INTERACTION OF [Fe,Fe] HYDROGENASES WITH SINGLE-WALLED CARBON NANOTUBES

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Single-walled carbon nanotubes (SWCNT) are promising candidates for use in energy conversion devices as active photo-collecting elements, for dissociation of bound excitons and charge-transfer from chromophores, or as molecular wires to transport charge. Hydrogenases are enzymes that efficiently catalyze the reduction of protons from a variety of electron donors to produce molecular hydrogen and may inspire new bio-hybrid materials for use in solar-driven fuel cells for direct conversion of sunlight into hydrogen. Here, we study changes in SWCNT optical signals upon addition of recombinant [FeFe] hydrogenases from *Clostridium acetobutylicum* and *Chlamydomonas reinhardtii*. We find evidence that novel and stable charge-transfer complexes are formed under conditions of the hydrogenase catalytic turnover, providing spectroscopic handles for further study and application of this hybrid system. The formation of a charge-transfer complex with hydrogenase sensitizes the nanotubes to the proton-to-hydrogen redox half-reaction. Thus, the experimental potential can be altered by changing the pH or molecular hydrogen concentration, and this tunability is utilized to study the effects of applied redox potential on the optical transitions of various semiconducting nanotubes in solution. (This work is supported by the U.S. Department of Energy, Office of Basic Energy Sciences.)