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Cleaning protocol for a FO membrane fouled in wastewater reuse

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ABSTRACT

Forward osmosis (FO) is an emerging technology which can be applied in water reuse applications. Osmosis is a natural process that involves less energy consumption than reverse osmosis (RO), and therefore can be applied as a dilution process before low-pressure RO; it is expected to compete favourably against current advanced water reuse technologies that use microfiltration/ultrafiltration and RO. The focus of this research was to assess the efficiency of different cleaning procedures to remove fouling from the surface of a FO membrane during the operation of a submerged system working in FO-mode (active layer (AL) facing feed solution) intended for secondary wastewater effluent (SWWE) recovery, using seawater as draw solution (DS), which will be diluted and can further be fed to a low-pressure RO unit to produce fresh water. Natural organic matter (NOM) fouling was expected to affect the AL, while for the support layer (SL), transparent exopolymer particles (TEP) were used as indicators of fouling due to their stickiness and propensity to enhance the attachment of other foulants in seawater on the membrane surface. The composition of the NOM fouling layer was determined after proper characterisation with a liquid chromatograph coupled with organic carbon detection (LC-OCD), showing biopolymers and protein-like substances as the main constituents. NOM fouling showed high hydraulic reversibility after a 25% flux decline was observed, up to 89.5% when in situ air scouring for 15 min was used as a cleaning technique. Chemical cleaning with a mixture of Alconox, an industrial detergent containing phosphates, and sodium EDTA showed to increase the reversibility (93.6%). Osmotic backwash using a 4% NaCl solution and DI water proved to be ineffective to recover flux due to the salt diffusion phenomena occurring at the AL. Part of the flux that could not be recovered is attributable to TEP fouling on the SL, which forms clusters clearly identifiable with an optical microscope and TEP-specific dyeing/staining of the membrane. Two solutions were tested to clean in place of the SL: a 1% NaOCl solution and the same detergent solution used to clean the AL. The former agent compromised the integrity of the FO membrane, as proved with flux and conductivity measurements; 94.5% of flux was recovered with the latter agent, showing that chemically irreversible fouling for the FO membrane

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is on the order of 5.5%, which might be associated with the adsorption of biopolymers on the AL and some TEP residuals on the SL. Physical cleaning (air scouring) of the AL proved to be the most effective way to control fouling.

Keywords: Forward osmosis; Fouling; Cleaning; Natural organic matter; Transparent exopolymer particles

1. Introduction

Membrane processes are now commonly used in water reuse and seawater desalination. Forward osmosis (FO) represents a new opportunity to solve the global water crisis because of its low-energy requirement compared to high-energy processes, such as reverse osmosis (RO) or nanofiltration (NF). Previous studies investigated fouling in FO membranes [1,2], showing that for low-pressure osmotic processes, fouling is highly reversible. Previous work by Zhang et al. [3] has determined that extracellular substances (EPS) are the first step of fouling, comprising polysaccharides and proteins mainly, and are considered to be precursors of transparent exopolymer particles (TEP). TEP fouling cannot be controlled by feed water disinfection or microfiltration [4-6], thus there is need for an effective cleaning method.

This research focuses on the cleaning efficiency of several methods to remove Natural organic matter (NOM)-fouling on the active layer (AL) of a FO membrane immersed in a municipal secondary wastewater effluent (SWWE). Seawater was used as draw solution (DS). In addition, identification and cleaning of TEP on both the active and the support layer (SL) of the membrane were performed.



Fig. 1. FO system layout. TC—temperature controller, CC—conductivity controller, and GP—gear pump.

2. Materials and methods

The FO membranes used for this study are made of cellulose triacetate (CTA) (HTI, Scottsdale, AZ, USA). The AL of the membrane was facing the FO feed, leaving the SL in contact with the DS. This configuration (FO mode) has been proven to be the most effective in preventing fouling of the FO membrane [7–10]. The feed for the FO process was SWWE from the city of Jeddah. For the DS, a batch of Red Sea water was used.

Fig. 1 shows a detailed scheme of the set-up to induce fouling in the membrane. This configuration has been used in previous studies with relevant results [11]. The FO dilution process takes 24 h recirculating the DS inside the cell, increasing the level in the DS tank, while the SWWE concentrates (volume decreases). The cycle was repeated by replacing the diluted draw solution (DDS) with DS every day, to ensure that a fouling layer was formed on the surface of the membrane.

An evaporator was used to concentrate the SWWE without involving a membrane process, in order to identify the NOM fraction causing fouling. This sample was referred to as evaporated wastewater effluent (EWWE) as compared to the concentrated feed water after the FO process (CFW). A liquid chromatograph coupled with organic carbon detection analyser (LC–OCD) was used to analyse the organic carbon content of the water samples, following the procedure established by Huber et al. [12].

Two hydrodynamic cleaning procedures were used to remove fouling from the surface of the FO membrane. First, osmotic backwash was performed replacing the feed with 4% NaCl solution and recirculating DI water inside the cell. Afterwards, cleaning with air scouring was performed for 15min with airstone diffusers at a rate of 12.5 L/h of air per liter of SWWE. This last procedure has been proved to be very effective to remove foulants in an FO membrane.

Two chemical cleaning solutions were used to clean fouling on the FO membrane after the use of hydrodynamic procedures. The first washing solution was a 1% aqueous solution of NaOCl. The second one contained 0.8% of sodium EDTA and 1% of Alconox (commercial detergent containing phosphates). Both



Fig. 2. LC–OCD chromatograms for the CFW, EWWE, SWWE, DS and DDS.

solutions were tested on both the active and the SL to remove NOM and TEP fouling.

The Villacorte et al. protocol was used to prepare the cationic dye Alcian Blue, with which the fouled FO membrane was stained in order to assess TEP accumulation [6]. Transmitted and reflected light microscopy was used with different objectives to determine the best option for TEP removal.

3. Results and conclusions

LC–OCD chromatograms of the CFW, EWWE and DDS suggest that biopolymers (protein-like substances and polysaccharides), present in the SWWE and in the EWWE, but not in the CFW or the DDS (no partitioning through the membrane is observed), were the main constituents of the fouling layer in the AL of the FO membrane (Fig. 2).

After achieving a flux decline of 25%, cleaning was performed. Osmotic backwash did not help to recover any flux. The interaction of the salt in the cleaning solution with the foulants might have enhanced the pore blockage and thus the water flux kept decreasing. A second hypothesis is the reduction of the osmotic pressure difference due to salt accumulation in the fouling layer, which eventually reduce the flux through the membrane. Nevertheless, NOM-fouling showed high hydraulic reversibility (89.5% flux recovery) when air scouring for 15 min within the concentrated wastewater effluent was used as a cleaning technique.

TEP forms clusters clearly identifiable with an optical microscope (Fig. 3(a) and (c)). After a 10-min cleaning of the FO membrane with the Alconox



Fig. 3. (a) TEP fouling in the AL of an FO membrane ($60 \times$); (b) AL of a chemically cleaned FO membrane ($100 \times$); (c) TEP fouling in the SL of an FO membrane ($100 \times$), and (d) SL of a chemically cleaned FO membrane ($100 \times$).

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+ Sodium EDTA solution, TEP was significantly removed (Fig. 3(b) and (d)); nevertheless, traces of these particles can still be observed on both layers of the membrane after the cleaning.

Chemical cleaning of the AL with Alconox + Sodium EDTA proved to slightly increase the reversibility (93.6%). Chemical cleaning of the SL increased the chemically reversible fouling up to 94.5%. The irreversible fouling in this set of experiments was 5.5%, which may be attributed to biopolymers that cannot be removed from the membrane surface, along with the trace TEP left on the membrane surface.

Cleaning with a 1% aqueous solution of NaOCl compromised the integrity of the membrane as proven by conductivity and flux measurements, even though the pH of the solution was kept bellow 8, as recommended by the manufacturer.

References

- [1] T. Shibutani, T. Kitaura, Y. Ohmukai, T. Maruyama, S. Nakatsuka, T. Watabe, H. Matsuyama, Membrane fouling properties of hollow fiber membranes prepared from cellulose acetate derivatives, J. Membr. Sci. 376(1–2) (2011) 102–109.
- [2] V. Yangali-Quintanilla, Z. Li, R. Valladares, Q. Li, G. Amy, Indirect desalination of Red Sea water with forward osmosis and low pressure reverse osmosis for water reuse, Desalination 280(1–3) (2011) 160–166.
- [3] H.-M. Zhang, J.-N. Xiao, Y.-J. Cheng, L.-F. Liu, X.-W. Zhang, F.-L. Yang, Comparison between a sequencing batch membrane bioreactor and a conventional membrane bioreactor, Proc. Biochem. 41(1) (2006) 87–95.

- [4] T. Berman, R. Mizrahi, C.G. Dosoretz, Transparent exopolymer particles (TEP): A critical factor in aquatic biofilm initiation and fouling on filtration membranes, Desalination 276 (1–3) (2011) 184–190.
- [5] E. Bar-Zeev, I. Berman-Frank, B. Liberman, E. Rahav, U. Passow, T. Berman, Transparent exopolymer particles: Potential agents for organic fouling and biofilm formation in desalination and water treatment plants, Desalin, Water Treat. 3(1–3) (2009) 7.
- [6] L.O. Villacorte, M.D. Kennedy, G.L. Amy, J.C. Schippers, The fate of transparent exopolymer particles (TEP) in integrated membrane systems: Removal through pre-treatment processes and deposition on reverse osmosis membranes, Water Res. 43(20) (2009) 5039–5052.
- [7] B. Mi, M. Elimelech, Chemical and physical aspects of organic fouling of forward osmosis membranes, J. Membr. Sci. 320(1–2) (2008) 292–302.
- [8] Y. Wang, F. Wicaksana, C.Y. Tang, A.G. Fane, Direct microscopic observation of forward osmosis membrane fouling, Environ. Sci. Technol. 44(18) (2010) 7102–7109.
- [9] E.R. Cornelissen, D. Harmsen, K.F. de Korte, C.J. Ruiken, J.-J. Qin, H. Oo, L.P. Wessels, Membrane fouling and process performance of forward osmosis membranes on activated sludge, J. Membr. Sci. 319(1–2) (2008) 158–168.
- [10] S. Zou, Y. Gu, D. Xiao, C.Y. Tang, The role of physical and chemical parameters on forward osmosis membrane fouling during algae separation, J. Membr. Sci. 366(1–2) (2011) 356–362.
- [11] R. Valladares Linares, V. Yangali-Quintanilla, Z. Li, G. Amy, NOM and TEP fouling of a forward osmosis (FO) membrane: Foulant identification and cleaning, J. Membr. Sci. 421–422(0) (2012) 217–224.
- [12] S.A. Huber, A. Balz, M. Abert, W. Pronk, Characterisation of aquatic humic and non-humic matter with size-exclusion chromatography—organic carbon detection—organic nitrogen detection (LC–OCD–OND), Water Res. 45(2) (2011) 879–885.