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(Invited) Understanding Redox Shuttle Photocatalysis in Z-Scheme Solar Water Splitting Reactors

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Abstract

Particle suspension reactors for solar water splitting are capable of generating hydrogen at a cost that is competitive with hydrogen produced from steam methane reforming.1-3 Our team has validated a reactor design that resembles Nature's Z-scheme where two stacked and connected photocatalyst particle suspension reactor beds together drive overall solar water splitting.3 Electron (and proton) management between the beds occurs by transport of a redox shuttle through a nanoporous separator. Efficient designs require that the redox shuttle is selectively oxidized and reduced at the particles that drive H2 evolution and O2 evolution, respectively. By device physics numerical simulations we showed that even for highly efficient reactor designs (10% STH efficiency) redox shuttle transport between the beds can be sustained with only passive diffusion.3

In my presentation I will report on our team's recent progress on this design. Using finite-element numerical analyses we modelled and simulated the transient mass transport processes, light absorption, electrochemical kinetics, gas crossover, and thermal transport in the proposed reactor. Experimentally, we synthesized, characterized, and evaluated the photo(electro)chemical performance of the most promising photocatalyst nanocrystallites (BiVO4, WO3, and Rh-doped SrTiO3) as mesoporous thin films and as particles in model reactors, and in the presence of several different redox shuttles and at various pH values. For H2-evolving Rh-doped SrTiO3, we demonstrated that in the presence of Fe(II) the limiting rate of Fe(III) reduction decreases and the rate of H2 evolution increases; however, these desired processes occurred along with undesired Fe(III) reduction and undesired H2 oxidation. Introduction of Ru cocatalysts enhanced performance by increasing the rate of H2 evolution and to a lesser extent undesired Fe(III) reduction. For O2-evolving WO3, we showed that O2 does not interfere with collection of electrons and that selectivity toward Fe(III) reduction is possible at moderate concentrations of Fe(III). Overall, results from several studies using a series of redox shuttles and photocatalyst particles will be presented.

Collectively, our efforts represent strides toward achieving a high-level of techno-economic viability in solar water splitting reactors.

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