



Improving the organic and biological fouling resistance and removal of pharmaceutical and personal care products through nanofiltration by using in situ radical graft polymerization

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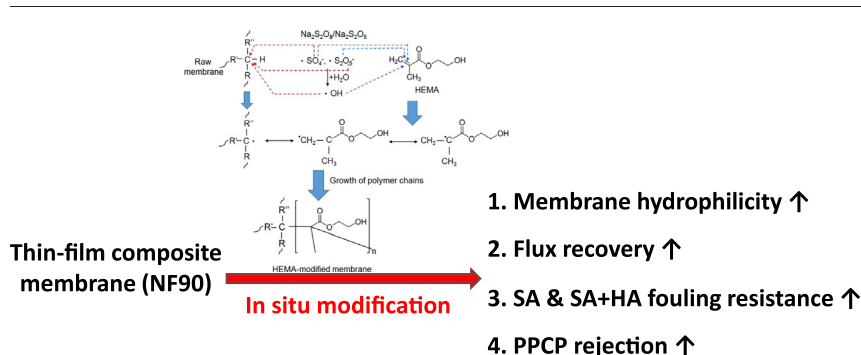
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HIGHLIGHTS

- Both fouling situations were effectively mitigated with enhance flux recovery of the in situ modified membrane.
- PPCP rejection was significantly improved by modified membrane before and after fouling.
- Both SPM and HEMA can increase the hydrophilicity of membrane surface by decreasing the contact angle.
- Triclosan adsorption on the modified NF90 after fouling increased significantly without much penetration across membrane.
- The fouling mechanism changed to intermediate blocking and complete blocking after membrane modification.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, an in situ radical graft polymerization technique using monomers of 3-sulfopropyl methacrylate potassium salt (SPM) and 2-hydroxyethyl methacrylate (HEMA) was applied to a commercial nanofiltration membrane (NF90) to improve its removal of six commonly detected pharmaceutical and personal care products (PPCPs) and mitigate organic and biological fouling by humic acid (HA) and sodium alginate (SA). Compared with the virgin membrane, the modified NF90 membrane exhibited considerably improved fouling resistance and an increased reversible fouling percentage, especially for SA + HA composite fouling. Moreover, the PPCP removal of the modified NF90 membrane was higher than that of the virgin membrane after SA and SA + HA fouling, respectively. Triclosan and carbamazepine, which are poorly rejected, could be effectively removed by modified membrane after SA or SA + HA fouling. Both monomers modified the membrane surface by increasing the hydrophilicity and decreasing the contact angle. The degree of grafting was quantified using attenuated total reflection Fourier-transform infrared spectroscopy. The mitigation in the fouling was evident from the low quantity of deposit formed on the modified membrane, as observed using scanning electron microscopy. A considerable amount of highly hydrophobic triclosan was adsorbed on the SA-fouled virgin membrane and penetrated through it. By contrast, the adsorption of triclosan was substantially lower in the SPM-modified membrane. After membrane modification, the fouling mechanism changed from solely intermediate blocking to both intermediate blocking and complete blocking after membrane modification. Thus, the in situ radical graft

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polymerization method effectively reduces organic and biological fouling and provides high PPCP removal, which is beneficial for fouling control and produces permeate of satisfactory quality for application in the field of membrane technology.

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1. Introduction

Membrane-based processes have been applied worldwide for water purification, reclamation, and desalination to solve the challenge of water scarcity (Tang et al., 2011). Nanofiltration (NF) is of considerable interest because it requires a much lower driving pressure compared with reverse osmosis (RO), results in high permeate flux and satisfactorily removes various emerging contaminants (Kimura et al., 2009; Kochkodan and Hilal, 2015; Lin et al., 2014; Michael et al., 2013; Mohammad et al., 2015; Nghiem and Coleman, 2008). However, the utilization of membrane technologies has been delayed because of membrane fouling (Kimura et al., 2009; Kochkodan and Hilal, 2015; Mohammad et al., 2015), deteriorating permeate quality and flux, increasing frequency of flushing and chemical cleaning, and decreasing membrane lifespan (Lin et al., 2006; Tang et al., 2011).

Eliminating membrane fouling entirely is impossible in practical application due to the ubiquity of organic and biological foulants in the aqueous environment. However, fouling can be mitigated by using membrane surface modification techniques, such as interfacial polymerization (Gol and Jewrajka, 2014; Kochkodan and Hilal, 2015; Soyekwo et al., 2017), surface grafting (Ben-David et al., 2010; Bernstein et al., 2012; Kochkodan and Hilal, 2015; Saeki et al., 2014; Tomer et al., 2009), surface coating (Kochkodan and Hilal, 2015; Lin et al., 2012), and incorporation of nanoparticles in composite membranes (Kochkodan and Hilal, 2015; Lee et al., 2013; Safarpour et al., 2015; Safarpour et al., 2016). Among these techniques, radical graft polymerization is simple, has versatile operation, and can be reproduced at relatively low cost (Ben-David et al., 2010; Bernstein et al., 2012; Mohammad et al., 2015). Various types of monomers and radical generation techniques can be employed, such as photo/chemical activation, irradiation, and plasma treatment (Ben-David et al., 2010; Kim et al., 2008; Tomer et al., 2009; Ulbricht and Belfort, 1996). Unlike some studies modifying membrane surface using physical methods (e.g., surface coating and incorporation of nanoparticles in composite membranes) that leach-out problem of coating compounds during long term operation can occur and compromise the performance of membrane operation, membrane modification using the chemical method—radical graft polymerization—can modify the functional groups on the membrane surface permanently. With proper operating procedure (e.g., no long term exposure to strong acid, base and oxidant), the modified membranes exhibit higher hydrophilicity with low biological fouling and increased membrane selectivity (Ben-David et al., 2010; Bernstein et al., 2011; Bernstein et al., 2012).

Numerous studies have focused on the surface modification of thin-film composite (TFC) RO membranes. However, fouling-resistant TFC NF membranes with high efficiency in removing pharmaceuticals and personal care products (PPCPs) removal have not yet been investigated in detail. Long-term exposure to trace PPCPs may cause toxicological or abnormal physiological effects in non-target organisms, such as humans, animals, and even plants (Fair et al., 2009; Herklotz et al., 2010; Kim and Tanaka, 2009; Kostich et al., 2014; Mukherji et al., 2011; Schwartz et al., 2003). Therefore, NF membranes have attracted considerable attention for mitigating biological and organic fouling and improving PPCP removal for water reclamation.

In this study, the modification of a commercialized NF membrane was systematically investigated by using the *in situ* radical graft polymerization technique. Very low bulk concentrations of monomers and initiators were required because of the rejection of solutes by the

membrane surface. This caused concentration-polarization and considerably increased the concentrations of monomers and initiators on the membrane surface, which was economical and beneficial for avoiding homopolymerization in the bulk solution (Bernstein et al., 2011; Bernstein et al., 2012). The surface characteristics, biological and organic composite fouling mitigation, and PPCP rejection efficiency of the modified membranes were studied. Moreover, the foulants along with the membrane coupons were extracted and analyzed for PPCP adsorption to elucidate the contribution of adsorption to PPCP removal.

2. Materials and methods

2.1. Membrane and chemicals

The modified NF membrane employed (NF90) was a commercialized semiaromatic piperazine-based polyamide (PA) TFC membrane with polysulfone (PSf) and polyester (PET) as the support layers (Dow FilmTec, USA). The permeability of pure water is 17.8 L/m² h bar, and the NaCl and MgSO₄ rejections are 40% and 85%–95%, respectively, according to the manufacturer. The physicochemical characteristics of NF90 have been well studied. NF90 has a molecular weight cut-off (MWCO) of 200 Da (Arsuaga et al., 2010; Lin, 2017; Mohammad et al., 2015), average pore radius of 0.34 nm (Lin et al., 2007; Nghiem and Coleman, 2008), contact angle of 59.4° (Nghiem and Coleman, 2008; Radjenovic et al., 2008), root mean square roughness of 142.8 ± 9.6 nm (Nghiem and Coleman, 2008), zeta potential of −27 mV (at pH 8) (Nghiem and Coleman, 2008), and isoelectric point of pH 4.0 (Nghiem and Coleman, 2008).

All the reagents used were of at least analytical grade and used without further purification. NaCl, NaHCO₃, KH₂PO₄, the membrane modification monomers [3-sulfopropyl methacrylate potassium salt (SPM) and 2-hydroxyethyl methacrylate (HEMA)] and the initiators [potassium persulfate (K₂S₂O₈) and potassium metabisulfite (K₂S₂O₅)] were purchased from Sigma Aldrich (USA). High purity (>99%) PPCP chemicals were purchased from Sigma Aldrich (USA). Their physicochemical properties are summarized in Table 1. These PPCPs were selected because of their high frequency of detection in Taiwan, small molecular weight (< 300 Da, effective removal is difficult in traditional water treatment plants), and different ionization (pK_a) and hydrophobicity (log K_{ow}), as indicated in Table 1. High-performance liquid chromatograph (HPLC)-grade acetonitrile and methanol were obtained from J.T. Baker (USA). Sodium alginate (SA) and humic acid (HA) were used to simulate biological and organic fouling, respectively. SA and HA were purchased from Sigma Aldrich (USA).

2.2. *In situ* membrane modification

Three parallel cross-flow cells were loaded with fresh flat-sheet membrane coupons and operated in the recycling mode with the cross-flow velocity, transmembrane pressure, and temperature maintained at 0.1 m/s, 690 kPa, and 25 °C ± 0.5 °C, respectively. The cross-flow cells were operated for at least 12 h, and distilled (DI) water was used to reach a steady-state permeate flux (J_{ps}). Subsequently, the feed was replaced with a monomer (0.005–0.05 M SPM or 0.01–0.02 M HEMA) and the initiators (0.01 M K₂S₂O₈ and 0.01 M K₂S₂O₅) in *in situ* membrane modification lasting 1 h (the optimal reaction time according to our preliminary experimental results) under the previously described filtration temperature, pressure and cross-

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