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Comment

Photocatalytic CO_2 conversion: Beyond the earth

Jingxiang Low [a,b], Chao Zhang [a], Ferdi Karadas [c], Yujie Xiong [a,*]

[a] *Hefei National Research Center for Physical Sciences at the Microscale, School of Chemistry and Materials Science, University of Science and Technology of China, Hefei 230026, Anhui, China*

[b] *Multidisciplinary Platform of Advanced Engineering, Chemical Engineering Discipline, School of Engineering, Monash University Malaysia, Bandar Sunway 47500, Selangor, Malaysia*

[c] *Department of Chemistry and National Nanotechnology Research Center, Bilkent University, Ankara 06800, Turkey*

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ABSTRACT

The issue of climate change attributed to CO_2 emissions has led to increased attention towards the study and development of artificial photosynthesis through photocatalytic CO_2 conversion to reconstruct the broken carbon cycle in nature. Photocatalytic CO_2 conversion can simultaneously reduce the CO_2 concentration in the atmosphere and produce valuable hydrocarbon fuels. With the recent discovery of abundant reserves of CO_2 and water at extraterrestrial sites, it has been proposed that photocatalytic CO_2 conversion can also be implemented at extraterrestrial sites to build up an artificial carbon cycle for providing propellants and life support for space missions. This comment presents our perspectives on the development of photocatalytic CO_2 conversion beyond Earth, with a focus on its general principles and potential challenges that may arise at extraterrestrial sites. Finally, a brief overview of the future research directions in this field is presented.

1. Introduction

Space exploration involves the discovery of outer space *via* manned or unmanned missions to expand the scientific knowledge and the potential for human habitation beyond Earth. Currently, space missions primarily rely on the transportation of basic needs from the Earth, which is extremely challenging and logistically impossible when we aim to develop stations or habitats on extraterrestrial sites, such as the Moon and Mars [1,2]. For example, astronauts need almost a kilogram of oxygen per day to sustain their lives. Therefore, tons of oxygen must be transported every year to build stations at extraterrestrial sites, raising the cost and risk of the mission [3,4]. In this context, *in situ* resource utilization (ISRU), which aims to rationally utilize extraterrestrial resources, has been proposed to reduce the burden of transporting earth-based resources and promote sustainable development at extraterrestrial sites. As such, the ISRU is regarded as a crucial technological advancement in the area of human space exploration.

At the present stage of ISRU development, carbon-based resources have attracted wide attention. Typically, carbon is the most essential element for organisms on Earth, as it not only serves as the primary component of all known life but also as an energy medium (*e.g.*, natural gas and petroleum) to power the Earth. These roles of carbon on Earth are achieved *via* the carbon cycle, which enables the transfer of carbon atoms from the atmosphere (present in gaseous carbon compounds such as CO_2 and CH_4) to Earth (present in the form of sugar, starch, *etc.*) and finally back to the atmosphere, thus completing the loop. Solar energy serves as the energy source for these

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* Corresponding author. E-mail: yjxiong@ustc.edu.cn (Y. Xiong).

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biogeochemical cycles, wherein plants and other organisms absorb the solar energy to assimilate CO_2 and H_2O and produce carbon-based compounds and oxygen *via* photosynthesis [5]. Considering the abundant sunlight irradiation and the presence of abundant CO_2 and H_2O reserves [6,7] at the currently targeted extraterrestrial sites (*i.e.*, the Moon and Mars), such a photosynthesis strategy can be adopted to build artificial carbon cycle systems at extraterrestrial sites to provide sufficient propellant and life support for space missions. In contrast to other potential strategies such as water splitting, which are limited to the production of gaseous propellants, this technique has the potential to generate gaseous, liquid, and solid products, thereby expanding its scope.

Given this background, artificial photosynthesis through photocatalytic CO_2 conversion holds great promise for achieving a sustainable cycle [8–10]. This strategy has the ability to imitate the role of photosynthesis in green plants and is expected to reconstruct the carbon cycle on Earth, which is currently disrupted by excess CO_2 emissions. This artificial photosynthesis strategy, if successfully implemented at extraterrestrial sites as a part of the ISRU (Fig. 1(a)) [11], can also allow the artificial carbon cycle to be built at such sites (Fig. 1(b)). Thus far, various products have been successfully obtained *via* photocatalytic CO_2 conversion, including CO, CH_4, CH_3OH, and HCHO [12]. However, the photocatalytic CO_2 conversion efficiency remains unsatisfactory for practical applications. Thus, the development of photocatalytic CO_2 conversion with excellent photoconversion efficiency and product selectivity is highly desired for applications not only on Earth but also on extraterrestrial sites.

This comment presents brief and clear guidelines for the development of photocatalytic CO_2 conversion and its applications beyond the Earth. First, the fundamental principles of photocatalytic CO_2 conversion are outlined. Then, a summary of the problems encountered in photocatalysis during its implementation at extraterrestrial sites is provided. Finally, a perspective on the development of this field is provided.

2. General principles of photocatalytic CO_2 conversion

2.1. Adsorption/activation of CO_2

The CO_2 bound to the surface of the photocatalysts has a higher chance of being reacted to form the intermediate compounds (Fig. 2(a), process (i)) [13–15]. Typically, according to the adsorption strength, the adsorption of CO_2 can be categorized into physisorption and chemisorption. The physisorption of CO_2 on photocatalysts is rather weak and contributes only marginally to the surface reaction. In the chemisorption of CO_2, CO_2 molecules can be adsorbed on specific binding sites of the photocatalysts by establishing chemical bonds that weaken the C=O bond strength and facilitate the subsequent conversion process.

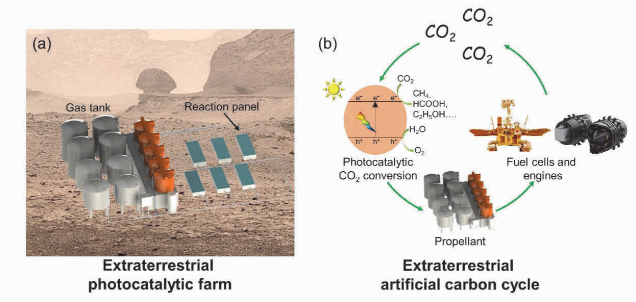

Fig. 1. Schematic for extraterrestrial artificial photosynthesis (i.e., photocatalysis) farm (a) and extraterrestrial artificial carbon cycle (b).

2.2. Excitation of the semiconductor

The electrons required for the reduction of CO_2 during photocatalysis are mainly derived from the excitation of semiconductors [16]. Therefore, a semiconductor should be photoexcited during the reaction to produce energetic photogenerated electron-hole pairs (Fig. 2(a), process (ii)). It is essential that the bandgap of the photocatalyst be neither too large nor too small. If the bandgap is too large, it can hardly be photoexcited by visible light, which constitutes the majority of incident sunlight. If the bandgap is too small, the reduction or oxidation potential of the semiconductor can be weak and insufficient to drive reduction and oxidation reactions, respectively. Recently, it was discovered that certain semiconductors on the regolith of the lunar or Martian surface meet this requirement, suggesting their potential to directly drive photocatalytic CO_2 conversion to provide propellant and oxygen for space missions.

2.3. Separation/migration of the photogenerated charge carriers

The separation and migration of photogenerated charge carriers are the most critical aspects that determine their utilization for CO_2 conversion and can eventually determine the photocatalytic CO_2 conversion efficiency [17,18]. Specifically,

Yujie Xiong *(University of Science and technology of China) received his Ph.D. in inorganic chemistry in 2004 from the University of Science and Technology of China (USTC). From 2004 to 2009, he worked as a postdoctoral fellow at the University of Washington in Seattle and as a research associate at the University of Illinois at Urbana-Champaign. He was the principal scientist at the Washington University in St. Louis in 2009–2011. He joined the USTC in 2011, and currently is the Chair Professor of Chemistry. His research centers on solar-driven artificial carbon cycle.*

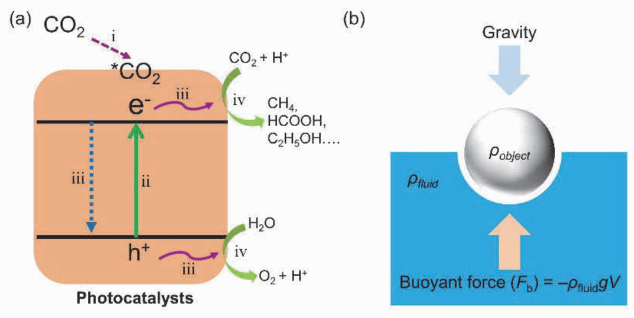

Fig. 2. Schematic illustration for photocatalytic CO_2 conversion pathways (a), including adsorption/activation of CO_2 (i), photoexcitation of photocatalyst (ii), separation/migration of photogenerated charge carriers (iii), and stabilization and conversion of intermediates (iv), and buoyancy (b), where ρ_{fluid}, ρ_{object}, g and V are fluid density, object density, acceleration due to gravity, and fluid volume, respectively.

the photogenerated electrons and holes have oppositely charged carriers, making them prone to annihilation, which reduces their ability to participate in photocatalytic CO_2 conversion (Fig. 2(a), process (iii)). Therefore, numerous strategies have been adopted to enhance the photogenerated charge carrier separation efficiency of photocatalysts [19,20]. Notably, the accumulation of (or migration of) photogenerated charge carriers at specific photocatalytic sites can also determine the selectivity of the photocatalytic CO_2 conversion reaction. For instance, the high density of photogenerated electrons at a specific site is beneficial for multi-electron reactions on photocatalysts to produce CH_4 (8 electron reaction), C_2H_4 (12 electron reaction), and C_2H_5OH (12 electron reaction).

2.4. *Stabilization of the intermediates*

Intermediates are the species formed during the surface reaction and product generation (Fig. 2(a), process (iv)). In a typical photocatalytic CO_2 conversion reaction, the intermediates can vary according to the reaction conditions, such as the photocatalyst, reaction phase (gas or water), light intensity, and temperature. Additionally, it is widely recognized that intermediates are the key to determining the selectivity of the photocatalytic CO_2 conversion reaction [21–23]. The photocatalytic CO_2 conversion reaction exhibits two extreme instances of weak and strong adsorption of intermediates. In weak intermediate adsorption, CO is normally produced because of the weak adsorption of intermediates on the surface of the photocatalyst, and the formed *CO intermediates can be rapidly desorbed from the reaction sites. For strong intermediate adsorption, CH_4 is typically produced. This was attributed to the strong adsorption of intermediates such as *CO, *CH, $*CH_2$, and $*CH_3$ on the photocatalysts, which resulted in the over-reduction of the intermediates to the low-oxidation state of carbon (i.e., CH_4). Thus, meticulous control of the intermediates must be achieved to obtain high-value products, particularly C_{2+} products such as C_2H_4 and C_2H_6O.

3. Issues encountered at the extraterrestrial sites

3.1. *Source of photocatalysts*

Although most currently available photocatalysts have demonstrated high stability and long-term application capabilities, they are not readily available at extraterrestrial sites. Therefore, it is important to develop a strategy for *in situ* resource utilization in photocatalyst preparation to reduce the transportation load of spacecraft. To achieve this, an extensive understanding of the composition of resources available at extraterrestrial sites is a prerequisite [24,25]. Recent blooming-space missions with sample returns have enhanced our understanding, thereby alleviating this problem. Taking the Moon as an example, our research group comprehensively analyzed the lunar regolith brought back by the Chang'E 5 (CE-5) mission [26], revealing that augite, plagioclase, olivine, and ilmenite are the primary constituents of the regolith. Furthermore, we demonstrated that CE-5 lunar soils can be directly utilized for photocatalytic CO_2 conversion for the production of hydrocarbon fuels and oxygen, demonstrating tremendous potential for using lunar soils as photocatalysts.

3.2. *Microgravity*

On Earth, gravitational force causes buoyancy (Fig. 2(b)), which eventually allows the formed gaseous products (*e.g.*, CH_4 and O_2) to detach from the photocatalysts after the reaction [27]. However, the situation is different at extraterrestrial sites [28]. For example, the gravitational acceleration on the Moon is only 1.625 $m\ s^{-2}$, approximately 16.66% of that on Earth. Under such conditions, the generated products can hardly be desorbed from the photocatalysts, blocking the surface-active sites for the next cycle of the reaction. In addition, the absence of convection flows induced by buoyancy may cause various problems in liquid-phase reactions, such as slow reaction kinetics and greatly suppressed photocatalytic performance. Therefore, various strategies, such as rotating and stirring the reaction system, have been proposed to create a gravitational force for phase separation to overcome these potential limitations.

3.3. *Extraterrestrial radiation*

Owing to the absence of an Earth-like atmosphere, the distribution of incident radiation at extraterrestrial sites can be different from that at the Earth's surface [29]. Taking Mars as an example, the thin CO_2 atmosphere can only screen for ultraviolet (UV) radiation shorter than 200 nm, allowing high intensities of UV-A, UV-B, and UV-C to reach its surface. This can be advantageous for the photocatalytic CO_2 conversion reaction because the incident light has high energy and can be employed for activating semiconductors with large bandgap values, which have longer photogenerated charge carrier lifetimes and stronger reduction-oxidation capabilities for the conversion of reactants. Therefore, the rational use of high-energy radiation should always be considered during the study of photocatalytic CO_2 conversion. In addition, the influence of extraterrestrial radiation on the stability and long-term application of photocatalytic CO_2 conversion should be systematically studied.

3.4. Temperature

Due to the absence of atmosphere on their surfaces, most extraterrestrial sites have a large day-night temperature difference (*e.g.*, approximately −133–27 °C for the Mars and approximately −130–120 °C for the Moon) [30,31]. Although a high temperature can be beneficial for facilitating surface reactions during photocatalytic CO_2 conversion, a low temperature can result in system malfunction. Therefore, thermal insulation is another critical problem that must be solved during the implementation of photocatalytic systems at extraterrestrial sites.

4. Conclusion and future perspectives

In summary, an artificial carbon cycle with photocatalytic CO_2 conversion as a key link can potentially provide fuel and oxygen during space missions. The recent deployment of several national or international space exploration projects has underscored the pressing need for successful photocatalytic CO_2 conversion to establish a carbon cycle at extraterrestrial sites. Therefore, there are numerous challenges in enhancing the efficiency and selectivity of the reaction as well as improving the workability and practicability of the system at extraterrestrial sites. Future research on photocatalytic CO_2 conversion should focus on the following areas:

(1) Poor photocatalytic CO_2 conversion efficiency and selectivity are the main reasons limiting its wide application, irrespective of the Earth or other extraterrestrial sites. Therefore, the photocatalytic CO_2 conversion efficiency must be further improved to meet practical requirements (*e.g.*, the benchmark photoconversion efficiency of 10% set by the US Department of Energy for commercialization). In addition, the selectivity of photocatalytic CO_2 conversion toward C_{2+} products, which have a high density and value, should always be targeted.

(2) The study of photocatalytic CO_2 conversion at analog habitats of extraterrestrial sites under extreme conditions, such as microgravity, cosmotic radiation, and extreme temperatures, should be carried out during these studies. Data regarding the efficiency, selectivity, and stability of the system should be collected as a reference for the future use of photocatalytic CO_2 conversion systems at extraterrestrial sites. Studies of the effects of extraterrestrial site environments on photocatalytic CO_2 conversion are among the most important directions in this field.

(3) The linking of photocatalytic CO_2 conversion systems with other power systems that can convert chemical energy into other forms of energy (*e.g.*, mechanical energy or electrical energy) must be developed to complete the artificial carbon cycle at extraterrestrial sites. Numerous alternatives exist for producing energy from hydrocarbons, such as fuel cells and engines. The workability of these technologies at extraterrestrial sites should also be investigated.

Declaration of interests

The authors declare no competing interests.

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Graphical Abstract

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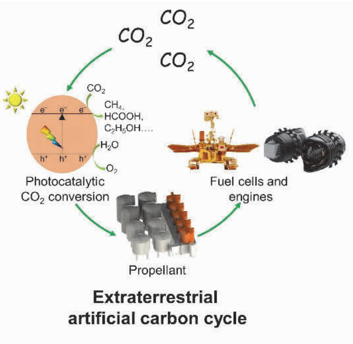
Photocatalytic CO_2 conversion: Beyond the earth

Jingxiang Low, Chao Zhang, Ferdi Karadas, Yujie Xiong *
University of Science and Technology of China, China;
Monash University Malaysia, Malaysia;
Bilkent University, Turkey



The discovery of CO_2 and H_2O reserves in extraterrestrial sites has prompted research on the implementation of extraterrestrial photocatalytic CO_2 conversion. This comment offers insights on extraterrestrial photocatalytic CO_2 conversion.

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地外光催化CO_2转化

刘敬祥[a,b], 张　超[a], Ferdi Karadas[c], 熊宇杰[a,*]

[a]中国科学技术大学合肥微尺度物质科学国家研究中心, 化学与材料科学学院, 安徽合肥230026, 中国

[b]马来西亚莫纳什大学工程学院化工学科多学科先进工程平台, 马来西亚

[c]毕尔肯特大学化学系和国家纳米技术研究中心, 土耳其

摘要: 随着人类社会对CO_2排放引起的气候变化问题愈发关注, 光催化CO_2转化因其能模拟自然界中的光合作用受到广泛研究, 以帮助修复自然界中已被破坏的碳循环. 光催化CO_2转化可以减少大气中的CO_2浓度, 并合成具有高价值的碳氢化合物燃料. 近年来, 随着地外空间发现了丰富储量的CO_2和水, 科学家提出了在地外也可以进行光催化CO_2转化反应, 进而在地外建立人工碳循环, 为太空任务提供推进剂和生命保障. 本文围绕光催化CO_2转化反应在地外空间应用的可行性进行探讨, 阐述光催化CO_2转化的基本过程, 包括CO_2分子的吸附与活化、半导体的激发、光生载流子的迁移和分离以及反应中间物种的稳定. 同时, 阐述了光催化CO_2转化在地外应用过程中可能存在的问题, 具体包括光催化剂的来源问题、地外微重力问题、地外辐射问题以及温度与温差问题.

随着近几年各国深空探测项目的提出与部署, 在地外空间探究光催化CO_2转化建立人工碳循环的可行性已成为一项紧迫的任务. 因此, 本文展望了地外光催化CO_2转化的未来发展方向, 具体包括(1)进一步提升光催化CO_2转化性能与选择性, (2)探究地外条件与因素对光催化CO_2转化过程的影响与(3)将光催化CO_2转化反应与其他技术匹配以拓宽相关的应用.

关键词: 光催化; CO_2转化; 碳循环; 原位资源化利用; 碳氢燃料

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*通讯联系人. 电子信箱: yjxiong@ustc.edu.cn (熊宇杰).

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