



## Organic fouling mechanisms in forward osmosis membrane process under elevated feed and draw solution temperatures



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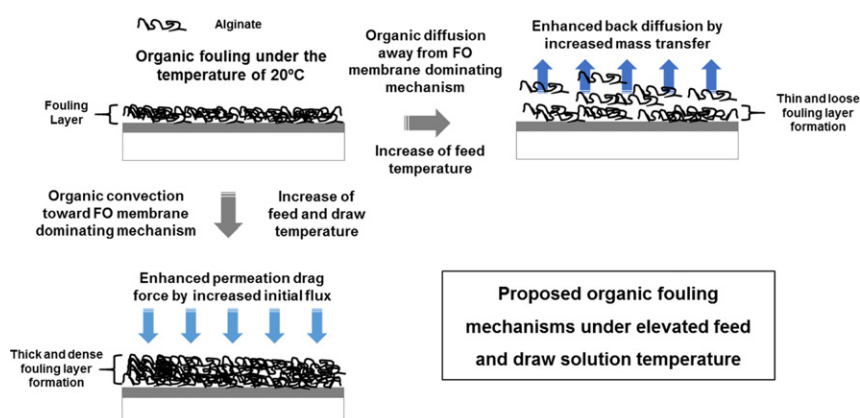
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### HIGHLIGHTS

- Organic fouling mechanism in FO was investigated at varying operating temperatures.
- Water flux and reverse salt flux were varied with increasing temperature.
- Enhanced mass transfer by increased feed temperature reduced organic fouling.
- Higher permeation drag by increased draw temperature increased organic fouling.
- Fouling was determined by interplay between convection and diffusion by temperature.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 10 June 2014

Received in revised form 24 October 2014

Accepted 28 October 2014

Available online 5 November 2014

#### Keywords:

Forward osmosis

Organic fouling

Draw and feed solution temperature

Organic convection and diffusion

Temperature polarization

### ABSTRACT

Organic fouling mechanisms in forward osmosis (FO) were systematically investigated at varying feed and draw temperatures. The effect of temperature variation on FO performance was first examined without foulants. When draw temperature increased, internal concentration polarization (ICP) decreased, which caused water flux to increase. Water flux was also improved with increasing feed temperature due to water permeability increased by decreased viscosity. Thus it can be deduced that water flux enhancement was induced by combined effects of reduced ICP and enhanced water permeability. A series of fouling experiments was then elaborately designed to fundamentally elucidate organic fouling mechanisms. Results demonstrated that organic fouling was significantly influenced by convective and diffusive organic transports induced by increasing temperature. Faster flux decline was observed with increasing draw temperature, primarily due to increased permeation drag. When increasing feed temperature, FO membrane was less fouled, attributing to enhanced organic back diffusion from membrane surface as well as increased organic solubility. Furthermore, fouling became more severe above certain critical flux at which organic convection by permeation drag dominated fouling mechanism as seen with escalating both temperatures simultaneously. Findings from this study can be utilized beneficially when FO temperature may be varied and needs to be optimized.

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## 1. Introduction

Reverse osmosis (RO) membranes have been employed in a wide range of applications including seawater and brackish water desalination as well as drinking and industrial water treatment [1–3]. However, the operation of RO plants is hampered by high energy consumption (i.e., typically above 4–5 kWh/m<sup>3</sup>), high fouling potential, and inherent limitations such as low recovery [4–8]. Recently, forward osmosis (FO) is being considered as a supplement for a low-energy, low-fouling, and high recovery process to conventional RO desalination. In the FO membrane process, a highly concentrated draw solution is utilized to induce the driving force of separation, and then is recovered at significantly reduced energy cost and fouling level [9–11].

For the effective reconstitution of draw solutes, FO is often integrated with thermally driven processes such as membrane distillation (MD), possibly using waste heat from power-generating and industrial manufacturing plants [12–19]. In such systems, FO may be operated at the elevated draw solution temperature, depending on the configuration and application. High feed water temperature can be also encountered when the FO membrane process is, for instance, applied for the treatment of shale gas flow back and produced waters of which temperature is typically above 50 °C–70 °C due to geothermal heat [12,17,20]. The basic performance and fouling behavior of the FO membrane process may be significantly altered when it is operated at elevated draw and feed solution temperatures.

Only limited studies have explored the effect of temperature on the FO process [21–24]. For example, Xie et al. reported that the dilution effect induced by higher water flux and the hindrance effect enhanced by higher reverse salt flux (RSF) led to an increase in the rejection of neutral trace organic contaminants at higher draw solution temperature [23]. Phuntsho et al. explained the influence of temperature on the FO process through changes in solution thermodynamic properties and various concentration polarization effects [24]. However, there is no study available in the current literature, which elucidates FO fouling mechanisms influenced by both draw and feed operating temperatures, fundamentally and technically.

Membrane fouling in the FO process is a complex phenomenon which is affected by the combined effect of several factors including operating modes and hydrodynamics, membrane and foulant characteristics, feed water solution chemistry, and draw solution properties [10,11,25–30]. In an early study by Mi and Elimelech [27], the role of various physical and chemical interactions, such as intermolecular adhesion forces, calcium binding, initial permeate flux, and membrane orientation, was first tackled in the organic fouling of FO membranes. The critical concentration concept was also suggested for FO process, indicating that relatively stable flux could be achieved when the concentration of draw solution was adopted below a critical value [25]. A recent study by Kim et al. [29] demonstrated that the absence of hydraulic pressure in the FO systems, compared to pressurized RO systems, was the main reason for better fouling reversibility by investigating fouling mechanisms of organic–colloidal aggregates. The reverse diffusion of draw solutes might also affect the fouling tendencies of FO processes [10,11]. RSF was reported to contribute significantly to flux decline through accelerated CEOP (cake enhanced osmotic pressure), in which the draw salts diffused were captured by the fouling layer and thus accelerated CEOP [10]. Despite the recent efforts to understand fouling mechanisms in the FO process, feed and draw solution temperature and/or their differences have not been investigated at all, in particular for organic fouling in the FO membrane process.

The main objective of this study is to fundamentally investigate organic fouling mechanisms in FO process under varying feed and draw temperatures. The effect of elevated solution temperatures on FO performance was first examined thoroughly without organic foulants. A series of fouling experiments were then carefully designed and conducted to assess the influence of operating temperature on organic fouling in the FO process. Lastly, organic fouling mechanisms induced by elevating

feed and draw temperatures was delineated and elucidated by analyzing the results obtained from performance and fouling tests. It is expected that findings from this work provide valuable information to design and operate FO process for such applications where feed and draw solution temperatures need to be varied and optimized.

## 2. Materials and methods

### 2.1. FO membrane

The FO membrane used in this study was provided by Hydration Technologies Innovations (Albany, OR, USA). This membrane is made of cellulose-based polymers with an embedded polyester mesh for mechanical strength. The total thickness of the membrane is approximately 50 μm. A detailed description of the structure and properties of the membrane can be found elsewhere [31].

### 2.2. FO membrane characterization

Properties commonly characterizing FO membranes are classified into the water permeability ( $A$ ) and salt permeability ( $B$ ) of the active (rejecting) layer, and the structure parameter ( $S$ ) of the support layer. The  $A$  and  $B$  coefficients were characterized based on the standard methodology suggested by Cath et al. [32]. The  $A$  and  $B$  coefficients were measured using deionized (DI) water and 2000 mg/L NaCl, respectively. Experimental measurements were conducted in a lab-scale RO unit modified for both feed and permeate flows. Feed and permeate temperatures were set at 20 °C, 35 °C, and 50 °C. The cross-flow velocity was maintained at 25 cm/s. Prior to each measurement, the membranes were compacted at 15 bar with DI water for at least 12 h until the permeate water flux had stabilized. Water flux was measured at 10 bar and the water permeability coefficient was determined by dividing water flux by the applied hydraulic pressure (Eq. (1)). NaCl was then added to the feed solution at a concentration of 2000 mg/L to determine the salt permeability coefficient at 10 bar. The RO system was stabilized for 2 h. The permeate water flux with 2000 mg/L NaCl solution was recorded, and the feed and permeate samples were taken to determine the observed NaCl rejection according to Eq. (2). The salt permeability coefficient was determined based on Eq. (3).

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

$$R = 1 - c_p/c_f \quad (2)$$

$$B = J_w \left( \frac{1-R}{R} \right) \exp \left( -\frac{J_w}{k} \right) \quad (3)$$

where  $A$  is the water permeability coefficient of the membrane,  $J_w$  is the water flux,  $\Delta P$  is the transmembrane pressure,  $R$  is the observed rejection,  $c_p$  is the concentration of the permeate,  $c_f$  is the concentration of the feed,  $B$  is the solute permeability coefficient of the membrane, and  $k$  is the mass transfer coefficient for the cross-flow channel of the RO membrane cell.

The  $S$  parameter was obtained based on the simplified equation (Eq. (4)) for water flux derived by McCutcheon and Elimelech [33]. For our experiments with DI water feed ( $\pi_{F,d} = 0$ ), this equation was further simplified and the  $K$  parameter (i.e., the resistance to diffusion within the porous support layer) was calculated as presented in Eq. (5). The  $S$  parameter was then simply determined by multiplying this  $K$  parameter by the solute diffusivity (Eq. (6)).

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