



More fouling resistant modified PVDF ultrafiltration membranes for water treatment

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ABSTRACT

Polyvinylidene fluoride (PVDF) membranes were modified using L2MM, a new type of hydrophilic polyurethane additive, to improve membrane performance. Membranes were prepared using 15 and 18% PVDF and small weight percentages (i.e., 1.5 and 3%) of L2MM. Their fouling resistance was evaluated by filtration/fouling tests with Ottawa River Water (ORW) using membranes incorporating two different types of L2MMs (L2MM(PEG-200) and L2MM(PEG-600)). For comparisons a commercial membrane (HFM-180) was also tested. A PVDF membrane prepared with 3% L2MM(PEG-200) which produced the highest cumulative permeate volume, had a lower flux reduction and its TOC removal was 24% higher than the HFM-180 membrane. L2MM produced more hydrophilic PVDF membranes which had pure water permeation (PWP) rates up to 128% greater than the control PVDF membranes. Many of the L2MM blended membranes had significantly higher total organic carbon (TOC) removals than the HFM-180 membrane tested (64–67% versus 46%). Modeling of the flux decline suggested the membranes fouled via a combination of pore blocking and cake filtration; scanning electron microscopy (SEM) of the fouled membrane surface confirmed the existence of a cake layer.

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1. Introduction

Membrane fouling, which reduces the permeate production rate over time, is an important concern in the operation of membrane separation processes. According to Zularisam et al. [1] fouling by natural organic matter (NOM) is considered a major contributor to membrane fouling in water treatment applications, as it can result in irreversible fouling. The extent of the fouling depends on the interactions between fouling substance and membrane and between fouling substances themselves [1]. Several alternatives can reduce the undesired effect of NOM fouling on membrane performance such as: (1) selecting a membrane material that minimizes attractive interactions between NOM and the membrane surface; (2) pre-treating the feed to selectively remove the most adhesive NOM substances; and (3) reducing fouling through improved hydrodynamics within the membrane modules [2]. Membranes intended for water treatment applications require high chemical resistance and many of them are hydrophobic. However, hydrophobic membranes foul at a considerably greater rate than hydrophilic membranes with similar pore sizes and separation characteristics [3–5].

Our research group has developed tailor-made polymeric additives that have hydrophilic functional tails to modify polyethersulfone (PES) membranes and increase their flux [6,7]. These polyurethanes were called LSMM, short for hydrophilic surface

modifying macromolecules. Dang et al. [8] showed that LSMM was thermodynamically compatible with polymers such as cellulose acetate (CA), polyetherimide (PEI), polysulfone (PS), PES, and polyvinylidene fluoride (PVDF). They showed that the addition of 3% LSMM in the casting solution of CA, PES, and PVDF membranes increased the final flux during pure water permeation (PWP) tests (by up to 12, 65, and 150%, respectively) but their contact angles did not significantly change. Recently, our group has developed a similar (and potentially more effective) hydrophilic additive to modify the PVDF membranes [9–10]. They were called L2MM which stands for hydrophilic 2nd generation surface modifying macromolecules. The primary objective of this study is to evaluate the performance of L2MM-modified-PVDF ultrafiltration membranes for the treatment of a river water containing substantial concentrations of natural organic matter.

2. Experimental

2.1. Materials

A L2MM can be described in general terms as a polyurethane with hydroxyl functional end-groups, these end-groups arising from the polyethylene glycol (PEG) used in its preparation. This study used two L2MM additives: L2MM(PEG-200) and L2MM(PEG-600). They were prepared using PEG with molecular weights of 200 and 600, respectively. Their preparation is discussed in Pezeshk et al. [9] and Pezeshk [10]. Based on nuclear magnetic resonance (NMR) the average molecular weights of L2MM(PEG-200) and L2MM(PEG-600)

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Table 1
Casting dope composition of experimental membranes prepared.

Membrane code	PVDF (%)	Concentration of additive (%)	Solvent (%)
18% PVDF control	18	0	82
18% PVDF + 3% L2MM(PEG-200)	18	3	79
18% PVDF + 3% L2MM(PEG-600)	18	3	79
18% PVDF + 1.5% L2MM(PEG-200)	18	1.5	80.5
18% PVDF + 1.5% L2MM(PEG-600)	18	1.5	80.5
15% PVDF control	15	0	85
15% PVDF + 3% L2MM(PEG-200)	15	3	82

were 4000 and 13,000 Da, respectively. PVDF (MW ca. 254,000 g/mol), donated by Elf Autochem Inc. (Oakville, ON), was used as the membrane's base polymer; and 1-methyl-2-pyrrolidinon (NMP) was used as the solvent (Sigma Aldrich Inc., Milwaukee, WI).

Ottawa River Water (ORW) was used for the filtration/fouling tests because its relatively high hydrophobic NOM content accelerates fouling and makes it a good challenge water [11]. ORW was collected from the intake of the Britannia Water Treatment Plant, located in Ottawa, Ontario.

2.2. Membrane preparation and testing protocol

Flat sheet PVDF membranes were prepared by the phase inversion technique that included the following steps: preparation of the membrane casting solution, manual casting of the membranes, and solidification of the membrane films by phase inversion in a mixture of ice and water. The casting dope compositions utilized are shown in Table 1.

The casting was accomplished by pouring the casting solution on a clean glass plate and spreading it with a brass casting rod with a 250 μm gap to form a thin film. Then, the glass plate and film were immediately immersed in a 4 °C ice-water bath where the film hardened to form the membrane [12]. Thus, the resulting membrane did not have a fabric backing and had a nominal thickness of 250 μm . Each casting produces a 15 cm by 25 cm membrane sheet; multiple

sheets were cast and six coupons (7 cm in diameter) of each type of membrane were cut for testing.

Membrane contact angles were determined using a VCA Optima Surface Analysis System (AST Products Inc., Billerica, MA), which uses a digital camera to monitor the addition of a 1.5 μL water drop on the surface of the membrane. Prior to the measurements, membrane samples were naturally dried and cut into smaller pieces (15 \times 50 mm). Fifty contact angle measurements were performed in different locations of the small coupons.

The river water's color was measured using a spectrophotometer with a built-in color standards (Hach DR2800, Hach, Loveland, CO). A UV-Persulfate TOC analyzer (Phoenix 8000, Tekmar/Dohrmann, Mason, Ohio) was used to measure the TOC, which was used to quantify NOM in ORW. UV absorbance was measured using a UV-vis spectrophotometer (DU-40, Beckman Instruments Inc., Irvine, CA). Turbidity was measured with a turbidity meter (Hach 2100AN, Loveland, CO). Morphology of the fouled membranes was characterized using scanning electron microscopy, the microscope (SEM EVO-MA10, Zeiss, Germany) was equipped with a secondary electron detector (SE) and a back-scattering detector (BSE) (Oxford Instruments, Abingdon, Oxfordshire, UK). To assess the quality of the new membranes developed in this study, they were tested in parallel with a commercially available PVDF flat sheet PVDF membrane (HFM-180, Koch Inc.).

The membrane filtration experimental set-up (Fig. 1) consists of a 20 L reservoir, a high pressure diaphragm pump, six membrane cells, valves, pressure gauges, a flowmeter and a heat exchanger. The water is pumped from the reservoir, through the six membrane cells in series and back to the reservoir. The system is operated at a constant feed pressure and a constant flowrate of 1.1 L/min. A sketch of the cross-flow membrane test cells is shown in Mosqueda-Jimenez et al. [13]. The high velocity flow (0.2 m/s) in the narrow channel (average height = 1.18 mm) adjacent to the membrane insures turbulent conditions in the membrane cell, and minimizes the impact of concentration polarization. Mosqueda-Jimenez et al. [13] found that these cells performed very similarly to SEPA cells in the filtration of ORW. They also showed that the performance of the second through sixth cells was statistically the same as that of the first cell in series. This is likely

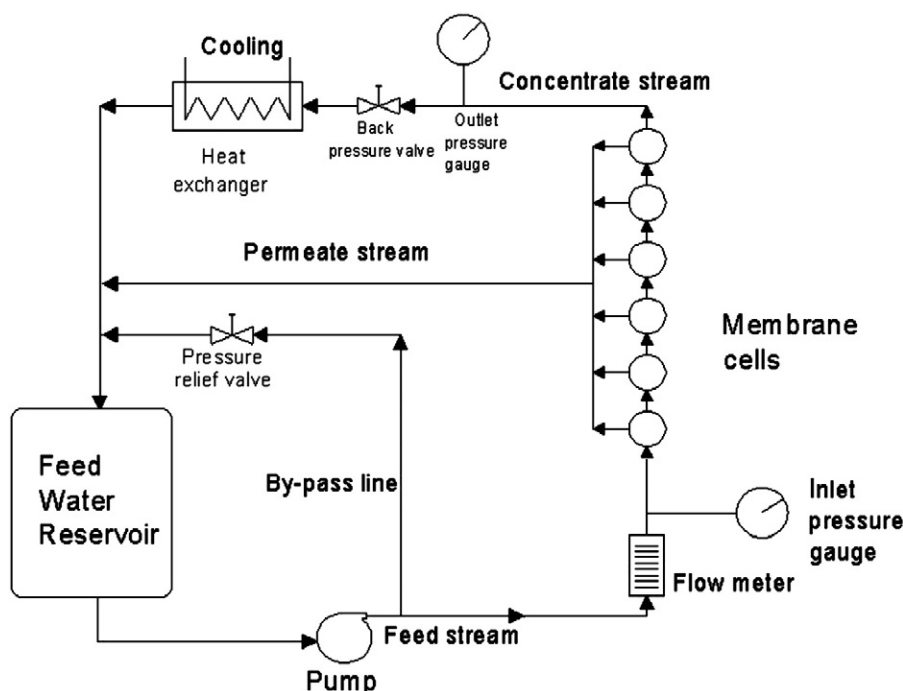


Fig. 1. Membrane experimental set-up (Mosqueda-Jimenez et al. [13]).

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