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Christian Tegner

Iron in plagioclase as a monitor of the differentiation of the Skaergaard intrusion

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Abstract Several recent publications suggest that the appearance of Fe-Ti oxides terminates iron enrichment and starts pronounced silica enrichment (the Bowen trend) during the differentiation of tholeiitic basalt. However, this does not appear to hold for the Skaergaard intrusion. New data from a ~950 m long drill core (90–22) through its Upper Zone reveal that: (1) iron in plagioclase increases from ~0.25 to ~0.45 wt% FeO_T with fractionation of evolved oxide ferrodiorites (An_{46-32}) and (2) the evolving liquid, which is modelled by incremental bulk-rock summation, increased its iron content from 20.1 to 26.5 wt% FeO_T *and* its silica content from 47.4 to 49.6 wt% SiO_2 with fractional crystallisation (the Fenner trend). Positive correlation between modelled iron-content of the magmas, and measured iron-content of plagioclase, confirms that iron enrichment is petrologically feasible even with Fe-Ti oxides in the fractionating assemblage. As suggested by previous authors, fractional crystallisation closed to oxygen exchange is the likely reason why some layered intrusions diverge from the Bowen mechanism of differentiation. It is emphasised that both trends seem to exist in nature.

Introduction

The evolution of tholeiitic magma has been controversial since the birth of igneous petrology. Initially, Bowen (1928) argued that silica-rich rocks in volcanic suites resulted from the differentiation of basalt. Many other workers (e.g. Fenner 1929), however, advocated that fractional crystallisation of basalt leads to iron-rich differentiates. Later, the work of Osborn (1959) and Presnall (1966) made it clear that the liquid line of descent of basalt is strongly influenced by the oxygen fugacity and magnetite crystallisation: a Fenner trend results if the oxygen fugacity is relatively low and the chamber is closed to oxygen exchange, while a chamber buffered at high oxygen fugacities leads to a Bowen trend. Recently, however, it has been questioned whether iron-enrichment is feasible at all subsequent to magnetite saturation (Hunter and Sparks 1987, 1990; Snyder et al. 1993; Thy and Lofgren 1994; Toplis and Carroll 1995, 1996).

The Skaergaard intrusion is central to this discussion because it represents the accumulated products of extreme fractional crystallisation under closed conditions (Fig. 1). Based on: (1) a balanced subtraction of average bulk cumulates from a preferred chilled margin composition (Wager 1960), (2) the composition of contemporaneous dykes (Brooks and Nielsen 1978, 1990), (3) partial melting of cumulates (McBirney and Naslund 1990), and (4) crystallised melt inclusions within plagioclase (Hanghøj et al. 1995), it has been suggested that iron-rich magmas existed during late-stage differentiation of the Skaergaard intrusion (Table 1, Fig. 2). The validity of these estimates, however, has been questioned (Hunter and Sparks 1987, 1990). The purpose of this paper is to use the iron content in plagioclases from a ~950 m long drill core, in combination with liquid compositions modelled by incremental bulk-rock summation of Upper Zone cumulates, to estimate the liquid line of descent of the most evolved Skaergaard liquids.

Drill core 90-22

The petrology of the Skaergaard intrusion (Fig. 1) presents overwhelming evidence for extreme, closed-system fractional crystallisation from a tholeiitic magma (Wager and Brown 1968; Naslund 1984; Hoover 1989; McBirney 1989, 1995). In addition, the presence of

C. Tegner[1]
Danish Lithosphere Centre, Øster Voldgade 10,
1350 København K, Denmark

[1] *Present address*:
Geological Survey of Norway, Leiv Eirikssonsvei 39,
Pb3006 Lade, 7002 Trondheim, Norway
E-mail: christian.tegner@ngu.no

Editorial responsibility: I. Parsons

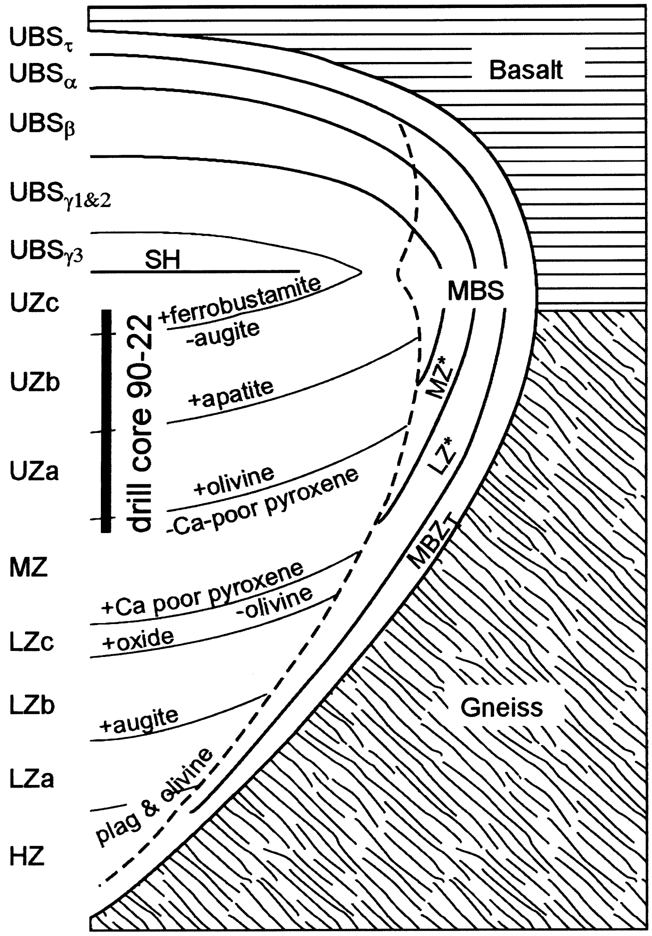

Fig. 1 Schematic cross section of the Skaergaard intrusion that solidified inwards from the walls (Marginal Border Series *MBS*), upwards from the floor (Layered Series, which in turn is divided into subzones, e.g. *UZa*, based on the appearance of cumulus minerals) and downwards from the roof (Upper Border Series *UBS*), with the last fractions of differentiated melt trapped between the advancing roof and floor cumulates in the Sandwich Horizon (*SH*). Modified from McBirney (1995)

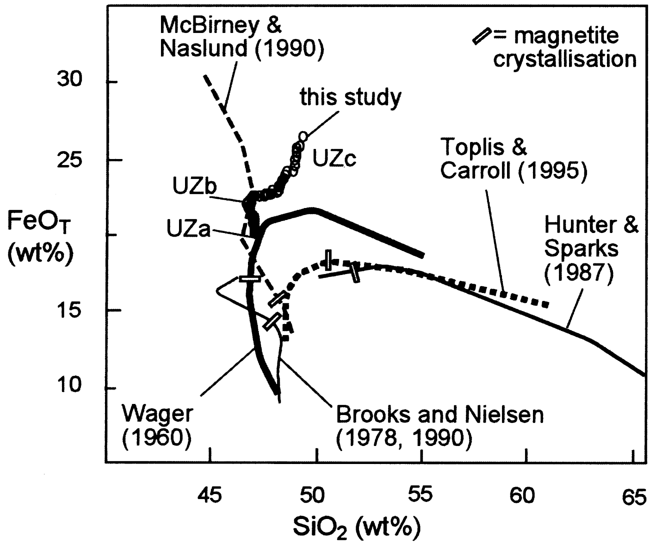

Fig. 2 FeO_T–SiO_2 relations of proposed liquid lines of descent for the Skaergaard intrusion. The starting points are either a preferred parental magma, or the inferred magma composition at the base of the Lower Zone. All liquid lines show the evolution towards the base of UZc with the exception of Brooks and Nielsen's (1978, 1990) line that stops at the base of UZa. The onset of magnetite crystallisation is marked on each line. The liquid line of the present study (UZa to UZc) is also shown

discordant gabbro pegmatites (Larsen and Brooks 1994) and pods of melanogranophyre believed to result from liquid immiscibility in an iron-rich ferrobasalt (McBirney 1975, 1989) indicate that no, or very little, material escaped from the chamber during its solidification. In hole 90-22, Platinova Resources Ltd. cored ~950 m of Upper Zone (UZ) cumulates within the central part of the intrusion (Fig. 1), representing the fractionation products from ~62% to ~95% crystallisation of the parental magma (McBirney 1975, 1995).

New electron microprobe data from plagioclase, and X-ray fluorescence (XRF) bulk-rock concentrations of SiO_2 and FeO_T, are summarized in Fig. 3 and representative analyses are given in Table 2 and 3. The smooth trend of decreasing An% (Ca/(Ca + Na)) in plagioclase (An_{46-32}) with stratigraphic height supports crystallisation in a closed system. The data clearly show that iron in plagioclase (~0.25–0.45 wt% FeO_T) increases smoothly with stratigraphic height through UZa and the lower two-thirds of UZb and correlates inversely with An% (Fig. 3a, b). In the upper one-third of UZb

Table 1 Estimated liquid compositions[a] at the base of the Upper Zone in the Skaergaard intrusion

	This study	H&al[b]	B&N[b]	M&N[b]	W&B[b]	T&al[b]	H&S[b]
SiO_2	47.4	45.7	46.3	46.7	47.4	51.1	53.8
TiO_2	3.7	4.6	5.9	3.6	2.2	4.0	1.9
Al_2O_3	11.9	11.8	11.1	9.5	12.5	10.3	11.9
FeO_T	20.1	17.1	17.1	22.7	20.7	18.5	18.2
MnO	0.3	0.2	–	0.4	0.3	–	0.2
MgO	3.3	3.5	6.3	3.2	2.5	2.9	1.8
CaO	8.5	12.8	10.0	10.0	9.5	8.2	6.6
Na_2O	3.4	3.0	2.4	2.4	3.2	2.9	3.5
K_2O	0.6	0.4	0.9	0.5	0.6	0.9	1.3
P_2O_5	0.8	0.8	–	1.2	1.2	1.1	0.9

[a] Recalculated to 100% volatile free.
[b] H&al = Hanghøj et al. (1995); B&N = Brooks and Nielsen (1990); M&N = McBirney and Naslund (1990); W&B = Wager and Brown (1968); H&S = Hunter and Sparks (1987); T&al = Toplis et al. (1994).

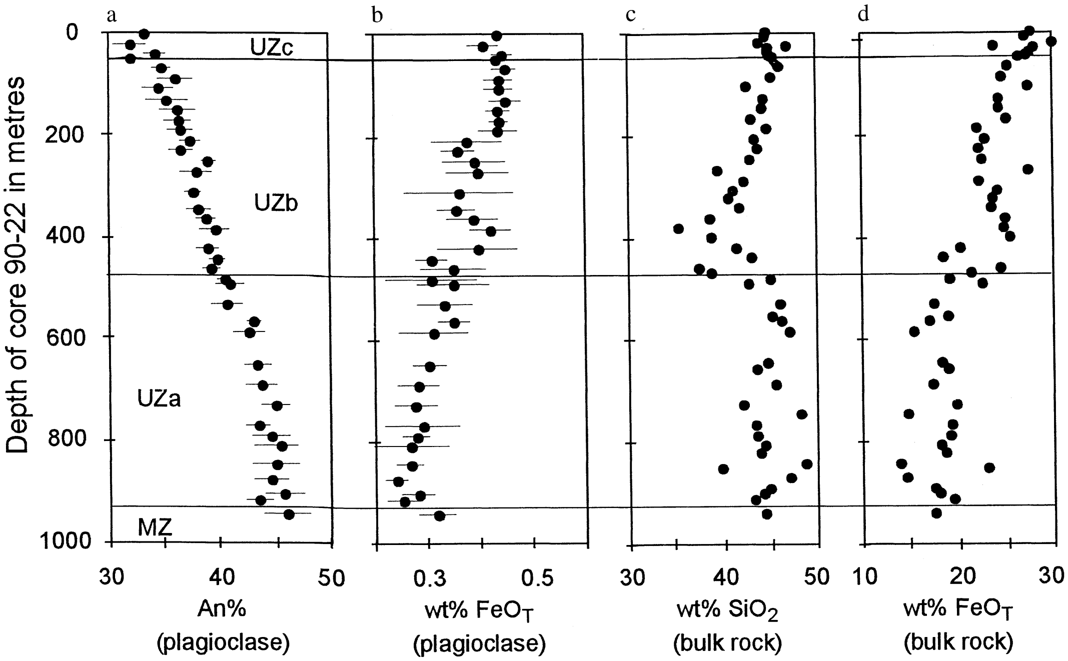

Fig. 3a–d New data plotted against depth of drill core 90-22. **a,b** Average An% and FeO_T in plagioclase, *horizontal bars* represent 2 standard deviation of 9 electron probe analyses within each sample. FeO_T in plagioclase was determined using a wavelength dispersive spectrometer with a precision of less than 0.05 wt%. **c,d** SiO_2 and FeO_T from bulk-rock XRF analysis. Electron probe and XRF analyses were carried out at the department of Earth Sciences, University of Aarhus, as described by Tegner et al. (1993)

and in UZc FeO_T in plagioclase is fairly constant. Bulk-rock SiO_2 content (Fig. 3c) varies between ~35 and ~48 wt%, showing the lowest values within the lower one-third of UZb (corresponding to a relatively melanocratic interval of the core) and a slight upwards increase in the upper half of the core. In contrast, the bulk-rock iron content (Fig. 3d) increases continuously from the base (~15 wt% FeO_T) to the top of the core (up to ~30 wt% FeO_T).

Iron in plagioclase

The substitution of iron in plagioclase [$Ca(Al_2Si_2O_8)$–$Na(AlSi_3O_8)$] occurs both on tetrahedral T-sites (Fe^{3+} and Fe^{2+}) and interstitial M-sites (Fe^{2+}) (Lindsley and Smith 1971; Longhi et al. 1976; Smith and Brown 1987; Xue and Morse 1994). The amount of substitution is strongly dependent on the oxidation state of the iron as demonstrated by experimentally determined partition coefficients (D^{iron} = wt% FeO_T in crystal/wt% FeO_T in melt) for calcic plagioclase ($>An_{75}$) which show a 12-

Table 2 Representative plagioclase analyses from the Upper Zone of the Skaergaard intrusion

Depth of Core 90-22 (m) Cumulate Zone	903.8 UZa	689.1 UZa	491.8 UZa	364.1 UZb	149.8 UZb	0.7 UZc
SiO_2	54.99	56.21	58.35	59.21	59.36	59.38
Al_2O_3	27.47	26.80	25.92	25.39	25.16	24.20
FeO_T	0.27	0.32	0.37	0.41	0.44	0.46
CaO	9.77	8.86	8.09	7.36	7.43	6.32
Na_2O	5.54	6.12	6.15	6.59	6.74	7.20
K_2O	0.51	0.47	0.43	0.55	0.51	0.67
Total	98.55	98.78	99.31	99.51	99.64	98.23
Cations per 8 oxygens						
Si	2.515	2.558	2.627	2.657	2.662	2.697
Al	1.480	1.438	1.375	1.343	1.330	1.296
Fe	0.010	0.012	0.014	0.015	0.016	0.018
Ca	0.478	0.432	0.390	0.354	0.357	0.308
Na	0.491	0.540	0.536	0.574	0.586	0.634
K	0.030	0.027	0.025	0.032	0.029	0.039
Total	5.004	5.007	4.967	4.975	4.980	4.992
An%[a]	49.3	44.4	42.1	38.2	37.9	32.7

[a] An% = Ca/(Ca + Na).

Table 3 Average rock compositions[a] used in liquid modelling

Rock	1[b]	2[b]	3[b]	UZa[c]	UZb[c]	UZc[c]
Depth of Core 90-22 (m)				809.2	249.2	10.6
SiO_2	54.01	59.06	49.34	44.97	43.61	45.39
TiO_2	2.35	1.18	2.14	5.33	3.56	2.96
Al_2O_3	12.76	14.69	8.30	11.97	11.95	8.95
FeO_T	16.03	13.45	26.70	18.50	23.04	27.70
MnO	0.20	0.24	0.40	0.26	0.34	0.73
MgO	1.43	0.58	0.20	6.32	2.94	0.27
CaO	7.45	4.57	9.02	9.79	9.34	10.20
Na_2O	3.78	3.83	2.68	2.64	3.20	2.71
K_2O	1.16	2.05	0.66	0.17	0.29	0.24
P_2O_5	0.82	0.35	0.56	0.05	1.73	0.83

[a] Recalculated to 100% volatile free.

[b] 1 = average of $UBS_{\gamma 1}$ and $UBS_{\gamma 2}$ (Naslund 1984), 2 = average melanogranophyre (Wager and Brown 1968), 3 = average of UZc, $UBS_{\gamma 3}$ and Sandwich Horizon (Wager and Brown 1968; Naslund 1984; McBirney 1989).

[c] Representative bulk rock samples, this study.

fold increase from ~0.030 at very low oxygen fugacities (f_{O_2}) through ~0.044 at the quartz-fayalite-magnetite (QFM) buffer to ~0.36 in air (Phinney 1992). All determinations of the oxidation-state of the Skaergaard magma suggest primary magmatic f_{O_2} values well below the QFM-buffer for the Upper Zone, and indicate a general tendency for f_{O_2} to decrease with fractionation to as low as 2–3 log units below QFM towards the Sandwich Horizon (Williams 1971; Morse et al. 1980; Frost and Lindsley 1992). Hence, it is concluded that the increase in the iron content of the Skaergaard plagioclases cannot be explained by changes in the oxidation of the magma; these changes would predict decreasing iron content in plagioclase with fractionation (Bernstein et al. 1992).

Experimental data on iron partitioning for intermediate and sodic plagioclase are apparently non-existent. Data from natural plagioclases, however, indicate that the iron content decreases with decreasing An% (Smith 1983; Smith and Brown 1987). This is consistent with the stoichiometry of plagioclase; increasing amounts of Si^{4+} leaves fewer available T- and M-sites for iron substitution with decreasing An%. It is therefore clear that the inverse correlation between FeO_T and An% in plagioclase cannot result from major element crystal compositional changes. It is therefore concluded that the iron contents of the Skaergaard plagioclases are related directly to the iron content (mainly Fe^{2+}) of the evolving magma.

Liquid modelling

The variability of published estimates of the composition of evolving Skaergaard liquids (Fig. 2) is evidence of the difficulties posed. The main problem is obviously related to the large degree of freedom in estimating the parental magma, cumulate compositions and volumes. Furthermore, the observed cumulate compositions might be in equilibrium with a variety of magma compositions. Bulk-rock summation of the near-continuous section cored in hole 90-22 is an attractive and alternative approach to estimate liquid evolution. The method uses a weighted, incremental summation of bulk cumulate compositions starting with the stratigraphically youngest rocks to calculate bulk-liquid compositions at successively older stratigraphic levels (Morse 1981). The method assumes firstly that the depth of the drill core is proportional to volume. Secondly, that the volume of magma remaining at the top of the core is equivalent to 40 m of core and that its composition was the average of UZc, Sandwich Horizon and $UBS_{\gamma 3}$ rocks (Table 3). This is the starting composition to which the cumulates of the drill core are added sequentially as described below. Thirdly, it is assumed that crystallisation took place in a closed system. A recent challenge to this assumption (Stewart and DePaolo 1990) was primarily concerned with the stratigraphically oldest rocks (HZ to LZb, i.e. < ~35% crystallised). In general, the isotopic compositions of the Skaergaard intrusion are remarkably constant (Stewart and DePaolo 1990; McBirney 1995; unpublished data by the author). Hence, there is no evidence for the assimilation of country rock and/or magma replenishment in cumulates younger than LZb. Fourthly, it is assumed that late-stage migration of interstitial magma, which has been shown to be significant for the highly incompatible elements (McBirney 1995), is insignificant for the major elements.

The main uncertainty with the present modelling is related to the estimation of rock volume and composition. The calculation uses bulk-rock compositions from fifty samples (i.e. one sample every ~20 m) of the drill core through the central part of the Upper Zone. However, cumulates were not only formed on the floor (the Layered Series, LS, to which the Upper Zone belongs) but also simultaneously on the roof (UBS) and walls (MBS) of the chamber. The volume ratio of UBS to LS cumulates is assumed to be 1:3 (Naslund 1984; McBirney 1995) whereas the MBS cumulates have been ignored because they are volumetrically minor at this stratigraphic level, and their bulk-rock compositions lie between that of LS and UBS (Hoover 1989). The UBS cumulates are relatively leucocratic compared to LS cumulates and the calculation uses an average of $UBS_{\gamma 1}$ and

Table 4 Representative Skaergaard liquids[a] calculated by bulk-rock summation

Cumulate Zone	Base UZa	Mid UZa	Base UZb	Mid UZb	Base UZc	Mid UZc
Depth of Core 90-22 (m)	914.2	769.1	461.8	270.3	46.2	0.7
SiO_2	47.4	47.4	47.1	48.2	49.4	49.6
TiO_2	3.7	3.6	3.1	2.7	2.3	2.2
Al_2O_3	11.9	11.8	11.2	11.0	9.3	8.6
FeO_T	20.1	20.6	22.2	22.9	25.2	26.5
MnO	0.3	0.3	0.3	0.4	0.3	0.1
MgO	3.3	2.9	2.2	1.5	0.5	0.4
CaO	8.5	8.4	8.0	7.5	4.9	1.3
Na_2O	3.4	3.5	3.6	3.9	5.7	8.3
K_2O	0.6	0.7	0.8	0.9	1.6	2.5
P_2O_5	0.8	1.0	1.4	1.0	0.7	0.6

[a] Recalculated to 100% volatile free.

$UBS_{\gamma 2}$ (Naslund 1984). Table 3 summarises the cumulate compositions used. The presence of exsolved melanogranophyres presents another complication. The amount of melanogranophyre increases towards the Sandwich Horizon (Wager and Brown 1968, McBirney 1989) and it is assumed that 2, 8 and 15 volume % of melanogranophyre exsolved within UZa, UZb and UZc, respectively. In summary the ratios of LS to UBS to melanogranophyre used in calculations are 74:24:2, 69:23:8 and 64:21:15 for UZa, UZb and UZc, respectively. However, even large volumetric changes (up to ~50%) for UBS and melanogranophyres do not significantly alter the results.

The modelled melt compositions (Fig. 2, Table 4) suggest a kinked path in which FeO_T increases continuously from ~20.1 wt% at the base of UZa to ~26.5 in UZc whereas the silica content decreases slightly (~47.4 to ~47.1 wt%) through UZa, followed by a steady increase to the top of the core (~49.6 wt%). The kink is obviously caused by the relatively melanocratic cumulate interval at the base of UZb compared to the rest of the Upper Zone. The origin of the melanocratic interval is not known but it is almost certainly *not* related to emplacement of new, primitive magma; this would have been recorded in the An% of plagioclase (Fig. 3a). Possible explanations may be: (1) that the melanocratic interval is limited to a small part of the intrusion sampled in the drill core, (2) a reflection of kinematic or other fluctuations, causing a change in the modal proportions, or (3) downward percolation of iron-rich liquids, see later discussion. It is therefore inferred that the kink in the modelled melt compositions (Fig. 2) is an artefact of crystallisation kinetics rather than a significant change in magma composition. Hence, bulk-rock summation strongly indicates that both the iron and silica contents of the evolving magma increased with fractional crystallisation of the Upper Zone. Two observations confirm that the modelled magma compositions are realistic. Firstly, the composition at the base of UZa (47.4 wt% SiO_2; 20.1 wt% FeO_T) is within the range of most published estimates (Table 1), except for the relatively silica-rich compositions of Hunter and Sparks (1987) and Toplis et al. (1994). Secondly, a positive correlation between measured FeO_T in plagioclase and FeO_T in the modelled liquids (Fig. 4) is reassuring. Only the rocks from UZc seem to diverge from this correlation. A best-fit line of the UZa and UZb data, however, does not project to the origin and suggests that D^{iron} (wt% FeO_T in crystal/wt% FeO_T in melt) varies from ~0.013 to ~0.019 from the base of UZa to the top of UZb. These D-values are smaller than the experimentally determined value of ~0.030 for D^{iron} (mainly D^{Fe2+}) in calcic plagioclase at low f_{O_2} (Phinney 1992) and the measured value of ~0.02 between calcic plagioclase and lunar basalt (Longhi et al. 1976). The obtained D^{iron} values for the Skaergaard rocks therefore seem to be realistic in view of the fewer T-sites available for iron substitution in sodic plagioclase relative to calcic plagioclase. However, a better knowledge of iron partitioning in sodic plagioclases is required before it is appropriate to interpret the correlation between FeO_T in plagioclase and FeO_T in the modelled liquids further.

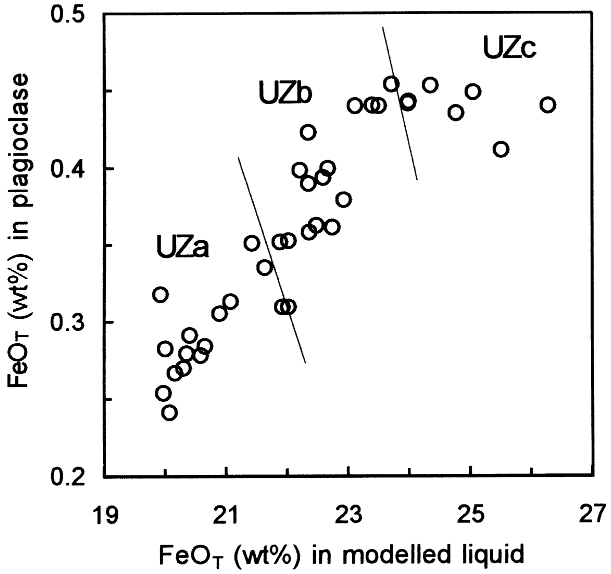

Fig. 4 FeO_T in plagioclase versus FeO_T in modelled liquids

The differentiation of the Skaergaard intrusion

The data presented here are different from previous studies of Skaergaard rocks because the measured iron is a trace element in plagioclase and hence directly monitors the iron content of the magma. The presented data (Figs. 2–4) are taken as evidence that the iron content of the Skaergaard magma continued to increase long after the crystallisation of Fe-Ti oxides began. This result is at odds with many recent publications suggesting that the appearance of Fe-Ti oxides terminates iron enrichment and starts silica enrichment (Hunter and Sparks 1987, 1990; Snyder et al. 1993; Thy and Lofgren 1994; Toplis and Carroll 1995, 1996). The importance of the present study is to emphasize that both trends appear to exist in nature (Morse 1980, 1990). Based on phase diagram considerations in the MgO-FeO-Fe_2O_3-SiO_2 and CaO-MgO-FeO-Fe_2O_3-SiO_2 systems Osborn (1959) and Presnall (1966) made it clear that systems buffered with oxygen follow a Bowen trend while systems closed to oxygen exchange follow a Fenner trend subsequent to magnetite saturation. Recently, Toplis and Carroll (1996) used experimental data, phase relations, mineral-melt partitioning and mass balance constraints to compute the liquid lines of descent under conditions open and closed to oxygen, and concluded that the Skaergaard was closed to oxygen. In the open system FeO_T increased to ~19 wt% (at QFM) and then dropped dramatically following magnetite saturation while in the closed system FeO_T increased to ~17 wt% (at QFM) and then decreased slightly. Considering their calculated modal proportion of fractionated magnetite (~43 wt% in the open system and ~26 wt% in the closed system at QFM), it is not surprising that the iron content of the computed liquids decreased following the onset of magnetite crystallisation. In the case of perfect fractional crystallisation closed to oxygen (starting at QFM) Toplis and Carroll (1996) compute that the bulk-rock composition (oxide gabbro) at the onset of magnetite crystallisation would contain ~22 wt% FeO_T. This value is significantly higher than the weighted average composition of LZc and $UBS_{\alpha2}$ cumulates (~18 wt% FeO_T; McBirney, 1989). In other words the calculations of Toplis and Carroll (1996) appear to overestimate the modal proportion of magnetite and, hence, may result in spurious liquid lines. Indeed, Toplis and Carroll (1996) showed that the modal proportion of magnetite is significantly smaller (~20 wt%) at QFM-1, but did not show the calculated liquid line for this scenario. In summary, the data presented here suggest that the iron content of the Skaergaard magma increased at least through to the upper part of UZb confirming that a Fenner trend is petrologically feasible in a closed-system plutonic environment.

Iron-rich tholeiites at rifted continental margins

Brooks et al. (1991) pointed out that iron-rich tholeiitic lavas are mainly found in large igneous flood basalt provinces related to continental break-up. This setting is certainly true for the Skaergaard intrusion, which was emplaced into the East Greenland continental margin during Early Tertiary rifting of the North Atlantic. Two principal features of continental margin evolution favour the genesis of iron-rich tholeiites. Firstly, the iron content of primary magmas increases with both increasing mean pressure of partial melting and increasing mean temperature at a given pressure (Langmuir and Hanson 1980). During continental rifting melting primarily occurs in a garnet-facies source at the base of a thick non-melting lithosphere (Fram and Lesher 1993), possibly as a consequence of upwelling hot mantle (Brooks 1973). Hence, melts produced during continental rifting are likely to be enriched in iron relative to primary magmas produced at shallower depth at mid-ocean ridges. Secondly, the presence of thick continental crust at passive margins allows the formation of shallow magma chambers, such a Skaergaard in which tranquil, prolonged fractional crystallisation may occur.

Why are iron-rich tholeiites rare?

Iron-rich tholeiitic magmas erupted on the Earth's surface are rare, and reported iron contents never exceed ~20 wt% FeO_T (Morse 1990; Brooks et al. 1991). This fact, however, does not negate the possible existence of more iron-rich melts at depth. The densities of the modelled magmas increase (~2.84–2.90 g/cm^3, not shown) for iron contents increasing from 20.1 to 26.5 wt% FeO_T. The magmas are therefore likely to be trapped in crustal magma chambers rather than rise to the surface (Stolper and Walker 1980; Brooks et al. 1991). This view is supported by growing evidence for downward percolation, or ponding, of iron-rich magmas in Fenner-trend layered intrusions. In the Bushveld Complex, for example, the discordant, iron-rich pegmatoidal pipes appear to form as a consequence of down-drainage of dense, evolved, iron-rich residual magma into cumulates underlying the crystallisation front (Scoon and Mitchell 1994). Also, in the Kiglapait intrusion Morse (1996) interpreted anomalously iron-rich cumulates in the layered sequence to result from ponding of iron-rich magmas at the chamber floor. A density-controlled reorganisation of liquid layers is also a likely scenario within the Skaergaard magma chamber (McBirney 1995). In drill core 90-22, petrographic and geochemical discontinuities at the base of UZc may suggest that dense, iron-rich magmas drained down from the roof-zone to be emplaced on top of the floor cumulates (Tegner, work in preparation). Furthermore, it may be speculated that the melanocratic interval at the base of UZb is related to downward percolation of iron-rich magmas.

Conclusion

Iron in plagioclases of oxide ferrodiorites of drill core 90-22 directly monitors the iron content of the evolving Skaergaard magma. The inverse correlation between iron content and An% in plagioclase suggests that the iron content of the magma increased with fractionation through to the upper part of UZb. Liquid modelling by incremental bulk-rock summation supports the plagioclase data and reveals that both iron (20.1 to 26.5 wt% FeO_T) and silica (47.4 to 49.6 wt%) increased during fractional crystallisation of the Upper Zone. These results confirm that iron enrichment is petrologically feasible even with Fe-Ti oxides in the fractionating assemblage. As suggested by Osborn (1959) and Presnall (1966) this trend is believed to result from fractional crystallisation closed to oxygen exchange.

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