



Effects of membrane compositions and operating conditions on the filtration and backwashing performance of the activated carbon polymer composite membranes



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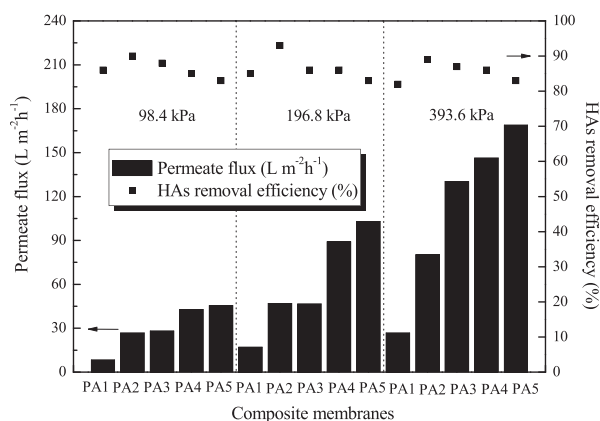
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HIGHLIGHTS

- We prepared the AC composite membranes with different composition ratios.
- Their filtration and backwashing performances were tested and studied.
- The AC composite membrane had great PWF, PF, FE, and FRR.
- The addition of AC improved the performance of composite membranes.

GRAPHICAL ABSTRACT



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ABSTRACT

This study prepares different activated carbon (AC) polymer composite membranes and investigates their operation characteristics of filtration and backwashing. Experimental results show that the addition of AC particles and hydrophilic polyethylene glycol (PEG) in the polymer membranes can improve the pure water flux (PWF) and permeation flux (PF) during filtration, as well as increase the flux recovery rate (FRR) after backwashing. The optimum transmembrane flux, filtration pressure and time of AC composite membrane were $45 \text{ L m}^{-2} \text{ h}^{-1}$, 196.8 kPa and 60 min; and the optimum backwashing pressure and time were 393.6 kPa and 10 min, respectively. Under such conditions, the flux recovery rate of the AC composite membranes achieved 87.5%.

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1. Introduction

Membrane filtration techniques are widely applied in the treatments of drinking water, waste water, and water recycling nowadays.

Compared with the traditional water treatment processes, membrane technology is more attractive due to its high efficiency, flexibility, and stability in removing various pollutants simultaneously [1–3]. However, membrane fouling is still a major problem in the application of organic polymer membranes. Membrane fouling is caused by the clogging of membrane pores and the accumulation of pollutants on the membrane surfaces, which increases the filtration resistances, declines the

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filtration flux, and reduces the validity period of membranes [4–10]. Therefore, membranes need to be cleaned and backwashed periodically or renewed.

To overcome such problems, modification of membranes by various methods is prevailing. The development of a composite membrane has become an attractive issue in membrane technology. With the addition of different materials and compositions, the chemical and physical properties, the antifouling ability, the filtration efficiency, and the backwashing efficiency of composite membranes are improved [11–13]. Table 1 summarizes the results of relevant studies on the preparations, compositions, properties, and operating characteristics of different composite membranes. Many studies indicated that the composite membranes prepared by the phase inversion method can significantly improve the surface morphology and structures of polymer membranes [14–18]. The composite membranes were made by mixing different hydrophilic polymers (such as polyetherimide (PEI), polyvinylpyrrolidone (PVP), PEG, cellulose acetate phthalate (CAP), and polyamideimide (PAI)) and inorganic materials (such as titanium dioxide (TiO₂), aluminum oxide (Al₂O₃), and silica (SiO₂)) to modify the hydrophobic polymer membranes (such as polyvinylidene fluoride (PVDF), polysulfone (PSF), polyethylsulfone (PES), and polyethylene (PE)) [19–25]. The results indicated that the modified composite membranes had better hydrophilicity, surface roughness, porosity, permeation flux and separation efficiency.

Membrane fouling during filtration processes usually resulted from pollutant precipitations, electrostatic adsorption, and the bio-fouling [4,6,10,26,27]. To remove the reversible and irreversible fouling on the surface and structure of polymer membranes, it is necessary to backwash the membranes with different backwashing pressure, frequency, reagents, and cleaning methods to enhance the permeation flux and flux recovery rates (UF/MF) [1,4,28–30]. The backwashing frequency usually depends on the filtration resistances and pressures. When the filtration pressure increases, the fouling of organic polymer membranes becomes worse and the backwashing efficiency declines. Pure water flux and permeation flux of membranes are thus difficult to recover [31–34]. The membrane structure property, filtration efficiency, and different membrane fouling mechanisms can be characterized through the operation profiles of filtration and backwashing procedures at different pressures and conditions [6,7,35,36].

The preparation and application of activated carbon (AC) composite membranes are seldom investigated in related literatures. The preparation and characteristics of different AC composite membranes have been studied in our previous works [16]. The results illustrated that the addition of activated carbon (AC) particles and hydrophilic polyethylene glycol (PEG) in the composite membranes can improve the porous structures, hydrophilicity, permeation flux, and filtration efficiency. To explore the filtration performance and backwashing characteristics of AC composite membranes, this study prepares different AC composite membranes to perform the filtration and backwashing

experiments for three cycles. The effects of different filtration and backwashing conditions on the pure water flux (PWF), permeation flux (PF), and flux recovery rate (FRR) of the AC composite membranes are also discussed.

2. Experimental

2.1. Materials

This study prepares five AC composite membranes with different composition ratios and investigates their performance at different filtration and backwashing conditions. The major components of the composite membranes were polyphenylsulfone (PPSU, MW: 53,000–59,000 g/mol) and polyetherimide (PEI, MW: 529 g/mol). Polyethylene glycol (PEG, MW: 200 g/mol) was used as the pore-formation agent and coconut shell-based activated carbon (AC) (China Activated Carbon Industries Ltd., Taiwan) was the additive particle. The solvent was N-methyl-2-pyrrolidone (NMP).

The particle size distributions of AC particles had three peaks of particle sizes at 0.01–0.1 μm, 1–10 μm, and 10–50 μm. The specific surface area, pore volume, micro-, meso-, and macro-pore volumes (percentages), and the average pore size of the AC particle were 524 m² g⁻¹, 0.431 cm³ g⁻¹, 0.226 cm³ g⁻¹ (52%), 0.194 cm³ g⁻¹ (45%), 0.011 cm³ g⁻¹ (3%), and 3.29 nm, respectively [16].

The prepared AC composite membranes were tested by filtering the simulated water with low concentrations of humic acids (HAs). The simulated water for the composite membrane tests was prepared by dissolving 1.4 g of HA powder (Sigma-Aldrich) into 1 L of deionized water and was pre-filtered using a filter with a mean pore size of 2.5 μm to remove the larger HAs. The pH value of simulated water was controlled at 6.7 ± 0.1 by 0.1 M HCl or NaOH [37–39]. The HA concentration in the feed solutions and resultant permeate solutions were determined by a UV-Vis spectrometer (UV-Vis/DRS; Perkin Elmer Lambda 35) at the wavelength of 254 nm [40–42]. The HA concentration in the pre-filtered solution was 476 mg/L.

2.2. Membrane preparation

All the AC polymer composite membranes were fabricated using the wet phase inversion method. The composition ratios of five different AC composite membranes are shown in Table 2. The major components of the membranes were 35 wt.% PPSU and 5 wt.% PEI polymers dissolved in N-methyl-2-pyrrolidone (NMP) solvent. The contents of the AC particles and pore-formation agent PEG were 0.1, 0.25 wt.% and 6 wt.%, respectively. The casting solutions containing PPSU, PEI, AC particles, PEG, and NMP were stirred at 500 rpm and heated at 60 °C for 2 h. Subsequently, the casting solution was carefully and uniformly spread on a glass plate using a casting knife. The solvent that remained in the casted membrane was evaporated at room temperature overnight. The casted

Table 1
Comparisons of the relevant studies on the compositions, properties, and operating conditions of different composite membranes. ^aPES: polyethersulfone; ^bDMMSA: hydrophilic *N,N*-dimethyl-*N*-methacryloxyethyl-*N*-(3-sulfopropyl); ^cBMA: *n*-butyl methacrylate; ^dMPC: methacryloyloxyethyl phosphorylcholine; ^eBMA: butyl methacrylate; ^fPVDF: polyvinylidene fluoride; ^gSPES: sulfonated polyethersulfone; ^hPVP: polyvinylpyrrolidone; ⁱTiO₂: titanium dioxide; ^jPSF: polysulfone; ^kPAI: polyamideimide.

Membrane materials	Compositions (%)	Preparation method	Contact angle (°)	Flow rate pressure	Flow rate time (min)	Backwash pressure	Backwash time (min)	Result	Refs.
PSf/MWNTs	15/4	Phase inversion	56	1–4 bar	120	–	–	PVP 55000 rejection = 63%	[19]
PES/PVA/PVP/TiO ₂	16/1.5/2/0.1	Phase inversion	43.2	5 bar	30 min	–	–	NaCl rejection = 41%	[20]
PVDF + SPES/PVP/TiO ₂	16/4/0.5	Phase inversion	72.2	1 bar	120	1 bar	10	BSA rejection = 79% FRR = 76.4%	[21]
PSf/PAI/UV–TiO ₂	69/30/1	Phase inversion	61.0	1725 kPa	120	1725 kPa	120	HA rejection = 86% FRR = 79.5%	[22]
PES/PEG/MPC–BMA	5.38/4.49/0.47	Phase inversion	35.0	100 kPa	30	150 kPa	20	BSA rejection = 59% RFR = 91%	[23]
PES/PEG /DMMSA–BMA	5.38/4.49/0.47	Phase inversion	49.0	0.1 MPa	60	0.1 MPa	60	BSA rejection = 95% RFR = 47.6%	[24]
PES/PVP/TiO ₂	15/5/0.3	Phase inversion	72.0	100 kPa	30	100 kPa	1	BSA rejection = 71%	[25]

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