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(Invited) Recent Progress in Fundamental Photoelectrochemical Studies Relevant to New Low-Cost Designs for 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. One reactor design resembles Nature's Z-scheme where two side-by-side and connected photocatalyst reactor beds together drive overall solar water splitting.1,2 The photocatalyst in each reactor bed also performs a half-reaction with a redox shuttle, i.e. oxidation or reduction, and therefore to prevent complete depletion of the redox shuttle, the electrolyte must transport between the beds via a nanoporous gas separator. While this design facilitates separation of the H2 and O2 reaction products, and therefore can circumvent formation of an explosive mixture of gases, transport of the redox shuttle between the beds requires active pumping which equates to additional capital expenditures that result in a near doubling of the reactor cost.1,2

Our team is evaluating the feasibility of new reactor designs where the beds are simply stacked. This generates a true tandem light-absorbing reactor where the theoretical maximum solar-to-hydrogen conversion efficiency is over 50% larger than a side-by-side or single light-absorber design. Each bed is projected to be < 10 cm tall and therefore, this design greatly decreases the distance required for redox shuttle transport, reducing or even eliminating the need for pipes, pumps, and forced convection.

In my presentation I will report on our team's recent progress on this design. We used finite-element numerical methods to model and simulate in two dimensions the transient mass transport processes, light absorption, and electrochemical kinetics in the proposed reactor. Model results suggest that a reactor operating at a ~4% solar-to-hydrogen conversion efficiency can operate indefinitely without complete loss of the active form of the redox shuttle at any location in the reactor beds. Experimentally, we are investigating and characterizing photocatalyst nanomaterials over many length scales, from single nanoparticles (~10 nm to ~1 μm in diameter) to mesoporous thin films (~10 μm thick) to laboratory-scale prototype particle-suspension reactors (on the scale of feet). On the single-particle level we jammed and covalently bound TiO2 nanoparticles into a single nanopore in a plastic sheet, wetted the particles with liquid electrolyte on both sides, and measured photovoltages that resulted from optical excitation of the particles. This was significant because it allowed, for the first time, in situ photoelectrochemical characterization of nanoparticle(s). We also synthesized, characterized, and evaluated the photo(electro)chemical performance of BiVO4, WO3, and Rh-doped SrTiO3 nanocrystallites as

mesoporous thin films and as particles in model reactors, and evaluated the transport properties of several redox shuttles. As a model system, H2-evolving Rh-doped SrTiO3 was evaluated as a mesoporous electrode and in particle form factor in the absence and presence of the ferric and/or ferrous iron redox shuttle. Based on this suite of results, we demonstrated that in the presence of FeII the limiting rate of reduction of FeIII is attenuated and the rate of catalysis for H2 evolution is enhanced. This is important because both halves of the iron redox shuttle are present in the reactor bed containing Rh-doped SrTiO3 and therefore this suggests that reduction of H+ can outcompete reduction of at least some FeIII under certain circumstances.

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

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