

Materials by Design Principles in Artificial Photosynthesis: Discovery and Synergistic Integration of Light Absorbers, Electrocatalysts and Membranes for a Complete, Stable, Efficient, and Safe Solar Fuels Generator

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Abstract.

We are developing an artificial photosynthetic system that will utilize sunlight and water as the inputs and produce hydrogen and oxygen as the outputs. We are taking a modular, parallel development approach in which three distinct primary components—the photoanode, the photocathode, and the product-separating but ion-conducting membrane—are fabricated and optimized separately before assembly into a complete water-splitting system. The design principles incorporate two separate, photosensitive semiconductor/liquid junctions that will collectively generate the 1.7-1.9 V at open circuit necessary to support both the oxidation of H₂O (or OH⁻) and the reduction of H⁺ (or H₂O). The photoanode and photocathode will consist of rod-like semiconductor components, with attached heterogeneous multi-electron transfer catalysts, which are needed to drive the oxidation or reduction reactions at low overpotentials. The high aspect-ratio semiconductor rod electrode architecture allows for the use of low cost, earth abundant materials without sacrificing energy conversion efficiency due to the orthogonalization of light absorption and charge-carrier collection. Additionally, the high surface-area design of the rod-based semiconductor array electrode inherently lowers the flux of charge carriers over the rod array surface relative to the projected geometric surface of the photoelectrode, thus lowering the photocurrent density at the solid/liquid junction and thereby relaxing the demands on the activity (and cost) of any electrocatalysts. A flexible composite polymer film will allow for electron and ion conduction between the photoanode and photocathode while simultaneously preventing mixing of the gaseous products. Separate polymeric materials will be used to make electrical contact between the anode and cathode, and also to provide structural support. Interspersed patches of an ion conducting polymer will maintain charge balance between the two half-cells.