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CONTACT (NAME ONLY): Suguru Noda

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Abstract

TITLE: A simple combinatorial method to discover Co-Mo binary catalysts that grow vertically aligned single-walled carbon nanotubes.

AUTHORS (ALL): Noda, Suguru [1]; Sugime, Hisashi [1]; Osawa, Toshio [1]; Tsuji, Yoshiko [1]; Chiashi, Shohei [2]; Murakami, Yoichi [2]; Maruyama, Shigeo [2].

INSTITUTIONS (ALL): 1. Department of Chemical System Engineering, The University of Tokyo, Tokyo, Japan.
2. Department of Mechanical Engineering, The University of Tokyo, Tokyo, Japan.

ABSTRACT BODY: Single-walled carbon nanotubes (SWNTs) are attracting much attention owing to their unique properties and potential applications. To establish their fabrication process, preparation of metal catalyst nanoparticles is a key issue, from which SWNTs grow during catalytic chemical vapor deposition (CCVD). Considering possible structural changes in catalyst nanoparticles on substrate surfaces at high temperatures during CCVD, we have proposed to utilize nanoparticles spontaneously forming from submonolayers of catalyst metals at the CCVD temperatures. By using our combinatorial method [1], we have discovered in a single experimental run that 0.05-0.1-nm-thick Co on SiO2 spontaneously forms Co nanoparticles that grow high-quality SWNTs during alcohol CCVD (ACCVD [2]) [3].
In this work, we extended our combinatorial method to binary catalysts and applied to the Co-Mo catalyst, which we previously prepared by dip-coating and found so active as to grow vertically-aligned SWNTs [4]. A mask with a slit simply set above a substrate during sputter-deposition yields thickness profiles of deposits in one-dimension perpendicular to the slit. We prepared thickness profiles of Mo (0.2- 4 nm) and Co (0.2- 8 nm) orthogonally on a SiO2/Si wafer. ACCVD changed the color of the catalyst wafer remarkably. By micro-Raman scattering measurement, we confirmed the formation of SWNTs and quantified their relative yield at 100 different points. When the nominal thickness of catalyst metals were converted into atomic concentrations, an interesting tendency was found; the relative yield was high for points with Co concentration slightly higher than the Mo concentration. This tendency is consistent with the model previously proposed [5] that the CoMoOx layer is formed and then the residual Co forms Co nanoparticle catalysts at a high number density on this layer. Then we analyzed the catalyst point with 1.5 nm Co and 1.4 nm Mo in more detail. Transmission electron microscope showed that the products were mainly SWNTs with some double-walled carbon nanotubes and amorphous carbon. Scanning electron microscopy revealed that the SWNTs were vertically aligned and formed a 2-um-thick film.
To realize SWNT-based devices, SWNTs should be grown under widely varied conditions and proper catalysts should be chosen for each condition. Our combinatorial method, which can yield an exhaustive catalyst library on a substrate, will accelerate the development of SWNTs growth processes.

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