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グラファイト→六方晶ダイヤモンドの相転移メカニズムの実験的考察 ーグラファイトの結晶性と静水圧性の影響ー
Experimental study on the phase transition of graphite to hexagonal diamond

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Hexagonal diamond (lonsdaleite) is a metastable polymorph of carbon and consists of ABAB... stacked sp3-bonded (tetrahedral) carbons. It occurs as microscopic crystals associated with graphite and cubic diamond in carbonaceous meteorites such as the Canyon Diablo meteorite and impact craters and can also be synthesized from well-crystalline graphite by high pressure experiments (e.g. Bundy and Kasper, 1967; Yagi et al., 1992). The phase transition of graphite to hexagonal diamond is considered to be a martensitic process, where [100] of hexagonal diamond is located parallel to [001] of the host graphite. However, we recent found a variety of such coaxial relations between graphite and hexagonal diamond based on TEM observations of samples synthesized by laser-heated diamond anvil cell (DAC). This suggests that the martensitic phase transition process is not always simple, but can be complex. Here, we conducted further research on the transition mechanism by using highly-oriented graphite samples with different crystallinities under different hydrostatic conditions.

We performed a series of high-pressure and high-temperature experiments at a pressure of 25 GPa and temperatures of 1800-2200K. Three types of highly-oriented graphite were used as starting materials: 1) highly-oriented graphite (HOG, Murakami et al, 1992), 2) highly-oriented pyrolitic graphite (HOPG) and 3) Kish graphite. Each sample was compressed in a DAC with/without an ethanol/methanol pressure transmitting medium and rapidly heated to a target temperature using fiber laser (1072 nm). The sample became transparent after laser heating, suggesting that the phase transition of graphite to diamond phase(s). The recovered samples were first examined by Raman spectroscopy for phase identification and then by transmission electron microscopy (TEM) for microtextural observations and electron diffraction analysis.

TEM observation revealed that the transparent area of recovered samples consists purely to mostly of hexagonal diamond with a layered structure similar to that of the graphite starting sample. A trance amount of cubic diamond also coexists in some cases. The electron diffraction (ED) patterns collected from pure hexagonal diamond synthesized from HOG and HOPG samples are complex and can be interpreted as a superposition of several types of reciprocal patterns in which [100], [002] and [-212] of hexagonal diamond are all arranged in a coaxial relation with graphite [001]. This suggests that the phase transition from graphite to hexagonal diamond proceeds mostly by $1/2(3a)^{1/2}$ or $1/(3a)^{1/2}$ shifts of graphene layers along graphite [100]. On the other hand, the ED patterns collected from Kish graphite are simple and can be indexed as a single reciprocal lattice where [100] of hexagonal diamond is parallel to [001] of the host graphite. It is likely that the crystallite size of hexagonal diamond synthesized correlates positively to the crystallite size (particularly in the a-axis direction) of initial graphite sources. Furthermore, the variety of the coaxial relation and transition process seems to be originated from the mosaicity (misorientation along c-axis) of the graphite sources.

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