

An improved method for surface modification of porous water purification membranes



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ABSTRACT

The surfaces of polysulfone and polyethersulfone ultrafiltration membranes were coated with polydopamine, yielding hydrophilic membranes that, under constant transmembrane pressure fouling conditions, have previously shown enhanced flux relative to unmodified membranes. When evaluated under constant permeate flux fouling, however, modified membranes exhibited higher transmembrane pressures than their unmodified analogs. This increased transmembrane pressure in the coated membranes was ascribed to the decrease in membrane permeance resulting from applying the polydopamine coating. The membrane permeance could be tuned by varying polydopamine deposition time and, even at the shortest deposition times studied here, a few minutes, a substantial increase in membrane hydrophilicity could be achieved. Therefore, polydopamine was deposited on a membrane of relatively high permeance until the pure water permeance of the modified membrane matched that of a membrane having lower native permeance, permitting a comparison of the fouling performance of a modified and unmodified membrane with the same pure water permeance. This approach was repeated, using a single, high permeance membrane as the base membrane for modification, to produce a family of modified membranes having the same initial pure water permeances as lower permeance, unmodified membranes. When unmodified and modified membranes of the same initial permeance were compared at constant flux fouling conditions, the modified membranes consistently exhibited lower transmembrane pressures and similar organic rejections to the unmodified membranes. Because many porous water purification membranes are operated at constant flux in industrial settings, an interesting methodology for membrane surface modification may be to surface-modify a membrane of high permeance until the desired permeance is achieved, rather than by surface modification of a membrane that natively has the desired water transport characteristics, since the surface modification procedures almost invariably lead to lower pure water permeance.

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1. Introduction

Water purification membranes often suffer from a decrease in productivity during filtration due to the accumulation of rejected material on the membrane surface [1]. While membrane filtration often offers many advantages over other separation techniques, such as relatively low energy use, a small footprint, and low capital investment, fouling remains a significant problem in many

applications [1]. Fouling is commonly addressed by various cleaning methods, such as backpulsing, air sparging, or chemical cleaning [1,2]. To reduce the operational expenses incurred with such procedures, the development of membranes inherently less susceptible to fouling remains a major goal of the membrane community.

Surface modification is one approach that has been widely explored to improve the fouling resistance of water purification membranes [2–5], including microfiltration [6–9], ultrafiltration [10–12], and reverse osmosis membranes [13]. Typically, polymeric ultrafiltration (UF) and microfiltration (MF) membranes are produced by a phase inversion process, where a solution of polymer in organic solvent is cast as a film and subsequently immersed in a nonsolvent (typically water), causing the polymer to precipitate

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from solution, forming a porous membrane [1]. Hydrophobic polymers such as polysulfone, polyethersulfone, polypropylene, and polyvinylidene difluoride are popular choices for phase inversion membranes because they are relatively inexpensive, offer good mechanical properties, and are chemically stable in most feeds of interest [1]. However, the hydrophobic nature of membranes made from these polymers exacerbates their susceptibility to fouling, since hydrophobic contaminants, such as emulsified oils, readily form strong hydrophobic–hydrophobic interactions with the membrane surface [14]. Therefore, hydrophilic surface modifications, which frustrate adsorption of hydrophobic foulants to the membrane surface while maintaining many of the advantages of the base membrane material, are the subject of wide interest [3,15].

Recently, polydopamine (PDA) has been explored as a surface modification agent that hydrophilizes many substrates, including metals, ceramics, and a wide variety of polymers [16–21]. PDA is deposited on a substrate surface by contacting the surface with a buffered, aqueous solution of dopamine at mild pH at ambient conditions [16]. Several recent studies have focused on characterizing the detailed chemical structure of PDA, which, due to its insoluble nature, is difficult to unambiguously determine using routine analytical methods, and this topic remains an area of active investigation [22–25]. Polydopamine has already garnered interest as a membrane surface modification material [26–33]. A pilot study, using polydopamine-modified membranes to purify flow-back water from hydraulic fracturing, showed improved UF membrane fouling performance [29].

We have previously modified a number of different porous and nonporous water purification membranes with polydopamine, including microfiltration, ultrafiltration, nanofiltration, and reverse osmosis membranes [30–32]. The fouling resistance of the modified membranes was evaluated using constant transmembrane pressure crossflow filtration, where the membrane was challenged with a fouling feed at a fixed transmembrane pressure (TMP), and the flux decline was monitored as the membrane fouled. An example from one such study is shown in Fig. 1. Here, a polysulfone ultrafiltration membrane, having a nominal molecular weight

cutoff of 20 kDa, was modified with polydopamine and its fouling performance compared to that of an unmodified, but otherwise identical, membrane during filtration of an oily water emulsion. The polydopamine-modified membrane, presumably due to its enhanced hydrophilicity, showed improved long-term flux relative to that of the unmodified membrane [31]. Similar results were obtained with several other microfiltration, ultrafiltration, nanofiltration, and reverse osmosis membranes [31,32].

While many laboratory fouling studies are performed using constant transmembrane pressure crossflow filtration experiments, most full scale ultrafiltration and microfiltration facilities operate at constant permeate flux with variable transmembrane pressure [34,35]. Few reports of constant flux fouling with surface-modified membranes exist in the literature [36]. In this paper, we describe the effect of PDA surface modification of polysulfone (PS) and polyethersulfone (PE) UF membranes on their fouling behavior under constant permeate flux conditions. As described elsewhere [30], the thickness of the PDA coating layer may be varied by adjusting the contact time between the dopamine solution and the membrane surface. As a result, the membrane permeance may be easily tuned, providing a mechanism for control of the water transport characteristics of the membrane while enhancing its resistance to fouling.

2. Materials and methods

2.1. Materials

Rolls of PS-20, PS-10, PE-20, PE-10, PE-5, and PE-2 UF membranes were purchased from Sepro Membranes, Inc. (Oceanside, CA). The rolls were stored in a cardboard box away from direct light exposure. PS-20 and PS-10 membranes were both polysulfone membranes, but PS-20 had a nominal molecular weight cutoff of 20 kDa, and PS-10 had a nominal molecular weight cutoff of 10 kDa. Similarly, PE-20, PE-10, PE-5, and PE-2 membranes were all polyethersulfone membranes with nominal molecular weight cutoffs of 20 kDa, 10 kDa, 6 kDa, and 4 kDa, respectively. Acrylic plastic plates, on which the membranes were mounted during surface modification, were 20 cm × 28 cm × 0.6 cm and obtained from Interstate Plastics (Austin, TX). Frames were made from some of the acrylic plates by cutting a 15 cm × 23 cm rectangle from their centers. Rubber gaskets were obtained from Advanced Gasket & Supply (Ft. Worth, TX) and had the same dimensions as the fabricated acrylic plastic frames.

Ethanol, dopamine hydrochloride, and Trizma HCl were purchased from Sigma Aldrich (St. Louis, MO); sodium hydroxide was obtained from Fisher Scientific (Pittsburgh, PA). Soybean oil (Wesson) was obtained from a local supermarket and Xiameter OFX-0193 non-ionic, silicone-based surfactant was purchased from Dow Corning (Midland, MI). *n*-Decane was purchased from Alfa Aesar (Ward Hill, MA). All reagents were used without further purification. A Millipore Milli-Q Advantage A10 water purification system (18.2 MΩ-cm at 25 °C, 1.2 ppb TOC) (Billerica, MA) was used to produce ultrapure water. TRIS buffer (15 mM) was prepared by dissolving Trizma HCl (2.634 g) in ultrapure water (1 L), then using sodium hydroxide to adjust the pH to 8.8.

2.2. Membrane pretreatment and modification

Sheets (20 cm × 28 cm) were cut from each of the membrane rolls. To wet the porous structure of the membrane, each sheet was placed, selective (i.e., feed) side down, into a shallow dish of ethanol so that ethanol wicked into the pores, permitting entrapped air to escape through the backing material. The sheets sank into the ethanol once the air inside the membrane was displaced, and

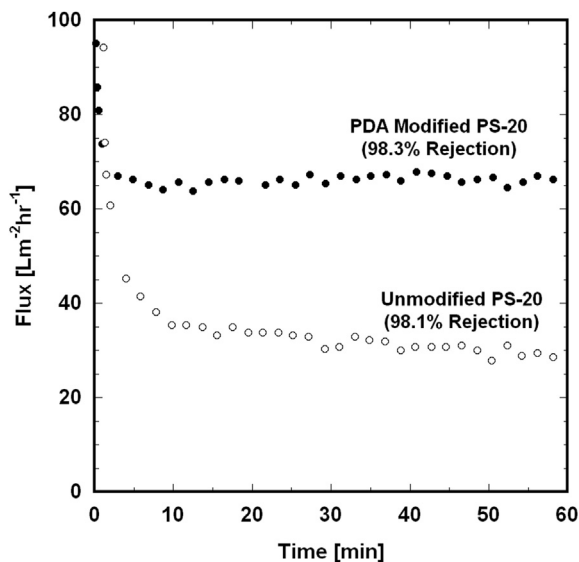


Fig. 1. Constant transmembrane pressure fouling of unmodified and PDA-modified PS-20 ultrafiltration membranes. PDA modification conditions: 2 mg/mL dopamine in 15 mM TRIS buffer, pH 8.8, 60 min. TMP: 30 psig, crossflow velocity: 0.18 m/s, foulant: 1500 ppm soybean oil emulsion prepared with non-ionic, silicone-based surfactant (9:1 oil:surfactant). PS-20 polysulfone UF membrane from Sepro Membranes, Inc. (Oceanside, CA). Reproduced with permission from Ref. [31].

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