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Artificial Photosynthesis

New Fuels: Artificial Photosynthesis
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What is Artificial Photosynthesis?

Global energy needs are expected to double over the next 40 years. To account for this rise in global energy usage and the consequent pressure on US domestic energy utilization, initiatives focused on finding cost-effective, carbon-neutral, geo-politically favorable energy sources are critically important. The most important renewable energy resource is the sun. The amount of sunlight that hits the earth in an hour could meet the entire world energy demand for a full year.[1] The conversion of only a small fraction of this available solar energy to a usable fuel source would dramatically change the inherent energy infrastructure, while helping to reduce emission of greenhouse gases associated with fossil fuel consumption.

Solar fuels are storable fuels produced using solar energy. Solar energy can *indirectly* generate usable fuels through biomass. Alternatively, the *direct* conversion of solar energy into fuels through a fully integrated system is known as artificial photosynthesis. Artificial photosynthesis applies the principles that govern natural photosynthesis to develop a man-made technology. It strives to be a viable fuel source based on the consumption of abundant resources: solar energy, water, and carbon dioxide. Artificial photosynthesis takes advantage of the efficient primary solar energy conversion steps of photosynthesis, but does not use energy to sustain life as does the natural process, nor does it necessarily require the land usage associated with biomass production.

Artificial photosynthesis produces fuel via two main pathways: carbon dioxide reduction to ultimately yield hydrocarbons and water oxidation to generate hydrogen. At pH = 0, water splitting can be described by the following overall equation:

$$2H_2O \leftrightarrow 2H_2 + O_2 \qquad E^\circ = 1.23\text{ V vs. NHE} \qquad (1)$$

In (artificial) photosynthesis, sunlight provides the required energy (kinetic and thermodynamic) to drive the reaction in the forward direction and split water into hydrogen and oxygen. Expansion of equation 1 demonstrates that it is the summation of two underlying half-reactions:

$$2H_2O \leftrightarrow O_2 + 4H^+ + 4e^- \qquad E_{ox} = 1.23\text{ V vs. NHE} \qquad (2)$$

$$2H^+ + 2e^- \leftrightarrow H_2 \qquad E_{red} = 0.00\text{ V vs. NHE} \qquad (3)$$

These reactions show that converting water to hydrogen and oxygen is a multi-step, multi-electron process that not only needs energy to perform redox chemistry, but also requires different redox catalysts. These catalysts facilitate each of the above multi-electron half-reactions, where one catalyst evolves molecular oxygen by oxidizing water (equation 2) and a second catalyst generates hydrogen (equation 3) by reducing protons.

Similar to natural photosynthesis, artificial photosynthesis uses light absorbing molecules and/or materials to capture light and produce a charge separation. Then, through a series of inter-/intra-molecular charge transfer reactions these charges are transported to catalytic sites to provide the requisite oxidizing/reducing energy to evolve oxygen or hydrogen. Depending on the architecture of the artificial photosynthetic device, the nature of the light capture, charge separation, charge transport and active catalytic sites vary greatly.

Currently there are no known molecular or heterogeneous catalysts for the reduction of CO_2 to fuels that have the combination of rates, overpotentials, selectivities, cost, and lifetimes that are needed solar fuels production. A phased parallel approach can immediately address issues associated with the integration of CO_2 reduction catalysts with light-harvesting and charge separation components, while allowing the simultaneous development of better catalysts for CO_2 reduction. Current efforts focus on developing catalysts for two-electron reduction reactions to produce CO (equation 4, at pH = 7), a key component of syngas used in existing commercial processes for hydrocarbon synthesis, and four electron processes to convert CO_2 to methanol for potential use in chemical synthesis and fuel cells (equation 5, at pH =7). The availability of syngas through partial reduction of CO_2 to CO may preclude the need for reducing CO_2 all the way to CH_4, but ultimately both its scientific feasibility and economics will dictate whether formation of CH_4 is a desirable endpoint. Although none of the known catalysts for these CO_2 reduction processes are optimal, some are sufficiently promising to begin studies of integrating them with solar energy capture and conversion systems to produce solar fuels.

$$CO_2 + 2H^+ + 2e^- \leftrightarrow CO + H_2O \qquad E^\circ = -0.53 \text{ V vs. NHE} \qquad (4)$$

$$CO_2 + 6H^+ + 6e^- \leftrightarrow CH_3OH + H_2O \qquad E^\circ = -0.38 \text{ V vs. NHE} \qquad (5)$$

Ultimately CO_2 should be recovered directly from the atmosphere to achieve a fully closed CO_2 cycle, as observed in photosynthesis. However, the knowledge or technology to do this efficiently currently does not exist. Research to achieve an integrated solar/CO_2 reduction system is focusing on using pure CO_2. In the intermediate term, it is possible to take advantage of technologies being developed for recovery of CO_2 from point sources such as power plants. The present state of the art for capturing CO_2 from flue gases is the use of an inefficient liquid amine based system. New approaches, such as Thermal Swing Adsorption (TSA) using tailored solid sorbents for CO_2 capture, are currently being developed, which do not waste energy on heating water as required for regeneration of the liquid amine systems. In the longer term, integrated photodriven CO_2 capture and reduction systems capable of removing and using CO_2 directly from the atmosphere need to be developed to achieve solar fuels formation.

Current Technological Maturity

To date, no comprehensive, cost-effective artificial photosynthetic system exists. Current capabilities involve linking a photovoltaic cell to a water electrolysis unit. In this system, the electricity generated from the photovoltaic cell provides the energy necessary to drive the water splitting reaction at two electrodes (equation 1). Two catalysts are still required for this reaction, with the oxidation of water (equation 2) occurring at the anode of the electrolysis cell and proton reduction at the cathode (equation 3). Although this system is able to perform water oxidation, both major components exhibit flaws that prevent widespread use. The most prevalent photovoltaic cells are made of single crystal, multi-crystalline, or amorphous silicon semiconductors both to absorb light and generate charges. These cells can operate at sunlight to electricity conversion efficiencies as high as 24%, but their manufacturing costs are high.[2] Promising alternative photovoltaic cells that use lower cost materials, such dye-sensitized solar cells and organic photovoltaics, are being developed, but their present working efficiencies are significantly lower than those of silicon-based cells. On the other end, current water electrolysis technologies tend to operate in hazardous alkaline conditions (concentrated KOH) and/or by using expensive noble metal catalysts, i.e. platinum in proton exchange membrane electrolyzers. These harsh environmental conditions and/or expensive catalyst/electrodes do not provide cost-efficient, ideal conditions for large-scale applications. Finally, wiring together these two *separate* devices, silicon-based photovoltaic cells and a water electrolyzer, only achieves overall efficiencies approaching ~13% with the potential for further losses being incurred through transmission of the electrical power, conversion from direct current to alternating current, and the voltage difference between the photovoltaic cells and the electrolyzer.[1,3] The ability to fully integrate both technologies into *one* system should increase the overall efficiency and therefore, increase its technological viability.[1]

A complete, highly efficient artificial photosynthetic system capable of photochemical hydrogen generation has yet to be established because significant knowledge gaps remain for components systems as well as their integration into an optimized system. As an example, Fig. 1 illustrates a proposed, fully-integrated model device architecture.[3] When high energy blue light is absorbed by a photoanode material, charge separation occurs that drives water splitting to generate protons and molecular oxygen. The remaining low energy red light then passes through the photoanode to reach a second molecular interface that selectively absorbs this light and serves to pump protons through a semi-permeable membrane. In the second compartment, a proton reduction catalyst reduces these protons to generate molecular hydrogen fuel. This example device architecture illustrates the complexity of integrating light capture, charge separation, and catalysis in a compartmentalized integrated artificial photosynthetic device that optimizes both efficiency and cost to increase the commercial viability of the technology.

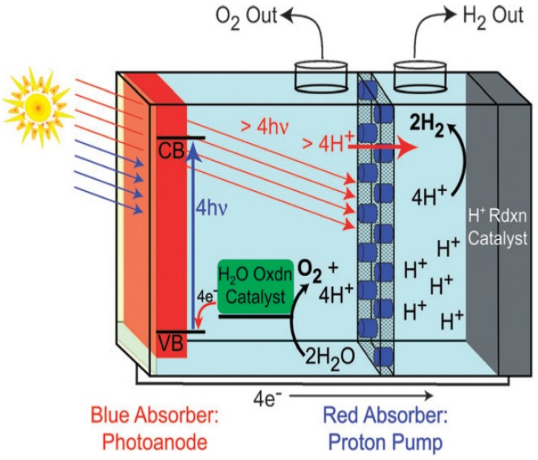


Fig. 1. Model artificial photosynthetic water splitter.[3]

Players and Research

Artificial photosynthesis is a large, diverse, and active research field. The combined expertise necessary to tackle this difficult problem has fueled an increasingly useful and important collaborative team research model. A number of recently established large U.S. research centers has decreased the projected lead-time for scientific advancements and has enhanced collaboration amongst leading scientists. Specifically, the National Science Foundation (NSF) recognized solar-to-fuels research as one of the "Grand Challenges" of the 21st century. In August 2008, the NSF funded the Powering the Planet: a Chemical Center for Innovation, which is composed of researchers from 13 universities, Brookhaven National Laboratory, BP Solar, and Southern California Edison.

Similarly, in 2001 the U.S. Department of Energy's (DOE) Basic Energy Sciences Advisory Committee conducted a study to address research needs pertaining to multiple DOE missions. In the following years, a series of ten workshops and published reports including the Basic Research Needs for Solar Energy Utilization report, which, in August 2009, ultimately led to the formation of 46 Energy Frontier Research Centers (EFRCs) with as many as 27 of them having some fraction of their ongoing work related to solar energy conversion and 7 of them largely focused on solar energy.

Most recently, the DOE established three Energy Innovation Hubs, including a solar research facility. In July 2010, the DOE awarded $122 million dollars over five years to researchers at California Institute of Technology in partnership with the DOE's Lawrence Berkeley National Laboratory for the formation of the Joint Center for Artificial Photosynthesis (JCAP). JCAP aims to rapidly move artificial photosynthetic research out of the laboratory setting, towards commercial viability, to create a direct solar fuels industry.

The level of effort required to fully develop economically viable solar fuels technologies requires significant private sector investments to leverage existing government support.

Table 1 lists researchers whose research impacts solar fuels. The sub-fields include solar energy capture and conversion by photovoltaics, molecular artificial photosynthetic systems, H_2O splitting, proton and CO_2 reduction catalysis, as well as photoelectrochemistry. The list is not comprehensive, but is illustrative, and includes individuals with significant activity in this field over the past 5 years.

Challenges

Extensive ongoing research efforts are focused on minimizing costs and increasing efficiencies through the use of earth-abundant robust materials, the generation new efficient materials/catalysts, and the increased understanding of how to fully integrate both photovoltaic and photocatalytic systems. Specifically, viable artificial photosynthetic technologies depend upon research discovery, development, and design in the following areas:

- Development of high performance, cost-effective light absorbing materials for use in photovoltaics
- Discovery and development of cost-effective catalysts that have long-term stability and can be linked to photovoltaic technologies
- Design and discovery of interconnected membrane networks that provide a physical support network for the overall process
- Design of interfacial materials that link light absorbers to catalysts to allow for efficient control of the integrated system
- Development and design of architectures that allow for scaling-up from the nanoscale to the macroscale

Given the current level of scientific and engineering effort, the complexity of this technology prevents its near-term contribution to the fuel supply without a significantly increased, focused and coordinated international effort. This emergent technology faces a multitude of challenges in each of the research areas listed above. Attention continues to focus on increasing efficiencies and decreasing costs of all device components, while devising effective methods for linking each component into an integrated system that functions under ambient conditions and offers long-term stability.

On a larger scale, difficulties arise when confronting how to store, distribute, and use hydrogen. Current commercial technologies convert hydrogen to hydrocarbon fuels using the Fischer-Tropsch process. However, this may not be the best long-term use of hydrogen because it introduces energy loses. Therefore, the development of an infrastructure to store and utilize hydrogen effectively is linked to the success of artificial photosynthesis as a useful transportation fuel source. Alternatively, the solar-driven conversion of CO_2 directly into liquid fuels that can be stored at ambient temperatures and can use existing infrastructure for distribution and consumption has distinct economic advantages in the near term.

Key Findings

Artificial photosynthesis is an very active research field where more than 450 related scientific research articles were published in peer-reviewed journals in 2010 alone. A detailed summary of all relevant finding exceeds the scope of this publication. As a consequence, this section will highlight six recent research advancements in the field and comment on their importance to the development of a complete cost-effective, efficient artificial photosynthetic device. The order in which they are presented does not reflect in any way upon their relative importance.

1. High-Throughput Combinatorial Approach to Catalyst Screening

As illustrated above, the solar energy derived water splitting into protons and oxygen is imperative to the development of an artificial photosynthetic system. Current research focuses on finding earth-abundant materials to perform this catalysis. A large range of metal oxides have been classified as candidates for use in photoelectrochemical cells as photoanodes for water oxidation (Fig. 1). However, a stable and efficient material, or combination of materials, has yet to be identified. Current theoretical capabilities prevent the identification of ideal semiconducting metal oxide candidates *a priori*. As a result, researchers have utilized high-throughput combinatorial approaches to discover and screen potential material(s), where automated depositions of metal oxide material combinations occur on electrodes and subsequently, their photoactivity is then assessed.[4-5] This method for

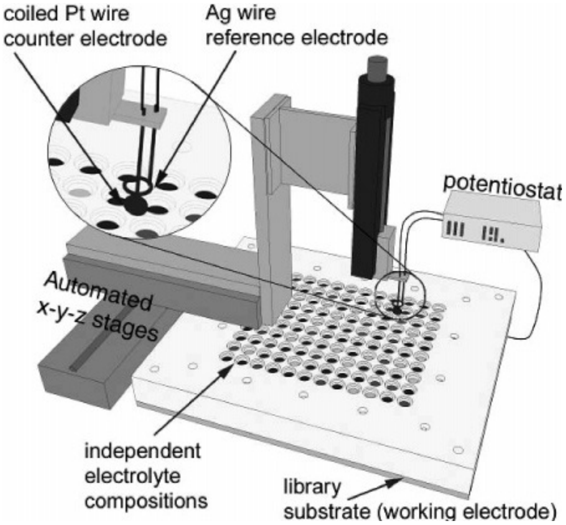


Fig. 2. An automated electrochemical deposition system for the serial production of metal oxides.[4]

the screening of photoanodes capable of water screening is one critical innovation for the rapid identification of potential metal oxide materials for water oxidation (Fig. 2).

2. Self-healing, Self-assembling Oxygen-Evolving Catalyst

One of the major hurdles for useful and efficient water oxidation is stability. One way natural photosynthesis overcomes this problem is by regenerating a key protein in the oxygen evolving complex every ~30 minutes. Therefore, achieving stable artificial photosynthesis requires materials and/or mechanisms in place that account for long-term chemical stability. Consequently, the discovery of a "self-healing," cobalt-phosphate water oxidation catalyst that operates under neutral conditions (and a wide pH range) introduces a new catalyst that increases catalytic stability, while operating under benign conditions and with earth-abundant materials (Fig. 3).[6] This catalyst also self-assembles *in situ*, which limits synthetic intensity and continues to push down costs.

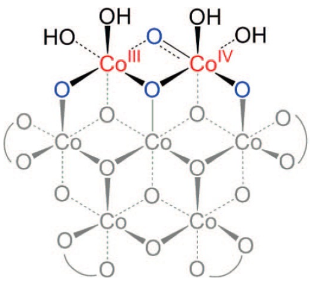


Fig. 3. Self-healing cobalt water oxidation catalyst.[6]

3. Molecular Level Understanding of Accumulative Electron Transfer

Advances in basic research have elucidated the fundamental principles that govern photoinduced single electron transfer reactions in both natural photosynthetic reaction centers and their synthetic (artificial) mimics. However, the catalytic oxygen evolution from water requires the accumulation of multiple oxidative equivalents at the catalyst, which introduces mechanistic complications. Mimics of the water splitting manganese-calcium complex in photosystem II (PS II) have been synthesized and linked to photosensitzers (light-absorbing molecules) to systematically determine how molecular structure, material energetics, and transfer kinetics influence accumulative electron transfer processes (Fig. 3).[7] Although the generation of a fully functional, efficient, molecular-level mimic has not been synthesized, this research illustrates the importance of creating a molecular scale mimic to gain basic mechanistic understanding before increasing system complexity.

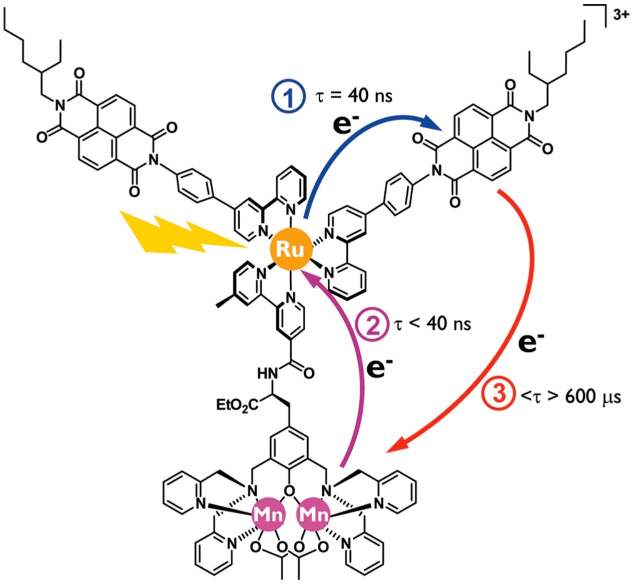


Fig. 3. Manganese-ruthenium trisbipyridinyl-dinaphthalenediimide triad PSII synthetic mimic.[7]

4. Self-Assembly of Photoactive Charge Conduits for Integrated Solar Fuels Systems

Complex, covalent molecular systems comprised of chromophores, electron donors, and electron acceptors, which mimic both the light-harvesting and the charge separation functions of photosynthetic proteins, have been used to study the dependencies of electron transfer rate constants on donor-acceptor distance and orientation, electronic interaction, and the free energy of the reaction. The most useful and informative systems are those in which there are structural constraints to control both the distance and the orientation between the electron donors and acceptors. Despite significant progress in the development of these covalent assemblies, the fundamental concepts of how to prepare individual light-harvesting complexes, reaction centers, or catalysts that can readily self-assemble into ordered supramolecular structures with the ability to function as complete artificial photosynthetic systems are largely unknown. Self-assembly provides a facile means for organizing large numbers of molecules into supramolecular structures that can bridge length scales from nanometers to macroscopic dimensions. For example, self-assembling charge conduits (Fig. 4),[8] can capture solar energy, separate charge, and deliver that charge to pendant catalysts or to electrodes. The central scientific challenge is to develop small, functional building blocks, having a minimum number of covalent linkages, which also have the appropriate molecular recognition properties to facilitate self-assembly of complete, *functional* artificial photosynthetic systems.

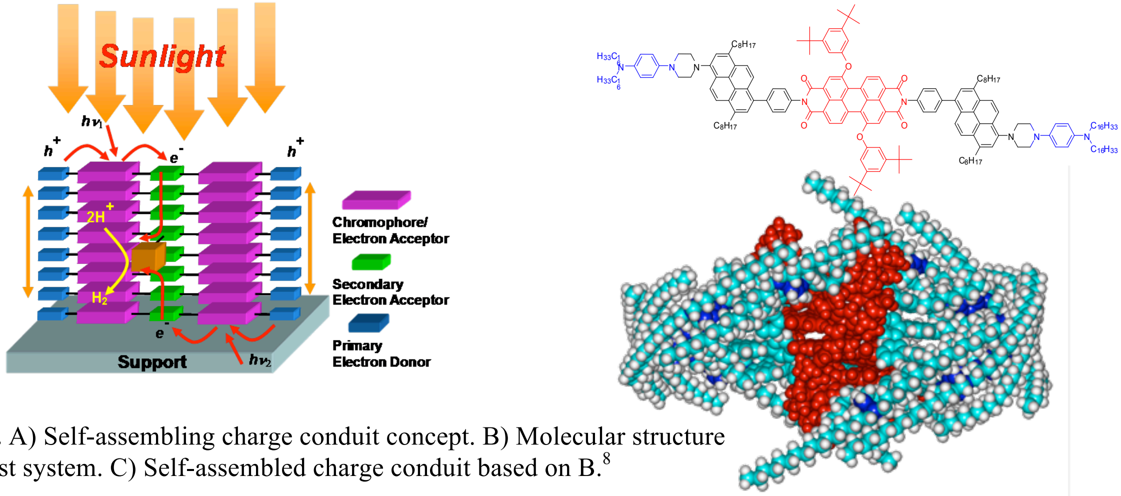
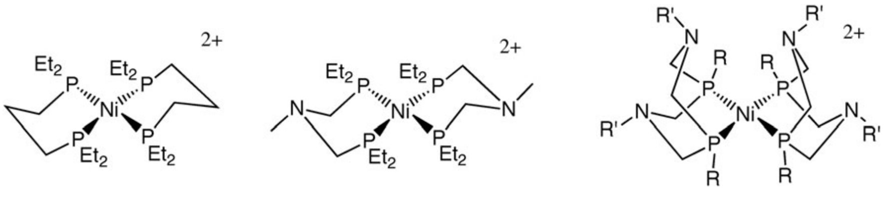


Fig. 4. A) Self-assembling charge conduit concept. B) Molecular structure of a test system. C) Self-assembled charge conduit based on B.[8]

5. *Pendant Base Incorporation in Molecular Catalysts for Hydrogen Production*

A continued theme of artificial photosynthetic research is the quest for earth-abundant, efficient catalysts for both oxygen as well as hydrogen evolution. Current state of the art synthetic hydrogen producing catalysts consist of expensive platinum metals. Therefore, to create a viable artificial photosynthetic technology, efforts to discover effective hydrogen evolution catalysts containing lower cost materials is imperative. Cheap cobalt and nickel metals have been incorporated into simple molecular derivatives, but have yielded limited hydrogen production success. As a result, research in this area has heavily focused on detailed mechanistic studies to expose the key features that increase hydrogen production and lead to the identification of better catalysts. These studies have determined that pendant bases incorporated in nickel/cobalt derivatives facilitate proton/hydride interactions as well as meet the structural requirements for



8 **9** **10**

Fig. 5. A series of nickel complexes for hydrogen production/oxidation that look to understand the effect of pendant bases on the second coordination sphere.[9]

the coupling of proton- and electron-transfer processes. Changes to the electronic and steric properties of these bases can further tune the reactivity of the catalyst.[9]

6. Singlet Fission for Enhanced Charge Generation

Theory is a powerful tool for predicting and describing chemical mechanisms, energetics, and rates. Theoretical calculations have helped to establish the maximum thermodynamic efficiency of single band-gap photovoltaic cells known as the Shockley-Queisser limit. At 1 sun, this theoretical power conversion efficiency limit is 32%. This efficiency maximum can be raised by increasing the number of excitons generated per photon absorbed. Specifically, utilization of light-absorbing molecules that are capable of singlet fission has been calculated to increase theoretical efficiencies to as much as 43%.[10] Singlet fission occurs when a molecule absorbs one photon to produce a singlet exciton, which then splits to form two triplet excitons (Fig. 6). These two excitons can then both inject charge into a semiconductor. Both theoretical and experimental research has begun to identify molecules that meet the energetic requirements for singlet fission. Discovery and development of these materials could both increase the efficiency of artificial photosynthetic systems and lower their cost by incorporating cheap, organic light absorbers.

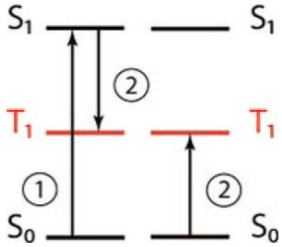


Fig. 6. Singlet fission: 1) The chromophore on the left is initially excited to S_1. 2) The excited chromophore shares its energy with the chromophore on the right, creating a T_1 state on each.[10]

Future Outlook

Artificial photosynthesis is a promising technology poised to revolutionize the availability and use of carbon-neutral, geo-politically favorable fuels. However, currently this technology remains in the research and development stages, relying heavily on the discovery of new materials and catalysts. Advances in basic research have already increased understanding of the fundamental principles that govern solar energy conversion. However, as yet, current technologies cannot produce efficient, scalable, and sustainable solar fuels that are economically viable. In the short term (five years), artificial photosynthetic fuels will continue remain in the research and development stage and rely heavily on closely-interacting teams of scientists and engineers. Long term effectiveness of artificial photosynthesis (technological capabilities by 2030) will depend heavily on the scientific advancements in the next five years that will lead to accelerated commercialization development and viability. This work must address the need to provide storable fuels that can make use of existing infrastructure. Without an immediate, intensive and focused research effort, solar fuels will have a difficult time impacting transportation fuels in the next 20 years.

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Table 1. Selection of Solar Fuels Researchers

Name	**Institution**	**Field of Expertise**
Paul Alivisatos	Lawrence Berkeley Laboratory	materials synthesis
Fraser Armstrong	University of Oxford, UK	hydrogen formation catalysts
Vincent Artero	CEA Grenoble	hydrogen formation catalysts
Harry Atwater	Caltech	nanomaterials
James Barber	Imperial College, UK	photosynthesis
Allen J. Bard	University of Texas	electrochemistry
John Birge	University of Chicago	engineering economics
Andrew Bocarsly	Princeton University	water splitting catalysts
Gary Brudvig	Yale University	water splitting catalysts
Bruce Brunschwig	Caltech	photocatalysis
Lin Chen	Argonne National Laboratory	catalyst structures
Fay Cook	Northwestern University	social policy
Marcetta Darensbourg	Texas A&M University	hydrogen formation catalysts
G. Charles Dismukes	Rutgers University	water splitting catalysts
Daniel DuBois	Pacific Northwest National Laboratory	CO_2 reduction catalysis
Kazunari Domen	University of Tokyo	photocatalysis
James Durrant	Imperial College, UK	photovoltaics
Richard Eisenberg	University of Rochester	hydrogen formation catalysts
Marc Fontecave	CEA Grenoble	hydrogen formation catalysts
Stephen R. Forrest	University of Michigan	organic photovoltaics
Heinz Frei	Lawrence Berkeley Laboratory	nanomaterials
Etsuko Fujita	Brookhaven National Laboratory	CO_2 reduction catalysis
Michael Grätzel	Ecole Polytech. Fed. de Lausanne	photovoltaics
Harry Gray	Caltech	photodriven molecular catalysts
Devens Gust	Arizona State University	photodriven catalysts
Leif Hammarström	Uppsala University	photodriven catalysts
Craig Hill	Emory University	water splitting to hydrogen
Michael Hoffman	Caltech	environmental sciences
Joseph T. Hupp	Northwestern University	photovoltaics
Ghassan Jabbour	KAUST, Saudi Arabia	photovoltaics
Rene Janssen	University of Eindhoven, Netherlands	organic photovoltaics
Paul Kenis	University of Illinois Urbana-Champaign	catalytic reactor design
Michio Kondo	AIST, Japan	photovoltaics
Phil Krein	University of Illinois Urbana-Champaign	systems engineering
Nathan Lewis	Caltech	photoelectrochemistry
Jeffrey Long	UC Berkeley	molecular catalysis
Wolfgang Lubitz	Max Planck Ins. für Bioanorg. Chemie	catalysis mechanisms
Hani Mahmassani	Northwestern University	transportation
Thomas Mallouk	Pennsylvania State University	solar fuels materials
Tobin J. Marks	Northwestern University	photovoltaics and catalysis
Eric McFarland	UC Santa Barbara	fuel cells
Michael D. McGehee	Stanford University	organic photovoltaics
Thomas Meyer	University of North Carolina	photocatalysis
David Milstein	Weizmann Institute, Israel	catalysis
Anna Moore	Arizona State University	materials for solar fuels
Thomas Moore	Arizona State University	materials for solar fuels

James Muckerman	Brookhaven National Laboratory	theory
Klaus Müllen	Max Planck Institut für Polymerforschung	organic materials
Daniel Nocera	MIT	water splitting catalysts
Jens Norskov	Stanford	theory
Arthur Nozik	National Renewable Energy Laboratory	photovoltaics
Ralph Nuzzo	University of Illinois Urbana-Champaign	photovoltaics
Bruce Parkinson	Colorado State University	photocatalysis
Michael Pellin	Argonne National Laboratory	materials synthesis and properties
Jonas Peters	Caltech	molecular catalysis
Mark A. Ratner	Northwestern University	theory and modeling
Notker Roesch	Technical University Munich	catalysis
John Rogers	University of Illinois Urbana-Champaign	electronic materials
Garry Rumbles	National Renewable Energy Laboratory	organic photovoltaics
Peter Stair	Northwestern University	CO_2 catalysis
Stenbjorn Styring	Uppsala University	photosynthesis
T. Don Tilley	UC Berkeley	molecular catalysis
John Turner	National Renewable Energy Laboratory	photovoltaics
Gregory Voth	University of Chicago	theory and modeling
Peng Wang	Changchun Inst. of Appl. Chem., China	photovoltaics
Michael R. Wasielewski	Northwestern University	photodriven catalysts
Frank Würthner	University of Würzburg	organic materials
Peidong Yang	UC Berkeley	nanomaterials
Junko Yano	Lawrence Berkeley Laboratory	catalyst structures
Luping Yu	University of Chicago	photovoltaic materials