New Reactor Designs for Z-Scheme Solar Water Splitting Photocatalysis

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Abstract

Particle suspension reactors for solar water splitting can be an economical alternative to photovoltaic-driven electrolysis. One design resembles Nature's Z-scheme where two photosystems work in concert to drive overall water splitting. A conceptual Z-scheme reactor has been reported where two compartments on the meter length scale are adjoined side-by-side, each containing photocatalyst particles that drive one half-reaction of overall water splitting, and are connected by a nanoporous material that allows mixing of the liquid electrolyte.[1,2] Electronic charge is mediated between the compartments by a dissolved redox shuttle that undergoes oxidation or reduction at the particles. While this design facilitates product separation (i.e. separation of H_2 and O_2) and therefore circumvents formation of an explosive mixture of gases, active transport of the redox shuttle over these distances has been projected to account for about half of the capital cost of the reactor.[1,2]

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

In my presentation I will report on our team's 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. The developed model provided insights into the influence of the reactor geometry and operating conditions on the overall performance. The Beer–Bouger–Lambert law was applied to obtain the spatial light-intensity field and volumetric reaction rates were obtained by coupling solid-state photodiode expressions with Butler–Volmer kinetics on the surface of the particles. Model results suggested that a reactor operating at a ~1% solar-to-hydrogen conversion efficiency can operate for greater than half a year without complete loss of redox shuttle at any location in the reactor.

Experimentally, we investigated materials over many size scales, from single particles (~10 nm 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 used bipolar electrodeposition to create Janus-type particles consisting of model carbon particles with metal and metal-oxide electrocatalysts for H_2 evolution and O_2 evolution at the poles. We also jammed and covalently bound TiO_2 nanoparticles into a single nanopore in a plastic sheet, wetted the

particles with liquid electrolyte on both sides, and measured photovoltages that resulted from excitation of few particles. We also synthesized, characterized, and evaluated the photo(electro)chemical performance of $BiVO_4$ and Rh-doped $SrTiO_3$ nanocrystallites as mesoporous thin films and particles in model reactors, and evaluated the transport properties of several redox shuttles.

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

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