

Generation history of carbonado inferred from photoluminescence spectra, cathodoluminescence imaging, and carbon-isotopic composition

HIROYUKI KAGI,^{1,*} SHUICHI SATO,^{2,4} TASUKU AKAGI,³ AND HISAO KANDA⁴

¹Geochemical Laboratory, Graduate School of Science, The University of Tokyo, Hongo, Tokyo 113-0033

²Itami Research Laboratories, Sumitomo Electric Industries, LTD, Itami, Hyogo 664-0016, Japan

³Environmental Science on Biosphere, Graduate School of Agriculture, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8509, Japan

⁴National Institute for Material Science (NIMS), Advanced Material Laboratory, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

ABSTRACT

Carbonado diamonds from the Central African Republic were investigated using spectroscopic observations and C-isotopic analysis. Based on photoluminescence (PL) spectra, carbonado samples were classified into two groups: Group-A, which exhibits an intense PL band at 504 nm; and Group-B, which exhibits PL bands at 504, 575, and 638 nm at room temperature. PL spectra measured at 120 K gave well-resolved side-band structures of 504 nm bands. Consequently, the 504 nm band of Group-A can be assigned to the 3H center attributable to self interstitials in diamond, whereas the 504 nm band from Group-B can be assigned to the H3 center attributable to a vacancy (□) trapped at nearest-neighbor substitutional nitrogen (N) pairs. The PL band at 575 nm, which is attributable to neutral N-□ pairs, is known to increase its width with increasing residual stress in diamond aggregates. The average FWHM of the 575 nm band and the standard deviations were 3.80 and 0.54 nm for Group-A carbonado, and 2.80 and 0.38 nm for Group-B carbonado. These values suggest that Group-A carbonado samples have higher residual stress than the Group-B samples. The presence of an H3 center and the lower residual stress in Group-B carbonado both suggest that they originated from higher temperatures compared to Group-A. Radiation halos were observed in cathodoluminescence (CL) images of both Group-A and Group-B samples. The CL halos are traces of radiation damage from radioactive nuclides. The texture of the haloes suggests that the radiation damage was a secondary event after formation of the carbonado diamonds. The average C-isotopic composition of Group-A is $-23.6 \pm 0.52\%$ in $\delta^{13}\text{C}_{\text{PDB}}$ and that of Group-B is $-26.3 \pm 0.65\%$. Group-A carbonado was enriched systematically in ^{13}C compared with Group-B carbonado. Isotopic fractionation might occur as a result of the different thermal history of carbonado.

Keywords: Carbonado, cathodoluminescence, photoluminescence, carbon isotope, diamond, radiation damage