Title

Mechanistic studies of technologically-relevant dye-sensitized and cocatalyst-containing photocatalytic materials

Authors

Cardon, Joseph
Gaieck, William
Tkaczibson, Kevin
et al.

Publication Date

2019

Mechanistic studies of technologically-relevant dye-sensitized and cocatalyst-containing photocatalytic materials

Joseph Cardon, William Gaieck, Kevin Tkaczibson, Nazila Farhang, Sam Keene, Simon Luo, Hsiang-yun Chen, Shane Ardo

Presented At 2019 Fall National Meeting Aug 22-29, 2019

Abstract

Particle suspension reactors for solar water splitting can generate H_2 at a cost that is competitive with H_2 produced by steam methane reforming. Motivated by this fact, my group recently proposed a dual-bed photocatalyst reactor with stacked photocatalyst beds that enables increases in efficiency due to serial light absorption and short redox shuttle mass transport distances. In my presentation, I will share recent exciting results from detailed-balance efficiency calculations that show that reactor efficiencies benefit from an ensemble of optically thin materials instead of using a standard single-light-absorber geometry. Experimentally we have shown that state-of-the-art Rh-doped $SrTiO_3$ photocatalyst particles are more selective for H_2 evolution when Ru cocatalyst are used and I will share our latest discoveries on mechanistic and structural details of their effective operation from studies using pulsed-laser spectroscopies and aberration-corrected transmission electron microscopy.

Thin films of inexpensive metal-oxide semiconductors containing surface-bound molecular dyes can serve as low-cost and robust alternatives to silicon for indoor photovoltaic applications. Motivated by this fact, my group is incorporating electrocatalysts to drive multiple-electron-transfer oxidation of redox shuttles at the dye-sensitized photoanode. By decreasing the overpotential for redox shuttle oxidation, this design enables use of dyes that are weaker oxidants when oxidized, therefore extending dye absorption into the near-infrared spectral region and increasing projected solar-cell efficiencies from ~10% to beyond 20%. In my presentation, I will share our latest results from studies of molecular iodide oxidation electrocatalysis using a modified foot-of-the-wave analysis and experimental (transient absorption spectroscopy) and numerical (Monte Carlo) results that identify design rules for effective charge accumulation and electrocatalysis using dye- and cocatalyst-modified photoelectrochemical mesoporous materials.

Collectively these results provide design rules and additional research pathways for the study of composite materials that can serve as active components in techno-economically viable devices for solar energy conversion into electricity, or storage as H_2.