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A novel membrane distillation–thermophilic bioreactor system: Biological stability and trace organic compound removal

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HIGHLIGHTS

- Salinity build-up occurred during MDBR operation.
- Salinity build-up could affect TN and TrOC removal by the bioreactor.
- However, MDBR achieved high performance regarding all water quality parameters.
- Biodegradation governed the removal of most TrOCs by the bioreactor.
- Physical separation by MD governed the removal of recalcitrant TrOCs.

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ABSTRACT

The removal of trace organic compounds (TrOCs) by a novel membrane distillation–thermophilic bioreactor (MDBR) system was examined. Salinity build-up and the thermophilic conditions to some extent adversely impacted the performance of the bioreactor, particularly the removal of total nitrogen and recalcitrant TrOCs. While most TrOCs were well removed by the thermophilic bioreactor, compounds containing electron withdrawing functional groups in their molecular structure were recalcitrant to biological treatment and their removal efficiency by the thermophilic bioreactor was low (0–53%). However, the overall performance of the novel MDBR system with respect to the removal of total organic carbon, total nitrogen, and TrOCs was high and was not significantly affected by the conditions of the bioreactor. All TrOCs investigated here were highly removed (>95%) by the MDBR system. Biodegradation, sludge adsorption, and rejection by MD contribute to the removal of TrOCs by MDBR treatment.

1. Introduction

Water reclamation is a pragmatic approach to address the scarcity of water supplies in urban areas due to population growth and irregular climate pattern (Shannon et al., 2008). Through water reclamation, municipal wastewater can be a reliable alternative source for clean water supply. However, development of advanced treatment processes is necessary to ensure adequate removal of common contaminants (e.g., organics, nutrients, minerals) and especially trace organic compounds (TrOCs) that occur ubiquitously in municipal wastewater. These TrOCs include steroid hormones, pharmaceuticals, personal care products, surfactants, pesticides, disinfection by-products, and UV filters (Tran et al., 2013b; Zhao et al., 2010) that have been widely detected in raw sewage and reclaimed effluent from conventional wastewater treatment plants. Their occurrence is of major health and environmental concern because of their potential adverse impact on living organisms (Schwarzenbach et al., 2006). Thus, the removal of TrOCs during water reclamation has been the subject of intensive research in recent years.

Membrane bioreactor (MBR) is an efficient wastewater treatment technology, capable of producing reuse standard effluent (Melin et al., 2006). MBRs can effectively remove TrOCs that are hydrophobic and/or readily biodegradable (Boonyaroj et al., 2012; Clara et al., 2005; Tadkaew et al., 2011; Tran et al., 2013a); however, recent studies have highlighted the challenges of removing recalcitrant TrOCs (e.g., carbamazepine and diclofenac) by

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biological treatment processes, including MBRs (Clara et al., 2005; Radjenović et al., 2009; Tadkaew et al., 2011; Wijekoon et al., 2013b).

Tadkaew et al. (2011) suggested that biodegradability of a TrOC can be qualitatively assessed based on the presence of electron donating functional groups (EDGs) or electron withdrawing functional groups (EWGs) in their molecules. They demonstrated that TrOCs with EDGs can be well removed in an MBR, whereas TrOCs with EWGs (such as chloride and amide) in their structure are usually poorly removed by MBRs. In a subsequent study, Wijekoon et al. (2013b) successfully extended this framework to elucidate the fate of TrOCs in the aqueous and sludge phases during MBR treatment. Given the resistance of some TrOCs to biodegradation, the use of post-treatment processes to specifically target these recalcitrant TrOCs has also been explored. Examples of these post-treatment processes subsequent to MBR treatment include reverse osmosis (Alturki et al., 2010), activated carbon adsorption (Nguyen et al., 2013a), and ultraviolet oxidation (Nguyen et al., 2013b).

Integration of a high retention membrane process such as nanofiltration (Choi et al., 2002), forward osmosis (Achilli et al., 2009; Alturki et al., 2012; Hancock et al., 2013), or membrane distillation (MD) (Goh et al., 2013a,b; Khaing et al., 2010; Phattaranawik et al., 2009) with a bioreactor constitutes a so called high retention MBR, which can be an efficient means to achieve high removal of pollutants. The working principles of these integrated processes have been demonstrated in recent studies; however, except for Alturki et al. (2012) and Hancock et al. (2011), the removal of TrOCs using these novel high retention MBRs has not been investigated.

MD is a low temperature distillation process that involves the transport of water vapour from a feed solution through the pores of a microporous and hydrophobic membrane to the distillate (product) side. Because mass transfer occurs in a gaseous phase, MD offers complete rejection of all non-volatile solutes (Curcio and Drioli, 2005). Membrane distillation bioreactor (MDBR) is a high retention MBR process where MD membrane can act as a barrier against the permeation of low molecular weight compounds and recalcitrant compounds. In the MDBR process, the biological reactor can be operated at thermophilic conditions to facilitate the integration of biological treatment with MD. In addition, the thermophilic bioreactor can also result in enhanced biodegradation of organics and low sludge yield (LaPara and Alleman, 1999).

The main aim of this study was to evaluate the performance of a novel hybrid MDBR process. Biological stability of the thermophilic bioreactor and the overall performance in terms of basic water quality parameters, as well as the fate and removal of TrOCs during MDBR treatment were elucidated.

2. Methods

2.1. MDBR experimental setup

A laboratory-scale MDBR system consisting of a glass bioreactor and an external direct contact membrane distillation (DCMD) module was used (Fig. 1). A peristaltic pump (Masterflex L/S, USA) was used to continuously transfer feed wastewater to the bioreactor. The bioreactor had an active volume of 5 L and was submerged in a water bath, which was equipped with an immersion heating unit (Julabo, Germany) to keep the temperature at 40 ± 0.1 °C. It was also covered with aluminium foil to avoid any exposure to sunlight and heat loss. The bioreactor was aerated using an air pump (Risheng RS 9801, China) connected to a glass diffuser, and an overhead mixer (Heidolph Instruments, Germany) was used to maintain homogeneity within the bioreactor. The mixed liquor of the bioreactor was used as the feed to the external DCMD module.

The DCMD module was made of acrylic glass to minimize heat loss to the surroundings. The flow channels were engraved in each of two acrylic glass blocks that made up the feed and distillate semi-cells. The length, width, and height of each channel were 145, 95, and 3 mm, respectively. The total active membrane surface area for mass transfer was 140 cm^2. Feed to the MD system (mixed liquor from the bioreactor) was continuously pumped to the membrane cell and recirculated back to the bioreactor. The temperature of the feed solution entering the MD cell was monitored using a temperature sensor connected to the feed line immediately outside the inlet. The temperature of the distillate leaving the membrane cell was monitored using another temperature sensor located immediately after the outlet of the distillate semi-cell. The temperature of the distillate was kept at 14.0 ± 0.1 °C using a chiller (Neslab RTE7, Thermo Scientific, USA) equipped with a stainless steel heat exchanging coil, which was directly immersed in the distillate reservoir. A glass container was used as the distillate reservoir and was placed on a digital balance (Mettler Toledo Inc, USA) to calculate the distillate flux. Excess distillate was pumped out from the distillate reservoir intermittently and collected in a stainless steel container for analysis. The MD feed and distillate flow rate were monitored using two rotameters and maintained at 1 L/min (corresponding to a cross flow velocity of 9 cm/s). Milli-Q water (2.25 L) was used as the initial distillate. The MDBR system was covered with insulation foam to minimize heat loss. A hydrophobic microporous polytetrafloroethylene (PTFE) membrane (GE, Minnetonka, MN) was used. The average pore size, porosity, thickness and active layer thickness of this membrane were 0.22 μm, 70%, 175 μm, and 5 μm, respectively (Nghiem and Cath, 2011).

2.2. Experimental protocol

The bioreactor system was inoculated with activated sludge from the Wollongong Wastewater Treatment Plant (Wollongong, Australia). A synthetic wastewater was used to simulate medium strength domestic wastewater and to maintain stable operating conditions. The synthetic wastewater was prepared daily by diluting a concentrated stock with Milli-Q water to obtain 100 mg/L glucose, 100 mg/L peptone, 17.5 mg/L KH_2PO_4, 17.5 mg/L $MgSO_4$, 10 mg/L $FeSO_4$, 225 mg/L CH_3COONa, and 35 mg/L urea (Alturki et al., 2012). The concentrated stock solution was prepared every week and kept at 4 °C in the dark.

Prior to the MDBR experiment, the bioreactor was acclimatised at 40 °C by operating the system in an MBR mode using a ceramic microfiltration membrane module (NGK, Japan). During the acclimatisation period, the bioreactor was operated at a hydraulic retention time (HRT) of 24 h and a solids retention time (SRT) of 88 d. The temperature, dissolved oxygen (DO) concentration, and conductivity of the mixed liquor were 40 °C, 2.8 ± 0.5 mg/L, and 425 μS/cm, respectively. The mixed liquor suspended solids (MLSS) concentration was 5.3 g/L, and under these operating conditions the mixed liquor pH remained stable at 7.6. More details about the ceramic MBR system are available elsewhere (Wijekoon et al., 2013b). After the bioreactor had been acclimatised for 75 d, the ceramic microfiltration membrane module was removed and the bioreactor was connected to the DCMD system. TrOCs were then continuously introduced to the influent at a concentration of approximately 5 μg/L of each compound. MDBR operation was initiated at temperature and DO concentration of 40 °C and 2.8 ± 0.5 mg/L, respectively, and operated for 38 d. The HRT of the MDBR was 9.6 d due to the low distillate flux of the DCMD system. The basic biological performance of the MDBR in terms of total organic carbon (TOC) and total nitrogen (TN) removal, conductivity/pH variation, and MLSS concentration was continuously monitored. The mixed liquor was collected weekly and centrifuged at