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

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By using alcohol as a carbon source in a catalytic CVD (ACCVD) [1], high-purity single-walled carbon nanotubes (SWNTs) can be grown at relatively low temperatures. The vertically aligned SWNTs film [2-4] with thickness up to 30 microns is grown on quartz or silicon substrates by employing the simple dip-coat preparation of catalytic metal particles. The growth mechanism of vertically aligned single-walled carbon nanotubes (VA-SWNTs) is discussed based on the *in-situ* growth monitoring by laser absorption [5,6] during CVD. The growth curves are characterized by an exponential decay of the growth rate γ, from the initial value γ_0 and decay time constant τ. The initial growth rate γ_0 is linearly proportional to pressure up to the critical value which is determined by CVD temperature. This result indicates the first order reaction below the critical pressure [6]. Beyond this critical pressure, the growth decay time drastically decreases probably due to the carbon over-coat on metal catalysts.

The non-flow CVD [7] 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. In fact, a sudden increase of growth rate can be observed by adding a small amount of acetylene during ACCVD. However, the deactivation rate of catalysts is also larger with acetylene. Nevertheless, the carbon conversion rate from ethanol to VA-SWNTs can be as high as 40 % in no-flow ACCVD. Hence, isotopically modified ethanol can be employed to study the growth process [8]. 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.

Finally, optical characterization of the VA-SWNT 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 and confusing and practically important Raman features are observed.

References

1. S. Maruyama, R. Kojima, Y. Miyauchi, S. Chiashi, M. Kohno, Chem. Phys. Lett. 360 (2002) 229.
2. Y. Murakami, S. Chiashi, Y. Miyauchi, M. Hu, M. Ogura, T. Okubo, S. Maruyama, Chem. Phys. Lett. 385 (2004) 298.
3. Y. Murakami, E. Einarsson, T. Edamura, S. Maruyama, Phys. Rev. Lett., 94 (2005) 087402.
4. E. Einarsson, H. Shiozawa, C. Kramberger, M. H. Ruemmeli, A. Gruneis, T. Pichler, S. Maruyama, J. Phys. Chem. C, 111 (2007) 17861.
5. S. Maruyama, E. Einarsson, Y. Murakami, T. Edamura, Chem. Phys. Lett. 403 (2005) 320.
6. E. Einarsson, Y. Murakami, M. Kadowaki and S. Maruyama, Carbon, 46 (2008) 923.
7. H. Oshima, Y. Suzuki, T. Shimazu, S. Maruyama, Jpn. J. Appl. Phys. 47 (2008) 1982.
8. R. Xiang, Z. Zhang, K. Ogura, J. Okawa, E. Einarsson, Y. Miyauchi, J. Shiomi, S. Maruyama, Jpn. J. Appl. Phys., 47 (2008) 1971.