東京大学グローバルCOEプログラム　機械システム・イノベーション国際拠点

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Parametric Studies towards Controlled Floating Catalyst CVD Synthesis of SWCNTs

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要旨

We review our current understanding of floating catalyst CVD synthesis of SWCNTs from CO using Fe catalyst clusters made via direct evaporation using hot wire generator as well as via thermal decomposition of ferrocene, and with the addition of trace amounts for CO_2 and water vapor. Here both the tube diameter as well as length can be tailored by changing the reactor temperature profile as well as CO_2 concentration. (n,m) distribution is biased towards large chiral angles with the maximum population at about 23 degrees. Then we proceed to explore the effect of carbon source gas, by adding C_2H_4 together with CO and looking at the effect of temperature when producing catalysts via ferrocene decomposition. Also, we present results when using C_2H_2 alone as a carbon source with H_2 carrier gas, and look at the effect of CO_2 addition.

To study the effect of Fe catalyst cluster size and concentration in the floating catalyst synthesis, we have developed a novel catalyst particle production method via physical vapor deposition, based on arc discharge between two electrodes i.e. the spark generator. This methods allows to control both the catalyst particle size and concentration when fed into the floating catalyst SWCNT synthesis reactor. Here we use the aerosol methods i.e. gas phase particle size measurements to observe in real time the changes of the produced tubes. Preliminary results show that when reducing catalyst particle gas phase number concentration, the bundle size of the produced tubes also is reduced.

To further understand the growth mechanisms, we have carried out parallel studies on SWCNT growth from carbon monoxide (CO) using supported CVD methods, both at ambient CO pressure in the in-situ Raman microscope as well as at 7 mbar pressure inside the dedicated, Cs-corrected environmental TEM (ETEM). When using supported bimetallic Fe-Cu catalysts, narrow chiral distribution SWCNTs were produced at ambient pressure growth. In addition, epitaxial formation of cobalt (Co) nanoparticles from $Co_xMg_{1-x}O$ solid solution reduction (when deposited into MgO via impregnation) in CO enables to grow SWCNTs also with a narrow diameter distribution. (ETEM) studies at reduced pressure reveal that the Co nanoparticles remain in metallic state and their epitaxial contact with MgO support remains coherent during SWCNT root growth process. Interestingly, when depositing Co via atomic layer deposition (ALD) onto MgO surface, we observe tip growth of SWCNTs inside the ETEM at 700 ^oC, with the Co catalyst nanoparticle shape as well as the tube growth direction fluctuating during the growth process. Also, then we have broader (n,m) distribution at ambient pressure growth conditions.

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