/UZb boundary, where extensive apatite crystallization occurs. Note the characteristic M-shape pattern of the UZb samples, which suggest a significant apatite effect. UBS samples, however, do not show any apatite effect in their REE patterns.

precipitated as an interstitial phase from trapped liquid.

Compared to mode dependent whole rock samples, individual mineral separates from the Skaergaard intrusion show much more systematic trace element trends with differentiation. Rare earth ele-

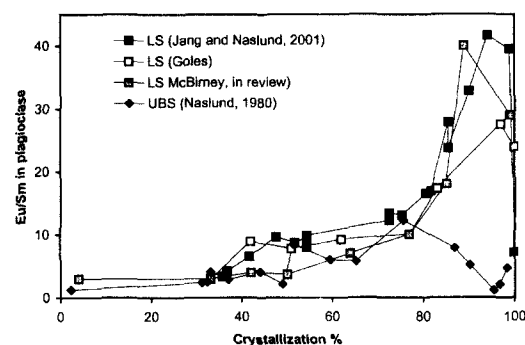
ment contents in LS plagioclase and pyroxene, which generally increase with differentiation of the intrusion, abruptly decrease at the UZa/b boundary, where abundant apatite first appears (Jang and Naslund, 2001; Jang and Naslund, in review; McBirney, in review). The rare earth element trend in LS plagioclase can be modeled using published apatite/magma  $K_{DjS}$ , assuming that the layered series magma precipitated 8% apatite from 80 to 85% crystallization, 4% apatite from 85 to 90%, 2.5% apatite from 90 to 95%, and 1.0% apatite from 95 to 100% (abundances in accord with observed modes for Layered Series samples) (Fig. 1). Crystallization %s are calculated assuming 0% crystallization at the Chilled Margin, 30% crystallization at the base of LZa, and 100% crystallization at the Sandwich Horizon. Relative volumes of the unit are assumed to be proportional to their stratigraphic thickness.

UBS plagioclase, however, follow a logarithmic enrichment trend for rare earth elements without any abrupt decrease at the equivalent stage to that observed in the LS. As a result, UBS plagioclase and pyroxene are more enriched in rare earth elements than LS plagioclase and pyroxene, at late stages of differentiation. The rare earth element pattern of UBS plagioclase is similar to the theoretical pattern calculated assuming the intrusion underwent Rayleigh fractionation with no apatite crystallization. In addition, the rare earth element patterns of UBS whole rock samples do not show the characteristic M-shaped pattern of apatite (Jang, 1999; McBirney, in review), and hence, show no significant apatite effect. These results suggest that apatite is not a primary phase in the UBS but an interstitial phase that does not significantly affect the rare earth element contents in associated UBS plagioclase or in the magma crystallizing at the roof. Previous attempts to model the partitioning of the rare earth elements in the Skaergaard intrusion (Haskin and Haskin, 1968; Paster, *et al.*, 1974) were unsuccessful, because the models did not consider the significant effect of apatite crystallization on the trend of rare earth element in both liquidus mineral phases and the coexisting Skaergaard magma, and did not recognize the contrasting effects of apatite at the floor and roof of the magma chamber.

#### 4. APATITE CRYSTALLIZATION AND CONVECTION

In terms of rheological experiments, thermodynamic calculations, and field evidence, such as magma current induced structures, convection is one of the most basic phenomena in basaltic magma chambers. Igneous layering is an almost ubiquitous feature in the Skaergaard intrusion. Although a number of types of layering have been described and numerous mechanisms have been proposed for their formation (c.f. Naslund and McBirney, 1996), convection related magma currents are one of the most commonly proposed causes for forming layering structures. It has been long recognized that the well developed layering in the LS suddenly disappears in UZb at a late stage of differentiation of the Skaergaard magma. In addition to the abrupt disappearance of layering structures, significant lateral mineralogical variations have also been reported in the rocks formed during the late stages of differentiation of the UBS and LS (Douglas, 1961; Naslund, 1977). An end to convection at the UZa/b boundary could explain the disappearance of layering, the development of lateral inhomogeneities, and divergence of LS and UBS rare earth element trends at this level.

Although UBS plagioclase separates are significantly enriched in rare earth elements relative to LS plagioclase during the final stages of differentiation, Eu in UBS plagioclase does not show a late stage enrichment but rather follows a trend similar to that in LS plagioclase (Jang and Naslund, 2001). The ratios of Eu/Sm in separated plagioclases from the LS and UBS show similar trends with differentiation for the first 80% of crystallization, until at the UZa/UZb boundary of the LS, and its equivalent in the UBS, the trends abruptly diverge (Fig. 3). Eu/Sm increases with differentiation during the final stage of crystallization in LS plagioclase separates and decreases in UBS plagioclase separates. The negative relationship between  $\text{Eu}^{2+}$  and oxygen fugacity,  $f\text{O}_2$ , and its effect on plagioclase and melt partitioning is well known (Drake and Weill, 1975). The observed trends suggest that the floor sequence may have become more reducing and the roof sequence more oxidizing with differentiation during the final 20% of crystallization. Alternatively, the Eu trend



**Fig. 3.** Eu/Sm in Skaergaard plagioclase plotted against percent of crystallization. Eu/Sm in both LS and UBS plagioclase gradually increases with differentiation of the intrusion, and then the two trends abruptly diverge at the UZa/UZb boundary, where extensive apatite crystallization occurs in the LS. The UBS magma apparently evolved separately from the LS magma after this point, suggesting that the Skaergaard magma may have ceased convection at that time.

was dominated by the crystallization of plagioclase, and therefore, was similar at the floor and roof while the Sm trend was dominated by apatite and therefore, was different at the floor and roof.

It is proposed that during the initial 80% of crystallization, convection within the Skaergaard magma chamber resulted in similar magma differentiation trends at the roof and floor.  $\text{P}_2\text{O}_5$  is an important factor in controlling viscosity of basaltic magma, and causes a significant increase in viscosity (Toplis *et al.*, 1994).  $\text{P}_2\text{O}_5$ , which was saturated at this stage of differentiation, could have contributed to an increase in viscosity that caused the Skaergaard magma to cease or nearly stop convection. As the Skaergaard magma ceased convection, or convected as small isolated cells, during the final stages of differentiation, volatile elements could have accumulated in the UBS magma, resulting in an increase in  $f\text{O}_2$ , and a decrease in Eu/Sm in UBS plagioclase. Apatite solubility is positively correlated with water (Hanchar and Watson, 1993), so an elevated water pressure in the UBS, could have prevented the UBS magma from crystallizing apatite as a primary phase, and instead suppressed apatite crystallization enough to cause apatite to only precipitate as an interstitial phase from the trapped melt.

Using published  $K_{\text{Di}}$  for rare earth elements in apatite, and the calculated rare earth element trend

in the Skaergaard, the theoretical trends of rare earth elements in LS and UBS apatite can be calculated (Fig. 1 and Table 1). In addition, the rare earth element content in LS apatite can be estimated by the inventory method using the rare earth concentrations of LS whole rock samples, the modal fractions of mineral phases, and the measured rare earth element contents of plagioclase and pyroxene. Both measured (Wager and Mitchell, 1951; Paster *et al.*, 1974; Nash, 1976; Naslund, 1980; McBirney, in review; this study) and estimated rare earth element contents of Skaergaard apatite follow the theoretical apatite trend. This close match between theoretical and observed trends suggests that LS apatite precipitated as a liquidus phase and, hence, significantly affected the rare earth element trend of the LS magma and the rare earth element trends for the other liquidus phases as well. UBS apatite apparently did not precipitate as a liquidus phase and, hence, did not affect the rare earth element trend of the UBS magma or the other UBS liquidus phases.

## 5. CONCLUSIONS

Trace element modeling of the Skaergaard intrusion using appropriate published  $K_{Dis}$ , the concentrations of trace elements in mineral separates and whole-rock samples, and the modes of coexisting mineral phases, demonstrates that the abrupt appearance of abundant apatite at the UZa/UZb boundary of the LS, caused a significant decrease in rare earth element abundances in the Skaergaard LS magma. Trace element modeling suggests that apatite in the UBS is not a primary liquid phase, but an interstitial phase that did not effect the enrichment trend of the magma. As the Skaergaard magma ceased convection, or convected as small isolated cells during the final stages of differentiation (Naslund, 1977; Jang and Naslund, 2001), an elevated  $P_{H_2O}$  induced by the accumulation of volatile elements in the UBS magma, inhibited or delayed the UBS from precipitating primary apatite. The Skaergaard magma in both the UBS and the LS followed the expected trace element trends for closed system crystallization, and, therefore, any differentiation model of the Skaergaard intrusion with significant amounts of injection or discharge from the chamber during the final 20% of crystallization can be ruled out.

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