

THE FRACTIONATION OF NITROGEN AND CARBON STABLE ISOTOPE RATIOS IN WESTERN AUSTRALIAN DIAMONDS

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Diamonds from the Argyle and Ellendale lamproites in Western Australia have a wide range in stable isotopic composition (See van Heerden *et al.*, this volume). Carbon stable isotope ratios can vary between $-22.1\text{‰} \leq \delta^{13}\text{C} \leq 0.0\text{‰}$, and the $\delta^{15}\text{N}$ values of these diamonds are between -22.1‰ and $+13.4\text{‰}$. Nitrogen content and state of aggregation are also highly variable in Western Australian diamonds. Examining the zonation patterns evident within single diamond crystals can elucidate the cause of some of the variability within these Western Australian diamonds.

Four diamonds that show simple, concentric growth layering in cathodoluminescence have been examined in detail. The nitrogen isotope composition, nitrogen content and state of aggregation as well as the carbon isotopic composition have been measured in traverses across plates cut from these 1 carat (5mm) diamonds. These parameters all vary regularly with position in the diamond crystal (Figure 1) and the general zonation pattern is one in which the core regions of the diamonds are characterised by having elevated concentrations of isotopically heavy (^{15}N -enriched) nitrogen that occurs in an advanced stage of aggregation. The outer reaches of these diamonds are marked by having lower nitrogen contents, by being relatively ^{15}N -depleted (that is isotopically "lighter") and by having nitrogen in a less aggregated form than the central regions of the stones. The carbon isotopic variations are much more subtle, however the general zonation pattern is also one in which the central regions of the diamond are characterised by an increase in the proportion of the heavier ^{13}C isotope.

The zoning profiles across these simple diamonds are consistent with Rayleigh fractional crystallization in which the heavier ^{15}N and ^{13}C isotopes are preferentially incorporated in the crystallizing diamond. This process has been quantitatively modelled, and fractionation factors have been determined. These are 1.001 for carbon and 1.004 for nitrogen fractionation during diamond precipitation. The initial carbon and nitrogen isotope compositions of the diamond source have also been modelled and these vary from $\delta^{13}\text{C} = -5.6\text{‰}$ to -10.3‰ and from $\delta^{15}\text{N} = +5\text{‰}$ to $+10\text{‰}$.

The fact that carbon and nitrogen stable isotope ratios and nitrogen concentration and aggregation state vary regularly across these diamond plates suggests that discontinuous, multi-phase growth is not responsible for the zonation patterns seen in these 3 Argyle and 1 Ellendale 9 diamond. There are, however, alternative explanations for these variations. Three component mixing, in which the 3 components have (i) positive $\delta^{15}\text{N}$ values and very negative $\delta^{13}\text{C}$ values; (ii) positive $\delta^{15}\text{N}$ values and "mantle" $\delta^{13}\text{C}$ values of about -5‰ and (iii) negative $\delta^{15}\text{N}$ values and "mantle" $\delta^{13}\text{C}$ values respectively, may explain the isotopic characteristics of these diamonds. These 3 components resemble Argyle eclogitic, Argyle lherzolitic and Ellendale 4 diamonds respectively. It is suggested that these three components may be representative of eclogitic, lherzolitic and harzburgitic diamonds in general. This hypothesis however needs to be tested by further analysis of inclusion bearing diamonds from as many different sources as possible.

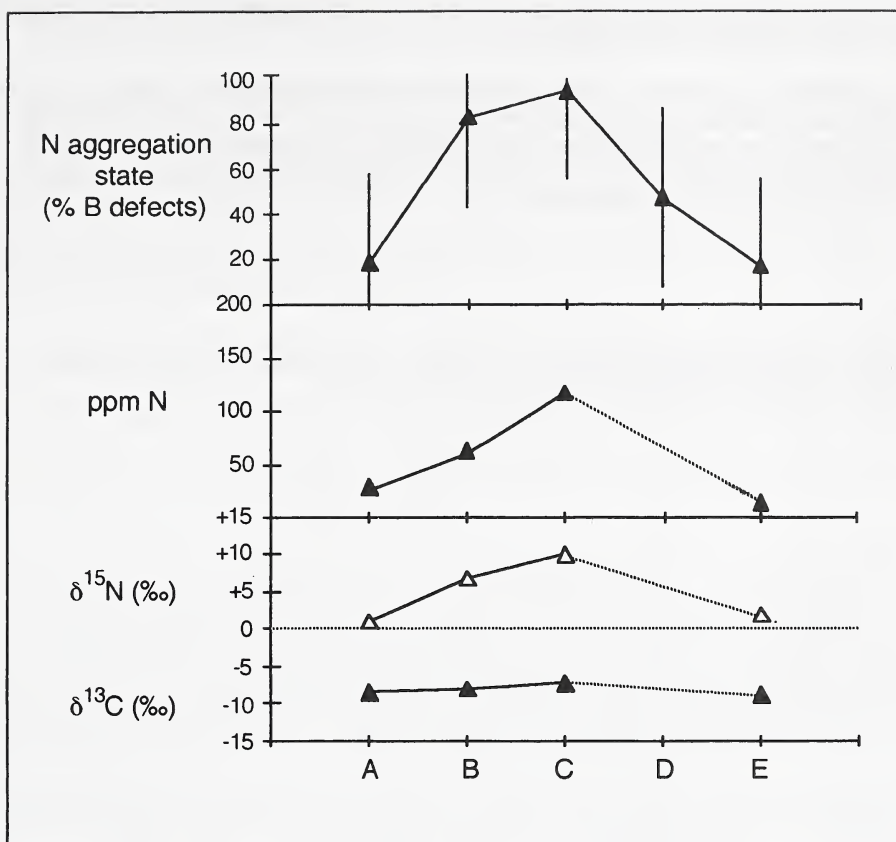


Figure 1: Representative zonation patterns across Argyle diamond plate 150701 #12-1. A to E represents and edge to edge traverse across the 5 mm diamond plate. Cathodoluminescence shows that block C is the centre of the diamond. Error bars are shown for estimates of the proportion of nitrogen occurring in B aggregates. Other uncertainties are smaller than the plot symbols.

The evidence that these diamonds grew in a fractional process has important implications. It supports the idea that diamonds grew in, or from, a volatile-rich fluid precursor, rather than as a result of sub-solidus crystallization. The high degrees of crystallization indicated place some constraints on the size of these “volatile bubbles”. If 100% crystallization is required to form a 5 mm diamond, then the initial reservoir must be of the same magnitude. Furthermore, the variability in the initial carbon and nitrogen stable isotope ratios indicates an initially heterogeneous

mantle. This shows that simple closed system fractional crystallization alone cannot explain all the stable isotope variability seen in diamonds.

Nitrogen aggregation is a function of temperature, time and N concentration, and the zonation in nitrogen aggregation state across these diamonds has implications for the ages of these diamonds. In the most extreme case, an age difference of 1.2 Ga exists between the core region and the outer rim of a diamond from Argyle. With crystal growth occurring on this time scale, a complex history for diamond genesis is to be expected.

References:

van Heerden L.A, Boyd S.R and Pillinger C.T (1995). The carbon and nitrogen isotope characteristics of Argyle and Ellendale diamonds. This volume.