Alcohol CVD Growth and Optical Characterization of Vertically-Aligned Single-Walled Carbon Nanotubes

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The vertically aligned single-walled carbon nanotubes (SWNTs) [1, 2] with film thickness up to 30 microns is grown on quartz or silicon substrates by alcohol catalytic CVD. The growth condition and growth mechanism of VA-SWNTs is discussed based on the *in-situ* growth monitoring by laser absorption [3, 4] during CVD. As shown in Fig. 1, film thickness L of VA-SWNTs film at various temperature, flow-rate, and pressure can be well fit by an empirical equation:

$$L = \gamma_0 \tau \left(1 - \exp(-t/\tau)\right)$$

where γ_0 and τ are initial growth rate and growth decay time, respectively. The initial growth rate γ_0 is linearly proportional to pressure up to the critical pressure which is determined by temperature [4]. This result indicates the first order reaction below the critical pressure.

The non-flow CVD [5] turned out to be very efficient, resulting a thicker film up to 100 μm. The growth curve is obviously different probably because of the contribution of small amount of acetylene thermally decomposed from ethanol. The carbon conversion rate from ethanol to VA-SWNTs can be as high as 40 %. Hence, isotopically modified ethanol can be employed to study the growth process [6] as in Fig. 2. The detailed chemical reaction process in gas-phase and on metal catalysts will be discussed based on CVD results using isotope labeled ethanol and acetylene as carbon source.

Recently developed removal and transfer technique of this film [7] enabled a direct TEM observation of free-standing vertically aligned SWNTs along the alignment direction. It was revealed that the film is comprised primarily of small SWNT bundles, typically containing 3-8 SWNTs [8]. This minimum bundling structure is ideal for various optical characterizations such as resonant Raman [9] and possible production of homogeneous composite materials [10]. Optical characterization of such a film using polarized absorption, polarized Raman, and photoluminescence spectroscopy will be discussed. Laser-excitation of a vertically aligned film from top means that each nanotube is excited perpendicular to its axis. Because of this predominant perpendicular excitation, interesting cross-polarized absorption [2, 11] and interesting and practically important Raman features as shown in Fig. 3 are observed.

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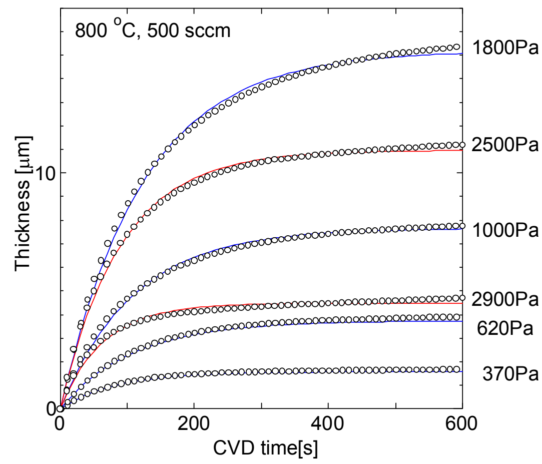

Fig. 1. *In-situ* measurement of film thickness of vertically aligned SWNTs by laser absorption during CVD.

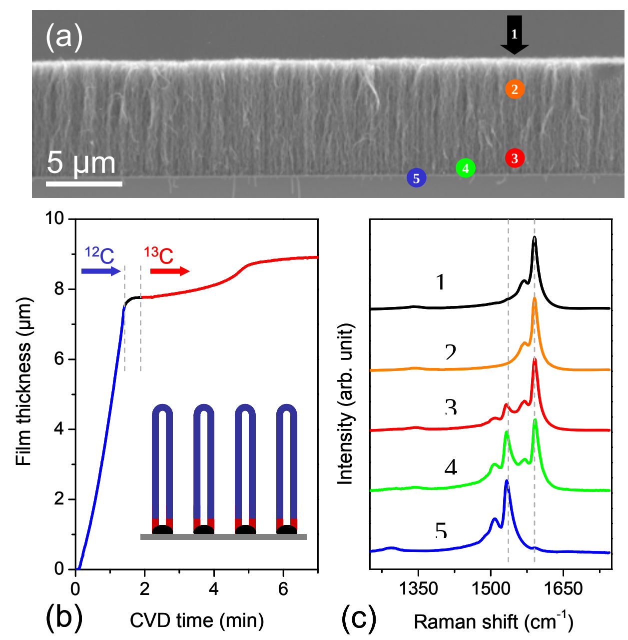

Fig.2 Demonstration of root-growth of VA-SWNTs by ^{13}C isotope. No-flow CVD was used for efficient growth.

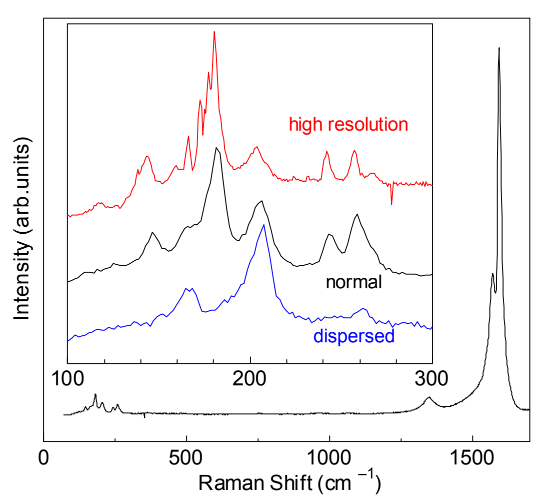

Fig. 3 Anomalous radial breathing mode of Raman scatterings excited from the top of a VA-SWNTs film.