



The role of forward osmosis and microfiltration in an integrated osmotic-microfiltration membrane bioreactor system



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HIGHLIGHTS

- By integrating MF with OMBR, salinity build-up in the bioreactor could be controlled.
- Hydrophobic and readily biodegradable TrOCs were well removed (>90%) by O/MF-MBR.
- OMBR offered better treatment than MF-MBR for hydrophilic/persistent TrOCs.
- Resistant TrOCs retained by forward osmosis leaked through MF channel of the O/MF-MBR.

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ABSTRACT

This study investigates the performance of an integrated osmotic and microfiltration membrane bioreactor (O/MF-MBR) system for wastewater treatment and reclamation. The O/MF-MBR system simultaneously used microfiltration (MF) and forward osmosis (FO) membranes to extract water from the mixed liquor of an aerobic bioreactor. The MF membrane facilitated the bleeding of dissolved inorganic salts and thus prevented the build-up of salinity in the bioreactor. As a result, sludge production and microbial activity were relatively stable over 60 days of operation. Compared to MF, the FO process produced a better permeate quality in terms of nutrients, total organic carbon, as well as hydrophilic and biologically persistent trace organic chemicals (TrOCs). The high rejection by the FO membrane also led to accumulation of hydrophilic and biologically persistent TrOCs in the bioreactor, consequently increasing their concentration in the MF permeate. On the other hand, hydrophobic and readily biodegradable TrOCs were minimally detected in both MF and FO permeates, with no clear difference in the removal efficiencies between two processes.

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1. Introduction

Water reuse is an important measure to tackle water scarcity and environmental pollution, which are key factors hampering economic development and threatening the natural ecosystem (Wintgens et al., 2008; Hochstrat et al., 2010). Safe and reliable water reuse requires adequate removal of salts, nutrients, pathogenic agents, and trace organic chemicals (TrOCs) from the reclaimed effluent. TrOCs are a diverse range of emerging organic chemicals of either anthropogenic or natural origin. They occur ubiquitously in municipal wastewater at concentrations in the range of a few nanograms per liter (ng/L) to several micrograms per liter (μg/L) (Luo et al., 2014). These TrOCs present arguably

the most vexing challenge to practical potable water reuse (Wintgens et al., 2008; Lampard et al., 2010; Drewes et al., 2013; Luo et al., 2014).

Adequate removal of TrOCs is also essential to facilitate water reuse for agriculture production. It has been demonstrated that the occurrence of pharmaceuticals, such as carbamazepine and triclocarban, in reclaimed wastewater (Tanoue et al., 2012) and biosolids (Wu et al., 2012) used to grow fruits and vegetables can bio-accumulate in edible parts of these produces. Therefore, a major technical challenge for the water industry is to develop new treatment processes that can reliably and cost-effectively remove these TrOCs during water reuse.

Recent efforts in wastewater treatment and reuse have led to the emergence of a novel osmotic membrane bioreactor (OMBR) process (Achilli et al., 2009; Cornelissen et al., 2011; Nawaz et al., 2013), which integrates forward osmosis (FO) with the

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conventional activated sludge treatment technology. In the OMBR system, the osmotic pressure difference between the mixed liquor and draw solution (e.g. NaCl) induces water diffusion through a semi-permeable FO membrane. The FO membrane can effectively retain small organic contaminants in the bioreactor, thereby facilitating their subsequent biodegradation (Alturki et al., 2013; Coday et al., 2014). Indeed, recent studies have shown the excellent performance of OMBR for TrOC removal, particularly the compounds featured with a negative charge (Alturki et al., 2012; Lay et al., 2012; Holloway et al., 2014). Thus, OMBR can potentially produce high quality reclaimed water for potable reuse, irrigation, or direct discharge in environmentally sensitive areas.

Despite the potential of OMBR, salinity build-up in the bioreactor caused by high rejection by the FO membrane and reverse transport of the draw solute remains a technical challenge for its further development (Van der Bruggen and Patricia, 2015). The high bioreactor salinity can reduce the driving force for water transport (Lay et al., 2010). Sludge characteristics and microbial community can also be altered with the elevated bioreactor salinity and subsequently worsen the biological treatment and membrane performance (Qiu and Ting, 2013). A short sludge retention time (SRT) is expected to control the build-up of salinity in the bioreactor. However, in an OMBR system with an operating SRT of 10 days, the bioreactor salinity still increased substantially, exerting inhibition on the microbial activity (Wang et al., 2014a). The short SRT could also adversely affect the biological performance (Grelrier et al., 2006). Several studies have recently proposed the integration of an microfiltration (MF) or ultrafiltration (UF) process with OMBR to bleed out inorganic salts from the bioreactor (Holloway et al., 2014, 2015; Wang et al., 2014b). By applying the approach, Holloway et al. (2014, 2015) showed a stable operation of a pilot UFO-MBR treating raw domestic wastewater over a period of four months. Removal to below the detection limit was reported for 15 out of 20 TrOCs investigated in their study using a pilot reverse osmosis process for draw solution and clean water recoveries (Holloway et al., 2014).

Building upon the existing literature on this topic, we aimed to evaluate the performance of an integrated osmotic and microfiltration membrane bioreactor (O/MF-MBR) by specifically comparing permeate qualities between the FO and MF processes and examining sludge stability in the bioreactor. The system performance was also assessed in terms of water flux, bioreactor salinity, and membrane fouling. TrOC removal was related to their hydrophobicity and molecular structures to mechanistically elucidate their fate within the integrated O/MF-MBR system. The interaction between FO and MF in the integrated system with regards to the fate and removal of TrOCs was also discussed.

2. Materials and methods

2.1. Representative trace organic chemicals

A stock solution containing 30 TrOCs (Table S1, Supplementary Data) were prepared in pure methanol and stored at $-18\text{ }^{\circ}\text{C}$ in the dark. The stock solution was used within a month. These TrOCs were selected to represent four major groups of chemicals of emerging concern – pharmaceutical and personal care products, endocrine disrupting compounds, pesticides, and industrial chemicals – that are ubiquitous in municipal wastewater. They have a diverse range of properties, including hydrophobicity, molecular weight, and functional groups (Table S1, Supplementary Data). Hydrophobicity of an organic compound can be measured by $\text{Log}D$, which is the effective octanol–water partition coefficient at a given solution pH (Nghiem and Coleman, 2008). Based on their

$\text{Log}D$ values at pH of 7, the selected TrOCs can be classified as hydrophilic (i.e. $\text{Log}D_{\text{pH } 7} < 3$) or hydrophobic (i.e. $\text{Log}D_{\text{pH } 7} > 3$).

2.2. FO and MF membranes

A flat-sheet, cellulose based membrane supplied by Hydration Technology Innovations (HTI, Albany, USA) was used in the FO process. The FO membrane is composed of a cellulose triacetate active (CTA) layer reinforced by a polyester mesh for mechanical support (McCutcheon and Elimelech, 2008). It is noteworthy that thin film composite (TFC) FO membranes with embedded polyester screen support have also been released by HTI and several other manufacturers in recent years. Both CTA and TFC membranes have their own positive attributes. Findings from this study are specific to the OMBR process rather than membrane properties. Thus, results from this study could be applicable to all FO membranes.

A hollow fiber, polyvinylidene fluoride MF membrane module from Mitsubishi Rayon Engineering (Tokyo, Japan) was submerged in the bioreactor. The effective surface area and nominal pore size of the MF membrane were 740 cm^2 and $0.4\text{ }\mu\text{m}$, respectively.

2.3. Experimental system

The integrated O/MF-MBR system used in this study was composed of a cross-flow FO configuration, a submerged MF membrane module, and a 10 L aerobic bioreactor (Fig. 1). An electrical air pump (Heilea, Ningbo, China) was used to continuously aerate the reactor via a coarse diffuser. A Masterflex peristaltic pump (Cole-Parmer, Vernon Hills, USA) was used to draw permeate through the MF membrane with an operation on/off time of 14/1 min. Transmembrane pressure (TMP) of the MF membrane was continuously monitored by a high resolution ($\pm 0.1\text{ kPa}$) pressure sensor (Extech Instruments, Nashua, USA).

A detailed description of the cross-flow FO configuration is available elsewhere (Alturki et al., 2012). Briefly, the FO configuration comprised two semi-cells made of acrylic plastic and draw solution delivery and control equipment. The FO membrane was placed between two semi-cells to seal the feed and draw solution channels with a length, width, and depth of 145, 95, and 2 mm, respectively. The effective membrane surface area was 138 cm^2 , with the active layer facing the feed channel (i.e. FO mode). The mixed liquor in the bioreactor was circulated to the feed channel by a Masterflex peristaltic pump (Cole-Parmer, Vernon Hills, USA). On the other side, a gear pump (Micropump, Vancouver, USA) was used to circulate a draw solution to the draw solution channel. The circulation flow rate of both feed and draw solutions was 1 L/min (i.e. a cross-flow velocity of 9 cm/s) monitored by rotameters (Cole-Parmer, Vernon Hills, USA). The draw solution reservoir was placed on a digital balance connected to a computer. During the experimental period, the draw solution concentration was kept constant by a conductivity controller equipped with a conductivity probe and a Masterflex peristaltic pump to automatically dose a concentrated draw solution to the draw solution reservoir. The controller accuracy was 0.1 mS/cm (i.e. 0.05 g/L NaCl). Both the concentrated and working draw solution reservoirs were placed on the same digital balance to avoid errors in flux estimation due to the concentration control equipment.

2.4. Experimental protocol

A submerged MF-MBR system was first seeded with activated sludge from the Wollongong Wastewater Treatment Plant (Wollongong, Australia). The initial mixed liquor suspended solid (MLSS) concentration in the bioreactor was approximately 5 g/L . A synthetic wastewater was used to simulate medium strength municipal sewage and consisted of 100 mg/L glucose, 100 mg/L

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