Growth Window and Possible Mechanism of Millimeter-Thick Single-Walled Carbon Nanotube Forests

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The water-assisted growth method, so-called "supergrowth", has an outstanding growth rate of single-walled carbon nanotubes (SWNTs) [1]. However, few research groups have succeeded in reproducing it and underlying mechanism of the growth rate enhancement is unclear. We recently reproduced the "supergrowth" by a parametric study on both reaction and catalyst conditions [2]. In this work, we report in detail the effect of the conditions determined, and discuss the novel mechanism essential for rapid growth of SWNTs.

Our standard condition is 8.0 kPa C₂H₄, 27 kPa H₂, 0.010 kPa H₂O, 67 kPa Ar at 1093 K for 10 min. Figure 1a shows the nanotube sample grown by the combinatorial catalyst library [3] of 0.2-3-nm Fe on Al₂O₃/SiO₂. 0.5-nm-thick Fe grew SWNTs with diameter around 4 nm as shown in Fig. 1(b), and thicker Fe grew thicker nanotubes. Nanotube forests grew up to millimeter and sub-millimeter thicknesses for Fe and Co catalyst, respectively, only when supported on Al₂O₃ layer. When catalysts were supported on SiO₂, the thickness of nanotube films was as small as sub-micrometers. The window for the rapid SWNT growth was narrow. Optimum addition of H₂O (0.010 kPa equals 100 ppmv) increased the SWNT growth rate but further addition of H₂O degraded both the SWNT growth rate and quality. Addition of H₂ was also essential for rapid SWNT growth, but again, further addition decreased the SWNT growth rate. Because Al₂O₃ catalyzes hydrocarbon reforming, Al₂O₃ possibly enhances the SWNT growth rate by dissociating and supplying the carbon species to the catalyst particles. The origin of the narrow window for rapid SWNT growth will also be discussed.

Fig. 1 (a) Photograph of Fe/Al₂O₃ combinatorial catalyst library after CVD under the standard condition. (b) TEM image of as-grown SWNTs at 0.5-nm-thick Fe under the same condition.


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