Graphitization of Diamond

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Introduction

Diamond is a form of carbon crystallized in the isometric system under a high pressure and high temperature. At normal pressure, diamond is thermodynamically metastable and changes its crystal structure to graphite when it is heated to high temperatures. By means of transmission electron microscopy, Evance and James examined graphitization behavior of diamond crystal fragments with 111 surfaces, thin enough for examination[1]. The fragments were heat-treated at temperatures from 1500 to 1900℃ for various times in a transmission electron microscope, and the observation and selected area electron diffraction were carried out. By heat treatment at 1600℃ for 45 min, graphite crystallite formation was detected in selected area electron diffraction pattern of a diamond fragment as the extra spots. Referring to the 220 diamond reflections, the interlayer spacing d_{002}, a half value of lattice constant c_0, and the lattice constant a_0 of the graphite crystallites transformed were evaluated to be d_{002} = 0.3348 nm and a_0 = 0.2456 nm. The d_{002} value obtained was not precise, however. Heat treatment at 1900℃ produced transformation to graphite in 5 to 10 min. The graphite crystallites had sizes in linear dimensions ranged between 10 and 15 nm. Graphitization of bulk diamond has not been studied extensively.

In the present study graphitization of the bulk diamond crystals in powder forms as well as a diamond jewel with the largest linear dimension of 4 mm was investigated by X-ray diffraction and scanning electron microscopy.

Experimental

Diamond powder specimens used were commercially available diamond powders with their grain size distributions of 0.25-0.75 μm (denoted hereafter Sample A) and 6-12 μm (Sample B). The sample A was natural diamond, and the sample B synthesized one. The samples A and B were heat-treated in the same furnace at temperatures 1700, 1800, 2000 and 2800℃, respectively, for 30 min in a flow of argon gas. For the original and heat-treated samples, X-ray diffraction was made at room temperature using a rotation anode type wide-angle diffractometer. For the heat-treated specimens, the interlayer spacing d_{002}(004), lattice constant a_0, and crystallite sizes L_c(004), L_c(112) and L_a(110) were determined. For the particles of the original and heat-treated samples, morphology was observed by a field emission gun type scanning electron microscope (FESEM).

As a reference material for the graphitization of the samples A and B, a diamond jewel described in the first section was heat-treated at 2000℃, and the X-ray parameters were determined. Morphology of this material was also observed by FESEM.

Results and Discussion

Both of the original diamond powder specimens consisted of well-crystallized polycrystalline diamond particles. Lattice constants for both specimens are the same value of 0.3560 nm. Both of the specimens transformed partially from diamond to graphite by heat treatment at 1700℃ for 30 min, but the graphitization degrees of the graphite crystallites transformed in the specimens were not high. The diamond crystallites were completely transformed to graphitic ones by heat treatments above 1800℃ for 30 min.

Figure 1 shows diffraction patterns of 2000℃-treated specimens of samples A and B, as examples of the patterns.

For the sample A heat-treated at 2000℃, separation of the 100 and 101 diffraction peaks for graphite is not distinct, and the 006 line does not appear at the high angle tail of the 112 line. At this heat treatment temperature, the sample B was graphitized better than the sample A was. This trend also appeared for the specimens heat-treated at other temperatures. The values of d_{002}(004), lattice constant a_0, and crystallite sizes L_c(004), L_c(112) and L_a(110) determined are listed in Table 1. The values of L_c(004) and L_a(110) in Table 1 for the specimens indicate that crystallites are plate-like, and graphitization degree of each of the samples A and B is improved a bit, as the heat treatment temperature (HTT) is risen.

The 2000℃-treated diamond jewel was completely broken into pieces by the heat treatment. X-ray parameters of this material are as follows; d_{002}(004) = 0.3364 nm, L_c(004) = 40.2 nm, L_c(112) = 2.2 nm, a_0 = 0.2462 nm and L_a(110) = 23.3 nm, respectively . The graphitization degree of the jewel-derived graphite crystallites is probably improved by heat treatment at higher temperatures than 2000℃, but pronounced improvement is not expected.

For the samples A and B, particles of each of them were not broken into pieces by heat treatments. Differences in morphology between heat-treated particles and the original ones are that the former are a bit rounding and contains pores of small sizes formed during heat treatments.

References

Evance, T., and James, P. F., *Proc. Roy. Soc.* A, 277, 260.

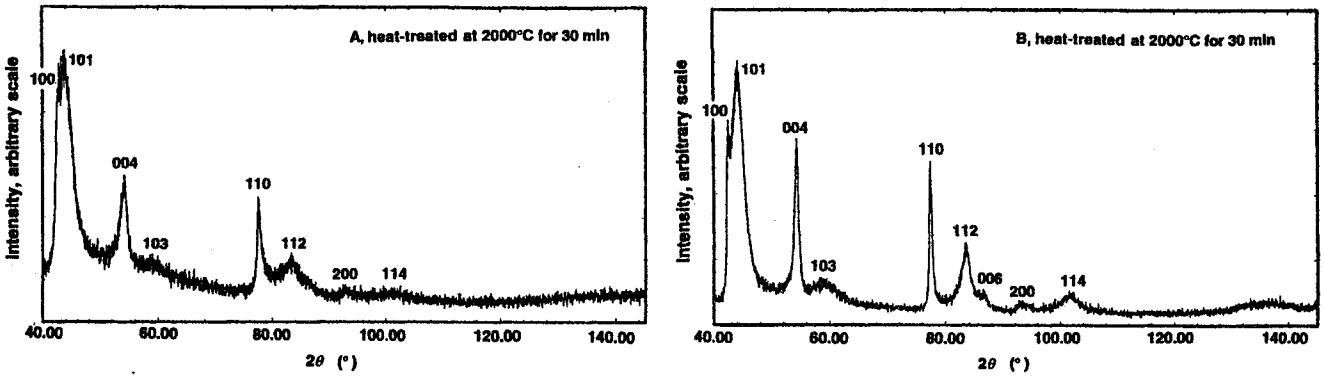

Fig. 1 X-ray diffraction patterns of samples A and B heat treated at 22000℃ for 30 min.

Table 1 X-ray parameters for heat-treated samples

HTT (℃)		1700	1800	2000	2800
d_{002}(004) (nm)	A	0.3396	0.3394	0.3387	0.3387
	B	0.3378	0.3378	0.3375	0.3369
L_c(004) (nm)	A	5.3	7.3	8.1	9.5
	B	13.0	13.0	13.8	20.5
L_c(112) (nm)	A	0.8	1.0	1.2	1.5
	B	1.8	2.2	2.4	9.4
a_0 (nm)	A	0.2459	0.2459	0.2462	0.3462
	B	0.2462	0.2462	0.2462	0.2462
L_a(110) (nm)	A	8.4	11.6	15.3	20.0
	B	24.6	26.8	30.8	47.5