

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 C₂H₄/H₂/H₂O/Ar reactant gas and Fe/Al₂O₃ catalyst, and found an essential role of Al₂O₃ 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 C₂H₄/ 200 Torr H₂/ 0.076 Torr H₂O/ Ar for the reactant gas and 1 nm Fe/ 20 nm Al₂O₃ 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 C₂H₄ and formation of C₂H₂ in a residence time of a few seconds. Then, we mixed 4 Torr C₂H₂ with 169 Torr H₂/ 0.076 Torr H₂O/ Ar instead of C₂H₄, and found that millimeter-thick nanotube forests actually grew without preheating. C₂H₂ is the actual precursor for the rapid nanotube growth from C₂H₄ feedstock.

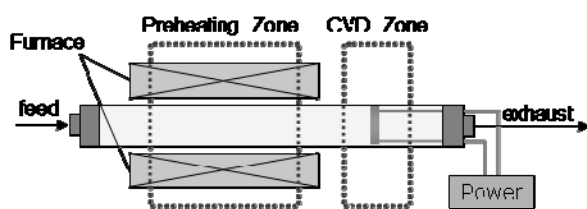


Fig.1 Experimental apparatus



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

^[1]K. Hata, et al., *Science* **306**, 1362 (2004) ^[2]S. Noda, et al., *Jpn J. Appl. Phys.* **46**, L399 (2007)