



# Polydopamine coating on a thin film composite forward osmosis membrane for enhanced mass transport and antifouling performance

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## ABSTRACT

We applied a polydopamine (PDA) coating on a thin film composite (TFC) forward osmosis (FO) membrane and investigated the effects of coating on FO mass transport and antifouling behavior. The PDA coating significantly improved membrane surface hydrophilicity as well as reduced membrane surface roughness. Using a short PDA coating duration of 0.5 h, the coated membrane TFC-C0.5 achieved enhanced FO water flux and reduced reverse solute diffusion simultaneously. The reduced reverse solute diffusion can be attributed to the enhanced membrane selectivity: TFC-C0.5 had better rejection and similar water permeability compared to the original TFC membrane. This reduction in reverse solute diffusion further reduced the internal concentration polarization inside the coated membrane, leading to an enhanced FO water flux. Nevertheless, longer PDA coating duration of 1–4 h resulted in reduced FO water flux due to the significantly increased hydraulic resistance of the coated membranes. The PDA coated membrane TFC-C0.5 also presented an improved antifouling performance compared to the control membrane using alginate as a model foulant. Our results reveal the great room for the development of effective coating materials in FO: a well-designed coating with high selectivity and low hydraulic resistance can improve solute rejection, reduce reverse solute diffusion, mitigate internal concentration polarization and enhance FO water flux in addition to control fouling. Such unprecedented opportunities break the traditional trade-off between water flux and antifouling performance when coating pressure driven reverse osmosis membranes.

## 1. Introduction

Forward osmosis (FO) is a membrane process using a draw solution with high osmotic pressure to extract water from a feed solution with low osmotic pressure [1,2]. In comparison with pressure-driven membrane processes such as reverse osmosis (RO) and nanofiltration (NF), FO requires lower energy input. This feature enables it being a potential alternative to address many water issues, such as seawater desalination [3,4], wastewater treatment [5–7], and power generation (i.e., by pressure retarded osmosis) [8–10]. Despite the promising applications of FO, its performance can still be significantly limited by membrane fouling [11–14] and mass transport limitations [15,16].

Surface modification is considered as a feasible approach for improving membrane antifouling performance [17–19]. One of the frequently used modification approaches is a mussel-inspired polydopamine (PDA) initiated coating [20–23]. PDA coating layer can be formed through the self-polymerization of dopamine on various substrates [24]. It can improve membrane surface hydrophilicity to reduce

fouling [25,26]. Therefore, PDA coating has been extensively investigated as an antifouling coating for pressure-driven membrane processes (e.g. RO, NF, and ultrafiltration) [20,27,28]. In recent years, PDA coating was also introduced on FO membranes to improve membrane performance. Arena et al. [29,30] reported the use of PDA for improving the hydrophilicity of membrane support layers, which effectively reduced internal concentration polarization (ICP) and improved water flux. Han et al. [31] also reported that PDA-modified substrate layer can enhance membrane performance in FO. Nevertheless, there is few study focusing on the use of surface coating (e.g. PDA coating) to modify the rejection layer of thin film composite (TFC) FO membranes. The transport phenomena (e.g. water transport and reverse solute diffusion) as well as the fouling behavior in the presence of a thin coating layer are still unknown in FO. Therefore, it is worthwhile to study the separation performance and antifouling behavior of a coated membrane and further elucidate the underlying mechanisms, since surface coating has been regarded as an important approach for enhancing membrane performance [17,28,32].

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In this work, we applied a PDA coating on a commercial TFC FO membrane from Hydration Technology Innovations (HTI). Membrane separation performance and antifouling behavior were systematically investigated under various PDA coating conditions. The results will provide mechanistic understanding on the influence of surface coating on FO membrane performance, and may open new insight for the design of effective surface coating for FO membranes.

## 2. Materials and methods

### 2.1. Membranes and chemicals

The TFC FO membranes used in this study were provided by HTI (Albany, OR). This membrane consists of a polyamide rejection layer and a porous supporting layer embedded on a polyester mesh [33].

Unless specified otherwise, all the chemicals used in this study are analytical grade. Deionized (DI) water was used for the preparation of all solutions. Dopamine hydrochloride (J&K Scientific Ltd.) and tris (Acros Organics, Geel, Belgium) were used to form PDA coating layer. Sodium chloride (Uni-Chem) was used to prepare draw solution (DS) and feed solution (FS). Sodium hydroxide (Uni-Chem), and hydrochloric acid (37 wt%, VWR, Dorset, U.K.) were used to adjust solution chemistry. Sodium alginate and calcium chloride were supplied by Sigma-Aldrich (St. Louis, MO) and used in fouling experiments.

### 2.2. Preparation of PDA coating

The preparation of PDA coating has been described in details in our previous work [26]. Briefly, a clean membrane coupon was placed in a container with only the rejection layer exposed in the coating solution. A 150 mL solution containing 0.2 wt% dopamine chloride and 10 mM tris at pH 8.5 was subsequently added to the container. The coating was performed under moderate shaking for a predetermined duration (0.5, 1, and 4 h). The coated membranes were denoted as TFC-C0.5, TFC-C1, and TFC-C4. All the coated membranes were thoroughly rinsed by DI water to remove unreacted residues before further testing.

### 2.3. Membrane characterization

A field-emission scanning electron microscope (FE-SEM, LEO 1530) was used to characterize membrane surface morphology. Dried membrane samples were sputter-coated with a thin layer of gold (BAL-TEC SCD 005). SEM was operated at an acceleration voltage of 5.0 kV. An attenuated total reflectance Fourier transformation infrared spectrometer (ATR-FTIR, Perkin Elmer Spectrum 100) was employed to characterize membrane surface functional groups over a wavenumber range from 650 to 4000  $\text{cm}^{-1}$ . An atomic force microscope (AFM, JPK Nano Wizard AFM) was used to resolve membrane surface roughness. A contact angle goniometer (OCA20, Dataphysics) was applied to determine water contact angles of membranes using a sessile drop method at 25 °C. The reported value of contact angle is an average value of ten replicates. A zeta potential analyzer (SurPASS, Anton Paar GmbH) was used to evaluate membrane surface charge with an adjustable gap cell and using 10 mM NaCl as background solution over a pH range from 3 to 10.

Membrane intrinsic properties including pure water permeability and solute permeability of the TFC FO membrane were evaluated in a pressurized RO mode using a lab-scale cross-flow filtration setup [26]. Briefly, a membrane coupon was placed in a filtration cell (CF042, Sterlitech) with an effective membrane area of 42  $\text{cm}^2$ . A 10 L feed solution (i.e., DI water or 10 mM NaCl) was then recirculated for 12 h at 10 bar with a cross-flow velocity of 22.4  $\text{cm/s}$  to pre-compact the membrane. Pure water permeability,  $A$ , and solute permeability,  $B$ , are determined by [34]

$$A = \frac{J_{v,RO}}{\Delta P - \Delta\pi} \quad (1)$$

$$B = \left(\frac{1}{R_{RO}} - 1\right) \times J_{v,RO} \quad (2)$$

where  $J_{v,RO}$  ( $\text{L m}^{-2} \text{h}^{-1}$ ) is the water flux under RO mode,  $\Delta P$  (bar) is the hydraulic pressure difference across the membrane,  $\Delta\pi$  is the osmotic pressure difference across the membrane, and  $R_{RO}$  is the solute rejection.

### 2.4. FO system

The FO membrane rejection tests and fouling experiments were conducted on a bench-scale cross-flow FO filtration system (Appendix A). An FO membrane coupon was placed in a cross-flow FO cell (CF042-FO, Sterlitech, effective membrane area of 42  $\text{cm}^2$ ). Diamond-patterned spacers were placed on the both sides to provide support for membrane and improve mass transfer [15,34]. Two gear pumps were used to recirculate the feed solution (FS, of 1.5 L 10 mM NaCl) and draw solution (DS, of 1.5 L NaCl over a concentration range of 0.5–2 M), respectively. The flow rates of both FS and DS were  $\sim 0.15 \text{ L/min}$ . Water flux was determined at specific time intervals by measuring the weight of the feed tank with a digital balance connected to a data recording program. The conductivity of the feed solution was monitored with a benchtop conductivity meter. The salt rejection,  $R_{FO}$ , in FO was defined as [35]

$$R_{FO} = 1 - \frac{J_s}{J_v C_{fs}} \quad (3)$$

where  $J_s$  ( $\text{g m}^{-2} \text{h}^{-1}$ ) is reverse solute flux,  $J_v$  ( $\text{L m}^{-2} \text{h}^{-1}$ ) is water flux in FO, and  $C_{fs}$  is the solute concentration in the feed solution.  $J_s/J_v$  is defined as the specific reverse solute diffusion.  $J_s$  was obtained as the slope of plotted  $C_{fs,t}(V_{fs,0} - J_v A_m t)/A_m$  versus  $t$ , where  $C_{fs,t}$  ( $\text{g L}^{-1}$ ) is the solute concentration in FS at time  $t$  (h),  $V_{fs,0}$  is the initial volume of FS (L), and  $A_m$  is the effective membrane area ( $\text{m}^2$ ).

Fouling experiments were conducted with a FS containing 10 mM NaCl, 20 mg/L sodium alginate, and 1 mM  $\text{CaCl}_2$ . Prior to the fouling stage, FO membrane was pre-equilibrated with foulant-free FS and DS for 0.5 h. Subsequently, bulk alginate solution and calcium solution were spiked to the FS to reach the targeted concentration. The fouling experiment was then continued for 6 h.

## 3. Results and discussion

### 3.1. Membrane surface properties

The surface morphology of various membranes were characterized by SEM (Fig. 1). The control membrane shows an appearance of typical “ridge and valley” structure (Fig. 1a), a characteristic of fully aromatic polyamide RO and NF membranes [36,37]. The average roughness was  $\sim 48.9 \text{ nm}$  (Fig. 2a), which is comparable to the reported roughness of polyamide-based TFC membranes [26,38]. The PDA coated membranes showed different morphology (Fig. 1b–d) where the polyamide layer was partially or fully covered. Correspondingly, surface roughness also slightly reduced to 36.1 nm of TFC-C0.5, 41.6 nm of TFC-C1, and 42.3 nm of TFC-C4.

Fig. 3a presented the surface charge of control and coated membranes. There was no significant difference between the control membrane and TFC-C0.5 while TFC-C1 and TFC-C4 presented more negative surface. Water contact angles decreased significantly from  $\sim 42^\circ$  of control membrane to  $\sim 25\text{--}29^\circ$  of PDA coated membranes (Fig. 3b), implying that the membrane surface became more hydrophilic with PDA coating. The improvement of hydrophilicity together with the decreased surface roughness might be beneficial to membrane anti-fouling [25].

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