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Impact of reverse nutrient diffusion on membrane biofouling in fertilizer-drawn forward osmosis

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A B S T R A C T

Biofouling in fertilizer-drawn forward osmosis (FDFO) for water reuse was investigated by spiking pure bacteria species *Pseudomonas aeruginosa PAO1 + GFP* and using three different fertilizers KNO_3, KCl and KH_2PO_4 as draw solutions. The performance of FO process for treating synthetic wastewater was assessed and their influence on the membrane fouling and in particular biofouling was evaluated relative to the type of different fertilizers used and their rates of reverse diffusion. FO performances using KNO_3 as draw solute exhibited severer flux decline (63%) than when using KCl (45%) and KH_2PO_4 (30%). Membrane autopsy indicated that the mass of organic foulants and biomass on fouled membrane surface using KNO_3 as draw solute (947.5 mg/m^2 biopolymers, 72 µm biofilm thickness and 53.3 mg/m^2 adenosine triphosphate) were significantly higher than that using KCl (450 mg/m^2 biopolymers, 33 µm biofilm thickness and 28.2 mg/m^2 ATP) and KH_2PO_4 (440 mg/m^2 biopolymers, 35 µm biofilm thickness and 33.5 mg/m^2 ATP). This higher flux decline is likely related to the higher reverse diffusion of KNO_3 (19.8 g/m^2/h) than KCl (5.1 g/m^2/h) and KH_2PO_4 (3.7 g/m^2/h). The reverse diffused potassium could promote the organics and bacterial adhesion on FO membrane via charge screening effect and compression of electrical double layer. Moreover, reverse diffused nitrate provided increased N:P nutrient ratio was favorable for the bacteria to grow on the feed side of the FO membrane.

1. Introduction

With the current world's economic growth, a 53% increase in the global water demand is expected by 2030, up to 6.9 trillion m^3/year as a total demand [1]. The agricultural sector currently consumes about 70% (global average) of accessible freshwater, however, 15–35% of this water is used unsustainably [2]. To alleviate this water stress around the world, wastewater reuse has been intensively investigated in the past two decades. However, it is essential to sufficiently treat wastewater before reuse for irrigation, not only to protect human health but also the quality and quantity of the crops grown using treated wastewater effluent. A variety of wastewater treatment methods have been investigated for irrigation reuse [3,4]. Some studies compared various technologies treating secondary wastewater effluent, including microfiltration (MF), soil-aquifer infiltration, and aerobic membrane bioreactor (MBR) [5–9]. However, these technologies are not effective in removing micro pollutants (i.e. constituents of emerging concern) and trace metals from wastewater, limiting the potential application of the reclaimed wastewaters [10] including for irrigation. Anaerobic membrane bioreactors (AnMBR) have been studied to treat wastewater because of its several advantages, including complete rejection of suspended solids, high organic rejection and biogas production and low sludge production [11]. However, high membrane fouling issues are still obstacles for both AnMBR and membrane filtration post-treatment (e.g., RO and NF), ultimately increasing the energy consumptions [12]. Osmotic membrane bioreactor (OMBR) has been proposed as a suitable alternative by integrating AnMBR with forward osmosis (FO) to overcome these issues [13–15]. A study has reported that trace organic compounds could be removed up to 80–90% by the FO process depending on the type of commercial membranes. With the increase of pressure on the feed side, the rejection of trace organic compounds decreased probably due to membrane deformation, increased external concentration polarization (ECP) and lower reverse salt flux (RSF) [16]. Bell et al. [17] has also demonstrated that the FO process using cellulose triacetate (CTA) membranes could achieve a steady-state water flux over 100 days. Because the rejection of ions by the FO membranes,

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the salinity of OMBR system might increase over time, but the incorporation of MF or UF units can substantially control the salinity build-up in OMBR and achieve a stable operation [18,19].

The FO process has been investigated for various applications in wastewater treatment, for example, activated sludge [20], drilling mud and fracturing wastewater from oil and gas industry [21], digester centrate [22], primary effluent [23], and secondary and tertiary treated effluents [24]. Although OMBR can provide high rejection of contaminants, it also exhibits some limitations, such as efforts to recover pure water from diluted draw solution (DS) and the inhibited biological process due to the reversely transported draw solute [13]. Recently, fertilizer-drawn forward osmosis (FDFO) has been investigated because the diluted DS can be used directly for irrigation purposes without the need for a draw solute separation and recovery process [25,26]. In FDFO, the FO process is driven by a concentrated fertilizer solution and thus the clean or treated water drawn from the wastewater is used to dilute the fertilizer solution which can then be directly used for fertigation. In this system, raw municipal wastewater and a highly concentrated fertilizer solution will be utilized as feed solution (FS) and DS in the FDFO process, respectively. Consequently, a diluted DS (fertilizer solution) can be obtained and supplied for greenhouse irrigation or fertigation [27]. Moreover, when the FDFO is used as a pretreatment of AnMBR, the concentrated wastewater can improve the digestion efficiency of downstream AnMBR and lead to a higher bio-methane production [28]. Three critical criteria affecting the success of the FDFO system are: i) maximization of water flux; ii) minimization of reverse solute flux and iii) control of membrane fouling (especially biofouling).

It has been reported that commercial solid fertilizers can be used as draw solutes for extracting reusable water from wastewater [29,30]. Although an effective fouling control by using chemical cleaning has been reported in the same study, the impact of RSF on fouling was not addressed.

This study is part of a project targeting wastewater reclamation through a sequential FDFO and AnMBR process. The FDFO process in this study aims to pre-concentrate the wastewater to be delivered to the AnMBR to improve its performance. When a raw municipal wastewater is utilized as feed, severe membrane fouling can be expected during the wastewater treatment by the FDFO process. There are numerous bacteria already present in raw wastewater and therefore represent a high biofouling potential. Besides, the FDFO process uses fertilizers as DS, which are mostly made up of nitrogen and phosphorous, nutrients necessary for bacteria growth. When fertilizers are used as draw solutes in a FO process, a reverse diffusion of nitrogen and phosphorus into the FO feed channel is inevitable. It has already been reported that reverse diffusion of certain fertilizer solutes can significantly enhance membrane scaling [31]. Bell et al. [29] demonstrated that FO is not a low fouling membrane process and proper pretreatments are required to protect the FO membranes from fouling. Moreover, the thin film composite (TFC) membranes exhibited a higher fouling propensity than CTA membranes despite their smoother, more hydrophilic, and more neutrally charged surfaces. Xie et al. [32] reported that reverse calcium diffusion could lead to a significant membrane fouling which was mainly due to the biofouling enhanced by calcium complexation. However, these studies did not address the impact of fertilizers which might have a stronger impact on biofilm formation depending on their corresponding diffusion capability through FO membranes.

The reverse diffused nutrients may also enhance the growth of bacteria in the presence of a carbon source on the FO membrane surface, ultimately leading to an increase of the development of biomass thereby enhancing biofouling. Phosphate nutrient limitation has been demonstrated as a measure to control membrane biofouling [33]. Besides the types of nutrients, the extent of biofilm formation and biofouling on FO membrane could also depend on other factors such as higher reverse diffusion rate of the nutrient and the N:P nutrient ratio. However, at present, it is not clear whether or not the reverse diffusion of fertilizer from the DS will influence the biofouling. To our knowledge, no studies have been reported demonstrating the potential effect of fertilizer reverse diffusion on biofouling in the FDFO process, and which type of fertilizers is preferable to be utilized to minimize membrane biofouling. To render the FDFO process economically feasible, the potential for biofouling of FO membranes should be fully understood and addressed.

Therefore, the main objective of the present study is to investigate biofouling in the FDFO process by spiking *Pseudomonas aeruginosa PAO1+ GFP* in a synthetic wastewater feed, using three different fertilizers potassium nitrate (KNO_3), potassium chloride (KCl) and potassium dihydrogen phosphate (KH_2PO_4) as draw solutes. The reverse diffusion of these three different fertilizer draw solutes was determined and compared. The impact of reverse diffusion of the different fertilizers on flux decline, foulants composition and biomass accumulation on the FO membranes were also investigated. The study provides insights to the impact of different fertilizers on biofouling of FDFO membranes and corresponding fouling mechanisms. This may assist researchers to identify an appropriate fertilizer for the FDFO process from the perspective of limiting biofouling.

2. Materials and methods

2.1. Draw solution

KNO_3 and KH_2PO_4 fertilizers were used in this study to investigate the impact of reverse diffusion of N and P nutrients on biofouling in the FDFO process. All fertilizers were reagent grade (Sigma Aldrich). The DS was prepared by dissolving 1 M chemical fertilizer in deionized water. The osmotic pressure of these three fertilizers was calculated by using the Morse equation (extension of van't Hoff Equation) (Eq. (1)). The details of DS are provided in Table 1.

$$\pi = i\varnothing CRT \tag{1}$$

where, i is the number of ions produced during dissociation of solute, ϕ is the unit less osmotic coefficient, C is the molar concentration of all solutes (moles/L), R is the universal gas constant (0.083145 L bar/moles K), and T is the temperature on Kelvin scale (K).

2.2. Feed solution

Synthetic wastewater was prepared in this study to mimic municipal wastewater effluent and used as FS of the FO process. The composition of synthetic wastewater is shown in Table 2. This synthetic wastewater recipe was also used in a reported FO-AnMBR process for wastewater treatment [34], so that the results of this study can be compared with other studies. FO membrane fouling consists of inorganic scaling, organic and biological fouling [35–37]. Inorganic scaling due to nucleation and crystallization of calcium carbonate and magnesium carbonate on the membrane surface could happen when these species exceed saturation concentration. In this study, however, Ca and Mg scaling is less likely because their concentrations in the feed were only 1.45 ppm and 1.18 ppm, respectively, which is much lower than the saturation concentration. Even at an elevated concentrations (i.e. feed volume reduced from 1 L initially to 760 mL during batch process) due to rejection of Ca and Mg by the FO membrane, Ca and Mg concentrations at the end of each FO run were 1.92 and 1.56 ppm, respectively, and not adequate to cause significant scaling issues.

Table 1
Details of draw solutions used in this study.

Draw Solution	Mole concentration (M)	Osmotic pressure at 20 °C (bar)
KCl	1	44.8
KNO_3	1	32.6
KH_2PO_4	1	32.3