Lateral Reactive Infiltration in a Vertical Gabbroic Crystal Mush, Skaergaard Intrusion, East Greenland

OLIVIER NAMUR1*, MADELEINE C. S. HUMPHREYS2 AND MARIAN B. HOLNESS1

1DEPARTMENT OF EARTH SCIENCES, UNIVERSITY OF CAMBRIDGE, DOWNING STREET, CAMBRIDGE CB2 3EQ, UK
2DEPARTMENT OF EARTH SCIENCES, UNIVERSITY OF OXFORD, SOUTH PARKS ROAD, OXFORD OX1 3AN, UK

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The Marginal Border Series of the Skaergaard intrusion (East Greenland) crystallized in situ on the vertical walls of the magma chamber. It is subdivided into an outer Unbanded Division and an inner Banded Division. The Banded Division contains abundant centimetre- to decimetre-thick bands dominated by fine-grained mafic minerals, with a morphology evolving from almost planar to deeply scalloped and fingered with increasing distance from the intrusion margin. The morphology of these bands is reminiscent of the reaction fronts described in sedimentary basins infiltrated by reactive fluids. We propose that the banding in the Skaergaard Marginal Border Series is produced by chemical disequilibrium resulting from the suction of primitive liquid from the main magma body into the crystal mush, driven by shrinkage of the mush during solidification. Liquid porous flow results in partial dissolution of evolved pre-existing mafic minerals in the mush. This changes the mush liquid composition to one capable of crystallizing mafic rocks with a very minor plagioclase component. Abrupt solidification of this liquid, resulting in the formation of the colloform bands, is explained by supersaturation of some mafic mineral components (e.g. olivine, clinopyroxene, Fe–Ti oxides) in the infiltrating melt. We suggest that the morphological evolution of the colloform bands is a consequence of increasing crystal mush thickness with progressive differentiation.

KEY WORDS: dissolution; fluid flow; magma chamber; porous media; disequilibrium

INTRODUCTION

Fluid flow through porous media is a process that occurs widely in the Earth, with examples ranging from CO2, water and hydrocarbon migration within sediments (Tissot & Welte, 1984; MacMinn et al., 2010; Minesecu et al., 2010) to extraction of partial melt from the mantle (McKenzie, 1984; Daines & Kohlstedt, 1994; Lundstrom et al., 1995; Kelemen et al., 1997; Dijkstra et al., 2003; Katz et al., 2006). When the infiltrating fluid is out of thermal or compositional equilibrium with the solid matrix (Jupp & Woods, 2003), it may react with its surroundings, potentially resulting in local solid dissolution and precipitation of new phases (Ortoleva et al., 1987a, 1987b; Chadam & Ortoleva, 1990; Aharonov et al., 1997; Maise & Pousin, 1997; Kang et al., 2003). This mechanism of reactive dissolution–precipitation is important for a range of geological processes such as cementation in sedimentary basins (Canals & Meunier, 1995; Chan et al., 2000; Wigley et al., 2012), formation of high-porosity melt-filled channels in the upper mantle (Daines & Kohlstedt, 1994; Kelemen et al., 1995a; Morgan & Liang, 2003; Liang et al., 2010; Van Den Bleeken et al., 2010), infiltration of metamorphic fluids in the crust (Bickle & Baker, 1990; Skelton et al., 1995), metasomatism of the mantle wedge above subduction zones (Maury et al., 1992; Kelemen et al., 1998; Widom et al., 2003; Ishimaru et al., 2007) and metasomatism of cumulate rocks from the oceanic crust and ophiolite complexes (Bédard & Hébert, 1996; Coogan et al., 2000; Lissenberg & Dick, 2008; Renna & Tribuzio, 2011).

In slowly solidifying igneous bodies, such as layered intrusions, sills and thick basalt flows, upward fluid flow through a porous crystal mush may exert a first-order control on both the geochemistry and texture of the cumulates.
Marginal Border Series (MBS), which grew inwards from the vertical walls (Fig. 1b). These units follow roughly parallel trends of differentiation into the center of the intrusion, converging at the Sandwich Horizon (Wager & Deer, 1939).

The Layered Series (LS) is divided into the lowermost and unexposed Hidden Zone (HZ), Lower Zone (LZ), Middle Zone (MZ) and Upper Zone (UZ). Some zones are further subdivided based on the nature of cumulus phases. The order of appearance of those phases is considered to be plagioclase + olivine (HZ, LZa) + augite (LZb) + Fe–Ti oxides (LZc) – olivine (+ pigeonite immediately afterwards; MZ) + olivine (UZA) + apatite (UZb) + ferro-hedenbergite (inverted from β-ferrobasaltite; UZc; Fig. 1b). Continuous fractional crystallization is also recorded by the compositions of the cumulus minerals, which change systematically with increasing stratigraphic height (plagioclase: An_{69–25}; olivine: Fo_{92–0}; clinopyroxene Mg_{#77–0}; Wager & Brown, 1968; Maaloe, 1976; Mc Birney, 1989; Thr et al., 2009).

The Marginal Border Series (MBS) is subdivided in an analogous manner to the LS into HZ*, LZa*, LZe*, MZ*, UZa* and UZb* (Hoover, 1989a). The thickness of the subdivisions generally increases with differentiation, reaching a maximum at the Sandwich Horizon. Although the exposure of the MBS is best on the western margin there appears to be no difference in thickness between equivalent units on the western and eastern sides of the intrusion. The MBS has higher concentrations of incompatible elements than the corresponding subzones of the Layered Series (Wager & Brown, 1968; Hoover, 1989a); this may be attributable to the absence of significant compaction at the vertical walls compared with the floor of the intrusion (Tegner et al., 2009, 2011). The minerals at the outer margins of the MBS are slightly more primitive (An_{32}; Fo_{73}; Cpx-Mg_{#40}) than those observed at the base of the LS (Hoover, 1989a); but in general mafic minerals are more evolved in the MBS than in the LS, possibly as a result of the high trapped liquid fraction (Hoover, 1989a).

The boundary between the MBS and the LS is gradational near the structural base of the intrusion, but elsewhere it is marked by a zone of localized, metre-scale faulting, slump structures, cross-beded laminations, and a gradual decrease of the dip of the layering to match that in the rest of the intrusion (Figs 1c and 2a). Wager & Brown (1968) called this transitional region the ‘cross-bedded zone’, and these features are attributed to physical failure of the steep-sided crystal pile (Wager & Deer, 1939; Irvine, 1980). In places (particularly on Ivnarmiut and Mellem; Fig. 1a) the boundary between the generally coarse-grained, homogeneous MBS and the adjacent LS is sharply defined, with a narrow, locally developed zone rich in oxides and isolated grains of plagioclase and
Fig. 1. (a) Simplified geological map of the Skaergaard intrusion showing the locations of the traverses investigated for this study (SP, Skaergaard Peninsula; SMP, Skaergaard Mini-Peninsula; KR-01, KR-02, KR-03, Kraemer Island). LZ, Lower Zone; MZ, Middle Zone; UZ, Upper Zone; MBS, Marginal Border Series; UBS, Upper Border Series. Map in the upper right corner shows the location of the map area in Greenland. (b) Schematic cross-section of the Skaergaard intrusion modified from Wager & Brown (1968) showing the spatial relationships between the Layered Series, Upper Border Series and Marginal Border Series. Subdivisions of each series and sequence of crystallization in the Layered Series are also shown. (c) Schematic cross-section of the Marginal Border Series as seen on the western side of the intrusion. Subdivisions between Banded and Unbanded (Tranquil Zone) Marginal Border Series, structural relationships with the Layered Series and relative position of various rock types and layering are shown. Modified from Hoover (1989a).
pyroxene separating the MBS from the gently onlapping, strongly modally banded, LS (Figs 2b and 2c).

The MBS was divided by Wager & Brown (1968) into an outer Tranquil Division, which they attributed to static crystallization (‘congelation’) of undifferentiated magma, and a Banded Division (Figs 1c and 2d), which they interpreted as the result of downward flow of magma during active magma convection. The boundary between the Tranquil and Banded Divisions occurs within LZa* (Fig. 1c). Here, we consider the term ‘Tranquil Division’ to be obsolete given both its connotation of a lack of fractional crystallization (Irvine, 1982; Hoover, 1989a), and the gradational manner in which the colloform banding appears (see below), and we prefer to describe rocks from this unit as being part of the unbanded MBS (Fig. 1c).

**PHYSICAL CHARACTERISTICS OF THE MARGINAL BORDER SERIES**

The Marginal Border Series is generally fine-grained close to the contact, and the chilled marginal gabbro is <2–3 m thick where present (Hoover, 1989b). The grain size of the rocks in the outer margin increases to a medium-grained gabbro over ~50 m (Wager & Deer, 1939). After this the
MBS rocks are generally coarser-grained than their LS counterparts and are not laminated.

The most striking features of the outermost, unband MBS are the ‘perpendicular feldspar rock’ and the ‘Wavy Pyroxene rock’, first described by Wager & Brown (1968), and the presence of abundant gabbro pegmatites (Fig. 1c).

The perpendicular feldspar rock occurs as vertical lenses or sheets of olivine gabbro that contain abundant elongate single crystals of plagioclase, typically oriented perpendicular to the vertical layering (Wager & Brown, 1968; McBirney & Noyes, 1979; Hoover, 1989a). The feldspar morphology has been ascribed variously to disequilibrium crystallization from a supercooled magma (Wager & Brown, 1968) or to constitutional supercooling (McBirney & Noyes, 1979).

The Wavy Pyroxene rock (Fig. 1c) forms lenses and pods of very coarse-grained, poikilitic pyroxene + plagioclase ± Fe–Ti oxides, defined mineralogically by the scarcity or absence of olivine (Wager & Brown, 1968; Humphreys & Holness, 2010). The lenses show a systematic morphological progression away from the contact, with a clear increase in thickness and spacing, and have recently been interpreted as late magmatic segregations formed by ductile tearing of partially consolidated mush (Humphreys & Holness, 2010).

Gabbro pegmatites form clots, pods and thin sheets containing abundant granophyre and locally large skeletal Fe–Ti oxide grains. They are interpreted as accumulations of mobile residual interstitial liquid (Larsen & Brooks, 1994).

The colloform banding of the MBS is marked by changes in modal mineralogy and grain size; the bands are always plagioclase-poor relative to the host-rocks and appear dark on weathered surfaces. Banding is generally steeply dipping or vertical. The ‘type example’ of banding is corrugated, with cuspat e or colloform outlines on horizontal rock surfaces (Fig. 2d), although the morphology of the colloform banding becomes more irregular towards the interior of the intrusion (see below).

In addition to the colloform banding, some rocks from the banded inner part of the MBS show well-developed modal layering. This is especially well exposed in Fe–Ti oxide-rich regions close to the edge of the MBS (e.g. LZc* on Ivnarmiut; Fig. 1a) where discontinuous, modally layered packets dip into the intrusion. These layered regions frequently contain discontinuous, layer-parallel, pegmatite bodies and gabbroic xenoliths. We consider these as a consequence of gravitational failure at the mush margin and do not discuss these features further in this paper.

FIELD CHARACTERISTICS OF THE COLLOFORM BANDING

Distribution

Colloform banding (Fig. 2d) first appears in LZa*, with the distance between the contact and the first colloform band generally increasing southwards (Table 1). The first few colloforms nearest the contact are widely separated (up to 60 m), but this distance decreases to a typical spacing of a few metres in the main set of colloforms, starting from the inner part of LZa*. Although some bands are poorly defined, most of the colloform bands in the outermost MBS can be traced along strike for 30–50 m, limited by the extent of outcrop.

The relative distribution of the colloform bands was investigated within LZb*, where they are best developed, along two sections perpendicular to the strike on Skaergaard Peninsula (SP traverse; 253 m; SP in Fig. 1) and Skaergaard Mini-Peninsula (SMP traverse; 70 m; SMP in Fig. 1). The sections were started 100 m away from the first colloform (towards the center of the intrusion) to avoid the unusually widely spaced distribution of the first few colloform bands nearest the contact. The relative homogeneity in the distribution of the colloform bands in the main set was assessed using fractal analysis (Fig. 3).

Following Manning (1994), the fractal dimension $D$ is a measure of how the transects are filled by colloforms and is defined as follows:

$$ D = \frac{d(\log N_r)}{d(\log(1/r))} $$(1)

where $N_r$ is the number of divisions of length $r$ (0.5–70 m) where at least one colloform band was observed. In a diagram showing the evolution of $\log(N_r)$ as a function of $\log(1/r)$, both traverses show two log–linear segments ($r^2$ 0.96–0.99) with slopes of $c$: 0.37 and 0.97. The break in slope occurs at $\log(1/r)$ of $c$: 0.25 (Mini-Peninsula) and $c$: 0.14 (Peninsula), corresponding the division lengths ($r$) of 3–10 m (Fig. 3). The linear segments indicate self-similar behaviour over two length-scales of observation. Average calculated values for $D$ are significantly higher for high $r$ values (3–10 m and above) than for low $r$ values, indicating a pervasive colloform distribution on high length-scales, whereas clustering is apparent over smaller length-scales.

Morphology

The morphology of the colloform bands changes significantly from the contact towards the center of the intrusion. Although this evolution is continuous and progressive, it can be described schematically as a succession of four stages (Fig. 4).

1. The colloform bands nearest the contact are typically almost planar in vertical cross-section, but are scalloped on horizontal rock surfaces, with smoothly rounded cusps that always point to the margin of the intrusion (Fig. 5a). In general, the amplitude ($\gamma$) of the scallops increases with their wavelength ($\lambda$; Fig. 5b and c).

2. With increasing distance from the contact, the colloform bands lose their subplanar length-sections and
develop a cuspat e shape in three dimensions (egg-box structure). They locally occur as a succession of closely spaced subparallel bands (Fig. 6a). These show similar wavelengths and amplitudes, although both parameters vary significantly from one set of colloforms to another (Fig. 6b and c). Within a single set of colloforms, the amplitude and wavelength of single bands may be either concordant, with cusps of one band lining up with the cusps of the next, or discordant, with cusps of one band forming between the cusps of the next. Some colloforms also display a secondary scalloping of short wavelength and low amplitude, superimposed on the main wavelength.

(3) Further from the contact (inner part of LZb* to UZa*), the colloform bands become less laterally continuous and more irregular in shape and orientation. The cusps become increasingly elongate and finger-like on horizontal surfaces (Fig. 7a). As with the outer cusps, the distribution of fingering in a succession of closely spaced colloforms may be concordant or discordant. The fingers generally have rounded ends and subparallel sides, although we observed more complicated fingers with shoulders or bifurcated ends (Figs 6e and 7b, c). Most fingers also contain an internal series of thin and locally discontinuous mafic bands, parallel to the rounded end and separating the homogeneous, more plagioclase-rich, internal gabбро (Fig. 6d, f and g). Some fingers, generally the longest, develop internal structures with a succession of subparallel, thin, mafic bands, homogeneous more leucocratic gabбро and nested pockets of pegmatite (Fig. 7d). Highly foliated and relatively plagioclase-rich gabбро is common along the sides at the end of the fingers.

(4) Further from the contact, the tips of the fingers are detached from the parent colloform. This process results in the development of structures that appear on horizontal surfaces as a series of nested mafic crescents or bowls, convex towards the intrusion margin (Fig. 8a and b). Typically each bowl is 1–4 cm thick and 15–100 cm long, with an approximately constant length to depth ratio (Table 1). In vertical cross-section, they form scoop-shaped structures dipping moderately towards the intrusion center. With increasing distance from the intrusion contact, the bowl structures become more abundant and more homogeneously distributed, locally linking to form chains of irregular cusps (Fig. 8c). Their morphology also changes, becoming less strongly cuspat e and not universally concave-inward.

### Table 1: Field characteristics of colloform bands and bowls

<table>
<thead>
<tr>
<th>Location: Kraemer</th>
<th>Ivannikut</th>
<th>Mini-Peninsula</th>
<th>Peninsula</th>
</tr>
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<tbody>
<tr>
<td>Traverse: KR-01; KR-03</td>
<td>IV SMP</td>
<td>SP</td>
<td></td>
</tr>
<tr>
<td>Units: LZa*–LZc*</td>
<td>LZa*–MZ*</td>
<td>LZb*–UZa*</td>
<td>LZA*-UZb*</td>
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<table>
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<tr>
<th>Colloforms (up to UZa*)</th>
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<tbody>
<tr>
<td>n</td>
<td>41</td>
<td>48</td>
<td>70</td>
</tr>
<tr>
<td>First colloform vs contact (m)</td>
<td>50–57</td>
<td>81–95</td>
<td>95</td>
</tr>
<tr>
<td>Average distance between colloforms (m)</td>
<td>3.6</td>
<td>1.5</td>
<td>3.2</td>
</tr>
<tr>
<td>Maximum distance between colloforms (m)</td>
<td>32</td>
<td>4.5</td>
<td>16</td>
</tr>
<tr>
<td>Minimum distance between colloforms (m)</td>
<td>0.3</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Average thickness (cm)</td>
<td>16</td>
<td>6.7</td>
<td>9.3</td>
</tr>
<tr>
<td>Maximum thickness (cm)</td>
<td>45</td>
<td>60</td>
<td>100</td>
</tr>
<tr>
<td>Minimum thickness (cm)</td>
<td>1.5</td>
<td>2.1</td>
<td>2</td>
</tr>
<tr>
<td>Wavelength (cm)</td>
<td>36–250</td>
<td>55–920</td>
<td>35–700</td>
</tr>
<tr>
<td>Amplitude (cm)</td>
<td>–</td>
<td>10–400</td>
<td>20–200</td>
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<tr>
<th>Bowls (LZc*–UZb*)</th>
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<tbody>
<tr>
<td>n</td>
<td>–</td>
<td>41</td>
<td>60</td>
</tr>
<tr>
<td>Average length (cm)</td>
<td>–</td>
<td>32</td>
<td>33</td>
</tr>
<tr>
<td>Average thickness (cm)</td>
<td>–</td>
<td>8.6</td>
<td>7.8</td>
</tr>
<tr>
<td>Average thickness (cm)</td>
<td>–</td>
<td>4.1</td>
<td>3.1</td>
</tr>
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1Location of traverses is shown in Fig. 1.
2All the measurements are from section SK-01.
3Measurements of distance have been started when colloforms become abundant.
The average wavelength of the colloforms was measured for 66 colloform bands from stage 1 and stage 2 on Kreamer Island \((n = 13)\), on the Skaergaard Peninsula \((n = 36;\) SMP in Fig. 1) and on the Skaergaard Mini-Peninsula \((n = 13)\). Values range from 20 to 1500 cm and wavelengths are randomly distributed between the lowest and highest measured values \((\text{Table 1})\). The average amplitude \((\gamma)\) of the colloforms was also measured in 41 bands \((35 \text{ in SMP traverse; six in SP traverse; Fig. 1})\) and ranges from 10 to 400 cm, with no normal distribution. A slight positive correlation \((r^2 = 0.72)\) is found between wavelength and amplitude, but those two parameters do not appear to evolve in any systematic way with distance from the contact.

Mineralogy and grain size

Colloform bands and bowls are marked by an abrupt increase in the ratio of mafic minerals to plagioclase. The bands typically have a sharp outer margin \((\text{i.e. nearest the intrusion wall})\), whereas the inner margin is sharp or gradational. The mineralogy reflects that of the host troctolite and gabbro; that is, no new phase appears and no phase disappears. Within the middle of the MBS, typically in upper LZb*, more mineralogically complex colloforms are locally observed. These are composite, with a thin, plagioclase-rich, outer part \((\sim 1/3 \text{ of the total thickness})\) and a strongly mafic inner part. Plagioclase grains in the outer band generally show preferred alignment parallel to the colloform band.

Colloform bands are generally much finer-grained than the adjacent troctolite and gabbro on the outer margin \((\text{i.e. towards the intrusion wall})\), with a grain-size ratio between 1:10 and 1:2. The contact-ward, outer, margin of the colloform is therefore defined not only by a change in mineral mode but also by a change in grain size compared with the external host-rock. Colloforms with a sharply defined inner margin tend to have a constant grain size from the outer to the inner side of the band. In contrast, colloforms showing a gradational change of mineral mode from the outer to the inner side of the band \((\text{i.e. gradational inner margin})\), generally also show an increasing grain size towards the center of the intrusion.

Relationship between colloforms, pegmatites and segregations

The MBS is relatively rich in centimetre- to metre-scale pegmatite bodies, and their shape and distribution are commonly controlled by the colloform bands and bowls. Lens-shaped pegmatites locally occur on the outer margin of the colloform bands \((\text{Fig. 8c})\), whereas rounded pegmatites are commonly observed within elongate fingers \((\text{Fig. 9a})\); that is, on the inner margin of the colloform band. In evolved regions of the MBS, pegmatites commonly concentrate at the outer margin of the outermost bowls of any bowl group \((\text{Fig. 8c})\). Whereas pegmatite bodies are never observed to cut the colloform bands, rare examples are observed of pegmatites cutting and displacing isolated bowls.
The colloforms closest to the intrusion margin occur within the zone containing the late magmatic segregations (the Wavy Pyroxene rock; Wager & Deer, 1939; Humphreys & Holness, 2010). Segregations are always absent in the colloform bands themselves, but their morphology changes in the vicinity of the bands. Inward of a colloform, segregations are smaller, more closely spaced and more regular in shape than outward of the band, with a shape and distribution similar to that of segregations close to the intrusion margin (Fig. 9b). Colloform bands therefore appear to reset the evolution of the segregations to a more ‘primitive’ morphology and distribution (Humphreys & Holness, 2010).

**SAMPLING AND METHODS**

**Samples studied**

Colloform samples for this study were collected during three field seasons (2006, 2008 and 2011; Supplementary Dataset 1, available for downloading at http://www.petrology.oxfordjournals.org) in the Skaergaard MBS on Kraemer Island, Skaergaard Peninsula and Skaergaard Mini-Peninsula in the stratigraphic interval ranging from L Za* to U Zb*. When possible, adjacent gabbros on the inward and outward colloform margins were also collected at a distance ranging from 1 cm to 1.5 m. We took detailed sample traverses across 18 colloform bands and adjacent gabbros, including traverses across groups of bands (e.g. C-SK11-C3; C-SK11-C14). The position of the samples and the colloform bands are shown in Fig. 10a and b, with details on traverse lengths and colloform thicknesses given in Fig. 10c and Table 2. Eighteen other colloforms were sampled with a single sample or two samples (colloform + outwards gabbro; Table 2). The complete sample set comprises 66 samples from the colloform bands and 60 samples of host gabbro (Supplementary Dataset 1). Twenty-three additional gabbros (Ivnarmiut traverse IV; seven samples; Kraemer Island KR-02 traverse, six samples; Skaergaard Peninsula SP traverse, 10 samples; Fig. 10c) in the stratigraphic interval L Za*–MZ* were collected relatively far from colloform...
Fig. 6. Outcrop-scale relationships of morphologically evolved (LZb*) colloform bands. (a) Series of closely spaced colloform bands with similar and concordant amplitude and wavelength; Skaergaard Peninsula (68°08′49″N, 31°45′32″W), looking west. (b) Series of closely spaced colloforms. It should be noted that the main bands show a secondary scalloping of low wavelength and low amplitude, superimposed on the main wavelength. Skaergaard Mini-Peninsula (68°09′04″N, 31°45′28″W), looking south. (c) Colloform band showing a morphologically simple tube-like finger; Skaergaard Peninsula (68°08′48″N, 31°45′20″W), looking NE. (d) Thick tube-like finger attached to a relatively thin (2–3 cm) colloform band. The successive banding (2–5 cm thick) inside the finger should be noted. Skaergaard Mini-Peninsula (68°09′04″N, 31°45′28″W), looking SE. (e) Morphologically complicated colloform finger attached to a thin (2–3 cm) colloform band. The finger is characterized by multiple ends, its external part is dominated by fine-grained mafic minerals (dark external band) and is thicker at the ends of the finger than along its sides. The finger also shows internal banding. Skaergaard Mini-Peninsula (68°09′04″N, 31°45′28″W), looking east. (f) Thick (3–5 cm) colloform band showing the development of a decimetre-long finger with internal banding; Skaergaard Peninsula (68°08′50″N, 31°45′38″W), looking east. (g) A 2–3 cm thick colloform band showing a c. 50 cm long finger with internal banding. (Note how the colloform on the external part of the finger is thicker than the sides of the finger; Ivnarmiut (68°09′25″N, 31°45′29″W), looking east.
bands (>2 m away from any colloform) to compare mineral modes in colloforms and ‘background gabbros’. Finally, a set of 77 gabbros forming a continuous traverse from LZa* to UZb* was collected through the MBS on the Skaergaard Peninsula (SP traverse; Supplementary Dataset 1). Some of these samples have already been presented by Humphreys & Holness (2010) and Holness et al. (2011). These samples were used for bulk-rock density measurements. Most of the samples were hammered, and a few traverses through colloform and adjacent gabbros were drilled using a petrol-powered backpack drill to produce cores 5–10 cm long and 3 cm in diameter.

**Analytical methods**

Mineral compositions for plagioclase, olivine, clinopyroxene and low-Ca pyroxene were obtained with a Cameca SX-100 electron microprobe at the University of Cambridge. A 15 kV and 10 nA beam was used with a spot size of 1 μm for olivine and pyroxenes, and 5 μm for plagioclase. Peak counting times were 20 s for major elements and 40 s for minor elements. When possible, at least three points in the cores of three grains (n > 9) were measured; the values reported here correspond to the mean of all the measurements that had a total between 98.5 and 101 wt%. Analyses out of this range were excluded from the average. The following standards were used for Kα X-ray line calibration: diopside for Si and Ca; rutile for Ti; corundum for Al; spessartine for Mn; fayalite for Fe; periclase for Mg; jadeite for Na; K-feldspar for K. Crystals used in the spectrometers were TAP for Si, Mg and Al; PET for Ti, Ca, Na and K; LIF for Mn and Fe. Raw data were corrected with the CATZAF software.

**In situ** mineral trace element analyses of olivine and plagioclase were performed using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the University of Cambridge. A pulsed 213 nm New Wave Research UP213 Nd:YAG laser with 100 mJ energy at a repetition rate of 10 Hz, coupled with an Elan DCR II quadrupole ICP-MS system, was used for ablation. Laser sampling was performed in a He–Ar atmosphere with a beam diameter of
Fig. 8. Outcrop-scale relationships of bowl-shaped colloforms in the Marginal Border Series. (a) Morphology of representative bowls on a horizontal surface. Skaergaard Peninsula (68°08′48″N, 31°45′00″W), looking east. (b) Morphology of representative bowls in cross-section on a steep wall to a north–south gully. The intrusion margin is out of the page, towards the viewer. Ivnarmiut, looking east. (c) Most-evolved morphology of the bowl structures where some bowls link up to form ‘bowl-chains’. The clustering of pegmatite bodies underneath chains of bowls should be noted; in contrast, isolated bowls do not have pegmatite underneath them. Skaergaard Peninsula (68°08′49″N 31°45′04″W), looking north.

Fig. 9. Outcrop-scale relationships between colloform banding, pegmatite bodies and late-stage segregations (Wavy Pyroxene Rock). (a) A 15 cm scale rounded pegmatite body located within a 30 cm long colloform finger. Skaergaard Peninsula (68°08′30″N, 31°43′39″W), looking east. (b) Schematic representation of the morphological evolution and distribution of late-stage segregations at the internal and external contacts of a colloform band. Segregations along the external contact of the band are larger and less closely spaced, suggesting that the colloform bands reset the evolution of the segregations.
Fig. 10. (a) Geological map of the western part of the Marginal Border Series. The locations of the traverses where samples were collected are shown. (b) Schematic section of the Marginal Border Series showing the stratigraphic interval exposed in each section and the locations where some colloform samples (and adjacent gabbros) were collected. (c) Schematic representation and length (cm) of some traverses where colloform (grey shaded areas) samples (white circles) and gabbros were collected. The '0 cm' reference value corresponds to the sharp outer contact of the colloform bands. Colloform contacts are represented by thick vertical lines. Dashed lines indicate that the internal contact is loosely defined. The direction of the intrusion margin is shown for reference. (d) Schematic section of the Marginal Border Series showing the locations where gabbros far from any colloform band were collected.
Table 2: Colloform samples collected for this study

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100 μm. Analyses were calibrated using 24Mg and 28Si as internal standard isotopes based on MgO and SiO2 concentrations measured by electron microprobe. During time-resolved analyses of minerals, possible contamination from inclusions and fractures was detected by monitoring several elements and only the ‘clean’ part of the signals was integrated. Two NIST glasses (610 and 612; Pearce et al., 1997) and BCR-2G (Norman et al., 1998) were used as external and secondary standards. The reproducibility of trace element results for the BCR glasses using the Cambridge analytical protocols is between 0.5 and 4% relative (1σ) for the majority of elements. Data were reduced using Glitter 4.0.
Bulk-rock compositions were measured by X-ray fluorescence (XRF). All the samples were carefully cleaned prior to crushing and milling with agate mortars. Major and some trace elements (Rb, Sr, Y, Zr, Nb, Cu, Ga, Ni, Zn, Co, Ba, Cr, V, P) were determined with an ARL 9400XP spectrometer at the University of Liège (Belgium). Major elements were measured in lithium tetra- and meta-borate fused discs, with matrix correction following the Trail–Lachance algorithm. Trace elements were measured on pressed powder pellets and data were corrected for matrix effects by Compton peak monitoring. Eight measurements were performed on each sample and the values reported here correspond to the average of the eight measurements. For phosphorus, the spectrometer was calibrated using 40 international standards ranging from 0.01 to 5 wt% P₂O₅. Repeated measurements of 15 international standards not included in the calibration curve indicate an accuracy relative to the reference values in the range between 0.3 and 10.5%, with an average of 3.4% (Namur & Charlier, 2012). For other trace elements, the spectrometer was calibrated using 64 international standards; analytical errors are estimated to be lower than 10% in most samples.

Mineral modes were determined by point-counting on polished thin-sections. For each sample 400–2000 points were counted on a grid of 0.3 mm × 0.3 mm. The estimated accuracy is better than 5% at the 99% confidence level (Howarth, 1998).

The density of bulk-rocks was determined by weighing the dry, clean sample in air and then in water, following Archimedes' principle. Repeated measurements on troctolite and gabbro samples (Namur & Charlier, 2012) indicate relative uncertainties between 0.001 and 0.012 g cm⁻³, with an average value of 0.007 g cm⁻³.

Augite–plagioclase–plagioclase dihedral angles (θcpp) were measured using a four-axis universal stage mounted on a Swift optical microscope, with a Leitz UM32 long working distance objective and a ×10 eyepiece. Populations of 50–130 single measurements were made on each sample, and we report the median value of these populations, θcpp, with the 95% confidence interval about the median calculated according to the method of Stickels & Hücke (1964).

RESULTS

Petrography

Background rocks

Background rocks are here defined as fractionated troctolites and gabbros cropping out in the interval between LZa* and UZb*; they form the host for the colloform bands, bowls, late-stage pegmatites and segregations. The petrography of these rocks has been described in detail by Hoover (1989a), and only the most important features are emphasized here.

Plagioclase, olivine, clinopyroxene, magnetite, ilmenite and apatite are the most abundant minerals, with accessory Ca-poor pyroxene, K-feldspar, quartz, biotite, amphibole and Fe–Cu–Ni sulfides. Background rocks from the MBS contain similar primocryst and interstitial phases to cumulates from the LS, but they differ from the latter in being more coarse-grained and unlaminated. They also contain more interstitial granophytic pockets (0–22 vol.%; Supplementary Dataset 2) dominated by apatite, quartz, K-feldspar and minor zircon and amphibole. Plagioclase is ubiquitous, forming randomly oriented and distinctively zoned, subhedral to euhedral, tabular grains, ranging in size from 0.2 to 15 mm. Olivine primocrysts occur from LZa* to LZc* and from UZa* to UZb*. They generally form 0.2–5 mm anhedral and slightly subhedral to subhedral grains. Some crystals are partially or completely replaced by serpentine. In MZ*, olivine is an interstitial phase occurring mostly as thin (e.g. 50–500 μm) coronitic textures around Fe–Ti oxides and clinopyroxene, as reported by Holness et al. (2011). Clinopyroxene is not a primocryst in LZa* and occurs as 0.5–10 mm zoned poikilitic grains, commonly enclosing rounded olivine and plagioclase laths. Clinopyroxene crystals very commonly contain abundant Ca-poor pyroxene exsolutions. Clinopyroxene from LZb* to UZb* forms large tabular anhedral to subhedral locally zoned primocrysts ranging in size from 0.5 to 8 mm. The largest grains are commonly strongly compositionally zoned and show evidence for overgrowth from interstitial melt. Ca-poor pyroxene (orthopyroxene and inverted pigeonite) is a minor interstitial phase in LZa*–LZb* and becomes a subordinate primocrystal phase from LZc*. Ca-poor pyroxene morphology evolves from relatively large (0.5–3 mm) poikilitic grains in the lowest stratigraphic parts to small (0.5–2 mm) subhedral tabular grains in the most evolved parts of the MBS. Fe–Ti oxides are represented by the ilmenite–hematite and magnetite—ulvospinel solid solutions. From LZa* to LZb*, they are interstitial phases and form small (<1 mm) anhedral patches dominated by ilmenite. From LZc* to UZb*, Fe–Ti oxides form large, commonly polycrystalline aggregates up to 2 cm across. Euhedral Fe–Ti oxides are locally present as inclusions in silicate phases. Apatite is an accessory interstitial phase in most of the MBS (LZa*–UZa*), and forms small (~50 μm) needle-shaped grains in the rims of silicate grains or in interstices between them. Significantly larger crystals are locally present in evolved late-stage liquid pockets and in patches of granophyre. In UZb*, apatite is a primocryst phase and forms euhedral millimetre-sized crystals.

Colloform bands and bowls

Plagioclase forms randomly oriented subhedral to euhedral lath-shaped grains, commonly with weak normal
zoning. Some colloform bands have a bimodal distribution of plagioclase size, with one group of grains of similar size to those observed in surrounding background rocks and a second group of significantly smaller grains. Olivine morphology varies from very anhedral, rounded and angular to perfectly euhedral. In MZ*, very few olivine grains are present and they form thin coronas around Fe–Ti oxides and clinopyroxene. In most samples where olivine is abundant (LZa*-LZc* and UZa*-UZb*), three-grain boundaries in olivine monomineralic regions are curved with 120° angles, suggesting textural equilibration. A bimodal grain-size distribution is common. Large olivine crystals are also locally sector twinned, reminiscent of those in rapidly cooled basaltic magmas (Dodd & Calef, 1971; Bryan, 1972; Clark et al., 1986). Clinopyroxene is ubiquitous in colloform bands, with morphology dependent on position within the MBS. In LZa*, clinopyroxene mostly forms oikocysts enclosing small plagioclase laths and rounded to euhedral olivine crystals. The grain size (0.5–20 mm) is generally similar to that in the surrounding troctolite, in contrast to olivine and plagioclase, which are finer-grained in the colloform than in the host-rock. To a lesser extent, clinopyroxene also occurs as millimetre-scale subhedral and randomly oriented tabular grains. Boundaries between adjacent clinopyroxene crystals (either oikocrysts or tabular grains) are commonly serrated and intergrown. From LZb* to UZb*, clinopyroxene is represented by 0.2–2 mm subhedral to perfectly euhedral tabular-shaped grains, commonly with simple twinning. Contacts between adjacent clinopyroxene crystals may also locally be serrated and intergrown. Low-Ca pyroxene (mostly inverted pigeonite) is a minor phase in most colloform bands and generally forms anhedral to slightly poikilitic, small (<d mm), crystals in close association with clinopyroxene. Fe–Ti oxide minerals are very minor phases in LZa*-LZb* colloforms, where they form small (<0.5 mm) anhedral patches generally dominated by ilmenite. Very small (<0.5 mm) euhedral Fe–Ti oxide grains were, however, observed in some LZa* colloform samples. From LZh* to UZb*, Fe–Ti oxides are much more abundant and generally form large (up to 2 cm) anhedral polycrystalline patches, but some colloforms also show small (1–5 mm) perfectly euhedral crystals of magnetite and ilmenite. Apatite is a minor phase from LZh* to UZa*, occurring as small (50–100 µm) needle-shaped grains. In UZb*, apatite is very abundant, forming euhedral millimetre-size crystals.

**Mineral modes**

Modal proportions have been determined for all the colloform samples, immediately adjacent gabbros on both sides of the colloform bands, and in gabbros at least 2 m away from any colloform (Supplementary Dataset 2). The colloform bands always contain a significantly higher proportion of mafic minerals ± apatite and a lower proportion of plagioclase than nearby gabbros (Figs 11 and 12). The amount of granophyre is also lower in colloforms than in surrounding gabbros (e.g. C-SKII-C12; Fig. 13; location shown in Fig. 10). The mineral association in colloforms is similar to that observed in gabbros, with olivine and clinopyroxene being the dominant phases in LZa* and LZb*, joined by Fe–Ti oxides in LZc*, LZa* and UZa* and apatite in UZb*. The outwards contact (i.e. closest to the intrusion margin) of the colloform bands always shows a sharp contrast of mineral modes with adjacent gabbros, whereas the inward contact can be sharp (e.g. C-SKII-C5; C-SKII-C7; Fig. 12; locations shown in Fig. 10) or gradational (e.g. C-SKII-C3; C-SKII-C5; Fig. 12; location shown in Fig. 10); that is, with a progressive inward modal
decrease of mafic minerals and a corresponding progressive modal increase of plagioclase. When the colloform contact is sharp on both sides, mineral modes in the adjacent gabbro on each side are generally similar (e.g. C-SK11-C5; Fig. 12; location shown in Fig. 10), although significant differences are locally observed, especially for olivine modal proportion (C-SK11-C1 and band A in C-SK11-C1; Fig. 12; locations shown in Fig. 10).

In a set of closely spaced colloform bands, mineral modes can be either relatively constant from one band to another, or can change significantly. This is clearly illustrated by the plagioclase/(plagioclase + mafic minerals) and olivine/(olivine + pyroxenes) ratios in the five colloform bands of the C-SK11-C14 colloform set (LZb*; Fig. 13).

**Augite–plagioclase–plagioclase dihedral angles**

The median augite–plagioclase–plagioclase dihedral angle, $\Theta_{_{\text{dih}}}^{\text{avg}}$, was measured across two colloform bands, one in LZa* (C-SK11-C3) and the other from MZ* (C-SK11-C5; locations shown in Fig. 10; Supplementary Dataset 3).
The colloform C-SK1 1-C3 is 36 cm wide, with a sharp outer margin, defined by a decrease in grain size and an increase in olivine mode, and a gradational inner margin. The mineral mode returns to the same as that of the background gabbro 30 cm from its sharply defined outer margin (Fig. 12). This colloform was sampled using a drill to collect six samples from the colloform itself, with three samples of the host gabbro on the outer margin and a further three on the inner margin. The host gabbro has \( \Theta_{\text{cpp}} \) of 86° ± 2°, typical of the gabbros in this region of the MBS (M. B. Holness, unpublished data). It increases sharply to 93° ± 2° at the outer, sharply defined edge of the colloform and decreases steadily back to 86° inward through the colloform; by extrapolation it returns to this background value 85 cm from the outer margin of the colloform, well outside the zone of plagioclase depletion marking the site of the colloform in the field (Fig. 14).

Colloform C-SK1 1-C5 is 6 cm thick, well-defined, and relatively oxide-rich, with a grain size almost the same as that of the host gabbro. We took two drilled samples from the colloform, two from the gabbro on the outer margin, and one from the gabbro on the inner margin (Fig. 10). The host gabbro has \( \Theta_{\text{cpp}} \) of 100° ± 2°, typical of the gabbros in this part of the MBS (M. B. Holness, unpublished data) whereas \( \Theta_{\text{cpp}} \) in the colloform is 104° ± 4°, and returns to the background value 4 cm from the well-defined inner margin (Supplementary Dataset 3).

**Bulk-rock compositions**

Colloform and gabbro samples are fresh when observed under the microscope, consistent with their low
loss-on-ignition (LOI) values (Supplementary Dataset 4). Major element composition reflects modal mineralogy and the colloform samples are significantly enriched in elements partitioning into mafic minerals (e.g. FeO, MgO, MnO) and depleted in elements enriched in plagioclase (Al₂O₃, Na₂O) compared with surrounding gabbros. The difference in CaO content between colloforms and gabbros is highly variable and depends on the plagioclase clinopyroxene ratio. In UZb*, the bulk-rock P₂O₅ is higher in colloforms compared with gabbros, reflecting a higher modal proportion of apatite. Trace elements that partition into mafic phases (Ni, Cr, V, Co) are higher in colloforms than in gabbros, whereas elements partitioning into plagioclase (Sr, Ba) are lower. Incompatible minor and trace element (Zr, Y, and P in LZa*–UZa*) contents are always low in colloform samples, suggesting a fraction of interstitial liquid of 4–6 wt%, with an average of 14 wt% [based on 43 samples and calculated using liquid compositions from Thy et al. (2009); Supplementary Dataset 4]. This interstitial liquid fraction is lower than that observed in the surrounding gabbros (10–70 wt%; average 29 wt% based on 48 samples). Both colloform and MBS gabbro samples, however, have an interstitial liquid fraction higher than most samples from the LS, especially from LZc to the top (Tegner et al., 2009).

Mineral compositions

We analyzed mineral core compositions (olivine, plagioclase, clinopyroxene, ± low-Ca pyroxene) in colloform and in adjacent gabbros on both sides of the colloform bands. The An content [molar Ca/(Ca + Na)] of plagioclase is, within error, the same in the colloform samples and surrounding gabbros samples (Fig. 13; Supplementary Dataset 5). Plagioclase from some colloform bands has slightly lower FeO and Or contents [molar K/(K + Ca + Na)] compared with adjacent gabbro samples, but no significant difference is observed for compatible (Sr, Ba; Supplementary Dataset 6) and incompatible (REE, Zr, Y, Rb) trace elements. Olivine is always characterized by a significantly different Fo content [molar Mg/(Mg + Fe)] in colloforms and gabbros (Figs 15a, 5b and 16a; Supplementary Dataset 7), generally 2–5% higher in the colloform compared with the gabbro. In contrast, the Fo content of olivine is 2% lower in section C-SKII-C2 (UZb*; location shown in Fig. 10). The chemical contrast between colloform and gabbro is always sharp on the outwards side of the colloform band and is generally gradational on the inward size of the band.

A few colloform samples (and more rarely host gabbros on the inward contact) show well-defined bimodality of olivine compositions. One endmember composition is
always identical to the olivine composition observed in the external gabbros, whereas the other is significantly more primitive (higher Fo content). Compatible trace elements (Ni, Co, V, Cr; Supplementary Dataset 8) are enriched in the colloform olivines compared with gabbro olivines, whereas incompatible elements (Y, Zr, Sc) are slightly lower. Other elements such as Li, P and Al show significant variability both from grain to grain and within grains, potentially obscuring any compositional contrast between colloform and gabbro. Comparing clinopyroxene composition in colloform and gabbros is not straightforward because clinopyroxene grains are characterized by significant core-to-rim and sector zoning and contain abundant exsolution lamellae of low-Ca pyroxene and Fe–Ti oxides at a scale generally smaller than the microprobe spot-size (1 µm). Nevertheless, in most traverses the Mg# [molar Mg/(Mg + Fe)] of clinopyroxene in colloform samples is slightly higher (1–4%) than in the surrounding gabbros (Figs 15a, b and 16b; Supplementary Dataset 9). No significant difference is observed for minor elements (Al₂O₃, TiO₂, MnO). Low-Ca pyroxene was analyzed in a single section from LZa* (C-SK11-C3; location shown in Fig. 10) and the Mg# is significantly higher (3–5%) in the colloform than in surrounding gabbros (Fig. 15a; Supplementary Dataset 10).

FORMATION OF THE COLLOFORM BANDS

The colloform bands are typically highly asymmetric, with abrupt changes in grain size, mineral mode, composition of mafic minerals and augite–plagioclase–plagioclase dihedral angle on their outwards margin, and a smooth progression back to more ‘normal’ values on their inner side. The colloform banding was previously interpreted as the result of crystal deposition during vigorous convection (Wager & Brown, 1968), but for several reasons this cannot be correct. First, the egg-box shape of the bands and the development of elongated fingers (up to 2 m long) are inconsistent with any convective process that might act on the surface of the steeply dipping MBS mush, which was broadly planar, as demonstrated by the smooth contacts between the different stratigraphic zones. Second, various geochemical and textural arguments (e.g. loss of rhythmic layering in the LS) suggest that convection ceased during the final stage of crystallization in the magma chamber (UZa–UZc; F <0·2; Boudreau & McBirney, 1997; McBirney & Nicolas, 1997; Jang & Naslund, 2001). Development of colloforms and bowls in the most evolved parts (Upper Zone) of the MBS therefore cannot be related to any convection-based process. For the same reasons, we can also rule out processes such as oscillatory nucleation (McBirney & Noyes, 1979; Morse, 1979) and contrasted rates of crystal growth (Boudreau, 1995), which also occur at the mush–magma interface. However, the clear asymmetry of the colloforms does suggest that they formed through a directional process.

The cuspate morphology of the colloforms, together with the fingers and bowls, is highly reminiscent of reaction fronts described both in natural porous rocks and in experimental analogues involving infiltration of liquid into a porous medium (Ortoleva et al., 1987a, 1987b;

Most of the morphological features we describe here have been documented in infiltrated sedimentary rocks, or modeled theoretically or experimentally, as follows.

1. The sinuousoidal or cuspatate shape of colloform bands (Fig. 4) is identical to that predicted for reaction fronts owing to reactive infiltration, where flow is focused in high-permeability pathways (Chadam & Ortoleva, 1990).

2. The development of fingers and isolated bowls (Fig. 4) has been described in uranium ore roll fronts (Granger & Warren, 1969) and interpreted as redox reaction fronts owing to water flows through a pyrite-bearing porous sandstone (Ortoleva et al., 1986). Fingering and development of high-porosity channels have also been recognized as important pathways of reactive liquid migration in the upper mantle (Kelemen et al., 1995b, 1997; Liang et al., 2010).

3. Repeated banding inside the fingers (Figs 6 and 7), and the sets of closely spaced bands (Fig. 4) have been described in sedimentary rocks (iron oxide banding in sandstones) and interpreted as self-organization owing to infiltration instability within a cycle of nucleation driven by supersaturation followed by depletion (Ortoleva et al., 1987a).

4. Offsets between fingers of a set of grouped reaction bands have been ascribed to decreasing permeability at the reaction front (finger tip) that diverts the flow between the first set of fingers and generates a second set downstream (Ortoleva, 1994).

These similarities between colloform morphology and reaction fronts observed in natural rocks and experimental porous media point towards an origin of the colloform banding by lateral fluid or melt migration within the crystal mush.

**Source of the infiltrating melt**

**Volatile-rich fluid vs silicate melt**

If colloform bands and bowls represent reaction fronts owing to fluid or melt migration within the MBS crystal mush, we must determine the nature of the infiltrating phase; that is, whether this is a volatile-rich, metasomatizing, aqueous fluid, an anhydrous silicate melt or a hydrous silicate melt. Supercritical CO2–H2O–F–Cl–S fluids are thought to exsolve from highly evolved interstitial melts within layered intrusions. Migration of such fluids may locally cause differential mineral dissolution (McBirney, 1987; Boudreau & McCallum, 1989; Boudreau, 1999; Kelemen et al., 2011). Volatiles (H2O, F, Cl) in silicate melts also have a major effect on mineral–liquid phase equilibria (Berndt et al., 2005; Feig et al., 2006, 2010; Botcharnikov et al., 2008), changing the relative stability of silicate and oxide phases and mineral compositions (Panjasawatwong et al., 1995; Berndt et al., 2005; Putirka, 2005). Collofoms could therefore be suggested to correspond to (1) relic rocks left behind after fluid migration or (2) rocks having crystallized from a hybrid liquid as a result of mixing between the resident mush liquid and the liquid produced by mineral dissolution during fluid migration. However, the absence of hydrous phases such as biotite or amphibole in collofoms and bowls and the absence of a plagioclase compositional contrast between collofoms and adjacent gabbros, together with unchanging mineral assemblages between colloform and surrounding gabbros, suggest that neither fluids nor hydrous silicate melts were responsible for colloform banding. The presence of abundant pyroxene and olivine in many collofoms and the surrounding gabbros is also inconsistent with the expected highly contrasted rates of dissolution for pyroxene and olivine during fluid migration. Furthermore, the presence of abundant colloform bands in the lowest stratigraphic part of the MBS (LZa*–LZb*) is not consistent with a significant aqueous component, because the proportion of residual interstitial melt must be extremely low at the saturation of a volatile-rich fluid phase. There is also no geochemical evidence (either bulk-rock or mineral compositions) supporting a high volatile content (i.e. high concentrations of K, REE, Sr, Ba).

We therefore conclude that neither hydrous silicate melts nor supercritical fluids can be responsible for the formation of the colloform bands. In contrast, the similar mineral assemblages in collofoms and gabbros point towards an infiltrating liquid that shared many chemical characteristics with the liquid that crystallized the background gabbros; in other words, a dominantly dry mafic silicate melt. The precise composition of this melt, and the mechanism driving melt migration, probably controlled the mineralogical and geochemical diversity we observe in the colloform bands.

**Direction of melt migration and driving force for infiltration**

Experimental and theoretical simulations have convincingly shown that self-organization in porous media results in scalloped reaction fronts with cusps and fingers always pointing in the direction of the flow (Ortoleva et al., 1986, 1987a; Wei & Ortoleva, 1990; Heinrich et al., 1991; Schneider & Beckermann, 1995). By analogy, in the Skaergaard MBS, the direction of the cusps and fingers, the marked asymmetry of mode and microstructure, together with the convexity of the banding within the fingers and the shape of isolated bowls, all indicate that silicate liquid flow was focused horizontally from the center of the intrusion towards the margin. This therefore suggests that the source of the infiltrating liquid was the main magma body, or at least, interstitial melt from younger parts of the crystal mush on the chamber walls.
Evidence for melt migration within crystal mushes has been observed in floor cumulates of numerous layered intrusions and has also been recognized in the upper mantle and oceanic crust (e.g., Wager & Brown, 1968; Bédard & Hébert, 1996; Caughter, 1996; Natland & Dick, 1996; Kellemen et al., 1998; Googan et al., 2000; Lissenberg & Dick, 2008). In these settings, the driving force for liquid migration is gravitational, generally through compaction (McKenzie, 1984; Meurer & Boudreau, 1998; Liang, 2008; Tegner et al., 2009; Namur & Charlier, 2012). However, buoyancy-driven flow is unrealistic in the MBS because the infiltration direction is lateral not vertical. However, experimental studies of metal alloy crystallization and theoretical simulations have shown that another possible mechanism driving liquid migration in porous media forming on walls is volume contraction, or shrinkage (Heinrich et al., 1991; Chiang & Tsai, 1992; Worster, 1992; Schneider & Beckermann, 1995). This mechanism has also been proposed for solidifying magma chambers, although no supporting field evidence has yet been presented (Petersen, 1987). Lateral flow in these cases is caused by the density difference between the liquid and solid phases.

In basaltic magma chambers such as Skaergaard, the density of the magma (Fig. 17a) is significantly lower than that of the solidcumulate that eventually forms (Fig. 17b), especially after the saturation of Fe–Ti oxide minerals. This means that the mush undergoes significant shrinkage during the early stages of solidification. When the mush becomes a coherent, rigid framework able to support its own weight, the bulk mush cannot shrink towards the wall to accommodate the volume change (Petersen, 1987), because the crystal framework cannot be deformed by the low resultant stress. Instead, interconnected melt within the mush responds to the pressure gradient by being sucked towards the solidifying margin (Petersen, 1987). This pressure gradient, and consequent liquid suction, should begin to act as soon as the mush forms a structurally coherent framework and is able to transmit stress (Petford, 2003; Lavallée et al., 2007; Champallier et al., 2008). The crystal fraction at which this occurs is poorly constrained but could be between c. 30 and 50%, depending on crystal shape (Philpotts & Carroll, 1996; Vigneresse et al., 1996; Philpotts et al., 1998; Saar et al., 2001; Jerram et al., 2003; Laumonier et al., 2011; Picard et al., 2011).

A semi-quantitative estimation of how much shrinkage, and therefore how much liquid suction, may have occurred in the Skaergaard MBS is provided by comparing the liquid density with the density of the fully crystallized rocks (Fig. 17; Supplementary Dataset 1). This is based on the assumption that the MBS solidified by in situ crystallization, so that the bulk-rock composition at any stratigraphic point reflects the composition of the bulk magma at the time of crystallization of that point. Considering a minimum initial porosity of 25vol.%, the difference in density between liquids and measured rocks is in the range of 5–10% for LZa*–LZb*, increasing to 10–16% in LZc*–UZb*. These values are lower estimates for shrinkage in the Skaergaard MBS, but might be reduced by 1–2% owing to thermal expansion (Niu & Batiza, 1991) at the liquidus temperature (1000–1170°C). They are found to be slightly higher than contraction values of c. 7vol.% observed in most metal alloys (Chiang & Tsai, 1992). This suggests that liquid-driven infiltration owing to shrinkage might be an important process in slowly solidifying igneous systems.

**DISEQUILIBRIUM IN THE CRYSTAL MUSH**

**Reactive infiltration in the crystal mush**

Mineral modes and compositions, together with the morphology of the colloform bands, suggest that banding results from a process of infiltration of a silicate liquid into the crystal mush owing to shrinkage, followed by crystallization. The mush liquid is probably either displaced by the infiltrating melt or mixed with it during the infiltration process. We can place some constraints on the most realistic scenario of these two end-members, using phase equilibria (see below). We first note that infiltration of a chemically unmodified Skaergaard silicate melt cannot have produced the parental magma of the colloform bands because no liquid along the Skaergaard liquid line of descent is able to crystallize rocks with the mineral modes we observe in the colloforms. This implies that the composition of the liquid must have been changed during infiltration and before crystallization. This can be achieved either by simple mixing between mush and infiltrating liquid or by dissolution of mineral phases in the mush owing to thermo-chemical disequilibrium between the infiltrating liquid and the crystalline framework surrounding it.

Simple mixing between the infiltrating melt and the melt initially present in the mush can be excluded using arguments based on phase diagrams. The anorthite–forsterite–diopside phase diagram of Osborn & Tait (1952) is adequate to investigate mixing processes in LZa*–LZb*. The crystal mush on the margins of the magma chamber comprises a strong crystal framework containing a fully interconnected, continuously evolving liquid. Mixing between a mush liquid either at the ternary plagioclase–olivine–clinopyroxene eutectic (LZh*), or anywhere on the plagioclase–olivine cotectic (LZa*), with a primitive LZa* liquid will produce hybrid liquids along the essentially planar plagioclase–olivine cotectic. The mixing process will therefore not change the cotectic proportions crystallizing from the mush liquid and cannot produce the observed colloform bands that are dominated by mafic minerals and depleted in plagioclase. In more
complicated systems, especially those saturated in Fe–Ti oxides (from LZc* to UZb*), cotectic reactions are generally not planar (Morse et al., 2004; Charlier et al., 2010). Magma mixing will produce liquids that fall into the stability field of a single phase, in contrast to the observation that colloform bands are polymineralic and therefore crystallized from multi-saturated melts.

As shown above, a simple magma mixing process is not able to produce a parental melt to the colloform bands. We therefore suggest that the resident mush liquid is continuously displaced by the infiltrating melt. However, given the hot and primitive nature of this infiltrating liquid, significant thermo-chemical disequilibrium is expected between liquid and solid, especially for evolved interstitial phases (Brandeis & Jaupart, 1986; Worster et al., 1990, 1993; Worster, 1992; Jupp & Woods, 2003). Depending on the extent of disequilibrium, this can result in reactive dissolution of pre-existing phases. Partial dissolution of
minerals in the crystal mush can explain the change in mush liquid composition to one capable of crystallizing the colloform bands with high proportions of primitive mafic minerals.

This interpretation is consistent with the presence of fingers in the colloform bands, which are thought to represent high-porosity channels owing to dissolution (Chadam et al., 1987, 1988; Ortoleva et al., 1987a, 1987b; Chadam & Ortoleva, 1990; Raw & Woods, 2003); it is also consistent with the presence of two distinct grain populations in most colloforms. Many colloforms contain two distinct populations of olivine and clinopyroxene grains, distinguishable by composition and/or size, with one population similar to the composition and/or size of the adjacent outer host gabbro and the other more primitive or smaller. This is consistent with the coexistence of relic grains from the original mush framework and mineral grains formed after infiltration and partial dissolution. The absence of bi-modal mineral populations in the other colloforms could be due to either complete dissolution of original mush grains before crystallization of the colloform (i.e. absence of relic grains), or re-equilibration of both the relic grains and the newly crystallized grains by the interstitial melt.

**Formation of the colloform parental melt**

To understand better the details of the proposed reactive process, we have modeled the geochemical consequences of mineral dissolution in the Lz*b* Skaergaard crystal mush using empirically and thermodynamically derived equations to predict liquidus mineral compositions (Toplis & Carroll, 1996; Wood & Blundy, 1997; Namur et al., 2012) and equilibrium cotectic proportions (PETROLOG, Dan-yushevsky & Plechov, 2011; MELTS, Ghiorso & Sack, 1995). We simulated the infiltration of a slightly hotter, more primitive, Skaergaard liquid corresponding to the base of Lz*b* (i.e. the outer part of Lz*b*) into a slightly cooler, more evolved, crystal mush containing solid phases typical of the top of Lz*b* (i.e. the inner part of Lz*b*), using liquid and mineral compositions from Thy et al. (2009) and Thy et al. (2006), respectively. In our simulation, the initial liquid mush is displaced during liquid migration. Minor liquid mixing between resident and infiltrating liquids would not significantly change the results presented below. We chose an Lz*b* liquid and crystal assemblage because (1) this unit is relatively simple, with only three coexisting primocryst phases, (2) there is a good agreement in the literature on the composition of Lz*b* liquids, unlike more evolved liquids (Hunter & Sparks, 1987; Toplis & Carroll, 1996; Tegner, 1997; Thy et al., 2009); and (3) the experimental calibration of MELTS and PETROLOG is robust for Fe–Ti oxide unsaturated melts (Gaetani, 1998).

We performed two sets of calculations (Supplementary Dataset 12): (1) liquid infiltrating into a mush of 75% crystals, 25% liquid; (2) liquid infiltrating into a mush of 25% crystals, 75% liquid. In each case, we simulated both mineral dissolution in cotectic proportions (Grove et al., 1992; Yang et al., 1996) and dissolution of plagioclase, olivine and clinopyroxene respectively. After each increment of dissolution, we calculated mineral cotectic proportions and mineral compositions that would be expected if some process caused rapid crystallization. Calculations for both initial porosities give similar trends for mineral cotectic proportions and mineral compositions (Figs 18 and 19), but the effects described below are greater for the system with low initial porosity (25%).

Progressive dissolution of minerals in cotectic proportions produces liquids that would crystallize increasing proportions of plagioclase and olivine and decreasing cotectic proportion of clinopyroxene (Fig. 18a and b). Mineral compositions in equilibrium melts are increasingly primitive (higher plagioclase An content, higher olivine Fo content and higher clinopyroxene Mg#) as dissolution proceeds (Fig. 19a and b). Progressive dissolution of plagioclase, olivine or clinopyroxene alone would produce hybrid liquids enriched in the dissolving mineral component. Crystallization of these liquids would therefore produce rocks dominated by a single phase (Fig. 18c–f), with a composition dramatically more primitive than that of the same phase that dissolves in the mush (Fig. 19c–f).

Results of calculations presented in Figs 18 and 19 indicate that the only way to obtain liquids capable of crystallizing the colloforms is preferentially to dissolve mafic minerals (olivine and clinopyroxene) from the crystal mush, without significant contribution from plagioclase dissolution. Crystallization of such liquids would form rocks with high proportions of primitive mafic phases, as observed in the colloform bands. The absence of significant plagioclase dissolution would explain the unchanged plagioclase composition from colloform bands to adjacent gabbros.

**Thermal or chemical dissolution?**

In the simplified phase diagram anorthite–forsterite–diopside (Osborn & Tait, 1952), the mechanism of dissolution described above can be understood in terms of dissolution of the low-melting fraction of the most evolved crystals of olivine and augite. This process temporarily drives the liquid to supersaturation in forsterite and diopside (i.e. along the cotectic curve), resulting in a liquid potentially capable of crystallizing the colloform bands. The dissolution process outlined above therefore requires differential dissolution rates for plagioclase compared with olivine and clinopyroxene. Experimental studies of thermally induced mineral dissolution in basaltic melts have been performed by adding crystals into a basaltic liquid at high temperature. Accurate measurements of crystal size before and after the experiments allow mineral dissolution rates to be constrained as a function of time and temperature. It was found that plagioclase dissolves faster than olivine and clinopyroxene in high-temperature magmas such as
Skaergaard basalts (Donaldson, 1985; Brearley & Scarfe, 1986; Edwards & Russell, 1996). Thermally induced mineral dissolution in the Skaergaard crystal mush would therefore produce hybrid melts with high-An plagioclase as the dominant liquidus phase (Figs 18 and 19). This is in strong contrast to the mineral modes and compositions observed in the colloform bands, and suggests that thermal dissolution cannot have been the dominant mechanism of mineral dissolution in the Skaergaard crystal mush. Instead, we suggest that dissolution was controlled by chemical disequilibrium, which has been shown to significantly alter mineral modes and chemistry during reactive porous flow (Kelemen et al., 1995a; Lissenberg & Dick, 2008). This process may be understood in terms of activities...
(a) of mineral component $i$ (e.g. An, Fo) in the liquid and the solid phase. At temperature $T$, liquid and a mineral phase are in thermodynamic equilibrium when the chemical potentials ($\mu$) of component $i$ are identical in the solid and the liquid. The chemical potential of $i$ is related to the activity ($a$) through a relationship involving temperature ($T$) and the Gibbs’ free energy ($G_i(T)$) of the pure component $i$ at temperature $T$. For a given mineral composition ($a_{i,Sol}$) there is therefore a single value of $a_{i,Liq}^{eq}$ at equilibrium. Any other value of $a_{i,Liq}$ results in chemical disequilibrium between the solid and liquid phase. Whether the solid phase will significantly dissolve or not depends on the difference between $a_{i,Liq}^{eq}$ and the actual value of $a_{i,Liq}$.

Activities of plagioclase (Ab, An components), olivine (Fo, Fa components) and clinopyroxene (Wo, Ens, Fs components) in the liquid phase depend on the activities of numerous oxides, the most important being $a_{CaO}$, $a_{Na_2O}$, $a_{Al_2O_3}$, $a_{SiO_2}$, $a_{FeO}$ and $a_{MgO}$. Currently existing thermodynamic models are insufficient to predict the way...
in which the activity of these oxide components varies as a function of liquid composition (Glazner, 1984; O’Neill, 2005; Toplis, 2005; Namur et al., 2012). Here, we use a simplified model and describe mole fractions \( X \) of melt components as \( X_{\text{An,Liq}} = \text{Ca} / (\text{Ca} + \text{Na}) \) and \( X_{\text{Fo,Liq}} = \text{Mg} / (\text{Mg} + \text{Fe}^{2+}) \). One-atmosphere experimental data from Toplis & Carroll (1995) show that \( X_{\text{An,Liq}} \) and \( X_{\text{Fo,Liq}} \) can be related to \( X_{\text{An,Sol}} \) and \( X_{\text{Fo,Sol}} \) through \( K_d \) values of 1.02 and 0.31, respectively. The evolution of \( X_{\text{An,Liq}} \) and \( X_{\text{Fo,Liq}} \) has been investigated as a function of the residual liquid fraction \( F \) (1–0) for the Skaergaard liquid lines of descent proposed by Toplis & Carroll (1995), Thy et al. (2009) and Tegner & Cawthorn (2010). \( X_{\text{An,Liq}} \) and \( X_{\text{Fo,Liq}} \) decrease continuously as a function of \( F \) (Fig. 20a and b), but the extent of \( X_{\text{An,Liq}} \) decrease is significantly less important than that of \( X_{\text{Fo,Liq}} \). This is presumably related to the high fraction of olivine and pyroxene crystallized during the first stages of differentiation of the Skaergaard magma (LZa and LZb; Tegner et al., 2009). As a consequence \( X_{\text{An,Liq}} / X_{\text{Fo,Liq}} \) increases significantly with liquid differentiation (Fig. 20c). This has a major influence on solid–liquid chemical disequilibrium during infiltration. For a given difference of liquid composition (or \( F \)) between the initial mush liquid and the new infiltrating liquid, the chemical disequilibrium between olivine (or clinopyroxene) and the new liquid will be much greater than for plagioclase. A significantly higher degree of olivine and clinopyroxene dissolution is therefore expected compared with that of plagioclase. Chemical dissolution of mafic phases was probably helped by rapid differentiation of the high mush liquid fraction in MBS rocks (Hoover, 1989a), efficient crystallization of evolved pore phases and strong compositional contrast between those phases and the infiltrating melt. The relatively large surface-to-volume ratio of the small interstitial grains will also contribute to their instability (i.e. high Gibbs’ free energy) compared with large cumulus grains, and will increase their propensity to dissolve. Such a process of chemical dissolution might also have been helped by pressure solution at crystal edges where stress is highest (Boudreau & Mc Birney, 1997).

**Solidification of the colloform bands**

In the previous sections, we have shown that it is possible to produce a colloform parent melt by (1) shrinkage of the crystal mush producing suction of primitive melt from the main magma chamber (or the internal, younger part of the mush) into the cooler, more evolved, crystal mush closer to the intrusion margin (Fig. 21) and (2) chemical disequilibrium between the infiltrating liquid and pre-existing evolved solid phases resulting in dissolution of evolved mafic minerals, whereas plagioclase is not significantly affected (Fig. 21). The invariably sharp external margin of the colloform bands, the small grain size of their constituent minerals and the unusual textures (e.g. sector twinning) demonstrate that the solidification of the infiltrating melt to form the colloform banding required an abrupt crystallization process (e.g. an overnucleation event), generally referred to as a quenching event. Quenching of the colloform bands is also consistent with previous interpretations of high \( \Theta_{\text{CIP}} \) values that have been observed in rocks where abundant nucleation was triggered by silicate liquid infiltration in a slowly solidifying crystal mush (e.g. the Wavy Horizon, Rum; Holness et al., 2007).

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**Fig. 20.** Chemical evolution of proposed Skaergaard liquid lines of descent as a function of residual liquid fraction \( F \) in the magma chamber. (a) Evolution of the molar ratio \( X \) of An in the liquid \( X_{\text{An,Liq}} = \text{molar Ca} / (\text{Ca} + \text{Na}) \). (b) Evolution of the molar ratio of Fo in the liquid \( X_{\text{Fo,Liq}} = \text{molar Mg} / (\text{Mg} + \text{Fe}) \). (c) Evolution of \( X_{\text{An,Liq}} / X_{\text{Fo,Liq}} \). Data are calculated from the Skaergaard liquid compositions of Toplis & Carroll (1995), Thy et al. (2009) and Tegner & Cawthorn (2010).
Quenching of the colloform-forming liquid might be related either to critical thermal disequilibrium between the crystal mush and the infiltrating liquid (Brandeis et al., 1984; Hort, 1998; Richet & Ottonello, 2010) or to supersaturation of some mineral components (e.g. mafic minerals) in the infiltrating liquid (Ballhaus, 1998). However, strong thermal disequilibrium in the crystal mush is not likely to have triggered the crystallization of the colloform-forming liquid. This is because the absence of significant plagioclase dissolution during liquid infiltration, as shown in the previous section, does not support a very steep thermal gradient in the MBS crystal mush.

Instead, we suggest, following experimental work demonstrating that addition of a chromite component to a mafic silicate melt drives the liquid towards chromite supersaturation and results in over-crystallization of chromite (Ballhaus, 1998), that crystallization of the colloform bands is triggered by supersaturation of olivine and pyroxene (e.g. olivine and clinopyroxene) components in the infiltrating melt. As dissolution of evolved mafic phases in the mush proceeds, the melt becomes increasingly supersaturated in those phases (Schmidt et al., 2012). When some critical supersaturation state is reached, the melt abruptly starts to crystallize primitive mafic phases in a high-porosity region of the mush. During quenching, mineral proportions are controlled not only by the liquidus phase assemblage but also by the relative complexity of mineral structures, which also contributes to a high ratio of structurally simple mafic minerals (olivine, clinopyroxene, Fe–Ti oxides) over plagioclase (Gibb, 1974; Corrigan, 1982; Tsuchiyama, 1983; Burkhard, 2005). The overall effect of this quenching event is to produce ultramafic bands with a composition strongly contrasting with the surrounding gabbros. We suggest that the thin plagioclase-rich band (1–2 cm thick) observed at the external contact of some colloform bands (Fig. 7) may arise from progressive depletion of the liquid in cations that are incorporated into the mafic minerals (e.g. Fe, Ti, Mg) such that the last fraction of residual liquid might eventually reach plagioclase saturation. Once fully crystallized, the fine grain size of the colloform bands means that they act as low-permeability barriers, hampering further liquid migration towards the intrusion margins.

**IMPLICATIONS FOR MUSH THICKNESS**

The unequivocal relationship between colloform bands and the development of late magmatic segregations (compare the Wavy Pyroxene rock; Humphreys & Holness, 2010) in the outer parts of the MBS suggests that crystallization of the colloform bands occurred relatively early in the solidification history of the Skaergaard mush. The presence of colloforms affects the evolution of the late-stage segregations (Fig. 9b), indicating that they formed after the crystallization of the colloform bands. Nevertheless, the morphology of the colloform bands clearly changes with increasing distance from the intrusion margin, suggesting that the physical characteristics of the crystal mush might have changed with differentiation. One possible explanation for this morphological evolution, from almost planar bands near the margin to deeply scalloped and fingered bands close to the inner margin of the MBS, could be a progressive increase of the crystal mush thickness and distance of liquid migration within the mush. Numerical simulations of flow in porous media with an
initially homogeneous porosity structure suggest that increasing the distance of liquid migration results in a propagating liquid front progressively changing from a planar morphology to one resembling a three-dimensional egg-box (Zhao et al., 2008). Moreover, increasing the mush thickness is likely to result in a heterogeneous distribution of the initial porosity (i.e. before liquid infiltration; Neufeld & Wettlaufer, 2008, 2011). Liquid infiltration and reactive dissolution preferentially occur in mushy regions with higher porosity, resulting in a progressively channelized and finger-like liquid flow as dissolution proceeds. The observed morphological evolution of the colloform bands with distance from the intrusion margin could therefore be explained by the crystal mush thickness increasing with progressive differentiation.

The data acquired for this study, however, do not allow us to place accurate constraints on the MBS mush thickness. We note that some of the colloform fingerling instabilities attain lengths of 2 m (from LZb* to MZ*), so the minimum mush thickness must be greater than this. Concerning the highest value, we regard the total absence of any change in primocryst assemblages in the colloforms compared with the surrounding gabbros as evidence for relatively short length-scale liquid migration within the mush. This is consistent with a maximum crystal mush thickness of a few metres to a few tens of metres.

CONCLUSIONS

Our re-examination of the Banded Division of the Skaergaard Marginal Border Series demonstrates that the colloform banding was not formed at the magma–mush interface by crystal-laden convective currents as envisaged by Wager & Brown (1968). Instead, it formed in a pre-existing crystal framework as a consequence of the infiltration of relatively primitive silicate liquids from the external part of the crystal mush or the main magma body. Fluid flow was driven by contraction of the crystal mush during solidification. Infiltration of primitive liquid into the crystal mush resulted in chemical disequilibrium between pre-existing solid phases and infiltrated liquid. This resulted in partial dissolution of small grains of evolved mafic phases, creating a liquid capable of crystallizing high proportions of primitive mafic phases. Critical supersaturation of some mafic mineral components in the infiltrating melt produced abrupt crystallization of the hybrid infiltrating liquid, producing a single colloform band, dominated by relatively mafic olivine and clinopyroxene, as well as Fe–Ti oxides and apatite in the most evolved parts of the Marginal Border Series. The numerous colloform bands point to the repeated action of this process during the formation of the Marginal Border Series. The morphological evolution of the colloform bands, from planar to deeply fingered with increasing distance from the intrusion margin, might potentially result from increasing crystal mush thickness with differentiation.

The Skaergaard Marginal Border Series thus records a wealth of phenomena related to horizontal fluid infiltration, resulting in chemical disequilibrium in the crystal mush of a slowly solidifying system pinned in the vertical dimension. Although this mechanism is thought to be the dominant process in producing mineral banding, other processes of mineral dissolution involving supercritical fluids (Boudreau, 1999) or pressure-solution at crystal edges (Boudreau & McBirney, 1997) might have enhanced pre-existing banding or been responsible for minor layering features within the Marginal Border Series.

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SUPPLEMENTARY DATA

Supplementary data for this paper are available at Journal of Petrology online.

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