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Constraints on the Origin of Carbonado from C and N Isotopes in Diamond Matrix and Studies of Inclusion Minerals

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Origin of carbonado, a porous aggregate of micro-meter size diamonds, remains an enigma. Their low $\delta^{13}C$ values (<-25‰) and mineral inclusions of crustal origin first suggested their formation within the crust. The most puzzling was crystalline diamonds as large as a few tens of micro-meter in the crust and tightly bond micro-grains without any glues in-between. Possible candidates for the crystallization process are shock syntheses by meteoric impact (Smith and Dawson, 1985), irradiation by α-particles of carbonaceous source materials from U and Th decay within the crust (Kaminsky, 1987: Ozima and Tatsumoto, 1997), and crystallization in the mantle which then is aggregated subsequently in the crust (Kagi et al, 1994). Newly developed hydrothermal method of diamond synthesis added another clue to aggregate micro-grains. However, none of the above explanations are yet generally accepted. Herein we studied four carbonados in conglomerate from Diamantina region (Minas Gerais, Brazil) through (1) Carbon and nitrogen isotopes in the diamond matrix, (2) Inclusion mineralogy and (3) Pb-Pb and REE analyses of the mineral inclusions in order to shed light on their formation processes and source materials. Typical nitrogen and carbon isotope compositions of gases released by bulk combustion of carbonados at 1000° were 0‰ (Air) and -27.5‰ (PDB) respectively. Carbon and nitrogen isotopes were homogeneous between grains compared with common terrestrial diamonds, suggesting carbonados' occurrence, at least in this region, as a single event from a relatively homogenized source. Bulk $\delta^{15}N$ values are distinct from extra-terrestrial components and more comparable to crustal materials than those from mantle. Among the crustal components, the value is within the range of values found for the atmosphere and volcanic gases. (Contamination of present atmosphere during experiments is excluded by $N_2/Ar > 2000$.) However, carbon isotopic compositions are depleted in ^{13}C, which is typical of organic C, and similar to previous reports on carbonados. This imbalance may be due to either mixing of more than one component or source fractionations during a chemical differentiation. Nitrogen contents in carbonado matrixes are relatively low (110-340 ppm) compared with common terrestrial diamonds and distinct from impact-related diamonds in El Mimbral (K-T boundary) (Hough et al, 1997) and the Popogai crater (Shelkov et al, 1998). Within a single grain, N/C ratio can vary by up to a factor of two, which showed a positive correlation with $\delta^{13}C$. This correlation was not interpreted as a mixing of two components, but as a trace of source evolution. This condition was reached, because of the frequency distribution of N/C ratio, which has its peak at the lower side (130 ppm), and $\delta^{15}N$ does not show any correlation within experimental error. Assuming such an evolution, crystallization must take place in a fluid environment, rather than a solid-state transition. This interpretation would also indicate that the original $\delta^{15}N$ value, of the source fluid, was about 0‰. From our data it is not possible to exclude contributions for carbon from biogenic component, with $\delta^{15}N$ value of +5‰, which will also be incorporated into the carbonado. Fluid of deep-seated origin reacting with crustal material to create another fluid seem to be the most reasonable explanation. Mineral inclusions identified were quartz, rutile, zircon, florencite, xenotime halloysite and clay-like aggregates of fine (< 1µm) particles, which contained Co and W in addition to Si, Al, Mg, Ca and C, confirming their aggregation in a crustal environment. The clay-like inclusions were commonly occurred for all 3 samples studied by EPMA, up to 60% of which are believed to be C of an unknown form. Positive correlation between $^{204}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ of the quartz inclusions indicates an age of 3179 ±870 Ma (2σ). It is consistent with the result of bulk cold combustion for African carbonado. (Ozima and Tatsumoto, 1997). Antiquity of carbonados would have broaden our speculation, for such an ancient crust may have distinct conditions such as redox. In addition, carbonado matrixes seem to be as old as the inclusion, suggesting these mineral inclusions to be co-genetic with diamond matrixes, though large uncertainty prevent us from further discussion.

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