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Modification of poly(vinylidene fluoride)/polyethersulfone blend membrane with polyvinyl alcohol for improving antifouling ability



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ABSTRACT

The modification of poly(vinylidene fluoride)/polyethersulfone (PVDF/PES) blend membranes was carried out by adding polyvinyl alcohol (PVA) to improve their antifouling ability. The physicochemical characteristics of the membranes in terms of porosity, contact angle, pure water flux, critical flux and fouling rate were systematically analyzed. The extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory was used to assess the antifouling ability of the modified membranes, and the membrane's long-term performance was also evaluated in a pilot-scale submerged anoxic/oxic membrane bioreactor for wastewater treatment. The results showed that the hydrophilicity and permeability of PVDF/PES blend membrane were efficiently improved by adding 0.3% PVA. The increase of critical flux and the decrease of fouling rate for the modified membrane were also observed in batch filtration experiments. XDLVO analysis indicated that the addition of PVA significantly improved the cohesion free energy of the membranes. The energy barrier between soluble microbial products and membrane surfaces was increased, which therefore reduced the adsorption of membrane foulants. The long-term filtration process also demonstrated that the antifouling ability of PVDF/PES membrane modified by 0.3% PVA was significantly enhanced. The study offers an effective way to enhance membrane antifouling ability of PVDF/PES membrane through the addition of PVA additives.

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1. Introduction

Poly(vinylidene fluoride) (PVDF) is a prevalent membrane material in microfiltration (MF) and ultrafiltration (UF) membrane preparation because of its excellent chemical resistance, high mechanical strength and thermal stability [1–4]. However, there are some problems of PVDF membranes, such as hydrophobic nature and low surface energy properties, which could affect their permeability and antifouling ability. Therefore, it is of significance to modify PVDF membrane for reducing its hydrophobic nature and improving its antifouling ability [5,6].

To date, a number of modification methods for PVDF membranes have been developed [7–10]. Among them, two methods are widely used, surface and blending modifications. Surface modification is usually achieved by coating or grafting a functional layer on the prepared membrane surface, in which most of the modified sites remained on the top and/or at the bottom layer of the membrane [3]. However, it is inefficient in modifying the pores

inside the membrane due to the limited diffusion of the modification agents. Blending modification, the simplest and most effective modification method, is usually used to achieve the desired functional properties along with the membrane preparation. Both the surfaces and inside-pores of the membranes can be modified synchronously through the synergy interactions between the basic polymer and additives. It can enable the prepared membrane to have the comprehensive characteristics of the blend materials [7,10].

Hydrophilic polymer materials and fine inorganic particles have been used as blend materials for modifying membranes [11]. For the polymer blend modification, it is expected to combine the advantages of each polymer and overcome the respective deficiencies of the original materials. Many polymer materials have been used to modify PVDF membranes, for instance polyacrylonitrile (PAN), polyether sulfone (PES), and amphiphilic hyperbranched-star polymer [12–15]. In recent years, polyvinyl alcohol (PVA) has also been used as a blend polymer since PVA is a nontoxic and biocompatible polymer with good film-forming properties and outstanding physicochemical stability [16]. Although PVDF membrane blended with PES polymer has been intensively investigated [15,17], the information of PVDF/PES blend membrane modified with PVA is very limited.

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In this study, the modification of PVDF/PES blend membrane by PVA was carried out to improve the membrane antifouling ability. PVDF/PES blend membranes were firstly prepared, and the optimal recipe for PVDF/PES blend membrane was selected by comparing contact angle, pure water flux and shrinkage ratio. Then, the PVDF/PES blend membrane was modified by PVA by adding different contents of PVA in the casting solution. The antifouling ability of the modified PVDF/PES/PVA membrane was evaluated by the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory, and the long-term performance of the membrane in a pilot-scale submerged anoxic/oxic membrane bioreactor (A/O-MBR) for wastewater treatment was also monitored for over 180 d. The results obtained in the preceding study are expected to provide a pathway for preparing antifouling PVDF membranes.

2. Materials and methods

2.1. Materials and experimental set-up

Commercial grade PVDF, with $\eta = 1.4\text{--}1.9$ Pa s, was purchased from Shanghai 3F New Material Ltd. (Shanghai, China). Polyether sulfone (PES) was supplied by BASF Shanghai Co. Polyvinyl alcohol (PVA, 1788) used as polymer additives was provided by Shanxi Sanwei Co., Ltd. Dimethyl acetamide (DMAC, analytical reagent) and dimethylsulfoxide (DMSO, analytical reagent), being the solvents, were obtained from Sinopharm (Shanghai, China). The polyethyleneglycole (PEG 600, analytical reagent), a pore-forming additive, was also purchased from Sinopharm (Shanghai, China). Deionized water was used throughout the membrane preparation.

A pilot-scale submerged anoxic/oxic membrane bioreactor (A/O-MBR) (as shown in Fig. 1) located in Quyang Municipal Wastewater Treatment Plant (WWTP) of Shanghai was used to evaluate critical fluxes, fouling rates and long-term performance of the membranes. The effective volumes of anoxic and oxic zones were 0.41 m^3 and 0.49 m^3 , respectively. The hydraulic retention time (HRT) of anoxic and oxic zones were 3.6 h and 4.4 h, respectively. The sludge retention time (SRT) was maintained at 60 d by daily discharging waste activated sludge. The specific aeration demand (SAD_m) was $1.0\text{ m}^3/(\text{m}^2\text{ h})$. The mixed liquor suspended solids (MLSS) concentration was about $6.01 \pm 1.04\text{ g/L}$ ($n=23$) in the reactor during the experiment. The raw wastewater was used as the influent, of which the characteristics are as follows ($n=36$): chemical oxygen demand (COD) $293 \pm 86\text{ mg/L}$, total

nitrogen $48.6 \pm 11.7\text{ mg/L}$, ammonium $39.2 \pm 9.3\text{ mg/L}$ and total phosphorus $5.0 \pm 0.9\text{ mg/L}$. The membrane modules each with an effective filtration area of 0.45 m^2 were mounted vertically between the baffle plates in the oxic zone. The effluent was extracted through the membranes by a peristaltic pump (Model BT-300, Baoding Longer Precision Pump Co., Ltd., China). The effluent flow rate and the trans-membrane pressure (TMP) were monitored by a water meter and a mercurial pressure gauge, respectively.

2.2. Membrane preparation

In the PVDF/PES blend casting solution, the sum of PVDF and PES concentration was 10 wt% and PES concentrations were set as 0%, 1%, 2% and 3%. The prepared PVDF/PES membranes were termed G1, G2, G3, and G4, correspondingly. DMAC and DMSO were used as solvents and PEG-600 was used as additive. The optimal recipe of PVDF/PES blend membrane was determined by analyzing contact angles, pure water fluxes and shrinkage ratios. According to the measurement results shown in Table S1 in the supporting information, G3 recipe was used as the basic membrane recipe for the following experiments. The modification of PVDF/PES membranes of G3 recipe was then carried out by adding PVA with the concentrations of 0%, 0.3% and 0.5% in the casting solutions. The modified PVDF/PES/PVA membranes were termed C1, C2 and C3, respectively.

In this study, all the membranes were prepared by phase inversion via the immersion precipitation method. The casting solutions were dissolved at $80\text{ }^\circ\text{C}$ for 48 h to obtain homogeneous solutions. The homogeneous casting solutions were subsequently casted on porous polyester non-woven fabrics with the scraper clearance $250\text{ }\mu\text{m}$. After shortly (30 s) exposed to ambient air ($25 \pm 1\text{ }^\circ\text{C}$, $30 \pm 5\%$ relative humidity) to allow partial evaporation of solvents, these solution films together with the non-woven fabrics were submerged into the coagulation bath (deionized water) at room temperature for forming the porous membranes.

2.3. Membrane characterization

2.3.1. Membrane morphology and functional groups

The surface and cross-section morphologies were observed by scanning electron microscope (SEM) (Model XL-30, Philips, Netherlands). Pore sizes of membranes were calculated by the Image-pro plus 6.0 software (Media Cybernetics, USA). The membranes

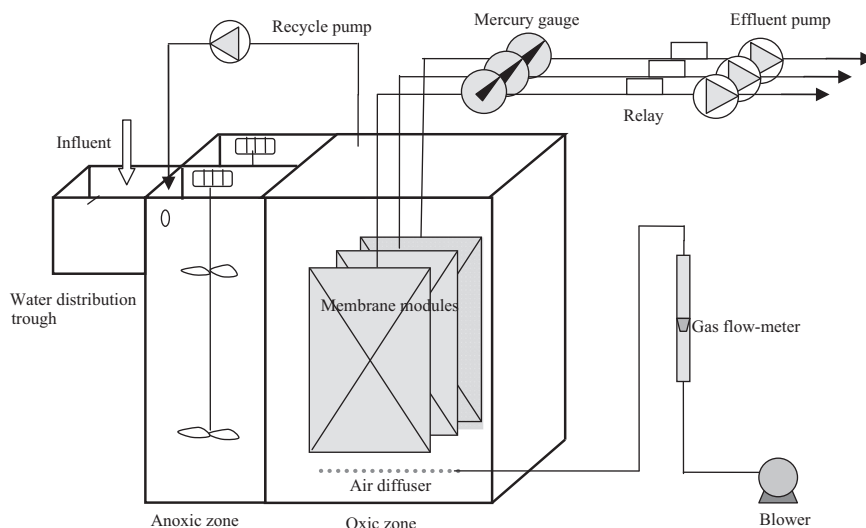


Fig. 1. Diagram of the submerged A/O-MBR.

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