CATALYST SIZE AND CARBON FEED-RATE CONSTRAINTS FOR CARBON NANOTUBE SYNTHESIS

M. H. Rümmeli[1], F. Schäffel[1], M. Löffler[1], C. Kramberger[1], T. Gemming[1], P. Ayala[2], A. Grüneis[1], E. Mohn[1], B. Rellinghaus[1], R. Kaltofen[1], B. Büchner[1], T. Pichler[1]

1. IFW Dresden, P.O. Box 270016, D-01171 Dresden, Germany.
2. Departamento de Fisica, PUC-Rio, CP38071, 22452-970 Rio de Janeiro, Brazil.

m.ruemmeli@ifw-dresden.de

Systematic spectroscopic and microscopic studies on single-walled carbon nanotubes synthesized via laser evaporation and laser assisted chemical vapor deposition (LA-CVD) were conducted. Our findings show upper and lower diameter limits that represent a nanotube nucleation window, which can be attributed to catalyst particle volume to surface area constraints. However, the precipitating carbon saturated cluster size distribution depends solely on thermal aspects. SWCNT are only obtained when both the nucleation window and the catalyst size distribution crossover. The degree to which these two windows overlap establishes the diameter distribution of the obtained SWCNT[1]. Figure 1 illustrates the proposed model.

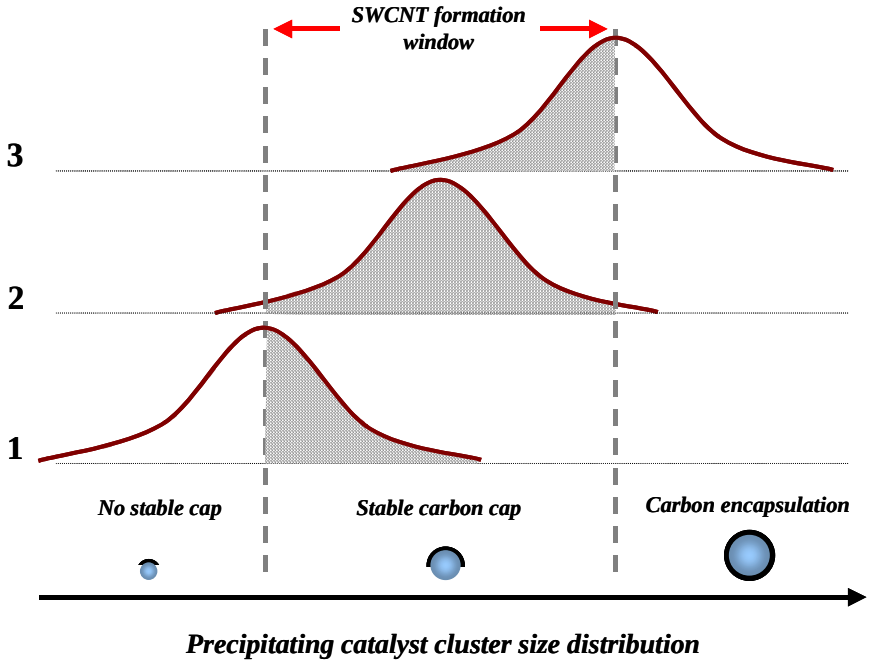

Figure 1. Proposed nucleation mechanism. The schematic shows the precipitating particle diameter distributions at three snapshots in time for 3 increased oven temperatures (1 to 3). Within the SWCNT formation window stable nucleation caps allow SWCNT growth. SWCNT are only formed in the overlap (shaded region) between the precipitating catalyst (size) distribution and SWCNT growth window. The degree of the overlap determines the SWCNT diameter distribution and mean diameter. Particles that are too large are encapsulated by graphite and particles that are too small are unlikely to form a stable cap.

Further the developed model points not only to catalyst geometrical constraints but also to the carbon feed rate determining if a nanotube can nucleate or not. LA-CVD pressure and feedstock flow rate experiments highlight the carbon feed-rate constraints.[2]

The same principles behind the proposed model can be applied to supported catalysts in CVD. Studies of ours in which we were able to deposit catalyst particles, as obtained from inert gas condensation, onto substrates in a controlled manner (size and density) enabled us to directly examine the catalyst particles prior to CVD synthesis and compare them with the resultant CNT in terms of their diameter and number of walls. The studies show that both the diameter and number of walls of the resultant CNT increase proportionally to the size of the catalyst particle. Figure 2 shows typical TEM images for catalyst (Fe) particles with different size distributions and their resultant CNT after CVD synthesis.[3] This behavior is fully consistent with catalyst volume to surface area constraints.

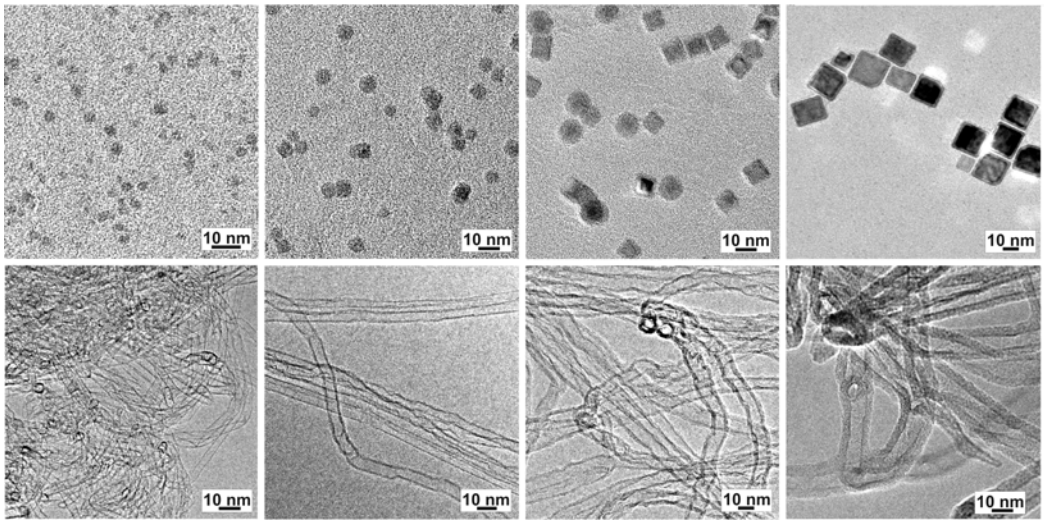

Figure 2. Upper panels: TEM images of the starting Fe catalyst particles. Increasing size from left to right. Lower panels, resultant CNT after CVD synthesis on $Si/SiO_2/Al_2O_3$.

In addition, our studies suggest that the root of a growing nanotube stems from the oxide support. Previous studies have shown that oxides typically used as supports in CVD can graphitize carbon in CVD synthesis conditions.[4]

References:

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