

Review Article

Highlighting Photocatalytic H₂-Production from Natural Seawater and the Utilization of Quasi-Photosynthetic Absorption as Two Ultimate Solutions for CO₂ Mitigation

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This paper reviews five potential solutions for CO_2 mitigation and theoretically analyzes related outstanding questions. Emission trading under the global mitigation objectives and policies is in dilemma because reducing emission while utilizing fossil fuels is a difficult balance. CO_2 capture and sequestration (CCS) technique offers a comprehensive solution, but it is risky and expensive. Analyses of the 100% renewable energy plan suggest hydrogen as a fuel of zero CO_2 emission. Photocatalytic splitting of natural seawater can be an ultimate scheme for the hydrogen production, providing that the associated technological constraints would be overcome. Soil CO_2 absorption in the arid regions (terms such absorption "quasi-photosynthetic absorption") implies a potential solution, but the kinetics and overall importance are undetermined. Further investigations of these unresolved issues are strongly needed to realize the global CO_2 mitigation target. Comparing the five potential solutions, photocatalytic H₂-production from natural seawater and the utilization of quasi-photosynthetic absorption are highlighted as two ultimate solutions.

1. Introduction

The burning of fossil fuels after the Industrial Revolution accelerated the polar ice cap melting and caused an increase of atmospheric CO_2 concentrations [1]. This will in turn cause an acceleration of the sea level rise. Ecosystems releasing/absorbing CO_2 to the atmosphere are therefore expected to be important to the future global/regional climate changes [2]. It is necessary to consider the anthropogenic impact on global climate by comparing the potential effect of human activities and their potentially negative aggregate effects on economic growth [3–5]. To make things worse, reproductive effort and seed quality also have significant implications on ecology systems [6]. In agricultural lands, conversation tillage and efficient utilization of fertilizers and irrigation have been proposed as techniques to increase the content of organic

carbon in soil (SOC) and mitigate atmospheric CO_2 increase [7]. These are also worthy of further investigations for atmospheric CO_2 mitigation since transferring carbon from the atmosphere to terrestrial ecosystems could offset some of the anthropogenic emissions from fossil fuel burning.

Global warming has become one major challenge to world's concerned scientific community. The disastrous global warming would raise the mixed-layer temperature in the sea and thermal expansion in turn could significantly raise the sea levels. Meanwhile, a large number of glaciers would be melt and enter into the sea, further enhancing the sea levels, which could in turn bring the disaster: the coastal areas would be flooded; the beach and coast would be eroded; the flooding would be intensified [8]. Since the early 20th century, the increase of the global air and sea surface temperature is up to 0.8°C and the Earth's surface has been successively warmer at each of the last three decades than any preceding decade since 1850 [9–11]. The disastrous global warming would also increase evaporation and then increase the atmospheric water vapor content [12]. Associated impacts of the future climate changes are the amount variations of precipitation, the probable expansion of subtropical deserts, and the more frequent extreme weather events [10–12]. By the year 2030 it is likely to be warmer than at any time in the last 120,000 years [13]. The disastrous global warming would even threat the life on Earth. The species that cannot adapt to such climate changes would be in danger of extinction, resulting in the species-diversity reduction. The disastrous global warming would also worsen the growth environment of the crop and hence fundamentally pose a serious hazard to agricultural production on Earth [14–16].

The combustion of fossil fuels, cement production, tropical deforestation, and other activities can largely contribute to the increase of radiative forcing from human activity [15]. In 1990s, these CO_2 emissions account for 80% of the current greenhouse effect, as compared with 57% of that in 1980s [16]. The ice core data shows that the atmospheric CO_2 varied from 180 ppm to 270 ppm over the past 800,000 years; that is, human activities after the Industrial Revolution should be recognized as a fundamental factor contributing the rapid increase of the atmospheric CO_2 level and influencing the global warming over the last century [17]. Therefore, CO₂ mitigation has become one of the major challenges related to the global warming. Many concerned scientific communities around the world have made a huge effort to investigate the CO_2 mitigation options [1–17]. However, people pay too much attention in ratifying the protocols and legally binding commitments, but few think of the ultimate solutions to limit the CO_2 emissions [17–19].

In this paper, we review five potential solutions for CO_2 mitigation: (1) emission trading under the global mitigation objectives and policies, (2) CO_2 capture and sequestration (CCS) technique, (3) the 100% renewable energy plan, (4) photocatalytic splitting of natural seawater to produce H_2 as zero-emission fuel, and (5) utilization of the energy driving soil CO_2 absorption in arid regions (termed "quasiphotosynthetic absorption"). The ultimate solutions are highlighted in comparison with these potential solutions and from the theoretical analyses of associated unresolved issues.

2. Theoretical and Potential Solutions

2.1. Emission Trading. Numerous scientists and policymakers began to realize the overlooked cousin of greenhouse effect. Adaptation became one of the most fashionable terms in the climate policy in this century [20]. According to a review from the United Nations Framework Convention on Climate Change (UNFCCC), the capacity of society's adaption has been closely related to whether it could tolerate the future climate change [21]. The UNFCCC and the Kyoto Protocol (UNFCCC in particular) as the first and foremost adaptation policy may help to find a solution. Meanwhile, the Intergovernmental Panel on Climate Change (IPCC) has developed some guidance and guidelines for the countries around the world to fulfil the UNFCCC reporting requirements [22].

The UNFCCC agreed to hold the atmospheric CO2 concentration at 450 ppm. Alternatively, before 2050, the global emissions need to be reduced below the emissions in the absence of any new policies [20-22]. Based on the setting of a quantitative restriction of CO₂ emissions, many countries started the international emissions trading. Different emissions trading schemes are implicitly designed in each country for the trade of the obligations in the UNFCCC. The international emissions trading in Japan was started in 2010 and run by TMG (Tokyo Metropolitan Government) [22]. The emission trading in the European Union and the national emissions trading in Norway both began from 2005 [23]. The Emission Trading Scheme was run from 2008 to 2012 in Switzerland, while the United Kingdom Trading Scheme was run from 2002 to 2006. Canada emissions trading started in 2007 [24]. Between 2009 and 2013, the Regional Greenhouse Gas Initiative started in ten Northeastern United States [25, 26]. The Australian scheme was run in 2003 by the State of New South Wales and the New Zealand scheme started in 2008 [27, 28]. Ever more countries "bank" the unused allowances for emissions in subsequent periods [29-42].

2.2. CO₂ Capture and Sequestration. As pinch technologies, transformation and utilization of CO2 are too expensive and cannot be widely used [43, 44]. One better solution is seeking for potential CO₂ sinks for carbon capture and storage (CCS: inject CO₂ into geological formations and then deposit it), reducing approximately 80-90% of emissions. In 2008, to investigate the technological feasibility for the long-term CO₂ storage, CCS was applied to a modern power plant in East Germany [45]. The estimated potential of CCS is 10% ~55% of the total mitigation before 2100 [46]. Carbon capture directly from the air which is feasible to reduce emissions from diffuse sources is still in its infancy, although a preprototype of air capture technology has been demonstrated [47]. Saline aquifers are also used for carbon storage, but the deep-ocean storage of CO₂ can exacerbate the ocean acidification and hence is illegal [48, 49].

CCS costs can be partly offset. After oxy-fuel combustion, the almost pure CO_2 stream is recirculated and injected into oil production fields to enhance oil recovery. It can also be stored in unmineable coal seams by the surface adhesion. The sale of recovered oil and methane can partly offset the costs. Mineral storage can be realized in a viable carbonation route, such as ultramafic mine tailings [50–52]. Stored CO_2 can be retained for 500 years at depths 1000 m~3000 m [53–63].

2.3. The 100% Renewable Energy Plan and a Zero-Emission Fuel. Renewable energy is defined as the energy that comes from resources which are naturally replenished on a human timescale such as sunlight, wind, rain, tides, waves, geothermal energy, and plant energy [64–66]. The oldest use of renewable energy in the form of traditional biomass as fuels starts from 790,000 years ago. In the mid-19th century (prior to the coal development), almost all used energy was still renewable. The use of solar energy was prompted by 1873 and its importance was recognized in 1911.

Today renewable energy has grown to replace fossil fuels for a substantial reduction of CO_2 emissions [67–75]. Since

the lifetime of CO_2 emission from renewable fuels is only a fraction of that from traditional fossil fuels, some economic analysts expected market gains for renewable energy from efficient energy use [71]. Many countries already started the economic exploitation of renewable energy sources. Particularly, 100% energy for electricity production in Iceland is from geothermal energy. In Brazil, hydroelectric power occupies 85% [72]. Renewable energy also contributes more than half of the electricity in Austria, New Zealand, and Sweden [73]. Wind power and solar water heating have been used around the world, especially in China [74]. The use of biomass for heating spreads and thorium-based nuclear power is also gaining interest [75–79].

2.4. Photocatalytic Splitting of Natural Seawater to Produce H₂ as Fuels. H₂ as a renewable fuel of zero-emission has been commercialized for cars, boats, airplanes, and even spacecraft [80–82]. Hydrogen can be delivered over long distances and can be stored in large quantities, at least as safe as gasoline [82-84]. It acts as a better energy carrier than electricity. Sun light is theoretically having enough energy to split water into H_2 and O_2 , but such a reaction is very slow [85–91]. A scientific topic occurs and attempts to replicate the process of photosynthesis and photocatalytic water splitting. It is one interesting way to produce hydrogen energy and no harmful chlorine gas was formed [49, 92-99]. In particular, photocatalytic splitting of natural seawater is environmentally cleaner (without release of CO2 or pollutants) and should be suggested as the ultimate solution of CO₂ mitigation in the future [100-102]. Nanostructured photocatalysts such as TiO₂ promoted with NiO (2.5%) and CuO (2.5%) have been employed and H₂ yields from photocatalytic splitting of seawater (containing oxalic acid as sacrificial reagents) on CuO/nano-TiO₂ and NiO/nano-TiO₂ are 8.53 µmol/gcat and 1.46 μ mol/gcat, respectively, after 5 h radiation [100].

Therefore, it is optimal to produce H_2 from the most abundant solar energy and the most abundant water resource on Earth (the natural seawater). The main objective in the future is to prepare more effective photocatalysts for splitting natural seawater. In such a scenario, the small regional plants around the world or even the local filling stations could produce hydrogen energy [101]. Difficulties associated with H_2 transportation can be overcome using on-site generation of hydrogen from off-grid renewable sources [102].

2.5. Quasi-Photosynthetic Absorption Utilization as a Potential Solution. As potential solutions for global mitigation, the circulation of CO_2 in the near-earth-surface reservoirs, the atmosphere, oceans, biosphere, and sedimentary rocks, also received widespread attention [103–107]. A CO_2 absorption process is found and is gaining more and more interest [108–115]. CO_2 sink from the inorganic carbon dissolution in the global water cycle has been demonstrated by Chinese scientists according to theoretical calculation and observed records [108]. This may be a new branch in the discussion of the missing carbon sink, highlighting the significance of the migration and storage process of the dissolved inorganic carbon in the arid regions [109]. In particular, soil respiration observations in a saline desert by Chinese scientists in 2006 revealed that soils in this desert admit a particular carbon process [110]. Scientists compared the dynamics of soil CO₂ fluxes measured before and after the sterilization treatment on the same soils; it was confirmed that the process is nonbiological and is as strong as the biological process [111]. After then, similar results were obtained in some other arid ecosystems [112–114]. It is indicated that CO₂ exchange in the global arid regions may be playing a more important role in the global carbon cycling and in modulating atmospheric CO₂ levels than previously assumed.

Since the sink size is as huge as photosynthetic CO₂ uptake [111, 112], we termed such CO₂ absorption "quasiphotosynthetic absorption." If estimates in previous publications represent an average of quasi-photosynthetic absorption, then soils in the global arid regions are absorbing up about half the amount emitted globally by burning fossil fuels [110]. Observations with chambers and open- or closepath eddy systems merely reflect the dynamics of CO₂ concentration in the soil-groundwater system. A long-time and large-scale observation of soil CO₂ fluxes and analyses of the soil profile data based on the isotope technology over arid ecosystems are still strongly encouraged [115]. Solar energy is the most abundantly available energy resource in the arid and semiarid biomes, which make up >30% of Earth's land surface [110–114]. The utilization of quasi-photosynthetic absorption for CO₂ mitigation demands further investigation on its relations to the cycle and conversion of solar energy.

3. Barriers and Unresolved Issues

3.1. Dilemma of Emission Trading. In the absence of CO_2 reduction policies, the global emissions would rise significantly during the 21st century [28–31]. Despite of CO_2 mitigation targets in the UNFCCC, till today, the vast majority of our energy is still supplied with fossil fuels. Political and social attitudes determine the difficulty to implement the effective policies. The future emissions depend on how socioeconomic development proceeds [32–34]. It is evident that the atmospheric CO_2 level is unlikely to stabilize in 21st century without major policy changes [34]. There are different proposals (the egalitarianism, basic needs and criteria, the proportionality and polluter-pays principle, historical responsibilities, willingness to pay, the comparable burdens and ability to pay, etc.) on how to allocate the responsibility for CO_2 emissions reduction [35–38].

The period of stabilizing atmospheric CO_2 concentrations not only depends on how quickly CO_2 is added to the atmosphere but also depends on how fast it is removed [36– 40]. Emissions trading has not delayed the phaseout of fossil fuels. Emissions trading requires CO_2 emitters to pay a tax for carbon emissions. Although such trading is theoretically cost-effective, it often falls disproportionately because of the potential unpopularity. A significant part of the emissions is from energy products and the people are insufficiently ready for the fees collection from a wide variety of emissions fees sources [40]. Increasing costs of carbon emitting fuels and activities can often result in an increase of electrical power prices (to cover the increasing costs of permits). This will in turn lead to missing motives and willingness



FIGURE 1: Schematic photocatalytic mechanism for pure TiO₂, Ni-TiO₂, and Ni/Cu-TiO₂.

because of increase in the costs of alternative power sources. The trading cannot directly guarantee a particular level of emissions reduction and the people have not fully understood the relationships between emissions and climatic processes. Successful strategies deserve an environmental funding and financial support [41].

Emission trading emphasized too much individual lifestyles and carbon footprints. It distracted attention from the corresponding political action that needs to be taken to mitigate the emissions. It is an alternative approach to mitigation emissions and so the overall effect is based on the significant development of less emission technologies. Small emissions reductions can be achieved cheaply, where larger ones are much more expensive. There is a dilemma. A sudden reduction is detrimental to regional economies, while a gradual reduction will risk environmental damage [42]. It can be a difficult balance to reduce global emission while utilizing fossil fuels.

3.2. Risk and Costs of CCS and Barriers to Renewable Energy. CCS techniques may offer a comprehensive solution to the global warming. But CCS applications also raise environmental effects. The pipe could tear and then leak CO_2 at high pressure and there is a major pressure front that can break the seal and make faults unstable [54]. The released CO_2 by a malfunction can kill a person at distance of 300 meters. This recommends that limits be set to the amount of CO_2 leakage [55]. Moreover, it is necessary to implement alert meters around the project perimeter, which helps to measure accidental leakage accurately [56, 57].

There are three different technologies for carbon capture: the capture after the combustion is applied in fossil fuel burning power plants, the technologies for removing CO_2 before combustion are widely utilized in the gaseous fuels, and the oxy-fuel combustion burns fuels in oxygen. A power plant equipped with CCS requires 60–180% more energy than those without CCS. To make things worse, a largescale adoption of CCS will increase the resource consumption [57–60]. Because of the potential catastrophic effects and additional electricity cost, CCS is still risky and expensive. It is necessary to suggest other storage sites besides oil fields, unmineable seams and saline formations, the gas fields, and saline-filled basalt formations [54–57].

Theoretical merit of CCS systems is to reduce most of CO_2 emissions, but this heavily depends on the plant types.

Significant environmental funding has been made available to support the large-scale CCS demonstration projects around the world, but the overall levels of CO_2 abatement remain 80%–90% higher than a plant without CCS (taking the fuel penalty into account). More openly, free measures are desired to finance regional and global CCS projects in the future [61–63].

In comparison with CCS, renewable energy costs are similar to the current energy costs; there are almost no technological or economic barriers to the 100% renewable energy plan. It is time to consider using 100% zero-emission hydrogen fuel as an optimal renewable energy plan [49, 94–99].

3.3. Constraints of Photocatalytic Splitting of Natural Seawater. Due to the increasingly polluted environment and the gradual depletion of fossil fuel reserves, it is highly desirable to develop photocatalytic water splitting technologies. To enhance the effectiveness of photocatalytic water splitting and H₂ production, photocatalysts must conform to several key principles. The quantum yield (QY = photochemical reaction rate/photon absorption rate × 100%) is a reliable determination of the photocatalyst effectiveness and the rate of the gas evolution can also be used to assist in comparisons [94-96]. The best photocatalyst has a high quantum yield and gives a high rate of gas evolution. The range of light absorbed is another determinant for a photocatalyst. That is, a less efficient photocatalyst that absorbs visible light may ultimately be more useful than the more efficient photocatalysts that absorb solely light with smaller wavelengths.

For example, TiO_2 is an efficient photocatalyst (absorbs only ultraviolet light and outperforms most visible light), as it yields both a high quantum number and a high rate of H₂ evolution [97]. Assisted by photocatalysts suspended directly in water, the water splitting is well done in one step and can therefore be more efficient (Figure 1). As an optimal key technology for the production of renewable, zero-emission hydrogen energy, the photocatalytic water splitting can play a significant role in the 100% renewable energy plan. This associates the 100% renewable energy plan to the development of hydrogen economy (a concept of delivering energy by H₂ proposed by J. B. S. Haldane) [49]. In order to meet the requirements for current issues on environment and energy, photocatalysis process initiated on nanomaterials is crucial [116].

Nanomaterials have attracted considerable attention for photocatalysis due to inexpensive and clean nature by using



FIGURE 2: Nanostructure of TiO₂: from bulk (a) to nanosheets (b).



FIGURE 3: Mechanisms of structurally promoted TiO_2 for photocatalysis: from bulk and nanosheets.

abundant, cheap, and environmentally friendly chemical reagents, energy source, and catalysts without secondary pollution [117–119]. Both the chemical composition and morphology of nanomaterials are of significant importance in promoting specific photocatalytic activity [118]. For example, the chemical doping of foreign elements can regulate the optical property of resultant nanostructure [119]. However, the hole scavenging with chlorides in seawater may be associated with the less H_2 yielded from photocatalytic splitting of seawater if compared with photocatalytic splitting of water [100]. There are several technological constraints to elucidate the role of salts in the seawater [101, 102].

There are a lot of challenges, especially when altering nanostructures and surface properties of catalysts, extending the photoabsorption range, and reducing photoinduced electrons and holes recombination. For example, doping will cause the valence band and different nanostructure, which help to enhance the photocatalysis performance by changing the nanostructure of TiO_2 from bulk to nanosheets (Figure 2). There are a larger specific surface area and more reaction sites (Figure 3). This is just a special case. In more general cases, the reliability of the nanostructure and morphology analysis of nanomaterials with the visual and experience identification is limited and not convincing. For the global H₂ production by photocatalytic splitting of natural



FIGURE 4: Chemical structure of the well-known visible-light responsive $g-C_3N_4$.

seawater, it is necessary to utilize computer vision in structurally and elementally promoted nanomaterials for photocatalysis [118]. Moreover, TiO₂ as an excellent photocatalyst for splitting of natural seawater majorly performs well under UV light irradiation. But UV light only accounts for about 3% of the solar spectrum. It is also necessary to introduce some visible-light responsive photocatalyst, such as g-C₃N₄ of optimal chemical structure (Figure 4), in photocatalytic splitting of natural seawater. Furthermore, nonnoble metal cocatalysts are strongly suggested to be employed and tremendously enhance the H₂-production, such as $Zn_{1-x}Cd_xS$ and CdS [120, 121]. The calculated CB and VB edge potentials of $Zn_{1-x}Cd_xS$ samples are illustrated in Figure 5 (CdS being the particular case x = 1). It can be clearly seen that the CB edge potential becomes more negative with increasing Zn content.

3.4. Undetermined Mechanisms of Quasi-Photosynthetic Absorption. There is no doubt that results from the previous studies implied that the quasi-photosynthetic absorption has the huge effects on CO_2 mitigation [122–126]. A better understanding of quasi-photosynthetic absorption helps us understand the subterranean carbon sinks and sources in the arid ecosystems. The theoretic calculations and the



FIGURE 5: The calculated CB and VB edge potentials of the $Zn_{1-x}Cd_xS$ samples.

long-term, large-scale observations of quasi-photosynthetic absorption can provide some important datasets and methods in the future research of the regional and global carbon balance over arid ecosystems affected by the water cycle in the hyper-arid, arid, and semi-arid regions.

Nevertheless, the physiochemical and hydrological mechanisms for quasi-photosynthetic absorption remain undetermined. A series of conjectures of the new issue have been raised and resulted in interesting and intense debate and discussions. In order to understand the mechanisms, the isotope ecology of the CO₂ exchange between the groundwater-soil system and the atmosphere should be discussed. To do that, the correlation relationships of organic/inorganic carbon, CO_2 , and $\delta^{13}C$ in the soils and groundwater must be detected. To further understand the carbon migration, transformation, and assignment as the gaseous, liquid, and solid phases, it is emergent to establish the process model for the variations of soil carbon with the groundwater recharge and discharge. This outstanding question is very difficult to resolve because of the fact that it not only involves hydrology, photoenergy, geochemistry, physics, ecology biology, and soil science but also involves a series of mathematical subjects for advanced analyses and predictions.

Besides the soil texture, structure, alkalinity, and other physiochemical characteristics, the simultaneous happening biochemical reaction and biological feedback must be taken into account. To make things worse, both soil and groundwater are very complicated systems and the situations in different depths are different. Attempts for mathematical analyses are therefore encouraged and numerical modeling and stochastic simulation can be a research priority for a better understanding of the quasi-photosynthetic absorption and its relations with photoenergy cycling.

4. Summary and Perspectives

The control of CO_2 emission has become one major challenge to world's concerned scientific community. Moreover it is also of great significance to conserve energy and curb emissions [127]. Despite the efforts to control emissions at governmental and regional level, it can be a difficult balance to reduce the global CO_2 emission while utilizing the fossil fuels. Increasing CO_2 emission not only contributes to global warming but also contributes to other environmental issues [128]. This review article is giving an overall picture of potential solutions for CO_2 mitigation and finding more efficient measures to successfully achieve the CO_2 reduction target as promised.

Combining with some results in the previous studies, our reviews and analyses indicate the following.

- The global warming creates an incentive to use 100% renewable energy. H₂ production is an optimal technology of renewable, zero-emission fuels. The significance of photocatalytic splitting of natural seawater is increasing.
- (2) The utilization of quasi-photosynthetic absorption can be a supplement of photocatalytic splitting of natural seawater. The total area of sea is more than twice times that of continent, and more than 1/3 of the continent is hyper-arid, arid, and semi-arid areas (Figure 6).
- (3) The future research of photocatalytic water splitting should consider the harmony between the full hydrogen economy and the agricultural and ecological water requirements. Using natural seawater also helps to reduce the water resources risk.



FIGURE 6: Present-day distribution of the ocean, continent, and hyper-arid, arid, and semi-arid areas.

- (4) The application prospect for quasi-photosynthetic absorption and the photocatalytic H₂-production from natural seawater and the utilization of quasiphotosynthetic absorption are worthy of further investigation on regional and global scales.
- (5) Though still facing a series of challenges in the largescale applications, photocatalytic splitting of natural seawater can motivate full hydrogen economy in the future.

Therefore, photocatalytic H_2 -production from natural seawater and the utilization of quasi- photosynthetic absorption are highlighted as two ultimate solutions for future CO_2 mitigation. Photocatalytic hydrogen production and its utilization as fuels can be the major efficient measures to successfully achieve the CO_2 emission reduction target as promised, while the utilization of quasi-photosynthetic absorption can be a supplement of photocatalytic splitting of natural seawater in hyper-arid, arid, and semi-arid land areas.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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