Revisited Roles of Bimetallic Catalysts for Controlled CVD Growth of Single-Walled Carbon Nanotubes

S. Maruyama (Advanced Industrial Science and Technology, The University of Tokyo), R. Xiang, H. An, Y. Qian (The University of Tokyo), K. Cui (Massachusetts Institute of Technology, The University of Tokyo), A. Kumamoto, T. Inoue, S. Chiashi (The University of Tokyo), and Y. Ikuhara (Japan Fine Ceramics Center, The University of Tokyo)

Abstract Text:

Bimetallic catalysts such as Co-Mo have been used for efficient growth of vertically aligned single-walled carbon nanotube (SWNTs) for a decade [1]. Recently, different kinds of bimetallic catalyst such as Co-W [2] or Co-Cu [3] are employed for structure controlled growth of SWNTs. Different roles of bimetallic catalysts are revisited by newly proposed in-plane transmission electron microscopy (TEM) technique, which enables a direct TEM characterization of catalysts and CVD grown nanotubes on SiO2 TEM grid. Sputtered and dip-coated cobalt-based catalysts, i.e., Co, Co-Mo, Co-Cu, Co-W are used in alcohol catalytic CVD (ACCVD).

We found the Co oxide catalysts can efficiently grow narrower diameter and smaller average diameter SWNTs compared with pre-reduced Co catalysts. The in-plane TEM and X-ray photoelectron spectroscopy (XPS) reveal that Co catalysts are transformed to Co3O4 after reduction-calcination process and then decompose to CoO before growth at a typical growth temperature (800 ºC) in Ar atmosphere. We conclude that an in-situ reduction process occurred on low-mobility CoO after the introduction of ethanol is essential to activate small metallic Co catalysts. The in-plane TEM studies confirmed that Co-Mo bimetallic catalysts have the same mechanism as oxidized Co system, i.e. low mobility oxides and reduced metal, consistent with our previous report [1].

By using Co-Cu catalysts, we can synthesize vertically aligned SWNTs with subnanometer diameters on quartz (and SiO2/Si) substrates [3]. Scanning transmission electron microscopic energy-dispersive X-ray spectroscopy (EDS-STEM) and high angle annular dark field (HAADF-STEM) imaging of the Co/Cu bimetallic catalyst system showed that Co catalysts were captured and anchored by adjacent Cu nanoparticles, and thus were prevented from coalescing into larger size, which contributed to the small diameter of SWNTs.

High-melting point W6Co7 alloy is reported to grow a single chirality, (12,6) with over 90 % abundance through high-temperature (1030 ºC) reduction and growth [2]. Here, we show that a sputtered Co-W catalyst can selectively grow (12,6) SWNTs by CVD at lower temperatures. Statistical Raman mapping analysis and optical absorption spectrum of the as-grown SWNTs reveal that the abundance of (12,6) is over 50%. The morphology and structure of catalyst is investigated by the in-plane TEM, which discloses the complicated structure changes before and after growth.

References:


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Submitter’s E-mail Address: maruyama@photon.t.u-tokyo.ac.jp
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First Corresponding Author
Dr. Shigeo Maruyama
Affiliation(s): Advanced Industrial Science and Technology; The University of Tokyo
Address: 7-3-1 Hongo, Bunkyo-ku
Tokyo, 113-8656
Japan
Phone Number: 81-3-5841-6421
E-mail Address: maruyama@photon.t.u-tokyo.ac.jp

Second Author
Dr. Rong Xiang
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: xiangrong@photon.t.u-tokyo.ac.jp

Third Author
Ms. Hua An
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: anhua@photon.t.u-tokyo.ac.jp

Fourth Author
Mr. Yang Qian
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: qian@photon.t.u-tokyo.ac.jp

Fifth Author
Dr. Kehang Cui
Affiliation(s): Massachusetts Institute of Technology; The University of Tokyo
Phone Number: 
E-mail Address: cui@photon.t.u-tokyo.ac.jp

Sixth Author
Dr. Akihito Kumamoto
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: kumamoto@sigma.t.u-tokyo.ac.jp

Seventh Author
Dr. Taiki Inoue
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: inoue@photon.t.u-tokyo.ac.jp

Eighth Author
Shohei Chiashi
Affiliation(s): The University of Tokyo
Phone Number: 
E-mail Address: chiashi@photon.t.u-tokyo.ac.jp
Ninth Author

Dr. Yuichi Ikuhara

Affiliation(s): Japan Fine Ceramics Center; The University of Tokyo

Phone Number:

E-mail Address: ikuhara@sigma.t.u-tokyo.ac.jp

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