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学 位 論 文 要 旨
Dissertation Summary

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論　文　名 : Synthesizing Diamond onto Metal Substrate by in-Liquid Plasma CVD Method
(Dissertation Title)

Several methods of gas phase CVD such as Hot Filament, DC arc, Jet DC Plasma, Laser Plasma Oxygen–acetylene Flame, and Microwave Plasma are difficult to synthesize on a substrate which has low heat resistance, large devices, and considered very costly in large scale production. So, the synthesizing diamond thin film using In-liquid plasma technique was our target in this study. In-liquid plasma is generated plasma in a bubble in liquid and synthesizing diamond in liquid alcohol is available. A Remarkable effect of liquid plasma process is high growth rate because the molecular density of a liquid is much higher than that of a gas. And the substrate which has low heat resistance can be deposited because of the cooling effect of the liquid. Furthermore, the experimental device becomes simple and easy. Therefore, synthesis of diamond using in-liquid plasma CVD is expected as alternatives besides the conventional gas phase plasma CVD.

The theory of IL-MPCVD is relatively new. But in general, the theory is almost the same as gas phase CVD. Both use the Bachmann diagram as a reference. This submerged plasma is a proprietary technology of Ehime University, it uses a microwave or high frequency as a plasma energy source and generated in liquid.

The mechanism of diamond synthesis using an in-liquid plasma chemical vapor deposition (CVD) method has been investigated. Chemical reactions from a liquid mixture of methanol and ethanol (in-liquid plasma CVD) and a gas mixture of methane and hydrogen (gas-phase CVD) are used for analyzing. The procedure was, initially, synthesizing CO and then induces a chemical reaction using the remaining C and H to synthesize a carbon substance. From the experimental results of carbon deposits using various liquid and gas mixtures as the source materials, found that the region of the remaining H and C after CO synthesis satisfying $H/C \geq 20$ is necessary to synthesize diamonds using in-liquid plasma CVD method. The IL-MPCVD method has succeeded to synthesize single crystal diamond (100) and (111) plane. Deposited diamond film on (100) has film growth gradually and smooth surface. Whereas (111) face has a polycrystalline film with irregularity growth and rough surface. The surface suitable for homoepitaxial growth was found to be (100).

Growing diamond on ferrous base materials is challenging because of its difficulties due to several factors. First, carbon can diffuse into the substrate with relatively high diffusion rate, so diamond nucleation time can be long. Second, the catalytic effect of iron on the growth of Sp^2 dominated amorphous and nanocrystalline carbon. Third, poor adhesion and high residual stress appear because the thermal expansion coefficient between diamond and ferrous metal are not compatible.

Diamond nucleation on non-diamond substrate is generally proposed to occur mostly on an intermediate layer of amorphous carbon, metal carbides and graphite. In the current study, silicon as substrate and Titanium as interlayer were used. Both of them were successful for growing diamond.

The application of the IL-MPCVD method for the synthesis of diamond film on stainless steel using double interlayer Ti/Si was investigated. The thickness of the interlayer for appropriate diamond film formation has achieved, despite the fact that the result should be improved.

For the future experiment, nickel is one of the materials that should be used for growing diamond. Nickel is one of the few materials that have a close lattice-parameter match with diamond ($a = 3.52$ Å for Ni and 3.56 Å for diamond). However, its high solubility for carbon and its strong catalytic effect on hydrocarbon decomposition and subsequent graphite formation at low pressures have prevented CVD diamond nucleation on the Ni surface without the deposition of an intermediate graphitic layer.