

## REVIEW SUMMARY

## SOLAR ENERGY

# Research opportunities to advance solar energy utilization

Nathan S. Lewis\*

**BACKGROUND:** Despite providing a relatively small percentage of total global energy supply, solar energy systems generally receive enthusiastic support from technologists, regulators, politicians, and environmental groups. The energy in sunlight can be converted into electricity, heat, or fuel. Although the costs of solar panels have declined rapidly, technology gaps still exist for achieving cost-effective scalable deployment combined with storage technologies to provide reliable, dispatchable energy.

**ADVANCES:** The costs of Si-based solar panels have declined so rapidly that panel costs now make up <30% of the costs of a fully

installed solar-electricity system. Research and development (R&D) opportunities hence lie in the development of very high efficiency conversion materials, to advantageously leverage the associated reduction in area-related balance-of-systems costs. Such materials would optimally either leverage or mate with existing, low-cost Si photovoltaic (PV) technology. Ultralightweight, flexible, robust, and efficient materials could also greatly reduce the installation costs and could allow for enhanced automation and inexpensive support structures.

The development of cost-effective persistent grid-scale storage to compensate for the intermittency of sunlight is a major area for

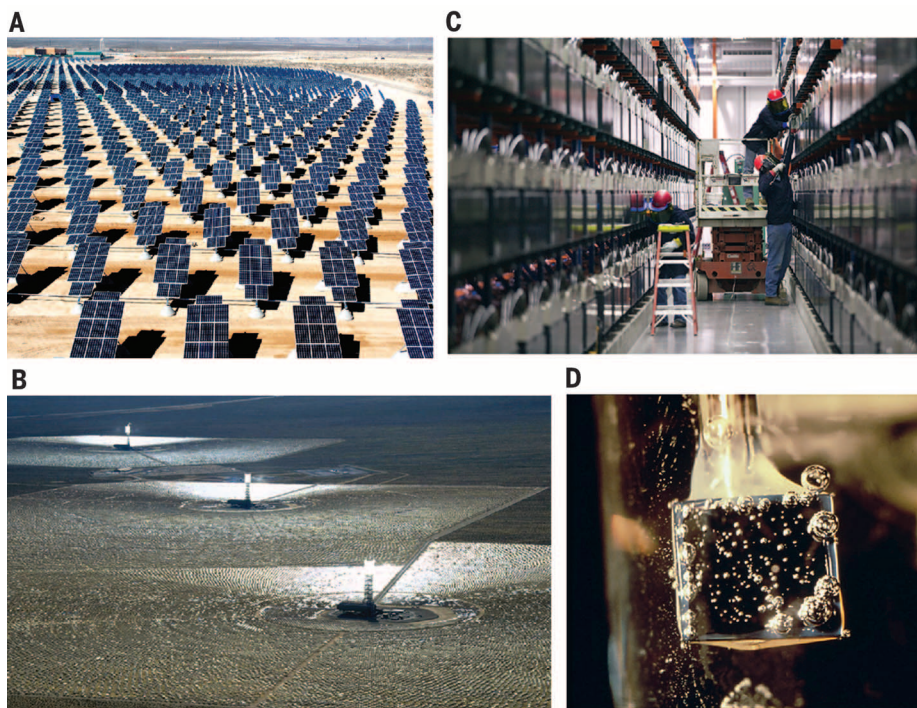
R&D. Possibilities include new types of batteries and flow batteries, as well as geologic storage of hydrogen, methane, or compressed air.

Opportunities also exist to improve the capabilities of concentrated solar power systems that convert sunlight into heat. Improved thermal storage fluids would provide longer-term storage to compensate for cloudy days in areas of high direct insolation. Thermoelectrics, in principle, could replace engines to provide efficient conversion systems that have no moving parts.

New thermochemical cycles could allow for the highly efficient, cost-effective conversion of solar heat into fuels by promoting endothermic reactions, such as water splitting, carbon dioxide reduction, or thermochemical conversion of feedstocks, such as methane to high energy-density liquid hydrocarbon fuels that are needed in the transportation sector.

Artificial photosynthetic systems that directly produce fuel from sunlight are in the proof-of-concept stage. Such technologies offer the potential to provide renewable hydrogen by solar-driven water splitting or to produce hydrocarbons directly from sunlight, water, and CO<sub>2</sub>. Key goals for R&D are development of materials that can absorb and convert sunlight efficiently that are seamlessly integrated with catalysts that promote the production of fuel, with the production of O<sub>2</sub> from water also required to complete a sustainable, scalable chemical cycle. Systems must simultaneously be efficient, robust, cost-effective, and safe.

**OUTLOOK:** Considerable opportunities for cost reduction that can achieve both evolutionary and revolutionary performance improvements are present for all types of solar energy-conversion technologies. Learning by doing and R&D will both be needed to produce an innovation ecosystem that can sustain the historical rate of cost reductions in PVs and concentrated solar thermal technology. Disruptive approaches to storage technologies are needed to compensate for the intermittency of sunlight and allow for development of a full clean-energy system. Solar fuels technology contains abundant opportunities for discovery of new materials and systems that will allow for deployable, cost-effective routes to the direct production of fuels from sunlight. ■



**Solar energy-conversion and storage technologies.** (A) Nellis Solar Power Station, a 14-MW PV installation at Nellis Air Force Base, NV. (B) Concentrated solar thermal power 392-MW installation at Ivanpah, CA. (C) World's largest battery (NiCd) storage installation (40 MW for 7 min, 26 MW for 15 min), Fairbanks, AK. (D) Solar fuels demonstration of a photoelectrode evolving hydrogen gas. [Image sources: (A) Nellis Air Force Base PV installation, [https://commons.wikimedia.org/wiki/Category:Nellis\\_Solar\\_Power\\_Plant](https://commons.wikimedia.org/wiki/Category:Nellis_Solar_Power_Plant). (B) Ivanpah solar electric generation installation, <http://i.ytimg.com/vi/M5ygzfCNpvM/maxresdefault.jpg>. (C) Fairbanks battery installation, <http://blog.gvea.com/wordpress/?p=1677>

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Major developments, as well as remaining challenges and the associated research opportunities, are evaluated for three technologically distinct approaches to solar energy utilization: solar electricity, solar thermal, and solar fuels technologies. Much progress has been made, but research opportunities are still present for all approaches. Both evolutionary and revolutionary technology development, involving foundational research, applied research, learning by doing, demonstration projects, and deployment at scale will be needed to continue this technology-innovation ecosystem. Most of the approaches still offer the potential to provide much higher efficiencies, much lower costs, improved scalability, and new functionality, relative to the embodiments of solar energy-conversion systems that have been developed to date.

**B**ecause of its unmatched resource potential, solar energy utilization has been the subject of intense research, development, and deployment efforts that have accelerated during the past decade (1). Efforts have focused on the development of photovoltaics (PVs) for production of solar electricity, on

conversion of solar energy into electricity or heat, and on artificial photosynthetic systems that directly produce fuels from sunlight. The dramatic increases in deployment and concomitant decreases in the cost of solar energy-conversion systems in the past decade attest to the importance of investments in innovation (1).

The cost-effectiveness of terrestrial solar energy systems is dictated by two fundamental constraints. First, as compared with fossil fuels or nuclear fission, the relatively low average terrestrial power density of sunlight, typically ~200 to 250 W/m<sup>2</sup>, requires very inexpensive materials and systems to cost-effectively cover the large areas needed to capture and convert solar power on a terawatt global scale (2, 3). Second, the intermittency of sunlight requires cost-effective energy-storage technologies to provide energy on demand with high reliability. This review provides an update on many of the developments that have occurred during the past decade (4) and identifies some of the promising opportunities for further research and development (R&D) in light of the present status and economics of solar energy-conversion technologies.

## Solar electricity Photoactive materials

Solar cells can be conveniently categorized on the basis of the type of light-absorbing material in the photoactive layer (Table 1). Devices based on crystalline silicon rely on a p-n junction formed through spatially directed doping of a

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**Table 1. List of PV materials and defining commercial and technical attributes.** n/a, not applicable.

| PV material                           | Maturity   | 2013 production (GWp) | Efficiency [best module (35), highest reported cell (7)]; attributes   |
|---------------------------------------|------------|-----------------------|--|
| CdTe                                  | Commercial | 1.9                   | 17.5%, 21.5%; thin film, sublimes congruently and enables monolithic module manufacturing  |
| a-Si:H                                | Commercial | 0.8                   | 12.3%, 13.6%; flexible modules when material is deposited onto stainless steel substrates; efficiency decays with time   |
| CuInGaSe <sub>2</sub>                 | Commercial | 0.8                   | 17.5%, 21.7%; requires stringent process control to maintain stoichiometry of four-element material over large areas   |
| Mono-Si                               | Commercial | 13.9                  | 22.9%, 25%; highest Si module efficiencies; implements technology for extensive control of bulk and surface recombination losses; high efficiency yields reduced area-related balance-of-systems costs   |
| Multi-Si                              | Commercial | 21.3                  | 18.5%, 20.8%; market leader  |
| Ribbon Si                             | Commercial | 0                     | Continuous instead of batch process to make the Si substrate   |
| GaAs                                  | Demo       | n/a                   | 24.1%, 28.8%; thin-film epitaxial layers require facile removal from the lattice-matched, expensive substrate; radiation tolerance and light weight are advantageous for space power applications        |
| Multijunction (high concentration PV) | Demo       | 0.05                  | 38.9%, 46%; high efficiency; limited to high locations with high direct normal irradiance, optimal performance requires complex dual-axis tracking and optical focusing                                  |
| Organic PVs                           | R&D        | n/a                   | 8.3%, 11.5%; readily processable, flexible cells; modest cell efficiencies; long-term decay of efficiency;   |
| Quantum dots                          |            |                       | n/a, 9.9%; potential for very high efficiencies through multiple exciton generation processes; growth of large single crystals not required  |
| Perovskites                           |            |                       | n/a, 20.1%; very rapid increase in demonstrated cell efficiency; stability unproven; soluble, toxic Pb salt; material dissolves in water   |
| Dye-sensitized solar cells            |            |                       | n/a, 11.9%; wet chemical processing of titania substrate followed by adsorption of dye; fabrication of cell requires sealing gel or liquid electrolyte; small improvement in efficiency over past decade |

planar silicon (Si) structure to effect charge separation and to allow for efficient production of photocurrent and photovoltage (Fig. 1) (5, 6). The cost of Si solar panels, measured in dollars per peak watt (\$/Wp) has decreased by ~20% for each doubling in cumulative global module production (2, 3). Commercially available Si panels, which accounted for ~90% of total solar panel production in 2013, now have an energy payback period of <2.5 years and ~16 to 21% power-conversion efficiencies (2, 3). The dramatic reductions in panel cost have been realized largely through sustained, systemic reductions in specific manufacturing costs, including those of the polymer encapsulant, the screen-printing of the silver electrical contacts, and even the production of the Si wafers themselves, along with economies of scale enabled by construction of very large panel-production facilities. Research opportunities to further lower costs include methods to integrate higher band-gap materials with Si to create a high-efficiency tandem device in a scalable, cost-effective process that is compatible with existing Si PV manufacturing methods.

Gallium arsenide and other “III-V” single-junction and multijunction PV devices (7–10) are highly efficient and used on satellites but are currently only considered cost-effective for terrestrial applications by combining small device areas with high-concentration-factor optics that utilize active solar tracking and optical concentrating systems. Opportunities exist for obtaining improved efficiencies through spectral-splitting approaches (11, 12), as well as through novel designs for both one-dimensional (1D) and 2D optical concentration and tracking systems and structures, as well as in development of new approaches to the low-cost growth of high-quality, high-performance III-V monolithic devices and structures.

In contrast to active materials based on single-crystal substrates, thin-film materials, such as cadmium telluride (CdTe), CuInGaSe<sub>2</sub>, amorphous hydrogenated Si, and organic PVs can provide flexible, lightweight modules that could result in reduced system installation costs. Engineered CdS/CdTe heterostructures provide control over junction recombination at the metallurgical interface (3). Toxicity concerns related to release of Cd into the environment have been raised, especially in Europe, but have been addressed by rigorous encapsulation of the active material, in conjunction with proposed panel-recycling programs (13). Scarcity issues related to the availability of Te may preclude scaling of CdTe PV technology to terawatt levels (14), but the lower CdTe module efficiencies of <15%, as compared with 16 to 21% efficiencies for Si panels, are presently more important considerations in determining the cost-competitiveness of the various PV panel technologies. Research opportunities involve grain-boundary passivation to allow thin films to exhibit the high mobilities and efficiencies of single crystals. Solar cells based on perovskites formed from lead salts with organic ammonium cations have demonstrated that extraordinary performance can be obtained using simple deposition techniques in novel materials systems

(7, 15–17). The long-term stability of these materials at the highest reported efficiencies remains to be established (17, 18). Intense efforts are currently being devoted to understanding the fundamental behavior of such systems, as well as to discover other similarly behaving classes of materials that are environmentally benign, do not release toxic lead ions upon dissolution in water, and combine efficiency with stability. Such considerations underscore the complex technological, political, and economic aspects associated with the development a sustainable, cost-effective solar energy-utilization system (1).

Earth-abundant light absorbers that could provide alternative materials options for solar absorbers, including Zn<sub>3</sub>P<sub>2</sub> (19, 20), ZnSnN<sub>2</sub> (21, 22), and Cu<sub>2</sub>O (23, 24), are receiving renewed interest after preliminary investigations in the early 1980s (25, 26). Research opportunities involve control over the bulk and surface properties of such materials to obtain high efficiencies, preferably in thin-film form.

The performance of organic PV cells, such as those based on composites of poly-phenylenevinylene (PPV) with a functionalized C60 that serves as the light absorber (27–30), in principle, can be systematically varied by chemical control over the composition and structure of the components of the device (29). Radical-related side reactions under visible and ultraviolet illumination of the organic materials that form the active components of the device structure must be minimized to obtain long-term stability while preserving high efficiency. Research opportunities for dye-sensitized solar cells include the development of molecular sensitizers that exhibit improved photovoltages, as well as enhanced stability, while retaining device efficiency (31, 32).

Quantum-confined systems, including inorganic quantum dots, can provide multiple electrons upon absorption of a photon having an energy greater than twice the band gap of the absorber (33). Organic materials exhibit an analogous process denoted as singlet fission (34). Both phenomena could form the basis for solar cells that exhibit efficiencies in excess of the conventional, Shockley-Queisser (S-Q), theoretical limit of 32% for a single-band gap material under unconcentrated sunlight (33). Research opportunities involve incorporation of this phenomenon into operational

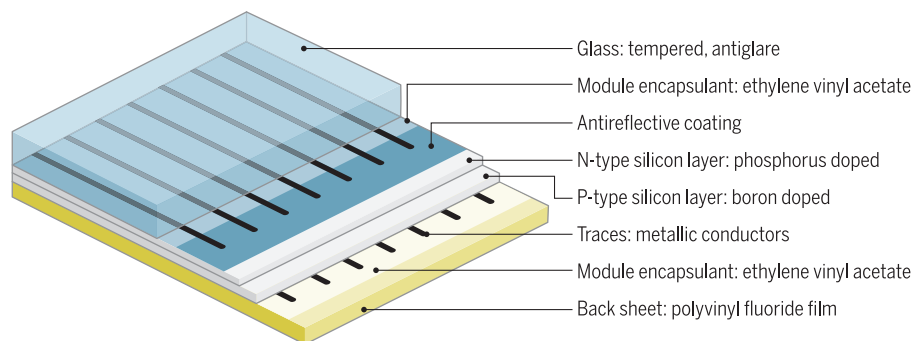
devices that exhibit high photovoltages in conjunction with quantum yields in excess of unity for photocurrent production. These materials systems will ultimately have to compete with other approaches to obtain device efficiencies that exceed the S-Q limit, such as multijunction cells, which have already exhibited efficiencies >40% under high concentration (7, 35).

### Balance of systems

The cost of Si solar panels now constitutes ~30% of the cost of a fully installed utility-scale system (Fig. 2) (36, 37). The “hard” materials costs, including the inverter, support structures, and electrical wiring, make up ~30% of the system costs. “Soft” costs, including installation labor, permitting, inspection and interconnection, financing, and customer acquisition, make up ~40% of the installed system costs. These balance-of-systems costs have not declined nearly as rapidly as module costs.

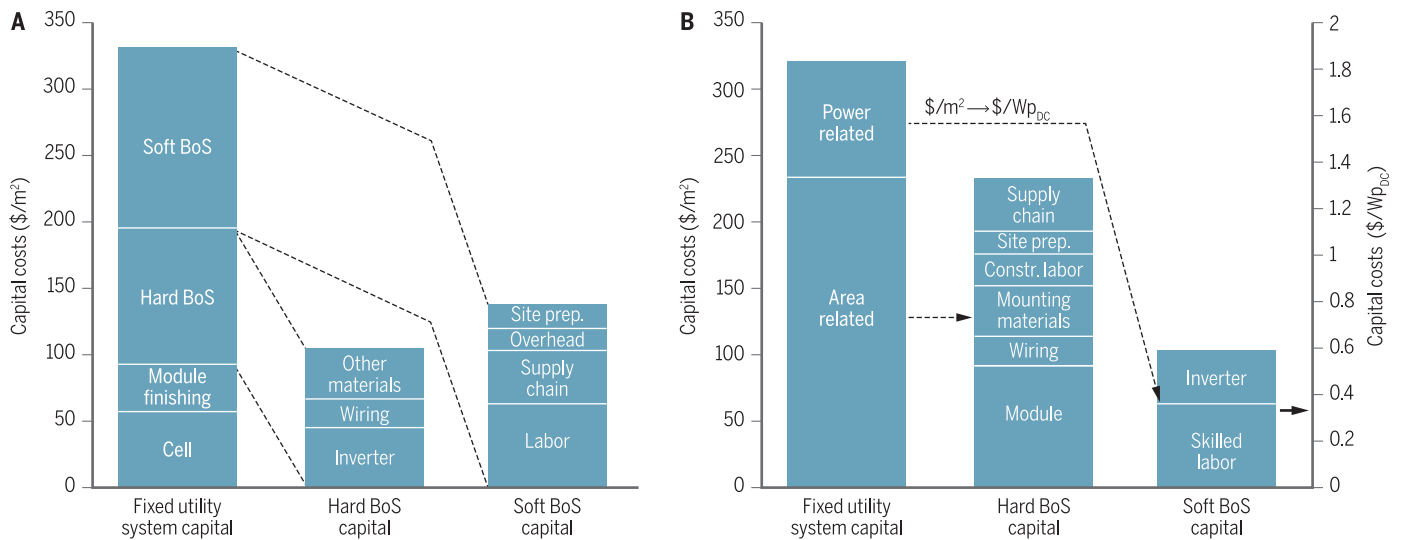
For an installation having a specific peak output power, increases in module efficiency would correspondingly reduce the area-related balance-of-systems cost (Fig. 2B). Any viable alternative to Si, or any PV technology that leverages or mates with Si PV technology will ultimately have to exhibit long-term stability and superior efficiency at competitive manufactured panel costs. Improvements in efficiency, especially through R&D, that result in the development of new materials and PV systems having efficiencies higher than the S-Q limit, would have more of an impact on lowering the cost of installed solar electricity than proportionate reductions in the manufacturing costs of present Si-based panels, as shown in Fig. 3.

To be certified for sale in the marketplace, solar panels are required to contain protective glass that can survive a hailstorm. The cost of the float-glass material is relatively low (38), but the stiffness and weight of the resulting panels produces sizable costs for shipping, requires the use of costly support structures, and produces substantial labor costs for installation. Soft costs are lower in Germany and Australia than in the United States, because permitting and installation processes and protocols have been streamlined (39, 40). Obtaining much lower installed PV system costs will not only require ultralightweight, flexible, robust, and efficient materials

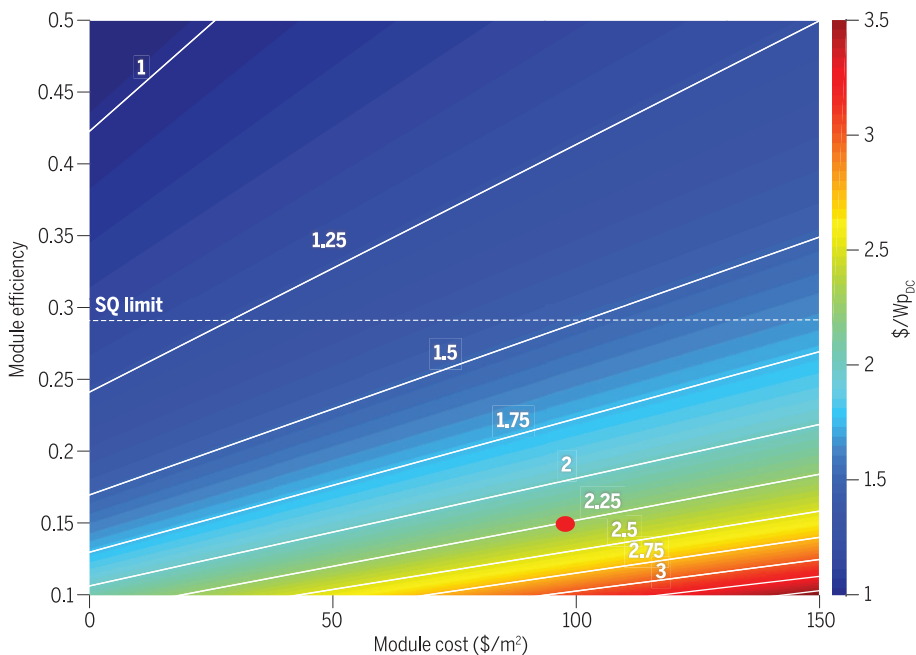


Note: material thicknesses not to scale

**Fig. 1. Components of a typical silicon solar cell.** The diagram shows the absorber layer, the p-n junction, antireflection coating, grid and contact lines, encapsulation, and glass support structures.



**Fig. 2. Breakdown of capital costs for installed utility-scale PV systems. (A)** Costs of finished modules (panels) made from Si solar cells, as well as hard and soft balance-of-systems (BoS) costs as tabulated by Goodrich *et al.* (35). The solar cell and module costs have been updated from those listed in (35) to reflect the rapidly declining price of these components (36). **(B)** Breakdown of the same capital costs for installed utility-scale PV systems as in (A), except that the costs are broken down into items that scale with the area of the installation (and hence, panel efficiency) and (area-independent) items that are related to the power produced by the installation and described in terms of dollars per peak watt of direct current.



**Fig. 3. Costs for installed utility-scale PV projects as a function of module efficiency and module cost.** Movement parallel to the abscissa assumes that all costs are held constant other than the module cost (Fig. 2A), whereas movement parallel to the ordinate is based on the assumption that there is a linear proportionality of all area-related costs (Fig. 2B) to the module efficiency. The red dot is representative as an example of recent utility-scale projects for which citable data are available (38, 39) with 15% efficient Si-based panels at a cost of \$90/m<sup>2</sup> and with a total installed project fixed capital cost of \$2.25/Wp. Utility-scale projects have been chosen because they are less expensive per Wp and less variable than rooftop installations, which have similar Si module costs but had, at the time, fully installed project costs of ~\$4.90/Wp in California as compared with \$2.20/Wp in Germany, with the difference caused primarily by differences in permitting, labor, inspection, and other soft costs.

and panel technology in the photoactive, encapsulating, and structural components, but also will require disruptive engineering approaches, including very inexpensive support structures, increasing automation, and streamlined protocols that minimize the skill level and effort associated with system installation (Fig. 3) (6).

Although the capital costs can be firmly established for a given PV installation (for example, Figs. 2 and 3), the levelized cost of electricity (LCOE) depends on the deployment site, which dictates the electrical energy that is produced by the panels over their useful lifetime and requires assumptions involving the discount rate (i.e., the time value of money), useful system life, and operating costs. For utility-scale Si PV systems having fully installed costs of \$1.80/Wp, conventional assumptions yield LCOEs of \$0.10 to \$0.15/kWh, with the lower value for favorable sites, such as in California (41). In 2014, the average U.S. electricity price for large industrial customers, including generation, transmission, and distribution costs, as well as profit, was ~\$0.07/kWh (42).

### Grid integration and energy storage

Regardless of the competitiveness on a levelized cost basis of solar electricity relative to fossil-based or nuclear-based electricity, the value of electricity produced from an intermittent resource is not the same as the value of energy that can be provided on demand. In some market scenarios, such as residential installations in areas with high peak electricity pricing, PV currently has a favorable value proposition, especially with tax incentives or other subsidies (41, 43). The full costs of solar electricity must also eventually include costs associated with grid integration, as

well as the accompanying cost of energy storage to compensate for daily, weekly, and seasonal variability in insolation (2, 41). To ensure reliability, utilities currently back up intermittent renewables, including solar and wind installations, to nearly their full rated peak capacity with dispatchable generation, generally derived from natural-gas-fired power plants. The cost of persistent grid-scale storage currently far exceeds the levelized cost of solar electricity (43, 44).

Pumped hydroelectricity is near its technoeconomic potential globally (44). Compressed-air energy storage will require cost-effective technologies to create robust, full-volume seals in very large underground geological cavities, such as salt caverns (44). Superconducting magnetic-energy storage and flywheels are best suited for high-power, low-energy applications (44, 45).

Batteries currently are expensive grid-scale electrical energy-storage technologies, especially when the costs are amortized over the >30-year lifetime of an installed solar electricity system (44). If one assumes a 15- to 20-year battery lifetime with one cycle per day, conventional or advanced lead-acid batteries have a levelized cost of energy storage (LCES) (not including generation costs) for renewable energy storage and/or time-shifting applications of \$0.80 to 1.0/kWh and \$0.40 to 0.50/kWh, respectively (46). Under the same assumptions, Na-S batteries, which require operation at ~350°C to obtain sufficient conductivity through the beta-alumina solid electrolyte that separates the liquid sodium negative electrode from the positive electrode composed of a surrounding layer of sulfur, have an LCES of \$0.30 to \$0.40/kWh (46). Capital costs of \$250/kWh for the battery cells are part of the \$1000/kWh current cost of a fully installed secondary lithium-ion (Li-ion) battery system for PV storage applications, which results in an LCES of \$0.49/kWh, if one assumes a 10-year service life, one cycle per day, a 10% discount rate, a linear capacity decline to 80% of rated capacity at the end of life, 92% charge-discharge energy-storage efficiency, and no operating costs (46, 47). Although the underlying battery chemistry and materials have been relatively unchanged for more than 20 years, Li-ion cells are following an ~25% learning curve, as calculated from sales and production data spanning 2000 to the present (48).

Given current average cell costs of \$270/kWh, at the historical rate of cost decline, obtaining a \$100/kWh Li-ion cell cost would require production and learning for >25 years, if one assumes a fixed 100 GWh worldwide annual capacity during that time, in accord with expectations for Li-ion battery-production capacity when new, large-production-capacity additions that are currently under construction become operational (48). Chemical reactions between the electrolyte and solvent in current Li-ion batteries fundamentally limit the useful lifetime of such batteries (49), regardless of the depth of discharge or power-management technology incorporated into the system. The costs associated with battery-pack integration and management, inverters, and other hardware and labor costs

that dominate the total installed system costs are currently much higher than the battery cell costs. Lithium-air (50), lithium sulfur (51), zinc-air (52), solid-state Li (53), and advanced Li-ion battery chemistries (53), among others, are all being explored.

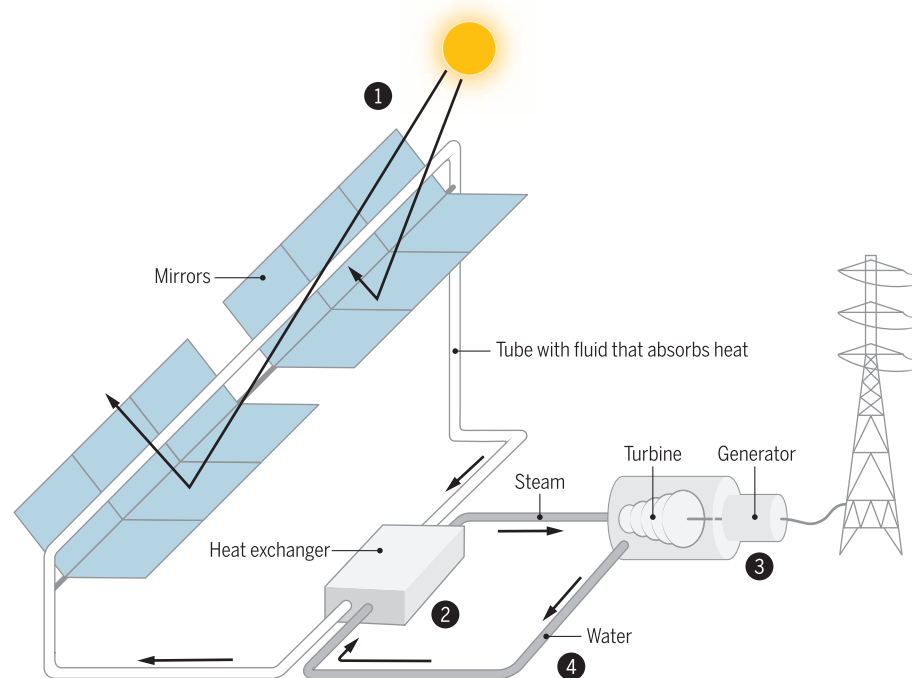
Flow batteries, based on electrically charging an anolyte and catholyte and storing the fluids in separate holding tanks until discharge, are a focus of major early-stage R&D efforts (44, 54). Electrochemical couples being developed include aqueous vanadium as  $V^{3+/2+}$ , Zn-Br, V-Cr, Fe-Cr, or  $H_2/Br_2$  systems (54), as well as quinone-hydroquinone redox species (55). Relatively large storage volumes are required because the energy density of flow batteries is typically 20 Wh/liter, compared with an energy density of >200 Wh/liter for Li-ion batteries and >12,000 Wh/liter for liquid hydrocarbon fuels, such as gasoline. If one assumes a 20-year lifetime and one cycle per day, at typical 65 to 75% charge-discharge efficiencies, Fe/Cr and V-based redox flow batteries have an estimated LCES of \$0.20 to \$0.30 and \$0.30 to \$0.40/kWh, respectively, with the latter costs resulting from higher materials costs for the V-based system relative to the Fe/Cr or Zn-Br systems (46). Electrically discharging the contents of the holding tanks rapidly will require pumping large quantities of liquid into effective contact with highly porous, high-surface area electrodes, which will add to the system cost. Notwithstanding associated carbon emissions, cost-effective generation technology to compensate for the intermittency of solar electricity and to provide back-up and firming capabilities for

daily and seasonal variability in new solar generation capacity currently uses natural-gas plants (46), with costs expected to remain low where shale gas is abundant.

### Solar thermal systems

The four main types of solar collectors are parabolic trough collectors, linear Fresnel reflectors, power towers (i.e., central receiver systems), and dish-engine systems, which produce local temperatures of 550°C, 550°C, >1000°C, and 1200°C, respectively (56–58). To generate electricity, either an oil or a molten salt heat-storage fluid, typically an eutectic mixture of 60 weight % (wt %) sodium nitrate to 40 wt % potassium nitrate, known as solar salt, is heated. The heat then is exchanged to produce steam, which is used to drive a turbine to generate electricity (Fig. 4) (5). Power conversion units are either separate or combined Rankine-Brayton cycles. As of 2014, the installed global capacity of PV was 177 GWp, as compared with <5 GWp of solar thermal capacity (59, 60). Solar thermal installations are preferably sited in regions with a high direct normal irradiance value, typically in desert regions of the southwestern United States or Australia, or, for example, Morocco or southern Spain (61).

Although large-scale solar thermal electricity projects were planned both in the United States and in Morocco, driven by mandates, renewable portfolio standards, and low-carbon electricity incentives, the generation costs have proven to be greater than \$0.15/kWh when all installation and operational expenses are included (62, 63).



**Fig. 4. Schematic of a typical 1D concentrating solar thermal system.** The sunlight is focused along one dimension to heat up a thermal fluid—typically, either an oil or a molten salt—which is then passed through a heat exchanger to produce steam that is used in a turbine to produce electricity.

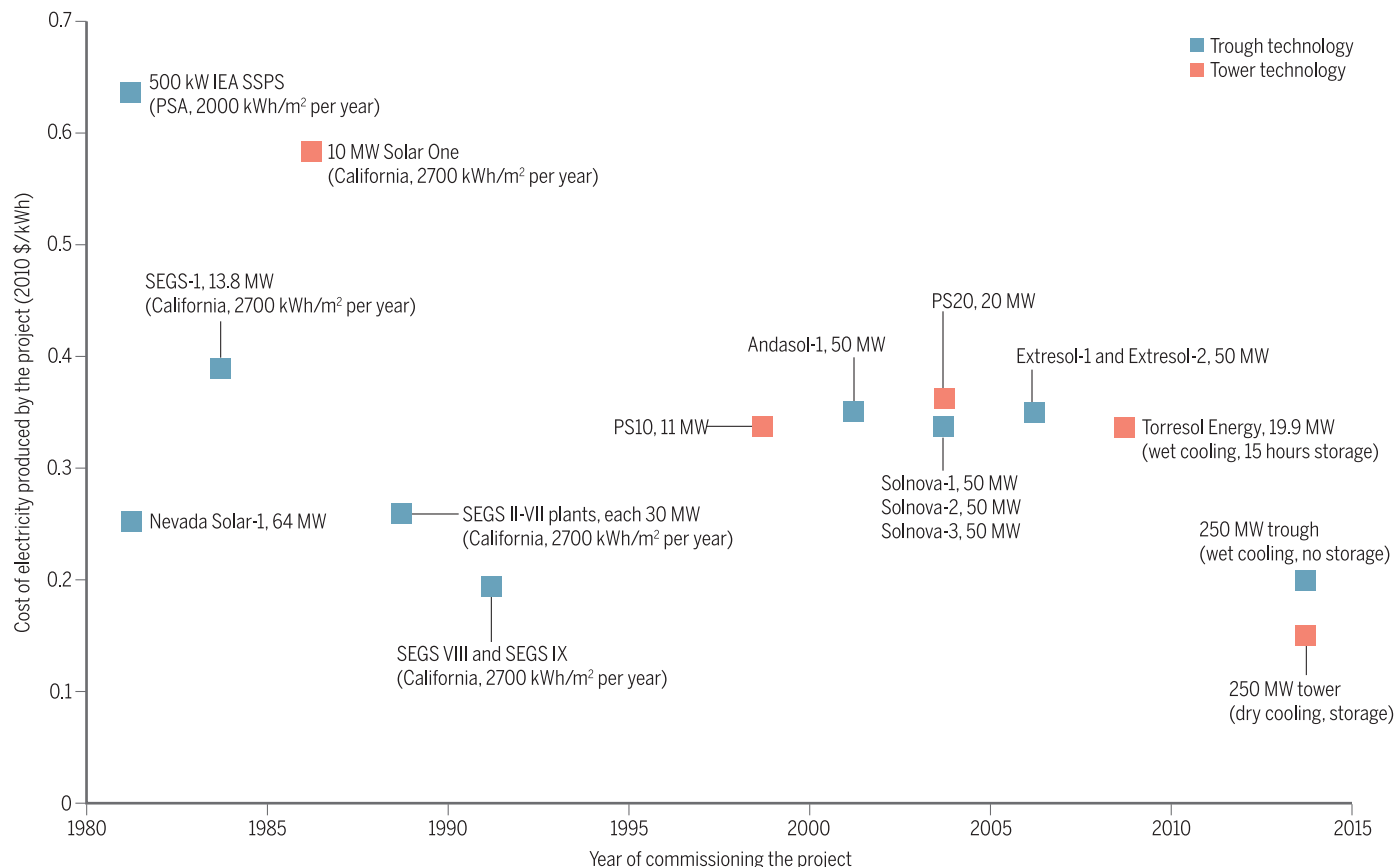
Between 6 and 8 hours of storage can be obtained by use of molten salts as the thermal fluid, but compensation for cloudy days requires 36 hours, or more, of storage. Improved thermal storage fluids are an active area of R&D (64). The all-inclusive costs for solar thermal systems have not declined substantially since the early 1980s (62) (Fig. 5), in contrast to the costs of PV modules and installed PV systems. For example, the Ivanpah Solar Electric Generation system commissioned in 2013 in the Mojave Desert of California is an air-cooled, 392 MW capacity, power-tower system consisting of 300,000 mirrors and three towers. The project had a capital cost of \$2.2 billion and is producing electricity under a 25-year power purchase agreement for a price estimated as  $> \$0.13/\text{kWh}$  to the cognizant utilities (41). Research efforts include development of power cycles that allow for higher-temperature operation, along with the development of advanced materials for the fabrication of the collectors (65), in conjunction with new engineering approaches to the design of the collectors and integration with the rest of the plant (66–68). Thermoelectrics could also serve as the technology that converts heat into electricity, provided that their performance can be improved to the necessary levels under the high temperatures that are produced in an operational solar thermal system (5, 69).

Extensive deployment of either PV or solar thermal electricity-generating systems in remote regions with high direct insolation would require installation of new transmission lines. The Sunrise Powerlink—a 117-mile-long, 1-GW capacity, 500-kV,  $>90\%$  above-ground transmission line built between 2010 and 2012 to provide solar and wind power from the Imperial Valley to San Diego—had a project cost of \$1.9 billion (70). Concentrated solar power can also be used to provide a source of heat to drive endothermic chemical reactions. In one demonstration system, solar heat is used as a supplement to process heat, to produce synthetic fuel from CO and H<sub>2</sub> (syngas) by using the Fischer-Tropsch process (71). For the solar-driven process to be economically viable, the cost of solar-derived heat must be less than the cost of heat derived from combustion of fossil energy, such as natural gas. The capital costs of the solar thermal part of the facility—including the costs associated with siting constraints, as well as underutilization of the plant during night time and periods of off-peak insolation—must be covered by the value of the solar-derived heat production.

Solar-derived heat can also be used to promote the formation of fuels, such as H<sub>2</sub> produced through solar-driven water splitting (72). In an exemplary two-step cycle, CeO<sub>2</sub> is first reduced thermally to Ce<sub>2</sub>O<sub>3</sub> with the production of O<sub>2</sub>,

followed by the thermally driven oxidation of Ce<sub>2</sub>O<sub>3</sub> by water to produce CeO<sub>2</sub> and H<sub>2</sub> (73). Other thermochemical cycles are based on sulfur iodine, hybrid sulfur, photolytic sulfur ammonia, zinc oxide, cadmium oxide, or FeAl<sub>2</sub>O<sub>4</sub> (74). Generally, the cycles exploit temperature swings to evolve O<sub>2</sub> and H<sub>2</sub> separately, which creates research opportunities to explore methods of obtaining high levels of heat recovery so as to ensure high system efficiencies. Isothermal redox cycles have been proposed to alleviate heat-rejection issues, by use of an open system in which a partial pressure change promotes the evolution of O<sub>2</sub> by the thermal reaction of CoFe<sub>2</sub>O<sub>4</sub> with Al<sub>2</sub>O<sub>3</sub>, and the CoAl<sub>2</sub>O<sub>4</sub> and FeAl<sub>2</sub>O<sub>4</sub> then react with steam to produce H<sub>2</sub> and close the chemical cycle (75). Many of these thermochemical water-splitting cycles are also of interest for the production of H<sub>2</sub> from the heat produced by gas-cooled nuclear fission reactors.

These thermochemical cycles to date have been demonstrated in process steps at the laboratory or pilot scales. Implementation of these thermochemical processes in deployed solar thermal systems will require R&D that enables the needed mass flows of reagents into the reactor while allowing facile product egress from the reactor, in addition to effectively confining the heat in the reactor while allowing for the optical excitation to enter, but not leave, the reactor. The reactor



**Fig. 5. Chronology of the total installed cost for some large solar thermal projects.** Names of projects and rated capacity of each installation. IEA SSPS, International Energy Agency Small Solar Power Systems (Europe); SEGS, Solar Energy Generating Systems (California); P10 (Spain).

**Table 2. List of solar fuel systems and their defining technical attributes.**

| Solar fuels system                | State of development  | Example systems  |
|-----------------------------------|---|--|
| Molecular systems                 | Persistent charge separation demonstrated; fuel-producing systems, including efficient, stable molecular electrocatalysts, must be discovered | Porphyrins linked to quinones either in diads, triads, or tetrads; Ru-bipyridyl inorganic chromophores linked to Pt particles or hydrogenases, and linked to molecular catalysts for water oxidation |
| Inorganic semiconductor particles | Solar-driven water-splitting devices demonstrated; efficiency, safety, stability need to be addressed simultaneously                          | GaN:ZnO w/ RuO <sub>2</sub> , Cr <sub>2</sub> O <sub>3</sub> -coated Rh cocatalysts (123)  |
| Semiconductor photoelectrodes     | Efficient, safe systems demonstrated; long-term stability, cost-effectiveness need to be improved   | Si/Al <sub>0.15</sub> Ga <sub>0.85</sub> As (124), GaAs/GaInP <sub>2</sub> (100)   |
| Photovoltaic electrolysis         | Demo  | Si PV coupled to electrolyzer (125, 126)   |

materials must also be cost-effective, efficient, and robust under extreme operating conditions and environments (5, 76).

### Solar Fuels

The direct production of fuels from sunlight could provide a scalable grid storage technology. Liquid fuels are required for ships, aircraft, and heavy-duty trucks, which collectively total 40% of current global transportation fuel demand. Solar fuels technology has thus received recent attention at the proof-of-concept R&D stage in various laboratories worldwide (5). The status of various approaches to solar fuels generation is summarized in Table 2. Exquisite chemical synthetic methods have been used to assemble various chlorophyll-based and -related light absorbers in conjunction with precisely connected electron donors and acceptors, to achieve separation of the light-induced electron-hole pairs produced by the chromophore of interest (5, 77, 78). The existence of the natural photosynthetic system implies that it ought to be possible to assemble a fully nonbiological photosynthetic assembly, which provides a goal for research efforts in synthetic organic, inorganic, and materials chemistry. Particles of inorganic semiconductors, in conjunction with heterogeneous cocatalysts, have been shown to act as artificial photosynthetic systems (5, 79). A research goal is to develop an efficient, stable photocatalytic system that does not produce stoichiometric, flammable mixtures of H<sub>2</sub> and O<sub>2</sub> (79).

In artificial photosynthetic systems based on photoelectrochemical (PEC) cells, (Fig. 6), the charge separation is facilitated by an electric field that is formed at or near a semiconductor-liquid interface (79, 80). Production of fuel generally requires coupling of the separated electrical charges with multielectron catalysts for the half-reactions of interest. In many respects, PEC cells represent the integration of a discrete PV module with an electrolyzer to achieve the conversion of sunlight into H<sub>2</sub> and O<sub>2</sub> by solar-driven water splitting (79, 81). Regardless of how it is produced, the solar H<sub>2</sub> could be used in a turbine or fuel cell for grid storage, and could be used as a transportation fuel either directly as H<sub>2</sub>, or indirectly by reacting the H<sub>2</sub> with CO<sub>2</sub> or other carbon-based feedstocks, such as biomass, to produce methanol or other reduced carbon-based

fuels (82, 83). Additionally, solar-derived H<sub>2</sub> could readily be used as a feedstock to produce ammonia, for use in agriculture, as well as in commerce and transportation.

For a solar-driven PEC-based water-splitting system to be deployed in the marketplace, the PEC system must be advantageous in cost relative to the established, discrete PV + electrolyzer (E) combination that provides the same system-level functionality (79). The levelized cost of H<sub>2</sub> (LCH) from a discrete PV+E system has been estimated to be \$7 to \$20/kg (84, 85), because of the relatively high cost of electricity from PV in conjunction with a low capacity factor for utilization of the electrolysis unit. In contrast, the LCH produced by steam reforming of natural gas is only \$2/kg (86). Research opportunities involve the development of a disruptive technology for electrolytic H<sub>2</sub> generation, noting that the chlor-alkali electrolysis process has been practiced at scale for more than a century (87). Storage in geologic formations has been proposed for renewable hydrogen produced by electrolysis (88).

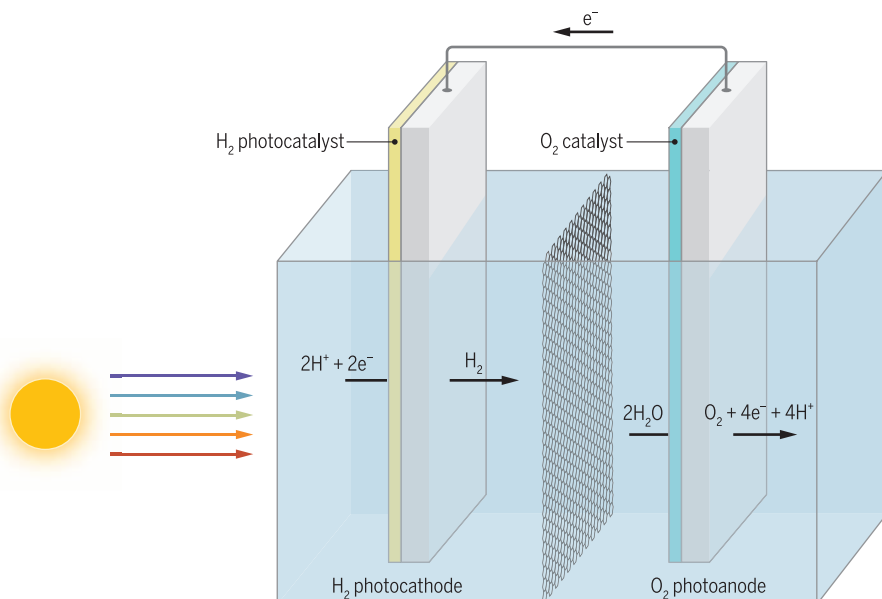
In state-of-the-art polymer-electrolyte-membrane (PEM) electrolyzers, the cost of the noble metal catalysts is <6% of the total system costs (89). Even less expensive catalysts—based on Ni-Mo and related alloys on the cathode and mixed Ni (Fe, La, and so on) oxides on the anode—are used in alkaline electrolysis (90), with a facility rated to produce 100 MW of electricity constructed in the 1980s at Aswan Dam (91). Moreover, available electrocatalysts in acidic or alkaline media display much lower overpotentials (92) than available electrocatalysts operated at near-neutral pH (93–95). Hence, an integrated photoelectrochemical system must take advantage of substantial cost reductions enabled by integration, other than purely a reduction in catalyst cost, to be advantageous in cost relative to a discrete PV+E combination. Opportunities for such cost reduction are provided by synergistic integration of very inexpensive materials in novel form factors and geometries, such as arrays of semiconducting microwires in flexible polymeric membranes (96, 97) coupled to earth-abundant electrocatalysts, with the system able to take up water from the atmosphere as an input feedstock (98).

A technology for an integrated solar fuels generator must be robust, efficient, safe, and cost-

effective. At present, viable integrated systems can simultaneously meet at most three of these desiderata. Systems that are very efficient and stable are currently made from expensive, complex materials assemblies, such as a high-efficiency III–V PV tandem cell connected electrically in series with, but isolated physically from, an electrolysis cell (99). Use of an analogous multi-junction III–V cell as a photoelectrode in an aqueous electrolyte in either a wired or wireless configuration produces H<sub>2</sub> transiently, because integration results in corrosion of the photoelectrode by the electrolyte (100, 101). Electrodes that are cheap and robust—such as spray-painted coatings of iron oxide as photoelectrodes by themselves or with coatings of electrocatalysts—are inefficient (5). Electrolysis or photoelectrolysis under bulk near-neutral pH conditions (93, 102–104) is inefficient and/or produces potentially explosive, stoichiometric mixtures of H<sub>2</sub> and O<sub>2</sub> over active catalysts for recombination of the products (95, 105–108).

Photocathodes made from p-type indium phosphide coated with nearly transparent noble metal electrocatalyst films have yielded >13% ideal regenerative cell efficiencies for the production of H<sub>2</sub> from 1 M HCl (aqueous) (109), which illustrates the possibilities for obtaining efficient and stable photoelectrodes for solar fuels production. Operation in acidic or alkaline liquid or polymeric electrolytes is important because intrinsically safe, efficient solar-driven water-splitting systems can be built in these media (81, 110). Some recent examples along this R&D path include the discovery of earth-abundant alternatives to Pt that are stable and highly active for H<sub>2</sub> evolution in acidic media (111) and the integration of earth-abundant electrocatalysts into Si microwire arrays for efficient H<sub>2</sub> production from acidic media (112). Research opportunities include methods to minimize the obscuring of light associated with typical metal and metal oxide electrocatalysts for fuel-forming half-reactions (113).

Porous films allow electrolyte to permeate to the underlying semiconductor and, thus, only partially mitigate deleterious corrosion or passivation processes (114). For oxidative processes, amorphous TiO<sub>2</sub> films in conjunction with Ni oxide islands (115), or alternatively the use of reactively sputtered Ni oxide films, have recently been shown to provide extended stability for photoanodes



**Fig. 6. Schematic of a tandem photoelectrochemical cell.** The photocathode where  $\text{H}_2\text{O}$ , or  $\text{H}_2\text{O}$  and  $\text{CO}_2$ , are reduced to a fuel, and the photoanode, where water is oxidized to produce  $\text{O}_2$ . The electrodes also generally have electrocatalysts to facilitate the desired multielectron half-reactions, and the membrane is present to ensure intrinsically safe operation as well as to facilitate beneficial collection of the  $\text{H}_2$  under a pressure differential to ensure its flow into a pipeline collection system.

performing water oxidation in alkaline media (114, 116, 117). These approaches to photoanode protection, which have shown operational stability and efficiency for water oxidation while operating continuously for thousands of hours under simulated sunlight, i.e., passing the charge equivalent to >1 year of outdoor operation (114), may allow for new combinations of materials to be utilized. These components will have to be combined with other materials and components at scale to realize the full potential of an integrated approach to solar fuels generation. Research opportunities also lie in the development of a cost-effective balance-of-plant that would provide feedstock water of the requisite purity, as well as facilitate the safe, cost-effective collection of the solar-derived  $\text{H}_2$  over relatively large areas for distribution and end-use, either as  $\text{H}_2$  or by conversion to carbon-containing fuels through known thermochemical conversion processes.

The multielectron reduction of  $\text{CO}_2$  to a liquid fuel will require the development of entirely new, unprecedented families of catalysts to effect such a transformation in an energy-efficient and selective fashion. To date, metal electrodes require high overpotentials, are generally unstable at the required reducing potentials, and also produce a wide array of trace organic products (118) that would require an expensive and energy-intensive process step to separate and concentrate the products. On specific electrode surfaces, substituted pyridiniums yield partial formation of methanol and other alcohols (119). An engineered bacterial system has been used to produce low concentrations of isopropanol from electrolytically generated  $\text{H}_2$  (120), complementing previous studies that have coupled the enzyme formate dehydrogenase

to a semiconductor electrode to demonstrate the direct enzyme-catalyzed photoelectrochemical production of fuel (121). Extensive research efforts aim to extend the stability of such enzymes in vitro.

Other challenges for  $\text{CO}_2$  reduction involve the flux limitations associated with utilization of atmospheric  $\text{CO}_2$  as a sustainable  $\text{CO}_2$  source, as well as the expense of concentration of  $\text{CO}_2$  for use as a reagent (122). In conjunction with advances in materials and methods, additional systems-based technoeconomic analysis is required to ascertain whether an artificial system can be constructed, in principle, with much higher efficiency, lower cost, and utility than either natural photosynthesis or direct or indirect solar  $\text{H}_2$  production followed by thermochemical conversion of  $\text{H}_2$  with  $\text{N}_2$ ,  $\text{CO}_2$ , or other carbon-containing reactants, such as ethylene or biofuels, to produce value-added, energy-rich fuels and/or chemicals.

### Promise and potential

The remarkable progress that has been made in cost reduction and commercial deployment of solar energy technologies underscores the benefits of investment in R&D and indicates the promise of, and necessity for, continued innovation to produce further advances in the field. Both evolutionary and revolutionary technology development, involving foundational research, applied research, learning by doing, demonstration projects, and deployment at scale will be needed to continue this technology-innovation ecosystem. Relative to the embodiments of solar energy-conversion systems that have been developed to date, higher efficiencies, lower costs, improved scalability, and new functionality are in principle

achievable. Hence, research, engineering and manufacturing will need to be pursued in harmony and in a sustained fashion to allow realization of the full potential of solar energy utilization, and to allow the energy in sunlight to make a material, and perhaps dominant, contribution to a sustainable, cost-effective, global energy system.

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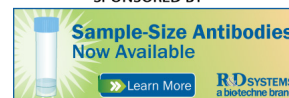
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## Research opportunities to advance solar energy utilization

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