A Review on Photocatalytic CO_2 Reduction using Nanomaterial

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Abstract

Photocatalytic CO_2 reduction using nanomaterials has emerged as a promising approach for addressing the global challenge of carbon dioxide (CO_2) emissions. This abstract provides an overview of recent advancements and key findings in the field of photocatalytic CO_2 reduction using nanomaterials. Various nanomaterials, such as quantum dots, metal oxides, graphitic carbon nitride (g-C3N4), and semiconductors, have been investigated for their photocatalytic activity towards CO_2 reduction. These nanomaterials possess unique properties such as high surface area, short charge transfer pathways, and tunable band gaps, enabling efficient absorption of solar energy and generation of electron-hole pairs. However, challenges such as charge carrier recombination, low surface area, and low electrical conductivity hinder the photocatalytic activity of some nanomaterials. To overcome these limitations, modifications and doping strategies have been employed to enhance their photocatalytic performance. These modifications include surface functionalization, co-catalyst deposition, and incorporation of carbon-based materials. Size-dependent effects have been observed, where nanoparticles of optimal sizes exhibit enhanced photocatalytic activity. Furthermore, the choice of solvent and reaction conditions plays a crucial role in achieving high selectivity and efficiency in CO_2 reduction. Selective reduction products, including methane (CH_4), methanol (CH3OH), formaldehyde (HCHO), and formic acid (HCOOH), can be obtained depending on the catalyst and reaction conditions.

Keywords: CO_2 reduction, Nanomaterials, photocatalysis, quantum dots, metal oxides, graphitic carbon nitride (g-C3N4), and semiconductors
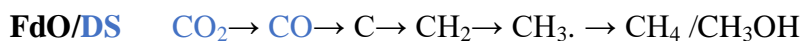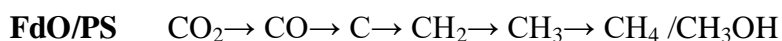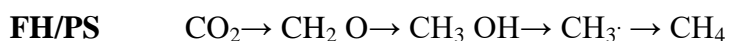
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Introduction: Photocatalytic CO_2 reduction using nanomaterials has emerged as a promising strategy for mitigating carbon dioxide (CO_2) emissions and addressing the global challenge of climate change. This approach harnesses the unique properties of nanomaterials to convert CO_2 into valuable carbon-based compounds, such as methane (CH_4), methanol (CH3OH), formaldehyde (HCHO), and formic acid (HCOOH), through solar energy-driven processes. Nanomaterials, characterized by their nanoscale dimensions and high surface area, offer several advantages for photocatalytic CO_2 reduction. One key advantage is their efficient light absorption, facilitated by their tunable band gaps and quantum confinement effects. This enables nanomaterials to effectively utilize a broad range of the solar spectrum, including visible light, for photocatalytic reactions. Quantum dots, semiconductor nanoparticles, and metal oxides are among the commonly studied nanomaterials in this field. [1-19]

Quantum dots, semiconductor nanoparticles with unique quantum confinement properties, exhibit high extinction coefficients for visible light, making them efficient photosensitizers. Their small size and large surface area provide abundant active sites for CO_2 adsorption and catalytic reactions. Additionally, their short charge transfer pathways minimize charge carrier recombination, leading to enhanced photocatalytic performance. Metal oxides, such as titanium dioxide (TiO_2), zinc oxide (ZnO), and tungsten trioxide (WO_3), are widely explored nanomaterials for photocatalytic CO_2 reduction. These metal oxides possess favorable band positions and catalytic properties, enabling the generation of electron-hole pairs upon light illumination. However, their photocatalytic activity is often limited by the recombination of charge carriers and their low surface area. To address these limitations, surface modifications, such as doping with metals or non-metals, and the introduction of co-catalysts have been employed to enhance their photocatalytic efficiency.

Graphitic carbon nitride (g-C3N4), a metal-free photocatalyst, has gained significant attention due to its unique properties. It is chemically and thermally stable, non-toxic, and abundantly available. Although pure g-C3N4 exhibits limited photocatalytic activity due to charge carrier recombination and low electrical conductivity, its performance can be enhanced through modifications, such as heteroatom doping or coupling with other materials. These modifications improve the separation and utilization of photogenerated charge carriers, leading to improved CO_2 conversion efficiency. Size-dependent effects have been observed in nanomaterials, where the photocatalytic performance is influenced by the particle size. Nanoparticles of optimal sizes exhibit enhanced light absorption, improved charge transfer kinetics, and increased active surface area, resulting in higher catalytic activity. However, excessively small particle sizes can lead to changes in the electronic and optical properties, potentially affecting the photocatalytic performance.

In addition to nanomaterial design and optimization, the choice of solvent and reaction conditions plays a crucial role in the efficiency and selectivity of photocatalytic CO_2 reduction. Solvents with appropriate polarity and solubility for CO_2 can facilitate its absorption and reaction on the catalyst surface. Moreover, the reaction conditions, including temperature, pressure, and reactant concentrations, need to be carefully controlled to optimize the conversion rates and selectivity towards desired products. Understanding the underlying mechanisms and reaction pathways involved in photocatalytic CO_2 reduction is essential for further improving the efficiency of nanomaterial-based catalysts. Computational modeling, spectroscopic techniques, and electrochemical analysis provide valuable insights into the charge transfer processes, CO_2 adsorption, and reaction intermediates. These studies help elucidate the key factors influencing the photocatalytic activity and guide the rational design of advanced nanomaterials. [1-25]

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Result and discussion: The increasing concentration of CO_2 in the atmosphere has been a significant concern for researchers over an extended period. Climate change primarily arises from the excessive presence of CO_2 in the environment, which is predominantly emitted through fuel combustion and deforestation. The escalating levels of CO_2 contribute to the greenhouse effect, resulting in global warming. Therefore, it is crucial to reduce the amount of CO_2 in the atmosphere. The conversion or reduction of CO_2 into alternative forms, however, presents a substantial challenge due to its inherent stability as a molecule. Under standard conditions, the thermodynamics of CO_2 reduction are unfavorable due to its negative electrode potential. Nonetheless, addressing global warming and energy scarcity issues can potentially be achieved by reducing CO_2 to hydrocarbons using solar light. [1] Through the photoreduction of CO_2 in the presence of water and solar light, it is possible to produce substances such as CH_4, HCOOH, CO, CH2O, CH3COOH, and CH_4, depending on the transfer of electrons and protons. Among the promising photocatalysts, TiO_2 stands out for its potential in CO_2 reduction. The reaction pathways for CO_2 reduction using TiO2 can be depicted as follows. [2]

FH/PS $CO_2 \rightarrow CH_2O \rightarrow CH_3OH \rightarrow CH_3 \cdot \rightarrow CH_4$

FH/DS $CO_2 \rightarrow CO \rightarrow CH_2O \rightarrow CH_3OH \rightarrow CH_3 \cdot \rightarrow CH_4$

FdO/PS $CO_2 \rightarrow CO \rightarrow C \rightarrow CH_2 \rightarrow CH_3 \rightarrow CH_4 / CH_3OH$

FdO/DS $CO_2 \rightarrow CO \rightarrow C \rightarrow CH_2 \rightarrow CH_3 \cdot \rightarrow CH_4 / CH_3OH$

In the context of photocatalytic CO_2 reduction, the abbreviations FH, FdO, PS, and DS represent different pathways and surface characteristics. FH refers to the fast-dehydrogenation path, while FdO stands for the fast-deoxygenation path. PS represents the perfect surface, whereas DS

represents the defective surface. Specifically, blue species are adsorbed at oxygen vacancies, while black species are adsorbed on the titanium surface.

Semiconducting photocatalysts like TiO_2, ZnS, and CdS suffer from a major drawback - their low quantum yield. However, certain transition metal complexes exhibit high quantum yields and serve as effective photocatalysts [3]. Recently, supramolecular metal complexes have been introduced for highly efficient CO_2 reduction, utilizing visible light, and demonstrating superior reaction rates compared to other photocatalysts [4]. Investigation into first-row transition metals for photocatalytic CO_2 reduction has garnered attention [5]. Notably, nickel supported on silica-alumina has shown efficacy as a photocatalyst for CO_2 reduction [6]. Another approach involves the low-temperature solar-driven reduction of CO_2 to CH_4 using H_2. Research in both photochemical and electrochemical fields aims to achieve high reaction rates by employing pure CO_2. Additionally, a promising technology is anticipated for the utilization of low-concentration CO_2 found in exhaust gases from power plants, typically containing only 3%-13% CO_2 [7]. Aqueous medium poses challenges for CO_2 reduction due to its poor solubility in water.

Nanomaterials and nanoparticles possess an exceptionally large surface area, making them highly efficient catalysts in both homogeneous and heterogeneous environments. In the realm of CO_2 reduction, semiconductor quantum dots (QDs) demonstrate remarkable photosensitization capabilities due to their high extinction coefficients for visible light and prolonged excited state lifetimes [8]. Researchers have proposed hybrid photocatalysts that effectively reduce CO_2 in aqueous media without the need for noble metals. A recent study highlights the preservation of catalytic activity in aqueous solutions through the selective attachment of $[Ni(terpy)_2]^{2+}$ complexes onto CdS nanocrystals. These hybrid photocatalysts exhibit over 90% effectiveness in CO_2 reduction when exposed to visible light irradiation [8].
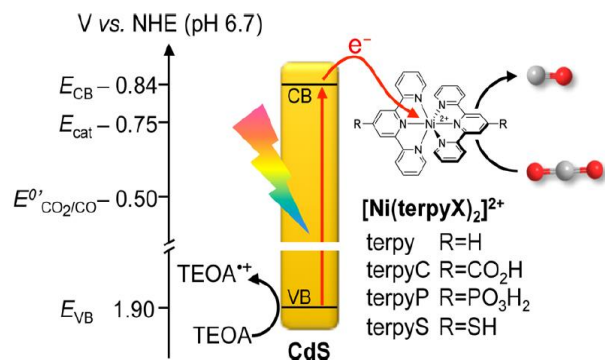
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Figure 1: a CdS quantum dot (QD) photosensitizer is shown with a molecular nickel bis(terpyridine) catalyst attached. The study examined various anchoring groups to evaluate their performance. [8]

Ru complexes supported on N-doped Ta2O5 substrates exhibit exceptional photocatalytic efficiency for CO_2 reduction. Researchers have conducted experiments to investigate the impact of anchoring groups, which bind to the semiconductor, on the efficiency of CO_2 reduction. The effectiveness of this reduction process is quantified by turnover numbers (TONs). A research group has reported significant insights into the electron transfer (ET) mechanism, providing valuable understanding of the photocatalytic reduction of CO_2 on N-doped Ta2O5 [9].

The shape-dependent photocatalytic activity of TiO2 has garnered attention. A recent study by the Mietek group has demonstrated enhanced photocatalytic CO_2 reduction activity in anatase TiO2 by co-exposing {001} and {101} facets [10]. {101} facets are thermodynamically favored, resulting in their predominance on the surface of anatase TiO$_2$ compared to {001} facets. However, a study by Han et al. reveals that {001} facets exhibit superior photocatalytic activity compared to {101} facets. This can be attributed to the higher density of unsaturated Ti atoms and active surface oxygen atoms on {001} facets. In {001} surfaces, the Ti-O-Ti bond angle is greater than that in

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{101} surfaces, which destabilizes the 2p states of surface oxygen atoms and enhances their reactivity. The fermi level of both {001} and {101} surfaces is equal, as depicted in Figure (b). However, in the case of {101} surfaces, the fermi level remains at the top of the valence band. Consequently, {001} and {101} surfaces form a heterojunction, facilitating the transfer and separation of electrons and holes [11]. The ratio of exposed {001} and {101} facets greatly influences the photocatalytic activity of anatase TiO_2.
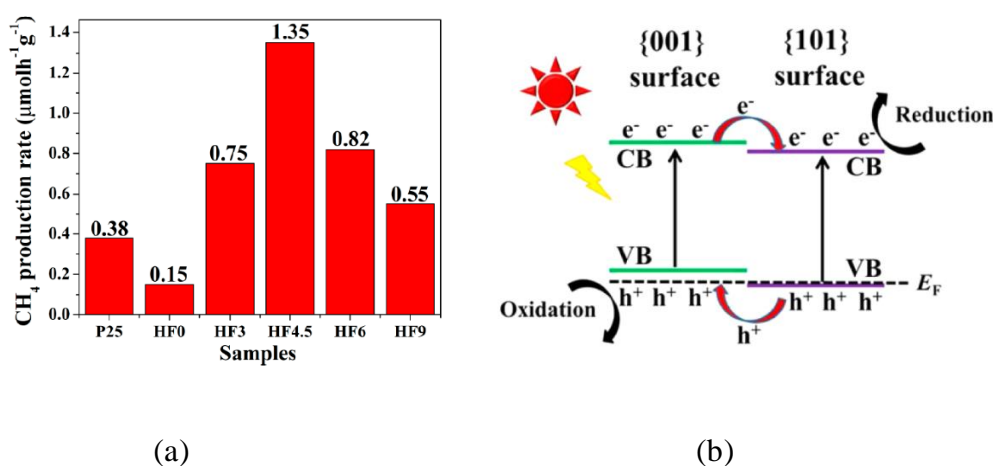

(a)   (b)

Figure 2: Presents the following information: (a) A comparison of the photocatalytic activity for CH_4 production between P25 and the prepared TiO_2 samples, and (b) the heterojunction between {101} and {001} surfaces. [11]

The influence of the ratio of {101} and {001} facets on the photocatalytic activity of TiO2 samples is evident from Figure (a). Increasing the amount of HF from 3 to 4.5 mL leads to an enhancement in photocatalytic activity due to the greater presence of {001} facets. However, further increasing the amount of HF beyond 4.5 mL results in a reduction in photocatalytic activity. This is attributed to the excessive abundance of {001} facets in anatase, leading to electron overflow towards {101} facets.

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The Ishitani research group has proposed a novel and efficient photocatalytic CO_2 reduction method utilizing Cu^+ diamine complexes as photosensitizers and Fe^{2+} diamine complexes as catalysts [12]. A heteroleptic Cu(I) diamine complex with both diamine and phosphine ligands has been chosen as the photosensitizer due to its longer lifetime and stronger oxidation state at higher energy levels. Although halide perovskite quantum dots (QDs) are promising optoelectronic materials for LED and photovoltaic devices, their stability is compromised in polar solvents or in the presence of moisture, hindering their use in photocatalytic processes. However, organic-inorganic halide perovskites exhibit a high extinction coefficient, wide absorption ranges, and low electron-hole diffusion lengths [13]. CsPbBr3 QDs, as a stable halide perovskite material, along with CsPbBr3 QD/graphene oxide composites, have been investigated by the Kuang group as photocatalysts for CO_2 reduction to ethyl acetate. This research represents the first artificial photosynthesis approach based on halide perovskite QDs.
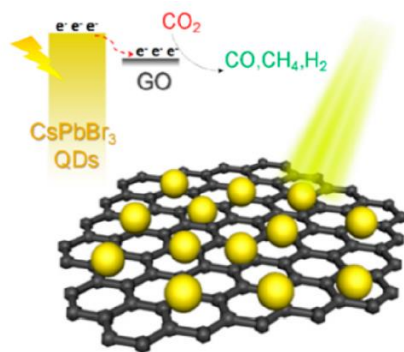

Figure 3: The provided diagram illustrates the CO_2 photoreduction process over the CsPbBr3 QD/GO photocatalyst. [13]

Quantum dots are highly effective photocatalysts due to their high surface area and short charge transfer pathways. In the study conducted by the Kuang group, ethyl acetate was chosen as the

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solvent because of its mild polarity, which stabilizes the CsPbBr3 quantum dots, and its high solubility for CO_2. The selectivity for CO_2 reduction was observed to be greater than 99%, highlighting the preference for halide perovskite materials over conventional metal oxide semiconductors. Ground state CO_2 reduction can be explained by the concept of frustrated Lewis pairs (FLPs) [14]. Despite the high cost of In2O3, it is considered an effective photocatalyst due to its superior chemical stability under light [15]. The photocatalytic activity of carbon-coated In2O3 Nanobelts was studied by the Goodenough group. In2O3 generates CO_2- from CO_2, which acts as a key species in the reduction process. Additionally, the reduction potential of In2O3 (-0.62V) is sufficient for the conversion of CO_2 to CO (-0.53V) and CH_4 (-0.24V) [16]. In2O3 exhibits a greater affinity for H_2 generation and lower activity for carbon-related products in the photocatalytic reduction of carbon dioxide with water. The coating of carbon on In2O3 nanobelts enhances its photocatalytic activity for CO_2 reduction to CO and CH_4, using water as a reductant and Pt nanoparticles as a co-catalyst, as summarized by the Goodenough group in their article. The carbon coating induces a higher eagerness for absorbing visible light, separates photoinduced electron-hole pairs, and suppresses proton reduction to H_2. These are the stepwise processes for CO_2 reduction to CO and CH_4.

P-type semiconductors CuO and Cu2O have narrow band gaps of 1.2eV and 2eV, making them suitable for visible light-driven photocatalysis. However, their low electronic conductivity may lead to recombination of electrons and holes, hindering catalytic activity. Modifying these oxides with carbon materials can enhance their conductivity [17]. The addition of reduced graphene oxide (rGO) as an additive improves the catalytic activity of CuO/Cu_2O for CO_2 reduction. The Wang group prepared CuO/Cu_2O nanowire arrays grafted with reduced graphene oxide using a thermal process. This composite effectively acted as a photocatalyst for the reduction of carbon dioxide to

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carbon monoxide under solar light irradiation. Nanostructured 2D materials have shown promise as efficient photocatalysts for CO_2 reduction. Graphene-based photocatalysts have gained attention due to their semiconductor properties with zero band gap, high surface area, and conductivity. Liang coupled graphene with Rutile for visible light photocatalytic CO_2 reduction. Graphene/P25 TiO2 photocatalysts, synthesized by Liang with low graphene defect density, exhibited efficient performance for CO_2 reduction under visible light, surpassing P25 alone by a 7-fold advancement in CO_2 reduction to CH_4 [18].

To enhance the photocatalytic activity, noble metal nanoparticles such as Pt, Pd, Ag, and Au can be dispersed on the photocatalyst. Kumar synthesized reduced graphene-coated gold nanoparticles, which demonstrated significant photocatalytic activity for CO_2 reduction under visible light. The photocatalytic activity was primarily attributed to the reduced graphene layers, which acted as highly efficient electron acceptors and transporters, facilitating the utilization of hot electrons in the plasmonic photocatalyst, as shown in the accompanying figure.
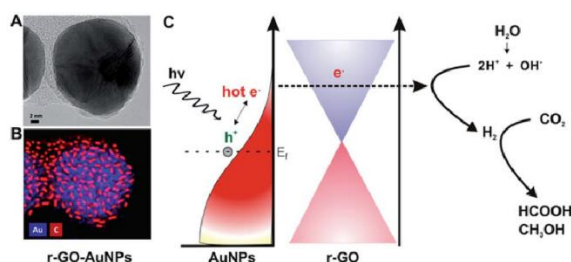

Figure 4: The given schematic represents the graphene-coated gold nanoparticles used for CO_2 photoconversion. [18]

Graphitic carbon nitride (g-C3-N4) is a metal-free photocatalyst that holds great potential for solar energy conversion. It has garnered significant interest among researchers due to its chemical and

thermal stability, non-toxic nature, and abundant availability. However, g-C3N4 exhibits limited photocatalytic activity characterized by charge carrier recombination, low surface area, and low electrical conductivity. Therefore, modifications to pure g-C3N4 are necessary. Unlike graphene, g-C3N4 is a metal-free material with a moderate band gap of 2.7eV. Its conduction band edge makes it a practical choice for CO_2 reduction into CH_4, CH3OH, HCHO, and HCOOH, as these reactions require a negative electrode potential [18].

The photocatalytic reduction of CO_2 with water using semiconductors follows a three-step process. Firstly, when semiconductors are illuminated with light energy equal to or greater than the band gap, electron-hole pairs are generated. These photogenerated electrons and holes then migrate to the surface of the semiconductors. Finally, the photogenerated electrons reduce the adsorbed CO_2 on the catalyst, while the holes contribute to the oxidation of water into oxygen [19]. The size of semiconductor nanoparticles plays a crucial role in their photocatalytic behavior. Koci et al. investigated the influence of particle size on the photocatalytic activity of TiO_2. Their research encompassed particle sizes ranging from 4.5 to 29 nm for the solid-liquid photocatalytic reduction of CO_2 with water. Among these, TiO_2 nanoparticles with an average size of 14 nm exhibited the highest effectiveness. While smaller particles might be expected to be more effective, their extremely small size changes both the optical and electronic properties. This can lead to an increase in the band gap energy, minimizing the efficiency of light utilization [20]. Furthermore, TiO_2 doped with coinage metals co-catalysts including Pt, Pd, Rh, Au, and Ag showed that the rate of CH_4 formation followed the order of Ag < Au < Rh < Pd < Pt. This observation can be attributed to Pt's superior ability to separate photogenerated electron-hole pairs.

Thus far, numerous approaches and models have been proposed or established for photocatalytic CO_2 reduction. It is challenging to define a specific standard or ranking for materials based on their performance in photocatalytic activity. No single parameter uniquely describes or determines the quantitative outcome. In our discussion, we have emphasized the significance of CO_2 reduction and highlighted various types of photocatalysts along with their performance analysis. Several criteria, including earth abundance, toxicity, availability, stability, and cost efficiency, contribute to the suitability of a photocatalyst. The adsorption ability and reactivity of CO_2 on the catalyst surface play a crucial role. The structure of semiconductors significantly influences CO_2 reduction as it affects the generation of electron-hole pairs and the catalyst's reactivity. Particle size is also a critical factor, where medium-sized nanoparticles of TiO_2 have demonstrated enhanced effectiveness. Additionally, the concentration of surface sites and the efficient photogeneration of electrons and holes on the surface greatly enhance photocatalytic activity. We previously observed that the higher reactivity of {001} facets makes them more active for CO_2 reduction to CH_4 due to their greater conduction band edge position and superior CO_2 adsorption capability. Further research is needed to improve the yield of CO_2 reduction soon.

In summary, nanomaterials show great potential as efficient catalysts for photocatalytic CO_2 reduction. Advances in nanomaterial synthesis, surface modification, and doping strategies have led to significant improvements in their photocatalytic performance. However, further research is required to optimize nanomaterial properties, explore new catalyst designs, and scale up the photocatalytic systems to achieve practical and sustainable CO_2 reduction. The development of highly efficient and selective nanomaterial-based photocatalysts holds promise for mitigating CO_2 emissions and addressing environmental challenges.

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Declaration

The authors have no competing financial interests to disclose.

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