



High permeation flux polypropylene/ethylene vinyl acetate co-blending membranes via thermally induced phase separation for vacuum membrane distillation desalination



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HIGHLIGHTS

- PP/EVA/soybean oil solutions can be a good compatible system via TIPS.
- The co-blending membrane exhibits excellent durability than neat PP membranes.
- A special cellular with thin dense cellular layer and thick loose cellular layer
- The co-blending membrane had lower resistance of mass transfer than neat PP.
- The higher flux of 27.6 kg/(m² h) during 36 h continuous operation was achieved.

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ABSTRACT

Isotactic polypropylene (iPP)/ethylene vinyl acetate (EVA) co-blending hydrophobic microporous membranes for vacuum membrane distillation (VMD) were prepared via thermally induced phase separation (TIPS), and the membranes were supported by a nonwoven polyester fabric with special cellular structures. These special cellular structures consist of two parts: a thin dense cellular layer and a thick loose cellular layer. These structures markedly decreased the resistance of mass transfer and improved flux performance during VMD. The co-blending membranes used for VMD led to superior long-term stability and excellent durability relative to neat PP membranes. This result can be attributed to the addition of EVA that significantly reduced membrane crystallinity. The thickness of the dense cellular layer and voids of the loose cellular layer showed a minimum and a maximum at a PP/EVA co-blending ratio of 6:1, respectively. Aqueous NaCl solutions (0.5 mol/L) were used in the VMD experiment at a feed temperature of 70 °C and a permeate side absolute pressure of 3 kPa. The flux of the iPP/EVA co-blending membranes was 27.6 kg/(m² h) within 36 h of continuous operation, whereas that of the iPP membranes was only approximately 16 kg/(m² h) within 6 h of continuous operation under the same condition.

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1. Introduction

Given its high separation efficiency, mild operating conditions, and low energy consumption, vacuum membrane distillation (VMD) has demonstrated promising results in seawater desalination, removal of heavy metal and volatile organic compounds, concentration of aqueous solutions, wastewater treatment and various separation processes [1–

4]. Compared with other MD configurations, VMD can reach higher fluxes and plant productivity for higher partial pressure gradients. Membrane materials are the key determinants of separation performance and water productivity for the future commercialization of MD [5]. Some studies developed new materials or enhanced the performance of conventional materials suitable for MD by either modifying hydrophilic membranes into hydrophobic membranes or using hydrophobic polymers [6]. Khayet [7] prepared hydrophobic polysulfone and polyethersulfone membranes with fluorinated surface-modifying macromolecules to treat radioactive wastewater solutions. However, the surface-modified membranes exhibit an extremely high liquid entry pressure (LEP) that restricts membrane wetting during MD in

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liquid radioactive waste processing. Wei et al. [8] obtained hollow fiber hydrophobic membranes from hydrophilic polyethersulfone through CF_4 plasma modification; the membranes showed a stable performance for 54 h in MD using 4 wt% aqueous NaCl at a feed temperature of 60.5 ± 0.2 °C. Popular intrinsic hydrophobic polymers used in MD membranes include polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polypropylene (PP) and polyethylene (PE) [9]. Among these materials, PTFE is mostly used in commercial and pilot MD modules because of its high hydrophobicity and excellent resistance toward harsh operation conditions [10,11]. However, the applications of PTFE are limited by its high cost and difficulties in module sealing. PVDF presents favorable hydrophobicity, mechanical strength and can be prepared using a wide array of methods, such as phase inversion immersion precipitation [12], thermally induced phase separation (TIPS) [13] and electro-spinning [14]. Considerable efforts have been exerted to enhance the porosity, LEP [15,16], Superhydrophobic [12, 17] and other properties of PVDF in VMD.

PP can be a semicrystalline polymer with excellent solvent resistance at room temperature, and porous PP membranes can be fabricated by stretching or TIPS. In TIPS, the polymer and the diluent are heated to a sufficiently high temperature to form a homogenous polymer solution. The selection of diluents, such as small-molecule nonsolvents, inorganic salts, or macromolecules and their combinations, influences membrane formation and separation performance. In our previous work [18], we prepared PP flat sheet membranes via thermally induced phase separation and found that soybean oil is a suitable diluent to control pore size and pore size distribution. However, neat PP cannot present long-term stability because of its weak durability. Neat PP membranes exhibit cracking and then leakage when applied in VMD. Polymer blending is an effective and economical method widely used to improve membrane performance in VMD. Simone et al. [6] obtained PVDF/PVP hollow fiber membranes with good structure, excellent mechanical properties and high porosity for VMD by using distilled water as feed. Chen et al. [15] fabricated composite membranes from a blend of high and low molecular weight PVDF membranes and found the highest flux of VMD with the thickest finger-like structure and the thinnest sponge-like structure. Dong et al. [17] prepared a superhydrophobic PVDF-PTFE nanofibrous membrane and tested its desalination for 15 h by VMD, yielding a stable flux of $18.5 \text{ kg}/(\text{m}^2 \text{ h})$ and salt rejection that exceeds 99.9%.

Blends of ethylene-propylene rubber, ethylene propylene diene monomer, polyethylene oxide, ethylene vinyl acetate (EVA) copolymer and styrene-ethylene-butylene-styrene have been prepared to improve PP toughness [19,20]. The commercially available hydrophobic polymer PP was used in the current study because of its strong solvent resistance and favorable mechanical strength [21]. PP also exhibits favorable performance in MD [22–28]. EVA was selected in the present study not only because of its better compatibility and lower cost than other elastomers or rubber but also because of its intrinsic hydrophobicity and low thermal conductivity. EVA shows good flexibility, impact resistance, nontoxicity and odorless feature [20]. Zhang et al. [29] assessed membrane thermal conductivity on the basis of membrane porosity and the thermal conductivity of polymer and gas (usually air).

In this work, PP/EVA co-blending hydrophobic microporous membranes were prepared via TIPS, and the membranes were supported by a nonwoven polyester fabric with special cellular structures. These special membrane structures presented lower resistance of mass

transfer than other general microporous membrane structure for EVA addition, which can provide long-time stability and higher flux to VMD. Subsequently, these PP/EVA co-blending membranes were tested in VMD.

2. Experimental

2.1. Materials

Isotactic PP (iPP) was supplied by Huabei Petrochemical Co. Ltd., China. The physical properties of iPP are listed in Table 1. EVA grade Elvax® 3120, Elvax® 460, and Elvax® 40 W with respective vinyl acetate (VA) contents of 8, 18 and 40 wt% were purchased from DuPont China Group Co., Ltd. The physical properties of EVA are listed in Table 2. Soybean oil-grade Fortune was purchased from China National Cereals, Oils, and Foodstuffs Corporation. The physical properties of soybean oil are shown in Table 3. Adipic acid (reagent purity), hexane (reagent purity), and NaCl (reagent purity) were purchased from Tianjin Chemical Reagent Co. Inc. All chemicals were used without further purification.

2.2. Preparation of PP/EVA co-blending membranes

PP/EVA flat hydrophobic microporous membranes were prepared via TIPS. Polymer particles were dried at 80 °C. A iPP/EVA solution (20–35 wt%) mixed with proper amounts of soybean oil (diluent) and adipic acid (nucleating agent) was stirred at 180 °C in a 500 mL reaction flask. Inert gas (nitrogen) was continuously introduced to avoid oxidation during melt-blending. Polymer particles were completely melted in soybean oil after 4 h, and bubbles simultaneously appeared. A homogeneous iPP/EVA-soybean oil phase formed after vacuum defoamation.

The polymer solution was cast on a nonwoven polyester fabric support material with a filler gauge under a preheated steel plate to achieve a thickness of 400 μm . Then, the cast film, together with the support material, was immersed into a coagulation bath (tap water at 20 °C) to solidify the sample. Then, the wet membranes were dried at room temperature for 24 h after the soybean oil was extracted using hexane. The compositions and codes of the samples are presented in Table 4.

2.3. Characterization of PP/EVA co-blending membranes

2.3.1. Scanning electron microscopy (SEM)

The morphology of the top surface and cross-section of the prepared flat membranes was observed via SEM (Hitachi SU1510, Japan) with an accelerating voltage of 40 kV. Membrane samples were fractured in liquid nitrogen to obtain the cross-section. All samples were coated with platinum by using a sputter coater (Hitachi E1020, Japan).

2.3.2. Fourier transform infrared spectroscopy (FTIR)

FTIR spectra were recorded on a Nicolet 6700 FTIR instrument from Thermo Electron Corp. within a transmittance range of $500\text{--}4000 \text{ cm}^{-1}$ at room temperature. The measurement was based on the absorption of infrared light at certain frequencies that correspond to the vibration modes of atomic groups present within the molecule.

2.3.3. X-ray diffraction (XRD)

The XRD spectra of the PP, EVA, and PP/EVA membranes were obtained at room temperature by using a fully automatic polycrystalline XRD (XD-3, Beijing Purkinje General Instrument Co., Ltd.). The

Table 1
The physical properties of iPP.

Parameters	Melt index (g/10 min)	Density (g/cm^{-3})	Melting point (°C)	Impurity (ppm)	Average molecular weight	Degree of crystallinity
Properties	11.20	0.946	165	≤ 167	8.6×10^5	56%

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