The Chemistry of Single Walled Carbon Nanotubes: Applications to biomolecule detection, nanotube separation, and electronic networks.

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Recent advances in the spectroscopy of single walled carbon nanotubes have significantly enhanced our ability to understand and control their surface chemistry, both covalently and non-covalently. Three distinct systems under investigation in our laboratory highlight these advances.

Molecular detection using near-infrared light between 0.9 and 1.3 eV has important biomedical applications because of greater tissue penetration and reduced auto-fluorescent background in thick tissue or whole-blood media. We have pioneered the use of carbon nanotubes as tunable near-infrared fluorescent sensors that are highly photo-stable. In one system, the transition of DNA secondary structure from an analogous B to Z conformation modulates the dielectric environment of the single-walled carbon nanotube (SWNT) around which it is adsorbed. The SWNT band-gap fluorescence undergoes a red shift when an encapsulating 30-nucleotide oligomer is exposed to counter ions that screen the charged backbone. We demonstrate the detection of the mercuric ions in whole blood, tissue, and from within living mammalian cells using this technology. Similar results are obtained for DNA hybridization and the detection of single nucleotide polymorphism. We also report the synthesis and successful testing of near-infrared β-D-glucose sensors that utilize a different mechanism: a photoluminescence modulation via charge transfer. The results demonstrate new opportunities for nanoparticle optical sensors that operate in strongly absorbing media of relevance to medicine or biology.

Covalent and non-covalent chemistries that are selective to single-walled carbon nanotubes of a particular electronic type have become increasingly important for electronic structure separation and on-chip modification of nanoelectronic devices. We have performed mechanistic studies to answer longstanding questions regarding their nature. Transient Raman and photoluminescence data collected in-situ reveal a series of two first-order reactions involving an adsorbed intermediate, and covalently bound product. The latter step can be deactivated by changing the structure of an absorbed surfactant phase, further supporting the mechanism. We discuss progress utilizing this chemistry to separate and sort single walled carbon nanotubes by their electronic structure using a 4-hydroxybenzene diazonium reagent.

Lastly, arrays and networks of metallic and semi-conducting carbon nanotubes are finding application in flexible electronics, as chemical and biological sensors, and as electronic interconnects. However, these systems demonstrate an environmental sensitivity and chemical reactivity that complicates processing, and facile incorporation into devices. We have studied the irreversible binding of adsorbates to such networks using electrical transport and Raman spectroscopy. The results are described theoretically using a continuum site balance model and mass action network involving the adsorbate-substrate interaction. All such devices in the literature and in our laboratory are shown to possess both reversible and irreversible binding sites that can be modeled independently. A simple analytical test is presented to distinguish between these two mechanisms from the transient conduction response of these arrays.


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2:00 – 3:00 p.m.
BOGGS ROOM 122
Refreshments will be served before the seminar.