

In situ investigation of combined organic and colloidal fouling for nanofiltration membrane using ultrasonic time domain reflectometry



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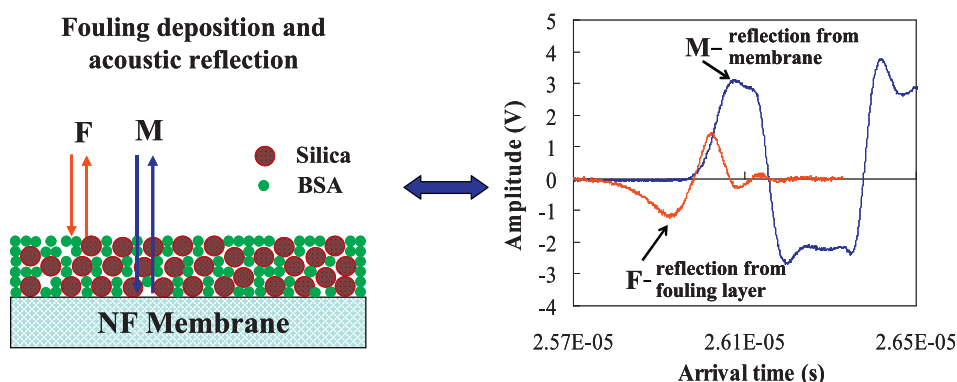
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HIGHLIGHTS

- UTDR was used to monitor the combined organic–colloidal fouling for NF membrane.
- Flux decline was greater for silica–BSA solution due to exceeding critical flux.
- Silica–BSA feed could induce denser fouling layer than silica–NaCl feed.
- Absorption of BSA on silica played a key role in formation of fouling layer.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 12 November 2014

Received in revised form 22 January 2015

Accepted 2 February 2015

Available online xxx

Keywords:

Ultrasonic time-domain reflectometry (UTDR)

Nanofiltration

Combined organic and colloidal fouling

Colloidal silica

Bovine serum albumin (BSA)

ABSTRACT

Ultrasonic time domain reflectometry (UTDR) was used to monitor the deposition of combined organic–colloidal fouling on a nanofiltration membrane. The fouling experiments were performed with different feed solutions: a mixture of 1000 mg/L silica and 1000 mg/L NaCl, a mixture of 1000 mg/L silica and 250 mg/L BSA, and 250 mg/L BSA alone, respectively. Results showed that the rate of flux decline obtained in the constant-pressure experiment with the mixture of silica + BSA was greater than that with the mixture of silica + NaCl and BSA alone. The acoustic measurements indicated that the fouling layer obtained from the combined organic–colloidal fouling was denser than that obtained from the colloidal fouling layer in the presence of NaCl. Furthermore, the mixed foulants rapidly deposited on the membrane surface in the early fouling phase, and then reached a plateau in the later fouling phase under a constant pressure operation, whereas the mixed foulants gradually deposited on the membrane surface as the fouling progressed under a constant flux operation. The main reason for the rapid foulant deposition during the initial fouling stage under constant pressure operation was because the initial flux was above the critical flux. The off-line AFM analysis and zeta potential measurements corroborated the ultrasonic measurements.

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1. Introduction

Nanofiltration (NF) is an attractive technology for brackish/seawater desalination and wastewater reclamation, as it can provide high multi-valent ion and organic contaminant rejection at much lower operating pressure than reverse osmosis (RO) [1]. However, membrane fouling is the major problem in the efficient operation of NF systems. Membrane fouling causes deterioration of both quantity and quality of the treated water and results in higher treatment cost. There are four main sources of fouling, namely colloidal materials, dissolved organics, inorganic precipitates, and microorganisms, of which organic and colloidal foulants are the most persistent problems in water and wastewater treatment [2]. To provide a better understanding of membrane fouling and fouling control, numerous studies have been performed to investigate the mechanisms of membrane fouling [3–8].

Most previous studies on NF membrane fouling have been focused on single foulants [3,4]. However, the fouling during water and wastewater filtration is always caused by more than one foulants with various particle sizes and surface characteristics, particularly colloidal materials and dissolved organic macromolecules. Additionally, there have been a limited number of studies on the combined fouling caused by inorganic colloids and dissolved organic matter simultaneously. The results exhibited very different fouling behaviors and mechanisms comparing with the fouling caused by the individual foulants [5–8].

Lee et al. [5] investigated the combined influence of natural organic matter (NOM) and colloidal particles on the fouling of high salt rejection NF membranes. They demonstrated that the rate of flux declined due to the combined fouling at the initial stage was greater than that caused by the colloidal particles and NOM individually. However, the discrepancy was reduced in the later filtration stages. This phenomenon was ascribed to the fact that the combined fouling was governed by not only cake-enhanced osmotic pressure but also NOM–calcium complexation. In a study on the combined fouling mechanisms of low salt rejection NF membranes, Li and Elimelech [6] found that the flux decline rate during the combined fouling was always higher than those based on the sum of colloidal and organic fouling alone. This synergistic fouling behavior was attributed to the hindered back-diffusion of foulants caused by the interactions between organic and colloidal foulants. Contreras et al. [7] further investigated the effect of the characteristics of organic foulants on the combined fouling behavior. They revealed that three hypothesized mechanisms were responsible for the enhanced flux decline in the presence of both colloidal materials and dissolved organic matter, including increased resistance of the mixed fouling layer, hindered back-diffusion, and organic foulant adsorption. These three mechanisms also had varying effects on combined fouling, and strongly depended on the molecular characteristics of the organic foulant. Mo et al. [8] studied the effect of bulk complexation between calcium and alginate on NF fouling. They found that there existed a critical calcium concentration with respect to initial fouling, below which increasing calcium concentration led to more severe fouling, and above which initial fouling was retarded.

Although the aforementioned studies provided insights into the combined fouling mechanisms, the actual mechanisms of fouling deposition have not been fully understood yet due to the lack of additional techniques to detect the foulant deposition processes. Therefore, the development of non-invasive, in-situ and quantitative technology to study the characteristics of the fouling layer growth is of great importance in order to better understand the synergistic effects of combined fouling on NF membrane, as well as to facilitate the strategies for prevention and control of membrane fouling.

For more than a decade ultrasonic time domain reflectometry (UTDR) has been successfully used to study membrane processes, especially NF or RO. Recently, Kujundzic et al. [9] comprehensively reviewed the use of UTDR technique for characterizing membranes and membrane processes that highlighted scaling measurement using UTDR for NF process. Mairal et al. [10,11] firstly employed UTDR technique for the real-time characterization of calcium sulfate (CaSO_4) fouling and

cleaning of flat-sheet RO membranes. The results showed a good correspondence between the change of ultrasonic signal amplitude and the development/removal of the scaling layer. Chai et al. [12,13] employed UTDR to study inorganic fouling and cleaning of commercial spiral-wound modules. In their studies, four particular peaks which emanated from specific surfaces were selected from the waveform responses as the observed signals to track their changes as the fouling progressed. Sanderson et al. [14] developed an UTDR technique for improving visualization of calcium carbonate fouling in RO modules. Based on the previous studies, Li et al. [15] further investigated the deposition processes of CaSO_4 on RO membranes using UTDR technique together with a simplified model. They reported that a fouling echo obtained in the time domain could indicate the actual state of a fouling layer on a membrane surface. However, it should be noted that these studies were mainly limited to a single foulant.

To further investigate the combined fouling behavior, Hou et al. [16] used UTDR to monitor the early-stage synergistic fouling of CaSO_4 and microorganisms on NF membranes. The results showed that bacteria could accelerate deposition of inorganic fouling on the NF membrane. Hou et al. [17] described the successful application of UTDR for investigating the impact of biofilm on CaSO_4 deposition on the NF membrane. The ultrasonic measurement indicated that the scaling layer of CaSO_4 formed in the presence of biofilm on the membrane surface was denser than that obtained from CaSO_4 alone. Recently, Sim et al. [18] have successfully adopted UTDR to detect biofouling by dosing of colloidal silica periodically as an acoustic enhancer. They confirmed that the colloidal silica had no effect on the viability of the bacteria using extrapolymeric substances (EPS) analysis, bacterial count analysis and confocal laser scanning microscopy (CLSM) analysis. Sim et al. [19] also used UTDR to monitor colloidal silica fouling and systematically investigated the effects of membrane operating parameters (crossflow velocity, flux, salinity and colloidal silica concentration) on the colloidal metastability. The results showed that lower crossflow velocity, higher flux and higher colloidal silica concentration could decrease the time required for the transition to a dense gel. Overall, the previous studies have demonstrated that UTDR is a powerful tool for the non-invasive monitoring of combined fouling.

Thus, the aim of the present study is to employ UTDR for the non-invasive real-time monitoring of combined organic and colloidal fouling during NF so as to fully understand the combined fouling mechanisms. Bovine serum albumin (BSA) and colloidal silica were used as model organic and colloidal foulants, respectively. Atomic force microscopy (AFM) and zeta potential analysis as independent methods were used to corroborate the UTDR measurements.

2. Materials and methods

2.1. Cross-flow NF and UTDR measurement systems

A schematic diagram of the cross-flow NF and UTDR measurement system in this study is shown in Fig. 1. The crossflow NF apparatus consisted of a 5 L feed tank for storage and supply of the feed solution, a diaphragm pump (GMB-78, QIAN, China) for pressurization of the feed solution, a peristaltic pump (BT 100-2J, Longer Precision Pump Co. Ltd., Baoding, China), a rectangular test module (custom-made, 200 mm × 110 mm) with an active membrane area of 60 cm² (120 mm × 50 mm), a hotplate/magnetic stirrer (Model 81-2, Sile, China), an electronic balance (PL6001-L/01, Mettler Toledo, Switzerland), a chiller (Model DLSB, Gongyi Yingyu Instrument Co. Ltd., China), a pulse dampener (custom-made) for minimizing pressure fluctuations, analog pressure gauge (Model YTN-60, ZOHA, China) and ball valves (Model Q11, CHANGRONG, Shanghai, China). The temperature of feed solution was controlled at 25.0 ± 1.0 °C using a hotplate/magnetic stirrer and a chiller, and measured with a digital thermometer (Model JM222L, Jinming Instrument Co. Ltd., China). The cross-flow membrane system had one feed stream entering the system, one rejection stream (brine) and one permeate stream (purified water) leaving the system. These three streams

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