

# Fast polydopamine coating on reverse osmosis membrane: process investigation and membrane performance study

Jianqiang Wang<sup>a,b</sup>, Hao Guo<sup>b</sup>, Xiaonan Shi<sup>b</sup>, Zhikan Yao<sup>b</sup>, Weihua Qing<sup>b</sup>, Fu Liu<sup>a</sup>, Chuyang Y. Tang<sup>b\*</sup>

a. Polymer and Composite Division, Ningbo Institute of Material Technology & Engineering, Chinese Academy of Sciences, Ningbo, P.R. China.

b. Department of Civil Engineering, The University of Hong Kong, Hong Kong, P.R. China.

\*Corresponding author: phone, +852 2859 1976; fax, +852 2559 5337; e-mail, [tangc@hku.hk](mailto:tangc@hku.hk)

## **Abstract**

We report a novel membrane surface modification method using a fast polydopamine coating (fPDAC) strategy. Specifically, NaIO<sub>4</sub> was introduced in the coating process to accelerate the polydopamine deposition rate. Surface properties and separation performances of fPDAC-coated reverse osmosis membranes were characterized and compared to those obtained using the conventional slow polydopamine coating (sPDAC) strategy. Quartz crystal microbalance measurements showed greatly increased polydopamine deposition rate using the fPDAC method, resulting in a reduction of 97% coating time to reach an areal mass of 2000 ng/cm<sup>2</sup>. Both fPDAC and sPDAC enhanced the surface hydrophilicity and reduced the membrane surface charge. At relatively low areal mass deposition (< 1000 ng/cm<sup>2</sup>), fPDAC-coated membranes showed improved NaCl rejection together with only mild loss of pure water flux. Nevertheless, this rejection enhancement effect was not noticeable when extensive polydopamine coating was applied due to the undesirable cake-enhanced concentration polarization effect. The extensive polydopamine coating was further accompanied with severe loss of membrane permeability, suggesting that shorter coating time (e.g., 4 min) is preferred using the fPDAC method. Our study provides a more rapid and effective membrane surface coating method compared to the conventional sPDAC method.

**Key words:** Polydopamine; Reverse osmosis; Coating

## **1. Introduction**

Polydopamine (PDA), an analogue of mussel adhesive protein, has attracted great attention due to its unique properties of adhesion, hydrophilicity, antifouling and versatility for post-functionalization [1-3]. PDA has been widely used as a coating material for membrane surface modification to enhance its surface hydrophilicity and separation performances through a dopamine polymerization process in a weak alkaline solution (normally pH=8.5) [4-10]. Although it is often considered facile and green [11, 12], the conventional PDA coating method is time consuming (often requiring several hours or even a few days [12]). Such long coating duration presents a critical barrier to its cost-effective implementation. Therefore, a fast polydopamine coating method is highly desirable.

Conventional PDA coating is achieved by the oxidation of the catechol groups of dopamine monomers and their subsequently polymerization [13, 14], where oxygen is used as the oxidant [1]. The relatively low dissolved oxygen concentrations in aqueous solution often limit the growth of the PDA coating. Therefore, much efforts have been devoted to improving the dopamine polymerization rate using alternative oxidants (e.g., pure oxygen [15]). Recently, alternative strategies have been proposed for fast dopamine polymerization, such as field induced acceleration (UV irradiation and electrochemical actuation [16-18]) and the use of stronger oxidants (ammonium persulfate [5, 19],  $\text{CuSO}_4/\text{H}_2\text{O}_2$  system [20],  $\text{FeCl}_3/\text{H}_2\text{O}_2$  [21] and sodium periodate [22]). Among these strategies, PDA produced using sodium periodate as the oxidant has the eumelanin-like character that best resembles the structure formed by conventional PDA coating [23]. This rapid coating method has a polymerization rate of approximately two orders of magnitudes faster than the conventional method. To the best knowledge of the authors, up to date, there is no report on the application of this rapid PDA coating technique in the context of membrane surface modification.

In this study, we report sodium periodate induced dopamine polymerization to prepare a fast PDA coating on a reverse osmosis membrane. The morphology, surface chemistry and the separation performance of the coated membrane was systemically investigated. Our study may open a new avenue for highly efficient membrane surface modification by PDA coating.

## 2. Materials and methods

### 2.1 Chemicals

Tris (hydroxymethyl) aminomethane (Tris, 99+%) was purchased from ACROS Organics. Dopamine hydrochloride (99%) was purchased from Alfa Aesar. A commercial RO membrane (XLE, Dow FilmTec) was used for the base membrane for polydopamine coating. The XLE membrane exhibits NaCl rejection (2000 mg/L) of  $94 \pm 2\%$  and pure water flux of  $85 \pm 3 \text{ L}/(\text{m}^2\text{h})$  at 10 bar. Sodium periodate ( $\text{NaIO}_4$ , 99%) and isopropanol (IPA, > 99%) was purchased from Dieckmann Co. Ltd. Sodium chloride ( $\text{NaCl}$ , 99%) was purchased from Sigma Aldrich. Potassium chloride ( $\text{KCl}$ ,  $\geq 99.99\%$ ) was purchased from Aladdin. Tween 80 (chemically pure) was bought from Sinopharm Chemical Reagent Co., Ltd, China. Soybean oil got from COFCO Co., Ltd, China.

### 2.2 Polydopamine coating on XLE membrane

Prior to PDA coating, a XLE membrane coupon was rinsed using 25% IPA aqueous solution for 20 min and stored in deionized water for at least 12 h before use. The rinsed XLE membrane coupon was mounted in a custom-designed container with only the rejection layer exposed for coating [24]. PDA coating experiments were carried out using two different ways (Figure 1): the conventional/slow polydopamine coating method (sPDAC) and the fast polydopamine coating method (fPDAC). For sPDAC process, a 300 mL of freshly prepared 2.0 g/L dopamine/tris aqueous solution (10 mM, pH 8.5) was poured into the coating container to start the coating process. The fPDAC method was performed in accordance to references [22, 25] with slight modifications. Specifically, a 150 mL of freshly prepared  $\text{NaIO}_4$ /tris aqueous solution (10 mM, pH 8.5) and a 150 mL of dopamine/tris aqueous solution (10 mM, pH 8.5) were added to the coating container to obtain a mixture with a dopamine concentration of 2.0 g/L and a dopamine to  $\text{NaIO}_4$  molar ratio of 0.5. The PDA coating experiments were performed under continuous shaking conditions over predetermined time intervals. Finally, PDA coated XLE membrane was rinsed with deionized water for 30 min before being stored in deionized water for further use. The PDA coated XLE membranes are denoted as XLE-sPDAC-n or XLE-fPDAC-n, where n stands for the coating time in min.

### 2.3 Membrane characterization

Membrane morphology was examined using a LEO 1530 FEG scanning electron microscope (SEM, Germany). Before SEM test, a thin layer of gold was sputter coated onto the samples. Zeta potential of the membranes were tested by using streaming potential (SurPASS 3 Electrokinetic Analyzer, Anton Paar GmbH, Austria) over a pH range of 3-10 (1.0 mM KCl as background electrolyte solution). Surface elemental composition of the membrane samples were analyzed by X-ray photoelectron spectroscopy (XPS, Shimadzu Axis Ultra D1d, England) using an Mg Ka excitation radiation. Water contact angle tests were carried out using a goniometer equipped with video capture device (Powereach, China). A 5  $\mu$ L droplet of deionized water was introduced to the membrane surface and the data was recorded after 10 s of stabilizing. The coating rate of polydopamine was determined through a Quartz crystal microbalance with dissipation monitoring (QCM-D, E1, Q-Ssence, Sweden) using an open module. Typically, the equipment was firstly started, and then about 3.0 mL of 2.0 mg/L dopamine aqueous solution (with or without NaIO<sub>4</sub>) was added into cell immediately. The deposited mass was calculated using Equation 1[26]:

$$\Delta F = -\frac{2f^2}{A\sqrt{\rho_q\mu_q}}\Delta m \quad (1)$$

where  $\Delta F$  is the frequency change (Hz),  $A$  is the piezoelectrically active wafer area (m<sup>2</sup>),  $f$  is the resonant frequency (Hz) of the wafer,  $\rho_q$  is the density of quartz (kg/m<sup>3</sup>),  $\mu_q$  is the shear modulus of quartz (Pa), and  $\Delta m$  is the mass of the coating amount (kg).

### 2.4 Separation performance test

Water flux and salt rejection performances of the membranes were tested using a high pressure cross-flow filtration system under a constant pressure mode [24]. Temperature of the feed solution was kept constant at about 25 °C using an immersion thermostat (J.P. Selecta S.A., Barcelona, Spain). Pre-cut membrane sample with an effective area of 42 cm<sup>2</sup> was mounted in a commercial cross-flow cell (CF042, Delrine acetal, Sterlitech, America). The membrane samples were pre-compacted using deionized water under 10 bar for 12 h before performance evaluation. The pure water flux can be calculated by measuring the mass of the permeate water collected over a specified time intervals according to Equation 2:

$$J_v = \frac{\Delta w}{\Delta t \times A \times \rho} \quad (2)$$

where  $J_v$  (L/ (m<sup>2</sup> h)) is the water flux,  $\Delta w$  (kg) is the mass of permeate water collected over a time period of  $\Delta t$  (h),  $A$  (m<sup>2</sup>) is the effective membrane area, and  $\rho$  (kg/L) is the density of permeate water.

Salt rejection performance of the prepared membrane was investigated using a feed solution of 2000 mg/L NaCl. The concentration of NaCl in the feed solution ( $C_f$ ) and the permeate solution ( $C_p$ ) was tested to determine the membrane rejection performance. The membrane rejection ( $R$ ) was calculated using equation 3:

$$R = \left(1 - \frac{C_p}{C_f}\right) \times 100\% \quad (3)$$

Fouling experiments were carried out using soybean oil-in-water (Tween 80 was used as surfactant) emulsion solution through a cross-flow filtration process. Firstly, the membrane was pre-compacted using deionized water for 12 h at 10 bar. And then NaCl was added into the feed solution to get a concentration of 2000 mg/L. After that soybean oil-in-water emulsions (1 v1%) were introduced into the feed solution to start the fouling experiment. The fluxes of the membranes were all adjusted to about 50 L/m<sup>2</sup>h through manipulating of the applied pressure for better comparison.

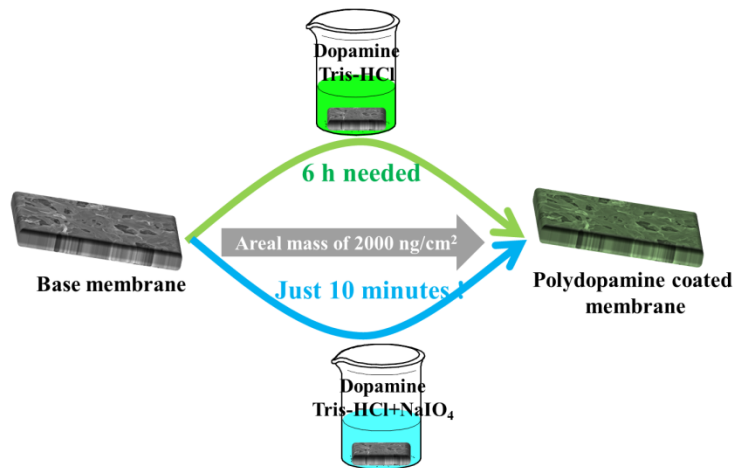


Figure 1. Schematic illustration of the slow (conventional) and fast polydopamine coating processes.

### 3. Results and discussions

### 3.1. Coating rate of polydopamine

The dynamic PDA coating process was monitored using QCM-D (Figure 2). In the conventional sPDAc process, the areal mass increased approximately linearly in the first ten minutes, after which the rate slowed down significantly. The coating reached an areal mass of 379 ng/cm<sup>2</sup> at a coating time of 50 min, corresponding to an average PDA growth rate of 4.5 nm/h (assuming a PDA density of 1 g/cm<sup>3</sup>). This value agrees reasonably well with the rate of 4-5 nm/h reported by H. Lee et al [1]. Compared to the sPDAc process, the mass deposition rate of polydopamine was much more rapid for the fPDAc process. The areal mass increased from 0 to approximately 2000 ng/cm<sup>2</sup> within the first 10 min for the fPDAc process compared to 360 min for the sPDAc process (supporting information Table S1). In this case, the fPDAc process provided a 97% reduction in coating time. The greatly increased coating rate can be attributed to the strong oxidative property of NaIO<sub>4</sub> used in fPDAc process, which can accelerate the self-assembly and covalent bond-forming reactions between dopamine and its oxidative product 5,6-dihydroxyindole [13, 22, 27]. In addition, the results also revealed that the polydopamine deposition rate phased over the coating process. The deposition rate was much faster in the initial 10 min and then gradually slowed down. This result may suggest that the coating rate is strongly affected by the degree of polymerization and/or the physical size of the assembled polydopamine, an effect that needs to be further investigated in future studies.

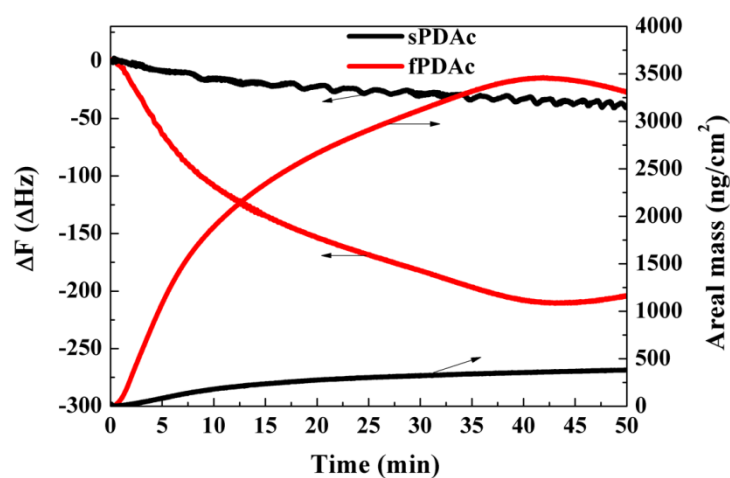


Figure 2. Rate of polydopamine coating based on QCM-D characterization. The areal mass was calculated based on the shift in frequency of the quartz sensor using Equation 1.

### 3.2. Morphology and surface properties of the membranes

XPS characterization revealed that the treatment of XLE by  $\text{NaIO}_4$  slightly increased the oxygen content from 11.4 to 12.1% (Figure 3), which can be attributed to the partial hydrolysis of polyamide (i.e., conversion of amide to more oxygen rich carboxylic groups) in the presence of the strong oxidant [28, 29]. Coating the membrane with polydopamine using either sPDAC or fPDAC method greatly increased oxygen content and reduced nitrogen content. The O/N ratio of both sPDAC- and fPDAC-coated membranes were approximately 2, which is consistent with the chemical composition of polydopamine [30]. This result confirmed the successful coating of polydopamine with both sPDAC and fPDAC methods.

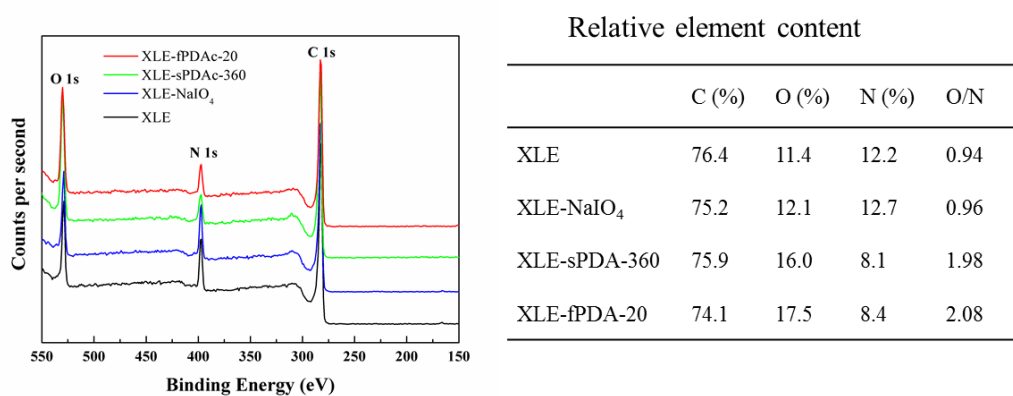


Figure 3. Surface elemental composition of the pristine XLE membrane and sPDAC- and fPDAC- coated membranes based on XPS measurements.

Figure 4 shows the membrane morphology before and after polydopamine coating. To allow a fair comparison, membranes with similar areal mass were paired. Compared to the pristine XLE membrane, the membrane coated with the sPDAC method did show clear morphology changes at an areal mass deposition of  $882 \text{ ng/cm}^2$  (corresponding to a coating time of 90 min). In contrast, the fPDAC-coated membrane surface with similar areal mass deposition ( $851 \text{ ng/cm}^2$  at a coating time of 4 min) had a grainy appearance. Furthermore, the grainy features became more extensive at increased coating time (10



min, 1893 ng/cm<sup>2</sup>). The morphological difference between membranes prepared by sPDAC and fPDAC can be attributed to their different polymerization kinetics, i.e., reaction-controlled vs. diffusion-controlled reactions [31, 32]. The polydopamine coating also significantly changed the membrane hydrophilicity. Compared to the pristine XLE membrane ( $48.6 \pm 2.2^\circ$ ), polydopamine coated membranes had much lower water contact angles ( $47.5 \pm 2.1^\circ$  for XLE-sPDAC-360 and  $43.6 \pm 1.4^\circ$  XLE-fPDAC-10 at similar areal mass deposition of  $\sim 2000$  ng/cm<sup>2</sup>).

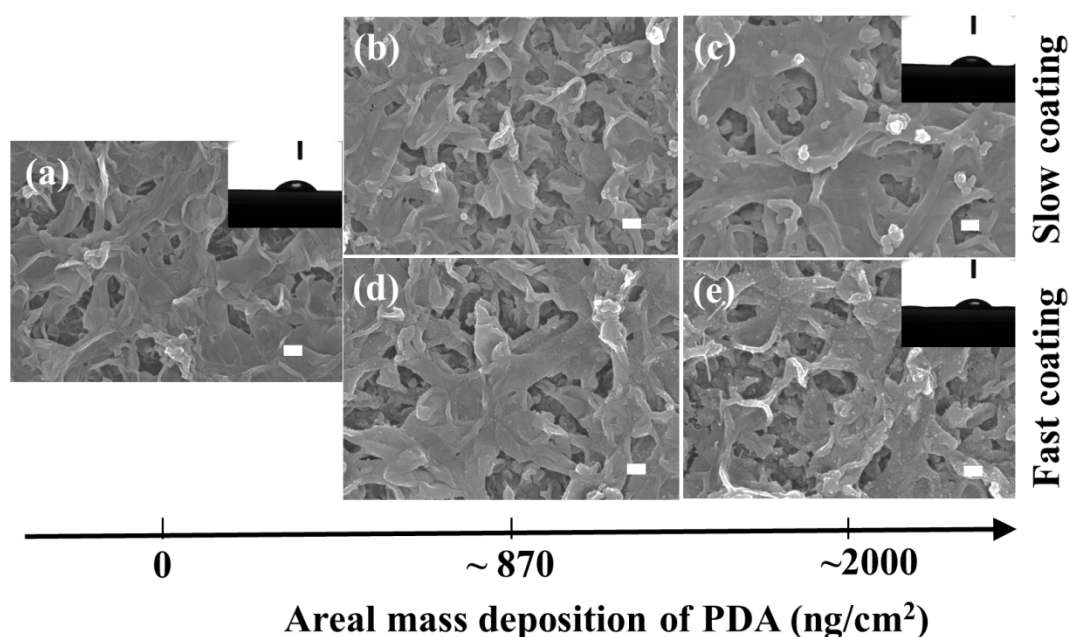


Figure 4. SEM micrographs of pristine XLE membrane (a) and polydopamine coated membranes with similar areal mass using slow and fast coating strategy respectively, (b) XLE-sPDAC-90, (c) XLE-sPDAC-360, (d) XLE-fPDAC-4 and (e) XLE-fPDAC-10 (all the scale bar is 200 nm, insets are the corresponding water contact angle results).

Zeta potential measurements showed that NaIO<sub>4</sub> treatment significantly enhanced the surface charge of XLE membrane (Figure 5). This result is consistent with the XPS result: the partial hydrolysis of polyamide results in increased density of charged functional groups (carboxylic and amine groups) [28, 33]. Coating the membrane with polydopamine reduced the membrane surface charge. Both sPDAC- and fPDAC-coated membranes had similar zeta potential. The neutralized surface charge property

combined with the increased hydrophilicity can enhance membrane antifouling performance [34-36].

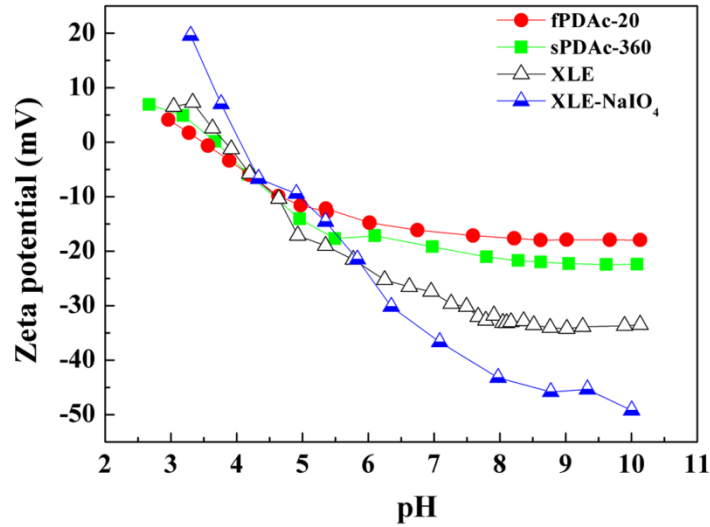


Figure 5. Zeta potential results of of the pristine XLE membrane and sPDAc- and fPDAc- coated membranes.

### 3.3. Separation performance of the membranes

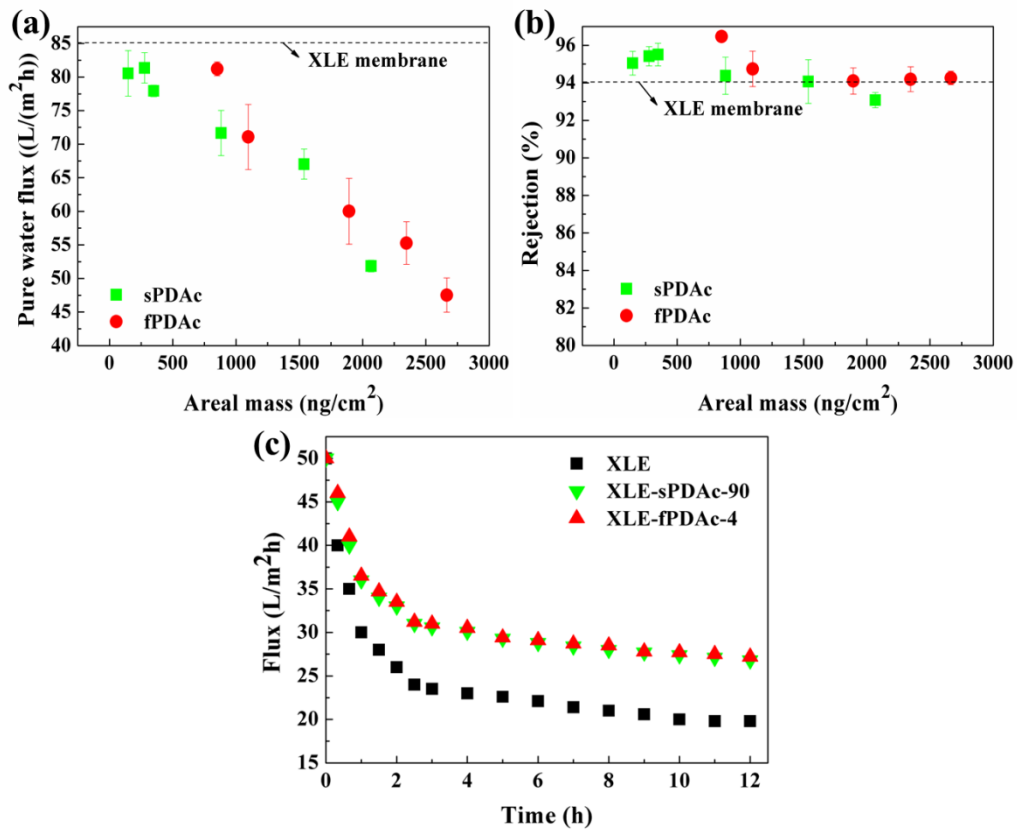


Figure 6. Pure water flux (a) and rejection (b) performance of the membranes; (c) Permeate flux change of the membranes during filtration of oil/water emulsion solutions.

Figure 6 presents the separation performances of membranes prepared by the two coating methods. Both sPDAC and fPDAC reduced the pure water flux of the coated membranes (Figure 6a). At a relatively low areal mass deposition of approximately  $1000 \text{ ng/cm}^2$ , the water flux was less affected for the fPDAC-coated membrane, which could be attributed to the partial hydrolysis of the amide bonds under the strong oxidant of  $\text{NaIO}_4$  [33, 37]. However, the difference in water flux became less obvious at higher areal mass deposition due to the increasing dominance of the hydraulic resistance of the coating.

$\text{NaCl}$  rejection values of the both sPDAC- and fPDAC-coated membranes were consistently above 93% (Figure 6b). Notably, when the polydopamine coating amount was relatively low ( $< 1000 \text{ ng/cm}^2$ ), the coated membranes had better  $\text{NaCl}$  rejection compared to that of the pristine XLE membrane (94%). This enhancement can be partially attributed to the sealing of the defects in the polyamide rejection layer by the polydopamine coating. Similar enhancement effect has been previously reported for polyamide membranes during their initial fouling stage by humic acid [38]. Compared to sPDAC-coated membranes, the fPDAC-coated membranes had better  $\text{NaCl}$  rejection. Despite its partial hydrolysis, the increased charge density in the polyamide rejection layer after fPDAC treatment resulted in stronger charge repulsion and thus better rejection of  $\text{NaCl}$  [37]. In the current study, extensive deposition of polydopamine was less effective in enhancing  $\text{NaCl}$  rejection (or even detrimental to  $\text{NaCl}$  rejection). This result can be explained by the cake-enhanced concentration polarization effect [39]. Since the polydopamine layer was more permeable to sodium chloride compared to the base XLE membrane [25, 40], the sodium chloride rejected by the polyamide layer will accumulate within the polydopamine layer. For membranes with extension polydopamine coating, severe concentration polarization can develop due to the lack of crossflow within the unstirred coating layer [39, 41].

Antifouling performance of the PDA coated XLE membranes were investigated using soybean oil as the foulant. As Figure 6c indicated that PDA coated membranes showed better antifouling performance than pristine XLE membrane despite its coating method (XLE-sPDAC-90 or XLE-fPDAC-4). The water fluxes of PDA coated membranes were about 36% higher than pristine XLE membrane after filtration of 12 h. The reason was mainly due to the hydrophilicity enhancement (as shown in Figure 4) after PDA coating and the strong repelling ability of PDA towards oil [6].

## **Conclusions**

Polydopamine modified XLE membranes were successfully prepared using a novel fPDAC method. The fPDAC process showed greatly accelerated polydopamine deposition rate in the presence of a strong oxidant  $\text{NaIO}_4$ . To reach a areal mass of approximately  $2000 \text{ ng/cm}^2$ , the fPDAC reduced coating time by 97% when compared to the conventional sPDAC method. Although both sPDAC and fPDAC coatings led to reduced pure water flux, the loss was milder for the fPDAC-coated membrane with an areal mass of approximately  $800 \text{ ng/cm}^2$ . In addition, this membrane also showed significantly better NaCl rejection compared to the base XLE membrane, thanks to the combined effect of defects sealing by the polydopamine coating and the enhanced charge repulsion effect by the partially hydrolyzed polyamide rejection layer. Moreover, the PDA coated XLE membrane showed better antifouling performance for treating the oil/water wastewaters. The current study also suggests that extensive polydopamine coating is unfavorable due to the severe reduction in water permeability and cake-enhanced concentration polarization.

## **Acknowledgements**

The authors thank the support from the General Research Fund (Project number 17207514) by the Research Grants Council of Hong Kong.

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