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Role of backwash water composition in alleviating ultrafiltration membrane fouling by sodium alginate and the effectiveness of salt backwashing

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ABSTRACT

To obtain a better hydraulic cleaning strategy for ultrafiltration (UF) membranes fouled by sodium alginate (SA), various types of backwash water, including UF permeate, ultrapure water, NaCl solution, $CaCl_2$ solution and SA solution, were compared with respect to hydraulic cleaning efficiency (HCE), hydraulically irreversible fouling index, Fourier transform infrared spectra analysis, normalized trans-membrane streaming potential coefficient and foulant release. The results indicated that UF permeate backwash or $CaCl_2$ solution backwash significantly decreased the HCE. On the contrary, NaCl solution backwash greatly alleviated the SA fouling, and backwashing with ultrapure water or organic compounds was also efficient when both Na^+ and Ca^{2+} were present in feed water. Moreover, the amount of released SA from the fouling layer during backwashing was closely related to that of released Ca^{2+}. Electric double layer release involved in backwashing with ultrapure water and SA solution, and ion exchange played significant roles in the effective hydraulic cleaning associated with Na^+. Further, monovalent salt backwashing was quite effective for a wide range of salt concentrations, and the approach was effective irrespective of monovalent salt types. Moreover, the results obtained from natural organic matter (NOM) and effluent organic matter (EfOM) further proved the efficiency of NaCl solution backwash.

1. Introduction

Ultrafiltration (UF) technology is increasingly applied in a variety of water treatment processes, including potable water production, wastewater reclamation, seawater or brackish water pretreatment [1–3]. In almost all these cases, membrane fouling caused by organic matter is a dominant operational problem [4]. The most problematic organics are macromolecular biopolymers such as extracellular polymeric substances (EPS) [5] which secreted by activated sludge or algae cells (especially during algal bloom periods) [6]. Polysaccharides are commonly used as representative of biopolymers as being the main constituent [7] and mostly approaching to the fouling behavior of EPS better than other constituents [8]. Sodium alginate (SA) has been previously used as representative of polysaccharides in waters to study its effect on membrane fouling [9–16].

The hydraulic irreversibility of UF membrane fouling, which is classified according to hydraulically cleaning, is rather problematic in long-term operation [17,18]. Compared to the well-documented researches on fouling by SA, systematic studies on fouling irreversibility are rarely reported. With exception of a few groups [11–13], many studies on SA so far have focused on single-cycle performance rather than multi-cycle operation. Meanwhile, most of bench-scale systems are also subjected to criticism because a backwash cycle was not included in these designs. Ye et al. [9]

Abbreviations: ANOVA, analysis of variance; CaBW, $CaCl_2$ solution backwash; DI, deionized; EDS, energy dispersive spectrometer; EfOM, effluent organic matter; HCE, hydraulic cleaning efficiency; HIFI, hydraulically irreversible fouling index; FTIR, Fourier transform infrared; MQBW, ultrapure water backwash; NaBW, NaCl solution backwash; NOM, natural organic matter; PES, polyethersulfone; RO, reverse osmosis; SA, sodium alginate; SABW, SA solution backwash; SA+Ca^{2+}, feed water consisting of SA and Ca^{2+}; SA+Na^++Ca^{2+}, feed water consisting of SA, Na^+ and Ca^{2+}; SEM, scanning electronic microscopy; SPC, streaming potential coefficient; TMP, trans-membrane pressure; UF, ultrafiltration; UFPBW, UF permeate backwash; w/o BW, without backwash

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stated that the reversibility of SA fouling changed during different filtration procedures, with the cake becoming more irreversible during the long-term subcritical filtration. Jermann et al. [10] performed dead-end filtration experiments with SA, showing that SA fouling was severe but mostly reversible. By investigating the influence of permeate flux on the irreversibility of SA fouling, Van de Ven et al. [12] demonstrated that the reversible filtration resistance at a low filtration flux turned to be irreversible at a high flux. Katsoufidou et al. [11] aimed at understanding of SA fouling and the efficiency of backwash technique, and stated that the reversibility of fouling increased with increasing Ca^{2+} concentration. Most scientific papers presented the filtration behavior at a constant pressure [10,11], but most industrial plants are operated under a constant flux. Hydraulic backwashing is important for improved understanding and control of hydraulically irreversible fouling, and for maximizing the benefits of UF technology [17].

The extent of irreversible fouling is largely dependent on the cleaning efficiency, which is closely related to the composition of the backwash water [19–21]. UF permeates have been widely used for backwashing in full-scale applications, but this approach is limited by the low backwash efficiency [19,20,22]. Most commonly, bench-scale studies on SA involving backwashing used deionized (DI) water as the backwash water for convenience [10–13]. Resosudarmo et al. [20] demonstrated that SA was easily removed from fouling layer by substituting UF permeate backwashing with DI water backwashing. Moreover, the effectiveness of backwashing with low-ionic strength water (e.g., DI water, reverse osmosis (RO) permeates) for cleaning UF membranes fouled during the filtration of natural water has also been reported by Li et al. [19,22]. The compositions in backwash water, such as organics, divalent cations (e.g., Ca^{2+}) and monovalent cations (e.g., Na^{+}), may play a great role in cleaning performance. Previous studies have demonstrated that backwashing with organic compound (e.g., UF permeates treated by dialysis) resulted in a high cleaning efficiency [19]. Li et al. [19,23] stated that the presence of cations (irrespective of Na^{+} or Ca^{2+}) in backwash water decreased the cleaning efficiency during filtration of canal water, and the authors attributed this to the charge screening effects of cations. However, Lee et al. [24] indicated that a high concentration of NaCl (0.1 mol/L) solution was effective for cleaning UF membranes fouled by hydrophilic organic matter. The positive effect of NaCl cleaning for removing hydrophilic gel fouling was also reported in published literature [25]. Obviously, the reported results in the previous literature are not always consistent, leaving a huge room for debate.

Therefore, to find a better backwash water composition for effective membrane cleaning, several types of backwash water, including UF permeate, ultrapure water, NaCl solution, $CaCl_2$ solution and SA solution, were tested in this study. Specifically, the work was performed with several objectives: (a) to evaluate the performance of hydraulic cleaning with various types of backwash water; (b) to explore the release of foulants and Ca^{2+} from the fouling layer for proposing mechanisms involving the backwashing process with different types of backwash water; and (c) to verify the effectiveness of monovalent salt backwashing on the control of membrane fouling by SA solutions as well as natural organic matter (NOM) and effluent organic matter (EfOM).

2. Methods and materials

2.1. Chemicals and materials

2.1.1. Chemicals and model foulants

Unless otherwise specified, all reagents and chemicals were analytical grade, and solutions were prepared by diluting with ultrapure water. NaCl, $NaNO_3$, Na_2SO_4, KCl, NH_4Cl, $CaCl_2 \cdot 2H_2O$, HCl and NaOH were purchased from Tianjin Benchmark Chemical Reagent Co., Ltd. (Tianjin, China). Ultrapure water (18.2 MΩ cm) was obtained using a Millipore water system (EMD Millipore Corp, USA). SA purchased from Sigma-Aldrich (USA) was used as the representative of polysaccharides in natural waters. The stock solutions (2 g/L) of SA was prepared by dissolving 2 g of SA into 1000 mL ultrapure water, followed by stirring for 24 h. The stock solution was stored at 4 °C in the dark. The polysaccharides (a main composition of biopolymers) in natural surface water (river, lake, canal, et al.) were usually detected in low concentration levels (less than 1 mg C/L) [26,27], so the SA solutions at low concentrations were also used in published literature [10,20,28]. According to published literature and our tests on the effect of SA concentrations on UF membrane fouling (see Fig. S2a, Supporting information), a working SA solution at the concentration of 2 mg/L (~0.8 mg C/L) was chosen.

2.1.2. Feed water and backwash water

In the most of the published literature, the ionic strengths (adjusting using NaCl) of SA solutions for UF fouling tests were 0–20 mmol/L [10,11,28,29], and the concentrations of Ca^{2+} typically used in SA fouling tests were 0–2 mmol/L [10–12,29]. According to published literature and our tests on the role of Na^{+} and Ca^{2+} in membrane fouling (see Fig. 2b and c, Supporting information), two levels of Na^{+} (0 and 10 mmol/L) were chosen to investigate the effect of ionic environment in feed solutions on membrane fouling and cleaning behaviors, and 0.5 mmol/L Ca^{2+} was employed in all SA feed solutions. Thus, two types of SA solutions with different types of cations were used as feed water: (a) 0.5 mmol/L Ca^{2+} ($CaCl_2 \cdot 2H_2O$) was added to the SA solution, and (b) 10.0 mmol/L Na^{+} (NaCl) and 0.5 mmol/L Ca^{2+} were both added to the SA solution. For convenience, the first SA solution is referred to as SA+Ca^{2+}, and the second SA solution is referred to as SA+Na^{+}+Ca^{2+}. The pH of each solution was adjusted to 7.5 using 1 mol/L NaOH or HCl. In order to ensure the solutions were mixed fully, the prepared solutions were stirred (300 r/min) for at least 2 h prior to use. In addition, to investigate whether the results of monovalent salt backwash obtained from SA solutions were still effective for natural waters, the efficiencies of NaCl backwash during filtration of NOM and EfOM were tested. NOM and EfOM were collected from Songhua River (China) and the secondary effluent of Wenchang wastewater treatment plant (Harbin, China) after filtration using a 0.45 µm cellulose ester filter (Taoyuan, China). The compositions of SA solutions and the main characteristics of NOM and EfOM were listed in Table 1.

When investigating the effect of backwash water composition, backwash water samples containing different compounds present in the UF permeate were prepared and used. In this study, five types of backwash water were employed, i.e., ultrapure water, NaCl solution, SA solution, $CaCl_2$ solution and UF permeate, referred to as MQBW, NaBW, SABW, CaBW and UFPBW, respectively, for convenience. The compositions of various types of backwash water are also listed in Table 1. The NaCl and $CaCl_2$ solutions were prepared by adding the appropriate amounts of NaCl and $CaCl_2 \cdot 2H_2O$, respectively, to ultrapure water. In the preparation of SA solution used for backwashing, the SA concentrations in the feed water were chosen to ensure that the SA content was equal to that in the permeates of each corresponding UF test. Besides, to compare with the filtration-backwashing results, UF tests without backwash (w/o BW) were also performed for both feed solutions.

To systematically evaluate the role of salt (including monovalent and divalent cations) concentrations in alleviating SA fouling, a serial of NaCl solutions (0.01–600 mmol/L, representing waters with different sources) and $CaCl_2$ solutions (0.01–0.50 mmol/L) with different concentrations were tested (Table 1).