Growth mechanism of multi-millimeter-tall single-wall carbon nanotube forests using Fe/Gd/Al catalysts

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Growth of vertically-aligned single-wall carbon nanotube (VA-SWCNT) forests by the catalytic chemical vapor deposition (CVD) is an attractive method for making applications. However, the growth termination of the CNT forests is an obstacle, and the deactivation of the catalyst nanoparticles due to the structure change of the catalyst nanoparticles is thought to be one reason for the termination. In general, maintaining smaller catalyst nanoparticles which are necessary for the SWCNT growth is more difficult because the smaller nanoparticles are less stable. As for the growth temperature, higher temperature is preferred for the high growth rate, but the structure change of the catalyst nanoparticles by migration, sintering, or Ostwald ripening is accelerated resulting in the shorter growth lifetime. To realize the longer growth lifetime, engineering catalysts is crucially important. So far, Fe-Gd catalyst on Al_2O_3 layer was reported to realize the growth lifetime of 13h and 22-mm-tall multi-walled CNT (MWCNT) forest at the growth temperature of 780 °C [1]. However, the growth rate is relatively low as ~0.5 μm s^{-1}, and the possibility of using Gd for SWCNT growth was not discussed in detail.

In this work, we applied the Fe/Gd/Al catalyst to the growth of SWCNT forests, and systematically studied the mechanism behind the enhanced growth (Fig. 1a). By optimizing the catalyst condition, we achieved a high initial growth rate of ~2 μm s^{-1} and long catalyst lifetime of ~50 min at 800 °C. Correspondingly, the areal mass continued increasing up to ~8 mg cm^{-2} in 60 min (Fig. 1b). It was found that Gd layer with the thickness of less than 1 nm is effective when it is deposited between Fe and Al layers. The Raman spectra showed the radial breathing mode (RBM) peaks from the top to the bottom of the CNT forests, which suggests the continuous growth of SWCNTs.

References

[1] W. Cho et al., *Carbon* **72**, 264 (2014).