Development of Catalyst Metal/Catalyst Underlayer Systems for a Highly Efficient Synthesis of Vertically-aligned Array of Single-walled Carbon Nanotube

Shunsuke Sakurai,[1]

[1] *CNT-Application Research Center, National Institute for Advanced Industrial Science and Technology, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan*

For the synthesis of long and vertically-aligned array (forest) of SWCNT, the densely-packed (> 10^{11} cm^{-2}) and small (c.a. 3 nm) catalytic matal nanoparticle should be formed in prior to the synthesis, and maintained for a long syntheis time (typically >10 min) at high temperature (> 700 ºC). To overcome the above strict requirements and achieve mm-tall SWNCT forest, iron (Fe) catalytic metal on alumina (Al_2O_3) underlayer was used as a common factor in the previous reports [1,2].

In this study, various metal oxide systems had been used as a catalyst underlayer, and the requirements for catalyst underlayer system to enhance the SWCNT synthesis had been investigated. First, we have shown an efficient synthesis of SWCNT forest using magnesia (MgO) underlayer as Al_2O_3 underlayer [3]. Through further study investigating the correlation between the SWCNT growth efficiency and the annealing temperature of the MgO underlayer, the importance of control on two diffusion processes, ‘on-surface’ diffusion (such as Ostwald ripening) and ‘subsurface’ diffusion has been suggested [4].

Therefore, we desinged and examined the double-layered catalyst underlayer systems to control two diffusion processes independently. First, pre-annealed layers of Al_2O_3, MgO, hafnia (HfO_2), or zirconia (ZrO_2) were prepared as ‘bottom’ underlayers, and capped with a thin (c.a. 1 nm) and as-sputtered Al_2O_3 or MgO as ‘top’ layers. Such systems examplified by $Fe/Al_2O_3/HfO_2$ enables an improved efficiecy (6.4 vs 2.6 mg/cm^{-2} for 30 min synthesis time) compared with a conventional Fe/Al_2O_3 system.

References:

[1] S. Sakurai, H. Nishino, D. N. Futaba, S. Yasuda, T. Yamada, A. Maigne, Y. Matsuo, E. Nakamura, M. Yumura, and K. Hata *J. Am. Chem. Soc.* **134**, 2148 (2012).

[2] S. Sakurai, M. Inaguma, D. N. Futaba, M. Yumura, K. Hata *Small* **9**, 3584 (2013).

[3] T. Tsuji, K. Hata, D. N. Futaba, and S. Sakurai, *J. Am. Chem. Soc.*, **138**, 16608 (2016).

[4] T. Tsuji, K. Hata, D. N. Futaba, and S. Sakurai, *submitted*.

Email: shunsuke-sakurai@aist.go.jp