Compositional Variation in Olivine in the Skaergaard Intrusion

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Skaergaard 암체의 감람석의 성분변화

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Abstract: Olivine from the Layered Series (LS), Upper Border Series (UBS), and Marginal Border Series (MBS) of the Skaergaard intrusion was analyzed to examine compositional variation. In general, olivine from all three series shows similar trends in major elements with differentiation: FeO* (total iron as FeO*), TiO_2, and MnO in the olivine, progressively increase, MgO and SiO_2 progressively decrease, and Na_2O, K_2O, Al_2O_3, CaO, and P_2O_5 remain unchanged at low abundances. No abrupt changes in the trends of major components in Skaergaard olivine are observed. The systematic compositional variations in Skaergaard olivine during differentiation preclude the possibility of any injection of new magma into the chamber during differentiation. Abrupt change in MnO, however, is observed in Skaergaard olivine during differentiation. MnO in Skaergaard olivine steadily increases with differentiation from LZ to UZc and then decreases in UZc of the LS and its equivalents of the MBS and UBS. The decrease of MnO in Skaergaard olivine during the final stage of differentiation is attributable to the abrupt and extensive appearance of ferrobustamite as a liquidus phase.

Key words: olivine, igneous differentiation, Skaergaard intrusion.

요 약: Skaergaard 암체의 감람석의 성분변화를 연구하기 위하여 Layered Series(LS), Upper Border Series (UBS), 및 Marginal Border Series(MBS)에서 산출되는 감람석의 성분을 분석하였다. 분화에 따른 원소조성변화는 일반적으로 암체 전체에서 유사한 원소조성변화를 보여주는데, 감람석의 FeO*, TiO_2, 및 MnO는 증가하고, MgO와 SiO_2는 감소하고, Na_2O, K_2O, Al_2O_3, CaO, 및 P_2O_5는 분화에 따른 주목할 만한 변화를 보이지 않는다. 분화과정의 전반에 걸쳐 Skaergaard 감람석은 어떠한 급격한 원소 변화 또한 보여주지 않는다. 따라서 마그마 분화에 따라 Skaergaard의 감람석이 체계적인 원소조성변화를 보이는 것으로 Skaergaard마그마 분화 도중에 새로운 마그마의 주입의 가능성은 배제된다. 주목할 만한 급격한 원소변화가 MnO에서 관찰된다. Layered Series(LS) 감람석의 MnO 성분은 분화가 진행됨에 따라 꾸준히 증가하다가 UZc에서 급격히 감소하는데, 유사한 경향이 UBS, 및 MBS에서도 관찰된다. 이와 같이 Skaergaard 마그마 분화 말기에 관찰되는 감람석의 MnO 성분의 급격한 변화는 liquidus상으로 갑자기 출현하는 ferrobustamite에 기인한다.

핵심어: 감람석, 화성분화, Skaergaard암체.

Introduction

The Skaergaard intrusion of East Greenland is one of the most intensely studied igneous bodies in the world. Since its first description (Wager and Deer, 1939; Wager and Brown, 1967), it is widely considered the type example of a strongly fractionated, layered intrusion, and a classic example of an igneous body that has undergone extensive in situ igneous differentiation. The Skaergaard, therefore, is one of the best examples in the world to study how liquids and coexisting mineral phases change as the

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differentiation of magma proceeds. Although the Skaergaard intrusion has been the subject of an extensive amount of publication, there is still debate about some of the most basic conclusions drawn by Wager and his coworkers (Wager and Deer, 1939; Wager and Brown, 1967), including: (1) the compositional trend of differentiation of the Skaergaard magma (Hunter and Sparks, 1987, 1990; McBirney and Naslund, 1990; Morse, 1990; Brooks and Nielsen, 1990; Toplis and Carroll, 1995, 1996; Loucks, 1996; Tegner, 1997, in review); (2) whether the intrusion remained as a open or closed system during differentiation (Hunter and Sparks, 1987, 1990; Stewart and DePaolo, 1990; McBirney, in review; Tegner, in review); (3) whether the magma convected during crystallization or was stagnant once crystallization started (Marsh, 1988, 1989; Huppert and Sparks, 1984); and (4) the temperature of crystallization during the solidification of the intrusion (Lindsley et al., 1969; Williams, 1971; Morse et al., 1980; Naslund, 1984; Lindsley, 1983; McBirney and Naslund, 1990).

Part of the reason for the continuing debate about some of the most basic features of the Skaergaard crystallization history is the inherent nature of the rocks, which are mixtures of early formed minerals and trapped melt. The importance of compositional variations of individual mineral phases for understanding the differentiation history of the Skaergaard intrusion has been recognized (Wager and Mitchell, 1951; McBirney, 1989, 1998; Jang and Naslund, 2001). Although a number of studies included analyses of one of the major mineral phases in the intrusion, olivine (Deer and Wager, 1939; Wager and Deer, 1939; Wager and Mitchell, 1951; Wager and Brown, 1967; Paster et al., 1974; Nwe, 1976; Naslund, 1984; Hoover, 1989a; McBirney, 1989, 1998; Tegner, in review), no previous study has systematically examined compositional variations in Skaergaard olivine.

In order to make comparisons between the floor, walls, and roof sequences, and to evaluate compositional variations, Skaergaard olivine has been examined using election microprobe analyses of samples from the Layered Series, the Marginal Border Series,

and the Upper Border Series of the intrusion. The major purposes of this study are to acquire reliable, systematic, geochemical data on Skaergaard olivine, over the entire range of crystallization, and to delineate how geochemical compositions in olivine change as the differentiation of Skaergaard magma proceeds.

Geology

The geology of the Skaergaard intrusion was first described by its discoverer, L. R. Wager and his coworkers (Wager and Deer, 1939; Wager and Brown, 1967) who outlined the general field relationships, mineralogical variations, and geochemical trends. Expeditions to the intrusion by later workers broadened and deepened the understanding of the intrusion and its relationship with regional igneous activities (McBirney, 1975, 1989; Naslund, 1984; Nielsen, 1978; Brooks and Nielsen, 1978; Hoover, 1989a, 1989b; Hirschmann et al., 1997; Tegner et al., 1998; and numerous others). Details about the general geology of the intrusion are available in these references.

The Skaergaard 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 along the sides of the chamber; and an Upper Border Series (UBS), which crystallized against the roof of the chamber (Wager and Brown, 1967; Naslund, 1984; Hoover, 1989a). The Layered Series is subdivided into Lower, Middle, and Upper Zones based on the disappearance and subsequent reappearance of olivine as a primary phase (i.e., the Middle Zone is generally olivine free). Further divisions of the Lower Zone into a, b, and c are based on the first appearances of subhedral clinopyroxene and abundant magnetite. The Upper Zone is similarly subdivided by the first appearances of apatite and ferrobustamite as primary minerals. The final unit of the intrusion, the Sandwich Horizon (SH), crystallized from the residual magma trapped between the upward crystallizing LS and the downward crystallizing UBS. The subdivisions of the Marginal Border

Series and the Upper Border Series are similar to those of the Layered Series, but more compressed. Although their mineral sequences differ in minor ways, the Layered Series, Upper Border Series, and Marginal Border Series all show similar trends of magma differentiation (Naslund, 1984; Hoover, 1989a; McBirney, 1989).

Petrographic studies of the Layered Series (LS), Marginal Border Series (MBS), and Upper Border Series (UBS) show that all three have followed similar sequences of crystallization. In general, the LS have better developed layering and wider ranges of modal abundances than the MBS or UBS, but mineral trends are similar in all three units. One major petrographic difference between and LS and UBS rocks is the apparent higher percentage of trapped melt in the UBS, which has crystallized as myrmekite, microgranite, and interstitial granophyre.

Olivine is one of the major cumulus phases in the Lower and Upper Zones of the LS and their equivalents in the UBS and MBS. Olivine is rarely found in the MZ of the LS or in its equivalents in the UBS and MBS, except as reaction rims between pyroxene and magnetite. Skaergaard olivine shows extreme compositional variation with fractionation from about Fo66 in LZa to Fo0 in the SH of the LS, from Fo57 in UBZ? to Fo0 in UBZ? in the UBS, and from Fo74 in MBSt to Fo13 in UZb equivalent rocks of the MBS (Wager and Brown, 1967; Nwe, 1976; Naslund, 1984; McBirney, 1989; Hoover, 1989a; Tegner, in review).

Olivine in the Lower Zone of the LS and its equivalents in the MBS is present as subhedral to euhedral primocryst, ranges from 0.2 to 5 mm in length, and makes up from 0 to 10 % by mode. Olivine in Upper Zone of the LS and its equivalent in the MBS is present as subhedral to euhedral primocryst ranges from 0.2 to 5 mm in length, and makes up from 0 to 20% by mode. Olivine in the UBS occurs as subhedral to poikilitic grains up to 15 mm across and makes up 30% or serpentine and opaque oxide pseudomorphs of olivine by volume of some samples, but in most samples make up less than 5% of the mode. Iron-rich olivines in many LS, MBS, and UBS samples contain fine exsolu-

tion lamellae of magnetite. In some UBS samples olivine is partly altered to ilvaite (Naslund *et al.*, 1983).

Geochmistry

Geochemical compositions of olivine from all three series and additional olivines that occur as reaction rims or as rare discrete grains in the Middle Zone of the LS and equivalents of the MBS and UBS were also analyzed using a JEOL 8900 electron microprobe in the Department of Geological Sciences at Binghamton University (Table 1). Operating conditions were 15 kv accelerating voltage, 15 nA sample current, and 1-3 micrometers beam diameter. The Bence-Albee method (1968) was used for ZAF matrix corrections. For each sample at least 9 points (3 points in three different olivine grains) were analyzed.

In general, major elements in olivine from the LS, UBS, and MBS show similar trends. FeO* (total iron as FeO*), TiO_2, and MnO in the olivine, progressively increase, and MgO and SiO_2 progressively decrease, with differentiation. Na_2O, K_2O, Al_2O_3, CaO, and P_2O_5 do not appear to change systematically and remain low during differentiation (Fig. 1). The general trend reported by previous studies from limited mineral separate data (Deer and Wager, 1939; Wager and Deer, 1939; Wager and Mitchell, 1951) and microprobe analysis (Nwe, 1976; Naslund, 1984; Hoover, 1989a; McBirney, 1989, 1998; Tegner, in review) is confirmed by this study.

The MgO content linearly decreases from 40% MgO in the least differentiated samples, to almost 0% MgO in the most differentiated samples, while FeO* systematically increases with differentiation, from 30 percent FeO* in the least differentiated samples to near 70 percent FeO* in the most differentiated samples. MnO in Skaergaard olivine steadily increases with differentiation from 0.2% in Lower Zone to almost 2% in UZc of the LS and its equivalents in the MBS and UBS, and then rapidly decreases with differentiation during the final few % of crystallization. The decrease in MnO may be related to the change from ferrohedenbergite to fer-

Table 1. Major element analyses of of olivine of the Skaergaard intrusion.

	Zone	Xl%	Fo#	SiO_2	TiO_2	Al_2O_3	FeO*	MnO	MgO	CaO	Na_2O	Total
KG-783	LZa	36	60	36.13	0.02	0.01	34.10	0.50	28.64	0.04	0.01	99.45
KG-782	LZb	37	58	35.99	0.03	0.01	35.16	0.46	27.77	0.06	0.02	99.48
KG-781	LZb	42	56	35.82	0.02	0.01	37.10	0.50	27.02	0.03	0.01	100.51
KG-776	LZc	52	54	35.59	0.05	0.01	38.67	0.49	25.71	0.06	0.00	100.58
KG-797	MZ	54	53	35.29	0.03	0.01	39.02	0.52	25.09	0.06	0.01	100.05
KG-813	MZ	54	45	34.82	0.05	0.01	44.19	0.62	20.47	0.04	0.01	100.20
KG-815	MZ	54	56	35.66	0.06	0.01	36.58	0.45	26.27	0.05	0.02	99.10
KG-778	MZ	54	53	35.32	0.03	<0.01	39.12	0.53	24.65	0.03	0.01	99.69
KG-899	MZ	54	52	35.35	0.04	0.02	40.14	0.57	24.22	0.13	<0.01	100.47
KG-772	MZ	72	38	33.48	0.05	0.01	48.44	0.75	16.67	0.07	0.01	99.47
KG-773	MZ	72	38	33.62	0.05	0.01	48.19	0.76	16.83	0.17	<0.01	99.64
KG-769	MZ	72	42	34.00	0.04	0.01	45.70	0.66	18.70	0.09	<0.01	99.20
KG-908	UZa	75	39	33.83	0.04	0.01	47.87	0.75	17.40	0.08	0.01	99.98
KG-830	UZa	81	35	33.02	0.05	0.01	50.07	0.74	15.40	0.15	0.01	99.44
KG-831	UZb	82	28	32.72	0.05	<0.01	53.79	0.86	11.76	0.25	0.02	99.45
KG-832	UZb	86	22	31.94	0.08	<0.01	56.71	1.02	9.05	0.21	0.02	99.02
KG-833	UZb	86	20	31.73	0.05	0.01	58.47	1.00	8.10	0.09	0.01	99.46
KG-834	UZb	90	13	31.21	0.10	0.01	61.33	1.12	5.34	0.03	0.02	99.16
KG-835	UZc	94	2	30.54	0.08	<0.01	65.78	1.98	0.59	0.02	0.02	99.01
KG-836	UZc	99	0	30.43	0.06	<0.01	66.59	1.37	0.19	0.34	0.02	98.99
KG-837	SH	100	0	30.36	0.08	<0.01	68.11	0.83	0.01	0.05	0.01	99.46
KG-107	UBZt	15	58	35.40	0.03	<0.01	35.62	0.51	27.76	0.06	<0.01	99.38
KG-110	UBZα	37	54	35.05	0.03	0.01	38.29	0.58	25.51	0.14	<0.01	99.62
KG-757	UBZα	76	24	32.33	0.04	0.01	55.51	0.97	9.95	0.20	0.02	99.01
KG-765	UBZγ	97	0	30.49	0.09	0.01	67.04	0.83	0.11	0.50	0.04	99.09
KG-806	MBSt	15	58	36.21	0.02	<0.01	35.56	0.50	27.83	0.10	<0.01	100.23
KG-804	LZb*	39	58	35.86	0.04	0.01	35.31	0.46	27.82	0.12	0.01	99.62
KG-803	LZc*	50	50	35.21	0.03	<0.01	41.25	0.58	23.35	0.14	0.01	100.57
KG-802	MZ*	63	42	34.24	0.02	<0.01	45.96	0.64	18.97	0.11	0.01	99.94
KG-800	UZb*	87	13	31.64	0.06	<0.01	62.05	1.13	5.08	0.24	0.01	100.21

Xl%-Crystallization percent calculated from stratigraphic thickness assuming 0% crystallization at the chilled margin, 30% at the bottom of LZa of the LS and equivalents in the UBS and MBS, and 100% at the SH.
FeO*-FeO* as total iron.
Fo#-mole percent of forsterite.

robustamite at the UZb/UZc boundary (Wager and Brown, 1967; Naslund, 1984; Jang, 1999; Tegner, in review). The SiO_2 content in Skaergaard olivine shows a steady but small decrease with differentiation in the LS, UBS, and MBS as a result of the change from forsteritic to fayalitic olivine.

Any significant variations in intensive parameters or magma composition during differentiation should be recorded as compositional changes in the crystallizing olivine. Except for MnO in UZc no abrupt changes are observed in the trends of the major components of Skaergaard olivine during differentia-

tion of the intrusion. The systematic variation of major elements in Skaergaard olivine suggests that the intrusion was a closed system in terms of magma recharge throughout its crystallization history.

Disscussion

Possibility of introduction of new magma

Since the original interpretation of the Skaergaard intrusion (Wager and Deer 1939), it has been considered to be an example of extreme, in situ, closed-system crystallization. Recently, however, the
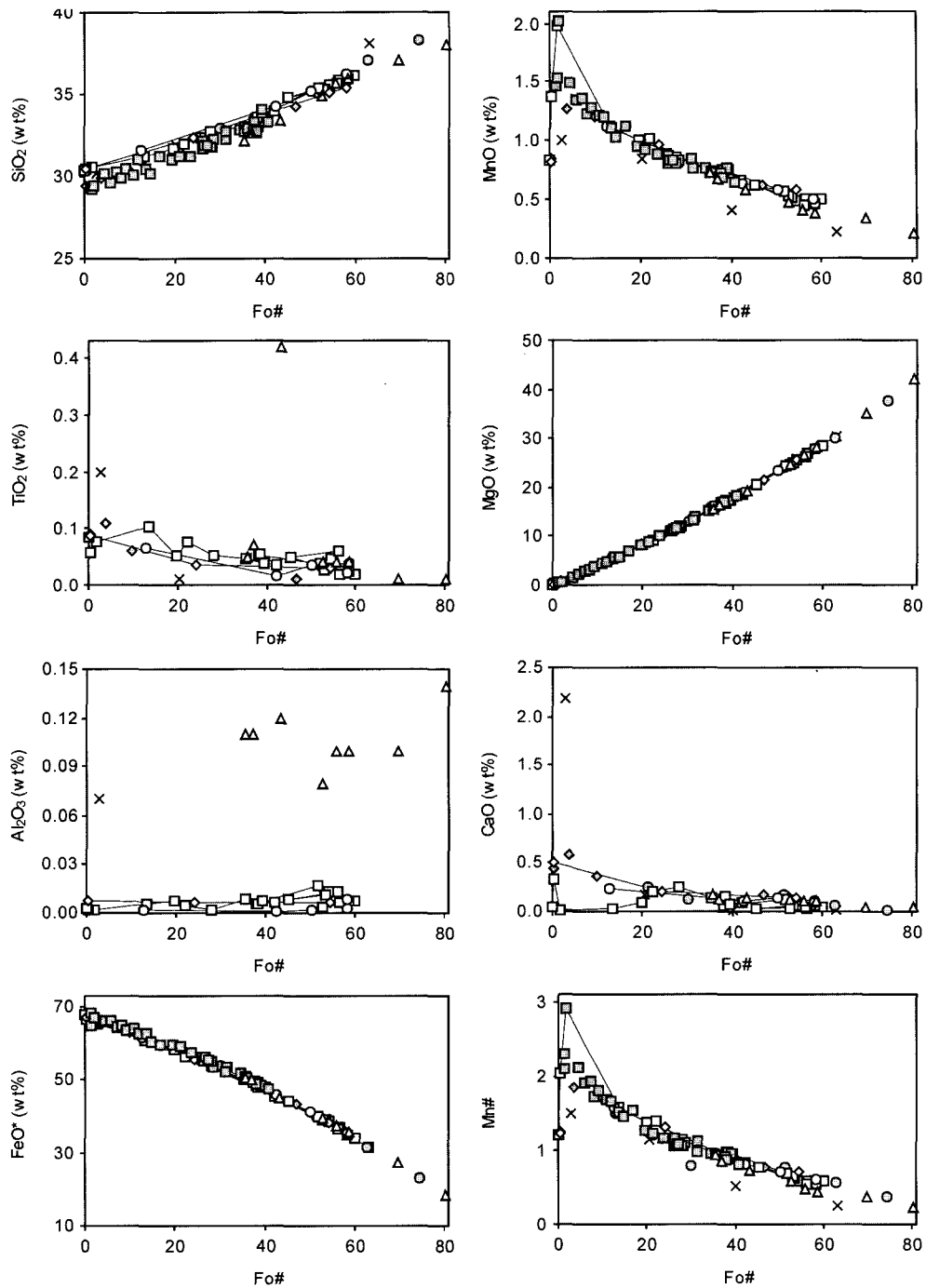
Fig. 1. Compositional variation of Skaergaard olivine plotted against mole percent of forsterite (Fo#). Layered Series
(LS)-open squares (this study by microprobe analyses), shaded squares (Tegner, in review), triangles (Nwe, 1976), and
x's (Wager and Mitchell, 1951); Upper Border Series (UBS)-open diamonds (this study by microprobe analyses), and
shaded diamonds (Naslund, 1984); Marginal Border Series (MBS)-open circles (this study by microprobe analyses),
shaded circles (Hoover, 1989a). Mn#=Mn/(Mg+Fe+Mn).

closed-system model has been challenged. Using theoretical mass balance calculations, Hunter and Sparks (1987, 1990) suggested that the Sandwich Horizon, does not represent the final Skaergaard liquid, but rather is a cumulate from a rhyolitic liquid that was erupted from the chamber. In addition, isotopic modeling of the Skaergaard intrusion (Stewart and DePaolo 1990) suggested that variations of isotopic ratios of Sr and Nd were caused by replenishment of new magmas into the Skaergaard chamber either continuously or in pulses over the history of crystallization.

Any large injections of new magma or discharges of magma through volcanic eruption as suggested by Stewart and DePaolo (1990) and Hunter and Sparks (1987, 1990) would be expected to produce petrological or field evidence, mineralogical interruptions, and/or geochemical spikes.

Drawing on their detailed field studies, McBirney and Naslund (1990) suggested that there is no evidence that a large mass of rhyolitic magma could precipitate the SH, and then erupt from the chamber. Experimentally determined Skaergaard liquids (McBirney and Naslund, 1990) suggest that low-SiO$_2$ trapped liquids are in equilibrium with the observed cumulate mineralogy through the SH, and that the proposed rhyolitic liquid is unnecessary to explain the Upper Zone and SH rocks. The many fold increases in excluded trace elements in Skaergaard whole rock samples, and plagioclase and pyroxene, and olivine during the final stages of crystallization indicate that any removal of magma during the final stage of crystallization must have been volumetrically very small (<1%).

Except for the phase change induced abrupt compositional changes in pyroxene at the UZb/c boundary (Jang, 1999; Tegner, in review), plagioclase, olivine, pyroxene, and ilmenite in the Skaergaard intrusion show very systematic continuous compositional changes with differentiation. Abrupt compositional changes in whole rock samples are attributed to modal variation of specific minerals, for example, Ni to olivine, Cr to pyroxene, V to magnetite, Ti to ilmenite, and P to apatite.

Recent isotopic studies of the intrusion (McBirney and Creaser, 2003; Tegner, in review) suggest

that isotopic ratios of Sr and Nd provide no evidence that a large batch of magma entered the chamber after its initial filling, and that the different isotopic ratios of many closely associated rocks seem to be related to their lithologic character.

Differentiation trend of the Skaergaard magma

Since the pioneering work on the trend of the Skaergaard magma by Wager and his coworkers (Wager and Deer, 1939; Wager, 1960; Wager and Brown, 1967), extreme iron-enrichment with no silica-enrichment until a very late stage of magma differentiation has been regarded as the typical example of iron-enrichment differentiation of basaltic magma at shallow levels in the crust. Recently, however, Hunter and Sparks (1987, 1990) proposed that the Skaergaard magma evolved on a trend of pronounced silica-enrichment after cumulus magnetite appeared at the top of the Lower Zone. Since the proposal by Hunter and Sparks (1987), opinions on the differentiation trend of the intrusion are divided into two different groups with many different ideas to approach the problem. Ideas used in favor of silica-enrichment trend include mass balance calculation based on magnetite crystallization (Hunter and Sparks, 1987, 1990), mineral-restoration technique (Loucks, 1996), and experimental work (Toplis and Carrol, 1995). Ideas used in favor of iron-enrichment trend include partial melting of the cumulates (McBirney and Nakamura, 1974; McBirney, 1975; McBirney and Naslund, 1990), relationship of olivine and silica activity (Irvine, 1976; Morse et al., 1980; Morse, 1990), the composition of contemporaneous dykes (Brooks and Nielsen, 1978b; Brooks and Nielsen, 1990), poly-mineralic melt inclusions (Hanghoj et al., 1995), iron in plagioclase (Tegner, 1997; McBirney, 1998), Mn/Fe in augite (Tegner, in review), and timing of magnetite crystallization (Jang et al., 2001).

Although lateral variations in mineral compositions have been reported in the LS (McBirney, 1975, 1989) and in the UBS (Douglas, 1961; Naslund, 1977, 1984), the changes in mineral composition with differentiation in the Skaergaard are generally consistent between the two series. Valu-

able information on the trend of the Skaergaard magma can be inferred from the sequence of appearances and disappearances of primocrysts in the stratigraphic section of the Skaergaard intrusion, and in the compositional variation of the solid-solution series whose compositions systematically vary in accordance with evolving contemporaneous magma (Naslund, 1980). In general, there are no prominent differences in the compositional trend of olivine primocryst solid-solution between the LS, MBS, and UBS, which implies that the composition of the Skaergaard magma was similar at the floor, walls, and roof of the intrusion. The sequence of olivine primocryst puts a limit on the range of liquid composition and the trend of magma differentiation. Reappearance of olivine as liquidus phase at top of Middle Zone and remains as equilibrium phase to the end of differentiation indicates that the activity of silica in Skaergaard magma probably remained low during differentiation (Morse, 1990; Tegner, in review). Although the extreme compositional change of Skaergaard olivine up to pure fayalite at final stages of differentiation can put limits on final melt to volumes, it can not tell if the intrusion follow Fe-enrichment or Si-enrichment trend.

Origin of MnO trend

MnO in Skaergaard olivine steadily increases from 0.2% in the least differentiated samples up to 2% in UZc of the LS and its equivalents of the MBS and UBS, and then abruptly decreases with differentiation. A similar MnO trend is observed in whole rock samples and in other mineral phases as well (Vincent and Phillips, 1954; Wager and Brown, 1967; Jang, 1999; Tegner, in review). MnO in Skaergaard pyroxene steadily increases from 0.3 to 0.5% with differentiation in UZc of the LS and equivalents of the MBS and UBS it abruptly increases up to 1.2%, and then decrease with further differentiation (Wager and Brown, 1967; Jang, 1999). The abrupt increase of MnO in pyroxene coincides with a phase change from ferrohedenbergite to ferrobustamite. In addition to the phase change, a significant increase in the modal percentage of pyroxene is observed at the UZb/UZc boundary (Wager and

Brown, 1967; Jang, 1999). The decrease of MnO in Skaergaard olivine during the final stage of differentiation is probably attributable to the abrupt and extensive crystallization of ferrobustamite, which has a high KDMn and causes a depletion of MnO in the magma. During the final stages of differentiation, Mn acted as a compatible element (Tegner, in review).

Conclusions

The composition of Skaergaard olivine changes systematically with differentiation of the Skaergaard intrusion. Similar compositional variation trends are observed in the olivine from the LS, UBS, and MBS. Systematic compositional variations of Skaergaard olivine during differentiation exclude the possibility of major injections into or discharges from the Skaergaard magma chamber. MnO in Skaergaard olivine steadily increases with differentiation from LZ to UZc and then decreases in UZc of the LS and its equivalents of the MBS and UBS. Similar decreases in MnO are observed in whole rock samples and in other mineral phases as well. MnO in Skaergaard pyroxene increases with differentiation from 0.3% to 0.5%, and then abruptly increases in UZc to 1.2%. The increase of MnO has been attributed to a phase change from ferrohedenbergite to ferrobustamite (Jang, 1999; Tegner, in review). A significant increase in modal pyroxene is also observed at the UZb/UZc boundary (Wager and Brown, 1967; Jang, 1999). The decrease of MnO in Skaergaard olivine during the final stage of differentiation is attributable to the abrupt and extensive appearance of ferrobustamite as a liquidus phase.

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