



Carbon and nitrogen isotope distribution in HTHP synthetic diamonds

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Variation of stable isotope ratios within natural diamond crystals is used to track growth events such as changes in source compositions, fractionation effects or kinetic factors. However, the interpretation of carbon and nitrogen compositions in natural crystals is hampered by insufficient evidence of growth behaviour under known conditions. In the present work we combine experimental growth of crystals under carefully monitored conditions with subsequent detailed analysis of the synthetic crystals by SIMS. Two large crystals with different nitrogen contents were grown at $P \sim 5.5$ GPa and $T \sim 1450^\circ\text{C}$ using a split-sphere multi-anvil apparatus at the UIGGM (Russia). The crystals were cut parallel to $\{110\}$ and were analysed using high spatial resolution ion microprobe techniques (SIMS) of NERC-EMMAC facility in Edinburgh University (UK).

Octahedral sectors of the crystals are shown enrichment in nitrogen content (with factor of 3) and depletion in heavy nitrogen isotope (for about 30 permil) in comparison with cubic sectors. However, despite an order of magnitude contrast in nitrogen abundance for large octahedral (255 ppm) and very small trapezohedral (17 ppm) sectors no difference in isotope ratio was found. A general tendency of decreasing nitrogen abundance from the seed region towards the edges was also observed in both crystals. The maximum carbon isotope fluctuations within individual crystals amount to 1.5 and 3.4 permil. The distribution of $\delta^{13}\text{C}$ values does not show close relationship with the sector structure but pronounced exponential trends in direction of growth was observed.

On the assumption that the high-pressure cell is a closed system for bulk and isotope

compositions, the distribution of $\delta^{13}\text{C}$ values along the direction of growth can be the result of isotope interaction between the growing crystal and other carbon-bearing phases in the cell. In this case the maximum difference obtained within a crystal is equal to true isotope partitioning between growing crystal and the Fe-Ni melt medium. The variations in $\delta^{13}\text{C}$ in the direction of crystal growth stem from the accommodation of the system to the isotope partitioning effect; after a while variation ceases when a steady state condition is reached between the C source, the transport medium and the growing crystal.

The lack of pronounced correlations of $\delta^{15}\text{N}$ with other crystal characteristics can not be declared finally because of relatively big data uncertainty. Fluctuation of nitrogen content within synthetic diamond crystals has been widely observed before but the good correlation with carbon isotope ratio in the present study indicates the possibility of a closer relation of carbon isotope and nitrogen behavior than often proposed. The isotope effect calculated from the range of carbon isotope composition (3.5 permil) in the diamonds is consistent with the scale of $\delta^{13}\text{C}$ fluctuations around main maximum (-5 ± 3 permil in many peridotitic natural diamonds. Three permil in average is also documented for internal carbon isotopic trend from core to rim in “coated” diamonds. Further detailed study of other model systems for HTHP diamond crystallization using the high spatial resolution SIMS technique will allow further understanding of the isotope records in diamond crystals and the processes responsible for their origin.

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