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Invited review article

Diamond formation — Where, when and how?

T. Stachel *, R.W. Luth

Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, AB, T6G 2E3, Canada

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ABSTRACT

Geothermobarometric calculations for a worldwide database of inclusions in diamond indicate that formation of the dominant harzburgitic diamond association occurred predominantly (90%) under subsolidus conditions. Diamonds in eclogitic and lherzolitic lithologies grew in the presence of a melt, unless their formation is related to strongly reducing CHO fluids that would increase the solidus temperature or occurred at pressure–temperature conditions below about 5 GPa and 1050 °C. Three quarters of peridotitic garnet inclusions in diamond classify as "depleted" due to their low Y and Zr contents but, based on $LREE_N$–$HREE_N$ ratios invariably near or greater than one, they nevertheless reflect re-enrichment through either highly fractionated fluids or small amounts of melt. The trace element signatures of harzburgitic and lherzolitic garnet inclusions are broadly consistent with formation under subsolidus and supersolidus conditions, respectively. Diamond formation may be followed by cooling in the range of ~60–180 °C as a consequence of slow thermal relaxation or, in the case of the Kimberley area in South Africa, possibly uplift due to extension in the lithospheric mantle. In other cases, diamond formation and final residence took place at comparable temperatures or even associated with small temperature increases over time.

Diamond formation in peridotitic substrates can only occur at conditions at least as reducing as the EMOD buffer. Evaluation of the redox state of 225 garnet peridotite xenoliths from cratons worldwide indicates that the vast majority of samples deriving from within the diamond stability field represent fO_2 conditions below EMOD. Modeling reveals that less than 50 ppm fluid are required to completely reset the redox state of depleted cratonic peridotite to that of the fluid. Consequently, the overall reduced state of diamond stable peridotites implies that the last fluids to interact with the deep cratonic lithosphere were generally reducing in character. A further consequence of the extremely limited redox buffering capacity of cratonic peridotites is that redox reactions with infiltrating fluid/melt likely cannot produce large diamonds or high diamond grades. Evaluating the shift in maximum carbon content in CHO fluids during either isobaric cooling or ascent along a cratonic geotherm, however, reveals that isochemical precipitation of carbon from CHO fluids provides an efficient mode of diamond crystallization. Since subsolidus fluids are permissible in harzburgites only, and supersolidus melts in lherzolite we suggest that CHO fluid metasomatism may explain the long observed close association between diamonds and harzburgitic garnets. In the absence of thermodynamic data we cannot evaluate if supersolidus carbonate-bearing melts, stable at fO_2 conditions below EMOD, would experience a similar decrease in maximum carbon solubility during cooling or ascent along a geotherm. The absence of a clear association between diamond and lherzolitic garnets, however, suggests that this is not the case. A very strong association between diamond and eclogite likely relates to the fact that the transition from carbonate to diamond stable conditions occurs at redox conditions that are at least about 1 log unit more oxidizing than EMOD. At this time we cannot quantitatively evaluate the redox buffering capacity of cratonic eclogites but given their much higher Fe contents it has to be significantly higher than for peridotites. Alternatively, diamond in eclogite may precipitate directly from cooling carbonate-bearing melts that may be too oxidizing to crystallize diamond in olivine-bearing lithologies.

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* Corresponding author.
E-mail address: tstachel@ualberta.ca (T. Stachel).

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1. Introduction

Following the discovery of the first kimberlite hosted diamond deposits in South Africa in 1870–1871, diamond formation was initially linked to reaction of the kimberlite magma with abundant carbonaceous shale fragments (crustal xenoliths) present in these diatremes (Lewis, 1887). The subsequent proposal of diamond representing a high-pressure phenocryst in kimberlite was widely accepted (e.g., Williams, 1932) till the advent of geochemical studies of inclusions in diamond (Meyer, 1968; Meyer and Boyd, 1972; Sobolev et al., 1969) and radiometric dating of diamond formation ages (Kramers, 1979; Richardson et al., 1984), both implying crystallization in Earth's mantle unrelated to host kimberlite magmatism. The seminal suggestion of a xenocrystic origin for diamond in kimberlite (based on the observation of diamondiferous eclogite xenoliths), however, already dates back to Bonney (1899). Since the 1970s, numerous studies covering all important deposits around the globe have provided a detailed picture on the mineralogical and chemical environment for diamond formation, including the attendant pressure–temperature conditions (reviewed in Gurney, 1989; Meyer, 1987; Stachel and Harris, 2008). Following the recognition of diamond as mantle-derived xenocrysts in kimberlite, the concept emerged that diamond forms via redox reactions (Eggler and Baker, 1982; Luth, 1993; Rosenhauer et al., 1977) that relate to migration of a fluid or melt through a mantle host rock (Haggerty, 1986) and in consequence, diamond is now generally viewed as a metasomatic mineral (e.g., Stachel and Harris, 1997; Taylor et al., 1998). Little, however, is known about the exact composition and the redox character (carbonate- versus methane-bearing) of the fluids or melts that precipitate smooth-surfaced monocrystalline diamonds.

In this contribution we use the large body of published data on diamonds and their mineral inclusions and less plentiful Fe^{3+}/Fe^{2+} determinations on minerals in cratonic garnet peridotites to discuss the “where, when and how?” of diamond formation and to place constraints on possible modes of diamond precipitation that invalidate some popular models.

2. Diamond substrates in Earth's mantle

The mineralogy and the mineral compositions of diamond host rocks in Earth's mantle are very well characterized through studies on mineral inclusions in diamond (reviewed in Gurney, 1989; Meyer, 1987; Meyer and Boyd, 1972; Shirey et al., 2013; Stachel and Harris, 2008). These studies have shown that diamonds derive from subcontinental lithospheric mantle extending into the diamond stability field (e.g., Boyd and Gurney, 1986) or may have originated at an even greater depth (extending to at least 700 km) in the sublithospheric mantle (Harte and Harris, 1994; Moore and Gurney, 1985; Scott Smith et al., 1984). Sublithospheric diamonds are, however, rare and the subcratonic lithospheric mantle represents the primary source of over 99% (by mass) of the worldwide diamond production (Stachel and Harris, 2008). In addition, the sublithospheric diamonds studied so far relate to the recycling of oceanic lithosphere into the deep mantle (Harte et al., 1999b; Stachel et al., 2000a, 2000b; Tappert et al., 2005; Walter et al., 2011) and hence can provide only very limited insights into diamond formation and storage in pyrolitic upper and lower mantle. For this contribution we will, therefore, focus exclusively on lithospheric diamonds.

Based on their mineral inclusion content, diamonds from the lithospheric mantle ($N = 2837$) are divided into peridotitic (65%), eclogitic (33%) and websteritic (pyroxenitic) suites (2%). Using garnet compositions ($N = 685$), the peridotitic inclusion suite can be subdivided into harzburgitic (56% of all diamonds), lherzolitic (8%) and wehrlitic (0.7%) parageneses. The 86:13:1 harzburgite:lherzolite:wehrlite split of the peridotitic suite is virtually unchanged from the original pioneering work of Gurney (1984). The 2:1 ratio of peridotitic:eclogitic suite diamonds is based on destructive studies on diamonds generally <3 mm in size; it has, however, been speculated that among larger diamonds the relative proportion of the eclogitic suite may increase (e.g., Gurney, 1989; Stachel and Harris, 2008). In any case, 33% of all diamonds hosted in eclogite far exceed the <1 to 5% estimated volumetric abundance of eclogite in subcratonic lithospheric mantle (Dawson and Stephens, 1975; McLean et al., 2007; Schulze, 1989). Equally, the ratio of harzburgitic:lherzolitic paragenesis diamonds (~7:1) reverses the relative proportions of harzburgite to lherzolite in diamond stable lithospheric mantle (ca. 1:4 for the Western Kaapvaal Craton; Griffin et al., 2003). This suggests that compared to lherzolite, harzburgite and eclogite are strongly preferred substrates for diamond (Grütter et al., 2004; Gurney, 1984).

3. Pressure–temperature conditions of diamond formation

3.1. Peridotitic suite

Diamond represents a closed system, with even the mobility of hydrogen being very low (Connell et al., 1998; Saguy, 2004). Non-touching