Predicting the Chiral-selectivity of SWCNTs on Bimetallic Catalyst Surfaces Using DFT Calculations

Debosruti Dutta[1], R. Mohan Sankaran[2], and Venkat R. Bhethanabotla[1]
[1]Department of Chemical & Biomedical Engineering,
University of South Florida
[2]Department of Chemical Engineering
Case Western Reserve University
e-mail: debosruti@mail.usf.edu

***Abstract*— The inability to grow single-walled carbon nanotubes (SWCNTs) with a specific chirality has been the biggest hindrance in their widespread usage in the micro-electronics industry. Chirality of a SWCNT defines its atomic-scale structure and is the single governing factor for the nanotubes to exhibit metallic or semiconducting properties. Theoretical and experimental studies have suggested that an epitaxial relationship exists between the catalyst metal nanoparticle and as-grown SWCNT. To further explore this idea, we have recently developed a synthesis technique to prepare compositionally-tuned bimetallic catalysts for SWCNT growth that shows the composition of the catalyst changes the chirality distribution of the as-grown SWCNTs. We have used density functional theory (DFT) calculations to study the nucleation of specific nanotube chiralities, as observed in our experiments with NixFe1-x catalysts, on our model bimetallic catalyst surfaces. Tuning the catalyst composition of a bimetallic nanoparticle changes the average bond length, which in turn determines the growth preference of a SWCNT chirality by epitaxial matching. Our modeling efforts successfully explain our experimental observations for changes in chirality distribution of Ni and NixFe1-x catalysts. Other experimental results reported in the literature have suggested that Cu nanoparticles and faceted Fe surfaces under specific reaction conditions enhance the growth of metallic SWCNTs. We have extended our DFT modeling to predict the growth of various metallic and semiconducting nanotube caps on NixCu1-x surfaces and various faceted Fe surfaces in an effort to explain previously reported experimental results. We have shown that nanotubes are stabilized at the nucleation stage by specific structures, i.e. compositions, of bimetallic catalyst surfaces.**