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EDTA-based adsorption layer for mitigating FO membrane fouling via *in situ* removing calcium binding with organic foulants

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Abstract

Forward osmosis (FO) is an emerging technology for wastewater treatment and reclamation. However, membrane fouling remains a strong hindrance to FO application. We proposed a novel approach for alleviating FO membrane fouling via *in situ* removing Ca^{2+} binding with organic foulants using the EDTA-based adsorption layer. Results suggested that the EDTA-based adsorption layer can effectively remove the Ca^{2+} binding with sodium alginate, and its adsorption capacity correspondingly increased as a function of Ca^{2+} concentration in the feed solution. Owing to the effective extraction of Ca^{2+} from the fouling layer by the EDTA-based adsorption layer, water flux of FO membrane was significantly enhanced, and fouling layer became easily removed by physical flushing, suggesting a remarkable alleviation of FO membrane fouling. Mitigation of FO membrane fouling by the EDTA-based adsorption layer was attributed to the fact that the fouling layer structure became more porous and looser after *in situ* removing Ca^{2+} from the alginate-Ca^{2+} gel networks. This study demonstrated a novel fouling control strategy via *in situ* removing Ca^{2+} binding with the organic foulants, providing a new avenue for FO membrane fouling management.

Keywords: Forward osmosis; Membrane fouling; EDTA; Organic foulants; Calcium

1. Introduction

An emerging osmotically-driven membrane process, forward osmosis (FO) exhibits great promise to address global challenges in water-energy nexus [1-6]. FO utilizes a draw solution (DS) with high osmotic pressure to drive the water molecular from a feed solution (FS) with low osmotic pressure passing through a semi-permeable membrane [6-8]. Compared to pressure-driven nanofiltration (NF) or reverse osmosis (RO) processes, FO process shows several advantages including lower energy consumption [9], lower fouling propensity [10-12], and higher water recovery [13]. However, membrane fouling remains a critical hindrance to FO process as well as osmotic membrane bioreactor (OMBR), which results in a decline of water flux, an increased cleaning frequency and a shortened membrane life [14-21].

FO membrane fouling is caused by deposition of suspended particles or colloids, microorganisms, organic macromolecules, sparingly soluble inorganic compounds, or their mixtures on (or even inside) the membrane [4, 17]. Organic fouling induced by macromolecular organic compounds such as alginate, protein, and natural organic matters is more complicated [22], not only because specific interactions between chemical functional groups on the FO membrane surface and those of the organic foulants may occur, but it was also found to be affected by the ionic composition of the feed solution [23]. As a result, many previous studies on FO membrane fouling are focused on organic fouling [4, 11, 23-27]. In these studies, it has been demonstrated that organic fouling of FO membrane is deteriorated with the presence of divalent cations, i.e., forming a dense, cross-linked organic fouling layer [6], and consequently resulting in a rapid flux decline [23]. It is hypothesized that if the divalent cations can be removed from the foulants on the FO membrane surface, the organic fouling layer might become loose and subsequently easily removed by a simply physical cleaning.

Ethylenediaminetetraacetic acid (EDTA) is an aminopolycarboxylic acid, which is colorless and water-soluble. As a hexadentate ligand and a chelating agent, EDTA can bind to divalent metal ions such as Mg^{2+}, Ca^{2+}, Mn^{2+} and Fe^{2+} to form a stable water-soluble chelate [28-30]. Thereby, EDTA has been commonly used as a chelating agent in various aspects such as electroplating [31], phytoremediation [32], food safety [33], and soil remediation [34, 35]. Compared with other chelating agent, EDTA is more attractive owing to its cheap, convenient and practical.

We are inspired by the combination of EDTA and Ca^{2+} to form a stable EDTA calcium chelate, where this concept can successfully extract Ca^{2+} from the foulants on the membrane surface. In this study, we attempt to combine EDTA with agar to prepare an EDTA-based adsorption layer for *in situ* extracting the Ca^{2+} binding with the organic foulants from the FO membrane surface, thereby for mitigating membrane fouling and facilitating subsequent membrane cleaning. Until now, studies on applying an EDTA-based adsorption layer for controlling organic fouling of FO membrane were rarely reported. The aim of this study is to evaluate the proof-of-concept for FO membrane mitigation through *in situ* removing Ca^{2+} combined with the organic foulants by the EDTA-based adsorption layer.

2. Materials and methods

2.1. FO membrane

Cellulose triacetate (CTA) FO membrane (supplied by Hydration Technologies Inc.) used in this study was fabricated from cellulose acetate embedded in a polyester woven mesh [4]. As reported in our previous study, its water permeability coefficients (*A*) and salt permeability coefficients (*B*) were 2.8×10^{-12} m/(s Pa) and 17.5×10^{-7} m/s, respectively [19]. Prior to experimental tests, the FO membranes were soaked in deionized (DI) water and stored at 4 °C.

2.2 EDTA-based adsorption layer preparation

Considering the low solubility of EDTA in water, EDTA-2Na was chosen as the adsorption substrate. The procedure of preparing the EDTA-based adsorption layer was summarized in Fig. 1. Firstly, 0.5% wt EDTA-2Na was dissolved in DI water at 95 °C. And then, 3% wt agar was adding into the EDTA-2Na solution by stirring well with the glass rod. After that, the EDTA-agar mixture was poured into a mold whose size same as the CTA FO membrane (11.4 cm × 5.7 cm × 0.5 cm in dimension). When the solution temperature was cooled to the room temperature (25 ± 2 ^{o}C), the EDTA-based adsorption layer was obtained through removing from the mold.

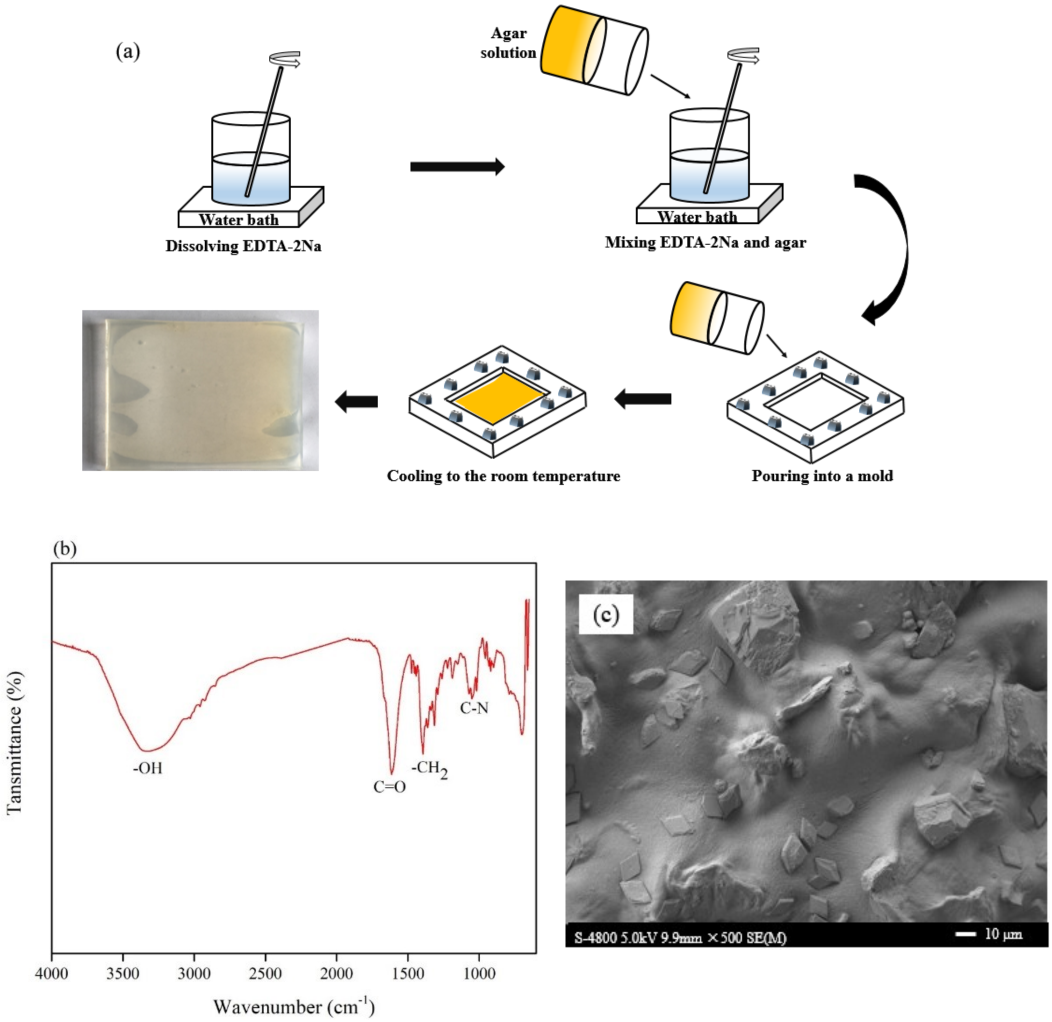


Fig. 1. Synthesis and characterization of the EDTA-based adsorption layer: (A) Flow chart of the

preparation of EDTA-based adsorption layer, (B) FTIR and (C) SEM images of the EDTA-based adsorption layer. The scale bar corresponds to 10 μm.

2.3. FO and EDTA-based adsorption experiments

Fouling behaviors of the FO membrane were evaluated in a bench-scale filtration system, as schematically shown in Fig. S1, Supporting Information. This test system included a cross-flow membrane cell with two symmetric flow channels (each of 85 mm × 39 mm × 2 mm in dimension). Membrane coupons were placed in the membrane cell between the two channels for the DS and FS, respectively. Two peristaltic pumps (Longer Precision Pump, China) were used to pump the DS and FS into the separate closed loops. The cross-flow velocity in both channels of the membrane cell was constant at 1.3 mm/s. Both DS and FS were kept at the temperature of 25 ± 2 °C. Change in the weight of FS was monitored by a digital balance (Mettler Toledo, China) and recorded in a computer by a data acquisition software (Mettler Toledo, China), which was converted into changes in water flux of FO membrane.

A 4 M NaCl solution was used as the DS in both fouling and baseline experiments. Sodium alginate (75-100 kDa) was selected as the model organic foulant. The baseline experiment was conducted before the fouling experiment with the DI water as the FS and the 4 M NaCl as the DS. In order to evaluate the adsorption efficiency of EDTA-based layer for Ca^{2+} binding with the sodium alginate, the FO membrane was fouled by the sodium alginate and $CaCl_2$ in sequence, which was different from the previous fouling protocol [4, 24]. The procedure of the FO membrane fouling tests was summarized in Fig. S2. Firstly, a new FO membrane coupon was sealed in the FO cell with active layer facing FS, and then a baseline experiment was performed for 4 h to obtain the initial flux of the FO membrane. After that, the fouling tests were started with 2 L of FS and DS.

With regard to the single organic fouling, the fouling filtration was operated for 48 h; while the enhanced organic fouling filtration was conducted with a $CaCl_2$ solution for 24 h after the single organic fouling experiments operated for 24 h. As for the single organic fouling, the FS consisted of 200 mg/L sodium alginate, 20 mM NaCl, 20 mM Na_2SO_4 and 1 mM $NaHCO_3$, while the FS was changed into the $CaCl_2$ solution with three concentration levels of 1, 15 and 35 mM, respectively, for the tests of enhanced organic fouling by Ca^{2+}.

The EDTA-based adsorption experiments were conducted after the fouling tests. As schematically shown in Fig. S3, Supporting Information, the fouled FO membranes were attached to the EDTA-based adsorption layer for 24 h during the *in situ* adsorption experiments. Just like the commonly used FO cell, the EDTA-based adsorption layer could have a sufficient physical contact with the fouled FO membrane and avoid destructing the fouling layer via adjusting the tightness of the four screws on the adsorption device. After that, the adsorption layer was removed into the eluent for 24 h. 1 M HCl was chosen as the eluent for removing Ca^{2+} from the EDTA-based adsorption layer. Meanwhile, the remaining foulants were removed from the FO membrane surface by sonication [36]. The extracted and remained Ca^{2+} concentrations were measured by an atomic absorption spectrometer (Shimadzu, Tokyo, Japan). The adsorption efficiency of Ca^{2+} was used for evaluating the EDTA-based adsorption layer. The specific descriptions on calculating adsorption efficiency of Ca^{2+} could be found in Section S1, Supporting Information.

2.4. Analytical method

Water flux through the FO membrane was obtained based on the weight change of the FS. After the fouling tests and the EDTA-based adsorption experiments, the water flux of the fouled and adsorbed FO membranes were determined by the FO cell for 4 h with the DI water as the FS and the 4 M NaCl as the DS. In order to eliminate the impacts of initial water flux for different FO membranes, a normalized water flux was used for characterizing the fouled and adsorbed FO membranes. It was obtained through the determining flux dividing by the initial flux before fouling tests.

The fouled FO membranes removed from the FO cell at the conclusion of experiment for characterization. A field-emission scanning electron microscopy (FE-SEM) (S4800, Hitachi, Japan) and an energy diffusive X-ray (EDX) analyzer (S4800, Hitachi, Japan) were used to capture the surface images and element compositions of the fouled and adsorbed FO membrane samples, respectively. Confocal laser scanning microscopy (CLSM) (LSM 710, ZEISS, Germany) was applied for analyzing the distributions of polysaccharides on the fouled and adsorbed FO membrane samples. Details of analytic methods of FE-SEM, EDX and CLSM analyses have been shown in our previous publication [19, 37].

3. Results and discussion

3.1. Flux performance of FO membrane during the fouling tests

A comparison of the results on FO membrane water flux profiles at different feed conditions is illustrated in Fig. 2. Water flux of FO membrane declined with the extension of the operating time regardless of the composition of the FS. However, the profile for FO membrane flux decline was significantly influenced by the foulants composition in the FS. Compared with the single organic fouling only induced by the sodium alginate, the flux decline became severer when Ca^{2+} was added into the FS. In addition, there was a noticeable water flux decline with an increase of the Ca^{2+} concentration. These experimental observations clearly showed that Ca^{2+} aggravated the organic fouling of FO membrane, which was consistent with previous literature [4, 24, 37]. This phenomenon could be attributed to the fact that Ca^{2+} and alginate form complexes with unique structure, thereby resulting in a high density gel network [38-40].

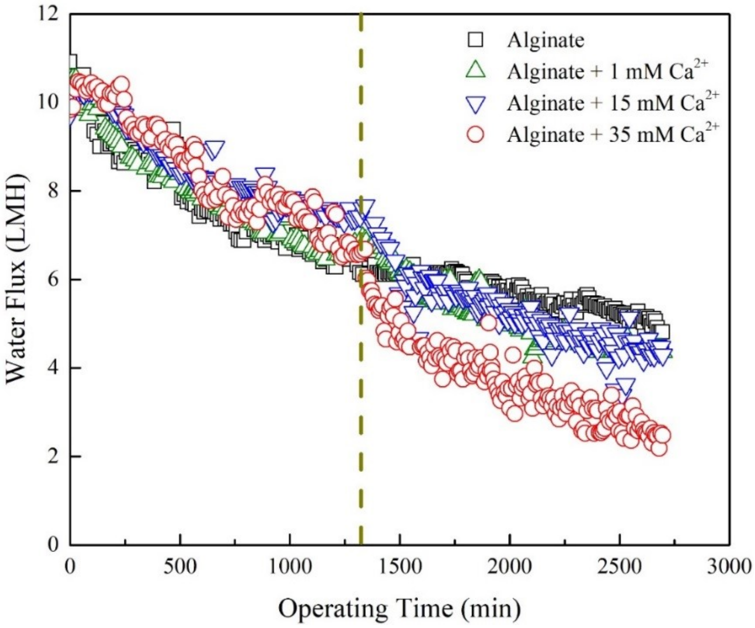
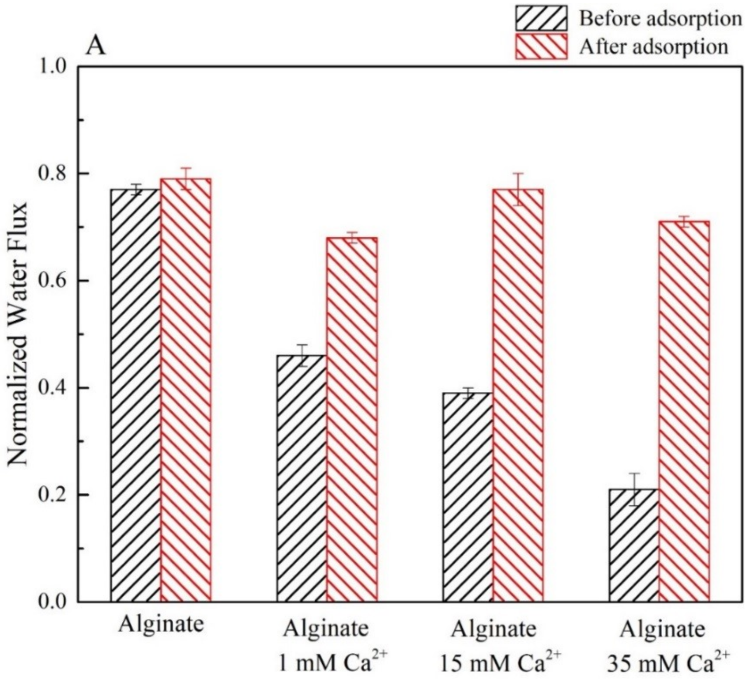


Fig. 2. Water flux profile of FO membranes during fouling filtration. FO fouling filtration was conducted with feed solution containing 200 mg/L sodium alginate, 20 mM NaCl, 20 mM Na_2SO_4 and 1 mM $NaHCO_3$, with varying calcium concentrations from 1 to 35 mM. Draw solution was 4 M NaCl. The filtration was operated for 48 hours.

3.2. Calcium removal and its impacts on the membrane fouling

After the fouling tests of the FO membranes, Ca^{2+} was extracted from the fouled FO membranes by the EDTA-based adsorption layer. From Fig. 3 (a), the normalized water flux of the fouled FO membrane induced by only alginate sodium was similar before and after adsorbing by the EDTA-based layer. By contrast, the normalized water flux had a significant increase after adsorbing Ca^{2+} via the EDTA-based layer for the organic fouling with Ca^{2+}, indicating an effective mitigation of FO membrane fouling. Variations of the normalized water flux for different fouling conditions were consistent with the adsorption efficiency of Ca^{2+} (Fig. 3 (b)). Specifically, there was no Ca^{2+} adsorption for the single organic fouling while the adsorption efficiency was ranged from 55.3% to 93.5% for the enhanced organic fouling with different concentrations of Ca^{2+}. These results implied that the EDTA-based layer could effectively adsorb Ca^{2+} from the alginate bound foulants. As a result, organic fouling of the FO membrane could be alleviated via *in situ* removing Ca^{2+} from the fouling layer.

Although the EDTA-based layer could effectively adsorb Ca^{2+} from the organic fouling layer of the FO membrane, the adsorption capacity and efficiency were limited by the feed concentration of Ca^{2+}. The adsorption capacity and efficiency of Ca^{2+} correspondingly increased with the feed Ca^{2+} concentration (Fig. 3 (b)), which was driven by more Ca^{2+} binding with the alginate at a higher addition of Ca^{2+}. It is notable that the adsorption efficiency was over 90% when the concentration of Ca^{2+} was up to 35 mM. Meanwhile, it was interesting to find that the Ca^{2+} adsorption efficiency was correlated well with the increase of the normalized water flux due to the Ca^{2+} adsorption, suggesting the significant contributions of Ca^{2+} to the organic fouling.



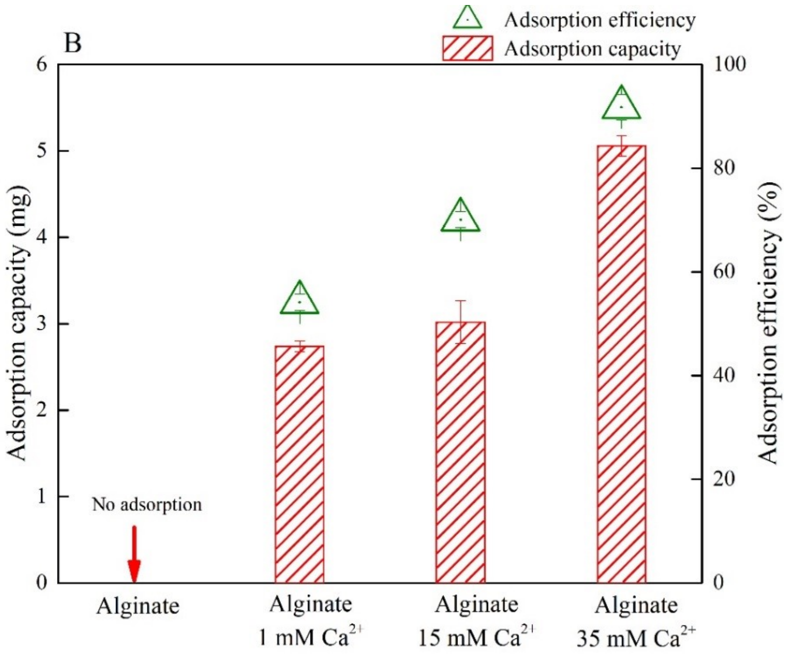
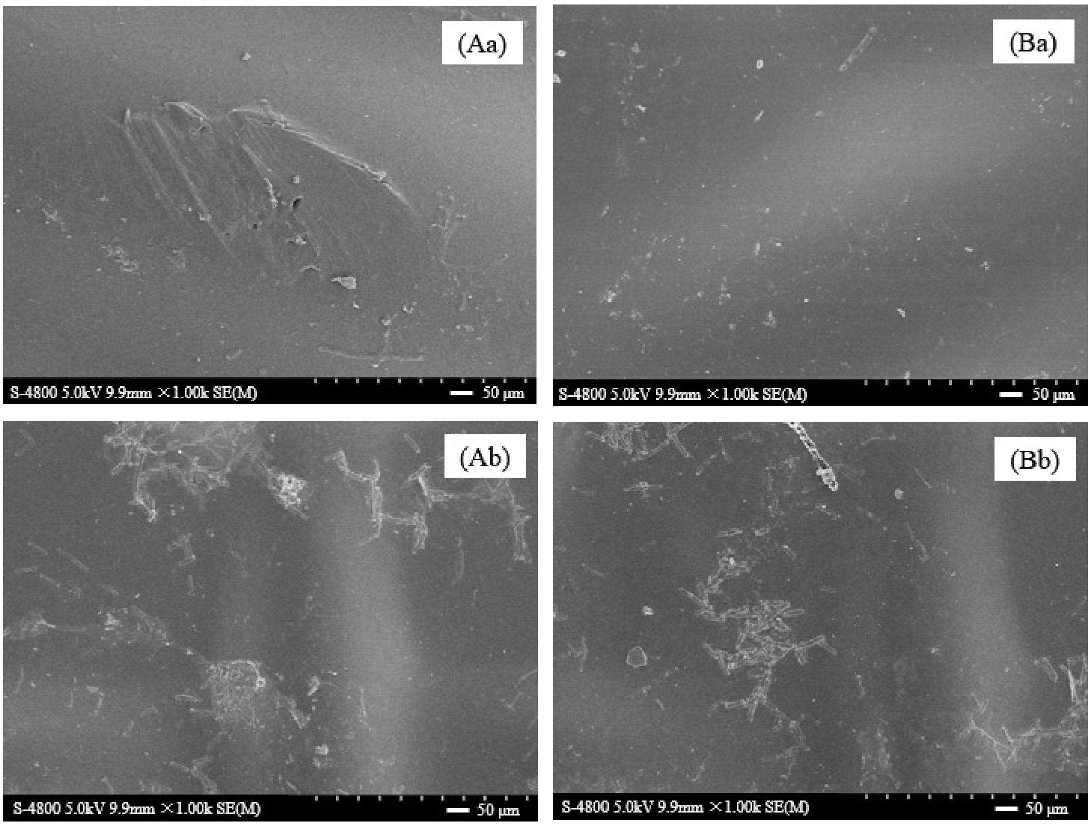


Fig. 3. EDTA-based adsorption layer performance in fouling mitigation: (A) Normalized water flux and (B) Adsorption capacity and efficiency at different fouling conditions. The FO fouling filtration conditions were described in Fig. 2. *In situ* Ca^{2+} adsorption was performed using the EDTA-based adsorption layer for 24 h, 1 M HCl as the eluent.

3.3. Changes in fouling layer structure after in situ removing Ca^{2+}

In order to evaluate the structure of fouling layer before and after Ca^{2+} adsorption via the EDTA-based adsorption layer, the morphology, element composition and structure of fouled FO membranes before and after adsorbing were analyzed by FE-SEM, EDX and CLSM, respectively. From Fig. 4, no obvious foulants were deposited on the FO membrane surfaces when only sodium alginate was used, while foulants deposition formed on the FO membrane surface when Ca^{2+} adding into the feed solution. This result further demonstrated that Ca^{2+} enhanced the organic fouling via forming alginate-Ca^{2+} gel networks. It is noteworthy to see that the alginate-Ca^{2+} gel networks became larger with the increase of Ca^{2+} concentration in the feed solution. After adsorbing Ca^{2+} via the EDTA-based adsorption layer, some gel networks on the enhanced organic fouling layer disappeared regardless of the Ca^{2+} concentration, which was consistent with the increase of normalized water flux of FO membrane (Fig. 3 (a)). From the EDX analyses (Fig. S4 and Table S1), the relative weight percentage of Ca on the fouled FO membranes correspondingly increased with the increase of the adding Ca^{2+} concentration, further suggesting the occurrence of more severe “egg-box-shaped” gel networks on the FO membrane surfaces. After removing Ca^{2+}, the relative weight percentage of Ca significantly reduced (Fig. S5 and Table S2). Both SEM and EDX results strongly proved the effective adsorption of Ca^{2+} from the fouling layer by the EDTA-based adsorption layer.



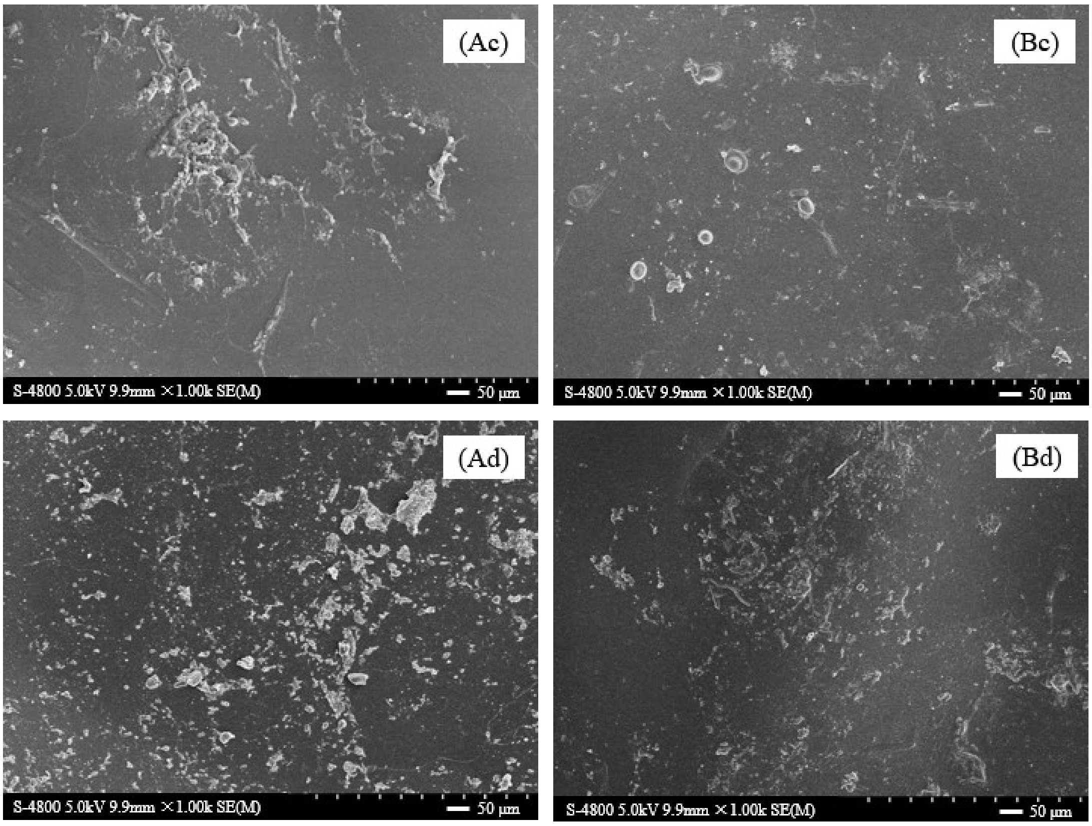


Fig. 4. FE-SEM images of the fouled (A) and adsorbed (B) FO membranes: (a) single organic fouling; (b) organic fouling + 1 mM Ca^{2+}; (c) organic fouling + 15 mM Ca^{2+}; (d) organic fouling + 35 mM Ca^{2+}. The scale bar equals to 50 µm.

We further examine the fouling structure to understand the impacts of the Ca^{2+} removal. Fouled FO membranes before and after the Ca^{2+} adsorption were analyzed by the CLSM observations. From Fig. 5, some aggregates of polysaccharides appeared on all the fouled FO membrane surfaces. The polysaccharide aggregates were formed assisting with the Ca^{2+} through forming “egg-box-shaped” gel networks. In addition, the aggregates of polysaccharides in some spots became more and larger with the increase of feed Ca^{2+} concentration, which was consistent with the increase of substratum coverage of polysaccharides (Table 1). It should be paid attentions that the deposited polysaccharide aggregates became less after adsorbing by the EDTA-based layer. It could be attributed to the fact that the Ca^{2+} was effectively removed from the fouling layer, resulting in a destruction of the alginate-Ca^{2+} gel networks supported by the reduction of substratum coverage of polysaccharides (Table 1). In fact, some polysaccharides also detached from the FO membrane surface accompanied by the destruction of the alginate-Ca^{2+} gel networks, which was evident by the decrease of polysaccharides on FO membrane surface after EDTA-based adsorption (Table 1). Thus, the mitigation of the fouling induced by the combination of alginate and Ca^{2+} with the EDTA-based adsorption layer was mainly attributed to the destruction of the alginate-Ca^{2+} gel networks and subsequent reduction of the amount of the deposited polysaccharides.

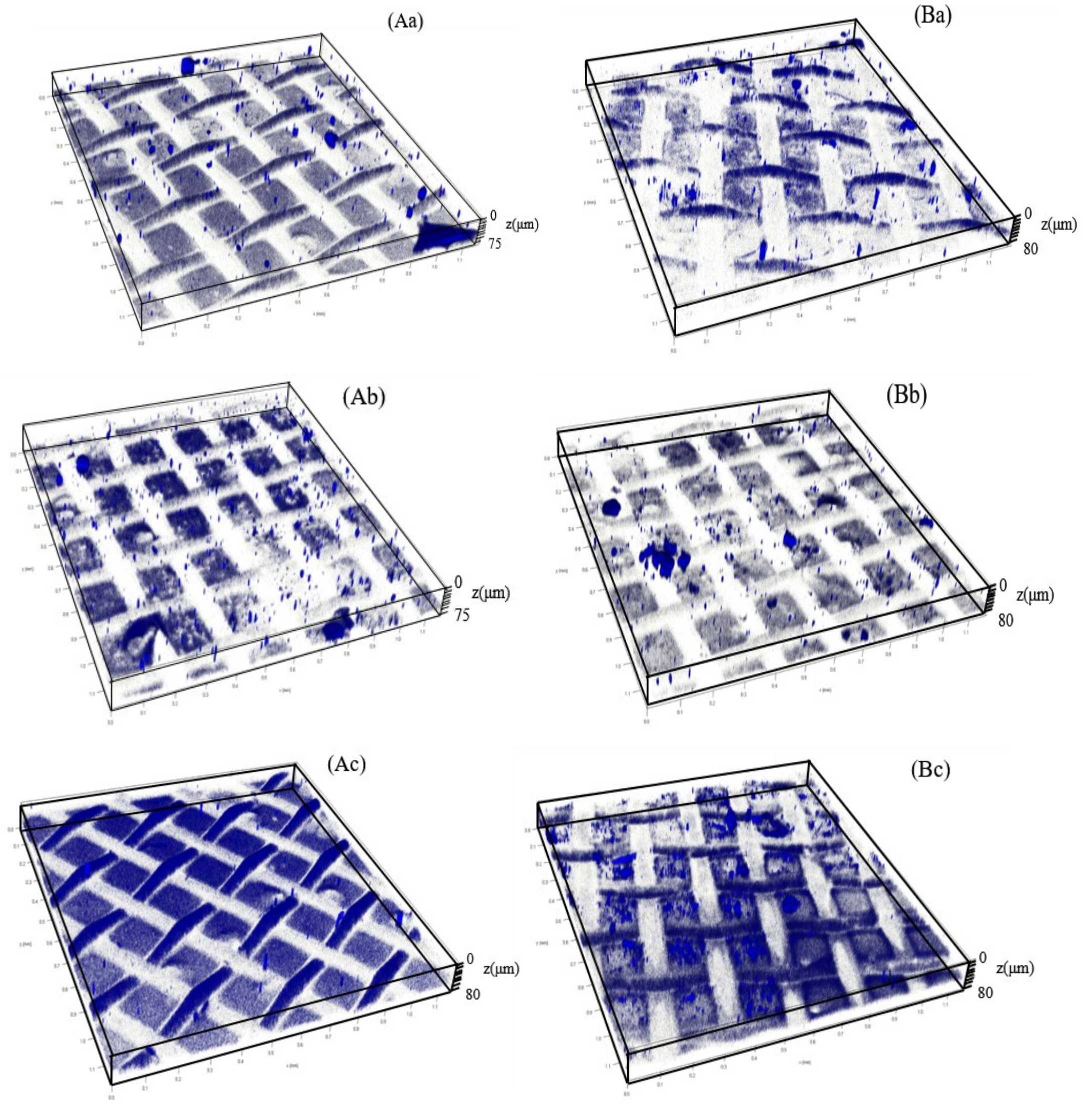


Fig. 5. CLSM images of the fouled (A) and adsorbed (B) FO membranes: (a) organic fouling + 1 mM Ca^{2+}; (b) organic fouling + 15 mM Ca^{2+}; (c) organic fouling + 35 mM Ca^{2+}.

Table 1 Structural parameters of the fouling layer before and after adsorption obtained from CLSM images via PHLIP[a].

	Fouling layer	Average amount of polysaccharide ($\mu m^3/\mu m^2$)	Substratum coverage (%)	Mean thickness (μm)
Before adsorption	organic fouling + 1 mM Ca^{2+}	14.78 ± 0.33	38.69 ± 1.48	52.34 ± 4.76
	organic fouling + 15 mM Ca^{2+}	15.84 ± 0.38	44.32 ± 0.93	55.89 ± 0.73
	organic fouling + 35 mM Ca^{2+}	18.03 ± 0.69	61.39 ± 0.64	57.54 ± 3.09
After adsorption	organic fouling + 1 mM Ca^{2+}	14.05 ± 0.24	33.86 ± 0.16	52.07 ± 2.75
	organic fouling + 15 mM Ca^{2+}	15.21 ± 1.01	41.35 ± 0.71	54.39 ± 1.71
	organic fouling + 35 mM Ca^{2+}	16.68 ± 1.25	52.40 ± 3.53	54.52 ± 1.59

[a] Values are given as mean values ± standard deviation (number of measurements: n=3).

3.4. Impacts of removing Ca^{2+} on cleaning efficiency

We also examine the physical cleaning efficiency after EDTA-based adsorption. Fouled and adsorbed FO membranes at the FS condition of sodium alginate and 35 mM Ca^{2+} were backwashed in the FO cell for 0.5 h with the 0.5 M NaCl as the FS and the DI water as the DS according to previous literature [11, 41]. There was no obvious variation of the normalized water flux for the fouled FO membrane after the backwashing; by contrast, it significantly increased from 0.71 to 0.89 for the adsorbed FO membrane after backwashing (Fig. S6). It suggested that the EDTA-based adsorption could enhance the physical cleaning of the fouled FO membranes. Such evidence strongly suggested that the EDTA-based adsorption not only mitigated the FO membrane fouling but also enhanced the membrane cleaning via changing the structure of the fouling layer.

3.5. Mechanisms on the in situ mitigating FO membrane fouling

With aforementioned experimental results, we conceptualized the mechanisms on the *in situ* mitigating FO membrane fouling (Fig. 6). For a fouled FO membrane with the alginate-Ca^{2+} gel networks, EDTA-based adsorption layer can directly destruct this structure by removing the alginate-bound Ca^{2+} via the chelation of the EDTA functional groups. As a result, after extraction of the alginate-bound Ca^{2+} from the organic fouling layer, the fouling layer structure became loose because of destruction of the alginate-Ca^{2+} gel networks and detachment of the deposited polysaccharides, both of which mitigated FO membrane fouling and further enhanced the cleaning efficiency. Moreover, the EDTA-based layer can be reused for adsorbing Ca^{2+} after being regenerated by HCl based on the results that the Ca^{2+} adsorption efficiencies of the fouled FO membrane at the FS condition of 200 mg/L sodium alginate and 35 mM Ca^{2+} were 93.51% and 90.03% for the new and the reused layers, respectively. However, considering the possible damage of HCl to its structure during several regenerations, its lifespan should be further evaluated in future works.

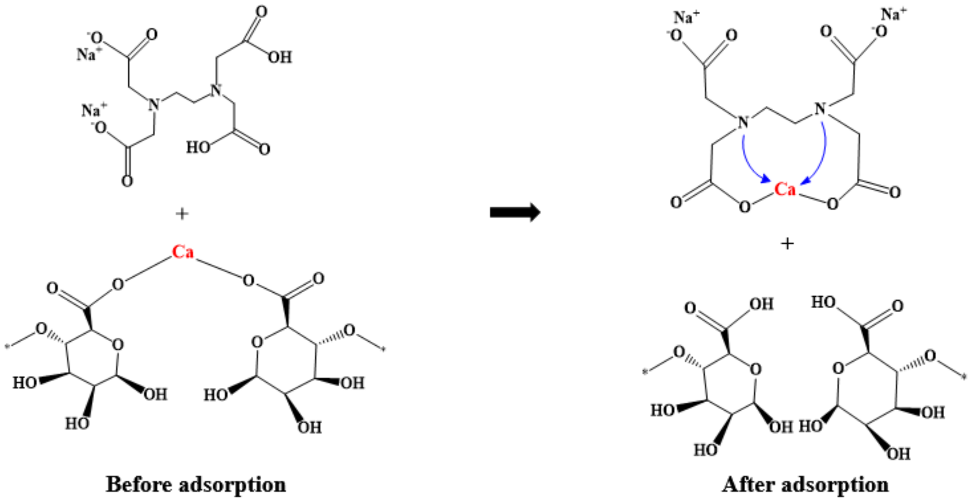


Fig. 6. Mechanisms of Ca^{2+} adsorption from the alginate Ca^{2+} gel networks by the EDTA-based layer.

4. Conclusions

The EDTA-based adsorption layer can effectively alleviate organic fouling of FO membrane via *in situ* adsorbing Ca^{2+} from the fouling layer. The success in alleviating organic fouling of the FO membrane demonstrated the feasibility of fouling control strategy via directly extracting Ca^{2+}

from the fouling layer. Extracting Ca^{2+} from the fouling layer played a critical role in altering membrane fouling layer structure, which shed light on novel fouling control and management. In addition, the EDTA-based layer exhibits higher adsorption efficiency owing to the stronger chelation of EDTA.

Acknowledgements

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Appendix A. Supplementary information

Detailed information on additional tables, figures and descriptions on calculating adsorption efficiency of Ca^{2+} via the EDTA-based adsorption layer can be found in the Supporting Information.

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