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Carbonate–silicate liquid immiscibility in the mantle propels kimberlite magma ascent

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Abstract

Kimberlite is a rare volcanic rock renowned as the major host of diamonds and originated at the base of the subcontinental lithospheric mantle. Although kimberlite magmas are dense in crystals and deeply-derived rock fragments, they ascend to the surface extremely rapidly, enabling diamonds to survive. The unique physical properties of kimberlite magmas depend on the specific compositions of their parental melts that, in absence of historical eruptions and due to pervasive alteration of kimberlite rocks, remain highly debatable. We explain exceptionally rapid ascent of kimberlite magma from mantle depths by combining empirical data on the essentially carbonatite composition of the kimberlite primary melts and experimental evidence on interaction of the carbonate liquids with mantle minerals. Our experimental study shows that orthopyroxene is completely dissolved in a Na₂CO₃ melt at 2.0-5.0 GPa and 1000-1200 °C. The dissolution of orthopyroxene results in homogeneous silicate-carbonate melt at 5.0 GPa and 1200 °C, and is followed by unmixing of carbonate and carbonated silicate melts and formation of stable magmatic emulsion at lower pressures and temperatures. The dispersed silicate melt has a significant capacity for storing a carbonate component in the deep mantle (13 wt% CO₂ at 2.0 GPa). We envisage that this component reaches saturation and is gradually released as CO₂ bubbles, as the silicate melt globules are transported upwards through the lithosphere by the carbonatite magma. The globules of unmixed, CO₂-rich silicate melt are continuously produced upon further reaction between the natrocarbonatite melt and mantle peridotite. On decompression the dispersed silicate melt phase ensures a continuous supply of CO₂ bubbles that decrease density and increase buoyancy and promote rapid ascent of the magmatic emulsion.

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1. INTRODUCTION

Kimberlites are very rare and small-volume igneous rocks, but are nonetheless important to the exploration and scientific communities because they are often associated with diamonds and their parental magmas originate from within or below the deepest continental lithosphere (Dawson, 1980; Mitchell, 1989; Haggerty, 1999). Kimberlite magmas,

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despite a prodigious load of mantle-derived dense cargo, appear to rise through the cratonic lithosphere at above average rates (e.g. McGetchin et al., 1973; Eggler, 1989; Canil and Fedortchouk, 1999). The transport and preservation of diamonds and xenoliths in these deepest derived mantle magmas is related to the chemical composition, temperature, density, viscosity and buoyancy of primary kimberlite melts. However, the assessment and quantification of these parameters is hampered by inevitable contamination of proto-kimberlite liquids en route to the surface by entrained lithospheric and crustal material, and by the effects of syn-emplacement stratification of the magmas and

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syn/post-emplacement alteration of kimberlite rocks. The compositions proposed for the primary kimberlite are diverse, thus, their properties remain poorly constrained.

A traditional view on the composition of the archetypal primary kimberlite melts as being hydrous, carbonated ultramafic/ultrabasic liquids (e.g. 18-32 wt% SiO₂, 22-31 wt% MgO, 5-12 wt% H₂O, 5-14 wt% CO₂; Edgar et al., 1988; Price et al., 2000; le Roex et al., 2003; Harris et al., 2004; Becker and le Roex, 2006; Kopylova et al., 2007; Mitchell, 2008; Nielsen and Sand, 2008; Kjarsgaard et al., 2009) has been challenged by recent empirical and experimental studies. Ubiquitous olivine in kimberlites, responsible for the previously inferred ultramafic composition of parental melts, has been demonstrated to largely (>50%) represent the lithospheric mantle invaded by an essentially carbonatite liquid (Kamenetsky et al., 2008; Brett et al., 2009; Patterson et al., 2009; Arndt et al., 2010; Pilbeam et al., 2013). The aluminosilicate-poor, Na-Ca carbonate composition of the kimberlite parental melt was suggested by compositions of melt inclusions entrapped in olivine, Cr-spinel and ilmenite from kimberlites in Siberia, Canada, South Africa and Greenland (see review in Kamenetsky et al., 2014). Additionally, carefully designed experimental studies of a putative primary kimberlite (e.g. Canil and Bellis, 2008; Sparks et al., 2009; Brooker et al., 2011) demonstrated that such compositions are inconsistent with their inferred high volatile solubility and low temperatures (Edwards and Howkins, 1966; Clement, 1975; Mitchell, 1986) at conditions of magma emplacement into the crust and diatreme formation. An essentially carbonatite melt, parental to most kimberlites (e.g. Kamenetsky et al., 2007, 2008, 2014), is undersaturated in silica and thus capable of dissolving mantle silicates, especially orthopyroxene, the most Si-rich mantle mineral, abundant in the mantle and absent in kimberlite rocks (e.g. Mitchell, 1973).

A consensus exists about the mechanism responsible for ultra-fast ascent of kimberlite magmas – it is buoyancy increased by volatile exsolution and related "bubbling" (see reviews in (Mitchell, 1986; Eggler, 1989). However, the composition of starting kimberlite melts is explicitly relevant to a dilemma of whether the kimberlite is a volatile-rich ultrabasic magma (Kopylova et al., 2007; Mitchell, 2008; Kjarsgaard et al., 2009) that catastrophically degasses CO₂ during decompression (e.g. Wilson and Head, 2007), or a carbonatite melt that undergoes decarbonation due to reaction with orthopyroxene (e.g. Wyllie and Huang, 1975; Eggler and Wendlandt, 1979).

The latter process of the carbonatite melt "silicification" by assimilation of mantle orthopyroxene with concomitant release of CO₂ is modelled by experiments at 1 atm and 1000–1100 °C, using molten Na₂CO₃ as an analogue of natural carbonatite liquids (Russell et al., 2012). Reaction between liquid Na₂CO₃ and silicates at 1 atm results in formation of highly reactive and unstable sodium metasilicate, Na₂SiO₃ (also known as "liquid glass") and abundant CO₂ gas. The observed vigorous exsolution of CO₂ from the decomposing Na₂CO₃ melt at 1 atm was assumed to represent the naturally occurring process in ascending kimberlite magma (Russell et al., 2012).

In this study we have tested whether the proposed mechanism of magma vesiculation and increased buoyancy related to CO_2 degassing during breakdown of Na_2CO_3 melt at 1 atm (Russell et al., 2012)is applicable to deep mantle conditions (\geq 2 GPa) where ascent of the primary kimberlite melt starts.

2. EXPERIMENTAL RATIONALE

The choice of Na₂CO₃ (melting point 854 °C at 1 atm) in preference to calcitic and dolomitic compositions in the experiments by Russell et al. (2012) was dictated by very high temperatures of melting of common carbonates (Irving and Wyllie, 1975; Huang and Wyllie, 1976), unrealistic for the lithospheric mantle. For the same reason sodium carbonate has been used in other analogue experiments for investigating the behaviour of carbonatite liquids at mantle conditions (Minarik and Watson, 1995; Hammouda and Laporte, 2000). It has been demonstrated that natural carbonate mantle melts represented by Na₂CO₃ remained interconnected in a fine-grained dunite at exceptionally low melt fractions (0.05 wt% melt; Minarik and Watson, 1995) and that such melts can fast percolate upward in polycrystalline olivine over long distances (Hammouda and Laporte, 2000).

Unfortunately Russell et al. (2012) provided no justification to why a sodium carbonate melt could be used as an analogue to primary kimberlite liquids given that the overwhelming majority of kimberlite rocks are Na-poor (<0.2 wt.% Na₂O; e.g. Kjarsgaard et al., 2009). This paradox can be partially rectified by demonstrating inevitable Naloss during alteration of kimberlites and recognising Narich carbonate compositions of melt inclusions in minerals of kimberlites (Kamenetsky et al., 2009b, 2013, 2014) and kimberlite-hosted mantle xenoliths (Golovin et al., 2008; Korsakov et al., 2009; Giuliani et al., 2012; Sharygin et al., 2013; Kamenetsky et al., 2014).

The analogue Na₂CO₃ composition is also significant given the role of Na in mantle-derived carbonate melts and related metasomatic transformations that has been emphasised in several experimental studies (e.g. Green and Wallace, 1988; Wallace and Green, 1988; Sweeney et al., 1995; Yaxley and Green, 1996; Yaxley and Brey, 2004; Litasov et al., 2013; Shatskiy et al., 2013a,b, 2015). In particular, even minor amounts of sodium in a carbonated mantle protolith may significantly reduce melting temperatures (by up to 400-500 °C; e.g. Dasgupta et al., 2004; Yaxley and Brey, 2004; Litasov et al., 2013), resulting in sodic carbonate compositions of near-solidus melts (45% Na₂O at 15 kb and 850 °C; Sweeney et al., 1995). The generation of the sodic liquid has been envisaged in the melting reaction between jadeite component in clinopyroxene and dolomitic liquid $(4NaAlSi_2O_6 + CaMg(CO_3)_2 =$ 2Na₂CO₃ + CaAl₂SiO₆ + MgAl₂SiO₆ + 6SiO₂; Yaxley and Brey, 2004). Additionally, a number of experimental studies emphasised the role of Na-rich carbonate melts in generation of kimberlites (e.g. Safonov et al., 2009, 2011; Sharygin et al., 2015) and natrocarbonatites (e.g. Sweeney et al., 1995), nucleation and growth of diamonds (e.g. Pal'yanov et al., 1999, 2002) and occurrence of

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