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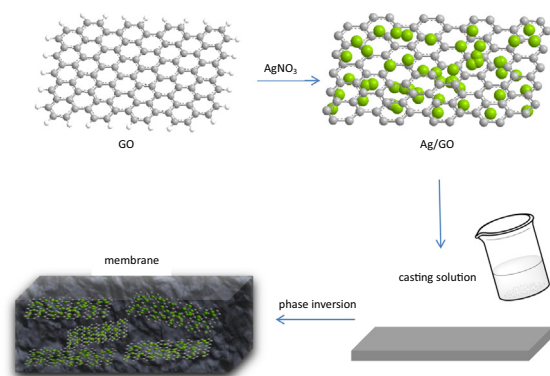
Anti-bacterial properties of ultrafiltration membrane modified by graphene oxide with nano-silver particles



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GRAPHICAL ABSTRACT



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ABSTRACT

To improve the anti-biofouling properties of PVDF membranes, GO-Ag composites were synthesized and used as membrane antibacterial agent by a simple and environmentally friendly method. As identified by XRD, TEM and FTIR analysis, AgNPs were uniformly assembled on the synthesized GO-Ag sheets. The membranes were prepared by phase inversion method with different additional amounts (0.00–0.15 wt%) of GO-Ag composites. The GO-Ag composites modified membranes show improved hydrophilicity, mechanical property and permeability than unmodified PVDF membrane. Specially, the antibacterial properties and inhibition of biofilm formation were greatly enhanced based on conventional inhibition zone test and anti-adhesion of bacterial experiment. The modified membranes also reveal a remarkable long-term continuous antimicrobial activity with slower release rate of Ag^+ compared to AgNPs/PVDF membrane.

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1. Introduction

Ultrafiltration (UF) has a wide application in drinking water production and wastewater treatment due to its low cost and high liquid separation efficiency [1,2]. However, membrane fouling

problems always exist in an emerging separation/desalination membrane process, which limits the practical applications. For all types of fouling, biofouling is the most complicated and inevitable in membrane separation process, since the biofilm is not easily removed and results in a decrease in water flux and selectivity [3,4]. In addition, higher operating and maintenance cost is required for cleaning of biofouling on membrane by mechanical or chemical approaches [5]. Hence, to overcome such disadvan-

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tages, various strategies have been proposed to suppress the biofouling of membrane process in recent years [6,7]. A common strategy employed to prevent biofilm growth and membrane fouling is the addition of antibacterial nanoparticles, such as titanium dioxide (TiO_2), silicon dioxide, and zinc oxide (ZnO), to the membrane backbone materials during membrane preparation [8–10]. Another effective way to mitigate the membrane fouling was to increase the membrane layer hydrophilicity [11]. A more hydrophilic membrane surface was usually less susceptible to be fouled under the same separation conditions [12].

Polyvinylidene fluoride (PVDF) is a common material for ultra-filtration membrane preparation for its excellent chemical stability and thermal performance [13]. However, the inherent hydrophobicity of the PVDF membrane is easy to be contaminated by bacteria and causes biofouling on the surface or in the pores of membranes during wastewater treatment process [14]. Based on this mechanism, a number of works have been reported to increase the hydrophilic properties of PVDF membranes by blending metallic and non-metallic nanoparticles into PVDF matrix [1,15,16].

In recent years, graphene oxide (GO) has been used as modifier in polymer membranes to enhance the mechanical strength and

hydrophilic properties of membranes due to its large specific surface area and abundant oxygen-containing surface groups [17–19]. Due to its strong inhibitory and biocidal effects [20,21] silver nanoparticles (AgNPs) and silver composite have been used to decrease membrane biofouling [22–24]. In our previous study, the hydrophilicity, permeability and in situ antibacterial properties were improved for the Ag-embedded nano-sized titanium dioxide (Ag-n-TiO_2) modified polyvinyl chloride (PVC) membranes [25]. However, the nanoscale AgNPs are hard to be fixed in the membrane firmly by physical blending and thus Ag^+ is easily isolated from chemical groups in the complex and actual water condition [26].

Silver nanoparticles assembled on graphene oxide sheets (GO–Ag) have been exploited as a novel antibacterial agent [27]. GO's functional groups provide ideal nucleation sites for AgNPs. Therefore, AgNPs can strongly attach to the GO surface [27]. In addition, GO can be easily dispersed into polymer matrix, and increases the compatibility of membrane casting solution. Therefore, in this paper we report the fabrication of a novel type of PVDF mixed membrane by embedding GO–Ag composites. Since few of literatures have reported the application of GO–Ag composites as

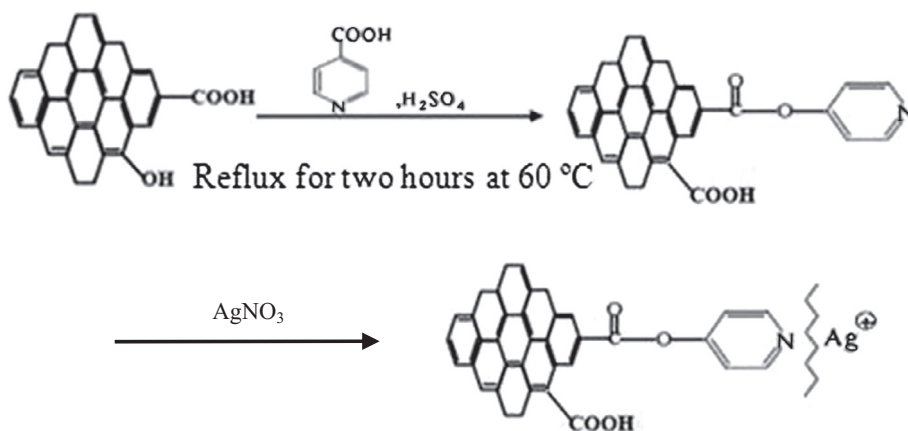


Fig. 1. Principles of the reactions of modified GO.

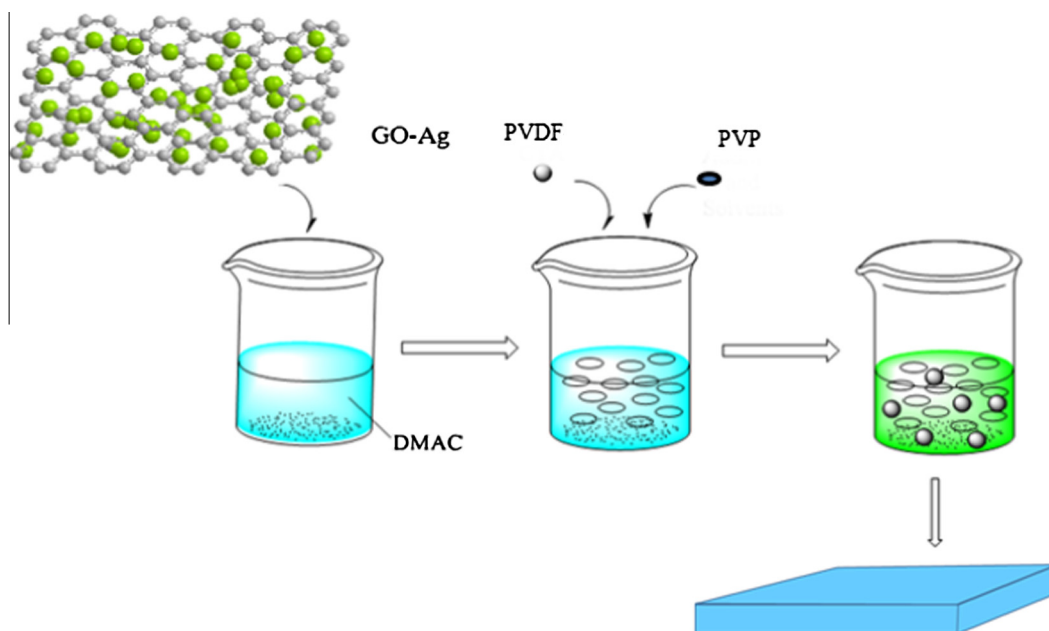


Fig. 2. Schematic diagram of the fabricated processes of GO–Ag/PVDF membrane.

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