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Research Progress on Water Treatment Membranes based on New Photocatalytic Materials

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Abstract

While achieving the separation requirements, the photocatalytic water treatment membrane can use its photocatalytic ability to degrade the pollutants on the surface of the membrane, inhibit the growth of bacteria, and reduce the membrane pollution, which shows unique advantages in water purification and wastewater treatment. Firstly, the basic mechanism of photocatalysis is briefly introduced, and the structure and performance characteristics of different new photocatalytic materials are summarized. Then, focusing on the design and development of photocatalytic water treatment membranes, the application and water treatment performance of graphite carbon-nitride (g-C_3N_4), carbon quantum dots (CQDs), layered dihydroxide (LDHs), metal-carbon/nitride (MXenes), metal-organic frameworks (MOFs) and covalent organic frameworks (COFs)-based photocatalytic water treatment membranes were emphatically discussed. Finally, the problems existing in the photocatalytic water treatment membrane are analyzed in depth, and its future development is prospected.

Keywords

Membrane Separation; Photocatalysis; Membrane Fouling; Water Treatment.

1. Introduction

In recent years, with the rapid development of industry and the sharp increase in population, a large amount of toxic and harmful pollutants have been discharged into the environment, leading to increasingly serious water pollution problems [1]. To address these issues, various water treatment technologies have been developed, such as activated sludge method, adsorption method, ion exchange method, membrane separation method, coagulation and precipitation method, etc. [2]. The membrane separation method, which is efficient, green, and easy to operate, can effectively remove salts, bacteria, and other organic pollutants (such as dyes, pharmaceutical intermediates, etc.) from groundwater, urban and industrial wastewater. It is a promising new separation technology [3]. However, membrane materials are easily contaminated by organic matter during water treatment, which affects the permeation flux and service life of the membranes [4]. Although physical cleaning and chemical cleaning can restore membrane flux to a certain extent and mitigate membrane fouling, these cleaning methods often require additional chemicals and energy consumption, which not only increases the process cost but also shortens the service life of the membranes, thereby weakening the competitiveness of membrane separation technology [5]. Therefore, the development of anti-fouling membrane materials has always been at the forefront and a hot topic in the field of membrane separation.

Photocatalysis is an efficient and sustainable advanced oxidation technology with high research and application value in wastewater treatment [6]. Anti-fouling photocatalytic membranes coupled with photocatalytic functions have received extensive attention and research in recent years [7]. These photocatalytic water treatment membranes can degrade pollutants adsorbed

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on the membrane through active radicals generated by photocatalysts under light irradiation, achieving separation while inhibiting the accumulation of pollutants on the membrane surface, reducing pore blockage, and achieving active anti-fouling. However, most photocatalytic water treatment membranes exhibit limited photocatalytic activity only under UV light irradiation [8]. Moreover, common metal semiconductor photocatalysts mostly contain rare metals, have complex preparation processes, or low solar energy conversion efficiency, resulting in poor practical application effects [9-11]. Therefore, finding efficient, stable photocatalysts responsive to visible light and integrating them into membrane materials appropriately is a key focus and challenge in the field of photocatalytic water treatment membranes. Currently, materials with excellent physical and chemical properties and photocatalytic abilities such as graphite carbon nitride (g-C_3N_4) [12-13], carbon quantum dots (CQDs) [14-15], layered double hydroxides (LDHs) [16-17], metal carbides/nitrides (MXenes) [18-19], metal-organic frameworks (MOFs) [20-21], and covalent organic frameworks (COFs) [22-23] have gained extensive attention in the field of photocatalysis. In summary, this review first introduces the mechanism of photocatalysis, then classifies and summarizes the performance characteristics of the above six new photocatalysts, and focuses on the preparation strategies and separation performance of photocatalytic water treatment membranes. Based on the broad application prospects of photocatalytic water treatment membranes, developing efficient, stable, and practically applicable photocatalytic water treatment membranes is the future development trend.

2. Mechanism and Materials of Photocatalytic Technology

2.1. Mechanism of Photocatalytic Technology

The strong redox capability of radicals is the foundation of photocatalytic technology. Under light irradiation, photocatalysts generate active radicals, which can undergo a series of chemical reactions with organic pollutants, degrading them and achieving water purification [24]. The entire process involves the following steps: First, when the energy (hv) absorbed by the photocatalyst is higher than its bandgap energy (Eg), electrons (e^-) in the valence band are excited and transition to the conduction band, leaving relatively stable holes (h^+), as shown in equation (1). Then, the excited electrons and holes migrate to the surface of the photocatalyst and initiate a series of redox reactions. Among them, electrons reduce O_2 to form superoxide radicals (O^{2-}). Due to their strong oxidative ability, holes can directly oxidize organic molecules or react with H2O to form hydroxyl radicals (OH). Ultimately, under the combined action of holes, superoxide radicals, and hydroxyl radicals, organic pollutants are decomposed into H_2O and CO_2, completing the photocatalytic degradation process. It is important to note that during the migration of electron-hole pairs, some electrons and holes may recombine and release energy. If the recombination rate of electron-hole pairs is high during the catalytic process, it will hinder the generation of radicals, thereby reducing the catalytic efficiency.

$$\text{Photocatalyst} + hv \rightarrow e^- + h^+ \quad (1)$$

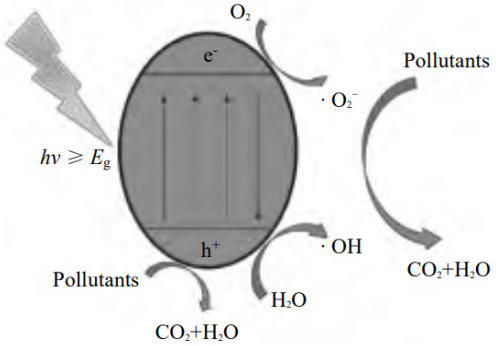

Fig 1. Mechanistic diagram of the photocatalytic process

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The basic mechanism of photocatalytic technology is briefly introduced here, but for photocatalytic membranes, the photocatalytic process is affected by many factors, including the band structure of the photocatalyst, the type of pollutants, and the presence form of the photocatalyst in the membrane. Therefore, specific photocatalytic mechanism analysis should be carried out for different reaction systems.

2.2. Materials for Photocatalytic Technology

The key to the development of photocatalytic membranes lies in the development of photocatalytic materials. In recent years, some new photocatalysts with excellent photocatalytic capabilities, good stability, and environmental friendliness, such as g-C_3N_4, CQDs, LDHs, MXenes, MOFs, and COFs, have received extensive attention and research in the design and preparation of photocatalytic water treatment membranes. Table 1 lists the basic characteristics of these six materials, such as structure, bandgap width, optical properties, and advantages. Among them, g-C_3N_4, LDHs, and MXenes have unique two-dimensional structures: g-C_3N_4 has a narrow bandgap and is resistant to photodegradation, making it widely studied as a metal-free semiconductor catalyst; low-cost LDHs and highly conductive MXenes have broad spectral absorption, adjustable bandgaps, and high specific surface areas, which allow them to function not only independently as photocatalysts but also in combination with semiconductor catalysts to enhance their development [25]. Zero-dimensional CQDs have upconversion photoluminescence properties and can serve as spectral converters to improve the solar energy absorption rate of UV-responsive semiconductor catalysts [26]. MOFs and COFs, with their porous organic structures, can be custom-designed at the molecular level to have different bandgaps and pore sizes, making them highly competitive in the field of catalysis. These materials all exhibit optical properties responsive to visible light, paving the way for the preparation of high-performance photocatalytic water treatment membranes.

Table 1. Three Scheme comparing

Photocatalytic Materia	Structure	Bandgap	Optical Properties	Advantages
g-C_3N_4	Amorphous or crystalline; layered structure	~2.7 eV	UV-visible spectral response	Non-toxic; photostable; easy to synthesize
CQDs	Amorphous or crystalline; layered structure	Size-dependent	Broad optical absorption; upconversion photoluminescence; UV-visible spectral response	Biocompatibility; high electron transfer capacity; easy to functionalize
LDHs	Crystalline; layered structur	2.2 -3.2 eV	Adjustable optical bandgap; UV-visible spectral response; broad spectral absorption	Low-cost; high chemical stability; large surface area
MXenes	Crystalline; layered structur	0.04-3.49 eV	Adjustable optical bandgap; UV-visible spectral response; broad spectral absorption	Hydrophilic; high electronic conductivity; large surface are
MOFs	Crystalline; mixed porous structure	1.0-5.5 eV	Strong light-harvesting ability; UV-visible spectral response; adjustable optical bandgap	High porosity; customizable structure; large surface area
COFs	Crystalline; organic porous structure	1.3-2.9 eV	Strong light-harvesting ability; UV-visible spectral response; adjustable optical bandgap	High porosity; strong thermal/chemical stability; large surface area

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According to the preparation method, the photocatalytic water treatment membrane can be divided into two types: mixed matrix type and surface supported type. The mixed matrix photocatalytic water treatment membrane is prepared by doping the catalyst in the polymer matrix, and the preparation process of this membrane is relatively simple and can effectively avoid the detachment of the catalyst. Surface-supported photocatalytic water treatment membranes are usually prepared by loading photocatalysts on the surface of the base membrane by surface grafting, self-assembly, vacuum filtration and in-situ growth, and often have a large photocatalytic area. In this section, the development of different photocatalytic water treatment membranes is summarized and analyzed from different types of photocatalysts.

3. Research Progress on Photocatalytic Water Treatment Membranes

3.1. Photocatalytic Water Treatment Membranes Based on g-C_3N_4

g-C_3N_4 is a visible-light-responsive porous sheet material whose porous characteristics can enhance the molecular sieving ability of water treatment membranes and reduce mass transfer resistance, providing unique advantages in the preparation of photocatalytic water treatment membranes [12]. Typically, g-C_3N_4-based photocatalytic water treatment membranes are prepared by phase inversion or surface loading methods such as grafting and vacuum filtration.

Yang et al. [27] physically blended mesoporous graphitic carbon nitride with polyvinylidene fluoride (PVDF) casting solution and prepared photocatalytic water treatment membranes by phase inversion. Increasing the catalyst content improved the hydrophilicity and antifouling properties of the membrane. Even under sunlight, the degradation rate of cefotaxime (CFX) by the photocatalytic membrane exceeded 97%, showing great potential in actual wastewater treatment. To further enhance the antibacterial activity and photocatalytic performance of the photocatalytic membrane, silver nanoparticles (AgNPs) with antibacterial activity can be used to modify g-C_3N_4, creating Ag/g-C_3N_4 composite catalysts with both antibacterial activity and photocatalytic degradation functions. Zhang et al. [28] introduced the modified Ag/g-C_3N_4 catalyst into polyethersulfone (PES) and prepared Ag/g-C_3N_4/PES ultrafiltration membranes by phase inversion. The prepared composite membrane exhibited excellent antibacterial, photocatalytic dye degradation, and antifouling performance under visible light irradiation.

When using conventional blending modification methods, the catalysts and polymer matrices may have high binding capabilities, but issues such as uneven catalyst distribution, incompatibility with the matrix, and inability to directly expose the catalyst to light may arise, reducing the separation and catalytic performance of photocatalytic water treatment membranes. To overcome these issues, Li et al. [29] used magnetic-induced freezing casting to prepare efficient Fe_3O_4/g-C_3N_4/PVDF membranes (FCMs) with macroporous structures. The magnetic Fe_3O_4/g-C_3N_4 composite catalysts can be directionally and highly exposed on the membrane surface under the action of a magnetic field, endowing the membrane surface with more photocatalytic active sites and forming an orderly network macroporous structure conducive to light penetration, thereby improving the visible light utilization rate, water flux, and stability of FCMs.

Additionally, chemical modification can be performed on the membrane surface to achieve stable catalyst loading through covalent bonds with specific functional groups. Kolesnyk et al. [30] achieved catalyst fixation by forming amino groups on the PVDF membrane surface activated with carbonate to form g-C_3N_4. Chi et al. [31] grafted polyacrylic acid (PAA) on polytetrafluoroethylene (PTFE) ultrafiltration membranes via plasma-induced grafting, using the carboxyl groups provided by the PAA layer to coordinate with Ti^{4+} to achieve g-C_3N_4/TiO_2 catalyst loading. The heterojunction formed by g-C_3N_4 and TiO_2 effectively expanded the visible light absorption range of TiO_2, improved the separation efficiency of photogenerated electron-

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hole pairs, and enhanced the photocatalytic degradation ability. During practical tests, this membrane exhibited high water flux, antifouling, and photocatalytic self-cleaning capabilities.

Compared with matrix modification, surface modification methods may face the problem of catalyst detachment in practical applications. To solve this issue, Huang et al. [32] added polyethylene glycol (PEG) and glutaraldehyde (GA) as crosslinkers to the g-C_3N_4/PVDF membrane surface, improving the self-cleaning ability of PVDF membranes while enhancing the stability of the catalyst. Additionally, some researchers used sodium alginate (SA) as an adhesive to achieve stable loading of g-C_3N_4 on the PVDF membrane surface [33]. Our research group used the condensation reaction between polyvinyl alcohol (PVA) and GA to form a stable coating on the surface of polyacrylonitrile (PAN) membranes modified with g-C_3N_4, thereby improving the poor adhesion between the catalyst and the base membrane.

In summary, further improving the performance and stability of photocatalysts is key to the application of g-C_3N_4-based photocatalytic water treatment membranes.

3.2. Photocatalytic Water Treatment Membranes Based on CQDs

CQDs are a new type of zero-dimensional carbon material [34] whose unique structure and rich surface functional groups can effectively enhance the hydrophilicity and permeability of the membrane surface. CQDs-based photocatalytic water treatment membranes are often prepared by surface grafting methods.

Using the coordination effect between the surface functional groups of CQDs and Ti^{4+}, Xie et al. [35] immersed the prepared TiO_2 membranes in nitrogen-doped CQDs (NCQDs) suspensions to prepare NCQDs/TiO_2 composite membranes. Under the influence of nitrogen doping, the composite membranes exhibited excellent photocatalytic performance. This is due to the fact that the doped heteroatoms reduced the work function of CQDs [36], increased the conduction band difference with TiO_2, and accelerated the separation of electron-hole pairs, thereby enhancing the light absorption and degradation ability of the NCQDs/TiO_2 photocatalyst in the composite membrane. In addition to photocatalytic degradation, CQDs have also been proven to have photo-induced sterilization functions [37]. Based on this, Chen et al. [38] successfully immobilized CQDs modified with antibacterial gold nanoparticles (AuNPs) on polyether sulfone (PSF) membranes modified with polydopamine (PDA) through the condensation reaction between PDA and carboxyl groups on the surface of CQDs. The synergistic effect of CQDs and AuNPs improved the antifouling ability of PSF membranes. Additionally, the CQDs/Au/PSF membranes exhibited excellent antibacterial activity against Escherichia coli, with a sterilization rate of 90.2%.

When CQDs-based catalysts are loaded on the membrane surface, they come into direct contact with organic pollutants, easily leading to catalyst pore blockage and resulting in catalyst inactivation. To address this issue, Shao et al. [39] constructed a polyamide layer on the surface of PES membranes modified with PDA-CQDs through the interaction between PDA and CQDs. This method not only alleviated pore blockage but also prevented the detachment of CQDs, extending the service life of CQDs photocatalytic membranes. Under visible light, CQDs improved the antifouling ability of PES membranes by degrading organic pollutants on the membrane surface. Additionally, blending catalysts with polymer matrices can also alleviate catalyst inactivation issues. Zhang et al. [40] mixed CQDs with PES casting solution and prepared CQDs/PES membranes through phase inversion assisted by a direct current electric field. Under the electric field, CQDs were uniformly distributed in the membrane matrix and highly exposed on the membrane surface, enhancing both the antifouling ability and water flux of the CQDs/PES membrane.

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3.3. Photocatalytic Water Treatment Membranes Based on LDHs

LDHs are a type of two-dimensional layered inorganic material with simple preparation processes and high chemical stability, making them ideal visible-light-responsive photocatalysts [41]. Currently, LDHs-based photocatalytic water treatment membranes are mainly prepared by surface self-assembly or phase inversion methods.

Mutharasi et al. [42] used ZnAl-LDH as a filler to prepare ZnAl-LDH/PEI mixed matrix ultrafiltration membranes and ZnAl-LDH/PEI polyamide (PA) nanofiltration membranes through blending methods, improving the water flux and antifouling ability of polyethyleneimine (PEI) membranes. Zong et al. [43] used the chelating effect between carboxyl groups on PDA and metal ions to load NiAlFe-LDH on PDA-modified PVDF membranes through in-situ growth, preparing NiAlFe-LDH/PDA/PVDF membranes capable of efficiently separating oil-water emulsions. NiAlFe-LDH not only significantly improved the hydrophilicity of the membrane surface but also exhibited excellent photocatalytic activity in degrading tetracycline under UV light irradiation. To treat mixed wastewater containing precious metal ions and dye molecules simultaneously, Wang et al. [44] prepared LDHs membranes doped with different metal ions (Ni, Al, Fe) on conductive fabrics and explored their separation mechanisms. In the mixed wastewater, silver ions were reduced by the LDHs membrane and deposited on the membrane surface, with silver nanoparticles acting as electron traps to reduce the recombination rate of photocatalyst electron-hole pairs, promoting the photocatalytic activity of the LDHs membrane for methyl orange (MO). Results showed that Fe-doped LDHs membranes exhibited the best visible-light photocatalytic performance.

To further investigate the photocatalytic reduction and degradation abilities of LDHs for heavy metal ions and organic compounds, Wang et al. [45] tested mixed wastewater doped with different heavy metal ions (Ag^{+}, Pb^{2+}, Cu^{2+}) and MO. Results showed that the higher the Fermi level of coexisting metal ions, the higher the degradation efficiency of MO by LDHs membranes, providing new insights into the removal of heavy metal ions and organic pollutants. To further enhance the catalytic activity of LDHs, Zhao et al. [46] prepared ZnWO4 and NiAl-LDH heterojunctions and loaded them on PVDF membranes through vacuum filtration. The synergistic effect of $ZnWO_4$ efficiently separated photogenerated electrons and holes, utilizing water molecules and free hydroxyl ions in LDHs interlayers to accelerate water molecule oxidation into hydroxyl radicals, further enhancing catalytic efficiency. Additionally, researchers often combine LDHs with carbon materials or conductive polymers with good conductivity, such as graphene oxide (GO), carbon nanotubes (CNT), polypyrrole (Ppy), etc., to improve the conductivity and photocatalytic performance of LDHs [47-49]. Li et al. [50] successfully prepared photocatalytic self-cleaning PAN/PEI nanofiber membranes coated with FeCu-LDH/GO catalysts through electrospinning and hydrothermal methods. Under visible light, the nanofiber membranes exhibited high antifouling ability, with a flux recovery rate (FRR) of 96.3% after photocatalytic degradation. Liu et al. [51] prepared LDHs@g-C_3N_4@PDA composite photocatalysts through dopamine modification and loaded them onto PVDF base membranes through vacuum filtration. Under visible light, the composite membrane achieved continuous dye separation and degradation in a short time, exhibiting excellent photocatalytic self-cleaning function.

4. Conclusion

This review first briefly introduces the basic mechanism of photocatalytic technology, then organizes the performance characteristics of six novel photocatalytic materials, including g-C_3N_4, CQDs, LDHs, MXenes, MOFs, and COFs. It then focuses on the research status of photocatalytic water treatment membranes based on these materials. Photocatalytic water treatment membranes possess both separation and catalytic functions, effectively solving

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membrane fouling problems in practical applications. Additionally, the successful development of novel photocatalytic materials and post-modification methods with visible-light responsiveness and excellent physicochemical properties provides new avenues for preparing photocatalytic water treatment membranes suitable for complex systems. Researchers have developed various new methods for loading photocatalysts to maximize their advantages in membranes, making significant progress in improving photocatalyst light absorption areas, uniform distribution, and stability. Based on broad application prospects, photocatalytic water treatment membranes are widely used in dye and antibiotic degradation in printing and dyeing wastewater and pharmaceutical wastewater, reduction of metal ions in mineral wastewater, degradation of oil contamination on membrane surfaces in oily wastewater, etc.

Although photocatalytic water treatment membranes based on novel photocatalytic materials have good development prospects, they still face challenges in practical preparation and application. First, there is a lack of long-term stability data for photocatalytic water treatment membranes, requiring further research. Second, the practical application of photocatalytic water treatment membranes requires corresponding production equipment, but currently used equipment is not suitable for industrial production. In summary, improving the stability of photocatalytic water treatment membranes and developing membrane equipment capable of photocatalytic water treatment remain hot research topics.

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