



## Diamond-forming fluids in fibrous diamonds: The trace-element perspective

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### ABSTRACT

We closely examine the trace-element concentrations of fifteen microinclusion-bearing diamonds from Canada, Guinea and South Africa that trapped low-Mg carbonatitic, silicic and saline high-density fluids (HDFs). The microinclusions trapped only HDFs, with no mineral microinclusions or mixtures of HDF+mineral; thus, their LA-ICP-MS analyses solely represent the compositions of the trapped fluids.

HDFs of different major-element compositions (silicic, carbonatitic and saline) are characterized by fractionated REE patterns and variable, mostly negative, anomalies in Sr, Ti, Zr, Hf and Y relative to REEs of similar compatibility, regardless of their host diamond's provenance. In the highly incompatible elements (Cs–Pr) two patterns are notable. “Ribbed” patterns are characterized by high levels of Ba, Th, U and LREEs and lower alkalis, Nb and Ta. “Planed” patterns are smoother and typically devoid of significant fractionation between elements of similar compatibility. Co-variation diagrams of (La, Ce)/(Nb, Rb) vs. (U, Th)/(Nb, Rb) and Rb/Nb vs. La/Nb ratios can be used to best distinguish between the two patterns.

Similarities of canonical ratios, such as Nb/(Th, U, La) and K/U, between MORB and OIB samples and HDFs with “Planed” patterns suggest an asthenospheric source for these HDFs. For silicic compositions, this idea is strengthened by calculating the sources in equilibrium with such HDFs which range in composition between the DMM and more fertile parts of the convecting mantle. Isotopic analyses may pinpoint this connection.

The trace-element patterns of MARID and PIC xenoliths show a mirror-image to the pattern of silicic, low-Mg carbonatitic and saline HDFs with “Ribbed” patterns. Assuming that the HDFs are the product of a 0.1% batch melting, we obtain a source that is similar to the MARID and PIC patterns. However, the decoupling between major- and trace-elements in the HDFs argues against melting or fractional crystallization as the main processes leading to the formation of the “Ribbed” patterns.

Percolation of an asthenospheric silicic HDF with “Planed” pattern through previously metasomatized lithosphere that carries accessory phlogopite and Fe–Ti oxides, closely reproduce the “Ribbed” pattern of silicic and low-Mg carbonatitic HDFs at fluid/rock ratios  $\approx 0.1\%$ . The initial trace-element pattern of the lithosphere influences the more compatible elements of the HDF (Sr–Lu). However, in the Cs–Pr range, the presence of phlogopite and Ti–Fe oxides controls the evolution of the “Ribbed” pattern. Percolation explains the observed decoupling between major- and trace-elements in HDFs and the resemblance of trace-element patterns in HDFs from different cratons. It may also explain the limited variation of  $\delta^{13}\text{C}$  in fibrous diamonds ( $-6 \pm 2\%$ ). The two patterns escape the circular “chicken and egg” reasoning that calls for an enriched source for the formation of highly fractionated melts: it suggests that diamond-forming fluids can come directly from the asthenosphere (with no need for a pre-metasomatized source) and that they can be further modified in the lithosphere.

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### 1. Introduction

Metasomatism involves the enrichment of a rock with new substances brought in from the outside (Goldschmidt, 1922) and in

most cases involves fluids. There are relatively few constraints on the chemistry, transport mechanism and the scale of this process within the Earth's mantle (Hofmann, 1984). Nevertheless, the nature of chemical modification and enrichment of mantle-derived xenoliths, xenocrysts and mineral inclusions in diamonds, indicates the involvement of enriched melts,  $\text{H}_2\text{O}-\text{CO}_2$  supercritical solutions or other C–H–O fluids in an open-system alteration of many mantle rocks (Menzies and Hawkesworth, 1987;

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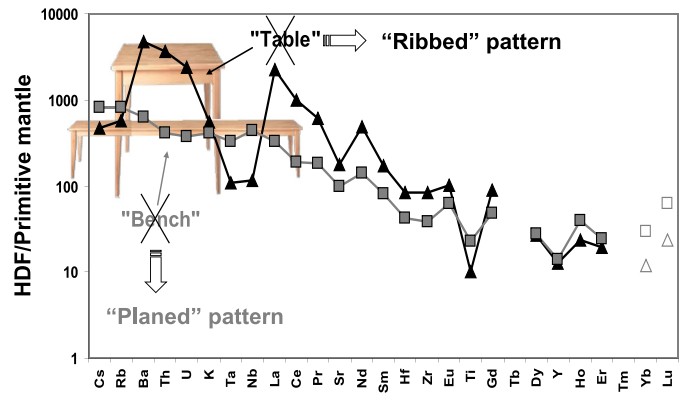
Pearson et al., 2003; Stachel and Harris, 2008; Griffin et al., 2009; O'Reilly and Griffin, 2013; and references therein). While many of the products of such fluid–rock interaction have been analyzed, the chemical composition of the fluids that promoted the metasomatism could only be assumed or calculated using mineral/melt partition coefficients.

The morphology of diamonds and their symmetric growth patterns (Sunagawa, 1981, 1984; Bulanova, 1995; Davies et al., 1999) suggest that they have precipitated from fluids, while their common association with veins and alteration zones (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) directly connects them with the process of mantle metasomatism. Fibrous diamonds that have trapped fluids in microinclusions act as “time capsules”: they grew during the metasomatic event and captured the fluid agents “in action”. Therefore, they provide direct chemical evidence on diamond-forming processes in particular, and on mantle metasomatism in general.

Secondary-ion mass spectrometry (SIMS) and electron microprobe analysis (EPMA) have revealed that the major-element compositions of these sub-micrometer inclusions vary between four end-members: saline high-density fluids (HDFs) which carry mostly K, Na and Cl and are depleted in carbonates and silicates, high-Mg carbonatitic HDFs characterized with high MgO and carbonate content and low silica and alumina, low-Mg carbonatitic and silicic HDFs with varying amount of carbonates, silicates and water (Navon et al., 1988; Schrauder and Navon, 1994; Izraeli et al., 2001, 2004; Shiryayev et al., 2005; Tomlinson et al., 2006; Klein-BenDavid et al., 2007, 2009; Zedgenizov et al., 2007, 2009; Weiss et al., 2009). Quantitative determination of the water content by infrared spectroscopy indicates that the high-Mg and the low-Mg carbonatitic to silicic HDFs are, in fact, hydrous melts with 10–25 wt% water (Weiss et al., 2010). However, in this paper, we will retain the more general term – HDF.

Following the early neutron-activation work of Fesq et al. (1975) and Bibby (1979), Schrauder et al. (1996) used INAA to measure the concentrations of 31 elements in fibrous diamonds from Jwaneng, Botswana. Akagi and Masuda (1988) burned Zairian fibrous diamonds and used isotope-dilution mass-spectrometry to determine their Sr isotopic composition and REE, Sr, Rb, K and Ba concentrations. These studies demonstrated that the HDFs have elevated levels of incompatible elements compared to primitive mantle (PM) values and that the REE patterns are similar to those of kimberlites. More data have been collected in the last few years with the development of laser ablation, allowing rapid “on-line” analysis of many trace elements in diamonds (Resano et al., 2003; Rege et al., 2005, 2010; Tomlinson et al., 2009), as well as “off-line” ablation followed by mass spectrometry (McNeill et al., 2009; Klein-BenDavid et al., 2010). Both “on-line” and “off-line” ablation methods found similar trace-element patterns in fibrous diamonds: fractionated REE patterns and variable, mostly negative anomalies (PM normalized) of the alkalis (K, Rb and Cs), high field strength elements (HFSE: Zr, Ti, Nb, Ta, Hf), Sr and Y relative to Ba, Th, U and REEs of similar compatibility (Zedgenizov et al., 2009; Weiss et al., 2009, 2011; Tomlinson et al., 2009; McNeill et al., 2009; Klein-BenDavid et al., 2010; Rege et al., 2010). As a rule, the trace-elements in fibrous diamonds reside in the microinclusions; thus, the measured values may be corrected using the EPMA data to obtain the trace-element concentrations of the trapped fluids themselves (Weiss et al. 2008a, 2009).

Weiss et al. (2008b) showed that all HDFs share similar incompatible-element characteristics regardless of their major-element compositions or the host diamond provenance and identified two principal patterns in the highly incompatible elements (Cs–Pr); one is mostly flat with no significant anomalies and a



**Fig. 1.** Primitive-mantle-normalized incompatible element spidergram demonstrating the two common patterns of HDFs in fibrous diamonds. The two patterns, originally designated as “Bench” and “Table” patterns by Weiss et al. (2008b) are here renamed “Planned” and “Ribbed” patterns, respectively, to better describe their geometry. The patterns were also named “fibrous-low” and “fibrous-high” (Rege et al., 2010) and “plateau” and “valley” (Klein-BenDavid et al., GCA, in review). For simplicity, only two silicic HDFs (diamonds ON-JWN-110 and ON-KAN-381) with end-member trace-element characteristics are shown, but the two patterns are common in carbonatitic, silicic and saline HDF compositions, regardless of the provenance of their host diamond (see Fig. 2). The uncertainties on the values of Yb and Lu (gray symbols) are large and they can only be regarded as qualitative. Primitive-mantle values are from McDonough and Sun (1995).

moderate decrease in concentrations from the most incompatible elements toward the more compatible ones. The other has elevated Ba, U, Th and LREE, depleted Nb and Ta and in most cases, highly depleted alkalis (Fig. 1). These patterns were designated first as “Benches” and “Tables” (Weiss et al., 2008b) and later as “fibrous-low” and “fibrous-high” (Rege et al., 2010). In order to accurately describe the geometry of the two trace-element patterns in fibrous diamonds and to unify the nomenclature used by different groups, we suggest renaming the patterns as “Planned” pattern and “Ribbed” pattern (Fig. 1) and use these expressions in future studies.

The overall patterns of the HDFs were explained in several ways: Tomlinson et al. (2009) discussed the possibility of wall-rock reaction and fractional crystallization of carbonate to explain the enrichment in light-REE and large-ion lithophile elements (LREE and LILE, respectively) and the differential enrichment of Sr. Rege et al. (2010) preferred immiscibility between carbonatitic and hydrous-silicic components to explain the variations in Nb/Ta, Zr/Hf, Ba/LREE and LREE/HREE ratios and the negative anomalies of Sr. The possibility of providing most trace-elements by saline solutions or by percolating hydrous fluids that interact with large volumes of mantle rocks, as well as the need for rutile in order to explain the large negative anomalies of Nb and Ta, were suggested by Weiss et al. (2009). Furthermore, they pointed out that the trace-element patterns of high-Mg carbonatitic HDFs trapped in diamonds from Kankan, Guinea and Siberia (Zedgenizov et al., 2007) were distinct from the low-Mg carbonatitic, silicic and the saline end-members by showing enrichment in Nb and Ta compared to LILE and LREEs. The deeper negative anomalies of K, Rb, Cs, Nb and Ta and the extreme enrichment in Ba, Th, U and LREE of high-Mg HDFs from Kankan, relative to those from Siberia were explained by Weiss et al. (2011) as the result of lower degrees of melting and the presence of phlogopite and traces of rutile in the sub-continental lithospheric mantle (SCLM) in the Kankan source.

In the present study we closely examine the trace-element concentrations of microinclusion-bearing diamonds that trapped low-Mg carbonatitic, silicic and saline HDFs. The data is used to explore the potential sources of diamond-forming fluids and to better understand the processes that created their specific chemical trace-element patterns.

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