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GUEST EDITORIAL

Artificial Photosynthesis and Solar Fuels

The development and well-being of society rely on sufficient energy supplies, and concerns about our strong dependence on fossil fuels have been rapidly increasing. At present, more than 80% of our primary energy is provided by fossil fuels. In addition to limitations in availability and geo-political problems associated with fossil fuels, the emission of green-house gases upon the use of fossil fuels has been recognized as a significant threat to our current living conditions. Thus, new sources of energy that provide a large-scale, sustainable energy supply must be developed. Solar energy is one of the few alternative energy sources that could be scaled up to meet our future demands. More solar energy reaches our planet in one hour than is consumed by mankind in a year, and consequently we would need to harvest less than 0.02% of this solar energy to fulfill all of our energy requirements. Solar energy can be converted into several different useful forms: heat, electricity, and fuels. Most of these technologies have not yet been fully explored and developed. Of particular interest is the development of solar fuels, which are concentrated energy carriers with long-term storage capacity produced by energy input from solar irradiation. These fuels would play an important role in balancing out the daily and yearly local variations in solar irradiation. Solar fuels are expected to become an important contributor to our future energy systems.

Fuels can be produced from solar energy by both indirect and direct pathways. Examples of indirect pathways include conversion of biomass to biogas, as well as hydrogen production by electrolysis of water using electricity from photovoltaics. Direct pathways produce the fuel directly in an integrated system, without intermediate energy carriers. Artificial photosynthesis is an example of a direct pathway for producing solar energy. Natural photosynthesis in green plants, algae, and cyanobacteria use solar energy to convert carbon dioxide and water to fuel, primarily energy rich carbohydrates. The basis of artificial photosynthesis is not to copy this process but rather to learn from it and reproduce the same principles in much smaller man-made systems.

A variety of fuels may be produced by artificial photosynthesis. Carbon-based fuels formed by reduction of carbon dioxide would be most attractive, but the multielectron catalytic chemistry involved makes this a challenging goal. Hydrogen production from protons is a two-electron process, which makes efficient fuel production less demanding, although its efficient storage is more difficult. Natural oxygenic photosynthesis uses water as raw material for the ultimate source of electrons in reductive fuel production. Use of this abundant substrate, however, requires mastering of the multielectron oxidation of two water molecules to molecular oxygen. This process is well-managed in Nature with good efficiency, but it has been very difficult to achieve with artificial homogeneous catalysts.

This special issue collects Accounts on natural photosynthetic principles, as well as biomimetic and artificial photosynthetic systems. The focus lies on predominantly molecular systems and processes, which could be part of an integrated artificial photosynthetic system for solar fuel production. Research directed toward understanding molecular mechanisms of photosynthetic energy conversion and biomimetic systems, which may provide strategies for artificial photosynthesis, is presented. The realization of photosynthetic principles in artificial light-harvesting and supramolecular assembly strategies for efficient light-induced charge separation in molecular and nanoscale systems is also described. Moreover, work on the coupling of artificial photosynthetic charge separation to multielectron/proton transfer and catalysis is reviewed. Finally, research on homogeneous catalysts for hydrogen production, carbon dioxide reduction, and water oxidation is presented.

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We hope that the breadth of the work compiled in this Special Issue of *Accounts of Chemical Research* will stimulate further research in artificial photosynthesis and solar fuels, an area that is both scientifically rewarding and of immediate societal urgency.

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