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Metasomatic diamond-forming events in the Limpopo Belt deep lithosphere recorded by HDFs in Venitia diamonds

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Introduction

Venetia diamonds sample the lithosphere underlying the Central Zone of the Limpopo Belt, at the junction of the Kaapvaal and Zimbabwe Cratons, and record its unique history of metasomatism (Viljoen et al., 2002; Koornneef et al., 2017). High-density fluids (HDFs) trapped in microinclusions in fibrous diamonds allow direct examination of such metasomatic fluids. The kinetics of nitrogen aggregation in the host diamonds allow the distinction between consecutive metasomatic events and their relation to large-scale magmatic and tectonic events (Weiss et al., 2022).

Results

The majority of diamonds analyzed in the present study (25 of 27) contain high-Mg carbonatitic HDFs, one diamond is silicic and one is saline (Fig. 1). Of those, sixteen diamonds (15 carbonatitic and one saline) carry all their nitrogen in A-centers, five diamonds carry 10-30% B-centers and four diamonds have ~60-80% of their nitrogen in B-centers (Fig. 2). The silicic diamond exhibits ~100% B-centers.

The carbonatitic HDFs vary in composition and form two groups. Both fall within the worldwide high-Mg carbonatitic array (Weiss et al., 2022). While the Mg concentration of the two groups overlaps, they are distinguished by their Si, Ca, Na, K and Cl (Fig. 1). Nineteen diamonds form a tight cloud with average composition: 10 ± 2 SiO₂, 10.3 ± 0.9 FeO, 23.6 ± 1.7 MgO, 26 ± 3.4 CaO, 8.4 ± 2.2 Na₂O, 13.5 ± 4.6 K₂O and 1.5 ± 0.6 Cl. The other six span an array in several variation diagrams. SiO₂, FeO, CaO and Na₂O show a negative correlation with K₂O, while Al₂O₃ and Cl correlate positively with K₂O. The chemical variation of the HDF is independent of the nitrogen aggregation. Many of the diamonds in all three populations contain micro-mineral inclusions (e.g. olivine, orthopyroxene, and clinopyroxene)

Discussion

The predominant abundance of high-Mg carbonatitic HDFs in Venetia diamonds (92%) is unique and different from diamonds carried by kimberlites in the western and central Kaapvaal craton, which are controlled by saline or silicic to low-Mg carbonatitic compositions. Previous studies show that diamonds with different aggregation levels are associated with different HDF types, attributed to separate

metasomatic events (Weiss et al., 2018a,b; Kempe et al., 2021). The present results are unique in that a large range of aggregation is found within a single HDF-type (100% A-centers – 80% B-centers).



Figure 1: Oxide variation diagrams (in wt.%) of individual HDF microinclusions in diamonds from Venetia. **a**: MgO *vs.* SiO₂ shows saline (green symbols), silicic (red symbols) and high-Mg carbonatite (blue-purple) HDF types. **b**, **c**, **d**: CaO, FeO and Cl *vs.* K₂O exemplify the chemical variation within the high-Mg carbonatitic HDF.

The kinetics of nitrogen aggregation provide a framework for constraining the time-temperature relations of Venetia diamonds. The Venetia geotherm that was determined based on xenoliths and mineral inclusions in diamonds allows the translation of temperatures to a depth of origin. The observed chemical variation within the high-Mg carbonatitic HDF does not fit the variation in nitrogen aggregation level and a range of B/(B+A) ratio is present in both groups. Therefore, the compositional variation cannot be explained by separate metasomatic events at different times.

We propose that the range of aggregation reflects heating of the carbonatitic HDF-bearing diamonds during a local thermal event. Van der Meer et al. (2013) studied xenoliths from Venetia and proposed a localized heating event involving the introduction of asthenospheric material to the lower part of the lithosphere prior to kimberlite eruption. They estimated that temperatures may have risen by ~200 °C. If the high-Mg carbonatitic diamonds were formed before this event, their nitrogen may have aggregated over a temperature gradient away from the penetrating hot body. Most diamonds were not affected by the heating and did not evolve beyond A-centers, but some experienced higher temperatures and aggregated to form the variety of B-center levels observed, depending on their proximity to the thermal source.

The silicic diamond with its high levels of nitrogen aggregation must be old and deep, as time alone does not allow reaching such aggregation level at near solidus temperatures at shallow depths. The saline diamond may have formed during a separate, late event or together with the carbonatitic diamonds.



Figure 2: Nitrogen aggregation from A-centers to B-centers as a function of time (t) and temperature (T) for fibrous diamonds from Venetia (this study). (a) Total nitrogen *vs.* the percentage of nitrogen in B-centres. Isotherms are calculated assuming a residence time of 500 Myr. The high-Mg carbonatitic diamonds show a wide range of nitrogen aggregation. (b) Time *vs.* temperature plot. Each line presents all possible t-T combinations for the content and aggregation state of nitrogen in a single diamond. The data lines in (b) are coded by color as in Fig. 1. Black dashed lines mark tectonic and diamond-forming events in the Limpopo Belt (Richardson et al., 2000; Koorneef et al., 2017).

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