Molecular hydrogenation of carbon nanotubes - Evidence of spillover mechanism for hydrogen storage

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Motivation

- Molecular hydrogenation of SWNTs with catalyst to split the hydrogen molecule.
- Detection of C-H bonds formed in the process and prove “spillover” mechanism, for hydrogen storage.

Samples and methods of preparation

- As grown CVD samples - 2Å thick cobalt metal deposited on silicon oxide wafers as catalyst, Isopropanol as carbon source, growth temperature was varied from 700°C to 800°C.
- Commercially obtained HiPCO samples (Used only conductivity measurements) - SWNTs dispersed in isopropanol (1mg/10ml), sonicated for 15 minutes, and then spin cast on a quartz slide, annealed to get rid of solvent.
- Langmuir Blodgett Films (LB Films) - method of centrifugation to unbundle the nanotubes so as to have maximum catalyst coverage on SWNTs.
- Platinum (Pt) nanoparticles were sputter deposited onto respective samples amount deposited is calibrated as nominal thickness (amount of film deposited on a flat substrate).
- SWNTs diameter range 0.8nm-1.6nm for all samples

In-situ 4 probe Conductivity Measurements

- In-situ 4 probe current kinetic measurements with time was recorded as samples exposed to ~700torr molecular hydrogen.

X Ray Photoelectron Spectroscopy (XPS) Measurements

- Ex-situ XPS was performed on Pt-SWNT composites with Pt loading of 0.6nm, at elliptical undulator beam line 13-2, SSRL, with a Scienta R3000 analyzer with a resolution of 250meV.
- Peak shape before and after hydrogen exposure (~120psi, 8.25bar) was deconvoluted into components which correspond to sp2, sp3 peaks.
- Deconvolution gives rise to a 3rd peak at 3.1eV higher than sp2 peak which we attribute to decrease of the electric conductivity which can cause a reduction of the core hole screening, resulting in a ~3.3eV higher final state energy. The creation of a band-gap can give rise to a shake-up line.
- The relative weights of the sp1 (sp3) peak is 0.84 (0.17) for the LB film and 0.87 (0.13) for the as-grown film and hence, the weight percentage hydrogen uptake is 1.2wt% for LB films and 1 wt% for the as-grown films.

Future work

- X Ray Raman Spectroscopy (XRS) - A droplet cell would be used to measure K-edge XRS (inelastic scattering) during application of electrochemical potential. Hard X rays of 6-7KeV would be used for measurement.

Conclusion

- Achieved molecular hydrogenation under near ambient condition in Pt-SWNT composites with hydrogen storage capacity of up to 1.2wt%.

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