



Oxide, sulphide and carbonate minerals in a mantle polymict breccia: Metasomatism by proto-kimberlite magmas, and relationship to the kimberlite megacrystic suite

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ABSTRACT

Polymict breccias entrained by kimberlites are mantle xenoliths comprising coarse-grained mantle minerals (porphyroclasts) and rock clasts, cemented together by ilmenite, rutile, phlogopite, olivine and minor sulphides. These unusual xenoliths are generally considered to result from ascending primitive/precursor kimberlite magmas that crystallised in the magma conduit at lithospheric mantle depths. To enhance our understanding of these processes, we investigated the oxide, sulphide and carbonate minerals of a new polymict breccia (DU-1) from the Bultfontein Dumps (Kimberley, South Africa).

Xenolith DU-1 contains heterogeneous domains of ilmenite-rich breccia surrounded by spinel harzburgite wall rock. The breccia domains also host resorbed Cr-diopside porphyroclasts and occasional Fe–Ni–Co sulphides segregations. Ilmenite occurs as large (up to 5 cm), chemically zoned lenses, associated with minor rutile. The ilmenite has Hf isotope values (initial $\epsilon_{\text{Hf}} = 2.1\text{--}3.0$) in the range of South African Group I kimberlites and associated megacrysts. The ilmenite lenses host primary multiphase carbonate-rich, olivine-dominated, phlogopite-dominated and sulphide mineral inclusions. Carbonate-rich inclusions host abundant magnesite and dolomite, with subordinate kalsilite, phlogopite, alkali-carbonates, phosphates and chlorides.

The occurrence of carbonate-rich inclusions suggests entrapment of a Ca–Mg-rich alkali-carbonate melt during ilmenite growth. However, geochemical modelling indicates that this melt was not parental to the ilmenite. Instead, it is suggested that the ilmenite (and other cementing phases) crystallised from ilmenite and the other cementing phases crystallised from a S-bearing Ti–Fe–K-rich ultramafic silicate melt, derived from an ascending proto-kimberlite melt, which was modified subsequently by wall rock assimilation and/or magma mixing, porphyroclast dissolution and crystal fractionation. The alkali-carbonate melt could be the residual liquid after these processes occurred. Alternatively, the ultramafic silicate and alkali-carbonate melts were produced by liquid immiscibility from a silicate-carbonate proto-kimberlite melt. The occurrence of ilmenite inclusions in sulphide segregations, and sulphide inclusions in ilmenite lenses is indicative of late-stage silicate-sulphide liquid immiscibility.

The major and trace element concentrations, Hf isotope composition and mineral inclusion content of ilmenite are consistent with a genetic relationship between polymict breccia formation and the kimberlite megacryst suite. We propose that polymict breccias and megacrysts were produced at different depths by common silicate-carbonate magmas during the early stages of kimberlite magmatism.

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1. Introduction

Kimberlite magmas intruded through ancient cratons commonly entrain fragments (xenoliths) of the Earth's mantle. The majority of mantle xenoliths are exhumed from the sub-continental lithospheric mantle (SCLM) or the lithosphere–asthenosphere boundary (LAB; e.g. Pearson et al., 2003). Mantle xenoliths generally display evidence of metasomatism by fluids and melts of variable composition. The most commonly inferred metasomatic agents are carbonate and

alkali-carbonate melts (Kogarko et al., 1995; Yaxley et al., 1998; Giuliani et al., in press-a), basaltic and alkali-basaltic melts (Harte et al., 1993; Burgess and Harte, 2004), kimberlitic and lamproitic melts (Waters, 1987; Kinny and Dawson, 1992), and C–O–H fluids (Andersen et al., 1984; Frezzotti et al., 2010; Giuliani et al., in press-b).

Carbonate melts are likely to play a prominent role in mantle metasomatism, because experimental studies have shown that carbonate melts are the first product of partial melting of carbonated peridotites and eclogites (Wyllie and Huang, 1975; Wallace and Green, 1988; Hammouda, 2003; Yaxley and Brey, 2004; Brey et al., 2008; Litasov et al., 2010) and have high mobility under mantle

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conditions (e.g. Dobson et al., 1996; Hammouda and Laporte, 2000). However, carbonate melts can also derive from liquid immiscibility in CO₂-rich silicate or silicate-carbonate melts, although there is limited textural evidence of carbonate-silicate liquid immiscibility in mantle rocks (Pyle and Haggerty, 1994; Schiano et al., 1994; Moine et al., 2004). In the mantle other common products of liquid immiscibility are sulphide melts; sulphur saturation of silicate and carbonate melts is commonly achieved upon cooling (Mavrogenes and O'Neill, 1999; Kogarko et al., 2001). The occurrence of sulphide minerals in several suites of mantle xenolith from oceanic and continental settings is considered indicative of the existence of immiscible sulphide melts (e.g. Kogarko et al., 1995, 2001; Lorand et al., 2004; Lorand and Gregoire, 2006). However, metasomatism by sulphide melts is unlikely to modify the mantle to any large extent, because mantle sulphide melts are not always wetting (i.e. mobile) under mantle conditions (Gaetani and Grove, 1999; Rose and Brenan, 2001).

The study of mantle xenoliths provides invaluable information towards understanding the processes that pre-date kimberlite emplacement. For example, some xenoliths exhibit evidence of metasomatism by melts of kimberlitic affinity, which occurred immediately prior to kimberlite entrainment. Two classes of xenolith that are often considered to be directly related to kimberlite or proto-kimberlite magmas, are the megacrystic suite and polymict breccia xenoliths (e.g. Lawless et al., 1979; Moore and Belousova, 2005). The megacryst nodules (up to 40 cm in size) entrained by kimberlite magmas appear to have crystallised at high pressures (commonly 45–55 kbar) that are broadly equivalent to the lithosphere–asthenosphere boundary (Nixon and Boyd, 1973; Gurney et al., 1979; Harte and Gurney, 1981; De Bruin, 2005). Radiogenic (Sr–Nd–Hf) isotope systematics of kimberlitic megacrysts indicate an asthenospheric origin for the megacryst magmas, with variable contamination by Archean lithospheric material (e.g. Griffin et al., 2000; Davies et al., 2001; Spetsius et al., 2002; Nowell et al., 2004; Tappe et al., 2011). Radiogenic isotope data and the close timing between megacryst formation and kimberlite entrainment suggest a cognate origin for the megacryst suite and the host kimberlite magma (e.g. Moore and Belousova, 2005; Kopylova et al., 2009). However, trace element modelling indicates that the megacryst minerals crystallised from melts with compositions distinct from that of the host kimberlite (e.g. Harte, 1983; Davies et al., 2001; Merry and Le Roex, 2007).

Polymict breccias are a rare type of xenolith only recorded from kimberlites in the Kaapvaal and Siberian cratons. They are described as mixtures of large mantle minerals (porphyroclasts) and rock clasts, cemented together by variable amounts of ilmenite, rutile, phlogopite, olivine and sulphides (Lawless et al., 1979; Höfer et al., 2009). The cementing minerals are thought to have precipitated from Ti–K-rich hydrous ultramafic melts (Wyatt and Lawless, 1984; Harte et al., 1993), and there is evidence for multiple pulses of melt addition (Höfer et al., 2009). The porphyroclast compositions reflect provenance from diverse lithologies, including harzburgite, lherzolite, eclogite, and minerals of the megacryst suite (Lawless et al., 1979; Zhang et al., 2003; Pokhilenko, 2009). Mineral thermobarometry has shown that the porphyroclasts included in polymict breccia xenoliths were entrained from multiple locations in the lithospheric mantle, encompassing a vertical profile of about 100 km (Höfer et al., 2009; Pokhilenko, 2009). Partial dissolution and overgrowth of porphyroclasts is common (Zhang et al., 2001a; Höfer et al., 2009), whereas evidence of in-situ melting is only reported occasionally (Wyatt et al., 1998; Pokhilenko, 2009). The preservation of mineralogical and chemical heterogeneities in polymict breccias, and chemical variations across single grains (e.g. overgrowths) imply that the processes involved in the amalgamation of the porphyroclasts within the polymict breccia, immediately pre-dated xenolith entrainment by the kimberlite magma (Lawless et al., 1979; Höfer et al., 2009; Pokhilenko, 2009). For this reason, and because of the broadly similar texture to hypabyssal kimberlites (i.e. mantle clasts

in a groundmass of magmatic origin), polymict breccias have been regarded as failed kimberlite intrusions, produced by proto-kimberlite magmas (Lawless et al., 1979; Pokhilenko, 2009). It follows that the investigation of polymict breccias can provide important information about the processes occurring in the mantle at the early stages of kimberlite magmatism.

In order to improve our understanding of the petrogenesis of polymict breccias, we have undertaken detailed petrological investigations of the oxide and sulphide minerals contained in a new polymict breccia xenolith (DU-1) collected from the Bultfontein Dumps (Kimberley, South Africa). Textural relationships, mineral inclusion studies, major element, trace element and Hf isotope analyses of ilmenite, rutile and sulphides were employed to constrain the composition and evolution of the melt that generated polymict breccias. Finally, we propose a connection between polymict breccias and the megacryst suite, and present a petrogenetic model that involves the generation of polymict breccias and megacrystic minerals from a common proto-kimberlite magma that interacted with mantle wall rocks at different depths (in this paper the expression “proto-kimberlite magma” refers to primitive or precursor kimberlite magma whose chemical composition cannot be defined unequivocally).

2. Analytical techniques

Xenolith sample DU-1 is relatively large (approximately 20 × 16 × 11 cm) and contains domains with distinct petrographic character and complex textures (see Section 3). For this reason, 18 thin sections were cut from different domains of the xenolith, and characterised under transmitted and reflected light, using a Nikon Labophot 2-Pol petrographic microscope. The textural relationships and compositions of the phases were then investigated in detail by Scanning Electron Microscopy (SEM) and Electron Microprobe Analysis (EMPA).

The study of xenolith microstructures and the preliminary investigation of inclusions in ilmenite were performed on carbon coated thin sections at the University of Melbourne, using a Philips (FEI) XL30 ESEM TMP, equipped with an OXFORD INCA energy-dispersive X-ray spectrometer (EDS). For more detailed examination of the inclusions hosted by ilmenite, several ilmenite grains were hand-picked under a binocular microscope, with the largest (>1 mm) mounted in epoxy. After polishing with liquid hydrocarbons (in the absence of water to preserve water-soluble inclusions) and carbon-coating, the ilmenite grains were examined at the Central Science Laboratory, University of Tasmania, using a Hitachi SU-70 field emission scanning electron microscope (FE-SEM) equipped with an OXFORD INCA-XMax80 EDS. The FE-SEM provides ultra-high resolution imaging, which enables examination of nm-sized surface features in secondary electron mode. The analytical conditions employed during the acquisition of X-rays for single-spot chemical analyses and elemental maps with the SEM and FE-SEM were as follows: beam acceleration voltage of 10–15 kV, pixel dwell time between 100 and 500 μs, and total scanning time between 5 and 15 min for each map.

Electron microprobe and LA-ICP-MS analyses were then undertaken to determine the chemical compositions of minerals in thin section. Major oxide analyses were performed using the University of Melbourne Cameca SX50 Electron Microprobe, which is equipped with four vertical Wavelength Dispersive Spectrometers. The analytical conditions were as follows: beam acceleration voltage 15 kV, beam current 35/25 nA, and beam diameter of 2 μm; counting times per analysis of 20 s on peak positions and 10 s on two background positions located on either side of the peak position; detection limits of 300 ppm for most elements.

The trace element abundances of DU-1 ilmenite and rutile were measured using a Varian 810 quadrupole ICP-MS located at the University of Melbourne. The instrument is interfaced with an excimer 193 nm UV laser ablation probe for in situ analyses of minerals – see Woodhead et al. (2007) for a detailed description of analytical procedures. Laser

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