Diamond-like carbon films and carbon nanomaterials deposition from organic liquid phase

Kiyoharu Nakagawa[1], Mikka Nishitani-Gamo[2], Hidenori Gamo[3], and Toshihiro Ando[1]*

[1]NIMS, 1-1 Namiki Tsukuba, Ibaraki 305-0044, Japan
[2]University of Tsukuba,1-1-1 Tennodai Tsukuba, Ibaraki 305-8577, Japan
[3]Toppan Printing Co. Ltd., Takanodai-minami Sugito, Saitama 345-8508, Japan

Dense and hard diamond-like carbon (DLC) films were deposited on silicon substrates by a novel liquid phase deposition method in alcohols. Carbon nanotubes (CNT) arrays were also grown on silicon substrates by using a novel catalytic method in alcohols with Fe catalysts. Extraordinary un-equilibrium condition between the substrate surface and the liquid phase gave significantly pure hard carbon films within a few minutes in methanol and/or ethanol. This very fast and dense production of DLC films can be attributed to the great difference of chemical potential between the substrate surface and reactant liquid.

Figure 1 shows experimental set-up of the liquid phase deposition system. The reactor chamber consists of glass flask, glass condenser and tubes. Silicon was used as a substrate to be resistively heated to 600~1300^{o}C in alcohol. The reactor chamber was set in cooling water bath to maintain the temperature of alcohol below its boiling point. The sample was immersed in cool liquid alcohol. Even though the sample was heated to very high temperature 600~1300^{o}C, the temperature of whole liquid alcohol was kept around at room temperature. The large difference in temperature of the interface between the substrate catalyst and the liquid phase causes a large gradient of the chemical potential to give very high nucleation density and high growth rate. Figure 2 shows the DLC films deposited in methanol (Fig. 2(a)) and in ethanol (Fig. 2(b)). The films were dense and smooth. EDX and XPS analyses revealed that these films consisted of pure carbon and the surface was oxidized. Raman spectra from these samples indicated typical two board band, namely D-band around at 1350cm^{-1} and G-band around at 1580 cm^{-1}.

By using the same system, carbon nanotubes were grown on the silicon substrate with a small amount of Fe catalyst. Catalytic decomposition in this system give dense carbon nanotube arrays. When diamond was used as a substrate, a novel composite material, CNT array/diamond, was formed. This vacuum-free process is adaptable to industrial levels, and may find a large field of applications in the synthesis of various kinds of carbon films and carbon nanomaterials through the use of organic liquid sources.

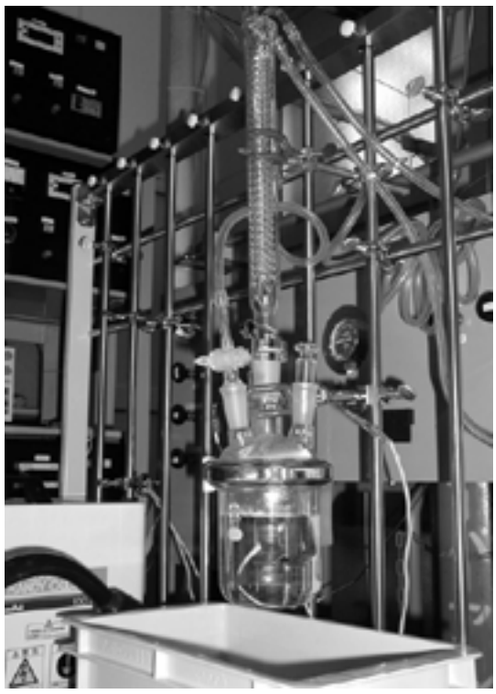

Fig. 1 Chamber for synthesis of DLC in organic liquid

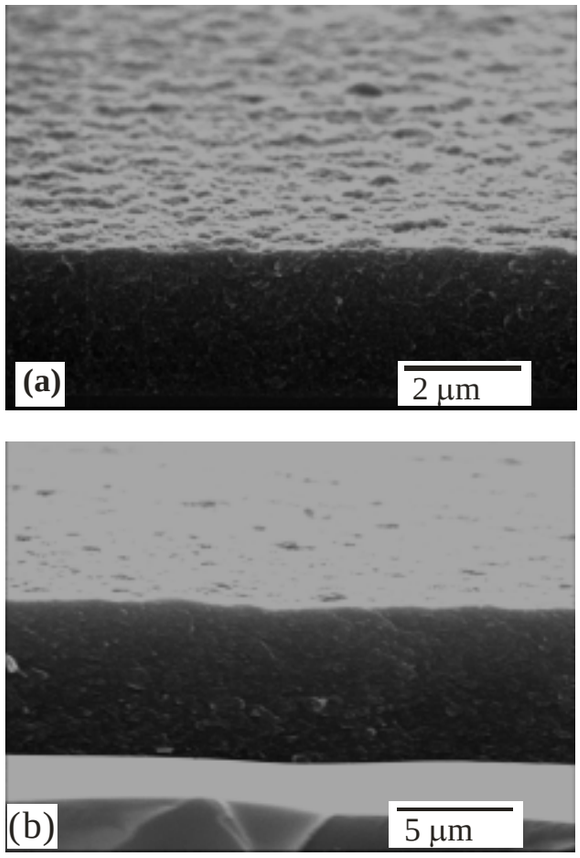

Fig. 2 SEM images of the DLC deposited in (a) methanol, and (b) ethanol at 780^{o}C

1) Y. F. Zhang, M. N.-Gamo, C. Y. Xiao, and T. Ando, J. Jpn. Appl. Phys., *41*, L408 (2002).
2) Y. F. Zhang, M. N.-Gamo, K. Nakagawa, and T. Ando, J. Mater. Res., *17*, 2457 (2002).

* **c-diamond@md.neweb.ne.jp**