

PhD Thesis Defense: Electrochemical hydrogen storage in graphene oxide and light-induced dehydrogenation of graphane

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15:00

ICFO Mir-Puig Elements Room

Renewable electricity is inherently intermittent, and without large-scale storage, it would be difficult to fully displace the use of fossil-based fuels. Hydrogen is a promising energy storage alternative, owing to its high gravimetric energy density. However, its poor volumetric density limits its practical use. Compressed-gas, cryogenic, and metal-hydride approaches address this partially and at a substantial energy penalty. Graphane (C1H1)n, GA), a fully hydrogenated sp3 carbon lattice with a theoretical 7.7 wt% hydrogen capacity in C-H bonds, offers a promising long lasting and safe hydrogen storage. Provided that graphene is chemically inert to hydrogenation, in this thesis we consider a storage strategy based on

graphene oxide (GO), whose oxygen functional groups (OFGs) create reactive sites that enable hydrogenation under milder conditions. This thesis examines this GO-to-GA pathway as a single hydrogen storage strategy, constructing an experimental cycle of electrochemical hydrogen incorporation, scaled continuous-flow processing, and thermal or optical hydrogen recovery.

The core of the thesis begins in Chapter 2 by examining electrochemical and solar-assisted hydrogenation of GO films. Such electrochemical studies demonstrate that hydrogen incorporation into GO is facilitated by adequately tuning the electrochemical conditions and by the presence of OFGs. We also demonstrate that hydrogenation by periodic cyclic potential modulation outperforms fixed-potential reduction. A performance attributed to continuously exposing fresh OFG sites rather than favouring the competing hydrogen evolution reaction. Coupling a $BiVO_4$/FeNiOOH photoanode with a solar cell shifts the required bias cathodically, indicating hydrogenation may be feasible at substantially reduced voltage. A continuous-flow configuration is explored in Chapter 3 to address upscaling and hydrogenation of GO suspensions. Copper foam outperforms nickel as the cathode material due to its higher overpotential toward H_2 evolution, yielding rGO-H at cell potentials as low as 1.5 V with negligible gas evolution.

Chapter 4 discusses a photon-based desorption mechanism to probe the C-H desorption dynamics, exploiting selective photon-matched bond destabilization. In this chapter we used GA to better isolate sp^3 C-H desorption from O-bound hydrogens present in rGO-H. We demonstrate that 1.55 eV photons (800 nm) dehydrogenate only once supercontinuum generation is reached, causing extensive lattice fragmentation. When using 2.35 eV photons (527 nm), closely matched to the 2.28 eV C-H desorption barrier, we demonstrated dehydrogenation while preserving the carbon framework. This selective removal of the C-H bonds may trigger a self-propagating cascade reaction where the resulting sp^2 clusters destabilize and lower the desorption barrier for neighbouring C-H bonds, letting dehydrogenation proceed at a certain point, largely independent of the photon flux.

Collectively, these three chapters establish a solar-assisted electrochemical hydrogen loading of GO, while photon-based studies offer mechanistic insight into selective C-H bond breaking and cascade propagation, pointing toward controlled hydrogen release from GA.

Thursday December 3, 15:00 h. ICFO Mir - Puig Elements

Thesis Director: Prof. Dr. Jordi Martorell and Dr. Carles Ros

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