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# Assessing the removal of organic micropollutants by a novel baffled osmotic membrane bioreactor-microfiltration hybrid system



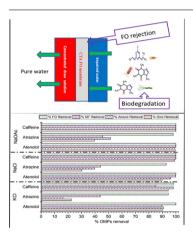
Nirenkumar Pathak<sup>a</sup>, Sheng Li<sup>b</sup>, Youngjin Kim<sup>b</sup>, Laura Chekli<sup>a</sup>, Sherub Phuntsho<sup>a</sup>, Am Jang<sup>c</sup>, Noreddine Ghaffour<sup>b</sup>, TorOve Leiknes<sup>b</sup>, Ho Kyong Shon<sup>a,\*</sup>

<sup>a</sup> Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Post Box 129, Broadway, NSW 2007, Australia

<sup>b</sup> King Abdullah University of Science and Technology (KAUST), Water Desalination and Reuse Center (WDRC), Biological & Environmental Science & Engineering Division (BESE), Thuwal, 23955-6900, Saudi Arabia

<sup>c</sup> Graduate School of Water Resources, Sungkyunkwan University, 2066 Seobu-ro, Jangan-Gu, Suwon, Gyeonggi-Do 16419, Republic of Korea

#### G R A P H I C A L A B S T R A C T



#### ARTICLE INFO

Keywords: Biofouling Microfiltration (MF) Osmotic membrane bioreactor (OMBR) Organic micropollutants (OMPs) Salinity build-up

#### ABSTRACT

A novel approach was employed to study removal of organic micropollutants (OMPs) in a baffled osmotic membrane bioreactor-microfiltration (OMBR-MF) hybrid system under oxicanoxic conditions. The performance of OMBR-MF system was examined employing three different draw solutes (DS), and three model OMPs. The highest forward osmosis (FO) membrane rejection was attained with atenolol (100%) due to its higher molar mass and positive charge. With inorganic DS caffeine (94–100%) revealed highest removal followed by atenolol (89–96%) and atrazine (16–40%) respectively. All three OMPs exhibited higher removal with organic DS as compared to inorganic DS. Significant anoxic removal was observed for atrazine under very different redox conditions with extended anoxic cycle time. This can be linked with possible development of different microbial consortia responsible for diverse enzymes secretion. Overall, the OMBR-MF process showed effective removal of total organic carbon (98%) and nutrients (phosphate 97% and total nitrogen 85%), respectively.

\* Corresponding author. *E-mail address*: Hokyong.Shon-1@uts.edu.au (H.K. Shon).

https://doi.org/10.1016/j.biortech.2018.04.044 Received 14 February 2018; Received in revised form 9 April 2018; Accepted 11 April 2018 Available online 14 April 2018 0960-8524/ © 2018 Elsevier Ltd. All rights reserved.

#### 1. Introduction

With rapidly growing world's population, climate change and extensive industrialization pressure is increasing on the limited fresh water resources (Kim et al., 2017). Increased water scarcity led to advancement of many technologies that utilize alternative water sources, such as domestic sewage. This can help in protecting environment and ease water paucity by indirect and direct reuse of impaired water (Luo et al., 2016). Nevertheless, wastewater treatment processes face some issues, specifically regarding organic micropollutant removal (Nguyen et al., 2016). Removal of OMPs from wastewater by current physicochemical technologies is prohibitively expensive (Singhal and Perez-Garcia, 2016). Wastewater treatment facilities using conventional activated sludge treatment are ineffective and inconsistent of degrading the wide range of organic micropollutants that could potentially pose risks to humans and the environment (Holloway et al., 2014; Singhal and Perez-Garcia, 2016).

Therefore, main concern to reclaimed water application is unpredictable and usually limited OMPs removal by traditional wastewater treatment processes (Luo et al., 2016). Novel OMPs removal or destruction options are activated carbon, ultraviolet (UV) disinfection and advanced oxidation processes (AOP). Also, membrane bioreactor (MBR), reverse osmosis (RO), nanofiltration (NF) and more recently, forward osmosis (FO) have been used extensively. In various studies it was found that the removal efficiency mostly depends on OMPs hydrophilicity, surface charge, and bio degradability (Besha et al., 2017; Coday et al., 2014; Luo et al., 2016). Advanced treatment processes based on MBR technologies can achieve higher and more consistent micropollutants removal compared to conventional systems (Besha et al., 2017). It has been investigated that MBR process efficiently remove moderately biodegradable and hydrophobic OMPs to conventional process (Park et al., 2015). Although MBR effectively removes some of the OMPs like ibuprofen, it cannot be used as an absolute barrier to effectively remove some poorly biodegradable OMPs (Alturki et al., 2012; Besha et al., 2017). In fact, MBR could not remove hydrophilic OMPs very well due to their persistent nature and low bioadsorption on sludge (Luo et al., 2017b).

More recently research effort has been dedicated to the development of an innovative osmotic membrane bioreactor (OMBR) process, which uniting activated sludge process with forward osmosis (FO) membrane (Luo et al., 2015); Nguyen et al., 2015). FO membrane appeared as a robust alternative for the indirect and direct potable reuse applications and a good barrier to OMPs in the bioreactor thereby aiding their consequent biodegradation (Coday et al., 2014). OMBRs have many advantages such as higher quality product water, lower fouling propensity than conventional membrane processes and better energy efficiency. However, the salt accumulation in the mixed liquor due to the reverse salt flux (RSF) from the draw solution (DS) and the osmotic concentration of the feed solution leads to a reduced driving force for permeate flux and also affect the biological activity inside the reactor (Lay et al., 2010; Luo et al., 2015a; Nguyen et al., 2015; Pathak et al., 2017; Qiu and Ting, 2013).

Nitrogen compounds are nutrients limiting sewage treatment and they can cause a variety of problems, including a reduction in concentrations of dissolved oxygen in water sources which can lead the death of fish and also bring about eutrophication (Derakhshan et al., 2018). Nitrogen removal by simultaneous nitrification/denitrification (SND) requires a separate anoxic tank in the process with circulation of the mixed liquor from the anoxic tank and membrane tank in a MBR plant. Circulation of the mixed liquor suspension accounts for 10–15% of total energy consumption in current MBRs. Kimura et al. (2007) proposed a baffled membrane bioreactor that can eliminate the energy needed for the mixed liquor circulation. In our previous work, Pathak et al. (2017) successfully demonstrated the performance of a novel baffled OMBR in which baffles are inserted in the reaction tank and feed water is drawn through FO membranes. By utilizing this method, SND can be promoted in a single reactor. As a result, sludge recirculation pump and anoxic tank mixer can be omitted (Pathak et al., 2017).

This paper investigated the technical feasibility of a baffled OMBR-MF hybrid system for wastewater treatment applications, in particular the efficiency of OMPs removal under oxic-anoxic conditions. This study also evaluated carbonaceous organic removal and nutrient removal in a baffled OMBR-MF system employing inorganic and organic draw solutes. The baffled OMBR-MF system performance was investigated in terms of permeate flux and salt accumulation, analysing growth of biofouling layer and microbial activity using confocal laser scanning microscopy (CLSM), extracellular polymeric substances (EPS) and floc size analysis respectively. Lastly, three model OMPs were analysed to evaluate the feasibility for wastewater reuse.

#### 2. Materials and methods

#### 2.1. FO and MF membrane characteristics

The Hydration Technology Innovations (HTI) (Albany, OR, USA) made Cellulose triacetate (CTA) FO membranes was employed in this work (Kim et al., 2017). FO membrane area was  $0.0264 \text{ m}^2$ . The submerged FO membrane module was custom designed and fabricated using stainless steel (SS). The hollow-fiber micro-filtration (MF) membrane (Uniqflux Membranes LLP, India) was employed in this study. The MF membrane had  $0.1 \text{ m}^2$  area and a nominal pore size of  $0.33 \,\mu\text{m}$  respectively.

#### 2.2. Feed solutions

All the chemicals used in this research were of reagent grade (Sigma Aldrich Saudi Arabia). The feed to the OMBR-MF hybrid system was a simulated sewage consisting of 300 mg/L glucose, 50 mg/L yeast, 15 mg/L KH<sub>2</sub>PO<sub>4</sub>, 10 mg/L FeSO<sub>4</sub>, 60 mg/L (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, and 30 mg/L urea. The simulated sewage was prepared from stock solution providing 350 mg/L chemical oxygen demand (COD), 16 mg/L ammonium nitrogen (NH<sub>4</sub>-N), 28 mg/L total nitrogen (TN) and 3.5 mg/L phosphate (PO<sub>4</sub>-P).

In lab-scale baffled OMBR-MF hybrid system three OMPs removal was examined. Atenolol, atrazine and caffeine were supplied by Sigma Aldrich (Saudi Arabia). All three OMPs commonly exist in sewage and considered as persistent to conventional activated sludge process. Atenolol is a beta-blocker retaining both electron donating groups (–OR, –CH) and electron withdrawing group (–CONH<sub>2</sub>). Similarly atrazine is a herbicide also has electron donating group (-NHR) and electron withdrawing group (–Cl). Wei et al. (2016) reported that caffeine being a stimulant and its removal was too less in the beginning and it would need longer adaptation time even though cyclic structure has strong electron donating group (–R).

#### 2.3. Draw solutions

The DS employed in this study were prepared to make 0.75 M by dissolving respective salts of three different chemicals sodium chloride (NaCl), potassium chloride (KCl) and sodium acetate (CH<sub>3</sub>COONa or NaOAc) in deionized (DI) water. The chemicals used in this study were reagent grade procured from Sigma-Aldrich, Saudi Arabia. NaCl and KCl were selected as inorganic DS and NaOAc was chosen as organic DS to compare the performance of baffled OMBR between inorganic and organic draw solutes. NaCl is commonly used inorganic ionic salt draw solute in most of all OMBR studies. KCl is inorganic salt with high water flux. These solutes are ideal for minimizing ICP and creating high osmotic pressures. Both NaCl and KCl possess high solubilities in water and no toxicity. The use of organic based draw solutes such as salts of NaOAc exhibits significantly lower reverse salt flux than that for inorganic DS. Additionally, organic DS are biodegradable and do not contribute toward salinity build-up in the bioreactor (Achilli et al.,

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