



## Diamond growth in mantle fluids



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### ABSTRACT

In the upper mantle, diamonds can potentially grow from various forms of media (solid, gas, fluid) with a range of compositions (e.g. graphite, C–O–H fluids, silicate or carbonate melts). Inclusions trapped in diamonds are one of the few diagnostic tools that can constrain diamond growth conditions in the Earth's mantle. In this study, inclusion-bearing diamonds have been synthesized to understand the growth conditions of natural diamonds in the upper mantle. Diamonds containing syngenetic inclusions were synthesized in multi-anvil presses employing starting mixtures of carbonates, and silicate compositions in the presence of pure water and saline fluids (H<sub>2</sub>O–NaCl). Experiments were performed at conditions compatible with the Earth's geotherm (7 GPa, 1300–1400 °C). Results show that within the timescale of the experiments (6 to 30 h) diamond growth occurs if water and carbonates are present in the fluid phase. Water promotes faster diamond growth (up to 14 mm/year at 1400 °C, 7 GPa, 10 g/l NaCl), which is favorable to the inclusion trapping process. At 7 GPa, temperature and fluid composition are the main factors controlling diamond growth. In these experiments, diamonds grew in the presence of two fluids: an aqueous fluid and a hydrous silicate melt. The carbon source for diamond growth must be carbonate (CO<sub>3</sub><sup>2-</sup>) dissolved in the melt or carbon dioxide species in the aqueous fluid (CO<sub>2(aq)</sub>). The presence of NaCl affects the growth kinetics but is not a prerequisite for inclusion-bearing diamond formation. The presence of small discrete or isolated volumes of water-rich fluids is necessary to grow inclusion-bearing peridotitic, eclogitic, fibrous, cloudy and coated diamonds, and may also be involved in the growth of ultradeep, ultrahigh-pressure metamorphic diamonds.

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### 1. Introduction

The growth mechanism of natural diamonds in the Earth's mantle is still an open and debated issue in Earth Sciences (e.g. Harte, 2010; Shirey et al., 2013; Stachel and Harris, 2009 and all references therein). Diamond is a metasomatic mineral that formed through redox reactions of mobile C-bearing phases (fluids/melts) that percolate in the mantle over a large range of depths suggesting different kind of carbon sources. The nature and storage modes for carbon at depth are still poorly understood. This is mostly because, unlike to hydrogen, carbon is not significantly incorporated into the major rock-forming minerals of the mantle but instead forms accessory phases. Depending on the depth these accessory phases may evolve from fluids/melts, carbonates, diamond, Fe-rich alloys, or metal carbide. For that reason diamond growth scenarios in the different mantle regions (upper, transition

zone and lower) are still undefined. A systematic investigation of the phases trapped in natural diamonds as inclusions is of primary relevance to constrain diamond genesis as well as to understand carbon storage and cycling in the mantle.

Most natural diamonds are brought up to the Earth's surface through kimberlitic volcanism in ancient cratonic lithosphere, but monocrystalline and sublithospheric diamonds can also be found in alluvial deposits. Due mainly to differences in their inclusions, we distinguish diamonds formed in the lithosphere from those that are formed below the lithosphere in the convecting mantle. The majority of the gem quality monocrystalline diamonds are from the lithosphere. They have eclogitic or peridotitic (the most abundant) associations depending on the mineral assemblage of their inclusions, and come from depths of at least 200 km, based on the phase equilibria of their solid inclusions (Harte, 2010; Stachel and Harris, 2009). Recently, fluid inclusions of high-density saline fluids have been found in very old monocrystalline diamonds (Weiss et al., 2014) possibly implying a morphological continuum between monocrystalline diamonds and fibrous diamonds, another species of lithospheric diamonds. The later are characterized by their

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high density of fluid and mineral inclusions (Klein BenDavid et al., 2010 and references therein). The fluids reflect oxidizing conditions and show a variety of compositions between carbonatitic, silicic and saline end-members. These fluids are the crucial deciphering tools for diamond growth mechanisms as they represent the “medium” in equilibrium during diamond growth (e.g. Klein BenDavid et al., 2010; Navon et al., 1988 and references therein). A final form in which diamonds are found is as polycrystalline assemblages (Heaney et al., 2005) such as framesites (e.g. Kurat and Dobosi, 2000) and carbonados (e.g. Sautter et al., 2011). Carbonados are exclusively found as pebbles within placers and are not known to be associated with kimberlitic rocks, so their origin is still a matter of debate. Moreover carbonados completely lack typical mantle-derived inclusions and may contain assemblages of native minerals and metals (protogenetic inclusions of augite and ilmenite – i.e. seeds for diamond nucleation), pointing to a genesis characterized by highly reducing conditions (FMQ-15 log units, Sautter et al., 2011 and ref. therein). These diamonds are characterized by tiny micrometer-sized crystals found in crustal rocks originally subducted to ultrahigh pressures (e.g. De Corte et al., 2002; Dobrzhinetskaya, 2012 for a review).

Diamonds grow through metasomatic processes involving C–O–H bearing fluids or melts. The nature of these fluids likely varies depending on the particular diamond forming conditions. It has been proposed, for example, that unlike diamonds from the upper mantle that grow within oxidized fluids, ultra-deep diamonds (i.e. from the lower mantle) derive from reduced fluids (Stachel and Harris, 2009). The recent discovery of an OH-bearing ringwoodite inclusion in a diamond from the transition zone (Pearson et al., 2014) implies that transition zone diamonds may grow from hydrogen-bearing melts or fluids. Diamonds therefore allow carbon behavior in the deep Earth to be traced but also provide crucial and unique information concerning the deep cycling of other volatile species. Water and halogens, for example, are commonly found as high density fluids (HDF) trapped as inclusions mainly in fibrous, coated and cloudy diamonds (e.g. Burgess et al., 2002; Izraeli et al., 2001; Johnson et al., 2000; Klein BenDavid et al., 2007; Navon et al., 1988; Pearson et al., 2014; Tomlinson et al., 2006, 2009; Weiss et al., 2009, 2014). Furthermore, whereas most HDFs are found in fibrous and coated diamonds, a recent report of HDF trapped in monocrystalline diamonds from the Phanerozoic carrying a peridotitic signature (Weiss et al., 2014) points to a role of HDF in the growth of these diamond gems.

Indications concerning the parental “fluids” of diamonds and the locus of their growth in the mantle can be obtained by studying the composition of natural inclusions trapped in diamonds (Novella et al., 2015; Pearson et al., 2014), and from the isotopic carbon and nitrogen signatures of diamonds (see the review after Cartigny et al., 2014; Mikhail et al., 2014). The association of diamond growth with subduction zones is frequently proposed because subduction recycles hydrous and relatively oxidized fluids (Stachel et al., 2005). However, this concept is highly debated, based on carbon and nitrogen isotope signatures of natural diamonds (Cartigny et al., 2014). Indeed, most of the natural diamond gems, including fibrous and coated diamonds, exhibit mantle-derived carbon  $\delta^{13}\text{C}$  ranges from  $-10$  to  $-5\%$  whereas sedimentary carbon, recycled at subduction zones, ranges from  $-45$  to  $+4\%$ , with the range for organic matter being  $-45$  to  $-15\%$  and  $-3$  to  $4\%$  for carbonates (Cartigny et al., 2014). Some diamonds exhibit a light signature with respect to carbon suggesting that the recycled carbon participates to the growth of these diamonds (e.g. Bulanova et al., 2014; Mikhail et al., 2013; Thomson et al., 2014; Walter et al., 2011; Zedgenizov et al., 2014). This is supported by the oxygen isotope composition of inclusions trapped in superdeep diamonds (Burnham et al., 2015).

An alternative possibility is that the isotopic variability recorded in diamonds might also reflect a fractionation process occurring during diamond formation (e.g. Cartigny et al., 2014). In summary, diamond formation from C–O–H–N–S fluids (or melts) is quite likely but the

source and mechanism remain poorly understood (Stachel and Luth, 2015 and references therein).

One way to address these issues is through laboratory experiments. Diamond growth experiments at pressures and temperatures relevant to the Earth's interior have been conducted by numerous researchers over the last decades. Pal'Yanov et al. (1999) were among the first to experimentally demonstrate the practicality of diamond-growth from a mixture of carbonates and water. Since this study, additional experimental studies have shown that diamonds can grow in complex fluid mixtures, including silicates and saline fluids (e.g. Arima et al., 1993; Pal'Yanov and Sokol, 2009; Pal'Yanov et al., 2007; Safonov et al., 2007; Sokol and Pal'Yanov, 2008 and references therein; Arima et al., 2010; Bureau et al., 2012; Fagan and Luth, 2010).

Paradoxically, experimental diamond growth has been achieved using numerous fluid mixtures, and this makes the identification of parental fluids of natural diamonds quite difficult. One approach to more tightly constrain the parental fluids of natural diamonds is to reproduce the inclusions found in diamonds during growth experiments. In a previous study Bureau et al. (2012) were successful in growing diamond bearing inclusions from a mixture of silicate, carbonates and water. Depending on experimental conditions (from 7 to 9 GPa, from 1200 to 1700 °C), these inclusions comprised aqueous fluids, silicate glasses or minerals. It was shown that water is a key parameter to grow diamond bearing inclusions in the upper mantle, and that diamonds having trapped inclusions must have experienced fast growth rates. The study has showed that depending on the pressure and temperature conditions, upper mantle diamonds are possibly growing from one single supercritical fluid or from two fluids, aqueous fluid and silicate melt, in equilibrium with each other. This preliminary study was performed with simple starting compositions (pure water, iron-free system) and therefore the inclusions trapped in the new experimentally grown diamonds were not representative in composition of the inclusions observed in natural diamonds because, for example, iron was not present in the system.

In the present work, we have experimentally grown diamonds from fluids containing significant amounts of water (pure and saline), together with silicates and carbonates (synthetic and natural). Mineralogical assemblages similar to those found as inclusions in natural diamonds are obtained as inclusions in experimentally grown diamonds.

## 2. Materials and methods

Synthetic and natural powders of various compositions were employed as starting materials, as described in Table 1. We have used: (1) the same mixture MELD of synthetic powders already used in a previous study (Bureau et al., 2012), which has the average composition of natural inclusions trapped in fibrous diamonds, from the study of

**Table 1**  
Starting materials.

Name	Description	Composition
MELD	Mixture of oxides and carbonates <sup>a</sup>	SiO <sub>2</sub> 40.47; Al <sub>2</sub> O <sub>3</sub> 4.36; MgO 21.07; CaO 9.29; Na <sub>2</sub> O 2.36; K <sub>2</sub> O 17.57; TiO <sub>2</sub> 4.89, about 15 wt.% of CO <sub>2</sub>
SIDB	Natural siderite <sup>b</sup>	(Fe,Mg)CO <sub>3</sub> In at.%, MgO 2.47; FeO 49.11; MnO 7.17; CaO 0.83
SED <sup>c</sup>	Natural pelagic sediment <sup>d</sup> MD85682 Indian ridge	DRX analysis: 97% CaCO <sub>3</sub> + 3% SiO <sub>2</sub>
MORB <sup>c</sup>	Natural mid ocean ridge basalt <sup>e</sup> Indian ridge	DRX analysis: 38.26% CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> 13.25% (Mg <sub>1.41</sub> Fe <sub>0.59</sub> ) SiO <sub>4</sub> 44.72% (Ca <sub>0.8</sub> Mg <sub>1.2</sub> )Si <sub>2</sub> O <sub>6</sub>

<sup>a</sup> After Bureau et al., 2012.

<sup>b</sup> After Boulard et al., 2011.

<sup>c</sup> DRX analysis after E. Foy.

<sup>d</sup> From the *Muséum National d'Histoire Naturelle's* collection.

<sup>e</sup> Courtesy of Damien Jaujard.

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