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Fouling and cleaning of high permeability forward osmosis membranes

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ABSTRACT

This study critically assesses the fouling behavior in forward osmosis (FO) when operating with new generation of high permeability commercial thin film composite (TFC) FO membranes. Low fouling behavior, commonly accepted in FO with the membrane active layer facing the fouling feed solution (AL-FS), is demonstrated to be mainly the consequence of operating at a low permeation flux (typically lower than 10 L m^{-2} h^{-1}). A higher water flux is observed to lead to severe fouling when using high permeability novel TFC membranes. Advanced membrane surface characterisation and FO fouling experiments over a range of initial fluxes (5–17 L m^{-2} h^{-1}) demonstrate that the intense fouling behavior of high permeability TFC membranes is mainly connected to the initial flux, whereas membrane surface properties only play a minor role. Like for any membrane processes, it is demonstrated that a (so-called critical) flux can be defined in FO systems operated in AL-FS, above which stable operation cannot be maintained over an extended period of time. At higher flux, the fouling cake deposed on the active layer creates additional hydraulic resistance to filtration and consequently significant flux decline is observed over time. A typical anti-fouling strategy, such as high cross flow velocity cleaning, is not efficient to sustain high flux, but fouled membranes can be successfully cleaned using extended osmotic backwashing.

1. Introduction

Due to the recent development of dedicated membranes, osmotically driven processes such as forward osmosis (FO) have demonstrated promising performances. The potential of FO to reduce the energy consumption of reverse osmosis (RO) in seawater desalination is of particular interest [1]. Practically, a specific configuration referred to as an FO–RO hybrid process has been suggested by several researchers as a promising method to combine water reuse and desalination [2–4]. In such hybrid system, FO is implemented as a pre-treatment step before RO, whereby the seawater is first pre-diluted in a controlled manner by impaired water, before being fed to the RO. As a result of the osmotic dilution, a lower driving force is needed in the RO system, lowering the overall energy consumption of the combined process compared to classical RO desalination. The FO–RO hybrid also offers a double-barrier protection, which could increase consumer confidence toward water recycling [4]. Membrane fouling remains a major concern of reusing impaired water with dense membrane processes and affects long term process efficiency as already widely studied and demonstrated for RO [5,6]. Lower fouling propensity in FO compared to RO is commonly reported in the literature, even when wastewater is used as feed [3]; However, most FO fouling studies have so far been performed with the cellulose triacetate (CTA) benchmark membrane from HTI (Albany, OR, USA), which features a relatively low permeation flux [7–10].

Recent studies have confirmed the need for higher permeation fluxes to support favorable FO economics and thus to encourage wider FO applications [11,12]. Consequently, many studies have focussed on the development of a new generation of membranes offering higher permeation fluxes. The intense research on improving membrane performance led to the development of thin film composite (TFC) FO membranes. Some of these membranes are commercially available [13] and present improved water permeability and higher salt rejection than the benchmark CTA membrane from HTI [14,15]. They are generally composed of a polyamide active layer that allows for a high selectivity, and a support layer

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which features limited internal concentration polarisation (ICP) for a more efficient use of the osmotic pressure [14,15]. As such, new opportunities to improve flux in FO process exist, although the impact of higher operating flux on fouling propensity is still to be fully evaluated.

The impact of the permeation flux on fouling behavior has been well studied in many pressure driven membrane processes [6]. Operating at high flux typically leads to enhanced fouling (i.e. decreasing flux over time during constant pressure operation); thus, lower flux operation is generally recommended for sustainable long-term filtration. The concept of critical flux [16] has been widely used to study the impact of flux on fouling for membrane bioreactors [17], but also for other hydraulic pressure driven membrane processes such as RO [18,19]. In FO systems, the occurrence of critical flux was mentioned in more recent studies using HTI CTA membranes, with the support layer facing the foulant feed solution and under elevated osmotic driving force [20,21]. One recent study on the rejection of trace organic contaminants using a commercial TFC membrane, also mentioned that at higher initial flux, the foulant layer was more compacted on the membrane surface [22]. However, the TFC membrane tested was not compared to benchmark CTA, its surface was not characterised and significant decrease of flux over time was observed even at the lowest initial permeation flux. Thus it is not clear to what extend initial flux and membrane surface properties contributed to fouling. Therefore, it is still necessary to assess the respective impacts of membrane surface properties and high FO flux operations, especially when using more challenging feed waters, and conditions such as expected in the FO–RO hybrid system, i.e. with the active layer facing the foulant feed solution and at moderate osmotic pressures.

Tackling fouling is a key aspect of membrane processes and is usually achieved via a combination of fouling mitigation (i.e. membrane and module development and/or optimisation of hydrodynamic conditions) and adapted cleaning strategies [23]. TFC membranes developed for FO have proven to initially enhance water permeation, but their much rougher surface was accounted for the more severe fouling observed [24,25]. Similar studies for NF and RO membranes highlighted that such behavior was related to the polyamide-based active layer of TFC membrane [26,27]. Thus, few membrane developments have recently been dedicated to fouling mitigation. Double skinned membranes were synthetised to allow operation of FO membranes with the active layer facing the draw solution (resulting in lower ICP and thus higher intrinsic fluxes), and demonstrated much lower fouling propensity than single skin membranes [28,29]. Membrane surface modification approaches using amine enriched, polyethylene-glycol enriched [28–30] and silver–titanium nanoparticles [31] also demonstrated significantly lower organic fouling behavior. Promising results have been observed, but studies remain scarce and limited to lab-scale and lab-made membranes. So far, no fouling studies related to commercially available TFC FO membranes with anti-fouling properties have been performed.

During FO operation at low permeation flux, low fouling behavior is generally observed and as a consequence, no specific fouling mitigation strategies have been developed. Typically, FO is operated at low cross flow velocities (CFV) and simple physical methods to increase turbulence during cleaning (i.e. high CFV, use of spacers or pulsed flow) are mostly sufficient to restore flux back to its original level after fouling [7]. Operating at higher permeation flux is expected to generate more intense fouling behavior (flux and/or pressure driven) and may require adapted anti-fouling strategies. Implementing complex or expensive cleaning protocols such as air scouring or chemical cleaning may be a serious drawback to the economic sustainability of FO. Osmotic backwash, extensively studied in the RO literature [32–37] may represent an adapted strategy to clean FO membranes [36,38].

There is therefore a clear need to further investigate fouling phenomena during high flux FO operation. This study provides, for the first time, a comparative assessment of fouling behavior of four commercially available membranes from HTI and Porifera Inc. (Hayward, CA, USA). In addition to membrane characterisation, a detailed study was performed to determine (1) the impact of initial flux on fouling propensity, (2) the impact of membrane surface properties, (3) the impact of physical cleaning strategies.

2. Material and methods

2.1. Membranes

Four commercially available FO membranes were used in these experiments: two from HTI (i.e. flat-sheet CTA and TFC FO membranes) and two TFC from Porifera Inc. with hydrophilic support layer (i.e. Porifera-FO and Porifera-FOm). Specific surface modification treatment to lower the fouling behavior has been applied on Porifera FOm membrane. After reception, membrane samples were stored at 4 °C and soaked in Deionised (DI) water for at least 1 h before use. No pre-compaction was applied.

The membranes pure water permeability (A) and salt permeability (B) were measured in a bench scale RO cross flow setup at 25 °C with a CFV of 0.3 m s^{-1}. This rig is referred to as "RO setup" from now on and was further described in our former publication [39]. Hydraulic pressure (ΔP) was applied on the FO membrane with active layer facing the feed solution (AL-FS) membrane orientation in the range of 2–6 bar. As described in [39], water and salt permeabilities vary depending on membrane support and as a consequence of membrane deformation. Therefore, A_0 and B_0 described hereafter were estimated value of pure water and salt permeability at 0 bar (based on average value of intercept obtained for both RO feed spacer and permeate spacers used as membrane support) [39]. Membrane resistance to filtration was calculated as in Eq. (1):

$$R_m = \frac{\Delta P}{\mu J_w} \quad (1)$$

With J_w the water permeation flux and μ the Newtonian dynamic viscosity. R_m can also be expressed as in Eq. (2):

$$R_m = \frac{1}{\mu A_0} \quad (2)$$

The structural parameter (S_0) value was evaluated based on FO tests using the setup described in our former work [39], DI water as feed solution and draw solution prepared based on dry Red Sea salts (RSS, Red Sea Inc., Isr) and concentrated at 35 g L^{-1} in DI water. The S_0 value for each membrane was calculated by implementing experimental flux obtained during FO tests as well as the A_0 and B_0 values in Eq. (3) adapted from [40,41]:

$$S_0 = \frac{D}{J_w} \ln \times \left(\frac{B_0 + A_0 \pi_D}{B_0 + J_w + A_0 \pi_F} \right) \quad (3)$$

With D the diffusivity of the draw solute, π_F and π_D the osmotic pressure of feed and draw solutions respectively (here π_F is equal to zero since DI water was used as feed).

Membrane surface morphology and surface roughness analysis were performed using an Icon atomic force microscope (AFM) (Bruker, CA, USA) in Scanasyst mode using an OTESPA probe. Dry membrane samples were mounted on a specimen holder and an image of a 10 × 10 mm area was scanned. Surface roughness of a 5 × 5 mm surface area was reported in terms of average roughness (R_a) values.

Membrane zeta potentials were determined from streaming potential measurements using a SurPASS Electrokinetic Analyzer from Anton Paar GmBH (Graz, Austria). All streaming-potential