A liquid-phase synthesis of various carbon nanofilaments in liquid hydrocarbons

Kiyoharu Nakagawa^a, Mikka Nishitani-Gamo^b, Hidenori Gamo^c, and Toshihiro Ando^{a,*}

a National Institute for Materials Science (NIMS), 1-1, Namiki, Tsukuba, Ibaraki, 305-0044, Japan b Department of Applied Chemistry and Sensor-photonics Research Center, Toyo University, 2100 Kujirai, Kawagoe, Saitama, 350-8585, Japan c Toppan Printing Co. Ltd. 4-2-3, Takanodai-Minami, Sugito, Saitama 345-8508, Japan

*E-mail: c-diamond@md.neweb.ne.jp

Carbon nanofilaments were grown on silicon substrates by using a novel catalytic method in an organic liquid hydrocarbon. Nonequilibrium catalytic deposition gave significantly pure carbon nanofilaments with very little soot within a few minutes in n-octane. Scanning electron microscope images indicate that the nanofilaments are grown perpendicular to the Si substrate surface. This very rapid and dense production of carbon nanofilaments can be attributed to the great difference of chemical potential between the substrate surface and reactant liquid.

The production and utilization of nano-carbon materials such as fullerene, filament, and carbon nano-tubes (CNTs) have attracted much recent attention. In general, various methods of synthesizing carbon filaments or CNTs--such as the disproportionation of CO, the thermal decomposition of hydrocarbons, arc discharge, laser ablation, and plasma CVD, have been reported. Until now, however, these have been no satisfactory method for the production of carbon filaments or CNTs.

Recently, we proposed one of the most effective method for synthesizing CNT using liquid-phase nano-amorhozo. In the liquid-phase nano-amorhozo method, CNTs were grown in organic liquids such as methanol and ethanol. This liquid-phase synthetic method enables great chemical flexibility and synthetic tunability, and the synthesis process is compatible with the existing semiconductor processes and easily adaptable to industrial production levels. CNTs produced by arc discharge must be separated from many impurities such as soot by various purification steps. In contrast, carbon nanomaterials produced using the liquid-phase nano-amorhozo technique do not need to be separated from products. Moreover, because carbon nanomaterials can be formed with high efficiently in large yields on Si substrate, they might be applied to the development of new and unique electronic devices. In this paper, we designed on effective technique for synthesizing of carbon nanofilament on Si substrate in liquid hydrocarbon using n-octane, an important ingredient in chemical feedstock. In our method, the organic liquid is rapidly converted into solid carbon nanofilament.

The apparatus consisted of a quartz chamber with outside cooling (Fig. 1). A direct current (DC) electric power supplier was used to apply a controlled current to the substrate. Highpurity n-octane (99.7%) was used as the organic liquid source. Si (100) substrates of low resistance (0.002 Ω -cm) and a size of $10 \times 20 \times 1$ mm3 were placed perpendicularly on the center of the chamber in the liquid and connected to the electric power supplier. The Si (100) substrate was ultrasonically cleaned in acetone and coated with Fe film 5 nm thick in a magnetron sputtering system using low-DC electric voltage (0.4 kV, 30 mA) and low Ar pressure (0.6 torr) for the production of nucleation sites for carbon nanofilament growth.

n-Octane was the only liquid hydrocarbon source used in our experiments. The as-grown carbon nanofilaments appeared as a dark black film on the Si substrate. Scanning electron microscope (SEM) images revealed that the sample consisted almost entirely of carbon nanofilaments (Fig. 2). The carbon nanofilaments were prepared in n-octane liquid at 1073 K. The reaction times were 10 and 300 s, respectively. Although their diameters and lengths differed, both nanofilaments had a zigzag structure.

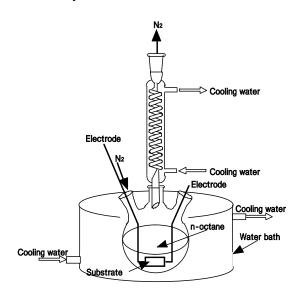
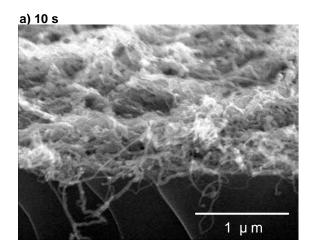


Fig.1 Schematic of the experimental setup chamber used to synthesize carbon nanomaterials in organic liquids. Substrate temperature was measured by optical pyrometers.



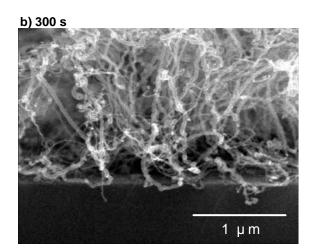


Fig.2 SEM images of carbon nanofilaments on a Si surface formed in noctane.

Acknowledgments

Some parts of this work were financially supported by Foundation for Promotion of Material Science and Technology of Japan (MST Foundation), Japan Science and Technology Corporation (JST), and Novel Carbon Project (NIIMS #166509).