MELT INCLUSIONS FROM THE DEEP SLAVE LITHOSPHERE: CONSTRAINTS ON THE ORIGIN AND EVOLUTION OF MANTLE-DERIVED CARBONATITE AND KIMBERLITE.

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INTRODUCTION

Megacrystalline clinopyroxene-rich lherzolite xenoliths, derived from the lithosphere beneath the Slave craton in Canada, were transported to the surface during the eruption of the A154N kimberlite ca 50 million years ago. The xenoliths host numerous globular carbonate-rich and ultramafic silicate-carbonate inclusions which were shielded from interaction with mantle wall-rock by the clinopyroxene host, providing rare samples of unmodified mantle-derived melts.

SAMPLE DESCRIPTION

CARBONATE-RICH GLOBULES

Analytical techniques are described by Van Achterbergh et al. (2002). Sample VR67103A, which contains fresh carbonate-rich globules, has been described by Van Achterbergh et al. (2002). It consists mostly of clinopyroxene (Cr-diopside) with large (> 7mm) grains of orthopyroxene (En₉₂) and garnet (Crpyrope). The compositions of the host phases are typical of compositional ranges for these minerals in mantle lherzolite. The globular inclusions occur along distinct planes with a bimodal size distribution (0.8-1.3 mm and 3.2-5.8 mm in diameter). The assemblage within the globules (Fig. 1a) consists of euhedral to subhedral Ti-rich phlogopite, radiating "spinifex" olivine (ol; Fo₉₀), Cr-spinel (sp), rare perovskite and sulfides set in a matrix of fresh calcite. Calcite occurs as two microstructural types: elongate randomlyoriented crystals (cc-1) and interstitial calcite (cc-2). Sr and Ba are concentrated in cc-1 (average 2.6 wt% SrCO₃ and 3.7 wt% BaCO₃), hence the brighter backscattered electron reflectance for cc-1 in Fig. 1a.

SILICATE-CARBONATE GLOBULES

Sample VR43445A is a 1.0 cm clinopyroxene macrocryst which contains ultramafic silicate globules

with subordinate carbonate. The globules vary in size from ca 100µm to 2mm in diameter. The silicate and carbonate components in the globules occur as concentric rings (Fig. 1b); the carbonate component is enriched in rare-earth elements (REE) with < 1 wt% SrCO₃ and BaCO₃ and < 0.1 MgCO₃ and separates an outer ring of silicate glass (silicate-a; mg# = 85.1) from a silicate-glass core (silicate-b; mg# = 91.4). The carbonate component crystallised as pure calcite (cc). Small grains of pyrite and pyrrhotite (< 4µm in diameter) occur in the silicate component of some inclusions.

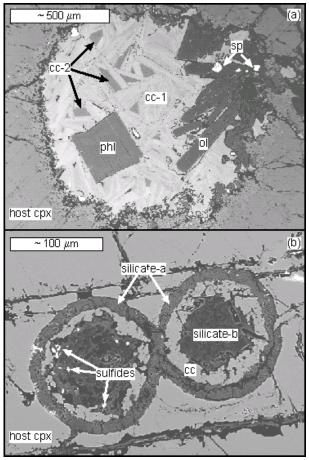


Figure 1: Backscattered electron images for (a) typical carbonate-rich and (b) typical silicate-carbonate globules described in this study. Abbreviations are given in the text.

DISCUSSION

ORIGIN OF CARBONATE-RICH GLOBULES

Van Achterbergh et al. (2002) interpreted the microstructures and bulk chemical compositions of the globules as evidence that the carbonate-rich globules may be defined as silica-rich Mg-carbonatites (based on the terminology of Woolley and Kempe, 1989). The inclusions are not in chemical equilibrium with the host, suggesting that the globules were trapped in the host shortly before the xenoliths were entrained in the kimberlite (Van Achterbergh et al., 2002). The depth from which the host lherzolite was derived can be estimated from the equilibration conditions (~ 1235°C and 60 kb) of the host minerals (clinopyroxene, orthopyroxene and garnet) using conventional geothermobarometry (Van Achterbergh et al., 2002) and is similar to the estimated base of the lithosphere beneath this area, i.e. approximately 200km (Griffin et al., 1999; Pearson et al., 1999).

Trace-element compositions of the globules are plotted in Fig. 2 and are depleted in terms of REE contents compared to the range for worldwide carbonatite compositions (Nelson et al., 1988). They are also depleted in Na (0.12 wt% Na₂O) relative to the host clinopyroxene (1.23 wt% Na₂O). The relatively low concentrations of Na and REE (these elements are incompatible in clinopyroxene and should concentrate the melt fractions) and the size and abundance of the inclusions in the xenoliths from this pipe (K. Kivi, unpubl. data) are inconsistent with an origin by lowdegree partial melting of carbonated peridotite (with low CO₂ content, e.g. 0.15% CO₂, Javoy, 1997). Dalton and Presnall (1998a) demonstrated that once melt fractions exceed 0.4% the melts are no longer dominated by carbonate and are best described as carbonate-bearing ultramafic silicates. However, if the source rocks were enriched in carbonate minerals, melts would remain carbonatitic at larger melt fractions (Dalton and Presnall, 1998a). Carbonates are always the first phase to be wholly consumed during melting (Canil and Scarfe, 1990) and the low REE abundance could be explained by melting of a large modal proportion of the carbonate minerals. We therefore propose that the source from which the melts formed was enriched in Ca-Mg carbonate minerals. Van Achterbergh et al. (2002) used oxygen, carbon, and strontium isotope data to demonstrate the involvement of recycled crustal material in the origin of these melt inclusions; this may provide the required carbonate-rich source.

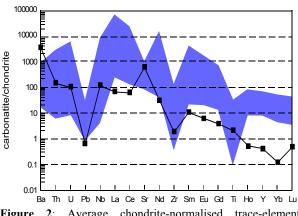


Figure 2: Average chondrite-normalised trace-element composition of the carbonatite globules and comparison with the compilation of Nelson *et al.* (1988) for world-wide occurrences of carbonatite melts (blue shaded area).

In summary, the carbonate-rich globules probably formed by partial melting of a carbonate-rich source and were mobilised and trapped in the host clinopyroxene near the lithosphere-asthenosphere boundary shortly before kimberlite eruption. The melting may have been a response to lowering of the solidus by upwelling C-O-H fluids and/or heat associated with kimberlite-precursor magmas. The source from which the melts formed contained recycled crustal material, consistent with suggestions by Barker (1996) that carbon recycling by subduction may be an important prerequisite for some types of carbonatite magmatism.

ORIGIN OF SILICATE-CARBONATE GLOBULES

The concentric microstructures of the silicate-carbonate globules are interpreted as evidence for the immiscible separation of silicate and carbonate melts after the inclusions were trapped in the host clinopyroxene. The outer silicate ring is interpreted either as a reaction rim of the melt with the host cpx or a zone of initial crystallisation; this is interpreted as shifting the bulk composition of the residual melt to higher CO₂ content and triggered immiscible separation of a carbonate component and a second silicate component, concentrated in the core. The change in bulk composition may have been accompanied by slow cooling which may have played a part in the immiscible separation. However, low alkali content in the globules and the presence of a pure calcite end-member are inconsistent with published experimental data on silicate-carbonate immiscibility (Lee et al., 1994; Lee and Wyllie, 1998), suggesting that there are additional components/elements in natural systems that play an important role in silicate-carbonate immiscibility and

which have not been incorporated in the experiments. The distribution of trace elements between the different components in these inclusions and the trace-element content of the carbonates are discussed elsewhere in these proceedings (Van Achterbergh *et al.*, 2003).

The bulk major-element composition of the silicatecarbonate inclusions is typical of Group 1 kimberlite. The average composition of nine inclusions is plotted on the MgO-CaO-SiO₂ (mole%) ternary in Fig. 3. and agrees well with the data compiled by Mitchell (1986) for uncontaminated kimberlite compositions. Also shown in Fig. 3 are the data from Dalton and Presnall (1998a), produced by melting experiments at 6 GPa in which they demonstrated that with increasing temperatures (indicated with an arrow in Fig. 3) the increasingly resultant melt fractions becomes kimberlitic in nature. The melts produced by Dalton and Presnall (1998a) at the highest temperatures in their experiments (≥ 1455 °C) are very similar in composition to the average value for the silicatecarbonate globules (Fig. 3). The bulk trace-element data are shown in Fig. 4 and also overlap with kimberlitic compositions (Mitchell, 1986; Grégoire et al., 2002).

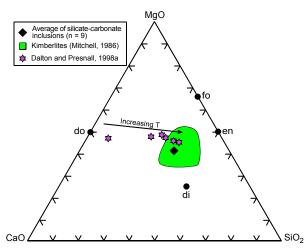


Figure 3: MgO-CaO-SiO₂ (mole%) ternary diagram (projected from CO₂) showing the position of the average bulk composition of the silicate-carbonate globules described in this study. The data plot within the field of kimberlite compositions of Mitchell (1986; green shaded area). The stars indicate the isobaric experimental melt compositions of Dalton and Presnall (1998a) produced by partial melting of carbonated peridotite at increasing temperatures (arrow). End-member minerals are indicated with black dots for reference (do = dolomite, fo=forsterite, en = enstatite, di = diopside).

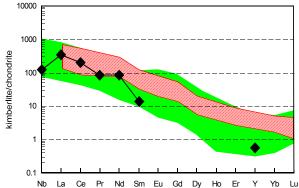


Figure 4: Average chondrite-normalised trace-element composition of 7 silicate-carbonate globules. Due to the small size and rarity of these samples no laser ablation analysis was carried out and the all data were obtained by non-destructive quantitative μ PIXE imaging (Ryan *et al.*, 2002); the REE Eu to Lu are below detection. The data are compared with the compilation of Mitchell (1986) for average kimberlite compositions (green shaded area) and the recent compilation of Grégoire *et al.* (2002; references therein) for Group I kimberlites (red cross-hatched).

The host clinopyroxene equilibrated at ca 750-800 °C and ca 35 kbar. The temperature was calculated from an algorithm (Griffin et al., unpubl. data) based on the temperature-dependent partitioning of Ni between clinopyroxene and olivine observed in 50 four-phase lherzolite xenoliths (W.L. Griffin and S.Y. O'Reilly, unpubl. data). The estimated uncertainty of this temperature is ca 75 °C. The pressure was estimated from the intersection of this temperature with the xenolith geotherm for this part of the lithosphere (Pearson et al., 1999). This temperature (at 35kbar) is more than 400 °C below the lherzolite-CO₂ solidus (Dalton and Presnall, 1998b) and about 200 °C below the lherzolite-CO₂-H₂O solidus (White and Wyllie, 1992). The geotherm at this depth is typical of a cool Archean 35 mW/m² geotherm (Pearson et al., 1999) with no evidence of a significant thermal perturbation that may have caused in situ partial melting. Furthermore, the host is enriched in Na (incompatible in clinopyroxene) relative to the melt (Van Achterbergh et al., 2003) which is inconsistent with equilibrium in situ partial melting of the host to form these melts. We suggest that the melts probably formed at deeper levels (lower lithosphere or asthenosphere) and higher temperatures and migrated upwards before being trapped in the host mineral. The similarity of the average bulk composition of these globules and the melts produced experimentally at 6 GPa and $T \ge 1455$ °C (Dalton and Presnall, 1998a) suggests that the source rocks may have been carbonated peridotite and according to Dalton and Presnall (1998a), mantle with 0.15% CO₂ (value suggested by Javoy, 1997) would be sufficient.

There is not enough evidence to establish or discount unambiguously a petrogenetic or timing link between these kimberlitic globules and the carbonatitic globules described in the previous section. In terms of major elements, the compositional continuum between carbonatites and kimberlites (derived from a common source) demonstrated by Dalton and Presnall (1998a) certainly suggests that a link is possible. However, it is inferred that these particular carbonatitic globules require a source enriched in CO₂, whereas the kimberlitic globules may be formed by melting mantle without unusually high levels of CO₂.

THE KIMBERLITE-CARBONATITE ASSOCIATION

Data obtained from the kimberlitic globules illustrate that in natural systems:

- 1. The composition of the carbonate-bearing ultramafic silicate melts is consistent with experimental predictions (Dalton and Presnall, 1998a) that such melts form from partial melting of carbonated peridotite at ca 70-100 °C above the solidus.
- 2. CaCO₃-rich melts can form by immiscible separation from a carbonate-bearing ultramafic-silicate parent melt.

These globules are interpreted as failed kimberlites, and it is noteworthy that the Sr and Ba content of the carbonate in these globules is markedly lower than in the carbonatitic globules. Low Sr and Ba -contents are characteristic of kimberlite carbonates in general (groundmass carbonate and vein carbonate) compared with typical carbonates from carbonatites (Kirkley, 1987). Therefore, if these globules do indeed represent primary kimberlite magmas, the inclusions provide evidence for a genetic relationship between carbonatebearing magmas and kimberlites. It is necessary to stress here that the concentric microstructure is indicative of super-solidus immiscible separation of melts, and that the carbonate component does not merely represent a calcite cumulate produced by fractional crystallisation. Van Achterbergh et al., (2003) provide further evidence for this, i.e. high concentrations of incompatible elements in the calcite.

Application of the evidence presented here to the debate on the association between kimberlites and carbonatites in general (e.g. Mitchell, 1979) depends on whether the carbonate component can be called a carbonatite or not. This depends in turn upon the preferred definition of carbonatite (e.g. Mitchell, 1979 vs. Woolley and Kempe, 1989) and is left to the reader.

It was shown that the carbonatitic globules described here probably did not form from the same source rock as the kimberlitic globules. This illustrates further that some mantle-derived carbonatites probably are primitive and unrelated to kimberlites. Some may form from melting of a carbonate-rich source, such as subducted crustal material.

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