



Interaction mechanisms of humic acid combined with calcium ions on membrane fouling at different conditions in an ultrafiltration system



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HIGHLIGHTS

- XDLVO approach can predict membrane fouling.
- Particle size and total surface energy of colloid influence membrane fouling.
- Increasing calcium ion concentration resulted in a larger colloid size.
- Increasing concentrations of calcium ions caused greater membrane fouling.
- Prechlorination reduced the interaction between calcium ions and humic acid.

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ABSTRACT

The Extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) approach was used to predict the mechanisms regarding the influence of humic acid together with calcium ions on membrane fouling in an ultrafiltration (UF) system. The polymeric UF membranes composed of polyvinylchloride (PVC) were selected to investigate membrane fouling. The results indicated that membrane fouling was aggravated when the total interaction energy caused by humic acid combined with calcium ions decreased. Calcium ions aggravated membrane fouling, compared with water samples without calcium ions. Increasing concentrations of calcium ions resulted in greater membrane fouling due to their chelation with humic acid. Increased pH influenced the interaction between calcium ions and humic acid, resulting in an increase in the total interaction energy, which reduced membrane fouling. Prechlorination reduced the interaction between calcium ions and humic acid, and resulted in an increase in the total interaction energy of the water sample, indicating that prechlorination may reduce membrane fouling, due to characteristic changes in organic matter.

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1. Introduction

Membrane technology has attracted considerable attention due to their capacity to produce a low-turbidity, pathogen-free filtrate from rivers, lakes and underground water supplies [1,2]. Nonetheless, membrane fouling is a serious impediment to use the low-pressure membrane systems as a substitute for conventional drinking treatment processes [3,4]. Previous studies have shown that natural organic matter (NOM) is the main cause of membrane fouling [5–8]. Humic acids are the predominant species in NOM, and are considered to be one of the major contributors to membrane fouling. Numerous studies have reported the physical and chemical aspects of membrane fouling caused by humic acid [9–11]. Previous studies have shown that membrane

fouling is affected by many factors, including the hydrodynamic and operating conditions, the characteristics of humic acid such as Zeta potential and particle size, and the pH and ionic strength of the feed water [12–14]. It is known that inorganic ions may contribute to the adsorption of organic colloids on membrane surfaces [15]. The adsorbed humic acid forms chelates with the inorganic ions which influence the behavior of membrane fouling [16,17]. Some studies have shown the effect of ionic strength on humic acid adsorption onto ultrafiltration (UF) membrane surfaces. Studies found that the adsorption of humic organic colloids increased with higher ionic strength of the feed water [13]. In addition, the pH value of feed water has been found to affect organic matter characteristics, permeability and retention, demonstrating that membrane fouling is pH-sensitive [18]. However, the mechanism of the influence of humic organic colloids combined with inorganic ions at different pH values on membrane fouling is still unclear.

The characteristics of humic acid may influence membrane fouling [19,20], and pretreatment has been used as a means of reducing membrane fouling due to the characteristic changes in organic matter. The

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development of effective pretreatment methods to transform humic matters is essential for improving the cost-effectiveness of membrane systems and for the broader application of membrane technology in water treatment systems. Several studies have developed potential pretreatment options for UF membrane fouling control including preoxidation [21,22], which has been widely investigated. Previous reports have demonstrated that oxidation pretreatment significantly mitigates organic fouling, which has been primarily attributed to changes in molecular characteristics [23]. Chlorine is widely used in water treatment facilities in China and prechlorination is a common pretreatment method. However, there are few publications available on the effects of prechlorination on membrane fouling when changing the interactions between humic organic colloids and inorganic ions.

Previous studies have shown that the Extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory can be used to predict membrane fouling in aqueous solutions [24]. The XDLVO theory can describe the attachment of a colloidal particle to a surface due to three interactions (as membrane fouling contributors): van der Waals (LW) interactions, electrostatic (EL) interactions and short-ranged acid–basic (AB) interactions [24,25]. The positive interaction force, between colloid and the UF membrane surface, represents a repulsive action to reject membrane fouling, while a negative one causes an attraction that aggravates fouling [26,27]. However, there is a paucity of available information on the interactions between humic organic colloids and inorganic ions and their effects on membrane fouling.

Calcium ion is a common inorganic ion in natural water, and its presence, especially its chelates with organic matter, may result in serious membrane fouling. The study by Seungkwan indicated that membrane fouling increased when calcium ions were added to humic acids [9]. Young found that only NOM and Ca^{2+} resulted in a fouling pattern similar to surface waters, while Mg^{2+} and Na^+ caused much lower fouling at the same ionic strength as the calcium solution, which indicated that calcium ions have an obvious effect on membrane fouling compared to other metal ions [28]. Ana demonstrated that the flux of UF rapidly declined in the presence of calcium ions [29], confirming the aggravated trend in membrane fouling caused by calcium ions. There is, however, a paucity of available information on the influence of interactions between humic matter and calcium ions on membrane fouling in different conditions, in relation to the XDLVO theory. In this study, the XDLVO approach was used to investigate the factors and mechanisms which influenced the interaction between humic acid and calcium ions causing UF membrane fouling.

2. Materials and methods

2.1. Theory

The XDLVO theory describes the total interaction energy, between a colloid and a membrane surface as the sum of LW, AB and EL interaction.

$$U_{mlc}^{\text{XDLVO}} = U_{mlc}^{\text{LW}} + U_{mlc}^{\text{EL}} + U_{mlc}^{\text{AB}} \quad (1)$$

where U^{XDLVO} is the total interaction energy between a membrane surface and a colloid immersed in water, U^{LW} is the LW interaction term, U^{EL} is the EL interaction term, and U^{AB} is the AB interaction term. The subscripts 'm', 'l' and 'c' correspond to the membrane, the bulk feed solution, and the colloid, respectively. The total interaction energy is calculated according to the described methods [24,30–32], expressed in mJ/m^2 .

2.2. Membrane and humic acid

Hollow fiber UF membranes, made of PVC, were provided by the Litree Purifying Technology Co., Ltd. (Suzhou, China) in this study. The UF membrane has a molecular weight cut-off (MWCO) of 50,000 Da

and an effective surface area of 0.133 m^2 . Detailed characteristics of the membrane used are summarized in Table 1.

The characteristics of analytical grade humic acid (Shijiazhuang, China) are summarized in Table 1. The powdered humic acid was dissolved in pure water to form a stock humic acid solution. This stock solution was stored in a refrigerator at $4 \text{ }^\circ\text{C}$ until use. The final concentration of humic acid in all water samples was $3 \text{ mg}/\text{L}$, expressed as dissolved organic carbon (DOC), which was determined by a TOC analyzer (Multi N/C 2100 TOC, Germany).

2.3. Concentration of calcium ions and pH values

The artificial water sample used in this experiment was composed of humic acid and calcium ions. Analytical grade calcium chloride (Shanghai, China) was used as the divalent calcium ion source. The calcium ion concentrations in the water samples were $100 \text{ mg}/\text{L}$, $200 \text{ mg}/\text{L}$ and $300 \text{ mg}/\text{L}$, respectively. The pH value of the water samples was 7.0 ± 0.2 .

The water sample with a calcium ion concentration of $200 \text{ mg}/\text{L}$ was used to investigate the role of pH on membrane fouling. The desired pH value was adjusted to 5, 7 and 9 by the addition $0.1 \text{ mol}/\text{L}$ HCl or $0.1 \text{ mol}/\text{L}$ NaOH, respectively.

2.4. Membrane fouling experiments

The experimental water samples were filtered by the ultrafiltration system, as shown in Fig. 1. The permeate flux was fixed at $40 \text{ L}/(\text{m}^2/\text{h})$ by using a peristaltic pump. Prior to the commencement of continuous UF, the trans-membrane pressure (TMP) (P_0) was measured by the filtration of pure water. The TMP of the sample (P_1) was compared with the pure permeate TMP (P_0), to provide a comparison between the different ultrafiltration conditions. The UF membrane system operated at a filtration cycle of 60 min, which was determined by the spot bench-scale trial. The following processes were performed at the end of each filtration cycle: first wash at $6 \text{ m}^3/\text{h}$ for 15 s, backwash at $8 \text{ m}^3/\text{h}$ for 30 s, and a second wash at $6 \text{ m}^3/\text{h}$ for 15 s. TMP may increase in the course of filtration, which reflects the resistance of the UF membrane due to membrane fouling. The constant-flux component of resistance-in-series model to determine resistance can be expressed as follows:

$$J = \frac{\Delta P}{\mu R} \quad (2)$$

where J is the permeate flux ($\text{L}/\text{m}^2/\text{h}^{-1}$), ΔP is the trans-membrane pressure (Pa), μ is the viscosity of permeate ($\text{Pa}\cdot\text{s}$), and R is the total (overall) filtration resistance (m^{-1}).

In the case of filtration, the following equation was used [33–35]:

$$R = R_m + R_f = R_m + R_{\text{rev}} + R_{\text{irr}} \quad (3)$$

where R_m is the intrinsic membrane resistance ($1/\text{m}$), determined from the experimental data relating to R in the filtration of pure water, and R_f is the total fouling resistance, which includes the physically-reversible fouling resistance (R_{rev} , $1/\text{m}$) and the physically irreversible fouling resistance (R_{irr} , $1/\text{m}$).

We can obtain the formula for calculating resistance of R_{irr} and R_{rev} :

$$R_{\text{irr}} = R_2 - R_m \quad (4)$$

$$R_{\text{rev}} = R_1 - R_2 \quad (5)$$

where R_1 is determined from the experimental data at the end of filtration, and R_2 is determined from the experimental data at the commencement of next filtration cycle after hydraulic cleaning. The range of water temperature was from $23 \text{ }^\circ\text{C}$ to $25 \text{ }^\circ\text{C}$.

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