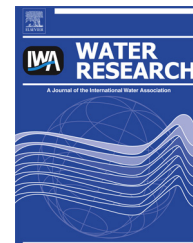




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Effect of low dosage of coagulant on the ultrafiltration membrane performance in feedwater treatment

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ABSTRACT

One of the critical issues for the widely application of ultrafiltration (UF) in water treatment is membrane fouling owing to the dissolved organic matter. The aim of the present study is to explore the effect of various particle sizes caused by low dosages of coagulant with dissolved organic matter on the UF membrane performance. Aluminum chloride was added to the synthetic water with the hydrophobic humic acid (HA), the hydrophilic bovine serum albumin (BSA) – a protein- and their 1:1 (mass ratio) mixture. The results showed that there was a critical dose of Al that could cause dramatic flux reduction by blocking the membrane pores after coagulating with HA/BSA. For HA or BSA, the critical dose of Al was relatively lower at pH 6.0 than that at pH 8.0. After coagulation, the flux decline caused by HA was slightly varied as a function of pH while that caused by BSA was greatly affected by pH. The flux decline caused by the 1:1 (mass ratio) HA/BSA mixture after coagulation was similar to that caused by HA after coagulation because BSA could be encapsulated by HA. In addition, the peak value of the molecular weight (MW) distribution of HA coagulated with Al was changed more drastically compared to that of BSA after filtration.

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1. Introduction

Ultrafiltration (UF) membrane has been widely used in drinking water treatment due to its excellent properties, such as removing a variety of waterborne viruses and much of the dissolved organic matter, etc. (Kimura et al., 2004; Jermann et al., 2007; Porcelli and Judd, 2010). However, a major impediment in the application of UF membrane technology is membrane fouling. The UF membrane could be polluted by three different ways: pore constriction, pore blocking and cake

layer (Jucker and Clark, 1994; Yuan and Zydney, 1999; Aoustin et al., 2001; Katsoufidou et al., 2007). The occurrence of different mechanisms of membrane fouling is related to particle size relative to membrane pore size (Huang et al., 2008) and other influence factors (Maximous et al., 2009; Katsoufidou et al., 2010; Masatoshi et al., 2011). The particle sizes which are close to the diameter of membrane pores can cause pore blocking, resulting in the most severe membrane fouling.

There are many different substances which co-exist in the complex natural water. Now more and more studies have

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focused on two or more contaminants in the solution, and some different pollution mechanisms have really been found compared to only one single pollutant in feed water (Jermann et al., 2007; Katsoufidou et al., 2010; Zazouli et al., 2010). Humic acid (HA) and bovine serum albumin (BSA) are used as model contaminants for their widely application in many studies (Tang and Leckie, 2007; Mo et al., 2008; Listiarini et al., 2009; She et al., 2009; Huber et al., 2011; Wang and Tang, 2011).

Coagulation remains the most common process to remove turbidity and nature organic matter (NOM) in water treatment, and Al/Fe salts are widely used as coagulants for pretreatment (Shon et al., 2004, 2005; Lee et al., 2009; Wang et al., 2009). Al-based or Fe-based salts could form a series of cationic hydrolytic species and weakly charged or uncharged precipitates after reacting with water (Stumm and Morgan, 1996; O'Melia, 1998). Al-Malack and Anderson (1996) determined the optimal coagulation conditions for wastewater which was 200 mg/L FeCl_3 at pH 9.0, and the chemical oxygen demand (COD) removal efficiency by this optimal dose of FeCl_3 was about 99.3%. Aguiar et al. (1996) found that the optimal dose of coagulant was 2.1 ± 0.2 mg Fe per mg of total organic carbon (TOC). In recent years, some studies have focused on finding an optimal dose in order to dramatically reduce the membrane fouling in the downstream treatment of membrane filtration. Tran et al. (2006) found that a specified dose of Al which could remove dissolved organic carbon (DOC) effectively also reduced the polyvinylidene fluoride (PVDF) micro-filtration (MF) membrane fouling. Lee et al. (2009) reported that there was an optimal PACl dosage with respect to fouling minimization, depending on the specific characteristics of wastewater. The optimal PACl dose was shifted to a higher level with increasing the ionic strength. They found that 0.0371 mM PACl (calculated as Al) was needed to achieve the highest UF membrane flux with a low ionic strength ($546 \mu\text{S}/\text{cm}$) while approximately 0.5 mM PACl could achieve the maximal flux when the ionic strength was much higher ($2390 \mu\text{S}/\text{cm}$). According to the results presented by Shon et al. (2005), there was an optimal dose of FeCl_3 which did not cause any flux decline during the whole operation of UF in 6 h. Finally they showed that at least 50 mg/L FeCl_3 was found to be necessary to avoid any significant flux decline and to obtain a better DOC removal efficiency. Up to now, more and more studies are focused on finding an optimal dose of coagulant to reduce the membrane fouling in the downstream treatment. However, few have been paid attention to the phenomenon that whether the membrane will be fully blocked by a critical dose of coagulant or not. Herein the key issue that we concern about is to know the membrane flux decline induced by the various particle sizes formed by HA/BSA coagulated with low dosages of Al.

This research focuses on the fouling mechanism of UF membrane at the low dosage coagulant. It aims at: (1) whether there is a critical dose of Al that can cause membrane flux reduce significantly or even fully block the membrane pores as a function of time, pH, etc. after coagulating with the hydrophobic HA or the hydrophilic BSA; if there is, then, (2) whether the critical dose of Al would be different for HA or BSA; and (3) what the critical dose of Al would be if the feed water is mixed by HA and BSA.

2. Materials and methods

2.1. Chemical reagents and materials

All chemicals were analytical reagent except for specific illustration and the deionized (DI) water was used for preparation of all stock solutions and membrane performance experiments. pH was adjusted by 0.1 M HCl or 0.1 M NaOH by pre-determined. Humic acids, sodium salt (HA, Aldrich, USA) was dissolved by DI water while BSA (Electrophoresis Pure, Sinopharm Chemical Reagent Co., Ltd, China) was dissolved by solutions of 0.15 M ionic strength phosphate buffered salt (Sean et al., 1993). Stock solutions of HA and BSA at a total concentration of 10 g/L were prepared. BSA working solutions were used within two days (Sean et al., 1993). Aluminum chloride ($\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, calculated as Al) was used as coagulant here and the stock concentration was 0.1 M. Working solutions were stored in the dark at 4 °C and all experiments were repeated for 2 or 3 times.

2.2. UF experiments

The UF membrane (PVDF) was used here with nominal molecular weight cutoff of 100 kDa. Each membrane was placed in DI water for at least 24 h to remove impurities and production residues. Immediately before the stirred cell test the DI water flux of the membrane was determined by filtering DI water through the membrane until reached a stable permeate flux. Normalized flux J/J_0 as a function of time was shown for the flux decline results from the stirred cell experiments, and J_0 was the initial membrane flux. Using UF stirred cell (Milipore, Amicon 8400) and keeping the pressure stable (0.1 MPa) by nitrogen gas, the fouling characteristics of the flat sheet UF membrane were studied by filtering dilute HA and BSA in the presence of different doses of Al in feed solution.

For the tests, the aluminum chloride stock solution was diluted in DI water (300 ml), in the UF stirred cell, with 0.1 M NaHCO_3 to give test solution. During the test, the pH of final solution was maintained by prior addition of a predetermined amount of 0.1 M NaOH or 0.1 M HCl. Afterward 10 ppm HA, 1:1_{ratio} 5 ppm HA/BSA mixture or 10 ppm BSA were formed by adding the corresponding stock solution to the feed water. Rapid mixing (250 rpm) lasted for 1 min while slow mixing (100 rpm) lasted for 14 min to allow floc growth to occur and taking out the stirrer carefully finally.

2.3. Analytical methods

pH was measured by Orion pH Benchtop; Variation of UF membrane flux as a function of time was recorded by data logger; Molecular weight distribution was determined by Gel Permeation Chromatography (GPC, Agilent Technologies, USA; Detector: UV₂₅₄; Column: TSK; Temperature: 25 °C); Particle size was measured by Zeta Sizer (Malvern Instrument, Nano Series, Nano-ZS, Model: ZEN3600); Ultrafiltration membrane pore size was analyzed by IB-FT, GmbH (POROLUX 1000, Germany).

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