



# A new approach to reconstructing the composition and evolution of kimberlite melts: A case study of the archetypal Bultfontein kimberlite (Kimberley, South Africa)

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## ABSTRACT

The compositions of kimberlite melts at depth and upon emplacement in the upper crust remain elusive. This can be attributed to the unquantified effects of multiple processes, such as alteration, assimilation, xenocryst contamination, and fractional crystallisation. The inability to accurately constrain the composition and physical properties of kimberlite melts prevents a comprehensive understanding of their petrogenesis.

To improve constraints on the compositions of kimberlite melts, we have combined modal analysis including the discrimination of xenocrystic from magmatic phases, with mineral chemistry determinations to reconstruct a whole-rock composition. We apply this approach to a sample of “fresh” macrocrystic hypabyssal kimberlite (sample BK-1) from the Bultfontein mine (Kimberley, South Africa). The accuracy of this whole-rock reconstruction method is validated by the similarity between reconstructed and measured whole-rock compositions. A series of corrections are then applied to account for the effects of post-emplacement serpentinisation, pre-emplacement olivine crystallisation, and the inclusion and assimilation of mantle material. This approach permits discernment of melt compositions at different stages of kimberlite evolution.

The primitive melt parental to the Bultfontein kimberlite is estimated to contain 17.4–19.0 wt% SiO<sub>2</sub>, 20.2–22.8 wt% MgO, 20.9–21.9 wt% CaO, 2.1–2.3 wt% P<sub>2</sub>O<sub>5</sub>, 1.2–1.4 wt% TiO<sub>2</sub>, 0.9–1.1 wt% Al<sub>2</sub>O<sub>3</sub>, and 0.6–0.7 wt% K<sub>2</sub>O, and has a Mg# of 83.4–84.4. Primary volatile contents (i.e., after an attempt to account for volatile loss) are tentatively estimated at ~2.1–2.2 wt% H<sub>2</sub>O and ~22.9–25.4 wt% CO<sub>2</sub>. This composition is deficient in SiO<sub>2</sub>, MgO and H<sub>2</sub>O, but enriched in CaO and CO<sub>2</sub> compared with most previous estimates of primitive kimberlite melts. We suggest that the primitive melt parental to the Bultfontein kimberlite was a transitional silicate-carbonate melt, which was progressively enriched in SiO<sub>2</sub>, MgO, Al<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, and Na<sub>2</sub>O through the assimilation of lithospheric mantle material. Comparisons with experimentally produced low-degree melts of carbonated lherzolite indicate that the Bultfontein kimberlite could have formed by ~0.5% melting of asthenospheric mantle at ~6.0–8.6 GPa (i.e., ~190–285 km) and ~1400–1500 °C. The low calculated Na<sub>2</sub>O contents (<0.2 wt%), which are inconsistent with derivation from low-degree melting of lherzolite, suggest that an alkali-bearing, volatile-rich fluid was exsolved during ascent or released after emplacement, and subsequently removed.

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## 1. Introduction

Kimberlites are volumetrically minor igneous rocks, but occur on all continents (e.g., Jelsma et al., 2009; Yaxley et al., 2013). Their deep derivation provides invaluable insights into the processes operating in the sub-continental mantle. However, despite extensive studies the compositions of kimberlite melts remain poorly constrained. Kimberlite melts have been described as volatile-rich ultramafic silicate melts (e.g., Kopylova et al., 2007; le Roex et al., 2003; Price et al., 2000); “carbonatitic” melts (e.g., Kamenetsky et al., 2008, 2014; Russell et al.,

2012); or transitional silicate-carbonate melts (e.g., Brooker et al., 2011; Nielsen and Sand, 2008). Uncertainty surrounding kimberlite melt compositions prevents a comprehensive understanding of many aspects of their petrogenesis, including the depth of melt generation (i.e., lithosphere – Becker and le Roex, 2006; le Roex et al., 2003; shallow asthenosphere – Bailey, 1980, 1993; Moore et al., 2008; Tappe et al., 2013, Tappe et al., 2017; or transition zone – lower mantle – Haggerty, 1994; Ringwood et al., 1992); and the causes of magmatism, which have been attributed to mantle plumes (e.g., Haggerty, 1994; le Roex, 1986), subduction of oceanic lithosphere (e.g., McCandless, 1999), or rapidly changing stress regimes during tectonic plate reconfiguration (e.g., Bailey, 1993; Jelsma et al., 2009; Moore et al., 2008; Tappe et al., 2017).

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Deciphering the composition of kimberlites is currently hampered by pre-, syn- and post-emplacement processes that modify the composition of kimberlites. These include: (1) hydrothermal alteration (e.g., Afanasyev et al., 2014; Giuliani et al., 2014, 2017; Stripp et al., 2006); (2) volatile loss through reactions with wall-rocks, fluid exsolution during ascent, and degassing upon emplacement (e.g., Brooker et al., 2011; Moussallam et al., 2016; Russell et al., 2012); (3) magmatic differentiation (e.g., Kjarsgaard, 2007; Mitchell, 2008); (4) inclusion and assimilation of xenogenic material (e.g., Brett et al., 2009; Giuliani et al., 2016; Kamenetsky et al., 2008; Schulze, 2001; Soltys et al., 2016).

Accordingly, parental kimberlite melt compositions cannot be determined directly from whole-rock geochemical analyses without taking the above processes into account. Previous studies have reconstructed kimberlite melt compositions using several approaches: (1) Whole-rock measurement of aphanitic kimberlites (e.g., Kopylova et al., 2007; Price et al., 2000; Shee, 1985); (2) subtraction of xenocrystic components from whole-rock compositions (e.g., Kjarsgaard et al., 2009; Nielsen and Sand, 2008); and (3) projections in geochemical plots using large whole-rock datasets from a single locality (e.g., le Roex et al., 2003). Most of these reconstructions imply that primitive kimberlites are Al- and Na-poor, volatile-rich, ultramafic melts containing ~25–35 wt% SiO<sub>2</sub> and MgO, and ~12–20 wt% CaO with variable H<sub>2</sub>O and CO<sub>2</sub> concentrations (e.g., Becker and le Roex, 2006; Kjarsgaard et al., 2009; Kopylova et al., 2007; le Roex et al., 2003; Price et al., 2000). However, these reconstructed melt compositions cannot be considered primary, because they are too Mg-rich to have been in equilibrium with potential mantle source rocks (e.g., Kopylova et al., 2007). Moreover, experimental studies have shown that these reconstructed compositions do not form pure melts at the pressure-temperature conditions of kimberlite emplacement (i.e., 100–200 MPa and 1100–1275 °C), and are unable to dissolve the measured whole-rock volatile contents (Brooker et al., 2011; Moussallam et al., 2016; Sparks et al., 2009).

Since the last attempts to reconstruct kimberlite melt compositions, there have been significant advances in our understanding of the origin of minerals contained in kimberlites (i.e., magmatic vs. xenocrystic vs. secondary/hydrothermal). These advances permit a new approach to estimating kimberlite melt compositions. This contribution focuses on a sample of “fresh” hypabyssal macrocrystic kimberlite (sample BK-1) from the diamondiferous (ca. 84 Ma; Kramers et al., 1983) Bultfontein kimberlite, Kimberley (South Africa). This sample was selected for melt reconstruction as the petrography, mineral chemistry and isotope systematics are well constrained and representative of the Kimberley kimberlites (Giuliani et al., 2016, 2017). In this study, we employed detailed image analysis to provide modal mineral abundances and to discriminate magmatic from xenocrystic components. These data were then combined with mineral densities and compositions to reconstruct the whole-rock composition. Finally, a series of corrections were applied to account for the syn- and post-emplacement alteration, the inclusion and assimilation of mantle material, and pre-emplacement crystallisation. The resulting reconstructed primitive melt composition is used to provide an assessment of the conditions of generation of the Bultfontein kimberlite.

## 2. Background

### 2.1. Primary kimberlite mineralogy

Kimberlites are holocrystalline and inequigranular igneous rocks that exhibit textures ranging from aphanitic to macrocrystic. They are notoriously susceptible to post-emplacement alteration; however, the origins of the fluids responsible for alteration are highly contentious (see next section). Kimberlites are hybrid rocks, containing a mixture of xenolithic and xenocrystic components of various origins (i.e., mantle and crustal) as well as magmatic and secondary (i.e. post-magmatic) phases. Discerning the origin of phases contained in kimberlites is challenging because many minerals can have multiple origins (e.g., Giuliani et al., 2016; Kamenetsky et al., 2008; Mitchell, 1986; Schulze, 2001). Nevertheless,

the mineralogy of hypabyssal kimberlites is broadly similar worldwide, typically including liquidus olivine, spinel group minerals, micas, perovskite, apatite, monticellite, and ilmenite, set in a cryptocrystalline matrix of carbonates (calcite and lesser dolomite) and serpentine (e.g., Mitchell, 1986, 2008).

### 2.2. Post-, syn- and pre-emplacement modification of kimberlites

#### 2.2.1. Serpentinisation

In this section, we refer only to pervasive syn- and post-emplacement alteration of ‘relatively fresh’ hypabyssal kimberlites and not to subaerial weathering (e.g., Mitchell, 2008). Serpentinisation is the predominant style of alteration, where serpentine may constitute up to 50 vol% of kimberlites and is the major host of H<sub>2</sub>O. Serpentine commonly replaces silicate phases (i.e., olivine, monticellite and phlogopite), but can also replace carbonates and apatite (e.g., Giuliani et al., 2017; Mitchell, 1986; Sparks et al., 2009). Serpentinisation reactions have the potential to increase whole-rock H<sub>2</sub>O/CO<sub>2</sub>, MgO/CaO, and SiO<sub>2</sub>/CaO ratios (e.g., Sparks et al., 2009), although quantifying these effects is challenging. In addition, serpentine occurs in the kimberlite matrix where it apparently does not replace a previous magmatic phase. Understanding the origin of this matrix serpentine is significant to modelling kimberlite melt compositions, because in otherwise “fresh” kimberlite samples (i.e., those with little pseudomorphic serpentine) it is volumetrically the most abundant form of serpentine.

There are two main opposing views on the origin of serpentine-forming fluids in hypabyssal kimberlite rocks:

- (1) Deuteric (i.e., late-stage magmatic) fluids, derived entirely from the kimberlite melt (Mitchell, 2008, 2013). In this model, serpentine is viewed as an inherent part of the kimberlite crystallisation sequence and should be included in melt reconstructions.
- (2) Fluids largely (or completely) derived from external sources (i.e. groundwater; Afanasyev et al., 2014; Brooker et al., 2011; Sparks et al., 2009; Stripp et al., 2006), which may mix with residual deuteric components (Giuliani et al., 2014, 2017). Here, the dissolved components (i.e., MgO and SiO<sub>2</sub>) are largely derived from the alteration of kimberlitic phases; whereas serpentine is a secondary, pore filling or replacement phase. In this case, most of the serpentine should not (or only partly) be included in melt reconstructions.

The experimental studies of Brooker et al. (2011), Moussallam et al. (2016) and Sparks et al. (2009) have shown that the amount of H<sub>2</sub>O required for an entirely deuteric origin of serpentine in some hypabyssal kimberlite rocks (i.e., ~5–10 wt% H<sub>2</sub>O) exceeds the solubility of H<sub>2</sub>O in a large spectrum of analogue kimberlite melts (i.e., 18–28 wt% SiO<sub>2</sub>; Moussallam et al., 2016) at the pressure-temperature conditions of emplacement. Kimberlites evidently retain some magmatic H<sub>2</sub>O, as they crystallise primary mica, and there may be a deuteric component in early generations of serpentine (Giuliani et al., 2017). However, following emplacement, there is likely extensive mixing of residual deuteric fluids with groundwater, which hampers accurate estimation of the H<sub>2</sub>O content of parental kimberlite melts. Therefore, it appears no simple end-member model (i.e., entirely deuteric or external) can accurately explain the origin of serpentine in kimberlite rocks.

#### 2.2.2. Incorporation and assimilation of xenocrystic material

Kimberlites host an assortment of xenocrystic and xenolithic components of diverse origins and it is evident from reaction textures between the host kimberlite and these entrained phases that kimberlite melts are highly reactive (e.g., Russell et al., 2012; Shee, 1985; Smith et al., 2004; Soltys et al., 2016). There is abundant petrographic and experimental evidence that orthopyroxene and, to a lesser extent, clinopyroxene and garnet are assimilated by kimberlite melts (Chepurov et al., 2013; Hunter and Taylor, 1982; Kamenetsky et al.,

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