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Fluid escape from diamond caught-in-the-act: toward the composition and origin of diamond-forming fluids

Luísa D.V. Carvalho [1], Thomas Stachel [1], Robert W. Luth [1], Andrew J. Locock [1], D. Graham Pearson [1], Matthew Steele-MacInnis [1], Richard A. Stern [1], Fabrizio Nestola [2], Ricardo Scholz [3], Tiago Jalowitzki [4], Reinhardt A. Fuck [4]

[1] *University of Alberta, Edmonton, Canada, luisa.carvalho@ualberta.ca, tstachel@ualberta.ca, rluth@ualberta.ca, alocock@ualberta.ca, gdpearso@ualberta.ca, steelema@ualberta.ca, rstern@ualberta.ca*

[2] *Università di Padova, Padova, Italy, fabrizio.nestola@unipd.it*

[3] *Universidade Federal de Ouro Preto, Ouro Preto, Brazil, ricardo.scholz@ufop.edu.br*

[4] *Universidade de Brasília, Brasília, Brazil, jalowitzki@unb.br, reinhardt@unb.br*

Introduction

Diamond is a metasomatic mineral that grows from the interaction between carbon-bearing fluids and mantle rocks. The nature of the diamond substrates is revealed by minerals captured during diamond crystallization, which also permits indirect constraints to be placed on the composition of the metasomatic agents. Direct samples of diamond-forming fluids occur as crystallized-fluid micro-inclusions in fibrous diamonds or, less commonly, in monocrystalline diamonds (for a recent review see Weiss et al. 2022).

An alluvial, gem-quality diamond from Brazil allowed the direct observation of an unusual process: the escape of trapped fluid (**Fig. 1a**). This diamond carried an inclusion assemblage with geochemical signatures inherited from serpentinite protoliths, including hydrous Mg-silicates coexisting with highly forsteritic (Mg# 97) olivine that has a non-mantle-like oxygen isotope composition ($\delta^{18}O$ of +6.2‰) (Carvalho et al. 2024). This assemblage is evidence of partially deserpentinized (dehydrated) peridotitic diamond substrates in subcratonic lithospheric mantle, and provides insights into the stability of hydrous phases and water storage in Earth's upper mantle.

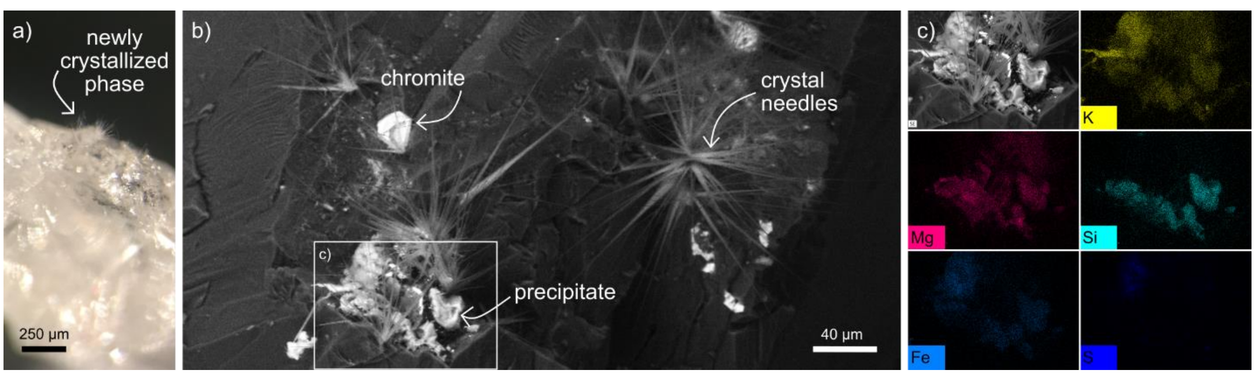

Figure 1: a) Optical image of the studied diamond showing a fibrous material on its surface. **b)** Secondary electron image of the surface of the same diamond, showing an exposed mineral inclusion (chromite) and radial aggregates that crystallized after fracturing the diamond (acicular crystals). An additional precipitate formed at the base of some aggregates (inset c). **c)** Energy dispersive X-ray spectroscopy (EDS) maps, showing the presence of K and Mg in the acicular crystals, in addition to Si, Fe and S in the precipitate.

What is that fibrous material?

Over the months following diamond breakage for inclusion release, acicular (needle-shaped) crystals grew from the surface of some diamond fragments. Scanning electron microscope (SEM) images revealed that the needles grew as radial aggregates out of cleavage planes and along inclusion/diamond interfaces (**Fig. 1b**). Energy-dispersive X-ray spectrometry (EDS) and Raman analyses showed that the newly formed phase is a hydrated K-Mg carbonate (**Fig. 1c**; Raman peaks for carbonate and water were recognized at ~1095 and 2900-3000 cm^{-1}, respectively). At the base of the radial aggregates, an additional solid containing K, Mg, Si, Fe, S, with minor Ba and Cl, precipitated (**Fig. 1c**).

X-ray micro-computed tomography (μCT) analysis was used to image the fluids still trapped within the diamond (**Fig. 2**). The fluid inclusions, recognized by contrasting grey values, occur both in isolation (**Fig. 2a**) and at the edges of mineral inclusions (**Fig. 2b**). The estimated volume of the largest connected fluid inclusions is 980 μm^3 (**Fig. 2c**).

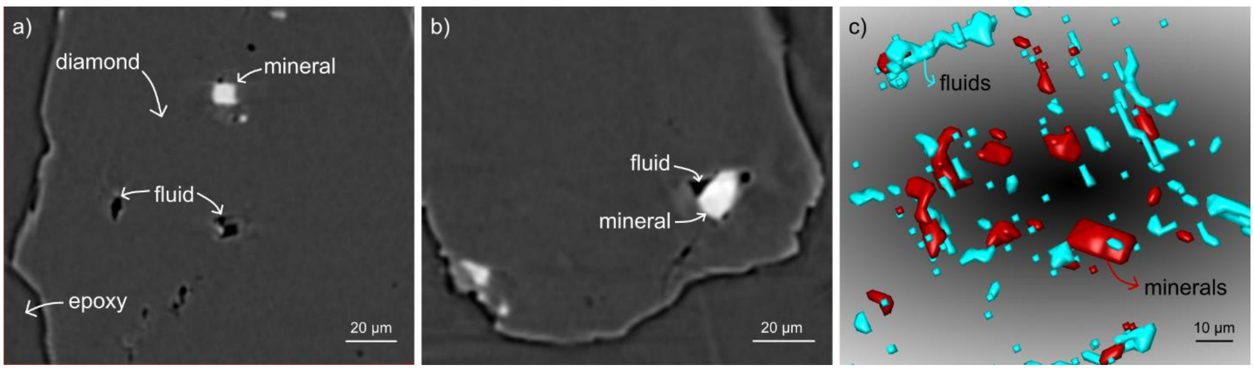

Figure 2: In **(a)** and **(b)**, 2D μCT slice of diamond fragments mounted in epoxy. Diamond and epoxy, as indicated in (a), present similar grey values, mineral inclusions are white, and less-dense material/fluids appear black. **c)** Segmented 3D image showing two distinct categories of inclusions in the studied diamond; pseudo-color is used to distinguish fluid inclusions (blue) and mineral inclusions (red). The voxel resolution for these images is ~0.38 μm^3.

The diamond forming fluids

Our current understanding, while still working on the identification of mineral and fluid phases trapped inside the diamond, is that the newly crystallized phases reflect the composition of high-density fluids (HDFs) captured during diamond growth. The high Mg and K contents and the carbonate nature of the crystallized phase indicate a composition closely related to high-Mg carbonatitic HDFs, similar to those found in fibrous diamonds (e.g., Klein-BenDavid et al. 2006; Logvinova et al. 2011). This type of HDF is usually linked to peridotitic sources (Weiss et al. 2011), in agreement with the serpentinized peridotitic affinity of the mineral inclusions recognized in this diamond (Carvalho et al. 2024).

The geochemical signatures of the mineral inclusions in this diamond establish a clear link to crustal alteration (Carvalho et al. 2024), documenting a long-inferred important role of serpentinite-derived fluids in diamond formation (Aulbach et al. 2011; Regier et al. 2023; Stachel et al. 2022). C and N isotope composition ($\delta^{13}C$ = –4.5‰; $\delta^{15}N$ = –1.2‰) of this diamond, however, indicates that carbon and nitrogen are chiefly mantle-derived. This mixed (crustal / mantle) signature indicates the interaction between a hydrothermally-altered section of subducted oceanic lithosphere and mantle-derived fluids infiltrating after incorporation into the cratonic mantle root.

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