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Review

Performance improvement of hybrid polymer membranes for wastewater treatment by introduction of micro reaction locations

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ABSTRACT

This article reviews the progresses on polymer membranes embedded with novel inorganic nanomaterials to provide micro reaction locations (MRLs) in the polymer matrices for wastewater treatment. Ultrafiltration (UF) membranes, especially polyvinylidene fluoride (PVDF) and polysulfone (PSF) membranes, are broadly applied in wastewater treatment. The strategy of embedding functional nanomaterials into the polymer matrices has been extensively investigated to enhance the integrated properties of polymer membrane. Nevertheless, the performance enhancement just comes from physical interactions between nanomaterials and wastes, while the chemical interactions are not involved, thus limiting further improvements. In order to further enhance the integrated properties of polymer membranes, functional inorganic nanomaterials that can chemically react with the wastes are proposed and embedded into the polymer membranes to form many MRLs. In this paper, the strategies for embedding functional nanomaterials such as sulfated TiO₂ deposited SiO₂ nanotubes, solid superacid porous ZrO₂ shell/void/TiO₂ core particles and porous Y_xFe_yZr_{1-x-y}O₂ coated TiO₂ solid superacid nanoparticles in polymer membranes were presented and the enhancement effect of MRLs on their integrated properties for wastewater treatments was discussed. Therefore, polymer membranes embedded with functional inorganic nanomaterials with MRLs are potentially applied in various wastewater treatments.

1. Introduction

The strategy of embedding functional nanomaterials into ultrafiltration (UF) membranes has been extensively investigated in order to enhance integrated properties of the hybrid membranes, such as hydrophilicity, anti-fouling and anti-compaction, and achieve attractive improvements in wastewater treatment.

UF is one representative membrane technology [1], and has received wide acceptance due to its various outstanding advantages, such as excellent separation efficiency, low energy consumption and easy maintenance [2]. UF membranes constructed with mesoporous structure are able to separate both the large particles, microbes, oil droplets and dissolved bio-macromolecules (including pyrogens, proteins, and viruses). However, the further applications of UF polymer membranes, especially PVDF and PSF membranes [3], have been restricted owing to their poor hydrophilicity, poor anti-fouling capacity and low anti-compaction property. As one prevalent strategy, embedding nanomaterials [4–7] into casting solutions has been intensely investigated to enhance anti-fouling and hydrophilic properties of polymer membranes. Thus various types of metal oxides, such as ZnO, SiO₂, TiO₂, and

Mg(OH)₂ [5,8–11], have been carefully examined in embedding into polymer membranes to enhance anti-fouling and hydrophilic properties. However, the application of these traditional metal oxides is restrained, which is ascribed to their limited hydrogen bonds on the surface. To overcome this drawback, Zhang et al. embedded non-stoichiometric oxides such as Ce_xSi_{1-x}O₂ [12], sulfated Y_xZr_{1-x}O₂ [13] and phosphorylated Zr_xSi_{1-x}O₂ [14] into polymer membrane and found that the integrated performance of the membrane is effectively improved. Unfortunately, embedding these nanomaterials into polymer membranes enhance the comprehensive properties only via physical interactions at the interface between aqueous solution and the polymer/nanomaterial, without any chemical reactions between them, restraining the further improvement of polymer membranes. According to the analysis of membrane fouling mechanism [15,16], membrane fouling is mainly caused by three classes of pollutants, namely organic pollutants, inorganic pollutants and microbes in the wastewater [17–21]. In the process of wastewater treatment, organic pollutants (hydrocarbon and soluble oil etc.) and microbes can form a gel layer on the surface of the membrane, which leads to the increase of membrane resistance and reduction of the permeate flux [22,23]. Inorganic

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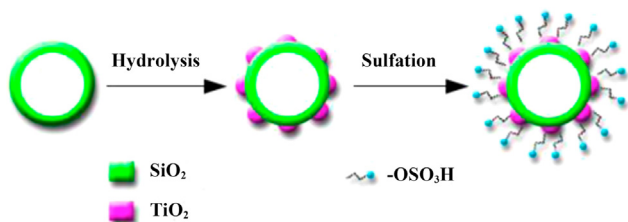


Fig. 1. Schematic diagram of STSNs preparation [26].

pollutants such as metal oxides can be adsorbed and deposited on the surface and inside channels of the membrane [24,25], causing the decline of the permeate flux. Therefore, in order to further enhance the hydrophilic and anti-fouling properties through involving both physical interactions and chemical reactions, novel functional nanomaterials are used to fabricate polymer membranes, forming micro-reaction locations (MRLs) on the channel wall and the surface of the membrane. The nanomaterials in MRLs discussed in this review paper have certain advantages. For example, TiO_2 with high photocatalytic activity can effectively degrade organic pollutants and microbes, delaying or eliminating the formation of the gel layer on the membrane surface. The embedded nanomaterials can also degrade and prevent the formation of inorganic pollutants such as metal oxides via reacting with strong solid superacid. Therefore, the performance of hybrid membranes by embedding such functional nanomaterials into polymer matrices has been evidently improved in wastewater treatment.

This article thus summarizes the recent progresses on the enhancement effect of MRLs on the properties of polymer hybrid membranes using sulfated TiO_2 deposited SiO_2 nanotubes, solid superacid porous ZrO_2 shell/void/ TiO_2 core particles and porous $\text{Y}_x\text{Fe}_y\text{Zr}_{1-x-y}\text{O}_2$ coated TiO_2 solid superacid nanoparticles as examples for embedded functional nanomaterials. This review has clearly demonstrated that these MRLs incorporated into polymer membranes evidently enhance the integrated properties, and such a membrane technology can be very potentially applied in wastewater treatment.

2. Sulfated TiO_2 deposited SiO_2 nanotubes (STSNs)

Sulfated TiO_2 deposited SiO_2 nanotubes (STSNs) with super strong acidity [26], tubular structure and high ratio of length to diameter were firstly prepared in our group and then added to PSF to prepare STSNs/PSF hybrid membranes. Typical STSNs with an average length of $8\ \mu\text{m}$ and inner diameter of around $300\ \text{nm}$ were prepared. The synthesis approach is schematically shown in Fig. 1. SNTs (SiO_2 nanotubes) were firstly prepared by a template-guided route, then $\text{SO}_4^{2-}/\text{TiO}_2$ solid superacid was deposited on the surface of SNTs to obtain STSNs by hydrolyzation and sulfation. The as-obtained STSNs were added to PSF to prepare STSN/PSF hybrid membrane by the phase inversion method.

Fig. 2 displays the TEM images of STSNs synthesized under the optimal conditions. As clearly seen in Fig. 2, the inner diameter of

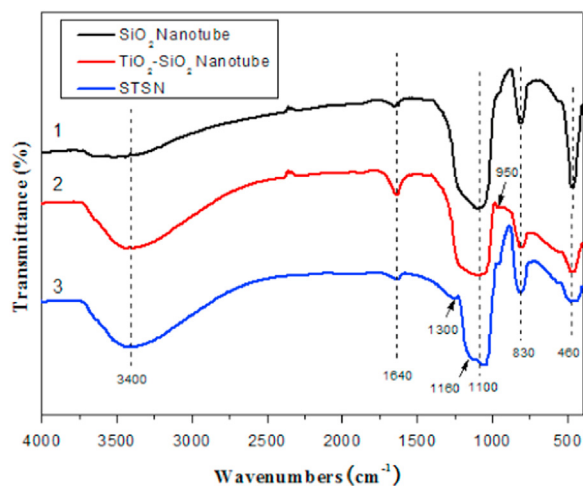


Fig. 3. FTIR spectra of SiO_2 nanotubes, TSNs and STSNs [26].

STSNs is $300\ \text{nm}$, the length is around $8000\ \text{nm}$, and the tube thickness is $50\ \text{nm}$, with the ratio of length to diameter being approximately 27. Moreover, the tube wall of STSNs is not as smooth as that of SiO_2 tubes, as there are many particles on the external surface, indicating the successful deposition of TiO_2 nanoparticles on the SiO_2 nanotube surface.

The infrared spectra of SiO_2 nanotubes, TSNs (TiO_2 deposited SiO_2 nanotubes) and STSNs are depicted in Fig. 3. The absorption peaks at $830\ \text{cm}^{-1}$, $1100\ \text{cm}^{-1}$ and $460\ \text{cm}^{-1}$ are the symmetric, asymmetric and bending vibrations of Si-O-Si, respectively [27,28], suggesting that TSNs and STSNs are SiO_2 based materials. Furthermore, the adsorption peaks at $3400\ \text{cm}^{-1}$ and $1640\ \text{cm}^{-1}$ are the stretching and bending vibrations of O-H on the surface of TSNs and STSNs, respectively. The strong O-H peaks may reveal that the hydrophilicity of SiO_2 nanotubes has been increased after the deposition of sulfated TiO_2 . In the spectrum of TSNs, the peak at $950\ \text{cm}^{-1}$ can be specifically assigned to Si-O-Ti stretching vibration [29], which indicates that the TiO_2 has been successfully deposited on SiO_2 nanotubes. In the spectrum of STSNs, the asymmetric and symmetrical stretching vibrations of $\text{S}=\text{O}$ are corresponding to the peaks at $1160\ \text{cm}^{-1}$ and $1300\ \text{cm}^{-1}$, suggesting that SO_4^{2-} groups have been grafted on the surface of TiO_2 . Consequently, the FTIR spectra clearly indicate that the sulfated TiO_2 nanoparticles are successfully deposited onto the surface of SiO_2 nanotubes.

Fig. 4 shows the SEM images of the cross-section of STSNs/PSF hybrid membranes. The as-prepared STSNs/PSF hybrid membranes have a typical asymmetric structure with a compact skin layer and a finger-like porous support. First, there exists a good interaction between the STSNs and PSF polymer matrices, which improves anti-compaction and hydrophilic properties, thus enhancing membrane performance. Second, as shown in Fig. 4(a) and (c), no obvious pore diameter change

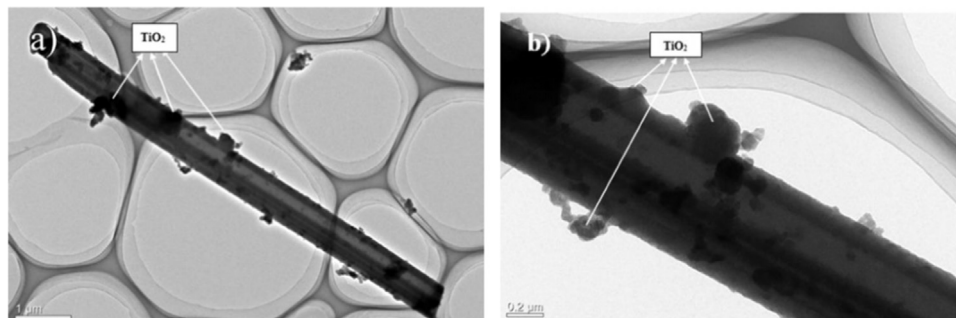


Fig. 2. TEM images of STSNs [26].

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