

Artificial photosynthesis: A pathway to solar fuels

Taking inspiration from nature and from the success of photovoltaic solar conversion, scientists are developing foundations for sunlight-driven synthesis of fuels, chemicals, and materials.

Harry A. Atwater



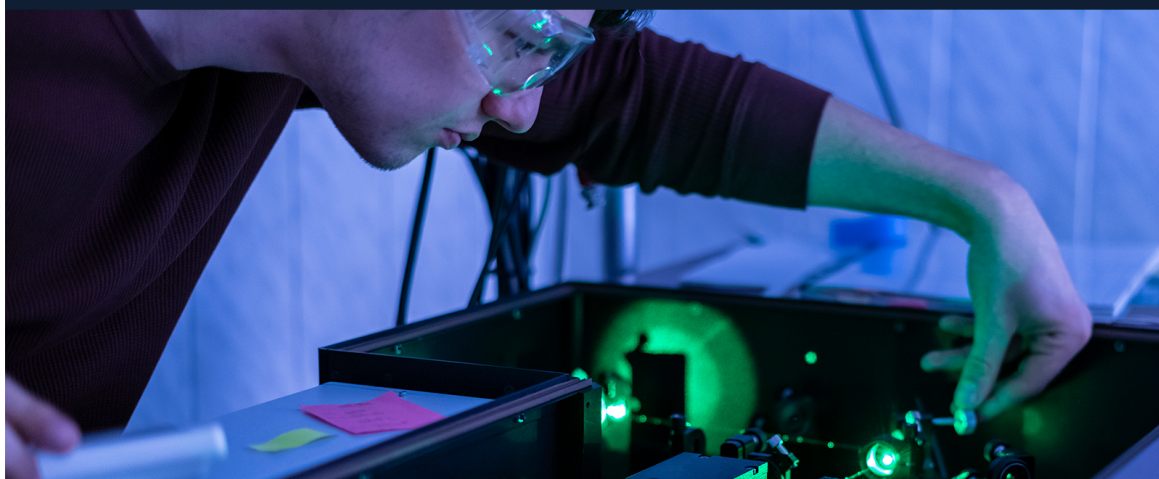
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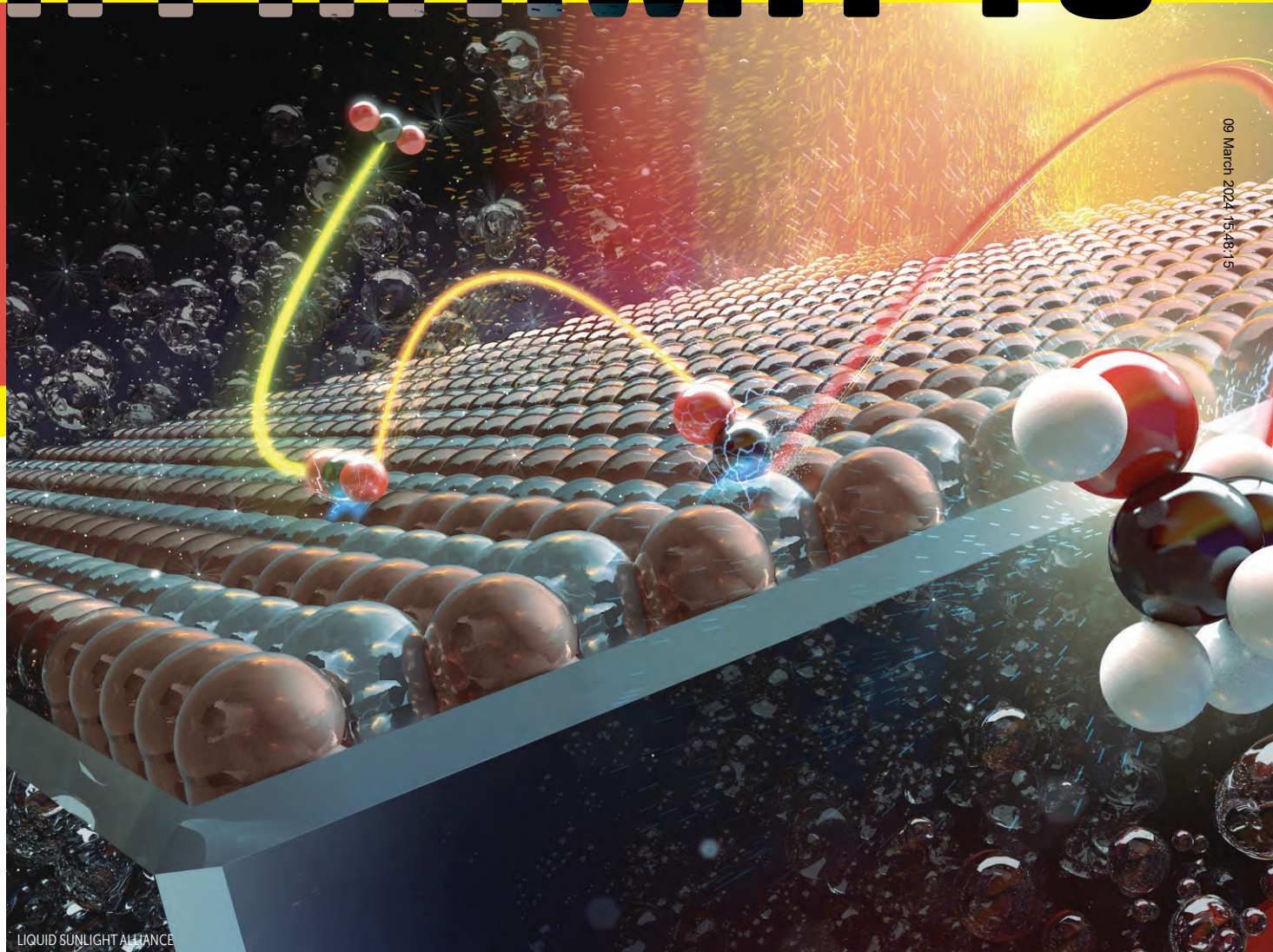
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ARTIFICIAL PHOTOSYNTHESIS: A PATHWAY TO



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SOLAR FUELS

Harry A. Atwater

Taking inspiration from nature and from the success of photovoltaic solar conversion, scientists are developing foundations for sunlight-driven synthesis of fuels, chemicals, and materials.

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The urgent need to address climate change has motivated an international effort to rapidly decarbonize our world's energy systems. In its most recent report,¹ the Intergovernmental Panel on Climate Change outlined the challenges we face. It articulates a two-part strategy: First, move away from reliance on fossil fuels as soon as possible, primarily by the electrification of large sectors of our economy. Second, capture carbon from the atmosphere to mitigate the warming associated with greenhouse gas emissions. During that effort, large sectors of the economy will likely remain difficult to decarbonize. For example, future long-haul aviation will continue to require hydrocarbon fuel.² For such sectors, a circular carbon economy will need to be established, in which the use of hydrocarbon fuels is balanced by carbon capture and conversion.

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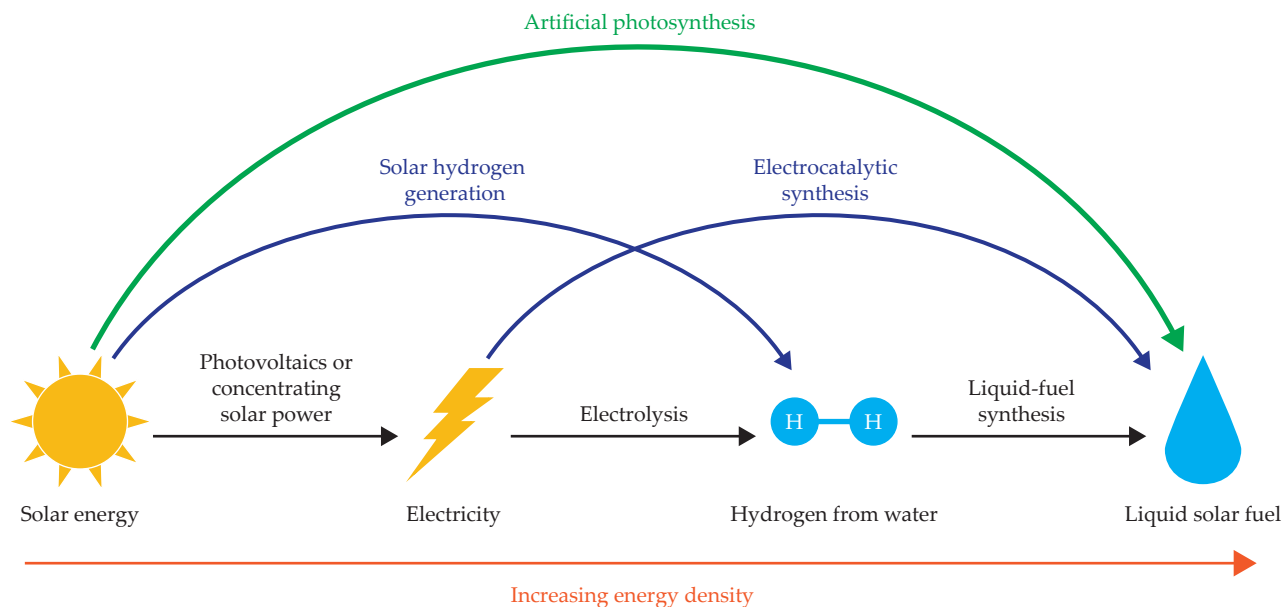


FIGURE 1. PATHWAYS for sunlight-driven chemical transformations of water and carbon dioxide into fuels. The green arrow indicating a direct pathway via artificial photosynthesis represents a single-step process for generating liquid-hydrocarbon and oxygenated fuels. The blue arrows indicate indirect pathways that can be combined to achieve liquid fuels. The processes include photoelectrochemical hydrogen generation, solar thermochemical hydrogen generation, photovoltaic or concentrating solar power for electricity production, electrolysis of water to make hydrogen, electrocatalytic synthesis of fuels using electricity, and liquid-fuel synthesis by chemically combining hydrogen and CO_2 . (Courtesy of Liquid Sunlight Alliance.)

Nature is remarkably adept at carbon capture and conversion through the photosynthesis of plants, algae, plankton, and other organisms that make up the biosphere. As scientists investigate new mechanisms for large-scale conversion processes to meet the needs of our energy transition, an important pathway to explore is that of artificial photosynthesis, which seeks to emulate nature's example by using engineered photoelectrochemical systems to synthesize solar fuels, chemicals, fertilizers, and other materials. Artificial photosynthesis thus has the exciting potential to create most of the chemical products required by our industrial civilization through using the ultimate source of abundant, renewable energy—our sun.

Energy-conversion landscape

Solar-fuel systems use photoexcitation, chemical transformation, and transport processes to produce fuel.³ A typical system includes light absorbers integrated with oxidation and reduction catalysts, membrane separators, and water-based electrolytes. Three central chemical reactions are involved in the artificial photosynthesis of carbon-containing products: the oxygen-evolution reaction, the hydrogen-evolution reaction, and the carbon dioxide-reduction reaction, as described in the box on page 35.

Each component must be designed so that the system efficiently uses the sunlight's energy to cause a reaction of water and CO_2 that produces fuel. The two primary types of solar-fuel systems are those generating H_2 as the fuel⁴ and those reducing CO_2 to gaseous, liquid, or oxygenated hydrocarbons, such as ethanol,⁵ as depicted by the green path in figure 1. Although they share common features, the two types have distinct advantages and challenges. The H_2 -generating system consumes only water and produces a single fuel (H_2), but it requires corrosive electrolytes to achieve high efficiency.

In natural photosynthesis, plants use sunlight to convert

water and CO_2 into sugars and carbohydrates. That process, however, is not efficient: Plants convert only about 1% of sunlight energy into stored fuel as plant biomass. Plants can also propagate themselves and use low CO_2 levels in the atmosphere. To be viable alternatives, nonbiological systems must achieve an efficiency of at least 10% by directly producing useful fuels rather than bulky biomass, because they cannot repair or replicate themselves.

Solar-fuel systems must be capable of operating outdoors for years, which requires that their active components be durable and withstand daily temperature and seasonal weather changes. Corrosion at the liquid–solid interface can be a problem caused by the acidic or basic conditions inherent in the electrolyte or generated by reactions at the electrode. Potential-dependent corrosion of the catalyst or light absorber can also occur.

The water-splitting process—the production of hydrogen gas from the decomposition of water—which is crucial for the systems, demands two catalysts: one for oxygen evolution, which produces O_2 molecules and protons, and another for hydrogen evolution, which assembles H_2 molecules from protons. Oxygen evolution usually occurs in either acidic or basic environments, but catalysts for the process tend to be unstable in highly acidic environments, except for those based on the rare precious metals iridium and ruthenium.

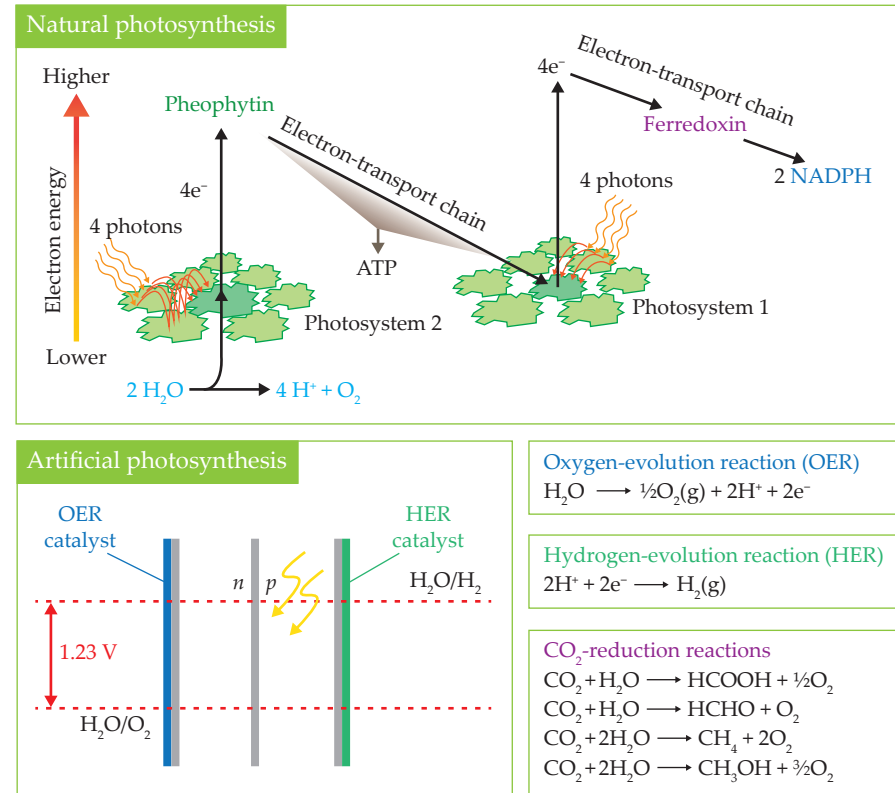
Storage and distribution of H_2 are challenging, because it must be carefully separated from the byproduct oxygen (O_2) gas to avoid forming an explosive mixture. On the other hand, CO_2 -consuming generators can operate with less-corrosive electrolytes and produce a fuel much like existing fossil fuels, such as natural gas and petroleum. That advantage allows engineers to leverage existing energy storage, distribution, and utilization infrastructure.

The reaction of CO_2 with water results in a mixture of different products and has low energy-conversion efficiency be-

Energy conversion in natural and artificial photosynthesis

Natural photosynthetic systems, such as chlorophyll, use two light-harvesting centers, photosystem 1 and photosystem 2, in a tandem configuration to create the chemical energy needed to produce adenosine triphosphate (ATP) and nicotinamide adenine dinucleotide phosphate (NADPH), two key sources of energy used by life at the cellular level. By analogy, efficient artificial photosynthetic systems like the one shown here can also use a tandem light-absorbing system consisting of two series-connected semiconductor photoelectrodes. The absorptions of photons (yellow) produce the photovoltage needed to drive the fuel-forming reactions—either water splitting (the production of hydrogen by the decomposition of water) or carbon dioxide reduction.

Alternatively, one can design a simpler but less efficient system (not pictured here) that uses a single semiconductor photoelectrode with a wider energy gap to absorb sunlight and produce the 1.23 V potential required for water splitting. Artificial photosynthetic systems also use either heterogeneous or molecular catalysts integrated onto the photoanode and photocathode surfaces to perform the oxidation and reduction reactions, respectively, needed



for fuel formation. Although photoelectrochemical water splitting has only two fuel-forming reactions, various pathways exist to produce fuel in CO₂ reduction, thanks to the richness of carbon chemistry.

Copper is the most important heterogeneous catalyst for CO₂ reduction be-

cause of its ability to produce multicarbon products. It typically produces them with little chemical selectivity, though. Thus a key challenge in solar-fuel systems designed for CO₂ reduction is achieving the required chemical selectivity to produce a single product with high yield rather than many products.

cause of low reactant concentrations and the complexity of the reaction mechanisms. The main technical challenges lie in achieving either high solar-to-fuel efficiency with multiyear durability or high product selectivity using CO₂.

Electrochemical conversion reactions can be coupled to sunlight indirectly—for example, by combining photovoltaics and electrolysis⁶—which optimizes the light-harvesting and catalytic processes that produce liquid fuels. They can also be directly coupled using integrated artificial photosynthetic systems, with the advantages those systems offer for integrated design, chemical control, and direct use of energy from sunlight.⁷

In many direct solar-fuel systems, bulk semiconductors absorb light and generate the photovoltage needed to drive fuel-forming electrochemical reactions. The photovoltage required to split water or drive the CO₂-reduction reactions at high (greater than 10%) solar-to-fuel efficiencies can be achieved using multiple photoabsorbers in a series-connected tandem configuration.⁸ With their exceptionally high photovoltages, multijunction III–V semiconductors have been a popular choice for more than two decades. Silicon, which is dominant in the microelectronics and photovoltaic industries, has also found extensive use. Some of the earliest work on photoelectrochem-

ical systems involved metal chalcogenides and chalcopyrite- and kesterite-type semiconductors because of their tunable bandgaps, which are ideal for solar-fuel generation.⁹

Computation, high-throughput synthesis, and data mining have accelerated the discovery and development of new semiconductor materials for solar fuels.¹⁰ Those semiconductors, however, still suffer from excessive corrosion in aqueous electrolytes when they're used as photocathodes, and that corrosion limits their durability.¹¹ Semiconductor photoelectrodes can be protected using inert overlayers such as titanium dioxide,¹² though flaws in the coatings can reduce their efficacy. The fundamental science underlying photocorrosion reactions and kinetics—and how to control them—is an important research area for artificial photosynthesis because of the need for durable materials, as I'll discuss later.

Water splitting

The first demonstration of direct solar-driven synthesis of fuels was reported in 1972 by Akira Fujishima and Kenichi Honda, who generated hydrogen by photoelectrochemical water splitting using a TiO₂ crystal as a semiconductor absorber.¹³ Their device also oxidized water on the TiO₂ surface and generated

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hydrogen gas with a separately wired platinum electrode. In 1983 Melvin Calvin, a chemistry professor at the University of California, Berkeley, published a groundbreaking paper establishing conceptual links between natural and artificial photosynthesis. In that work, he likened the components of a solar-fuel system to the components of biological systems containing micelles and vesicles.¹⁴

Over the past 15 years, researchers have made tremendous progress designing the architecture for photoelectrochemical, water-splitting solar-fuel generators. Modeling of solar-fuel prototypes has also been critical for guiding materials development because it defines the operational conditions and constraints for various cell designs. Researchers have used chemically selective membranes to develop strategies for stabilizing systems while ensuring greater than 10% solar-to-hydrogen conversion efficiency. The durability of photoelectrodes—in particular, their resistance to photo-induced degradation processes—was enhanced by the development of interfacial protection layers and hybrid composite coatings to prevent corrosion in acidic or basic electrolyte environments.

The advances in each of those areas, together with predictive models for design, have enabled researchers to make rapid progress increasing the efficiency of photoelectrochemical water-splitting devices. Figure 2 illustrates the result of that research: a device with a demonstrated 19% solar-to-hydrogen efficiency.¹⁵

Advanced water-splitting prototypes combine photonic design, transparent-catalyst synthesis, semiconductor band engineering, and protective antireflective coatings. Tailored multifunctional crystalline TiO_2 interphase layers provide corrosion protection, with favorable band alignment between the semiconductor conduction band and the energy level for water reduction, which facilitates electron transport at the cathode-electrolyte interface. Under a standardized level of simulated solar irradiation, solar-to-hydrogen efficiencies of 19.3% and 18.5% were obtained in acidic and neutral electrolytes, respectively. Such systems can now reach a value of 85% of the theoretical limit for photoelectrochemical water splitting.

CO₂ reduction

The architecture of the solar-fuel generator directly affects the product selectivity and activity for CO₂-reduction catalysis. In a water-splitting cell, the concentration of water is 55 molar; the reactant concentration thus poses no limitation on the reaction rate. But the low solubility of CO₂ in liquid-water-based electrolytes (30–40 millimolar, typically) constrains the reaction rate in a reduction system. Thus solar-fuel generators for CO₂ reduction typically use vapor-phase environments rather than liquid-electrolyte ones. Gas-diffusion-electrode (GDE) and membrane-electrode-assembly (MEA) architectures feature porous conductive membrane architectures like the one shown in figure 3; they can overcome not only the CO₂ solubility issue but also the inherent ohmic losses associated with liquid layers.¹⁶

Cells with liquid between the electrodes are labeled as diagnostic in figure 3 because they allow researchers to explore specific physical phenomena in laboratory experiments. To reach higher current densities and exploit the other benefits of vapor feed—for example, low water concentrations and the use of CO as an alternative reactant feed gas—high-efficiency cells are advantageous. Typically, MEA and GDE cells use a copper or silver heterogeneous catalyst, and the CO₂-reduction reac-

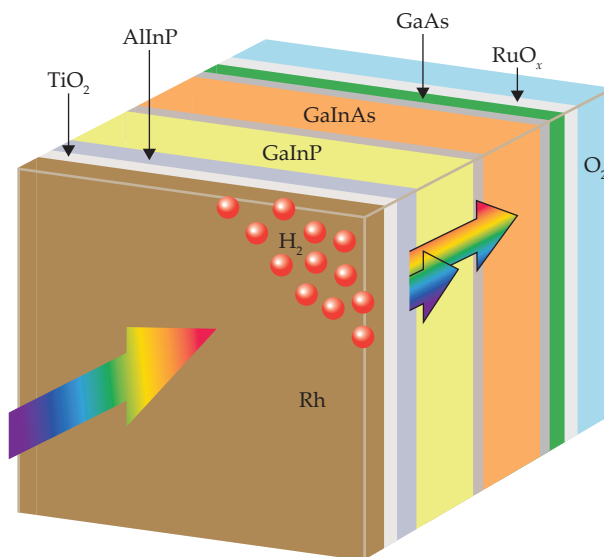


FIGURE 2. A SOLAR-FUEL GENERATOR splits water to produce hydrogen gas from sunlight at an efficiency that exceeds 19%. Sunlight strikes a front-facing tandem GaInP/GaAs photoelectrode that is functionalized with a rhodium catalyst to trigger the hydrogen-evolution reaction and with a ruthenium oxide catalyst to trigger the oxygen-evolution reaction. A titanium dioxide coating on the GaInP photocathode serves as a chemical protective layer and antireflection coating. The blue–green part of the solar spectrum is absorbed in the GaInP photoelectrode and the red-IR portion of the spectrum is absorbed in the GaAs photoelectrode. (Figure adapted from ref. 15.)

tion occurs at a gas-solid-liquid interface in the membrane layer. Such CO₂ conversion devices have been able to produce chemical products that are dominated by dicarbon-containing molecules, such as ethylene and ethanol, and high current densities are possible at relatively low cell potentials.

Integration of all those components in a structure as complex as a GDE can introduce significant variations in the electrode's activity, selectivity, and durability, even for well-studied catalysts such as copper. Mass-transfer effects and the local environment can lead to changes in the product distribution that could be incorrectly attributed to catalyst deactivation—a common concern in such studies. Further progress in developing next-generation architectures will likely require a combination of multiphysics modeling and measurements of local pH and reactant species under reaction conditions. Such modeling and experimental studies reveal the importance of the local chemical environment.

The voltage efficiency of a solar-driven electrochemical cell depends on the physical properties of its components—namely, catalysts, the electrolyte, and the membrane; operating conditions, which are the CO₂'s flow rate and pressure and the current density; and the cell's physical dimensions. To make progress designing complex reactors, scientists have developed multi-dimensional models for GDE and MEA electrochemical CO₂-reduction cells—more specifically, for cells that simulate electrocatalysis for CO₂ reduction and oxygen evolution, ionic transport, and chemical acid-base reactions in liquid- or ion-conducting polymer electrolyte layers.

The ion-transporting membranes in such devices are situated close to the gas-solid interface. Thus hydrating the membrane

with liquid water is an important concern because the liquid- and ion-containing exchange solutions help to lower the cell voltage. An important area for future research is to assess and evaluate the CO₂ utilization efficiency in those complex chemical environments. It can be low in the exchange solutions because of CO₂ pumping across the anion-conducting membrane.

Making liquid solar fuels from CO₂

Compared with water splitting, CO₂ reduction requires addressing a much more complex set of issues related to the transport and interfacial reactivity of reactants, intermediates, and products. When the solar-fuel community was primarily focused on water splitting, the appropriate scientific approach was to address mechanistic questions related to processes, such as reactions around a catalytic center or charge-carrier generation and transport. And researchers largely focused on processes occurring in a single sunlight-driven microenvironment, such as a photoabsorber-catalyst cathode interface.

Such approaches generated small-molecule gaseous fuel products, such as molecular hydrogen or carbon monoxide, for which small numbers of electron and proton transfers are needed. Most of the effort in the solar-fuels arena has previously addressed conditions in which concentrated feedstocks are present. They include liquid water as a source for generating hydrogen or pure CO₂ as a source for generating related products of CO₂.

The ultimate goal of direct solar generation of liquid fuels, such as multicarbon reduced products of CO₂ using dilute sources, however, is a far greater challenge that requires a conceptually different approach. To control the complex interplay of sequential or coupled transfers of multiple electrons, protons, and photons needed for liquid solar fuels requires a rethinking of the microenvironments that bring reactants to the reaction center.

Designing solar-fuel systems as assemblies of microenvironments is a new paradigm. Previous advances in solar fuels have led to success in building single microenvironments suited to water splitting and CO₂ reduction to two-electron products on the one hand and electrocatalysis to multicarbon products on the other. Until recently, though, the design of microenvironments for direct, efficient, and selective sunlight-driven generation of multicarbon or nitrogen-containing fuel products has been out of reach.

A new focus for solar-fuels research is to couple multiple microenvironments with an integrated or cascaded format—that is, to directly use sunlight to create liquid multicarbon fuel products in a precisely tailored manner and over a broad range of scales and ambient operating conditions. The microenvironment assemblies include photocatalysts, CO₂ capture media, membrane and porous media for ion and molecular transport, media for proton and cation management, and molecular and inorganic photo- and electrochemical reaction centers.

An example of such a coupled microenvironment architecture is illustrated in figure 4, which shows a tandem cascade of a photoelectrochemical reactor for CO₂ reduction to ethylene, coupled with a sunlight-driven thermochemical reactor that synthesizes ethylene-based fuel products. The mixture has a carbon concentration in the C₄ to C₁₇ range, which resembles the typical molecular weight distribution of jet fuel.

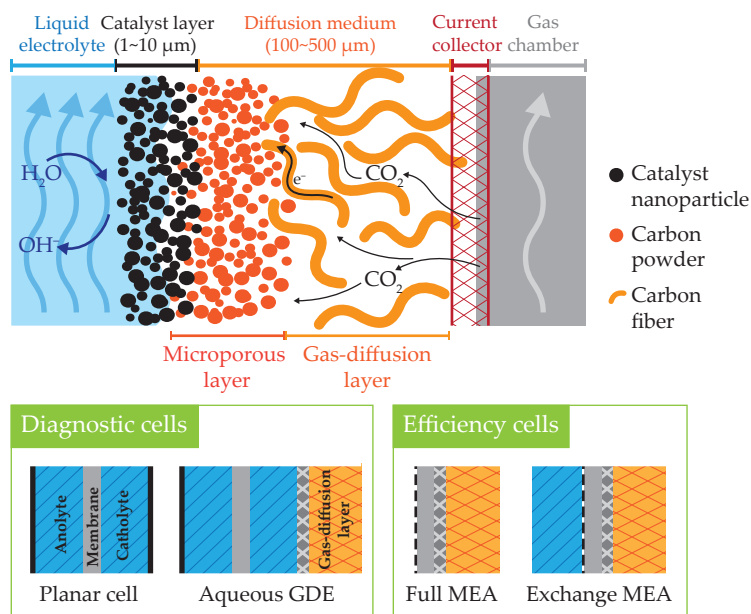


FIGURE 3. A GAS-DIFFUSION ELECTRODE (GDE) with different membrane-electrode-assembly (MEA) cell designs. The top panel depicts an aqueous liquid electrolyte that supplies protons to the catalyst, which is in contact with a porous gas-diffusion medium that facilitates the transport of carbon dioxide gas to the catalyst layer. The bottom panels illustrate (at left) a liquid electrolyte (planar) cell and a GDE used to characterize the voltage in the cell. At right are membrane-electrode assemblies, whose layer thicknesses and configurations are tailored to optimize the device efficiency. (Adapted from L.-C. Weng, A. T. Bell, A. Z. Weber, *Phys. Chem. Chem. Phys.* **20**, 16973, 2018.)

The goal is to demonstrate how the coupling of photoelectrochemical and photothermal reactors produces a liquid fuel from sunlight, CO₂, and water vapor. To date, that coupled microenvironment reactor has synthesized butene and hexene with high efficiency.

Selective synthesis of multicarbon products

A key requirement for complex product synthesis in solar-fuel systems is the precise synthesis of a single product with high selectivity and the suppression of minor or undesired products. Achieving that catalytic selectivity could in principle eliminate the need for separations, which is one of the costliest steps in any chemical synthesis process. A key advance in the past few years has been to apply molecular additive layers and ionomer coatings during the synthesis in order to tune the product distribution. The tuning is achieved, in turn, by controlling the availability of CO₂ and water at the reaction interface. Fortunately, molecular additives, such as pyridinium and Nafion ionomer coatings, can control the chemical microenvironment at the CO₂-reduction cathode interface.

Mechanistic studies are helping researchers understand how nitrogen-substituted pyridinium films can tune the rich profile of CO₂-reduction products on copper and silver electrodes. Silver surfaces catalyze primarily the CO₂-to-CO conversion in aqueous electrolytes, with the concomitant production of H₂ and a small amount of formate (HCOOH), depending on the potential that's applied. Whereas copper produces multicarbon products, silver typically produces only CO and H₂ in

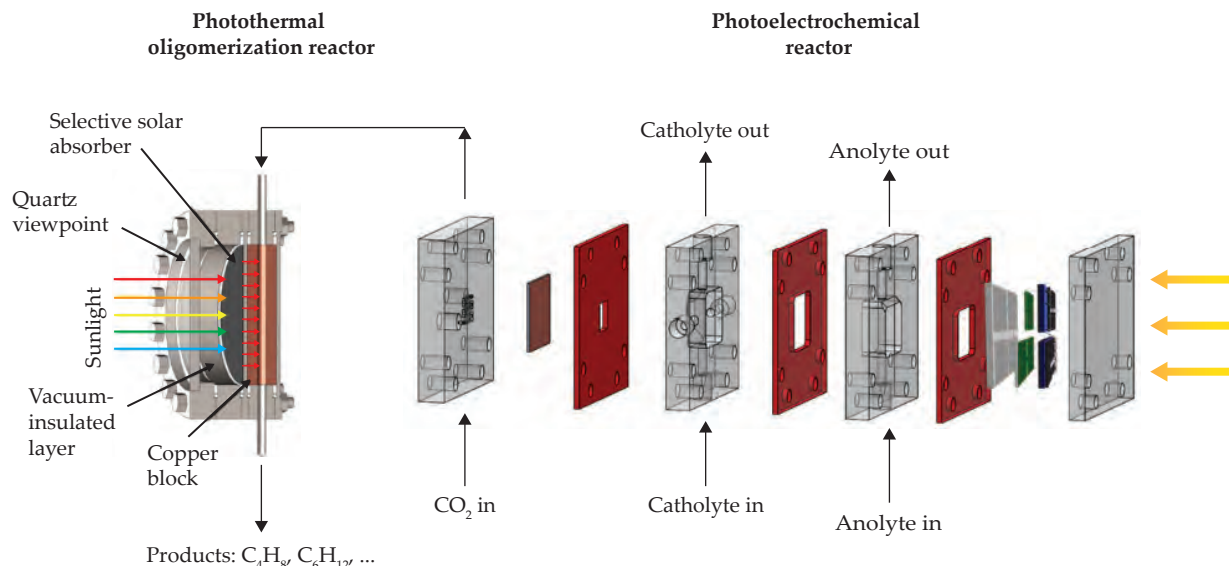


FIGURE 4. REACTORS IN TANDEM synthesize liquid fuel. This microenvironment features two reactors situated in a cascaded configuration: A photoelectrochemical reactor (at right) takes in sunlight to electrochemically reduce carbon dioxide to ethylene, and then a solar photothermal reactor (at left) uses harvested sunlight to heat a thermal heterogeneous catalyst, which then transforms the ethylene into alkene products. (Courtesy of Liquid Sunlight Alliance.)

electrocatalysis of CO_2 reduction. Silver thus offers a simpler system for understanding chemical processes.

Certain nitrogen-substituted pyridinium additives on silver foils produce CO with extremely high selectivity by inhibiting proton reduction—that is, hydrogen evolution—but not CO reduction. The data from electrokinetic studies suggest that hydrogen production was selectively inhibited by the growth of hydrophobic organic layers on the silver surface that limits proton transport but not CO_2 transport. Although the situation is more complex for CO_2 reduction on copper, researchers have found that selected pyridinium molecular additive films suppress hydrogen evolution and certain CO_2 -reduction products, such as methane, and favor the formation of ethylene and ethanol instead. The data point to proton transfer as the rate-determining step in catalysis instead of the commonly observed electron-transfer step, for example, in the case of planar silver electrodes.

Durability

The performance demands for generating liquid solar fuels are significant. They require that generators operate efficiently and stably for up to 100 000 hours under real-world conditions. Despite substantial research on artificial photosynthesis over the past several decades, a major bottleneck in designing long-lasting systems for liquid solar fuels is the durability of components. Corrosion is well recognized as a failure mechanism, but little is understood about the myriad other processes that can influence the photocatalyst or reaction-center microenvironment and thus drive changes in performance over time.

Researchers need to understand the fundamental chemical and physical mechanisms that cause degradation and compromise the function of photocatalysts, membranes, and coatings. If such an understanding could be achieved, a set of tools would likely emerge for designing components for both stability and performance in photo-driven microenvironments. Knowing the corrosion mechanisms, the operational limits of

durability, and details about the evolution of catalyst shapes will provide feedback for such models and enable predictive design of future systems.

Several approaches have been taken to understand and control corrosion in solar-fuel systems. Quantitative methods for studying corrosion rates were developed by detecting degradation products in the electrolyte or by measuring the mass loss of the component. *In situ* observation of corrosion processes by scanning probe techniques is also possible, and when combined with theory, it can reveal underlying mechanisms. Many anode and cathode materials are metastable under water-splitting or CO_2 -reduction conditions, and their resistance to corrosion can be incorporated as a selection criterion in computational and experimental searches for new materials for CO_2 -reduction photocathodes.

Photocorrosion is not the only process that can lead to performance loss. Photocathodes that drive CO_2 reduction usually incorporate a cocatalyst that favors CO_2 reduction over hydrogen evolution and controls product selectivity. Often the cocatalysts are metal nanoparticles; copper is used in many studies because it is the only metal catalyst that facilitates the C–C coupling reactions necessary to form C_{2+} products.

It is well known that nanocrystalline, copper-based CO_2 -reduction catalysts can change shape or sinter during operation and lose their selectivity. Identifying the mechanisms responsible for that performance degradation might allow researchers to design systems resistant to those effects. The corrosion mechanism involves coupled charge and atom transfer from a solid interface into solution. Theories have been extended to include proton-coupled electron transfer, which is central to water oxidation. They do not, however, address the processes that initiate corrosion in the first place or help identify corrosion-resistant materials.

Another major gap in the science of solar fuels and electrochemical systems is understanding the rare events that cause material instability or passivation. During macroscopic corro-

sion, the rare events occur orders of magnitude faster than researchers can observe them. To correct that problem, researchers are starting to explore model systems with *in situ* time-resolved experiments.

A path to scaling up

Engineers have now developed photovoltaic energy systems up to the terawatt scale, and their manufacture is supported by comprehensive supply chains, distribution networks, and a worldwide infrastructure. An important question is whether solar fuels can follow a similar trajectory to large-scale production. Most conceptual work on the scalability of solar-fuel systems has focused on water splitting. To date, solar-fuel researchers have developed prototype systems at the 100 m² scale¹⁷ and produced a detailed description of a hypothetical 1 GW hydrogen plant that generates 220 000 metric tons of H₂ per year with a photoelectrochemical generator design.¹⁸

Materials, initial energy requirements, and estimates of the annual net energy for the plant came from an initial study. A key finding was that the most significant factors affecting the energy returned on energy invested are the solar-to-hydrogen efficiency and panel lifetime. That study and similar ones point to the importance of ensuring high efficiency and a long lifetime for sustainable photoelectrochemical hydrogen production.

Significant advances have been made in solar-fuel research in the past decade for both water splitting and CO₂ reduction. Although no manufacturing industry currently exists, solar fuels have the potential to contribute to a sustainable circular carbon economy in a climate-stable world. Researchers in the field can take inspiration from the example of photovoltaics,

which has undergone a millionfold expansion in production capacity over the past 30 years. If many of the same manufacturing methods and infrastructure resources that are now available for photovoltaics can be reused for solar fuels, they may offer a pathway for rapid acceleration of this important renewable-energy technology.

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