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### **Chemical Geology**

journal homepage: www.elsevier.com/locate/chemgeo

# Chemical abrasion of zircon and ilmenite megacrysts in the Monastery kimberlite: Implications for the composition of kimberlite melts

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#### ARTICLE INFO

Article history: Received 3 March 2014 Received in revised form 29 May 2014 Accepted 4 June 2014 Available online 19 June 2014

Editor: K. Mezger

Keywords: Kimberlite Mantle Megacryst Melting Mineralogy Melt inclusions

#### ABSTRACT

Ilmenite and zircon megacrysts, among other minerals representing the subcontinental lithospheric mantle, are exclusively delivered to the surface by kimberlite magmas. The intimate association of ilmenite and zircon with their transporting kimberlite melts still remains perplexing, as these minerals do not belong to the kimberlite liquidus assemblage at crustal pressures. The ilmenite and zircon megacrysts from the Monastery kimberlite (South Africa) represent a textbook example of the megacryst suite. The megacrysts show substantial chemical modification along contacts with the host kimberlite. Fine-grained "reaction" assemblages, comprising minerals rich in Zr (baddeleyite and sodium-zirconium silicates) and Ti (Ti-Fe oxides, perovskite, sphene, kassite), are present around zircon and ilmenite, respectively. At the zircon-ilmenite contact, chemical contributions from both minerals are recorded in Zr-Ti-rich phases such as calzirtite and zirkelite. The megacrysts contain crystallised melt pools and secondary melt inclusions in healed fractures; their mineral assemblage is dominated by alkali-bearing phases, including silicates (nepheline, kalsilite, sodalite, phlogopite-tetraferriphlogopite), titanates (priderite, freudenbergite), zirconium silicates (khibinskite, parakeldyshite), carbonates (zemkorite, eitelite), phosphates (apatite, bradleyite, nahpoite), sulfates (aphthitalite) and chlorides (halite, sylvite). These inclusions and melt pools are interpreted to be produced by reaction between the megacrysts and the transporting kimberlite melt, which infiltrated fractures in the megacrysts. Most secondary minerals at contacts with kimberlite require a supply of Ca, which is readily available in the carbonatite component of the kimberlite magma. The enrichment of the encapsulated mineral assemblages in alkali and volatile elements (Na, K, S, Cl) also appears to originate from the kimberlite melt.

The similar U–Pb ages and identical Hf-isotope compositions of the megacryst assemblage ( $89.2 \pm 2.8$  Ma;  $\epsilon$ Hf -0.4 to +1.3), the reaction assemblage ( $98 \pm 7$  Ma) and the host kimberlite ( $90 \pm 4$  Ma;  $\epsilon$ Hf -0.6 to +1.7), imply their close genetic affinity. Although the megacrysts and kimberlite magma originated from the same source at the same time, the chemical disequilibrium recorded in the alteration of megacrysts precludes a simple "parental melt-cognate crystal" relationships. This apparent paradox can be resolved by considering the unmixing of a protokimberlite melt into silicate-oxide and carbonate liquids at mantle conditions.

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#### 1. Introduction

Large crystals and their intergrowths (so-called megacrysts, up to several cm in size) are delivered by kimberlite and alkali basalt magmas from the lithospheric mantle worldwide. The most common megacryst minerals are garnet, clinopyroxene, orthopyroxene and ilmenite, whereas olivine, phlogopite and zircon are less common (e.g. Gurney et al., 1979).

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http://dx.doi.org/10.1016/j.chemgeo.2014.06.008 0009-2541/© 2014 Elsevier B.V. All rights reserved. The megacryst assemblages entrained by kimberlites 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). As summarised by Bell and Moore (2004), kimberlite megacrysts may have formed by a combination of processes, including: i) fractional crystallisation of discrete magmatic intrusions with very little wall rock interaction; ii) melt hybridisation in an ascending diapir of asthenospher-ic material; and iii) metasomatic conversion of peridotites by channelized "megacrystic" magmas. Compositional homogeneity is common in







megacrysts and suggests residence at high temperature and/or long time in the mantle before being entrained by the kimberlite magma (Gurney et al., 1979).

A cognate origin for the megacryst suite from the host kimberlite magma is supported by i) radiogenic isotope data (e.g., Nowell et al., 2004; Kopylova et al., 2009); ii) the presence of inclusions of kimberlitic material in megacrysts (Gurney et al., 1979; Rawlinson and Dawson, 1979); and iii) the close timing often inferred to occur between megacryst formation and kimberlite entrainment (e.g., Kinny et al., 1989; Kopylova et al., 2009). However, trace-element modelling indicates that the megacryst minerals crystallised from melts that are compositionally distinct from the host kimberlites (e.g., Davies et al., 2001; Merry and Le Roex, 2007). On the other hand, the intimate association between megacrysts and transporting kimberlite magmas is unlikely to be a coincidence, and thus invites further exploration.

The "classic" megacryst suite in the Monastery kimberlite, renowned for the size of its crystals, diversity and preservation (Whitelock, 1973; Gurney et al., 1979), contains zircon and ilmenite; unlike other megacryst minerals, these are typically absent in common mantle lithologies. Significant alteration recorded on the rims of zircon and ilmenite (e.g., Kresten et al., 1975; Raber and Haggerty, 1979; Pasteris, 1980; Haggerty, 1987, 1995; LeCheminant et al., 1997; Patchen et al., 1997; Golubkova et al., 2013, Anashkin et al., 2013a,b) provides an opportunity to further investigate the genetic relationships between the assemblage of kimberlite-hosted megacrysts and their host kimberlite magma. In this paper we present compositional and geochronological data for the mineral assemblage occurring at contacts of zircon and ilmenite with the kimberlite groundmass, and seek answers to when, where and why the modification of megacrysts took place. Additionally, we use the chemical signature of the alteration products to constrain the composition of the kimberlite melt. The analytical methods employed in this study are presented in Supplementary Materials.

#### 2. Ilmenite-zircon megacryst assemblage

The megacrysts minerals are more abundant in the Group I Monastery kimberlite pipe in South Africa than in any other studied kimberlite on the Kaapvaal craton (e.g., Moore et al., 1992; Griffin et al., 1997). In the main Cr-poor megacryst suite at Monastery, coexisting garnet, clinopyroxene and orthopyroxene crystallised early, joined by ilmenite and phlogopite, then joined by zircon, and finally by Fe-rich olivine (Gurney et al., 1979; Moore et al., 1992). The well-known Monastery ilmenite–zircon megacryst assemblage (e.g., Mitchell, 1973; Moore et al., 1992; Griffin et al., 1997; Belousova et al., 1998) comprises large crystals (>5–15 mm) that commonly form intergrowths and are characterised by an almost complete absence of crystal faces (Figs. 1A, 2). They are irregular to roughly ovoid and even rounded in shape. Zircon inclusions (>0.5 mm in size) are observed in ilmenite but not vice versa. The ilmenite is typical kimberlitic picroilmenite with MgO up to 10– 15 wt.% (e.g., Mitchell, 1973; Griffin et al., 1997). Zircon megacrysts and zircon inclusions in ilmenite are compositionally similar (Supplementary Table S1). They have trace-element contents within the range defined for zircons from other kimberlites in South Africa, Siberia and Australia (Belousova et al., 1998, 2002). Remarkably low levels of most trace elements (total REE < 50 ppm, Y 20–75 ppm, U 6.5–38 ppm, Th 1–13 ppm, both Nb and Ta < 3 ppm) and chondrite-normalised REE patterns with low and flat HREE, combined with the almost complete absence of Eu anomalies (Supplementary Table S1), make them clearly distinct from crustal zircons (e.g., Belousova et al., 1998, 2002). The zircon megacrysts analysed during this study show a narrow range in Hf-isotope composition with  $\varepsilon$ Hf varying from + 0.2 to + 1.3 (Supplementary Table S2), which is similar to the range (from - 1.9 to + 0.9), previously reported for the Monastery zircon (Griffin et al., 2000).

The titanium content of the Monastery zircon was used to calculate crystallisation temperature using the Ti-in-zircon thermometer (Watson and Harrison, 2005; Watson et al., 2006). The variation in Ti content (11 to 40 ppm) in the megacrysts produces a range of apparent temperatures (750 to 850 °C; Supplementary Table S3). These are broadly similar to the temperatures reported for 84 zircon megacrysts from South African kimberlites (758  $\pm$  98 °C; Fu et al., 2008).

The mineralogically complex alteration features of the megacrysts have been described previously (e.g., Kresten et al., 1975; Raber and Haggerty, 1979). In our study these features include the rims around the megacrysts and alteration patches occurring along fractures and trails of crystalline melt inclusions.

#### 3. Alteration of megacrysts: types and mineralogy

A prominent feature of the Monastery ilmenite and zircon megacrysts is the alteration affecting their rims at the contacts with the host kimberlite groundmass and with each other, as well as internal fractures and "melt pools" in both minerals (Figs. 1–6). The alteration rims are up to 5 mm wide, typically wider around zircon and much narrower around ilmenite, and have diverse phase assemblages (Tables 1, 2). Alteration at the contact between zircon and the kimberlite groundmass (Figs. 1B, C, 2) is the most developed in terms of width, zonation and compositional complexity, and has been effective in smoothing the edges of zircon.

At least five subparallel zones, differing in texture, grain sizes and mineral proportions can be recognised in the alteration rims (Fig. 2). The main mineral constituent is Zr oxide (tentatively, baddeleyite) interspersed in a fine-grained aggregate of Al-bearing Mg-silicate (serpentine?), calcite, phlogopite and apatite. This assemblage resembles the mineralogy of the host kimberlite groundmass, whereas other kimberlite minerals – euhedral olivine (now serpentinised), perovskite, Cr-spinel and Fe-oxides do not appear in the alteration products. The outermost rim of the alteration zone, composed mainly of acicular



Fig. 1. Optical photograph (A) and back-scattered electron images (B, C) displaying fragments of intergrown zircon (zr)-ilmenite (ilm) megacrysts, showing alteration bands along contacts with the host kimberlite (kimb), with each other and along fractures.

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