In situ observation of diamonds in ureilites by Raman spectroscopy

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Introduction: Ureilites are unique in containing relatively large amounts of C occurring as graphite or diamond [1]. The mode of occurrence and X-ray properties of carbon minerals in ureilites show that diamond in ureilites formed by high-pressure conversion of graphite that crystallized during igneous or metamorphic processes on an ureilite parent body or bodies [2]. Nakamuta et al. [3], [4] suggested that catalytic mechanism dominated in the diamond formation in some ureilites and non-catalytic one in others. Their results are mainly based on X-ray diffraction patterns of each C-rich grain. In this study, *in situ* observation of graphite and diamond crystals in each grain was made by Raman spectroscopy in order to confirm the formation mechanisms of diamond in ureilites.

Sample and Experiment: Polished thin sections of six Antarctic ureilites, Y-74123, A 77257, Y-8448, A 881931, Y-74659 and Y-74130, among which X-ray properties of diamond vary in a wide range, were observed under an optical microscope and MicroRaman spectra of graphite and diamond in each C-rich grain were obtained. MicroRaman spectra were recorded with a Jobin-Yvon T64000 triple-grating spectrometer. A microscope was used to focus the 514.5 nm Ar excitation laser beam to a 1 micron spot. Accumulations lasting 120 to 600 seconds were made. The laser power on the sample was 2 mW.

Results: C-rich grains in Y-74123, A 77257 and Y-74659 show amoeboid shapes and those in Y-8448, A 881931 and Y-74130 show blade-like shapes. The sizes of C-rich grains are about 1-2 mm in length and 0.1-0.05 mm in width. In each grain, diamond crystals can be observed in high relief under an optical microscope of high magnification. Raman spectra of diamond and graphite crystals were recorded. Raman spectra of graphite in all samples are composed of E_{2g} band at 1580-1600 cm^{-1} and D band at about 1350 cm^{-1} and agree well with that of relatively well crystallized graphite [5]. The full width of half maxima (FWHM) and peak positions of the E_{2g} bands vary among ureilites and also among crystals in a grain. The intensity ratio between D and E_{2g} bands is known to be used as an index of crystal size of graphite [5]. However, the intensity ratio is largely affected by grinding when a polished thin section is prepared [6]. Then, the intensity ratio is not considered as an index of crystal size of graphite in this study. The Raman spectra of diamonds in each sample appear around 1332 cm^{-1}. The FWHM of the Raman band vary among ureilites and also among crystals in a grain. The band of Y-74130 is conspicuously broader than those of others. Raman spectra of diamonds in Y-74123 and A 881931 are similar to that of diamond synthesized by static experiments with a catalyst or by a CVD method and the spectra of Y-74130 to that of diamond synthesized by shock or detonation experiments. The results of Raman spectroscopy suggest that diamonds in Y-74659 and Y-74130 may have formed by shock at the conditions near to those of shock or detonation experiments and those in Y-74123, A 77257, Y-8448 and A 881931 have formed at the conditions near to those of static experiments with a catalyst or at a high temperature and a high pressure near to a melting point of graphite. These results agree well with the results obtained by X-ray analyses of C-rich grains [4].

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