

Studies of SWNT bundling and growth mechanisms during floating catalyst CVD synthesis

Esko I. Kauppinen

Department of Applied Physics, Aalto University School of Science
Puumiehenkuja 2, P.O. Box 16100, FI-00076 Aalto, FINLAND

esko.kauppinen@aalto.fi

We report recent studies on the synthesis of high quality, narrow helical distribution single walled carbon nanotubes from CO with a ferrocene-based floating catalyst chemical vapor deposition (FC-CVD) reactor and show that SWNT networks consisting of long, clean and highly individualized SWNTs exhibit substantially improved transparent conducting film (TCF) performance [1]. Interestingly, the tube growth rate is reduced when increasing tube bundling during the synthesis. We explore the effect of CO₂ concentration on the (n,m) distribution as well as on the catalyst-to-tube diameter ratio, as determined by HR-TEM. In addition, we present recent studies on novel floating catalyst synthesis route for individual, i.e. non-bundled, small diameter single-walled carbon nanotubes (SWCNTs) with narrow chiral angle distribution close to armchair [2]. An *ex situ* spark discharge generator was used to form iron particles with geometric number mean diameters of 3-4 nm and fed into a laminar flow chemical vapor deposition reactor for the continuous synthesis of long and high-quality SWCNTs from ambient pressure carbon monoxide. The Raman G/D intensity ratios up to 48 and mean tube lengths up to 4 μm were observed. The chiral, i.e. (n,m) distributions, as directly determined by electron diffraction in the transmission electron microscope clustered around (7,6), (8,6), (8,7) and (9,6) tube species with up to 70% of tubes having chiral angles of 20° or greater. The tube mean diameter was reduced from 1.10 to 1.04 nm by reducing the growth temperature from 880 to 750 °C, simultaneously increasing the fraction of semiconducting tubes from 67 to 80%. Limiting the nanotube gas phase number concentration to ~10⁵ cm⁻³ successfully prevented nanotube bundle formation due to collisions induced by Brownian diffusion. Up to 60-80 % of the total of 500 as-deposited tubes observed by atomic force and transmission electron microscopy were individual. Tubes grow perpendicular to catalyst particle surface, based on detailed HR-TEM analyses of tubes and both active and non-active catalyst particles. We explore the fraction active catalyst particles based on detailed gas phase size distribution measurements of tubes and catalyst particles as well as on aerosol dynamics modeling.

[1] A. Kaskela et al. (2015) Submitted.

[2] K. Mustonen et al. (2015) Appl. Phys. Lett. **107**, 013106.