

In situ X-ray Photoelectron Spectroscopy Study of Catalyst-Support Interaction and growth of Carbon Nanotubes

Axel Knop-Gericke

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Dept. Inorganic Chemistry
Faradayweg 4-6, 14195 Berlin, Germany

The catalyst-support and catalyst carbon interaction during the chemical vapor deposition of single-walled carbon nanotubes was investigated by combining environmental transmission microscopy and in situ, time-resolved X-ray photoelectron spectroscopy. The behavior of Ni, Fe, Pd and Au model catalyst films on SiO₂ during preannealing in O₂ and NH₃ and during C₂H₂ decomposition was compared. The catalyst metal surface supplies sites to dissociate the hydrocarbon precursor and then guides the formation of a carbon lattice and the liftoff of a carbon cap. The catalysts are sharply distinguished by their reactivity towards activation of the hydrocarbon precursor, following trends known from heterogeneous catalysis. For Fe and Ni, the active state of the catalyst is a metallic nanoparticle. Graphitic networks do not form on oxidized Fe. Pd forms a silicide on SiO₂ under reducing conditions. Pd (silicides) and Au nanocrystals are catalytically less efficient in terms of precursor dissociation, while the low adhesion of C on Au surfaces impedes nanotube nucleation[1].

Fe²⁺ and Fe³⁺ interface states were detected for metallic Fe on Al₂O₃ in the absence of measurable Al reduction. This support interaction is much stronger than that on SiO₂, and it restricts Fe surface mobility. The resulting much narrower Fe catalyst particle size distribution on Al₂O₃ leads to a higher carbon nanotube nucleation density and a vertical nanotube alignment due to proximity effects [2].

References:

- 1 S. Homann et al. J. Phys. Chem C 2009, 113, 1648-1656
- 2 C. Mattevi et al. J. Phys. Chem C 2008, 112, 12207-12213