Abstract

The development of robust membranes that govern fluxes of water, electrolyte ions, and half-reaction reactants and products throughout the microenvironment assembly comprising a solar fuels device is essential to the design of solar fuels devices. Critical science gaps in understanding how to achieve exquisite control of molecular and ionic transport in semipermeable soft matter must be bridged by embracing an interdisciplinary, cross-cutting approach that leverages multi-physics modeling, material synthesis, advanced characterization, and system integration. A key outcome is to understand how to design highly selective, semipermeable soft matter capable of rapidly transporting charge-carrying electrolyte ions between electrodes, which supports high device current density, while simultaneously suppressing crossover of half-cell reduction products, which enhances device efficiency by minimizing parasitic re-oxidation.

Introduction

Photoelectrochemical devices for water oxidation and CO2 reduction often utilize polymeric membranes, which must be:

• Conductive to electrolyte ions
• Relatively impermeable to CO2 reduction products
• Resistant to degradation by electrolyte, light, and heat

Designing materials that have all of these properties is challenging. Significant tradeoffs among these properties are commonly observed with traditional materials.

Recent Results and Critical Science Gaps

Membranes with high ionic conductivity are attractive because they generally support high current densities. However, membranes with high ion conductivities also often exhibit high permeabilities to CO2 reduction products, such as alcohol fuels. Significant CO2 reduction product crossover may jeopardize device efficiency, because reduced products may migrate to and be re-oxidized at the anode. Transport of both electrolyte ions and CO2 reduction fuel products is strongly correlated with membrane water content, frustrating design of membranes with high conductivity to electrolyte ions and low permeability to CO2 reduction products.

Bipolar membranes are of considerable interest because each of their two laminated layers promotes transport of ions of differing valences; we hypothesize that use of a bipolar membrane in a solar fuels device will enable tailoring of electrolyte environments to particular oxidation and reduction catalysts.

Perspectives on Membranes for Solar Fuels Devices

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Outlook

• Membrane ionic conductivity and permeability to CO2 reduction products can be tuned over a wide range by varying membrane functionality, but both are strongly correlated with water content, resulting in a tradeoff between ionic conductivity and CO2 reduction product crossover.
• New strategies to de-couple the transport of charge carriers and neutral CO2 reduction products are needed.
• While bipolar membranes enable the use of anolyte and catholyte at different pH values, new bipolar membranes with reduced resistivities must be developed for use in CO2 reduction devices.
• Membranes resistant to photo-degradation and chemical degradation (e.g., alkaline hydrolysis) will likely need to be developed for practical solar devices.

Acknowledgments

This material is based upon work performed by the Joint Center for Artificial Photosynthesis, a DOE Energy Innovation Hub, supported through the Office of Science of the U.S. Department of Energy under Award Number DE-SC0004993.

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