RESEARCH ARTICLE

Reductant-assisted polydopamine-modified membranes for efficient water purification

Feng Sun^{1*}, Jinren Lu^{1*}, Yuhong Wang², Jie Xiong¹, Congjie Gao¹, Jia Xu (🖂)¹

1 Key Laboratory of Marine Chemistry Theory and Technology (Ministry of Education), School of Materials Science and Engineering,

College of Chemistry and Chemical Engineering, Ocean University of China, Qingdao 266100, China

2 National Center of Ocean Standards and Metrology, Tianjin 300112, China

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Abstract Surface engineering with polydopamine coatings has been considered a promising surface functionalisation tool. However, it is difficult to control the selfpolymerisation for polydopamine formation, which usually causes severe interparticle aggregation. In this study, polydopamine self-polymerisation was controlled by adjusting its reducing environment using a reductant (NaBH₄) to fabricate mixed cellulose ester (MCE)/ polydopamine membranes. An oxidising environment using NaIO₄ was additionally tested as the control. The results showed that a thin polydopamine coating with small polydopamine particles was formed on the skeleton frameworks of the MCE membrane with NaBH₄, and the self-polymerisation rate was suppressed. The polydopamine coating formed in the reducing environment facilitated excellent water transport performance with a water permeance of approximately 400 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$ as well as efficient organic foulant removal with a bovine serum albumin rejection of approximately 90%. In addition, the polydopamine coating with NaBH₄ exhibited both excellent chemical stability and anti-microbial activity, demonstrating the contribution of the reducing environment to the performance of the MCE/polydopamine membranes. It shows significant potential for use in water purification.

Keywords membrane, water purification, polydopamine, reducing environment, self-polymerization control

1 Introduction

As one of the most important processes of separation, water purification is critical to address the growing global

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E-mail: qdxujia@sina.com.cn

*These authors contributed equally to this work.

concerns pertaining to water pollution and shortage [1]. Membrane technologies such as ultrafiltration and nanofiltration have been employed successfully in various applications to remove pollutants from contaminated water and produce fresh water, such as in wastewater treatment, seawater desalination, reclamation, and drinking water production [2–5]. Membranes form the core of membrane technology. Extensive research has been conducted on improving the separation performance of membranes by developing advanced membrane materials and fabrication approaches. There are two approaches to membrane fabrication: surface modification, and phase inversion. Surface modification such as coating and grafting is considered a versatile approach to tailoring membrane surfaces with desirable properties [6,7].

Recently, inspired by the adhesive nature of catechols and amines in mussel adhesive proteins, dopamine (3,4dihydroxyphenethylamine), as a precursor monomer of polydopamine (polydopamine) formation, was used to form a robust and water-resistant polydopamine layer via *in-situ* self-polymerisation, which bound firmly to diverse substrate surfaces [8,9]. Therefore, polydopamine is considered a uniquely adaptable and simple surface functionalisation tool owing to its versatility, simplicity, and broad potential for use in the biomedical, energy, consumer, industrial, and military sectors [10-14], especially in membrane-based separation. In addition, polydopamine is popularly known as a bio-glue owing to its high adhesion between the dense layer and substrate [15,16] and its facile post-introduction of reactive polymers [8,17], inorganic nanoparticles (Ag, Cu, TiO₂) [18-23], proteins, polysaccharides, and hydroxyapatites [24-30]. For instance, we fabricated an efficient nanofiltration membrane via polydopamine deposition followed by polyethyleneimine cross-linking [8]. As a substrate of thin-film-composite membranes for water purification, polymeric porous membranes with polydopamine coatings can achieve increased water permeability owing to the

hydrophilic modification by polydopamine [31–35]. Furthermore, based on its synergistic interaction with aromatic amine molecules [36], 3,4-dihydroxyphenethylamine can be used as one of the mixed monomers to participate in interfacial polymerisation to fabricate improved thin-film-composite membranes. All the above-mentioned research demonstrates the effectiveness of surface engineering with polydopamine coatings [37–39].

Dissolved O₂ is essential for traditional polydopamine formation via auto-oxidation at alkaline pH [40]. In this case, it is difficult to control the self-polymerisation for polydopamine formation and this causes severe interparticle aggregation, which inevitably increases the thickness and roughness of the polydopamine coatings [41]. As is well known, a thin coating layer with a well-distributed micro/nano-structure is essential for efficient water purification. Therefore, researchers have invested tremendous effort in customising the structure, thickness, and properties of polydopamine coatings based on dopamine concentration, coating time, pH, solvent, and chemical additives, including metal ions [42]. For instance, employing a lower concentration (< 0.5 mg/mL) [43] and shortening the coating time from 24 h to 1–3 h in Tris buffer [44] could effectively reduce polydopamine particle formation. Additionally, Xu et al. developed a rapid polydopamine/polyethyleneimine co-deposition approach to suppress the precipitation of polydopamine aggregates and thus obtain an even polydopamine coating surface [45,46]. However, although there is a general knowledge of the crucial role of oxidising/reducing environments in polydopamine formation [47,48], few researchers have focused on the influence of the reducing environment on polydopamine self-polymerisation to suppress polydopamine aggregates.

To better understand the polydopamine formation behaviour and elucidate its controllable parameters, we were motivated to control the modification of polydopamine by adjusting its reducing atmosphere using a reductant. NaBH₄ was used as the reductant in this study owing to its moderate reductive nature and water solubility in alkaline solutions. For comparison, the influence of the oxidising atmosphere achieved via the addition of NaIO₄ into 3,4-dihydroxyphenethylamine solution was additionally tested. The effects of the reducing atmosphere on the surface structure and separation performance of the polydopamine coating were investigated. The pH-dependent stability of the polydopamine coating and its antimicrobial activity were additionally investigated, which are crucial to a real water purification application.

2 Experimental

2.1 Materials

Unless stated otherwise, all the reagents were used without

further purification, including 3,4-dihydroxyphenethylamine (98%, Sigma), NaIO₄ (99%, Aldrich), NaBH₄ (98%, Aldrich), and bovine serum albumin (BSA, 67 kDa, Sigma). Mixed cellulose ester (MCE) microfiltration membranes with a diameter of 6 cm, average pore size of 0.1 μ m, and thickness of 135.2 μ m were purchased from Beijing Shenghe Membrane Tech Co. Ltd. To avoid the incorporation of Tris buffer into polydopamine [49], deionised water was used as the sole solvent without any buffer; it was prepared using a Millipore Milli-Q water purification system with a resistivity of 18.0 M Ω ·cm.

2.2 Polydopamine-coated membrane fabrication

To prepare fresh 3,4-dihydroxyphenethylamine solution, NaBH₄ or NaIO₄ in a predetermined dosage was first dissolved in deionised water. The solution pH was adjusted to approximately 9 using 1 mol· L^{-1} NaOH, and then 3,4-dihydroxyphenethylamine was dissolved to reach a concentration of 1.0 $g \cdot L^{-1}$. A clean MCE membrane was soaked in deionised water for 1 h and then immediately immersed into a sealed vessel, which was filled with the freshly prepared 3,4-dihydroxyphenethylamine solution with continuous stirring at 180 r · min⁻¹ and 15 °C for 4 h without contact with atmospheric O₂. Thereafter, the MCE membrane was taken out and washed with deionised water to remove residual polydopamine particles, which were not firmly adsorbed. The resultant polydopamine-coated MCE membrane (MCE/polydopamine) was stored in deionised water for further permeation tests and dried overnight at room temperature for characterisation.

2.3 Membrane characterisation

A Fourier transform infrared (FTIR) spectroscopy instrument (TENSOR27, Bruker, Germany) with attenuated total reflection mode was used to analyse the chemical compositions of the samples with a resolution of 4 cm⁻¹ and wavenumber in the range 400–4000 cm⁻¹. The surface morphologies of the membranes were observed via scanning electron microscopy (SEM, S-4800SEM, Hitachi, Japan). The samples were dried overnight and sputtercoated with Au nanoparticles prior to the SEM measurements. Water contact angle analysis was conducted using a contact angle goniometer (DSA 100, KRUSS, Germany). Each sample was fixed flat on a glass slide and a water drop with a volume of 2 μ L was dropped on to the membrane surface. At least five measurements were taken for each sample and their average value was calculated.

2.4 Separation performance

The water transport property of the MCE/polydopamine membranes was evaluated based on their pure water permeance (L_p), which was measured using a lab-scale ultrafiltration set-up [50] with an effective membrane area

of 19.63 cm². The membrane sample was pre-pressurised with deionised water at 0.15 MPa for 2 h until it reached a steady state of permeation. Four transmembrane pressures of 0.08, 0.10, 0.12, and 0.14 MPa were applied to the ultrafiltration set-up and the pure water permeance was measured. The value of L_p (L·m⁻²·h⁻¹·bar⁻¹) was calculated using Eq. (1).

$$L_{\rm p} = \frac{Q}{A \times \Delta t \times P},\tag{1}$$

where Q is the permeate volume over a certain period of filtration time Δt ; A is the effective membrane area; P is the feed pressure.

The separation performance, including permeation flux (*J*) and organic foulant rejection (*R*), was measured via filtration experiments in dead-end and constant-pressure modes. A total of 200 mg \cdot L⁻¹ of BSA solution was used as an organic foulant feed solution and each filtration experiment was conducted at an operating pressure of 0.10 MPa and room temperature for 2 h. The definitions of permeation flux and BSA rejection are presented in Eq. (2) and Eq. (3), respectively.

$$J = \frac{Q}{A \times \Delta t},\tag{2}$$

$$R = \left(\frac{C_{\rm F} - C_{\rm P}}{C_{\rm F}}\right) \times 100\%. \tag{3}$$

2.5 Chemical stability and anti-microbial activity

It is reported that polydopamine coatings are sensitive to solution pH, especially at higher pH [48]. In the present study, HCl solution (pH 3) and NaOH solution (pH 11) were selected as representative solutions to investigate the chemical stability of the MCE/polydopamine membranes. Chemical stability is important in real water purification applications, wherein chemical cleaning (using acidic or basic solutions) of fouled membranes is routinely carried out.

Escherichia coli was employed as a model microorganism to evaluate the anti-microbial activity of the MCE/ polydopamine membranes. The counting method of colony-forming units has been described elsewhere [51]. *E. coli* was incubated in a Luria-Bertani (LB) liquid culture medium, which was shaken at 37 °C for 24 h and then diluted with additional LB liquid culture medium to a predetermined concentration. An MCE membrane (as a blank sample) and an MCE/polydopamine membrane were cut into small pieces (1.0 cm × 2.0 cm). They were sterilised together with all the glassware using ultraviolet radiation (40 W, 253.7 nm) for 1 h. The membrane samples were placed in separate cuvettes containing 10 mL of *E. coli* suspension, which were then placed in a shaking incubator at 37 °C for 24 h. Thereafter, the membranes were removed from the cuvettes, and the *E. coli* cells were detached from the membranes using 15 mL of LB liquid culture medium, which was collected and diluted with additional LB liquid culture medium to 0.1% of the original concentration. Then, 200 μ L of the diluted solution was spread on the LB solid culture medium and incubated at 37 °C for another 24 h. The colonies on the culture dish were enumerated by the plate count method using a biological microscope. Each sample was tested thrice. The anti-microbial efficiency (*E*_b) was calculated as follows:

$$E_{\rm b} = \left(\frac{N_{\rm b} - N_{\rm m}}{N_{\rm b}}\right) \times 100\%,\tag{4}$$

where $N_{\rm b}$ and $N_{\rm m}$ are the numbers of colonies corresponding to the MCE membrane (as a baseline) and the MCE/ polydopamine membrane, respectively.

3 Results and discussion

3.1 Characterisation of MCE/polydopamine membranes

In this study, the MCE membrane was immersed in a fresh 3,4-dihydroxyphenethylamine solution with continuous stirring, and the polydopamine coating was gradually formed via 3,4-dihydroxyphenethylamine self-polymerisation adjusted using an oxidising/reducing environment (Fig. 1). To avoid the incorporation of Tris buffer into polydopamine [49] and intervention by O_2 in air, deionised water was used as the sole solvent without any buffer, and the vessel filled with the 3,4-dihydroxyphenethylamine solution was sealed.

The concentration of NaBH₄ or NaIO₄ was adjusted to investigate the effect of the oxidising or reducing environment, respectively, during polydopamine selfpolymerisation on the membrane morphology, and the results are shown in Fig. 2. The MCE/polydopamine membranes appeared darker in colour as the environment became more oxidising. The membrane with 0.1% NaIO₄ appeared black-brown and that with 0.1% NaBH₄ appeared light brown, indicating that NaBH₄ can suppress polydopamine formation. This phenomenon is in accordance with the SEM images (Fig. 2). Compared with the pristine MCE membrane, the MCE/polydopamine membranes exhibited denser morphologies to different extents, indicating that immersion in 3,4-dihydroxyphenethylamine solution with NaBH₄ or NaIO₄ could lead to polydopamine modification on the MCE skeleton surface. When NaBH₄ was added, much smaller polydopamine nanoparticles were attached to the skeleton of the MCE membrane and could be observed partially. When NaIO₄ was added, more and larger polydopamine nanoparticles were formed; the entire MCE membrane was covered when 0.2% NaIO₄ was used. This demonstrated that



Fig. 1 Illustration of fabrication and structure of MCE/polydopamine membranes via controlling the oxidating/reducing environment.

polydopamine self-polymerisation was decelerated in a reducing environment but promoted in an oxidising environment. It should be noted that although the polydopamine coating on the membrane surface in this work was not continuous owing to the non-continuous MCE skeleton, a continuous polydopamine coating layer can be obtained if a continuous substrate is used.

The chemical structures of the membranes were analysed using FTIR (Fig. 3(a)) and their water contact angles were shown in Fig. 3(b). Compared with the MCE membrane (Fig. S1), the MCE/polydopamine membranes with NaIO₄ exhibited new strong characteristic peaks at 3300 cm⁻¹ ascribed to N-H/O-H, at 1696 cm⁻¹ ascribed to C = C in benzene ring, and at 1200 cm⁻¹ ascribed to C–N, all of which were attributed to the polydopamine formation and were reinforced with increase in NaIO₄ concentration. All these peaks significantly weakened when NaBH₄ was employed, indicating that the reductant restrained 3,4-dihydroxyphenethylamine self-polymerisation. The surface wettability of membranes is essential for separation performance and was evaluated using water contact angle measurements (Fig. 3(b)). Although the polydopamine coatings with NaBH₄ yielded much higher water contact angle $(50^{\circ}-70^{\circ})$ than those of the polydopamine coatings with NaIO₄ ($15^{\circ}-25^{\circ}$), the values were still lower than that of the pure MCE membrane (approximately 90°), confirming successful polydopamine modification in a reducing environment with NaBH₄. The good surface wettability ascribed to the inherent hydrophilicity of the amide and catechol groups of the polydopamine particles would enable the membranes to be rapidly infiltrated by water, thereby enhancing their water permeation performance during water purification.

3.2 Separation performance of MCE/polydopamine membranes

The effects of various polydopamine coatings on the water transport property of the MCE/polydopamine membranes were evaluated via the pure water permeation test in the dead-end mode at 0.10 MPa. The results are shown in Fig. 4(a). All the MCE/polydopamine membranes yielded lower water permeance than that of the pristine MCE membrane (approximately 500 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$). In a more oxidising environment (less NaBH₄ and more NaIO₄), the water permeance decreased from approximately 400 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$ to approximately 190 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$. This was primarily attributed to the pore shrinkage of the MCE membrane owing to the polydopamine modification (Fig. 2) despite its improved hydrophilicity (Fig. 3(b)), indicating that pore shrinkage played a more crucial role than that of hydrophilicity in the permeation performance of the MCE/polydopamine membranes.

A filtration experiment of 200 mg·L⁻¹ BSA solution was performed to evaluate the separation performance of the MCE/polydopamine membranes. Figure 5(a) presents that the MCE/polydopamine membrane exhibited lower water flux with decrease in NaBH₄ and increase in NaIO₄ concentrations, which was the same tendency as that of pure water permeance. The BSA rejection of the membranes is shown in Fig. 4(b). With decrease in NaBH₄ and increase in NaIO₄ concentration, the membranes exhibited improved BSA rejection. The highest BSA rejection of 97% was achieved with the highest NaIO₄ content of 0.2%. It is interesting that even in the strongest reducing environment employed in this study



Fig. 2 SEM images and photos of MCE/polydopamine membranes.



Fig. 3 Surface properties characterization of MCE/polydopamine membranes: (a) FTIR spectra; (b) water contact angle.

 $(0.2\% \text{ NaBH}_4)$, the MCE/polydopamine membrane yielded a BSA rejection of approximately 90% at 100 min of filtration, which was much higher than that of the pristine MCE membrane (approximately 59%). This demonstrated the significant contribution of the reducing environment to polydopamine modification, facilitating excellent water transport performance with a water permeance of approximately 400 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}and$ efficient organic foulant removal with a BSA rejection of approximately 90%. In addition, for each filtration pass in the dead-end filtration experiment, the increase in rejection and decrease in flux with filtration time were both primarily attributed to the additional mass transfer resistance owing to membrane fouling. Thus, the amount of increase in BSA rejection with filtration time could represent the anti-fouling property of the membranes. In Fig. 4(b), the MCE/polydopamine membranes exhibited lower rejection variations than that of the MCE membrane, and this variation in rejection declined further with decrease in NaBH₄ and increase in NaIO₄. This indicated that irrespective of the reducing/oxidising environment, polydopamine modification enhanced the anti-fouling property of the membranes. Compared with the reducing environment, the oxidising environment contributed more to the enhancement of the anti-fouling property.

3.3 pH stability of MCE/polydopamine membranes

The chemical and structural stabilities of membranes play an important role in practical applications. It has been reported that polydopamine coatings are sensitive to solution pH, especially at higher pH, and those formed in more oxidising environments are more unstable [48]. To investigate the effects of a reducing environment on the stability of polydopamine coatings, various MCE/polydopamine membrane samples were immersed in acidic (pH = 4) and alkaline (pH = 11) solutions, separately. As shown in Fig. 5(a), the solutions appeared colourless and



Fig. 4 Separation performance of MCE/polydopamine membranes: (a) pure water permeance and the ratio of water flux to pressure with 200 mg \cdot L⁻¹ BSA solution as feed; (b) BSA rejection with 200 mg \cdot L⁻¹ BSA solution as feed.

transparent at pH = 4 although the solutions with 0.1% and 0.2% NaIO₄ were found to be slightly yellow, indicating excellent stability of all the MCE/polydopamine membranes in the acidic solution. In contrast, murky solutions were observed for the MCE/polydopamine membranes at all concentrations of NaIO₄ tested, which darkened with increase in NaIO₄ concentration and became identical to the colour of the MCE/polydopamine membranes with $NaIO_4$ (Fig. 2). This indicated that the stability of the membranes with NaIO₄ decreased tremendously at higher pH values, which was confirmed by the filtration tests (Fig. 5(b)). This was probably owing to the large number of polydopamine nanoparticles, which were loosely adhered to the MCE membrane and easily released in the alkaline solution. However, it should be noted that at pH = 11, the solutions remained colourless and transparent in the case of the MCE/polydopamine membranes with NaBH₄, confirming the contribution of the reducing

environment to the stability of polydopamine modification. This might be related to the thinner polydopamine coating with smaller polydopamine particles (Fig. 2), which could attach to the MCE membrane firmly.

3.4 Anti-microbial activity of MCE/polydopamine membranes

In this study, gram-negative *E. coli* was used as the model microorganism to evaluate the antimicrobial activity of the MCE/polydopamine membranes. A pristine MCE membrane was tested as a control. As shown in Fig. 6(a), compared with the control MCE membrane, all the MCE/polydopamine membranes showed considerable reduction in *E. coli*, indicating that the polydopamine coating conspicuously decreased the number of cells attached to the membrane surface. The corresponding anti-microbial efficiencies ($E_{\rm b}$) were approximately 60%–70% (Fig. 6(b))



Fig. 5 Stability evaluation of MCE/polydopamine membranes in acid (pH = 4) and base (pH = 11) solution: (a) photos; (b) separation performance.



Fig. 6 Petri dish results of E. coli growth inhibition: (a) photos; (b) colony number and anti-microbial efficiency.

compared with that of the control. Compared with the MCE/polydopamine membranes with NaBH₄ ($E_b = 60\%$ -65%), the oxidising environment facilitated better antimicrobial activity with $E_b = 65\%$ -70%, which might be attributed to the greater number of polydopamine particles attached to the surface and the higher hydrophilicity of the MCE/polydopamine membranes with NaIO₄. It should be noted that the difference in E_b between the membranes with NaIO₄ and NaBH₄ was not obvious, indicating the effectiveness of the reducing environment on the antimicrobial activity.

4 Conclusions

In summary, we fabricated a series of MCE/polydopamine membranes, wherein a reductant (NaBH₄) or oxidant $(NaIO_4)$ in various concentrations was introduced to form the polydopamine coatings. The MCE/polydopamine membranes in the oxidising and reducing environments showed distinct properties in terms of morphology, wettability, separation performance, pH stability, and anti-microbial activity. Compared with the membranes prepared in an oxidising environment, the MCE/polydopamine membrane with NaBH₄ exhibited excellent water transport performance with a water permeance of approximately 400 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$ as well as efficient organic foulant removal with a BSA rejection of approximately 90%. In addition, the MCE/polydopamine membrane with NaBH₄ achieved both excellent chemical stability and anti-microbial efficiency, showing great potential for use in water purification. This demonstrated that the oxidising/reducing environment can be a significant parameter influencing polydopamine formation.

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