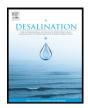
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Impact of membrane bioreactor operating conditions on fouling behavior of reverse osmosis membranes in MBR–RO processes

Bing Wu^{a,*}, Tamotsu Kitade^b, Tzyy Haur Chong^a, Tadahiro Uemura^b, Anthony G. Fane^{a, c}

^a Singapore Membrane Technology Centre, Nanyang Technological University, 1 Cleantech Loop, CleanTech One #06-08, 637141, Singapore

^b Toray Singapore Water Research Center, 1 Cleantech Loop, CleanTech One #03-09, 637141, Singapore

^c Department of Civil and Environmental Engineering, Nanyang Technological University, 639798, Singapore

HIGHLIGHTS

- ► High F/M-MBR permeate led to higher RO fouling rate.
- ► Soluble polysaccharides and TEP on the RO membranes were associated with RO fouling.
- ▶ Propagation of bacterial cells on the RO membranes did not determine RO fouling.
- ▶ The presence of inline filters in the RO systems alleviated RO fouling.
- ► FA-like and microbial by-product-like substances were predominant in RO foulants.

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ABSTRACT

This study compares fouling propensities of reverse osmosis (RO) membranes in two parallel MBR–RO systems. Two MBRs were operated at different food to microorganism (F/M) ratios and the permeate was fed to the respective RO membrane. The results show that greater amounts of organic substances in the high F/M (0.50 g/g day⁻¹)-MBR permeate led to higher RO fouling rates (>4.5-fold) compared to the low F/M ratio (0.17 g/g day⁻¹)-MBR permeate. The presence of filters (~5 μ m) in the RO feed line and recycled RO concentrate line significantly alleviated RO fouling. Chemical analysis of RO foulants indicated that the soluble polysaccharides and transparent exopolymer particles (TEP) accumulated on the RO membranes were strongly associated with RO fouling. However, propagation of bacterial cells on the membranes did not determine RO fouling development. This finding was further confirmed by confocal laser scanning microscopy images. Furthermore, excitation–emission matrix (EEM) fluorescence spectroscopy was used to trace the fate and transport of the potential soluble foulants in the MBR–RO system.

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1. Introduction

Enhanced wastewater reclamation has an increasingly important role because of freshwater supply scarcity and population growth. To meet the strict criteria for reclamation water, pressure-driven membrane separation units such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) or combined processes of these units are utilized in water reuse processes [1]. For example, a dual membrane bioreactor (MBR)–RO process has been applied to reclaim municipal wastewater due to the improved quality of effluent achieved in MBRs. An MBR process can remove above 95% organic carbon and completely remove suspended solids from wastewaters by biodegradation and membrane retention. Moreover, less waste sludge is produced compared to the conventional active sludge process. Subsequently, the RO membrane eliminates dissolved solids, organic compounds, nutrients and pathogens in MBR effluent to produce high quality reclaimed water [2–5].

However, the universal appeal of this hybrid technology is limited by membrane fouling, which reduces productivity and increases energy costs, in particular due to membrane fouling in RO systems [6]. The colloidal, organic, and inorganic substances in MBR effluent promote organic fouling and scaling of RO membranes. In addition, biofilm development on RO membranes becomes biofouling that deteriorates RO performance [1,7,8]. A number of studies have investigated the RO fouling propensities of lab-scale or pilot-plant MBR–RO processes treating various wastewaters [1–4]. Kent et al. found that proteins were the predominant RO foulants in the initial fouling stage of the RO membrane, but polysaccharides deposition on the membrane surface became dominant after operating the RO membrane for a few weeks in a MBR–RO system [9]. In the previous work investigated by Jacob et al., it was observed that the compositions and molecular weight distributions of the MBR permeate were related with the fouling behaviors of the RO membranes [10].



Corresponding author. Tel.: +65 91950394; fax: +65 67910756.
E-mail address: wubing@ntu.edu.sg (B. Wu).

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As RO feed water, MBR effluent components such as total organic carbon (TOC, indicating nutrient level), extracellular polymeric substances (EPS, indicating chemical composition), or transparent exopolymer particle (TEP, indicating the physical nature of some organics) are thought to be associated with fouling propensity of RO membranes [11-13]. Furthermore, the MBR permeate quality could be influenced by the operating conditions (e.g., hydraulic retention time (HRT), sludge retention time (SRT), substrate loading (i.e., food to microorganisms ratio) and composition, filtration flux, membrane pore size, etc.) that determine biomass characteristics (e.g., concentration, viscosity, microbial community, EPS production) and/or membrane fouling conditions [14–17]. For example, higher levels of protein and polysaccharides in the MBR permeate were detected at sub-critical flux compared to critical flux and super-critical flux in the initial filtration period of an MBR [16]. If MF membranes were used in the MBRs, they likely allowed more organic substances to pass through compared to UF membranes [5]. Therefore, the MBR design and operation could have a potential impact on RO fouling development in a MBR-RO process. However, to date, there have been few studies on the effect of MBR operating conditions on downstream RO membrane performance. The relationship between RO fouling tendency and RO foulant properties (organic substances accumulation/biofilm growth) is also not well understood.

This research aims to compare the fouling propensities of RO membranes fed with various MBR permeates, which were obtained from two MBRs operated at different ratios of food to microorganisms (F/M). The relationship between MBR permeate quality and RO fouling rate was investigated. The contributions of soluble organic substances and viable cells in the RO biofilm layers to transmembrane pressure (TMP) increase were examined. The information on fouling behavior of the RO membranes offers opportunities to reduce RO fouling in the MBR–RO processes by optimization of MBR operating conditions.

2. Materials and methods

2.1. MBR-RO description and operating conditions

A schematic of a lab-scale MBR–RO setup is shown in Fig. 1. Two MBRs were seeded with the activated sludge (\sim 6 g/L) taken from a wastewater treatment and reclamation plant. The biomass concentrations in both MBRs were maintained at \sim 6 g/L by regulating the biomass wastage (i.e., adjusting sludge retention time (SRT)). A concentrated synthetic

wastewater was fed into the two MBRs at an organic loading of 1.0 and 3.0 gCOD/L day respectively by regulating their individual feed pumps. At the same time, tap water was supplied into each MBR to maintain an effective reactor volume of 30 L, whose condition was controlled by a level sensor in the reactor. The composition of synthetic wastewater was CH₃COONa (32 g/L), NH₄Cl (4.8 g/L), MgSO₄·7H₂O (0.4 g/L), K_2HPO_4 (3.5 g/L), $CaCl_2 \cdot 2H_2O$ (0.55 g/L), and $FeSO_4 \cdot 7H_2O$ (0.02 g/L). All chemicals were from Daejung (Korea). Each MBR had six submerged flat-sheet membrane modules (Polyvinylidene Fluoride (PVDF), hydrophilic, 0.08 µm nominal pore size, 0.0288 m² of effective surface area for each membrane module, Toray Industries, Inc., Japan, details in the supplementary data Table A.1). A permeate flux of 20 L/m² h (LMH) was maintained for each membrane module by regulating the flowrate of individual suction pumps (on 9 min/off 1 min, controlled by a timer). The TMP of each membrane module was monitored by a pressure transducer, which was connected to a personal computer equipped with data logging system (Msystem, Japan). Hydrochloric acid (Daejung, Korea) was automatically added into the reactor when the pH was higher than 7.1, whose condition was controlled by a pH sensor in the reactor. The MBR operating conditions are summarized in Table 1.

Two parallel RO cells with commercial brackish water RO membranes (UTC-70, 32 mm \times 7 mm, 0.0186 m² of effective surface area, Toray Industries, Inc., Japan, details in the supplementary data Table A.1) and feed channel spacers were used. For each RO unit, the MBR permeate was collected and stored in a feed tank (10 L) with a stirrer (IKA, Germany) at a temperature of 25 ± 1 °C (controlled by a cooling water system). The MBR permeate was delivered from the feed tank using a high-pressure pump (Winston Engineering Corporation, Singapore) to the RO cell. The feed pressure was controlled by a back-pressure regulator (Swagelok, USA) and monitored by a pressure transducer (Ashcroft, USA). The cross-flow rate of feed (20 L/h, equivalent to 0.1 m/s) was regulated by a flow control valve (Swagelok, USA) and recorded by a flow meter (Brooks Instrument, USA). The permeate flowrate was monitored and controlled by a mass flow controller (Brooks Instrument, USA) to maintain the permeate flux of 20 LMH automatically. The permeate pressure was monitored by a pressure transducer (Ashcroft, USA). The conductivities of feed and permeate were measured by conductivity meters (Thermo Scientific, USA). Due to the limited productivity of MBR permeate, the RO concentrate and RO permeate were recycled back to the feed tank and overflow of this mixture was conducted. The pressure transducers, mass flow controllers, and conductivity meters

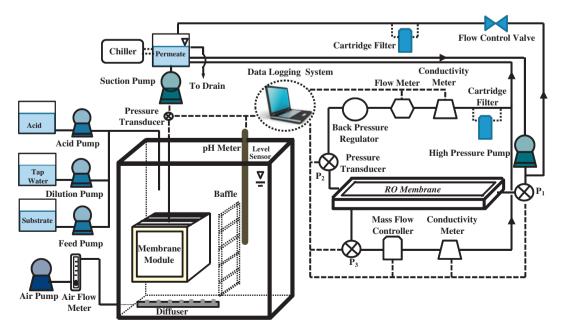


Fig. 1. Schematic diagram of a lab-scale MBR-RO system.

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