Crucial Role of Gas-Phase Pyrolysis of Ethylene in Rapid Growth of Carbon Nanotubes

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Rapid growth of millimeter-thick SWNT forests in a few minutes was realized by water-assisted ethylene CVD [1]. We previously reproduced such growth using $C_2H_4/H_2/H_2O/Ar$ reactant gas and Fe/Al$_2$O$_3$ catalyst, and found an essential role of Al$_2$O$_3$ under layer [2]. In this work, we studied the effect of gas phase and substrate temperatures separately, and found a crucial role of gas phase reaction in forming actual precursor from ethylene.

Figure 1 shows the experimental apparatus used in this study. The reactant gas was once heated by flowing through an externally-heated quartz tube, cooled down, and then fed to a resistively-heated graphite substrate on which the catalyst was supported. The typical condition was 60 Torr $C_2H_4$/ 200 Torr $H_2$/ 0.076 Torr $H_2O$/ Ar for the reactant gas and 1 nm Fe/ 20 nm Al$_2$O$_3$ for the catalyst.

Figure 2 shows the side-view images of the graphite substrates after CVD at 800 °C substrate temperature for 10 min. Nanotubes did not grow efficiently at preheating temperatures of 700 °C or below, but they grew efficiently to millimeter-thickness at higher preheating temperatures. CHEMKIN simulation showed the decomposition of $C_2H_4$ and formation of $C_2H_2$ in a residence time of a few seconds. Then, we mixed 4 Torr $C_2H_2$ with 169 Torr $H_2$/ 0.076 Torr $H_2O$/ Ar instead of $C_2H_4$, and found that millimeter-thick nanotube forests actually grew without preheating. $C_2H_2$ is the actual precursor for the rapid nanotube growth from $C_2H_4$ feedstock.

Fig.1 Experimental apparatus

Fig.2 Images of SWNT forests (a) no preheating, (b)preheating: 700 °C, (c)preheating: 850 °C

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