



High-density fluids and the growth of monocrystalline diamonds

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Abstract

The chemical nature and composition of the growth medium of monocrystalline (MC) diamonds is still a matter of debate, partially because carbonate-bearing high-density fluids (HDFs) that are common in fibrous diamonds have not been found in MC diamonds. Here we report the first finding of HDF microinclusions in a MC octahedral diamond from Finsch, South Africa and in the MC octahedral core of a coated diamond from Kankan, Guinea; both diamonds carry nitrogen in B-centers.

Numerous microinclusions in diamond Finsch_2a_cap1 are restricted to two thin layers parallel to the (1 1 1) face, ~20 and 200 μm from the diamond rim. Low-Mg carbonatitic HDFs are found along the inner layer while the outer layer trapped saline compositions. The major and trace element compositions of the inclusions and their infrared spectra are highly similar to those of microinclusions found in fibrous diamonds. A few isolated microinclusions of saline compositions are scattered around a sulfide inclusion in the center of the octahedral core of diamond ON-KAN-383.

This evidence for the involvement of oxidized fluids in the formation of MC diamonds adds to previous reports on the antiquity of HDFs in fibrous diamonds, the presence of carbonate and halide phases in inclusions in MC diamonds and the similarity of trace element pattern of a MC diamond to those of low-Mg carbonatitic HDF in fibrous diamonds. In addition, we show that the interaction of HDFs with depleted garnets can produce sinusoidal REE patterns which are one of the primary features of Iherzolitic and harzburgitic garnet inclusions in MC diamonds. Together, these observations suggest that HDFs are involved in the formation of many types of diamonds from the Archaean to the Phanerozoic. HDFs are trapped in large quantities during rapid, fibrous growth, but must also be present during the growth of many MC diamonds.

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1. INTRODUCTION

It is broadly accepted that diamond formation in the Earth's mantle is related to metasomatic processes induced by C–O–H-bearing fluids or melts. This theory is based on the concentric growth patterns of diamonds as revealed in cathodoluminescence (CL) images (Sunagawa, 1981, 1984; Bulanova, 1995; Davies et al., 1999; Tappert and Tappert,

2011), which suggest they grew undisturbed into surrounding fluids/melts; on their association with veins and alteration zones in xenoliths (Meyer, 1987; Schulze et al., 1996; Keller et al., 1998; Taylor et al., 2000; Taylor and Anand, 2004; Anand et al., 2004; Bulanova et al., 2004); and on evidence that mineral inclusions in diamonds show enrichment in incompatible trace elements, derived from the fluids that also deposited their host diamond (Shimizu and Richardson, 1987; Stachel et al., 2000, 2004; Promprated et al., 2004; Stachel and Harris, 2008; Klein-BenDavid and Pearson, 2009).

Two principal species of C-bearing fluids are relevant to the formation of lithospheric diamonds: carbonate-rich (CO₂) and methane-rich (CH₄) fluids. Under the

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oxygen-fugacity (fO_2) conditions of the subcratonic lithosphere, which vary between ~ 1 log unit below the fayalite–magnetite–quartz (FMQ) buffer reaction at ~ 100 km to ~ 4 units below FMQ at depths of ~ 220 km (Woodland and Koch, 2003; McCammon and Kopylova, 2004; Frost and McCammon, 2008), both CO_2 -rich and CH_4 -rich fluids/melts may lead to the formation of diamonds through redox reactions (Frost and McCammon, 2008; Stachel and Harris, 2009). Experimental modeling has shown that diamonds can nucleate from CO_2 -rich fluids/melts and grow by such reactions (Kumar et al., 2000; Sokol et al., 2001; Pal'yanov et al., 2002; Tomlinson et al., 2004; Pal'yanov and Sokol, 2009; Bataleva et al., 2012; Bureau et al., 2012), from CH_4 -rich fluids (Tomilenko et al., 1998; Sokol et al., 2009) or from the reduction of carbonates by CH_4 (Yamaoka et al., 2002).

Natural diamonds occur in three main textural varieties (Sunagawa, 1984, 2005; Wilks and Wilks, 1991; Tappert and Tappert, 2011): monocrystalline (MC) diamonds, mostly of octahedral morphology, but also of cubic habit (twins -mostly maccles- and parallel intergrowths oriented along the same crystallographic planes are commonly grouped with the MC diamonds); polycrystalline diamonds, including carbonados and framesites (diamondites; Kurat and Dobosi, 2000); fibrous diamonds, including diamonds with cube-like morphology, fibrous coats overgrown on MC diamond (coated diamond) and MC diamonds with internal zones of fibrous growth (cloudy diamonds). Direct sampling of the growth medium is enabled by high-density fluids (HDFs) trapped as microinclusions, commonly found in all types of fibrous diamonds. These inclusions indicate that fibrous diamonds precipitated from oxidized fluid/melts (i.e. carbonate-bearing fluids; Navon et al., 1988; Schrauder and Navon, 1994; Shiryaev et al., 2005; Tomlinson et al., 2006; Klein-BenDavid et al., 2009). Throughout this paper, we use the term HDFs when referring to such carbonate-bearing fluids/melts. The formation mechanism for polycrystalline diamonds is not clear (Haggerty, 1999; Heaney et al., 2005); however, fluid microinclusions were recently found in one framesite from Orapa, Botswana (Jacob et al., 2011) and two carbonado diamonds from Brazil and central Africa (Kaminsky et al., 2013), suggesting the involvement of HDFs in polycrystalline diamond formation. Trace element analyses of a few framesite diamonds, presumably from Botswana, which show similarities to kimberlite and carbonatitic melts corroborate this idea (Dobosi and Kurat, 2002; Rege et al., 2008), but additional data are needed for any firm conclusion. To date, microinclusions carrying HDFs have not been documented in MC diamonds, furthering the debate on whether HDFs are responsible for the formation of all types of diamond. Indeed, it has been suggested that MC diamonds grow from the oxidation of asthenosphere-derived reduced C–H–O fluids (i.e. methane-rich fluids; Deines, 1980; Cartigny et al., 1998; Cartigny, 2005; Thomassot et al., 2007; Stachel and Harris, 2009). This suggestion is based mainly on variations in $\delta^{13}C$, $\delta^{15}N$ and the N contents of such diamonds, which are consistent with the stable-isotope fractionation predicted for diamond growth from methane-rich fluid. However, except for a single

report of direct detection of reduced gases in inclusions within a Siberian diamond (Tomilenko et al., 1997), inclusions of CH_4 -bearing fluids have not been found in natural diamonds.

Another notable difference between fibrous and MC diamonds is that almost all nitrogen in the former resides in A-centers, whereas most MC diamonds carry both A- and B-centers. The higher aggregation state of nitrogen (B-centers) is usually attributed to the antiquity of MC diamond compared with the short residence time of fibrous diamonds at mantle temperatures (Boyd et al., 1987; Navon, 1999; Gurney et al., 2010). Based on these differences and the age of silicate and sulfide inclusions in MC diamonds, Stachel and Harris (2009) associated Paleo-Archean to Meso-Archean ages with methane-bearing fluids and younger Proterozoic and Phanerozoic ones with carbonate-bearing melts (HDFs).

Here, we report the first finding of HDF microinclusions in a MC octahedral diamond from Finsch, South Africa and in the MC octahedral core of a coated diamond from Kankan, Guinea. Both diamonds carry nitrogen in A- and B-centers. Using FTIR, EPMA and LA-ICP-MS analyses we characterized the compositions of these HDFs and show that they share great similarities with common HDFs that form fibrous diamonds.

2. SAMPLES AND ANALYTICAL TECHNIQUES

We report the analyses of two samples of MC diamonds. The first is a cleaved piece of an octahedral diamond from the Finsch mine in South Africa (Finsch_2a_cap1), which is 4.6 mm in length, 1.2 mm in width and has D grade color. This diamond contains numerous microinclusions that are restricted to two thin layers, ~ 10 μm thick, parallel to the (111) face and located ~ 20 and 200 μm from the rim of the diamond (Fig. 1 and Figs. A and B in Supplementary material). The second is a coated diamond from Kankan, Guinea (ON-KAN-383), which was described in detail by Weiss et al. (2009). This diamond has a zoned, microinclusion-bearing fibrous coat and contains numerous mineral inclusions in its octahedral core (Fig. G in Supplementary material).

The diamonds were laser-cut and polished on both sides to create a thin slab. They were then cleaned ultrasonically in HF 60% and HNO_3 69% for 2 h and washed with ethanol and distilled water before analysis. FTIR analyses were made using a Bruker IRscope II microscope coupled to a Nicolet 740 FTIR spectrometer (Gloar source, KBr beamsplitter, He–Ne laser, MCT detector) at the Institute of Earth Sciences, the Hebrew University of Jerusalem, Israel (HUJI). Spectra were collected in the range of 550–4000 cm^{-1} with readings every 2 cm^{-1} . The presence of nitrogen B centers was evaluated after subtracting the diamond bands and the appropriate absorption of pure IaA spectrum. Nitrogen concentration and aggregation states were determined using a computer program supplied by D. Fisher (De Beers Technologies UK, Maidenhead) and the absorption coefficients of A-centers, B-centers and C-centers (Boyd et al., 1994a, 1995; Kiflawi et al., 1994; Zaitsev, 2001). The uncertainties on the A- and B-centers

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