

Diamond origin and genesis: A C and N stable isotope study on diamonds from a single eclogitic xenolith (Kaalvallei, South Africa)

Mederic Palot^{a,*}, Pierre Cartigny^a, Fanus Viljoen^{b,c}

^a Laboratoire de Géochimie des Isotopes Stables de l'Institut de Physique du Globe de Paris, Université Paris 7, 2 Place Jussieu, T54-64, 75251 Paris cedex 05, France

^b Group Exploration Macrodiamond Laboratory, De Beers Consolidated Mines Ltd., Southdale, 2135, South Africa

^c Now at: DST Research Chair in Geometallurgy, Department of Geology, University of Johannesburg, PO Box 524, Auckland Park 2006, South Africa

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ABSTRACT

In order to study the geochemistry of diamonds formed during similar growth conditions in a very localised environment in the mantle, we carried out a detailed study of the variation in $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, N content, and N aggregation state of 35 diamonds in a single eclogite xenolith from the Kaalvallei kimberlite in South Africa. Diamond nitrogen contents determined by infrared spectroscopy range from 239 ppm to 1272 ppm, and are positively correlated with nitrogen aggregation states which vary from 11.5% to 43.7% of IaB defects. Modelling of these parameters using second order reaction kinetics suggests that the diamonds likely represent a single population which has been resident in the mantle at temperatures of 1090 °C to 1190 °C. $\delta^{13}\text{C}$ values of the diamonds analysed range from -6.0% to -4.2% , while $\delta^{15}\text{N}$ values vary from -8.9% to -4.1% , with no correlation between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. These values account for 5% and 15%, respectively, of the worldwide isotopic range for diamonds. The limited variability of C and N isotopic compositions for the diamonds analysed are compatible with a model of metasomatic diamond formation from a single, homogeneous fluid, and this is also supported by the infrared data. Although the absence of any clear correlation between $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ and diamond N content precludes the accurate identification of the fluid species involved in diamond growth, the lack of correlation may indicate the involvement of a carbonate- or CO_2 -type fluid.

The observed range in negative $\delta^{15}\text{N}$ values for all the diamonds analysed are within the limits of the so-called mantle range ($\delta^{15}\text{N}_{\text{mantle}} = -5 \pm 2\%$) which is consistent with a mantle origin for these diamonds. Negative $\delta^{15}\text{N}$ is inconsistent with diamond formation from recycled (crustal) material which is enriched in ^{15}N (i.e. positive $\delta^{15}\text{N}$).

In contrast, positive europium and strontium anomalies in silicate minerals of the host xenolith, as well as oxygen isotopic data which deviate from the mantle range, are consistent with a protolith consisting of recycled oceanic crust. It is therefore concluded that our data supports a model of metasomatic diamond crystallisation from a mantle-carbon source.

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1. Introduction

The high strength and inert nature of diamond makes it an ideal substance on which to study the interior of the earth from e.g. the base of the lithosphere (150–250 km) to the transition zone, and also in to the lower mantle (>660 km; Stachel et al., 2000a,b). Thus, diamond and its impurities can provide insight into aspects of mantle mineralogy as well as the origin of the lithosphere and its evolution. (Deines et al., this issue; Kaminsky et al., this issue; Batumike et al., this issue; Sobolev et al., this issue). Based on the mineralogy and major element chemistry of inclusions in diamonds, it is by now well established that diamond crystallises primarily in two differing lithologies, namely peridotite and eclogite (Sobolev, 1977; Stachel

and Harris, 2008; references therein). Over the last 30 years, these studies have also helped constrain our understanding of e.g. the temporal evolution of the lithospheric mantle (Shirey et al., 2002). Such studies, however, face a series of difficulties. Diamonds are usually recovered as mixed populations from the host kimberlite, and may represent a variety of high-pressure host rocks (as attested by e.g. the variability in the Cr and Ca content of peridotitic garnets occurring as inclusions in diamonds), derived from distinct temperature regimes in the mantle, which may correspond to varying diamond mantle residence times. The presence of such mixed populations might explain why typically only weak correlations between e.g. $\delta^{13}\text{C}$ and diamond N contents are observed.

In order to constrain models of diamond genesis more accurately it is necessary to focus on diamond populations characterised by similar growth conditions e.g. those from diamond-bearing eclogite and peridotite xenoliths (see for instance Thomassot et al., 2007). Eclogite represents a comparatively minor component of the earth's mantle

* Corresponding author Tel.: +33 (0)1 44 27 60 88.

E-mail addresses: palot@ipgp.jussieu.fr (M. Palot), cartigny@ipgp.jussieu.fr (P. Cartigny), fanusv@uj.ac.za (F. Viljoen).

(Schulze et al., 1996), yet may account for a significant proportion of the xenolith and diamond population at many kimberlites worldwide. Diamond-bearing eclogite also tends to predominate amongst diamondiferous xenoliths (Gurney, 1989). This over-abundance of diamond in eclogite was explained by Luth (1993) as being a consequence of two combined effects: (1) the relatively more reduced nature of eclogite compared to peridotite (O'Neill et al., 1993) would result in enhanced stability of diamond in eclogite (Luth, 1993), and (2) olivine-bearing mantle xenoliths such as peridotites are probably more easily altered and disaggregated relative to eclogite (Luth, 1993).

Diamond-bearing eclogites are unusually abundant at some of the kimberlites in South Africa e.g. the Kaalvallei kimberlite. Eclogites at the locality have also been well-studied (Viljoen, 2005). The current investigation therefore focuses on 35 diamonds extracted from a single eclogite (sample K8/109) from Kaalvallei, and serves to complement an earlier study on diamonds from a diamondiferous peridotite recovered at the Premier diamond mine (Thomassot et al., 2007). It also aims to provide insights into the other major high-pressure diamond host, namely eclogite. Observed relationships between carbon and nitrogen isotopic composition, and diamond growth, place constraints on (a) the origin of eclogitic diamonds and their relationship with their eclogitic host, (b) diamond genesis in relation to metasomatic processes, and (c) the implications thereof for models of diamond growth in the subcratonic lithosphere.

2. Xenoliths and diamonds at Kaalvallei

The Kaalvallei kimberlite is located in the Free State region of South Africa, on the farm Kaalvallei12, approximately 7.5 km from the town of Welkom along the Welkom–Virginia road (Fig. 1 in Viljoen, 1994; Viljoen, 2005). It occurs on the Kaapvaal craton, and intrudes shales and sandstones of the Ecca Group. The occurrence comprises of a roughly circular main pipe with surface area of 2 ha, and a satellite intrusion located 300 m west of the main pipe (Stiefenhofer, 1989). Monticellite-bearing kimberlite varieties predominate and the main pipe is classified petrographically and isotopically as a Group I kimberlite (Smith, 1983; Skinner, 1989), with an Rb–Sr mica age of 85 ± 1 Ma (Viljoen, 1994).

The diamond-bearing xenolith under consideration, K8/109, was previously described by Viljoen (2005). It is biminerally, comprising mainly of clinopyroxene (70%) and garnet (30%), with trace diamond (<1%). Based on texture, it is a Group I eclogite (i.e. coarse rocks with

large rounded garnets, set in a matrix of coarse clinopyroxene; MacGregor and Carter, 1970). As is typical for Group I eclogite xenoliths (including many diamond-bearing eclogites), elevated Na₂O in garnet as well as K₂O in clinopyroxene is observed (McCandless and Gurney, 1989; Viljoen, 2005). Garnet in the xenolith is comparatively Fe-rich (15.32 wt.% FeO), contains substantial TiO₂ (0.44 wt.%) but has low Cr₂O₃ (0.20 wt.%; Viljoen, 2005). The occurrence of diamond in a Group I eclogite from Kaalvallei is consistent with the view that these represent a major high-pressure source rock for diamond (Robinson et al., 1984; McCandless and Gurney, 1989).

Most of the diamonds in Kaalvallei eclogite xenolith K8/109 are colourless with unresorbed octahedral shapes, and with weights ranging from 0.12 to 1.65 mg (0.0006 to 0.0082 carats). The occurrence of small, unresorbed octahedral diamonds in the xenolith is also typical of diamonds from other eclogites at Kaalvallei (Kiviets, 2000), as well as for eclogites from other localities worldwide e.g. the Orapa kimberlite in Botswana (Robinson et al., 1984). No inclusions were encountered in any of the diamonds examined.

3. Analytical techniques

Diamonds were analysed by micro-Fourier transform infrared (FTIR) spectroscopy using a Nicolet Magna-IR 550 to determine both nitrogen content (N_{FTIR}) and nitrogen aggregation state. Spectrum acquisition was accomplished over a spectral range of 650 cm^{-1} to 4000 cm^{-1} , with 300 scans collected at a resolution of 4 cm^{-1} . Nitrogen spectra were deconvoluted utilising a least squares approach, and nitrogen contents and aggregation states determined using an absorption coefficient of $16.5 \text{ at ppm cm}^{-1}$ for A defects (Boyd et al., 1994a) and $79.4 \text{ at ppm cm}^{-1}$ for B defects (Boyd et al., 1995a). Accuracies are better than 10% for nitrogen content and better than $\pm 3\%$ for nitrogen aggregation state.

Mass-spectrometer-based analysis of diamonds for nitrogen content, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ followed the experimental procedure of Boyd et al. (1995b) with accuracies of 0.5‰ and 0.1‰ (2σ) for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, as well as 5% for total nitrogen measurements. Each diamond was combusted in a pure oxygen atmosphere at a temperature of $1100 \text{ }^\circ\text{C}$, and nitrogen was then separated from carbon dioxide using a mixture of CaO and Cu, with nitrogen oxides reduced to N_2 (Boyd et al., 1994b). CO_2 generated was analysed for carbon isotopic composition with a dual-inlet gas source mass spectrometer. Nitrogen concentrations in the gas produced by burning the diamond were measured with a

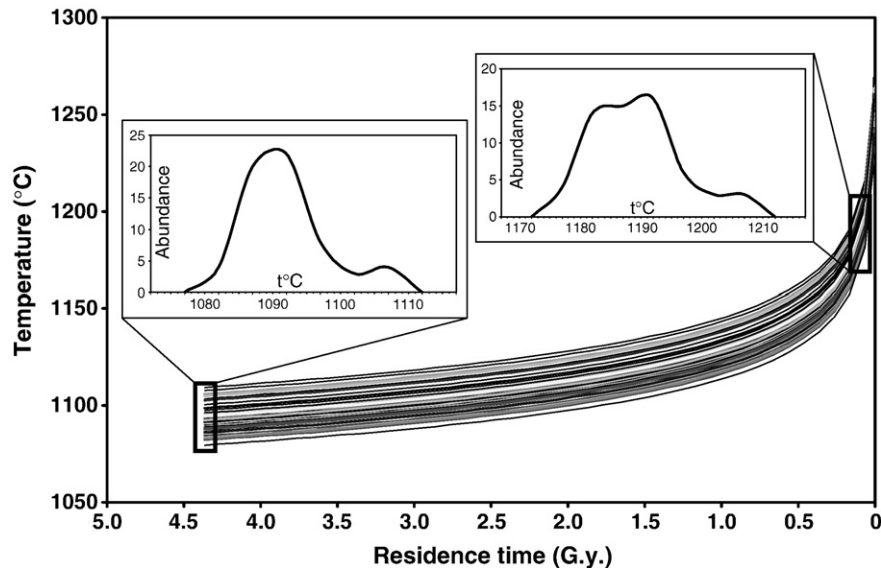


Fig. 1. Diamond nitrogen aggregation plotted as a function of average mantle residence time, and calculated utilising second order reaction kinetics (Chrenko et al., 1977). Temperature distributions have been calculated for assumed mantle residence times of 4365 M.y. and 80 M.y. Note that temperature is a little dependent on time over a wide spread of residence time but are sensitive for very small residence time in the mantle. Each solid line represents a single FTIR measurement on one diamond.

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