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Towards a better hydraulic cleaning strategy for ultrafiltration membrane fouling by humic acid: Effect of backwash water composition

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ABSTRACT

As a routine measurement to alleviate membrane fouling, hydraulic cleaning is of great significance for the steady operation of ultrafiltration (UF) systems in water treatment processes. In this work, a comparative study was performed to investigate the effects of the composition of backwash water on the hydraulic cleaning performance of UF membranes fouled by humic acid (HA). Various types of backwash water, including UF permeate, Milli-Q water, NaCl solution, CaCl₂ solution and HA solution, were compared in terms of hydraulically irreversible fouling index, total surface tension and residual HA. The results indicated that Milli-Q water backwash was superior to UF permeate backwash in cleaning HA-fouled membranes, and the backwash water containing Na⁺ or HA outperformed Milli-Q water in alleviating HA fouling. On the contrary, the presence of Ca²⁺ in backwash water significantly decreased the backwash efficiency. Moreover, Ca²⁺ played an important role in foulant removal, and the residual HA content closely related to the residual Ca²⁺ content. Mechanism analysis suggested that the backwash process may involve fouling layer swelling, ion exchange, electric double layer release and competitive complexation. Ion exchange and competitive complexation played significant roles in the efficient hydraulic cleaning associated with Na⁺ and HA, respectively.

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Introduction

Worldwide water pollution has boosted the problem of water scarcity, especially for safe drinking water. Under this circumstance, more effective water treatment processes are required in potable water production. With pore sizes less than 20 nm, the ultrafiltration (UF) membrane, which can totally retain the pollutants such as colloids, protozoa, bacteria and virus (Jacangelo et al., 1997), has been one of the most attractive alternatives to the conventional water treatment process. However, the retained pollutants may

accumulate on the membrane surface, inevitably leading to severe membrane fouling. Membrane fouling would definitely boost energy demand and operating cost, making the membrane technology less cost-effective.

To address the problem of membrane fouling, a variety of measures have been taken, including membrane modification (Rana and Matsuura, 2010), pretreatment (Gao et al., 2011; Huang et al., 2009), hydraulic cleaning and chemical cleaning (Porcelli and Judd, 2010; Shi et al., 2014). However, membrane modification and pretreatment, which may retard the accumulation of foulants by improving the anti-fouling ability of

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membranes and the quality of feed water, respectively, cannot completely eradicate the fouling. Therefore, the cleaning of membranes is indispensable for the sustainable application of UF systems. As strong acids and alkalis are involved, the widely employed chemical cleaning is not only unfriendly to surrounding environment but also harmful to membrane life-span (Crozes et al., 1997). Therefore, it is essential to explore the hydraulic cleaning so as to minimize the frequency of chemical cleaning.

The hydraulic cleaning consists of two aspects, i.e. a backward hydrodynamic force related to backwash intensity and a dispersion potential associated with solution chemistry of backwash water. A variety of studies have been performed to optimize backwash conditions, and the cleaning performance proved to be improved at a higher backwash strength (Crozes et al., 1997; Kennedy et al., 1998). Nonetheless, the gains in membrane permeability by applying the stronger backwash intensity may be easily offset by the increase in water/energy consumption (Akhondi et al., 2014; Crozes et al., 1997; Kennedy et al., 1998). Thus, backwash water composition is still a fertile area to explore. To date, several types of backwash water have been investigated, including tap water (Chen et al., 2003), UF permeate (Abrahamse et al., 2008; Li et al., 2009, 2012b), nanofiltration permeate (Li et al., 2010), reverse osmosis (RO) permeate (Chen et al., 2003; Li et al., 2012a), deionized (DI) water (Abrahamse et al., 2008; Li et al., 2009, 2010, 2012a, 2012b; Resosudarmo et al., 2013) and some other types of water with different ions. In consideration of convenience, UF permeates have been widely used for backwashing in full-scale applications, but this approach is limited by the low backwash efficiency (Abrahamse et al., 2008; Li et al., 2009, 2012b). Previous studies have demonstrated that backwashing with UF permeate treated by dialysis resulted in a high backwash efficiency (Li et al., 2009). Compared with UF permeate, RO permeate exhibited much greater capacities in cleaning the membranes fouled during filtration of sea water or surface water (Li et al., 2012a; Ma et al., 2013). Moreover, Li et al. (2009, 2012a) verified the effectiveness of backwashing with DI water in fouling control via both pilot- and bench-scale UF experiments treating canal water as well as brackish water. However, as reported by Resosudarmo et al. (2013), backwashing with DI water was ineffective for removing fouling caused by humic acid (HA).

Obviously, the differences among the mentioned backwash water lie in the composition. So it is reasonable to propose a question about the backwashing of fouled membranes. Is more purer water necessarily better for backwashing? By investigating the effects of monovalent and divalent cations on the backwash performance, Li et al. (2011) found that the cleaning efficiency was impaired to some extent perhaps due to the charge screen effects. However, for UF membranes fouled by hydrophilic organic matter, Lee et al. (2001) demonstrated that NaCl (0.1 mol/L) was much more effective than DI water, with a cleaning efficiency even greater than that achieved by chemical agents (e.g., NaOH or citric acid). Moreover, by determining the intermolecular foulant–foulant adhesion, the exposure of fouled membranes upon salt solution could induce a structural breakup of cross-like gel layer by ion exchange (Lee and Elimelech, 2007). Apparently, the reported results in the previous literature are not always consistent, leaving a huge room for debate.

Existing studies involving the composition of backwash water mainly focused on controlling membrane fouling to treat specific natural water, and controversial results were typically related to the complexity and diversity of natural surface waters. Because the effects and mechanisms of various components (such as organics and cations) in removing the retained foulants during backwash have been unknown, the design of backwash water composition for specified target foulants has been under debate. There are more uncertainties regarding the case of a complex foulant matrix in natural water. Therefore, the effect of backwash composition on the alleviation of UF membrane fouling by HA was investigated. Overall, the objectives of this study were to find a better backwash water composition for effective membrane cleaning. Several types of backwash water, including UF permeate, Milli-Q water, NaCl solution, CaCl₂ solution and low-concentration HA solution, were used in hydraulic cleaning, and the effects of the compositions were discussed.

1. Methods and materials

1.1. Feed water and backwash water

Unless otherwise specified, all reagents and chemicals were analytical grade, and prepared by diluting with Milli-Q water. NaCl, CaCl₂·2H₂O, HCl and NaOH were purchased from Tianjin Benchmark Chemical Reagent Co., Ltd (Tianjin, China). Milli-Q water (18.2 MΩ·cm) was obtained from a Millipore water system (EMD Millipore Corp, Billerica, MA, USA). To better understand the relevant mechanisms involved in the backwashing process, HA, which was purchased from Sigma-Aldrich Co., LLC (St. Louis, MO, USA), was used as a model foulant to represent natural organic matter (NOM), following previous studies (Jermann et al., 2007; Yuan and Zydney, 2000). A stock HA solution (2 g/L) was prepared as follows. First of all, 2 g of HA was dissolved in 1000 mL of NaOH solution (pH = 12). Next, the solution was stirred for 24 hr, and the pH of the solution was adjusted to 7.0 using 1 mol/L HCl. Then, the stock solution was stored at 4°C in the dark. A HA concentration of 5 mg/L was chosen to simulate the organic matter content in surface water (Sutzkover-Gutman et al., 2010; Tian et al., 2013) and was prepared by diluting the stock solution with Milli-Q water.

Two types of HA solutions with different types of cations were used as feed water, as listed in Table 1. To ensure a sufficient amount of deposited HA for the backwashing tests, feed solutions containing Ca²⁺ (0.5 mmol/L) were employed. The typical concentrations of Ca²⁺ and Na⁺ were chosen following previous literature (Tian et al., 2013). The pH of feed water was adjusted to 7.5 using 1 mol/L NaOH or HCl. Meanwhile, backwash water samples containing different compounds that were in the UF permeate were prepared and used. The compositions of the various types of backwash water are also summarized in Table 1. The abbreviations MQBW, NaBW, HABW, CaBW and UFPBW represent backwashing with Milli-Q water, NaCl solution, HA solution, CaCl₂ solution and UF permeate, respectively. The NaCl and CaCl₂ solutions were prepared by adding the appropriate

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