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RESEARCH PROGRESS OF SUPPORTED PHOTOCATALYTIC MEMBRANES

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Abstract: Photocatalytic technology and membrane separation technology are two new water treatment technologies. The photocatalytic membrane formed after coupling the two can not only fix the catalyst and alleviate membrane pollution, but also produce a synergistic effect and reduce pollutants in the water. degradation efficiency. The types of photocatalysts and supported membranes are reviewed, as well as the research status of photocatalytic supported membranes.

Keywords: Photocatalysis; Membrane separation; Supported photocatalytic membrane

1 CATALYST OF LIGHT

With the advancement of industry and the development of science and technology, people's living standards continue to improve, and environmental problems have gradually become more prominent. Reducing pollution from the source and improving pollution control measures are serious problems currently faced. Against the backdrop of global surface water quality deterioration and drinking water quality standards becoming increasingly stringent, cascade utilization of water resources and sewage reuse are important ways to solve problems such as water shortages. However, as the number of industrial enterprises continues to increase, the composition of wastewater has become more and more complex, such as dye wastewater, medical wastewater, farmland runoff rainwater, pharmaceutical wastewater, etc. The discharge of complex pollutants not only hinders the full utilization of water resources and reuse of sewage, but also causes serious harm to humans and aquatic animals.

Nowadays, traditional sewage treatment methods (such as activated sludge method) can no longer meet the requirements for degrading complex pollutants. Advanced oxidation technology is also a common sewage treatment technology, which uses ultraviolet light, ozone and H_2O_2 and other oxides to oxidize pollutants into CO_2 and H_2O and inorganic small molecules [1]. This technology has the advantages of high degradation efficiency and good effluent quality, but has the disadvantages of high energy consumption and high cost in actual industrial use. Photocatalytic oxidation technology can effectively degrade a variety of complex pollutants. In the sewage system, the hydroxyl radicals and oxygen ions generated by the photocatalyst after illumination will mineralize the pollutants into CO_2 and H_2O and some inorganic ions. Experimental studies have shown that photocatalytic technology can better degrade antibiotics [2], cell inhibitors [3], dyes [4], pesticides [5], endocrine disruptors [6], and Escherichia coli [7] in water. wait. Since photocatalytic oxidation technology uses light sources to degrade pollutants, especially organic matter, there are no secondary pollutants during the photocatalytic process, and it is non-toxic, has low energy consumption, and has low operating costs [8].

Another technology used in wastewater treatment is membrane separation. Membrane separation technology refers to the process of separating different components in water by using the selective permeability of the membrane material itself under the action of external pressure [9]. Membrane separation is a completely physical process and does not involve the degradation of pollutants. By combining membrane separation with photocatalysis, the synergistic effect of the two can be revealed. The photocatalyst can effectively degrade pollutants deposited on the surface of the membrane and effectively alleviate membrane pollution, while the membrane can fix the photocatalyst so that the photocatalyst can fully contact the pollutants to achieve the effect of degrading pollutants.

1.1 Mechanism of Action

FUJISHIMA A et al.[10] published an article on the water splitting experiment of N-type semiconductor materials in 1972, which attracted widespread attention to semiconductor photocatalytic materials. Including the degradation of organic and inorganic substances in the gas phase and liquid phase, hydrogen production, and photoreduction of CO2 wait.

Photocatalyst is essentially a type of semiconductor material with an energy band gap between its valence band (VB) and conduction band (CB). When the energy of illumination on the semiconductor surface is greater than the energy of its energy band gap, the electrons in the valence band are excited and transition to the conduction band, generating electron holes (h^+). The excited electrons are called photogenerated electrons (e^-). The photogenerated electrons and holes migrate to the surface of the semiconductor material and can react with other substances, showing reducing and oxidizing properties [11]. In water treatment, the oxidation property of electron holes is widely used. Since the oxidation property of the valence band is higher than that of general organic matter, hydroxyl radicals and superoxide radicals with high oxidative activity are generated [12-13].

1.2 Research Status

Common photocatalysts include TiO , C_3N_4 , ZnO, $Ag_3 PO_4$, CdS , CuO, ZnS, $CuWO_4$, VS_4 , V_2O_5 , Cu_2O, and Bi-based photocatalysts, etc. [14-24]. Among them, the most widely used is TiO_2 [25].

TiO_2 It is a type of N-type semiconductor material and has three crystal forms: brookite, rutile and anatase [26]. Under the irradiation of ultraviolet light (wavelength <385 nm), the electrons in the valence band are excited and transition to the conduction band (the bandgap width of anatase is 3.2 eV, and the bandgap width of rutile is 3.0 eV).). When anatase TiO2 TiO2 with rutile ore When the ratio is 4:1, its photocatalytic activity is optimal[27]. TiO_2 The reason why it is widely used is mainly because it is low in price, non-toxic and has stable physical and chemical properties. It absorbs light stably and does not produce optical radiation. It can show good photocatalytic activity under ultraviolet light irradiation. WANG X et al. [28] used N and P co-doped TiO_2 Loaded onto expanded graphene, a composite material with carbon layers floating on water was created. This photocatalytic material can efficiently utilize light sources and is easy to recycle. Experimental results show that the removal rate of microcystin by the composite photocatalytic membrane can reach 99.4%.

In addition, graphite phase carbon nitride (g-C_3N_4) has also attracted the attention of researchers because of its unique layered structure [29]. g-C_3N_4 It is also a type of non-metallic polymer semiconductor material with a forbidden band width of 2.7 eV (valency band width of 1.4 eV and conduction band width of-1.3 eV). It can be excited by 460 nm light to produce photogenerated electrons. and electron holes, has photocatalytic activity, and can be used to degrade pollutants in sewage, photolyze water to produce hydrogen, and reduce CO_2 and disinfection, etc.[30-33]. g-C_3N4 It has the characteristics of low cost, low toxicity, environmental friendliness, and the ability to respond to visible light [34]. However, because it is essentially a type of polymer material with relatively low quantum efficiency and low redox potential, g-C_3N_4 is limited. $_4$ Applications. ZHAO H et al. [35] used vacuum filtration and high-pressure technology to purify g-C_3N_4 The loaded graphene oxide nanosheets are loaded onto the cellulose acetate membrane, and the resulting g-$C_3N_{4/}$RGO composite cellulose acetate membrane exhibits strong dye removal rate and antibacterial properties.

ZnO is a type of N-type semiconductor material with a bandgap width of 3.2 eV. It has the advantages of diverse forms, high electron transfer efficiency, low price, and no pollution to the environment [36]. The preparation conditions of ZnO are different, and the crystallinity and specific surface area of the material are also different, which in turn affects its photocatalytic activity. ZnO is widely used because of its easy-to-control morphology. CANTARELLA M et al. [37] used co-precipitation method to prepare ZnO. During the preparation process, acetaminophen was added, which changed the morphological structure of ZnO and significantly enhanced the adsorption efficiency of acetaminophen.

Bi System catalysts mainly include $BiVO_4$, Bi_2WO_6, Bi_2MoO 6 , BiOBr, BiOI, $BiFeO_3$, $CaBi_2O_4$, Bi_2O_3, Bi_2S_3, $Bi_2Ti_2O_7$, BiOCl, etc. [38-40]. Among them, BiOI is a type of P-type semiconductor material with a bandgap width of 1.8 eV (valence band width of 2.35 eV, conduction band width of 0.54 eV) and a unique layered structure. BiOI is composed of two layers [Bi_2 O2] 2 + It is composed of ion layer, with Bi-ion layer in the middle. Since BiOI has a narrow bandgap, it has obvious absorption of visible light, but pure BiOI has limited ability to remove pollutants. The degradation effect of a single Bi-based photocatalyst cannot meet the needs. Most studies have constructed two photocatalysts into a heterojunction structure through semiconductor compounding. HUANG HW et al. [38] prepared BiVO 4 by hydrothermal method and BiOI by sol-gel method. The nanoparticles are combined with BiOI nanosheets to build an N-P type heterojunction structure. When degrading rhodamine B and phenol, it shows better performance than using $BiVO_4$ alone. Or better results when BiOI degrades.

Bi-based catalysts can also be combined with other catalysts to construct heterojunctions. For example, LI B et al. [41] used hydrothermal method to prepare BiOI/TiO_2 Heterojunction. Experiments show that BiOI/TiO_2 Compared to BiOI or TiO_2 alone has better photocatalytic performance, and when BiOI and TiO_2 When the molar ratio is 1:5, the highest pollutant degradation rate is achieved. This is because BiOI can interact with TiO_2 Form a P-N heterojunction structure to effectively reduce TiO_2 The bandgap width exposes more active sites.

2 PHOTOCATALYTIC FILM

2.1 Mechanism of Action

Membrane separation technology is a process that uses the selective permeability of membranes to separate and purify products. Membrane separation technology is widely used in many fields such as chemistry, petroleum, energy, biology, and environmental protection [42], including oil-water separation, nitrate hydrogenation, water purification, etc. [43-44]. The application of membrane separation technology to water purification has the advantages of low energy consumption, small footprint, and no secondary pollution. It has been actually used in various industrial wastewater and domestic sewage reuse projects [45]. The most important problem in the process of treating wastewater in ordinary membrane separation reactors is membrane fouling [46]. The essence of membrane fouling is the deposition of trapped substances on the membrane surface, which can be divided into physical deposition, chemical deposition, organic matter deposition, and biological deposition. Body deposition, etc.[42]. The occurrence of membrane fouling reduces the membrane flux, increases the transmembrane pressure, further shortens the service life of the membrane, and significantly increases the application cost [47].

2.2 Research Status

The photocatalytic process is combined with membrane separation technology to form a photocatalytic membrane system. The coupling of the two can not only efficiently treat wastewater, but also effectively alleviate the membrane fouling problem caused by membrane separation [48]. According to the pore size of the membrane, it can be divided into ultrafiltration, nanofiltration, microfiltration and reverse osmosis. Kind[49]. According to the material of the membrane, it can be divided into inorganic membrane and organic membrane [50-51].

At this stage, researchers at home and abroad have developed a variety of membrane base materials that can be used to support photocatalysts: inorganic membranes, including alumina, silicon carbide, etc.; metal mesh materials, including titanium mesh, copper mesh, etc.; glass fiber; Carbon fiber; High molecular polymer membrane; Cellulose and its derivatives, such as cellulose acetate, nitrocellulose, etc.

Inorganic membranes have the advantages of acid and alkali resistance, high temperature resistance, stable chemical properties, high mechanical strength, and easy cleaning [52], so they are widely used in petroleum, industry, food and other fields. Inorganic membranes are made of inorganic materials such as alumina, zirconia, silicon carbide, titanium dioxide, etc., added with appropriate amounts of additives and calcined at high temperatures. Its main structure consists of three parts: support layer, transition layer and membrane layer. Loading photocatalysts onto ceramic membranes can not only degrade pollutants, but also effectively alleviate membrane fouling. ZHANG Q et al. [53] used dipping-coating method to convert TiO2 The nanofibers are loaded onto the surface of the hollow fiber ceramic membrane, and the removal rate of humic acid by the loaded membrane can reach 90%. This proves that loading photocatalysts onto the surface of inorganic membranes can make the inorganic membranes self-cleaning and effectively alleviate membrane fouling. This self-cleaning membrane has also been used in practical separation and purification processes [54]. However, since the preparation process of inorganic membranes requires high-temperature calcination, the cost is high and the number of reuses in experiments is small, thus limiting its wide application.

Metal mesh materials have a uniform and dense mesh structure, good chemical stability, thermal stability and mechanical properties. LIN YQ et al. [55] used TiO2 Loaded onto the Ti film, the catalyst and film in the resulting photocatalytic film overcome the mismatch in thermal expansion coefficients between the catalyst and the film in traditional ceramic photocatalytic films, and have good photocatalytic degradation of dyes. QIAN DL et al[56] combined silver ions, sulfonated graphene oxide and TiO2 Loaded onto copper mesh respectively, the prepared copper mesh has bidirectional repellency and can effectively separate water-oil emulsion. Moreover, during the degradation process, dense TiO2 The clusters are loaded on the copper mesh to prevent copper from being corroded and oxidized. Metal mesh materials can be fixed by welding during industrial use. Therefore, this metal grid photocatalytic film has good practical application prospects.

Glass fiber is usually made of SiO2 Made with good UV light transmittance. Compared with other membrane materials, photocatalytic membranes made of glass fiber have a larger light contact area and thus have better catalytic activity. RAO GY et al. [57] synthesized TiO2/Fe2O3/GO loaded glass fiber membrane for degradation of humic acid. The degradation efficiency of humic acid under 2 h of UV light irradiation was 98%.

Carbon fiber is a common material that has been widely used as a support for various catalysts. The use of carbon fiber materials in photocatalytic membranes can not only increase the specific surface area of the catalyst, but also has good mechanical properties, electrical conductivity and corrosion resistance [23]. Shen

There are many types of polymer membranes, which can be roughly divided into polyolefins [59], polyamides [60], polysulfones [61-62] and fluorine-containing polymer materials [63]. This type of photocatalytic membrane is usually prepared by interfacial polymerization or phase inversion. The composite membrane produced has the advantages of good chemical and thermal stability, rich membrane pores, and the photocatalyst is not easy to fall off. YU S et al[62] prepared g-C3 N4/TiO2 through phase inversion method Composite polysulfone membrane degrades sulfamethoxazole to simulate pharmaceutical wastewater. Experimental results show that the composite polysulfone membrane can successfully degrade sulfamethoxazole into 7 intermediate products. However, the cost of polymer membranes is relatively high, and the reusability and durability of the membranes are poor.

Cellulose and its derivatives (such as cellulose acetate, cellulose nitrate, etc.) are a relatively common type of laboratory filter membrane. Its production process is mature, low-priced, and easy to obtain. In terms of filtration, it has good thermal stability and low adsorption capacity. LIF et al.[64] modified RGO/g-C3 N4 with polydopamine Composite photocatalyst, and then use vacuum filtration method to load the composite catalyst onto the cellulose acetate filter membrane. The composite membrane has a high interception efficiency for methylene blue dye wastewater and still has good photocatalytic activity after being recycled for 5 times under ultraviolet light irradiation. This type of cellulose filter membrane can not only achieve firm loading and maintain the photocatalytic activity of the photocatalyst, but the raw materials are easily available and the preparation process is simple. MOHAMED MA et al. [65] used old newspapers as cellulose raw materials and prepared N-modified TiO2 through phase inversion method. The composite cellulose acetate filter membrane was used to degrade phenol wastewater and achieved good experimental results.

3 CONCLUSION

There are many types of supported photocatalytic membranes that can be used for membrane separation and photocatalysis at the same time, and composite membranes have great application potential in water treatment. However, there are still many problems that need to be solved, such as the preparation process that enables the photocatalyst to be firmly loaded, The development of efficient and stable membrane materials and the coupling mechanism of photocatalytic membrane separation, etc. As environmental problems become increasingly severe, multifunctional photocatalytic membranes have broad development prospects in the fields of degradation of pollutants and reuse of sewage.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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