Semiconductor Materials for Photocatalytic Reduction of CO₂

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The photocatalytic reduction of CO₂ is one of the most effective methods to control CO₂ pollution. Therefore, the development of novel high-efficiency semiconductor materials has become an important research field. Semiconductor materials need to have a structure with abundant catalytic sites, among other conditions, which is of great significance for the practical application of highly active catalysts for CO₂ reduction. The photocatalytic reduction of CO₂ is a surface/interface reaction. It is important to find and use raw materials which are environmentally friendly and effective as catalysts.

semiconductor materials photocatalytic carbon dioxide

1. TiO₂ Photocatalyst

As an n-type semiconductor material, TiO₂ is regarded as an ideal semiconductor material for environmental pollution control due to its good chemical stability, non-toxicity and environmental friendliness. It has great potential application value in the field of environment and energy $\begin{bmatrix} 1 \\ 2 \end{bmatrix}$. In 1972, Fujishima and Honda reported $\begin{bmatrix} 2 \\ 2 \end{bmatrix}$ that TiO₂ could decompose water molecules into hydrogen and oxygen under ultraviolet light, bringing extensive attention to TiO₂ as a photocatalyst material. TiO₂ has been widely used in the photocatalytic decomposition of decomposition of hydrogen in aquatic products [3][4][5], the degradation of pollutants [6][7][8], the reduction of CO₂ [9][10][11][12] etc., and a series of research achievements have been made. TiO₂ photocatalyst materials are also very common in daily life; they are widely used in solar cells, coatings, cosmetics, antibacterial materials and air purifiers, as examples.

 TiO_2 can be found in the form of three crystal polymorphs: anatase, rutile and brookite [13]. The three crystals have a twisted octahedral structure of six oxygen atoms around a titanium atom. Anatase and rutile forms have tetragonal crystal structures. The different electronic structures of the three crystal types of TiO2 lead to great differences in their photocatalytic performance. Tang et al. [14] studied the effect of different crystal types on the photocatalytic performance of TiO₂. It was found that the degradation rate of pollutants was nearly 100% when anatase or mixes with anatase and rutile forms was used as photocatalyst. The degradation rate was less than 15% when pure rutile TiO₂ was used as photocatalyst. Jin et al. [15] prepared PbO-decorated TiO₂ composites by a one-pot method with highly photoactive CO₂ conversion. The heterojunction formed by the catalyst could effectively inhibit the recombination of photogenerated charge, while the PbO could improve the adsorption of CO₂ on the catalyst. Therefore, the photocatalytic activity of the heterojunction complex for CO2 reduction was significantly improved.

Morphology is also an important factor affecting the photocatalytic performance of TiO₂. The specific surface areas, active sites, charge transfer rates and exposed crystal surfaces of TiO₂ catalysts with different morphologies are significantly different, leading to great differences in their performance. Cao et al. $\frac{16}{10}$ prepared a TiO₂ photocatalyst with nanorods and nanorod-hierarchical nanostructures. The catalyst had better photocatalytic performance for CO₂ reduction than commercial TiO₂ (P25). The high catalytic activity was mainly attributed to the improved charge transfer performance, specific surface area and light absorption performance of the catalyst lent by the nanorodhierarchical nanostructures. Tan et al. $\begin{bmatrix} 17 \\ 2 \end{bmatrix}$ synthesized an Ag/Pd bimetal supported on a N-doped TiO₂ nanosheet for CO₂ reduction. Due to the modification of the Ag/Pd bimetal and the N doping, the absorption of visible light in the TiO₂ nanosheets was improved. This system also provided abundant surface defects and oxygen vacancies for the TiO₂ nanosheets, causing the catalyst to exhibit high performance for the photocatalytic reduction of CO₂ to CH_4 . Kar et al. [18] prepared a TiO₂ nanotube photocatalyst which showed a highly efficient photocatalytic reduction of CO₂ to CH₄. The high photocatalytic activity was mainly attributed to the enhancement of visible light absorption by the nanotube structure. In addition to the crystal size and morphology of TiO₂ catalyst, the intensity of the external light source also has an effect on the photocatalytic activity of TiO₂—generally, as the light intensity increases, more photogenerated electrons are generated by the catalyst's excitation, and the photocatalytic reaction is promoted as a result. Yang et al. ^[19] found that the photocatalytic degradation rate of TiO₂ to paracetamol attenuated with the decrease of light intensity.

2. Co/Ni-Based Catalysts

The transition metals cobalt and nickel, which have various redox states, have been widely used in the study of photocatalytic CO₂ reduction due to their rich crustal content, low cost, high catalytic activity for CO₂ and strong adsorption capacity ^{[20][21][22]}. In 2015, Wang et al. ^[23] prepared Co-ZIF-9 by a solvothermal method for the study of photocatalytic CO2 reduction. The results showed that the three-dimensional MOFs structure of Co-ZIF-9 was beneficial for CO₂ enrichment. The Co²⁺ ions and imidazole groups played a synergistic role in the photocatalytic reduction of CO₂. The Co²⁺ ion was beneficial for electron transport, and the imidazole group was beneficial for the activation of CO₂ molecules. Wang et al. [24][25] also studied the photocatalytic CO₂ reduction performance of cobalt-based spinel oxides. MnCo₂O₄ microspheres and ZnCo₂O₄ nanorods were prepared by solvothermal calcination and hydrothermal methods, respectively. The results showed that both catalysts exhibited excellent catalytic stability, which confirmed the possibility of Co-based spinel oxides for photocatalytic CO₂ reduction. Zhang et al. ^[26] studied the performance of six Co-MOFs with different coordination environments applied to photocatalytic CO₂ reduction. In pure CO₂ atmosphere, the MAF-X27/-OH material had high CO selectivity (98.2%). When the relative pressure of CO₂ dropped to 0.1 atm, the conversion rate of MAF-X27/-OH remained at about 80%, while the CO conversion rate of Co-MOF materials without this ligand decreased significantly. These results suggest that MAF-X27/-OH has excellent photocatalytic CO₂ reduction performance. These conclusions provide a theoretical basis for future studies on the photocatalytic CO₂ reduction of Co-based nanocatalysts.

In addition to Co-based nanomaterials, many Ni-based nanomaterials have also been applied in the study of photocatalytic CO_2 reduction. Niu et al. ^[27] synthesized a spongy nickel-organic heterogeneous photocatalyst

(Ni(TPA/TEG), which could effectively adsorb CO₂. This novel Ni-based photocatalyst significantly inhibited the production of H₂, CH₄ and CH₃OH during the photocatalytic reduction of CO₂. Thus, efficient CO₂ conversion was achieved, and the selectivity of CO was close to 100%. Yu et al. ^[28] obtained a covalent organic framework bearing single Ni sites (Ni-TpBpy) for the selective reduction of CO₂ to CO. The results showed that Ni-TpBpy effectively promoted the formation of CO in the reaction medium, and the selectivity of CO was 96% after 5 h of reaction. More importantly, the catalyst maintained 76% CO selectivity in a low CO₂ atmosphere. Ni-TpBpy showed good photocatalytic performance for the selective reduction of CO₂, which was mainly attributed to the synergistic effect of single Ni catalytic site and TpBpy supporter.

3. Metal Halide Perovskites (MHPs)

In recent years, metal halide perovskite (MHP) materials have become attractive in the field of optoelectronics and energy conversion ^[29]. Compared with traditional semiconductor nanocrystals, these materials have high extinction coefficient, narrow band emission, long carrier diffusion length and high defect tolerance. In addition, the diversity of perovskite structures also allows the band gap to be adjusted in order to enhance light capture ^[30]. The crystal structure of MHPs is similar to that of oxide perovskite. The chemical formula is ABX₃, where A is a monovalent cation, B is a divalent metal cation (the most common are Pb²⁺ and Sn²⁺) and X is a halogen ion. MHP nanocrystals exhibit a halogen-rich surface structure. Perovskites can be classified as inorganic or organicinorganic hybrid halogenated perovskites according to the type of cations in their chemical structure. The emergence of MHPs with unique photoelectric characteristics brings new opportunities for efficient photocatalytic CO₂ reduction.

Since the reduction potential of MHPs for CO₂ reduction usually changes with the nanocrystal size, the catalytic activity of MHP nanocrystals is also affected by their size. Sun et al. [31] first synthesized CsPbBr₃ quantum dots of different sizes to study the effect of quantum dot size on CO₂ reduction. It was found that CsPbBr₃ with a diameter of 8.5 nm had the longest carrier lifetime, more negative band bottom potential and the highest catalytic activity. Xu et al. [32] designed CsPbBr₃/GO composites by combining graphene oxide (GO) with CsPbBr₃ and used them for photocatalytic CO₂ reduction. The CsPbBr₃/GO composites exhibited higher CO₂ reduction activity than pure CsPbBr₃ nanocrystals. It was found that the improved CO₂ reduction performance was mainly attributed to the existence of efficient charge transfer in CsPbBr₃/GO composites, and charge injection graphene oxide effectively promoted the charge injection from MHPs to GO. Other studies have shown that the catalytic activity of MHPs can be improved by coupling various modifiers with MHPs nanocrystals. Man et al. [33] used NH_x-rich porous g-C₃N₄ nanosheets (PCNs) to stabilize CsPbBr3 nanocrystals. The formation of a N-Br bond leads to close contact with g-C₃N₄ nanocrystals and CsPbBr₃ nanocrystals. In order to suppress the serious charge recombination in MHPs, Jiang et al. [34] designed a novel Z-scheme alpha-Fe₂O₃/amine-RGO/CsPbBr₃ catalyst for high-efficiency CO₂ reduction. The construction of the Z-scheme heterojunction promoted charge separation and retained strong reducing electrons in CsPbBr₃ as well as strong oxidation holes in α-Fe₂O₃. Finally, it promoted the activity of artificial photosynthesis.

4. Metal-Organic Frameworks (MOFs)

Metal-organic framework materials (MOFs) are organic-inorganic hybrid materials with intramolecular pores formed by self-assembly between inorganic metal ions or clusters and organic ligands through coordination bonds. MOFs' large specific surface area, high porosity and tunable structure make them energy storage materials with strong development potential. The structure of MOFs can be controlled by changing the central metal atoms and the interaction between different organic ligands. Transition metals such as Fe, Co and Ni are often selected as central metal sources. As the raw materials of MOFs, such transition metals are abundant, widely distributed and easily available on Earth, which to some extent makes the raw materials cost of MOFs low. Therefore, MOFs are widely used in catalytic energy conversion and other applications ^[35]. In photocatalytic processes, MOFs can be used as photocatalysts or modifiers to promote the photocatalytic reaction.

MOFs are a relatively new type of frame material, and their uniformly dispersed metal nodes are conducive to the adsorption and activation of gas molecules. The combination of semiconductors and MOFs to form inorganicorganic nanocomposites can not only satisfy the absorption of photons by semiconductor materials to generate carriers, but also enable the materials to adsorb and activate highly stable CO_2 molecules. Xiong et al. ^[36] developed a method for synthesizing Cu₃(BTC)₂@TiO₂ core-shell structures. The loose shell structure of TiO₂ facilitates the passage of CO₂ molecules through the shell, while the strong CO₂ absorption of Cu₃(BTC)₂ in the core favors the efficient CO2 reduction of nanocomposites. Wu et al. [37] designed and constructed inorganic perovskite quantum dots and organic MOF complexes. A series of MAPbI(3)@PCN-221(Fe-x) composite photocatalytic materials were prepared by encapsulating quantum dots in MOF channels by a deposition method. In the aqueous phase, the MAPbI(3)@PCN-221(Fe-0.2) composite structure showed a high photocatalytic activity for CO2-much higher than that of PCN-221(Fe-0.2). At the same time, hole oxidation oxidized H2O to O2 to achieve the total decomposition of CO₂. Alvaro et al. [38] reported that MOF-5 produced by the coordination of Zn²⁺ and terephthalic acid has semiconductor-like properties. There is an electron-hole separation and transfer process in which electrons migrate from ligand to adjacent metal nodes for the photocatalytic degradation of phenol. Subsequently, a great deal of research was conducted on the construction and development of MOFs with semiconductor properties. On the one hand, MOFs act as light absorbers. On the other hand, due to their unique structural characteristics, they can be used as highly efficient catalysts integrating light absorption and catalytic active sites [39]. Fang et al. [40] designed and synthesized a pyrazolyl porphyrinic Ni-MOF (PCN-601) with active sites of light capture exposure and high specific surface area. The experimental results showed that PCN-601 had high-efficiency CO₂ photoreduction performance under the condition of CO₂-saturated water vapor. Under the condition of simulated solar irradiation (AM 1.5 G), the CH_4 yield of PCN-601 reached 92 μ mol·h⁻¹·g⁻¹ and the apparent quantum yield (AQY) of CH₄ was 2.18%. Therefore, the accumulated research suggests that MOFs have the best interfacial charge transfer and kinetic process among the materials evaluated for photocatalytic CO₂ reduction. The optimal catalytic activity was obtained in the catalytic reaction system, which provided a new direction for guiding and controlling the synthesis of new MOF materials.

5. Other Semiconductor Photocatalysts

Compared with commonly used photocatalysts (MHPs, MOFs, TiO₂, etc.), other efficient photocatalytic materials for the reduction of CO₂ need to be developed and studied. The conduction band position of SiC has a comparatively more negative potential, which can produce photogenerated electrons with stronger reduction ability for the photocatalytic reduction of CO₂. However, the synthesis of SiC in a high-temperature protective atmosphere is not conducive to the regulation of its nanostructure [41][42]. Layered double hydroxides (LDHs) such as Zn-Al LDH [43], Mg–AI LDH [44] and Zn–Cu–Ga LDH [45] have been used for the photocatalytic reduction of CO₂. Teramura et al. [46] synthesized a variety of LDHs with surface alkaline sites for the photocatalytic conversion of CO_2 to CO. Their activities tend to be higher than that of pure hydroxide. Graphite carbon nitride (g-C₃N₄) is a metal-free polymer material considered to be a promising visible-light catalyst [47][48]. Hsu et al. [49] used graphene oxide as a catalyst for the efficient photocatalytic conversion of CO₂ to methanol, and synthesized a graphene catalyst with an improved method to improve the activity. The yield of CH₃OH was six times that obtained using pure TiO₂. The photocatalytic activity of highly porous Ga_2O_3 for the reduction of CO_2 was found to be more than four times that of commercial Ga_2O_3 without the need for additives ^[50]. The performance of porous Ga_2O_3 was improved by doubling the surface area and tripling the adsorption capacity. Tanaka et al. [51] used H₂ instead of H₂O as a reducing agent to photocatalytically reduce CO₂ on Ga₂O₃, and the product was CO instead of CH₄. Notably, nearly 7.3% of the surface-adsorbed CO₂ was converted.

An effective strategy is to develop new semiconductor photocatalysts with visible-light response to improve the utilization rate of sunlight, so as to obtain higher photocatalytic activity. Zhou et al. ^[52] prepared Bi_2WO_6 square nanoplates by a hydrothermal method, and the product obtained by reducing CO_2 was CH_4 .

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