

<b>Name</b>	<b>Tokuyuki TERAJI</b>
<b>Organization</b>	<b>National Institute for Materials Science, Japan</b>
<b>Division/Department</b>	<b>Wide Bandgap Material Group, Optical and Electronic Materials Unit</b>
<b>Title</b>	<b>Dr.</b>

<b>Name</b>	<b>Takashi TANIGUCHI</b>
<b>Organization</b>	<b>National Institute for Materials Science, Japan</b>
<b>Division/Department</b>	<b>Ultra-High Pressure Processes Group, Materials Processing Unit</b>
<b>Title</b>	<b>Dr.</b>

<p><u>Presentation Title</u></p> <p>Synthesis of isotope controlled diamond crystals for quantum device application</p>
<p><u>Abstract</u></p> <p>NV center in diamond gives us suitable properties for quantum computing. <math>^{12}\text{C}</math> enriched diamonds are proper crystal network for these applications because reduction of <math>^{13}\text{C}</math> with nuclear spin makes the coherent time of the NV centers longer. Reduction of nitrogen contents and improving the crystalline quality of diamond are also key issue for the device applications. In this presentation, technological achievement on the isotope-controlled diamond synthesis will be described. We employed two different synthesis methods for obtaining these diamond crystals; one is low-pressure chemical vapor deposition (CVD) method and the other is high pressure / high temperature (HPHT) method.</p> <p>[Low-pressure CVD]</p> <p>Homoepitaxial single crystal diamond (100) films were grown by means of microwave-assisted chemical vapor deposition (MPCVD), using isotopic enriched methane (<math>^{12}\text{C}:5\text{N}</math> grade) as a source gas. In order to obtain rather thick diamond films with higher crystalline quality, high-power MPCVD condition was applied [1]. In addition, UHV compatible metal chamber was utilized to minimize the unintentional impurity incorporation in diamond. Methane concentration, reaction pressure and reaction temperature are 10%, 140 Torr and 950-1000°C, respectively. Highly purified IIa (100) diamond crystals whose top surface was mirror polished were used as substrate.</p> <p>As shown in Fig. 1, carbon was well converted isotopically from methane into diamond film (<math>^{12}\text{C}:99.998\%</math>). Other elements such as hydrogen, nitrogen, silicon and boron were not detected by SIMS, meaning a highly purified <math>^{12}\text{C}</math> enriched crystal. ESR</p>

measurement reveals isolated nitrogen contents is less than 0.5 ppb. High crystallinity of the homoepitaxial diamond films was evidenced by intense free exciton emission in the CL measurement. Coherent time of NV center created by implantation in there isotope controlled crystals was, however, not as long as expected at this moment. Understanding of the cause of shortening the coherent time and farther improvement for removing impurities/defects are in progress.

#### [HPHT]

Synthesis of diamond crystals was carried out by using temperature gradient method near 5.5GPa and 1450°C region using Belt-type HP apparatus. The source materials of  $^{12}\text{C}$  enriched carbon used were CVD diamond obtained by hot filament method using isotopic enriched methane ( $^{12}\text{C}:5\text{N}$  grade) as a source gas. Fig.2 shows typical HPHT crystal after growth duration of 42hrs in this study. At the present stage, growth rate of the crystals was approximately 0.3-0.6mg/hr by using HP chamber for the sample space of 7mm in diameter.

The key issues to control the crystals' quality (size, residual nitrogen and boron impurities, metal inclusions and residual stress) are to optimize the composition of metal solvents and HPHT growth condition. Variety of solvent compositions such as Ti and Cu contents was tested so as to control the nitrogen content in the grown crystals. Nitrogen getter of Ti content of 0.2 to 5wt% in the solvent exhibits correlation of color in grown diamond crystals, while 7wt% Ti in the solvent seems degraded growth condition.

On the other hand, due to the lack of appropriate boron getter so far, the control of residual boron impurity can only be carried out by selecting high purity starting materials system. The quantitative study of residual boron content in the crystals should be important for the future work.

By using Co-Ti-Cu solvent system, diamond crystals of  $^{12}\text{C}:99.995\%$  were obtained as characterized by SIMS study. Compared  $^{12}\text{C}$  isotopic purity between source materials and grown crystals, some unknown  $^{13}\text{C}$  contamination (i.e.0.005%) may affect the isotopic impurity in the growth circumstance. Since the possible origin of the  $^{13}\text{C}$  contamination is carbon diffusion from graphite furnace in HPHT chamber, an attempt is in progress to minimize this impurity effect.

[1] T. Teraji, "Chemical vapor deposition of homoepitaxial diamond films", Phys. Stat. Sol. (a) **203**, 3324-3357 (2006).

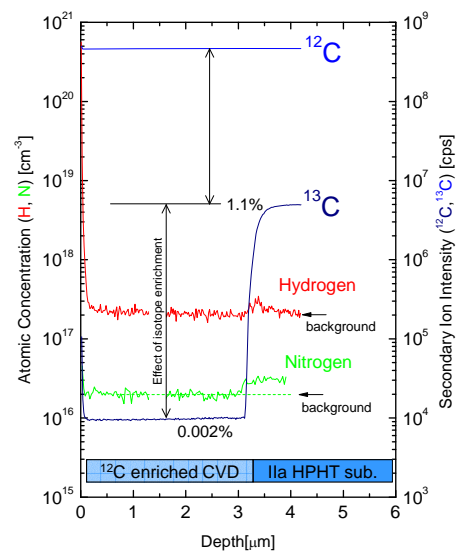


Fig.1 SIMS depth profile of homoepitaxial diamond deposited on Ila diamond (100).

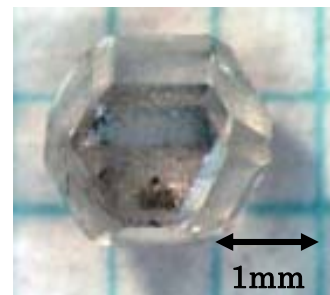


Fig.2 HPHT as grown diamond crystal.