Journal of Membrane Science 499 (2016) 555–562

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Membrane fouling and long-term performance of seawater-driven forward osmosis for enrichment of nutrients in treated municipal wastewater

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ARTICLE INFO

Article history:
Received 23 August 2015
Received in revised form
31 October 2015
Accepted 11 November 2015
Available online 17 November 2015

Keywords:
Forward osmosis
Nutrients recovery
Municipal wastewater
Membrane fouling
Disinfection

ABSTRACT

Forward Osmosis (FO) has been designed and studied for various applications due to the advantages it offers when compared to traditional pressure-driven membrane processes. In our previous study, seawater-driven forward osmosis was successfully applied to concentrate nutrients in treated municipal wastewater. In this study, the long-term operational stability and performance of the seawater-driven FO process was investigated by a 2-month continuous operation of bench-top FO reactors. Nitrification inhibition was recommended to maintain the concentration performance of nitrogen by preventing ammonia from being oxidized by microorganisms. With nitrification inhibition, concentration performances of 3- to 4-fold of total phosphorus and 2.1-fold of ammonia were stably achieved at a feed volume reduction of 75%. Membrane fouling, with the active layer of the FO membrane facing the feed solution, had only a mild impact; there was approximately 5% water flux reduction but no significant effect on nutrient enrichment performance. The analytical results by FTIR and SEM-XMA suggested that bio-fouling with a combined structure of bacterial clusters, biopolymers, and slight inorganic scales was the main FO membrane fouling pattern. Furthermore, pre-disinfection performed in feed solution effectively retarded the occurrence of membrane fouling in the FO process.

1. Introduction

Forward osmosis (FO), an osmotic energy driven membrane process, has been attracting increasing attention as a novel energy-saving water-treatment technology [1–4]. FO provides advantages over traditional pressure-driven membrane technologies such as reverse osmosis (RO) and nanofiltration (NF). In particular, (i) the operational cost and equipment investment for FO is reduced because it operates under low or no hydraulic pressure conditions; (ii) FO achieves high water recovery because of a reduced impact of scaling and high rejection of a wide range of contaminants; and (iii) FO is expected to have a lower membrane fouling propensity than pressure-driven membrane processes [5–7]. A number of studies and applications of FO have been reported during the past decade, including involvement in seawater and brackish water desalination [8–10], commercial materials production [11–14], and wastewater treatment [15–17].

Among the various applications of FO, the separation and recovery of nutrients (e.g., nitrogen and phosphorous) from various types of wastewater is a potential solution for natural resources and energy sustainability. Positive results were obtained with respect to the performance of FO on concentrating nitrogen and phosphorous in previous studies [15,17–19]. High levels of phosphate retention (> 90%) and improvements in nitrogen retention (up to 80%) from nutrient-rich wastewater source (e.g., urban source-separated urine) and dilute wastewater source (e.g., treated municipal wastewater) have been achieved in laboratory scale FOs [18,19]. In our previous study, the high performance of a seawater-driven FO on enriching nitrogen and phosphorous in treated municipal wastewater was further demonstrated using modeling approaches [19]. However, previous studies were mostly conducted based on batch FO tests. The long-term performance of FO on nutrient concentrations, which possibly differs from that in a batch test, has not been reported. To address the knowledge gap and comprehensively evaluate the feasibility of the proposed FO application, it is necessary to assess the nutrient enrichment stability and the performance of the FO process over a long-term period.

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http://dx.doi.org/10.1016/j.memsci.2015.11.009
0376-7388/

Fouling is a critical drawback for all membrane processes, as it causes dramatic deterioration in membrane performance and subsequently increases operational and maintenance costs. Existing bench-scale studies on FO membrane fouling have been conducted using model foulants such as alginate, proteins, humic substances and colloid particles [6,7,20–22]; these studies confirmed that membrane fouling in FO was more reversible than that in RO. It was also revealed that FO membrane fouling could be strongly affected by membrane structure and orientation in addition to other operational conditions. Cornelissen et al. [5] reported that during 48 h of contact with activated sludge in an FO membrane bioreactor (FoMBR), neither reversible nor irreversible fouling was observed on the FO membrane under the selected operational condition. Zhang et al. [23] observed significant flux decline when the active layer faced the draw solution (AL-DS) in a FoMBR. Mild membrane fouling in FoMBR and FO with secondary wastewater effluents as the feed solution was also reported in several publications [24,25]. Still, further understanding of fouling propensity and the properties of FO associated with real wastewater is desirable, particularly during long-term operations. While a significant number of studies focusing on the chemical cleaning of RO and NF membranes have been reported in scientific literatures [26–28], there are few studies that refer to the membrane fouling control strategy applied to the FO process.

The objectives of this study were to evaluate the performance and stability of a seawater-driven FO on enriching nutrients from treated municipal wastewater over a long-term period of operation. In addition, membrane fouling was characterized to deepen the understanding of the mechanisms and properties of membrane fouling, as well as to propose an effective membrane fouling control strategy in the FO process.

2. Materials and methods

2.1. FO membrane and unit configuration

A commercial flat sheet FO membrane provided by Hydration Technologies, Inc. (Albany, OR) was used in this study. The membrane has an asymmetric structure with the cellulose triacetate active layer embedded in a polyester screen mesh support. The membrane sheet was cut into proper sizes and soaked into Milli-Q water (Millipore, USA) for 24 h before use to remove residual preservative reagents.

A cross flow membrane unit (C10-T, Nitto Denko Co., Japan) was adopted. This membrane unit contained channels on both sides of the membrane for feed and draw solutions. The effective dimension of each channel was 167 mm (length), 36 mm (width), and 1.4 mm (height). Mesh spacers were applied on each side of the FO membrane to increase the turbulence at the membrane surface during the filtration process.

The FO membrane was installed with the active layer facing the feed solution and the support layer in contact with the draw solution (synthetic seawater). This membrane orientation (i.e. AL-FS) has been demonstrated to be more effective than the other membrane orientation to prevent membrane fouling in FO [5,20,22].

2.2. Draw solution and feed solution

Standard synthetic seawater [29] was prepared following the composition in Table 1 and was used as the draw solution. The synthetic seawater containing approximately 3.5 wt% of dissolved salts, generates an electric conductivity of 42.8 ± 0.3 mS cm^{-1} with an average pH of 8.12 ± 0.08 at ~25 °C. Four primary solutes, NaCl, $MgCl_2$, $CaCl_2$, and Na_2SO_4 contribute more than 97% of the total osmotic pressure of the draw solution. Additionally, a 4-fold concentrated synthetic seawater sample was prepared for the online re-concentrating of the draw solution diluted during the operation.

Table 1
Composition of synthetic seawater as FO draw solution.

Components	Concentration (g/L)	Osmotic pressure[a] $\times 10^5$ (Pa)	Contribution to the total osmotic pressure (%)
NaCl	21.03	15.17	75.5
Na_2SO_4	3.52	1.57	7.8
KCl	0.61	0.35	1.7
KBr	0.088	< 0.1 (~0.03)	0.1
$Na_2B_4O_7 \cdot 10H_2O$	0.034	< 0.1 (0.009)	< 0.1
$MgCl_2 \cdot 6H_2O$	9.50	2.38	11.8
$CaCl_2 \cdot 2H_2O$	1.32	0.50	2.5
$SrCl_2 \cdot 6H_2O$	0.02	< 0.1 (0.005)	< 0.1
$NaHCO_3$	0.17	< 0.1 (0.09)	0.4
Total		20.10	100

[a] Calculated by an online tool (http://www.lenntech.com/calculators/osmotic/osmotic-pressure.htm).

The effluent of a pilot-scale membrane bioreactor (MBR) was collected from a wastewater treatment plant in Japan and used as the feed solution. This MBR was designed in combination with an in-line sludge thickener for biomass production and nutrients recovery in wastewater [30]. By applying short hydraulic retention time (e.g., 1–2 h), nitrification was successfully suppressed at less than 50% of ammonia removal in the effluent. Despite of the short HRT applied, robust organic removal at > 90% was obtained in this MBR. During the sampling period, the DOC, TN and TP concentrations in the effluent of the MBR were observed at approximate 8.1 mg L^{-1}, 2.7 mg L^{-1}, and 20.0 mg L^{-1} respectively. Water quality of the influent is presented in Table 2. The pH was 7.9 ± 0.2, with an electric conductivity of 1.6 ± 0.1 mS cm^{-1}. An FO ($FO_{ATU+NaClO}$) with an additional of 5 mg L^{-1} allylthiourea (ATU) in its influent was operated in parallel with the one without any treatment (FO_{NT}). ATU was added to identify the impact of nitrification on the nitrogen enrichment performance in FO. The slightly higher dissolved organic carbon (DOC) concentration in the $FO_{ATU+NaClO}$ influent was attributed to the addition of ATU. Furthermore, to investigate the performance of disinfection on membrane fouling prevention, pre-disinfection was conducted for the $FO_{ATU\text{-}NaClO}$ feed solution by injecting 20 mg free Cl L^{-1} after 18 days of operation. The residual chlorine concentration in the $FO_{ATU\text{-}NaClO}$ feed tank was below 0.02 mg L^{-1}.

2.3. Laboratory-scale FO set-up and operation

The schematic of the bench-scale FO filtration system employed in this study is illustrated in Fig. 1. The feed solution was pumped at a certain flow rate into a retentate tank in which the solution was circulated inside the FO membrane unit. The draw solution was similarly circulated in the opposite side. The cross-

Table 2
FO feed solution qualities.

Contaminant	FO_{NT}	$FO_{ATU+NaClO}$
pH	7.9 ± 0.2	
Conductivity (mS cm^{-1})	1.6 ± 0.1	
DOC (mg L^{-1})	8.1 ± 0.9	10.0 ± 0.7
TP (mg L^{-1})	2.7 ± 0.5	2.7 ± 0.5
TN (mg L^{-1})	20.1 ± 2.2	20.0 ± 2.6
Ammonia-N (mg L^{-1})	10.3 ± 2.4	9.8 ± 2.1
Nitrite-N (mg L^{-1})	3.1 ± 1.8	2.6 ± 2.1
Nitrate-N (mg L^{-1})	12.8 ± 3.5	13.0 ± 5.0