Laboratory Name: Savannah River National Lab
B&R Code: KC0201030

FWP and possible subtask under FWP:
Elucidation of Hydrogen Interaction Mechanisms with Metal-Doped Carbon Nanosturctures

FWP Number: LAB 04-20
Program Scope:
This research work is aimed at obtaining a better understanding of the nanoscale level of hydrogen sorption behavior of Metal-Doped Carbon Nanostructures MD-CNT. The experimental work is closely linked to relevant modeling studies of these materials. The program also involves investigating the thermodynamics governing formation of complex metal hydrides. The nature of the proton dynamics as well as the local structure was probed by NMR techniques while the phonon vibrations and the effect of the catalyst on the local lattice structure were investigated by incoherent inelastic neutron scattering (IINS) spectroscopy. Pressure effects to the structure of metal hydride materials were analyzed with high pressure XRD and Raman instrumentation.

Major Program Achievements (over duration of support):
We are interested in the interaction of CNT with H_2, and creating metal dopants with a narrow size distribution is non-trivial. An obvious solution to this is to find a way to study *one* nanotube at a time, so the size, metal dopant, etc. is very well known for that particular sample. In collaboration with A. Rao and Dr. J. Gaillard at Clemson University, we developed a method of attaching a single carbon nanotube, or a small array of nanotubes to act as a cantilever for harmonic detection of resonance (HDR). This is used to investigate the interaction of the materials with hydrogen and deuterium gases.
Ti, and a variety of transition metal additives have been investigated for enhancing the formation and decomposition of $NaAlH_4$. Carbon nanotubes mixed with $NaAlH_4$ were found to improve the hydrogen sorption properties of $NaAlH_4$. CNT with a variety of diameters were found to have similar behavior in reversing the hydrogenation of $NaAlH_4$. We also found that graphitic carbon alone will reverse the hydrogenation of $NaAlH_4$, though not as well as CNT. This is an intriguing result, and suggests that there is much to be learned about H_2 interactions with carbon surfaces and their catalytic role.
$NaMgH_3$ was used as a model compound to understand the formation of novel complex hydrides. An IINS vibrational spectra study at 10 K indicated a new finding that two phases coexist with disorder and two distinct hydrogen sites within the material. NMR studies discovered a captivating and unexpected phenomenon where the proton motion in $NaMgH_3$ is much faster than in either of the binary constituents. Structural studies of $NaAlH_4$ have also been conducted at elevated pressures in collaboration with FIU. A high pressure phase transition was discovered in un-milled $NaAlH_4$ at 6.35 GPa followed by amorphization at 13 GPa .

Program impact:
Provides insight into likely metal-carbon structures for hydrogen storage materials. And provides insight into the structures and interactions dynamics of H_2 with complex hydrides and their formation.

Interactions:
Internal: Weekly progress meetings, technical meetings as needed
External: Clemson University, University of South Carolina, Virginia Commonwealth U, LANL, WSTLU

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):
Invited plenary presentation - Dr. Polly Berseth presentation at ASME Energy Nanotechnology International Conference, September 2007.
Dr. Ashley Stowe - invited lecturer at the Los Alamos Neutron Summer School which focused on BES work.

Personnel Commitments for FY2006 to Nearest +/- 10%:
Ragaiy Zidan 35%
Puru Jena 10%
Polly Berseth (post-doc) 80%
Ashley Stowe (Post-Doc) 50%
Steve Serkiz 10%
Scott McWhorter 10%

Authorized Budget (BA) for FY00, FY01, FY02:
FY05 BA $300 **FY06 BA $300** **FY07 BA $ 300**