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Understanding Growth Termination of Single-Walled Carbon Nanotube Carpets by Documenting the Evolution of Catalyst Morphology with the Transmission Electron Microscope

Seung Min Kim*, Cary L. Pint**, Placidus B. Amama***, Dmitri N. Zakharov****, Robert H. Hauge*****, Benji Maruyama***, and Eric A. Stach*

* School of Materials Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47906.
**Department of Physics and Astronomy and Richard E. Smalley Institute for Nanoscale Science and Technology, Rice University, Houston, TX 77005
*** Air Force Research Laboratory, Materials and Manufacturing Directorate, AFRL/RX, Wright-Patterson Air Force Base, OH 45433
****Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47906
*****Department of Chemistry and Richard E. Smalley Institute for Nanoscale Science and Technology, Rice University, Houston, TX 77005

Following Hata's pioneering work regarding water-assisted "supergrowth" [1], vertically aligned single-walled carbon nanotube (SWNT) carpets have received enormous attention, as they have a large number of potential applications. However, a lack of understanding of the mechanisms of growth termination remains a limiting factor in optimizing these structures for widespread usage. Recently, our group has investigated the role of water in supergrowth and hypothesized Ostwald ripening induced growth termination mechanism [2].

In order to further understand the growth termination phenomena, we have investigated the evolution of catalyst morphology using both standard and in-situ transmission electron microscopy techniques. A series of tightly controlled annealing and growth studies were performed in a dedicated chemical vapor deposition using three different gas ambients (hydrogen, hydrogen and water, and hydrogen, water, and atomic hydrogen). Quantitative characterization of the catalyst sizes as a function of annealing time leads to an explanation for growth termination in terms of not only Ostwald ripening, but also in terms of catalyst diffusion into the substrate as two important processes involved in growth termination.

These results are corroborated by real time observations of nanotube growth and growth termination in a dedicated environmental cell transmission electron microscope, where both ripening and substrate diffusion are directly observed. We will correlate these results with a simple computational simulation of the catalyst size evolution that incorporates both of these effects.

References

1. Hata, K., et al., *Water-Assisted Highly Efficient Synthesis of Impurity-Free Single-Walled Carbon Nanotubes.* Science, 2004. **306**: p. 1362-1364.
2. Amama, P.B., et al., *Role of Water in Super Growth of Single-Walled Carbon Nanotube Carpets.* Nano Lett., 2009. **9**(1): p. 44-49.

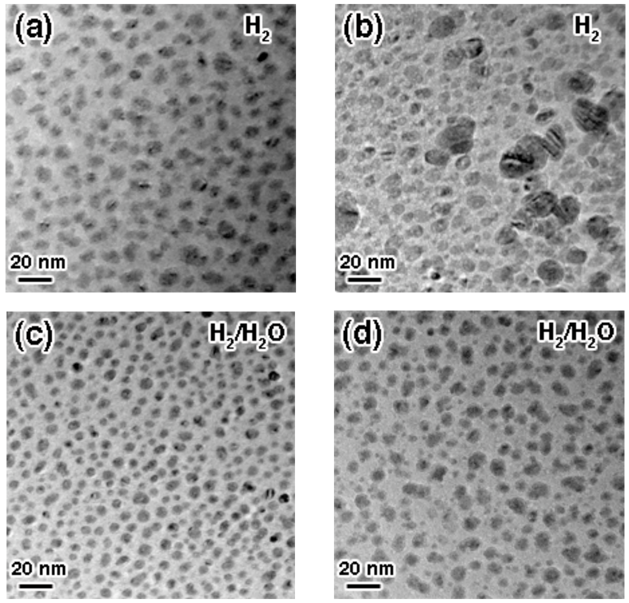

Figure 1. Plan-view TEM images of catalyst nanoparticles formed on the substrate after annealing in H_2 for (a) 30 s and (b) 5 min, and in H_2/H_2O for (c) 30 s and (d) 5 min. A reduction in the number of catalyst particles occurs from 30 s to 5 min. Each catalyst particle that disappears can no longer support nanotube growth. The number of catalyst particles in the H_2/H_2O is higher than that of H_2 alone, implying a higher number of growing nanotubes [2].

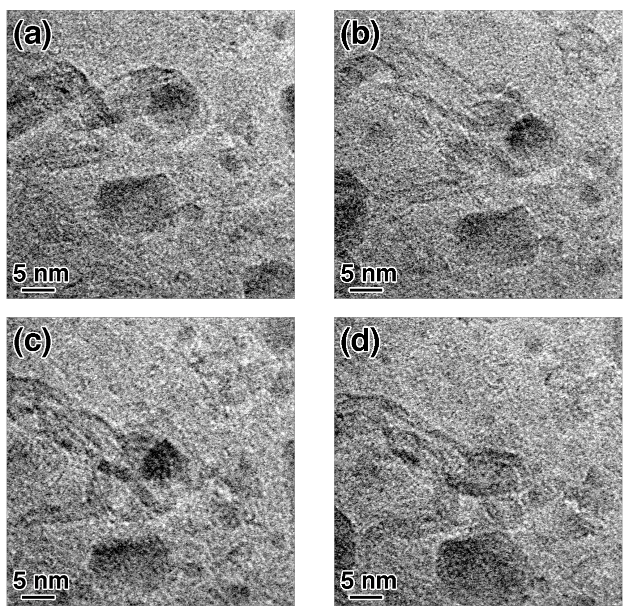

Figure 2. In-situ video frames showing evolution of a catalyst from which carbon nanotube is growing. In (a) and (b), carbon nanotube is growing and it almost stops growing while the catalyst are dissolving in (c). In (d), the catalyst is completely dissolved out and growth is terminated. Growth was performed at 650 ˚C in 2.5 mTorr C_2H_2 and 7.5 mTorr H_2 using the environmental cell transmission electron microscope.