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Emerging Trends of Artificial Photosynthesis and Environmental Challenges

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ABSTRACT

Artificial photosynthesis is a technology with immense potential that aims to emulate the natural photosynthetic process. The process of natural photosynthesis involves the conversion of solar energy into chemical energy, which is stored in organic compounds. Catalysis is an essential aspect of artificial photosynthesis, as it facilitates the reactions that convert solar energy into chemical energy. In this review, we aim to provide an extensive overview of recent developments in the field of artificial photosynthesis by catalysis. We will discuss the various catalyst types used in artificial photosynthesis, including homogeneous catalysts, heterogeneous catalysts, and biocatalysts. Additionally, we will explore the different strategies employed to enhance the efficiency and selectivity of catalytic reactions, such as the utilization of nanomaterials, photoelectrochemical cells, and molecular engineering. Lastly, we will examine the challenges and opportunities of this technology as well as its potential applications in areas such as renewable energy, carbon capture and utilization, and sustainable agriculture.

KEYWORDS-artificial photosynthesis, environmental challenges, trends, sustainable, agriculture

I. INTRODUCTION

The photosynthetic reaction can be divided into two half-reactions of oxidation and reduction, both of which are essential to producing fuel. In plant photosynthesis, water molecules are photo-oxidized to release oxygen and protons. The second phase of plant photosynthesis (also known as the Calvin-Benson cycle) is a light-independent reaction that converts carbon dioxide into glucose (fuel). Researchers of artificial photosynthesis are developing photocatalysts that are able to perform both of these reactions. Furthermore, the protons resulting from water splitting can be used for hydrogen production. These catalysts must be able to react quickly and absorb a large percentage of the incident solar photons.^[1]

Whereas photovoltaics can provide energy directly from sunlight, the inefficiency of fuel production from photovoltaic electricity (indirect process) and the fact that sunshine is not constant throughout the day sets a limit to its use.^{[2][3]} One way of using natural photosynthesis is for the production of a biofuel, which is an indirect process that suffers from low energy conversion efficiency (due to photosynthesis' own low efficiency in converting sunlight to biomass), the cost of harvesting and transporting the fuel, and conflicts due to the increasing need of land mass for food production.^[4] The purpose of artificial photosynthesis is to produce a fuel from sunlight that can be stored conveniently and used when sunlight is not available,^[5] by using direct processes, that is, to produce a solar fuel. With the development of catalysts able to reproduce the major parts of photosynthesis, the only inputs needed to produce clean energy would ultimately be water, carbon dioxide and sunlight. The only by-product would be oxygen, and production of a solar fuel has the potential to be cheaper than gasoline.^[6]

One process for the creation of a clean and affordable energy supply is the development of photocatalytic water splitting under solar light. This method of sustainable hydrogen production is a major objective for the development of alternative energy systems.^[7] It is also predicted to be one of the more, if not the most, efficient ways of obtaining hydrogen from water.^[8] The conversion of solar energy into hydrogen via a water-splitting process assisted by photosemiconductor catalysts is one of the most promising technologies in development.^[9] This process has the potential for large quantities of hydrogen to be generated in an ecologically sound manner.^[citation needed] The conversion of solar energy into a clean fuel (H₂) under ambient conditions is one of the greatest challenges facing scientists in the twenty-first century.^[10]

Two methods are generally recognized for the construction of solar fuel cells for hydrogen production:^[11]

- A homogeneous system is one such that catalysts are not compartmentalized, that is, components are present in the same compartment. This means that hydrogen and oxygen are produced in the same location. This can be a drawback, since they compose an explosive mixture, demanding gas product separation. Also, all components must be active in approximately the same conditions (e.g., pH).
- A heterogeneous system has two separate electrodes, an anode and a cathode, making possible the separation of oxygen and hydrogen production. Furthermore, different components do not necessarily need to work in the same conditions. However, the increased complexity of these systems makes them harder to develop and more expensive.

Another area of research within artificial photosynthesis is the selection and manipulation of photosynthetic microorganisms, namely green microalgae and cyanobacteria, for the production of solar fuels. Many strains are able to produce hydrogen naturally, and scientists are working to improve them.^[12] Algae biofuels such as butanol and methanol are produced both at laboratory and commercial scales. This method has benefited from the development of synthetic biology,^[12] which is also being explored by the J. Craig Venter Institute to produce a synthetic organism capable of biofuel production.^{[13][14]} In 2017, an efficient process was developed to produce acetic acid from carbon dioxide using "cyborg bacteria".^[15]

Artificial photosynthesis is a process that mimics a very famous biochemical reaction: natural photosynthesis. The artificial photosynthesis system includes an enzyme bed reactor to fix CO₂ in the air (or any other source needing CO₂ to be removed). This reactor is fueled by hydrogen energy and bioelectric transducers. The key components of an overall artificial photosynthesis system are:

Electricity production via PV panels to meet any electrical energy needs of the systems.

Dry agriculture: carbohydrates (food), liquid fuels, chemical feedstocks, and polymers for fiber manufacture can be produced with near or absolutely minimum water requirements. Artificial photosynthesis can lower conventional agricultural water use by almost thousands of times.

Hydrogen production: electrochemical water dissociation into H₂ and O₂ is accomplished by mimicking photosynthesis.

Even though artificial photosynthesis technology is in its early stages and cannot yet be used in large-scale plants, it has a substantial prospective to reduce global water demand and substantiate clean energy systems by producing power and hydrogen from photonic energy directly.^[1,2,3]

Artificial photosynthesis as a chemical process replicates natural photosynthesis to reduce anthropogenic carbon dioxide (CO₂), increase fuel security, and provide a sustainable global economy (Faunce et al., 2013). Because of the development of some suitable technologies, we can develop artificial photosynthesis using artificial leaves for the efficient conversion of solar energy into H₂ and other fuels (Centi & Perathoner, 2010, 2011). This method may be a potential technology for H₂ production by mimicking natural photosynthesis by green leaves. Therefore, research has been carried out to harvest solar energy to produce H₂ by artificial photosynthesis (Lewis & Nocera, 2006; Nocera, 2009). In artificial photosynthesis, the artificial leaf must be able to use sunlight and water to reduce CO₂ and water into H₂ (Centi & Perathoner, 2010, 2011; Roy, Varghese, Paulose, & Grimes, 2010). Hence, artificial photosynthesis has created great interest owing to the employment of photoelectrochemical cells (PEC) to use sunlight to produce solar fuel (Bensaid, Centi, Garrone, Perathoner, & Saracco, 2012; Listorti, Durrant, & Barber, 2009; Walter et al., 2010). In practice, solar-driven electrochemical fuel generation needs the integration of light-absorbing and electrochemical components able to separate product fuels (Modestino et al., 2013). Natural photosynthesis synthesizes glucose and oxygen in the presence of sunlight. In the presence of sunlight, the excitation of chlorophyll molecules in PSII and PSI generates ATP and NADPH along the ETC with the involvement of different protein complexes. Similarly, in artificial photosynthesis, the use of solar energy to produce NADPH as an electron donor by PSI must be achieved (Amao, Hamano, & Shimizu, 2012). Thus, we have to know all biochemical pathways before constructing an artificial photosynthetic system, because natural photosynthesis consists of the involvement of several pigments, proteins, and enzymes. The main barrier is producing artificial leaves for artificial photosynthesis to mimic the overall reactions of natural photosynthesis (Cogdel et al., 2010).^[4,5,6]

In artificial photosynthesis, model molecular systems, electron donor and acceptor assemblies, solar energy conversion by photo-induced charge separation, examination of photoelectrochemical cells to convert solar energy into fuels, light-harvesting component, charge separation, and catalysis in photosynthesis have been compared with natural photosynthesis. The three key components for solar energy conversion in artificial photosynthesis are light harvesting, charge separation, and catalysis (McConnell, Li, & Brudvig, 2010). According to Nocera (2012), a light-dependent reaction is required to mimic photosynthesis. Hence, artificial leaves were constructed by adding catalysts (Co-OEC

and NiMoZn) to a light-harvesting silicon-based semiconductor to split water. Those silicon semiconductors in artificial leaves may be considered photosynthetic membranes in natural photosynthesis. The silicon junction harvests sunlight and converts it into a wireless current through Si. Then, Co-OEC is integrated with Si that functionalizes single-junction PEC. Several photochemical reactions that occur in this solar cell generate the photovoltage used for the water-splitting reaction to generate H₂. In addition, electrocatalysts have been designed for a light-driven charge separation system for solar fuel production (Tran, Wong, Barber, & Loo, 2012). [7,8,9] Based on the current situation, the concept of an artificial leaf has been developed as a source of clean fuel. An artificial leaf can be developed using a silicon-coated sheet that splits water into hydrogen and oxygen. Silicon has become an attractive material with which to design an artificial leaf because silicone is cheap and abundant and such cells can capture and store energy. Although the development of an artificial leaf is still a scientific challenge, some concepts have been developed to overcome technical difficulties. To overcome those difficulties, some researchers have focused on artificial leaves for artificial photosynthesis to generate clean hydrogen. Hybrid strategies for solar water splitting based on dye-sensitized photoelectrosynthetic cells (Alibabaei et al., 2013), by proposing natural to artificial photosynthesis with artificial photocatalysts, hybrid photocatalysts for water oxidation/proton reduction and hydrogen evolution, as well as construction of complete photocatalytic system for hydrogen and oxygen evolution from water (Barber and Tang, 2013), used a photovoltaic reactor for artificial photosynthesis, based on water electrolysis to produce high-energy hydrogen atoms (Nong et al., 2014). An artificial photosynthetic system using TiO₂ was developed that has the features of photosynthesis to overcome the challenge of solar-driven water splitting and CO₂ reduction. In addition, the photoreduction of CO₂ into hydrocarbon fuels (CO and CH₄) has been demonstrated, which solves the architectural model of artificial photosynthesis to produce solar cells, fuel cells, or battery electrodes (Zhou et al., 2013).[7,8,9]

II. DISCUSSION

The Joint Center for Artificial Photosynthesis developed an idea for artificial photosynthesis using a water-splitting photoconversion system that produces H₂ directly from water. As in plants, water oxidation takes place in PSII with the help of visible light and electrons are transferred to the PSI with the help of electron transport mediators in which carbon dioxide is reduced to carbohydrates during a dark reaction. Plants prepare food (carbohydrates) with the help of water and carbon dioxide in the presence of visible light. Artificial photosynthesis deals with the understanding of the molecular aspects of mimicking the different steps present in natural photosynthesis to use sunlight in driving the conversion of CO₂ and H₂O to (typically) carbohydrates and oxygen. Molecular assemblies bioinspired from photosynthesis have been studied by various research groups, for example, Djokic and Soo [9], Wang et al. [10], Whang and Apaydin [11], and Barber [17]. These systems contain a chromophore, such as a porphyrin, which performs the first step of light harvesting. These molecules are covalently linked to one or more electron acceptors, such as fullerenes or quinones, and secondary electron donors. After chromophore excitation, the photoinduced electron transfer generates a primary charge-separation state. Electron transfer chains spatially separate the redox equivalent and reduce electronic coupling, slowing recombination of the charge-separated state to make possible their use in the redox catalytic processes.[10,11,12]

The reaction rates of the redox catalytic step are typically two or even more orders of magnitude slower than the charge creation/separation processes. This aspect is the critical issue in artificial photosynthesis processes, because these charged species, if not quickly consumed in the redox processes, can recombine, reducing the overall efficiency, or may instead favor side reactions, including the degradation of the components of the artificial leaf device itself. For these reasons, more robust inorganic systems have to be preferred over organic complexes and supramolecular systems. These allow a better understanding, but still, over 20 years of studies have failed to produce practically applicable artificial leaf devices, e.g., systems with a high enough rate of conversion and stability. The contributions of these studies to understanding relevant aspects, such as (1) antenna effects, by using chromophores that absorb light throughout the whole visible spectrum and then transfer electrons or energy to the charge-separation component, to increase overall efficiency, and (2) advanced catalytic redox centers with high turnover number, have to be remarked. Progress has also been made in supramolecular assembling of these components and mimicking of natural photochemical systems, but still progress is limited in crucial elements like those for the control and photoprotective elements borrowed from photosynthesis. These are the key elements for the self-regeneration behavior of natural systems and thus a key factor in the long-term stability.

The concept of artificial leaves is instead focused on the system architecture as the basis for the design of the devices, rather than on the development of the single elements and their subsequent assembling into a functional device. The different perspective implies a higher consideration of the system functionality, rather than that of the single elements, but in artificial leaf devices there is a mutual influence between these elements determining the overall behavior. The system performance is not derived by simply putting in sequence the single components. In addition, a system approach emphasizes aspects such as charge transport and system reliability, which are instead less investigated in an artificial

photosynthesis approach. These aspects are amplified in importance in moving from water splitting to the more challenging conversion of CO₂.

On the other hand, the complexity of the problems to be addressed indicates the need to simplify the problem by analyzing single aspects, while having a clear picture of the full device, and especially of the constraints related to its applicability, in terms of materials, cost effectiveness, possibility of recovering the products, etc. For this reason, we prefer to speak about artificial photosynthetic leaves (APLs), i.e., the need to combine the two types of approaches.[13,14,15]

In energy terms, natural photosynthesis can be divided in three steps:^{[11][23]}

- Light-harvesting complexes in bacteria and plants capture photons and transduce them into electrons, injecting them into the photosynthetic chain.
- Proton-coupled electron transfer along several cofactors of the photosynthetic chain, causing local, spatial charge separation.
- Redox catalysis, which uses the aforementioned transferred electrons to oxidize water to dioxygen and protons; these protons can in some species be utilized for dihydrogen production.

A triad assembly, with a photosensitizer (P) linked in tandem to a water oxidation catalyst (D) and a hydrogen evolving catalyst (A). Electrons flow from D to A when catalysis occurs.

Using biomimetic approaches, artificial photosynthesis tries to construct systems doing the same type of processes. Ideally, a triad assembly could oxidize water with one catalyst, reduce protons with another and have a photosensitizer molecule to power the whole system. One of the simplest designs is where the photosensitizer is linked in tandem between a water oxidation catalyst and a hydrogen evolving catalyst:

- The photosensitizer transfers electrons to the hydrogen catalyst when hit by light, becoming oxidized in the process.
- This drives the water splitting catalyst to donate electrons to the photosensitizer. In a triad assembly, such a catalyst is often referred to as a donor. The oxidized donor is able to perform water oxidation.

The state of the triad with one catalyst oxidized on one end and the second one reduced on the other end of the triad is referred to as a charge separation, and is a driving force for further electron transfer, and consequently catalysis, to occur. The different components may be assembled in diverse ways, such as supramolecular complexes, compartmentalized cells, or linearly, covalently linked molecules.^[11]

Research into finding catalysts that can convert water, carbon dioxide, and sunlight to carbohydrates or hydrogen is a current, active field. By studying the natural oxygen-evolving complex (OEC), researchers have developed catalysts such as the "blue dimer" to mimic its function. However, these catalysts are still inefficient.^[6]

Photoelectrochemical cells that reduce carbon dioxide into carbon monoxide (CO), formic acid (HCOOH) and methanol (CH₃OH) are under development.^[53] Similar to natural photosynthesis, such artificial leaves can use a tandem of light absorbers for overall water splitting or CO₂ reduction. These integrated systems can be assembled on lightweight, flexible substrates, resulting in floating devices resembling lotus leaves.^[54]

Phycobilitproteins from algae are under development for renewable energy production.^[55]

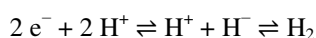
III. RESULTS

Artificial photosynthesis is envisioned as a promising strategy to convert sunlight, a practically unlimited and sustainable source of energy, into chemical fuels. In this scheme, the oxidation of water molecules is necessary to provide the electrons that will be employed in the synthesis of chemical fuels. Water oxidation is a particularly challenging reaction because it is a thermodynamically uphill multielectronic process with large activation barriers, but it is key for the realization of artificial photosynthesis because water is the only earth abundant molecule that can provide electrons in a massive and sustainable manner. Therefore, catalysts are needed for eluding the large intrinsic kinetic barriers of the reaction. In nature, water oxidation is catalyzed by a Mn tetrameric species, which enables O–O formation under the inherent mild physiological conditions through a putative high valent manganese oxo species. Taking natural water oxidation as model, molecular catalysts operating under homogeneous conditions have been explored with the objective of providing basic understanding at molecular scale of the factors that govern this reaction, which eventually will receive utility in the design of efficient water oxidation devices. [16,17,18]Traditionally, water oxidation has been studied with ruthenium and manganese based systems, but more recently attention has been shifted

toward catalysts based on iridium and first row transition metals: the former due to their extraordinary performance and the latter because of their favorable cost, availability and environmental impact when compared with second and third row transition metals. The topic has been very actively explored and important lessons have been gained. Catalysts based on first row transition metals pose specific problems in terms of stability because generally their metal-ligand bonds are labile and because reaching their high oxidation states require high oxidation potentials. Consequently, high valent states of these metals are exceedingly reactive, readily prone to engage in oxidative decomposition paths. Catalyst design is crucial for circumventing these problems and has enabled the discovery of extraordinarily reactive yet reasonably stable catalysts, comparable to the best examples based on second and third row transition metals. The following paper reviews key contributions to the field. The manuscript does not intend to be comprehensive, but instead, selected and in our opinion representative examples are discussed.

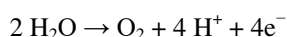
Artificial photosynthesis is referred as “a mimic of photosynthesis process” that harvest natural sunlight and in turn convert the CO₂ into fuels like hydrogen and carbon based. The hydrogen is produced from water splitting while carbon-based fuels like methanol, methane, or CO are usually produced from the reduction of CO₂ in the presence of water. Since hydrogen production will be discussed as separate section, this section will focus on the reduction of CO₂ to solar fuels and their application in other industries.

Photocatalyst, a light-driven material is widely used to realize the photocatalytic conversion of CO₂ under sunlight irradiation. The CO₂ conversion using photocatalyst was achieved through many means and some includes: CO₂-saturated aqueous solution containing suspended TiO₂ [38,39], high-pressure CO₂ system with TiO₂ powders suspended in isopropyl alcohol [40], and humidified gas-phase CO₂ [19,20,21]. Among these, the gas-phase conversion overwhelms the rest owing to its strong commercial ability, that is, zero recovery of catalyst from aqueous phase and overcome the low solubility of CO₂. As like artificial photosynthesis, this process reduces CO₂ through the photoexcited electrons (e⁻) in the presence of water (H₂O) to yield energy-bearing products such as methane (CH₄), methanol (CH₃OH), formaldehyde (HCHO), formic acid (HCOOH), etc. [22]. Hydrogen is the simplest solar fuel to synthesize, since it involves only the transference of two electrons to two protons. It must, however, be done stepwise, with formation of an intermediate hydride anion:



The proton-to-hydrogen converting catalysts present in nature are hydrogenases. These are enzymes that can either reduce protons to molecular hydrogen or oxidize hydrogen to protons and electrons. Spectroscopic and crystallographic studies spanning several decades have resulted in a good understanding of both the structure and mechanism of hydrogenase catalysis.^{[56][57]} Using this information, several molecules mimicking the structure of the active site of both nickel-iron and iron-iron hydrogenases have been synthesized.^{[11][58]} Other catalysts are not structural mimics of hydrogenase but rather functional ones. Synthesized catalysts include structural H-cluster models,^{[11][59]} a dirhodium photocatalyst,^[60] and cobalt catalysts.^{[11][61]}

Water oxidation is a more complex chemical reaction than proton reduction. In nature, the oxygen-evolving complex performs this reaction by accumulating reducing equivalents (electrons) in a manganese-calcium cluster within photosystem II (PS II), then delivering them to water molecules, with the resulting production of molecular oxygen and protons:



Without a catalyst (natural or artificial), this reaction is very endothermic, requiring high temperatures (at least 2500 K).^[8]

The exact structure of the oxygen-evolving complex has been hard to determine experimentally.^[62] As of 2011, the most detailed model was from a 1.9 Å resolution crystal structure of photosystem II.^[63] The complex is a cluster containing four manganese and one calcium ions, but the exact location and mechanism of water oxidation within the cluster is unknown. Nevertheless, bio-inspired manganese and manganese-calcium complexes have been synthesized, such as [Mn₄O₄] cubane-type clusters, some with catalytic activity.^[64]

Some ruthenium complexes, such as the dinuclear μ-oxo-bridged "blue dimer" (the first of its kind to be synthesized), are capable of light-driven water oxidation, thanks to being able to form high valence states.^[11] In this case, the ruthenium complex acts as both photosensitizer and catalyst. This complexes and other molecular catalysts still attract researchers in the field, having different advantages such as clear structure, active site, and easy to study mechanism. One of the main challenges to overcome is their short-term stability and their effective heterogenization for applications in artificial photosynthesis devices.^[65]

Many metal oxides have been found to have water oxidation catalytic activity, including ruthenium(IV) oxide (RuO_2), iridium(IV) oxide (IrO_2), cobalt oxides (including nickel-doped Co_3O_4), manganese oxide (including layered MnO_2 (birnessite), Mn_2O_3), and a mix of Mn_2O_3 with CaMn_2O_4 . Oxides are easier to obtain than molecular catalysts, especially those from relatively abundant transition metals (cobalt and manganese), but suffer from low turnover frequency and slow electron transfer properties, and their mechanism of action is hard to decipher and, therefore, to adjust.^[7]

Recently Metal-Organic Framework (MOF)-based materials have been shown to be a highly promising candidate for water oxidation with first row transition metals.^{[66][67]} The stability and tunability of this system is projected to be highly beneficial for future development.^[68]

V. CONCLUSION

However, the primary and foremost challenge is the lack of active and stable multifunctional catalysts that can efficiently harvest abundant solar energy. On the other hand, interfacial charge separation and CO_2 activation to facilitate proton-coupled electron transfer for the generation of fuels is also a part of this challenge. Further the conversion is uphill reaction processes ($\Delta G > 0$) which require high energy input due to its ultrahigh stability [26,27,28]. Although the existing technology of CO_2 conversion is far from practicability, numerous researches have been conducted to address the challenges in realizing this process for industrial application. Most of the reports focused on the development of novel photocatalyst are efficient in capturing and channeling sunlight. Numerous approaches like low dimension, porous structured, layered structured, hierarchical ordered, or hybrid-structured photocatalysts have been developed to improve the performance of CO_2 reduction [23,24,25]. In addition, the concept of employing cocatalysts and plasmonic photocatalysts has been reported in the recent years [29]. The catalytic system employed includes highly ordered mesoporous materials (SnO_2 and TiO_2) with 3D pore system embedded with either of this CdS, CdSe, ZnS, or ZnSe [31,32,33]. Among them, ZnS embodiment showed the highest yield rate for CH_4 ($3.620 \mu\text{mol gcat}^{-1} \text{h}^{-1}$) while the CdSe showed for CO ($5.884 \mu\text{mol gcat}^{-1} \text{h}^{-1}$).

Li and coworkers in 2016 proposed a dual-chamber reactor configuration separated by two-side Cu_2O /graphene/TNA photocatalyst along with proton exchange membrane [30]. Nature uses pigments, mainly chlorophylls, to absorb a broad part of the visible spectrum. Artificial systems can use either one type of pigment with a broad absorption range or combine several pigments for the same purpose.

Ruthenium polypyridine complexes, in particular tris(bipyridine)ruthenium(II) and its derivatives, have been extensively used in hydrogen photoproduction due to their efficient visible light absorption and long-lived consequent metal-to-ligand charge transfer excited state, which makes the complexes strong reducing agents.^[11] Other noble metal-containing complexes used include ones with platinum, rhodium and iridium.^[11]

Metal-free organic complexes have also been successfully employed as photosensitizers. Examples include eosin Y and rose bengal.^[11] Pyrrole rings such as porphyrins have also been used in coating nanomaterials or semiconductors for both homogeneous and heterogeneous catalysis.^{[7][53]}

As part of current research efforts artificial photonic antenna systems are being studied to determine efficient and sustainable ways to collect light for artificial photosynthesis. Gion Calzaferri (2009) describes one such antenna that uses zeolite L as a host for organic dyes, to mimic plant's light collecting systems.^[69] The antenna is fabricated by inserting dye molecules into the channels of zeolite L. The insertion process, which takes place under vacuum and at high temperature conditions, is made possible by the cooperative vibrational motion of the zeolite framework and of the dye molecules.^[70] The resulting material may be interfaced to an external device via a stopcock intermediate[31,32,33]

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