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Large-scale silicate liquid immiscibility during differentiation of tholeiitic basalt to granite and the origin of the Daly gap

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ABSTRACT

The dearth of intermediate magmatic compositions at the Earth's surface, referred to as the Daly gap, remains a major issue in igneous petrology. The initially favored explanation invoking silicate liquid immiscibility during evolution of basalt to rhyolite has lost support because of the absence of any firm geological evidence for separation of Fe- and Si-rich liquids in igneous rocks. This work presents a record of large-scale magmatic differentiation due to immiscibility in the tholeiitic Sept Iles intrusion (Canada), one of the largest layered plutonic bodies on Earth. Gabbroic cumulate rocks from the Critical Zone of this intrusion show a bimodal distribution in density and P_2O_5 content, despite identical major element chemistry of the principal magmatic phases. Immiscibility is supported by the presence of contrasting Fe-rich and Si-rich melt inclusions trapped in cumulus apatite. Phase diagrams and well-documented occurrences of small-scale immiscibility confirm that liquid-liquid unmixing and the separation of Fe-rich and Si-rich liquids may contribute significantly to the Daly gap along the tholeiitic liquid line of descent.

INTRODUCTION

Silicate liquid immiscibility during late-stage differentiation of terrestrial and lunar basalts has been recognized to produce contrasting Fe-rich (ferrobasalt) and Si-rich (rhyolite) liquid compositions, both experimentally and in natural rocks (Roedder and Weiblen, 1970; Roedder, 1978; Dixon and Rutherford, 1979; Philpotts, 1982). However, only microscopic segregations have been observed, either as immiscible glass droplets in the groundmass of basalts or as distinct melt inclusions in silicates and apatite from plutonic rocks. Interest in immiscibility has been revived in the light of recent evidence for coexisting conjugate liquids in late-stage differentiates of ferrobasaltic rocks of the Skaergaard intrusion, Greenland (Jakobsen et al., 2005, 2011; Holness et al., 2011; Humphreys, 2011). However, even in this case, although meter-scale segregations of ferrogabbros are found (McBirney, 1975), the overall effect on magmatic differentiation processes remains unclear (Veksler et al., 2007; Morse, 2008; Philpotts, 2008). The general lack of evidence for large-scale immiscibility has led to the consensus that immiscibility does not play an important role on the relative paucity of intermediate compositions along the basalt-rhyolite liquid line of descent (Bowen, 1928). This is supported by experiments showing that the formation of silicic melt by fractional crystallization of basalt is possible (Sisson et al., 2005), and by the fact that the variation of liquid composition appears continuous from basalt to rhyolite in some volcanic suites (Carmichael, 1964), even if the proportion of intermediate compositions is low. Other alternatives include liquid viscosity and density barriers that prevent eruption of intermediate liquid compositions (Geist et al., 1995), critical crystallinity that only enables crystal-liquid separation in a narrow compositional range (Brophy, 1991; Dufek and Bachmann,

2010), or the consequence of a rapid evolution of liquid composition for intermediate silica contents, producing low volumes of monzonitic melts (Peccerillo et al., 2003). However, these possibilities do not exclude a role of immiscibility in creating the Daly gap (Daly, 1914).

SEPT ILES LAYERED INTRUSION

In this study, records of large-scale silicate liquid immiscibility and its effect on differentiation of basalt in the Sept Iles layered intrusion, Canada, are reported (Higgins, 2005). This igneous body is a circular intrusion ~80 km in diameter, with a thickness of 5.5 km and an estimated volume of 20,000 km³, making it one of the largest layered intrusions on Earth, after the Bushveld complex in South Africa and the Dufek intrusion in Antarctica (Loncarevic et al., 1990). It is composed of a lower Layered Series made up of troctolite and gabbro, and an Upper Border Series comprising anorthosite capped by cupolas of granite (Namur et al., 2010) (Fig. 1A). This large magmatic event is dated at 564 ± 4 Ma (Higgins and van Breemen, 1998), contemporaneous with opening of the Iapetus Ocean. Chilled margin rocks suggest a parental tholeiitic basalt composition, close to that of the Skaergaard intrusion (Namur et al., 2010). The stratigraphic fractional crystallization trend of liquidus phases is interrupted by two large and several small reversals to more primitive compositions interpreted as resulting from magma chamber replenishments by undifferentiated primitive basalt and mixing with the resident magma (Fig. 1A).

A liquid line of descent for the Sept Iles layered intrusion has been obtained from consideration of the composition of fine-grained rocks that occur at the margin, in dikes within the layered series, and in the upper granitic cupolas (Namur et al., 2011). This evolution reveals the following characteristics: (1) the parental magma (49 wt% SiO_2, 15 wt% FeO_{tot}) first crystallizes plagioclase and olivine and evolves on a tholeiitic trend with Fe enrichment and no significant variation of the SiO_2 content; (2) the saturation of Fe-Ti oxides and clinopyroxene drives the residual liquid to iron depletion and silica enrichment; (3) intermediate compositions (57–67 wt% SiO_2) produced by fractional crystallization are absent (the Daly gap); and (4) granites with 67–76 wt% SiO_2 are abundant in the upper part of the intrusion.

GABBROS OF THE CRITICAL ZONE

Rocks analyzed for this study were gabbros from the ~200-m-thick Critical Zone, which is at the top of the Layered Series. These rocks are made up of plagioclase, olivine, clinopyroxene, magnetite, ilmenite, and apatite (Figs. 2A and 2B), and their compositions are the most evolved of the Layered Series. Samples were collected along 20 cores, 65–190 m long, regularly drilled along a 3 km traverse (materials and methods in the GSA Data Repository[1]). They display clear bimodality in their P_2O_5 con-

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Figure 1. Schematic stratigraphy of Sept Iles layered intrusion (Canada) and compositional and density variations of gabbros from Critical Zone. A: Stratigraphy and zones subdivision of intrusion (after Namur et al., 2010). The 0 m reference corresponds to appearance of apatite in Megacyclic Unit 2 (MCU II) and marks base of Critical Zone. Curve displays schematically evolution of cumulus phase compositions. Light gray bands in MCU II and at base of MCU III represent mixing zones between resident magma and new magma inputs. UBS—Upper Border Series; US—Upper Series; Pl—plagioclase; Ol—olivine; Cpx—clinopyroxene; Mt—magnetite. B: Stratigraphic variations of P_2O_5 content in gabbros from Critical Zone. Stars indicate positions of samples studied for their melt inclusions at 7.3 and 78.1 m. C, D: Histograms of density (n = 228) and P_2O_5 content (n = 895) of gabbros from Critical Zone. E–G: Stratigraphic evolution of compositions of plagioclase, olivine, and clinopyroxene in Critical Zone. Black symbols are for dense P_2O_5-rich gabbros and white symbols are for light P_2O_5-poor gabbros.
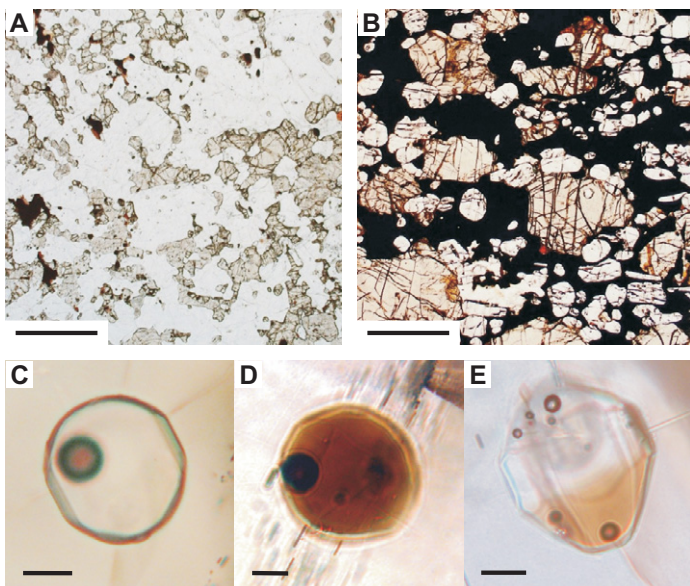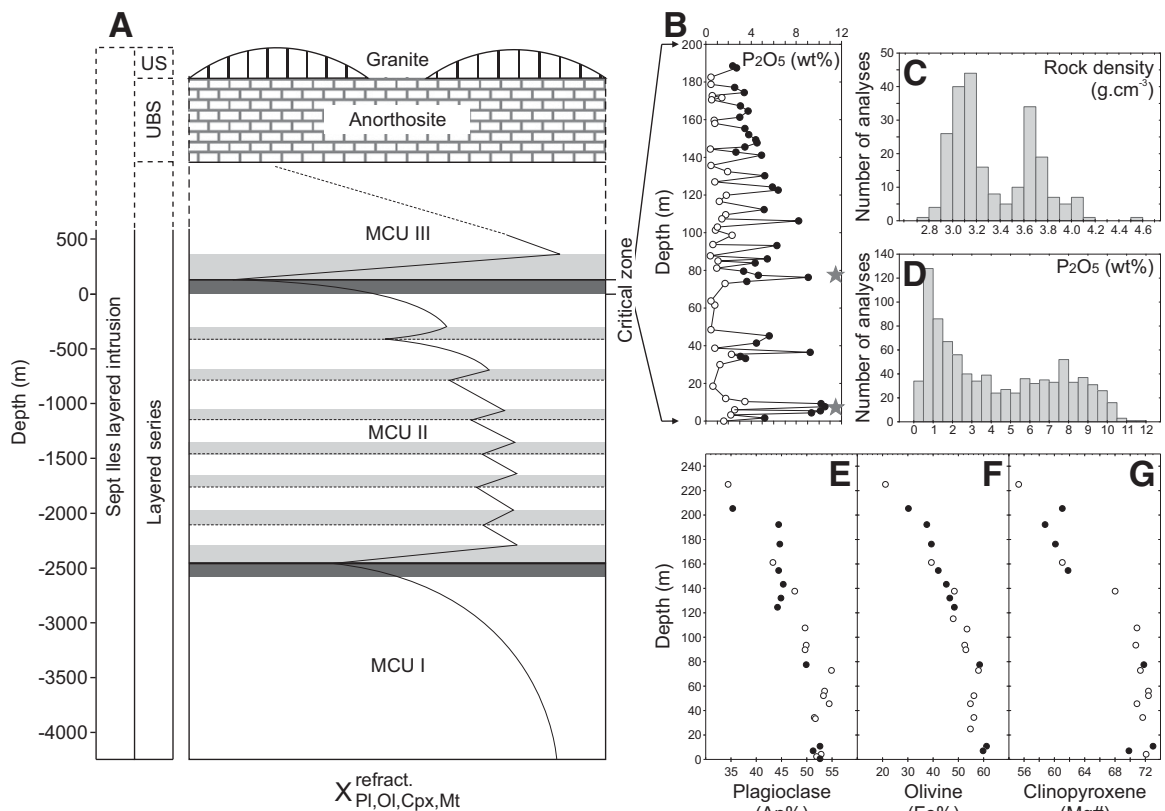

Figure 2. Microphotographs of apatite-bearing gabbros and types of melt inclusions in Sept Iles layered intrusion (Canada). A: Polarized-light microphotograph of Si-Al-Na–rich gabbro. Scale = 4 mm. B: Polarized-light microphotograph of Fe-Ti-P–rich gabbro. Scale = 4 mm. C: Si-rich granitic homogenized melt inclusion (sample at 7.3 m). Scale = 20 μm. D: Fe-rich ferrobasaltic homogenized melt inclusion (sample at 7.3 m). Scale = 20 μm. E: Homogenized melt inclusion containing two immiscible liquids (sample at 78.1 m). All melt inclusions have shrinkage bubble. Scale = 20 μm.

centrations and rock density, i.e., modal proportions of dense versus light minerals (Figs. 1B–1D). One group (Fig. 2A), representing ~66% of the samples, has an average density of 3.0–3.2 g/cm³ and contains 0–3.5 wt% P_2O_5. The other group (Fig. 2B), ~33% of the samples, has an average density of 3.6–3.8 g/cm³ with 3–12 wt% P_2O_5. Samples from the longer drill core and two additional samples collected in the field were selected for the determination of mineral compositions, as they provide a continuous record of the Critical Zone. The mineral compositions (Figs. 1E–1G; Tables DR1–DR3 in the Data Repository) display continuous evolution from the base to the top of the Critical Zone, with olivine ranging from Fo_{60} to Fo_{20}, plagioclase from An_{52} to An_{34}, and clinopyroxene from $Mg\#_{73}$ to $Mg\#_{55}$. The composition of the minerals does not distinguish in any way the two groups of rocks.

IMMISCIBILITY IN MELT INCLUSIONS

Silicate melt inclusions are ubiquitously trapped in apatite of these gabbros and display negative crystal shapes. These features, as well as the distribution of the inclusions, indicate that they represent droplets of liquids, trapped during the growth of the host phase. Two types of inclusions are clearly distinguished and both have crystallized daughter phases, grown from the trapped melt, in addition to glass. Dark inclusions contain chlorite, amphibole, plagioclase, Fe-Ti oxides, and fine-grained material. Their sizes range from 50 to 120 μm. Smaller light inclusions range from 10 to 80 μm and contain albite, orthoclase, biotite, amphibole, fine-grained material, and rare quartz.

Inclusions trapped in apatite from two samples located at stratigraphic heights of 7.3 and 78.1 m were rehomogenized using an optically controlled heating stage and a vertical drop-quench gas-mixing furnace.

Homogenization temperatures ranged from 1060 to 1100 °C. After the experiments, the quenched melts were analyzed for major elements using an electron microprobe (Tables DR4 and DR5). They display bimodal compositions: granitic Si rich (Fig. 2C) and ferrobasaltic Fe rich (Fig. 2D). Single inclusions also show clear evidence of unmixing between the two compositions (Fig. 2E). We consider that the inclusions represent conjugate immiscible liquids, albeit on a small scale. Compositions of these liquids are presented in Figures 3A–3D and compared to well-documented occurrences of silicate liquid immiscibility in both natural and experimental systems relevant to terrestrial (Dixon and Rutherford, 1979; Philpotts, 1982) and lunar basalt petrogenesis (Longhi, 1990).

DISCUSSION

Unmixing of two immiscible melts results from the intersection of the liquid line of descent and a subliquidus immiscibility field (Fig. 3E). The onset of liquid immiscibility in the Sept Iles layered intrusion occurs when the liquid line of descent reaches monzonitic compositions (55–57 wt% SiO_2 and 10–11 wt% FeO_{tot}), thus after significant silica enrichment and iron depletion, when the fraction of residual liquid was ~30% (Namur et al., 2011). The onset of silicate liquid immiscibility corresponds to the start of the Daly gap. With decreasing temperature, crystallization products of the two immiscible liquids, i.e., the same phases with the same compositions but different proportions, are subtracted from the bulk liquid composition, enriching the bulk composition in silica. One unexpected and original feature of our data is that the compositions of immiscible pairs converge with decreasing temperature, as illustrated by the less contrasted ferrobasaltic and granitic compositions of melt inclusions in samples at 78.1 m compared to those at 7.3 m (Tables DR4 and DR5). The crystallization products of the immiscible melts also become more similar with decreasing temperature, as illustrated by their P_2O_5 content (Fig. 1B).

Residual granitic rocks located at the top of the Sept Iles intrusion do not show any evidence of immiscibility, and their origin can be satisfactorily modeled by continuous fractional crystallization from the parental tholeiitic basalt (Namur et al., 2011). This observation provides further evidence in favor of the idea that the immiscibility field closes at low temperature, behavior perfectly consistent with phase relations in the system K_2O-FeO-Al_2O_3-SiO_2 (Visser and Koster van Groos, 1979), although we know of no data to assess whether this is true in complex natural systems. Closing of the miscibility gap at low temperature would have the consequence that liquid unmixing would have no bearing on the geochemical signature of residual granitic compositions, but would nevertheless be responsible for the absence of intermediate liquid compositions, i.e., the Daly gap.

Silicate liquid immiscibility does not appear to have affected the entire mass of residual liquid in the magma chamber at the same time. Such a process would have resulted in simple segregation between the dense iron end member at the base of the Critical Zone and the light silica end member at the top. In the Sept Iles intrusion, the crystallization products of each end member alternate at a scale of 20–5 m (Fig. 1B), suggesting that immiscibility may occur repetitively in a basal boundary layer. This is supported by the thermal regime of large basaltic intrusions, which have static boundary layers unaffected by convection at the interface between the cumulate pile and the main magma body (Jaupart et al., 1984). The standard interpretations of layer-forming processes in mafic intrusions by crystal settling and sorting or in situ oscillatory nucleation can produce the bimodal distribution of rock densities and bulk P_2O_5 contents with the same mineral compositions in the contrasted layers. Indeed, cumulates produced by the crystallization of a homogeneous melt followed by crystal sorting cannot be distinguished from the crystallization products of two immiscible melts. This is because these two liquids are in equilibrium and crystallize the same liquidus phases in different proportions. However, the presence of immiscible liquids in
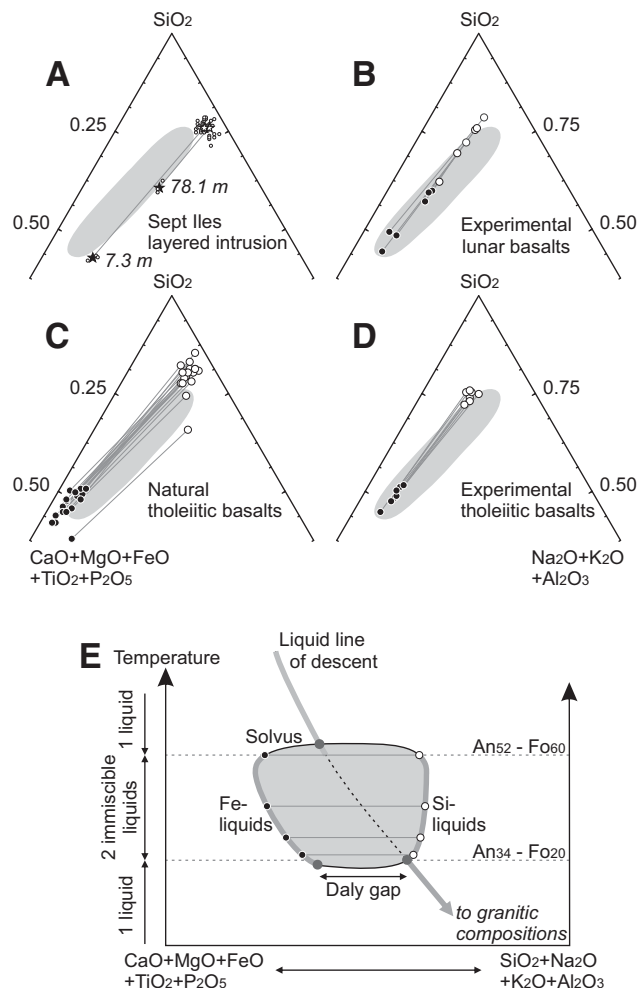

Figure 3. Pseudoternary diagrams (wt%) with tie lines for various conjugate immiscible melt pairs and schematic model for development of silicate liquid immiscibility. A: Immiscible ferrobasalts and granitic liquids obtained from melt inclusion compositions from samples at 7.3 and 78.1 m of Sept Iles layered intrusion. Single analyses are small circles and average compositions are large stars. B: Experimental immiscible pairs in lunar basalts (Longhi, 1990). C: Natural immiscible pairs of glassy globules in tholeiitic basalts (Philpotts, 1982). D: Experimental immiscible ferrobasalt and granitic liquids (Dixon and Rutherford, 1979). Gray area is projection of low-temperature immiscibility field in system leucite-fayalite-SiO_2 (Roedder, 1978). E: Schematic pseudobinary diagram illustrating development of silicate liquid immiscibility during liquid line of descent of Sept Iles tholeiitic parental magma. In the course of differentiation, liquid reaches solvus and splits into Fe-rich and Si-rich liquids. Bulk liquid composition follows silica enrichment due to subtraction of crystallization products of immiscible liquid (Critical Zone). With decreasing temperature, solvus closes and immiscibility terminates. Black circles are Fe-rich ferrobasalts and white circles are Si-rich granites.

melt inclusions supports the idea that rocks of the Critical Zone crystallized from an emulsion of two liquids.

The sole expressions of the immiscible excursion are the Daly gap and the occurrences of contrasted melt inclusions and cumulates that contain the same liquidus phases in different proportions. This stealthy expression of silicate liquid immiscibility compels a revision of processes responsible for the dearth of intermediate melts in tholeiitic suites and in other geological settings (Reubi and Blundy, 2009). Our results may also stimulate ideas about the differentiation of extraterrestrial basalts, particularly during the late-stage evolution of the lunar magma ocean (Shearer et

al., 2006), and for the origin of highly silicic compositions on the moon (Glotch et al., 2010) as well as during the crystallization of martian basalts that are rich in iron and phosphorous (McSween et al., 2004), two elements that expand the immiscibility field (Naslund, 1983).

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