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Comparison of fouling behaviors of hydrophobic microporous membranes in pressure- and temperature-driven separation processes

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ABSTRACT

Hydrophobic hollow fiber membranes were systematically investigated in two different operation modes including microfiltration (MF) and membrane distillation (MD). Using synthetic feed waters containing model foulants, wastewater secondary effluent, and seawater, the differences in fouling behaviors and its reversibility for the membranes between MF and MD operation modes were compared. Our results demonstrate that fouling patterns in MF were completely different from those in MD. The rates of flux decline were higher in MF than in MD in most cases. No flux declines were observed by model foulants such as alginate, humic acid, and kaolin in MD although thick foulant layers were observed by SEM analysis. This can be attributed to the difference in water transport mechanisms between MF and MD. It was found that the flux decline due to the foulants in the wastewater was more reversible in MD than in MF and could be recovered not only by chemical cleaning but also by physical cleaning.

1. Introduction

Hollow fiber membranes have been extensively used for various microfiltration (MF) applications, including water and wastewater treatment [1–4]. As seawater desalination has become an important source of fresh water [5,6], the hollow fiber membranes are increasingly applied for the pretreatment of feed water to seawater reverse osmosis (SWRO) [1]. Recently, the hollow fiber membranes are also considered for use in membrane distillation (MD) systems [7–10], which have the capability of removing ions and other impurities from seawater or reclaimed wastewater.

Membrane fouling is a common problem in most membrane-based systems. When hollow fiber membranes are used in MF operation modes, it has been reported that serious fouling occurs [11,12]. This is generally caused by the deposition of particles and colloids, adsorption of organics matters, and pore blocking [11]. On the other hand, there is relatively little information on the fouling of hollow fiber membranes in MD operation modes [13,14]. If MD is applied for the treatment of wastewater [15], it may suffer from fouling. Fouling of MD membranes may occur together with wetting [16,17], which is a process that liquid water penetrates into the pores of the membrane. The efficiency of membrane cleaning and the reversibility of fouling in MD processes have been considered [18] but still need to be further investigated.

Recently, the fouling propensities and wetting properties for hollow fiber MD membranes were investigated in our previous works [19,20]. Nevertheless, a more comprehensive understanding of MD fouling is required for its widespread application. One of the interesting points is that hydrophobic hollow fiber membranes are used in both MF and MD processes. MF is a pressure-driven membrane process while MD is a thermally-driven membrane process. Although their separation mechanisms and capabilities are different, the same membranes may be used to treat the same feed waters such as seawater or reclaimed wastewater. However, little information is available regarding the difference in fouling propensities between MF and MD processes that use the same membranes. Accordingly, the purpose of this study was to examine the fouling mechanisms for hollow fiber membrane in MF and MD operation modes. To the best of our knowledge, this is the first work that systematically compares pressure-driven and temperature-driven separation processes using the same membranes for various feed solutions.

2. Materials and methods

2.1. Seawater and wastewater

Two types of waters were used for the experiments including seawater and wastewater. The seawater was collected from the west coast of Korea. It was filtered through Whatman GF/C glass fiber filters (Sigma-Aldrich). The wastewater, which was the effluent from the primary treatment, was collected from a Korean sewage treatment

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Table 1
Water quality parameters.

Category	pH	COD (mg/L)	SS (mg/L)	T-N (mg/L)	T-P (mg/L)	Conductivity (mS/cm)	Turbidity (NTU)
Seawater	8.15	1.55	8.3	< 1.0	< 0.1	48.2	< 0.1
Wastewater	7.32	46.0	111.1	30.3	3.2	0.71	32.3

plant. The compositions of the seawater and wastewater are summarized in Table 1. The water quality analysis was commissioned by a specialized agency in Korea (KEWWI, Korea Environment & Water Works Institute).

2.2. Synthetic feed waters

Three synthetic feed solutions were also used, which contain kaolin, alginate, and humic acid, respectively. Humic acid technical (Sigma-Aldrich), alginic acid sodium salt from brown algae (Sigma-Aldrich), and kaolin (Samchum Chemical, Republic of Korea) were used as received. All synthetic wastewaters were prepared by dissolving 100 mg/L of the standards in ultrapure DI water and stirring overnight.

2.3. Membrane properties

The experimental tests were carried out using commercial capillary membranes made of polypropylene (average pore size of 0.2 μm; thickness of 0.4 mm; inner diameter of 1.8 mm) purchased from Membrana (Germany). The membrane module was prepared by inserting it into a PVC socket and sealing it with a urethane hardener. The membrane area was around 150 cm^2. The same membrane module was used for both the MF and MD process. Since the membrane is highly hydrophobic, it should be pretreated to be used for MF. Accordingly, prior to MF tests, the membrane was immersed in a 50% ethanol solution for 30 min, and then filtered several times using a syringe for 5 min. In MD tests, the membrane was used without any pretreatment.

2.4. VMD process: operating conditions

The experiment was undertaken by submerging a vacuum-pressured membrane distillation module in the feed water. The volume of the feed tank was 5 L and the feed temperature was 80 °C. While the temperature of the chiller was 20 °C, the temperature difference with the feed tank was 60 °C. Stirring in the feed tank was set at 300 rpm. The initial flux was set to be 27 ± 3 kg/m^2-h. To obtain this flux, the vacuum pressure was adjusted in the range of 50 to 100 mbar using a vacuum pump (PC 3001 VARIO PRO, Germany). At the end of each test, the equipment was cleaned by flushing DI water for 30 min and running the VMD with DI water for 30 min at 60 °C. VMD tests with DI water were then carried out to check the membrane performance and to evaluate membrane fouling. A schematic diagram of the VMD device at lab scale is shown in Fig. 1.

2.5. MF process: operating conditions

In the case of the MF process, the experiment was conducted using the same device as that used for the VMD to proceed with the static pressure method. The initial flux was set to be 30 ± 3 L/m^2-h, which was similar to that of the MD tests. To obtain this flux, the vacuum pressure was adjusted in the range of 950 to 970 mbar using a vacuum pump. For this experiment, the unnecessary chiller was removed and the feed tank was maintained at room temperature (25 °C). Stirring in the feed tank was also set at 300 rpm.

2.6. Analytical methods

Conductivity was analyzed using a Multi 3420 (Wissenschaftlich-Technische Werkstätten GmbH, Germany) digital precision meter. Turbidity was analyzed using a handheld Turb 430 IR (Wissenschaftlich-Technische Werkstätten GmbH, Germany) turbidimeter. Water quality analysis was conducted immediately after the end of each experiment.

2.7. Liquid entry pressure

Liquid entry pressure (LEP) is a critical parameter for membrane distillation because it indicates the pressure at which liquid can enter the membrane pores [16]. According to the Young–Laplace model [17], membranes consisting of cylindrical pores have an LEP given by

$$\Delta P = \frac{-2\gamma}{r_t}\cos(\theta) \tag{1}$$

where ΔP is the LEP, γ is the surface tension of the liquid, r is the pore radius, and θ is the intrinsic advancing contact angle between the liquid and the membrane material. However, it is difficult to accurately calculate the LEP using this equation. The LEP in the membrane was, therefore, directly measured using a lap-scale device as shown Fig. 2. The indigo dye (Sigma-Aldrich) was put inside the module and pressure was applied. The pressure was maintained for at least 10 s and then pressurized. The pressure was measured when the dyes appeared on membrane surface.

2.8. Scanning electron microscopy

Membrane structures and surface analysis were investigated using a field emission electron microscope (JSM-7610F, JEOL Ltd., Japan). Prior to coating the membrane with platinum, membranes were completely dried at 50 °C for 2 h. The membranes were then coated with platinum by sputtering for 120 s. Scanning electron microscopy (SEM) can provide high-throughput and high-performance analysis using high-power optics. It is also suitable for high-spatial-resolution analysis.

3. Results and discussion

3.1. Water quality analysis

Table 2 compares the water quality parameters for treated waters before and after the experiment. The measurements were repeated three times for reproducibility and the mean value was used. As expected, the turbidity removal efficiencies were high in both MF and MD but the conductivity removal efficiencies were high only in MD.

Although MD is not generally used for particle separation, it can definitely reject particles, leading to formation of foulant cake layers on the MD membrane surface in these cases. Since the initial flux values were similar and the same membranes were used, the hydrodynamic and physicochemical conditions for the formation of foulant layer seem to be similar in MF and MD. The only difference is the ion rejection, which will increase the ion concentrations near the membrane surface in MD due to concentration polarization.

3.2. Changes in permeate flux

3.2.1. Changes in MF flux

The changes in flux with VCF in MF operation are shown in Fig. 3. The flux was presented as a form of normalized flux (J/J_0). The initial