



Modeling salt accumulation in osmotic membrane bioreactors: Implications for FO membrane selection and system operation

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ARTICLE INFO

Article history:

Received 21 July 2010

Received in revised form 7 October 2010

Accepted 9 October 2010

Available online 15 October 2010

Keywords:

Salt accumulation

Forward osmosis (FO)

Osmotic membrane bioreactor (OMBR)

Solute reverse diffusion

Volumetric concentration factor

ABSTRACT

Novel osmotic membrane bioreactors (OMBRs) have been recently reported in the literature. An OMBR uses a dense salt-rejecting forward osmosis (FO) membrane, which exhibits high retention of organic matter and various other contaminants. Meanwhile, the high rejection nature also leads to the accumulation of salts in the bioreactor, which can adversely affect the biological activities as well as the FO water flux. A salt accumulation model is developed in the current study. Our model suggests that both the bioreactor salt concentration and the FO water flux are controlled by membrane properties (water permeability A , salt permeability B , mass transfer coefficient K_m , and membrane orientation relative to the draw solution) and the OMBR operational conditions (salt concentration of the influent wastewater, draw solution concentration, hydraulic retention time (HRT), and sludge retention time (SRT)). The salt accumulation is contributed by both the influent wastewater and the reverse diffusion of solutes from the draw solution, and is directly proportional to the volumetric concentration factor (i.e., the SRT/HRT ratio). The relative importance of reverse diffusion over contribution from influent solutes is governed by the membrane selectivity. For a relatively selective membrane ($B/A \ll$ the osmotic pressure of the influent water), solute reverse diffusion has negligible effect on OMBR performance. In contrast, the salt accumulation and FO water flux reduction are governed by reverse diffusion for B/A greater than the osmotic pressure of the influent water. The current study reveals the critical importance of the B/A ratio and HRT/SRT ratio for optimized OMBR operation.

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1. Introduction

Forward osmosis (FO) has emerged as an alternative membrane process to the conventional pressure-driven membrane processes in the recent years [1,2]. Unlike the pressure-driven processes that rely on a hydraulic pressure to “push” water through a membrane, the FO process is driven by an osmotic pressure difference across a semi-permeable dense membrane that retains solutes but allows water to pass through the membrane [2,3]. Pure water permeates through the FO membrane spontaneously from the low-osmotic-pressure solution (i.e., the feed water (FW)) to the high-osmotic-pressure solution (i.e., the draw solution (DS)) without the need of an externally applied pressure [2,3]. The elimination of the hydraulic pressure in FO means that a significantly lower amount of prime energy (i.e., electricity) is required to run an

FO process compared to that used for a typical reverse osmosis (RO) process. Thus, FO can be highly attractive where a high pressure osmotic DS is naturally available (e.g., seawater) or if the DS can be regenerated cost-effectively. On the other hand, like RO membranes, dense FO membranes have nearly complete rejections against a wide range of contaminants including dissolved salts and organic matter [4]. Consequently, FO has found many potential applications in water purification [5], wastewater treatment [6,7], seawater desalination [8] and osmotic power generation (using a derivative process known as pressure retarded osmosis, see Refs. [3,9,10]).

A novel type of FO-based membrane bioreactor (MBR) has been recently reported in the literature [7,11–13]. Unlike the conventional MBRs that typically use porous microfiltration (MF) or ultrafiltration (UF) membranes to retain biomass, the osmotic MBR (OMBR) system uses an FO membrane in the bioreactor [12]. With a high-concentration draw solution, pure water is extracted from the mixed liquor and diffuses across the membrane into the DS, while contaminants from the feed wastewater are nearly completely rejected. If wastewater reclamation is needed, pure water can be obtained while the draw solution is also regenerated

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via a downstream process such as thermal distillation [8]. Where a low grade heat source is available, this treatment scheme can be potentially attractive [14]. Compared to the conventional MBRs, the OMBR offers much higher rejection (dense FO membranes versus porous MF/UF membranes) at a lower hydraulic pressure [4,7]. The better retention of organic matter by FO may lead to 1) improved biological degradation of recalcitrant organics due to the prolonged retention time for organic matter, 2) reduced dissolved organic content in treated effluent, and 3) a likely enhanced removal for micropollutants [4,7]. Several studies have also suggested that FO is a low-fouling alternative to pressure-driven membranes due to the lack of hydraulic pressure [4,12], though more systematic studies are still required to investigate the underlying fouling mechanism(s) [15,16].

Despite the many advantages offered by OMBRs compared to conventional MF- or UF-based MBRs, the high salt rejection property of the FO membrane also introduces a unique new challenge – the dissolved salts rejected by the FO membrane will accumulate inside the bioreactor. In addition, solute will also diffuse from the high concentration DS into the bioreactor under the concentration gradient across the FO membrane [15,17]. This will further enhance the salt accumulation in the bioreactor. The elevated salt concentration in the bioreactor not only reduces the FO permeate flux (due to reduced the effective osmotic driving force and increased internal concentration polarization (see Refs. [15,18] and Section 2)) but also may adversely impact the physical and biological activities in the bioreactor [13]. Thus, a systematic study on the salt accumulation in the novel OMBR system is warranted.

This study aims to investigate the salt accumulation behavior in OMBR and its effect on FO performance. A theoretical model is developed based on the internal concentration polarization theory as well as the solute mass balance for the bioreactor. Model verification was performed via independent bench scale FO experiments. The salt accumulation model was then applied to simulate the OMBR performance under various conditions, and the effects of membrane properties and operational conditions were systematically investigated to reveal the consequent constraints on membrane selection as well as reactor design and operation. This study focuses on the effect of salt accumulation on the FO permeate flux behavior. On the other hand, membrane fouling [15] and biological activities [13] are beyond the scope of the current study. To the authors' best knowledge, this is the first systematic study to model the salt accumulation in OMBR and its implications for OMBR design and operation. While the model is developed primarily for OMBR, the basic principles applied in the model can also be extended to other osmotic reactors.

2. Model development

2.1. Internal concentration polarization and modeling FO water flux and solute reverse transport

Numerous studies have reported on the experimental testing and modeling of FO water flux [15,19–21]. These studies revealed that the typical FO flux is significantly lower than the ideal flux determined from the apparent driving force (osmotic pressure difference between the DS and FW) using the solution-diffusion model alone. Such non-ideal flux behavior is caused by concentration polarization of solutes inside the porous support layer (also known as the internal concentration polarization or ICP) [15,18,19]. For example, when the active rejection layer is oriented towards the draw solution (the AL-DS membrane orientation), solutes from the feed water is accumulated in the porous support layer due to the high retention nature of the FO rejection layer. Meanwhile, some solutes also diffuse through the rejection layer from the high concentration DS into the support. This causes the solute concentration

inside the support layer to be much greater than the bulk feed water concentration (concentrative ICP, refer to Fig. A1 in Appendix A), which results in reduced effective driving force (the osmotic pressure difference across the active rejection layer) as well as lower permeate flux. Similar reduction in FO water flux also happens for the active-layer-facing-FW (AL-FW) orientation due to the dilution of draw solution inside of the porous support (dilutive ICP, refer to Fig. A1 in Appendix A). The ICP phenomenon was first modeled by Lee et al. [19] in the context of pressure retarded osmosis. A simplified version of ICP model was later developed by Loeb et al. [1] for forward osmosis:

$$J_v = K_m \ln \left(\frac{A\pi_{\text{draw}} - J_v + B}{A\pi_{\text{feed}} + B} \right) \quad (\text{AL-DS, concentrative ICP}) \quad (1)$$

and

$$J_v = K_m \ln \left(\frac{A\pi_{\text{draw}} + B}{A\pi_{\text{feed}} + J_v + B} \right) \quad (\text{AL-FW, diluted ICP}) \quad (2)$$

where J_v is the volumetric flux of water; K_m is the mass transfer coefficient for the membrane porous support layer; A and B are the water permeability and solute permeability of the rejection layer, respectively; and π_{draw} and π_{feed} are the osmotic pressure of the draw solution and that of the feed water, respectively. It has been demonstrated that the AL-FW orientation suffers greater ICP level and consequently its water flux is lower than that of the AL-DS orientation under identical testing conditions [15,22,23]. In addition, ICP is more severe for higher draw solution concentrations, higher feed water concentrations, as well as lower mass transfer coefficient values [15,22,23].

The mass transfer coefficient K_m in Eqs. (1) and (2) can be related to the solute diffusion coefficient D and the membrane structure parameter S by [1,15]:

$$K_m = \frac{D}{S} \quad (3)$$

where the membrane structural parameter S is a property of the FO support layer, and it is defined as the product of membrane support layer thickness (l) and tortuosity (τ) over its porosity (ε) [1,15]:

$$S = \frac{\tau l}{\varepsilon} \quad (4)$$

Loeb's ICP model (Eqs. (1) and (2)) has been widely adopted by other research groups and has been verified experimentally for a wide range of membranes including both cellulose acetate asymmetric membranes and thin film composite membranes [15,18,20,22,23]. For high draw solution concentration and high feed water concentration, the osmotic pressure terms ($A\pi_{\text{draw}}$ and $A\pi_{\text{feed}}$) in Eqs. (1) and (2) can be orders of magnitude higher than J_v and B . Under such special conditions, the water flux in the two orientations (AL-DS and AL-FW) become nearly identical and it is determined by [1,22]:

$$J_v = K_m \ln \left(\frac{\pi_{\text{draw}}}{\pi_{\text{feed}}} \right) \quad (\text{both AL-DS and AL-FW}) \quad (5)$$

Similar to the water flux J_v , the solute flux J_s is also strongly affected by ICP [15,17]. Tang and coworkers [15,22,23] demonstrated that J_s is directly proportional to J_v according to the following equation:

$$\frac{J_s}{J_v} = \frac{B}{A\beta R_g T} \quad (\text{both AL-DS and AL-FW}) \quad (6)$$

where β is the van't Hoff coefficient, R_g is the universal gas constant, and T is the absolute temperature. In Eq. (6), the J_s/J_v ratio is largely determined by B/A (in unit of Pa). The B/A ratio is an important property of the membrane rejection layer. This ratio can be considered as a constant for a given rejection layer and solute type at fixed temperature. In general, a lower B/A ratio (i.e., a more

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