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 C_2H_4, 27 kPa H_2, 0.010 kPa H_2O, 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 Al_2O_3/SiO_2. 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 Al_2O_3 layer. When catalysts were supported on SiO_2, the thickness of nanotube films was as small as sub-micrometers. The window for the rapid SWNT growth was narrow. Optimum addition of H_2O (0.010 kPa equals 100 ppmv) increased the SWNT growth rate but further addition of H_2O degraded both the SWNT growth rate and quality. Addition of H_2 was also essential for rapid SWNT growth, but again, further addition decreased the SWNT growth rate. Because Al_2O_3 catalyzes hydrocarbon reforming, Al_2O_3 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.

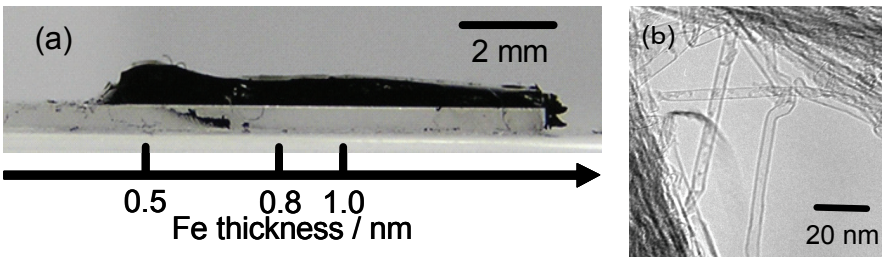

Fig. 1 (a) Photograph of Fe/Al_2O_3 combinatorial catalyst library after CVD under the standard condition. (b) TEM image of as-grown SWNTs at 0.5-nm-thick Fe under the same condition.

[1] K. Hata, et al., *Science*, **306**, 1362 (2004). [2] S. Noda, et al., *Jpn. J. Appl. Phys.*, **46**, L399 (2007). [3] S. Noda et al., *Carbon*, **44**, 1414 (2006).

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