



Fluorescent natural organic matter responsible for ultrafiltration membrane fouling: Fate, contributions and fouling mechanisms



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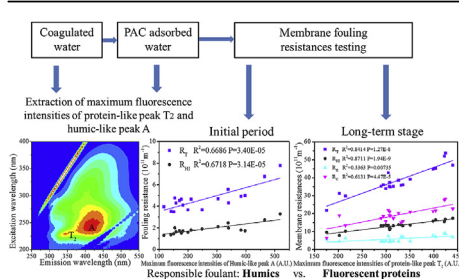
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HIGHLIGHTS

- The fate of fluorescent natural organic matter was depicted during the UF process.
- Humic-like substances contributed to the membrane fouling of initial period.
- Protein-like substances were highly concentrated, dominating the fouling behavior.
- A protein-governing cake layer was developed during the long run.
- Protein was strongly correlated with irreversible fouling in the long-term stage.

GRAPHICAL ABSTRACT



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ABSTRACT

Membrane fouling has been a main obstacle to the success of ultrafiltration (UF) technology. Recently, fluorescent natural organic matter (FNOM), including humic-like substances (HS) and protein-like substances, has been recognized as substances responsible for membrane fouling. In this study, the matrix of FNOM in natural river water was substantially modified by combined coagulation and powdered activated carbon adsorption to enhance the diversity of the FNOM matrix. Fluorescence excitation emission matrix spectroscopy was employed to characterize FNOM components during the UF process. The correlations between FNOM components of the feedwater and membrane fouling were evaluated for the initial period and long-term operation. Reliable correlations of the maximum fluorescence intensity of HS with initial membrane fouling indicated that HS were major foulants in the initial period. Furthermore, the protein-like component exhibited significant correlation with the concentration effect fouling ($R^2 = 0.6131$) and with irreversible fouling ($R^2 = 0.8711$). We found that the fouling mechanism changed from pore obstruction to a protein concentration polarization layer followed by protein cake layer filtration. Total fouling of the UF membrane over long-term operation was alleviated with powdered

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activated carbon (PAC) adsorption; however, the mitigation of irreversible fouling was dependent on whether PAC adsorbed protein-like substances.

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1. Introduction

Fluorescent natural organic matter (FNOM) has received increasing attention in the identification and monitoring of the dissolved organic matter (DOM) in various natural and engineered aquatic systems in the last decade (Henderson et al., 2009; Cohen et al., 2014). FNOM can be rapidly, sensitively, and selectively detected (even online) using a fluorescence spectroscopy technique (Peiris et al., 2013). Fluorescence spectroscopy was applied to predict the amount of trace organic compounds (Anumol et al., 2015), to monitor membrane bioreactor (MBR) performance (Liu et al., 2011; Galinha et al., 2012), and to improve the efficacy of anion exchange resins (Li et al., 2014). Notably, some components of FNOM (e.g., humic substances (HS) and protein-like substances), appear to be responsible for ultrafiltration (UF) membrane fouling, which causes a reduction in membrane permeability and increases the operation cost associated with high trans-membrane pressure (TMP) (Peiris et al., 2011; Peldszus et al., 2011). Therefore, understanding the fouling behavior of FNOM constituents is essential for prediction and control of UF membrane fouling.

Currently there is no clear consensus concerning the contribution of FNOM fractions to the membrane fouling (TMP increase or decline in filtration flux). Previous study demonstrated that the absorption of HS into the inner pores, or its accumulation on the membrane surface, caused severe decline in the flux; especially HS–Ca²⁺ aggregations resulted in exacerbated irreversible membrane fouling (Sutzkover-Gutman et al., 2010). However, it was suggested that fluorescent protein-like substances in natural surface water exhibit stronger correlations with UF irreversible fouling (on the basis of changes in TMP, permeability, or fouling index) than HS does (Peldszus et al., 2011; Peiris et al., 2013; Shao et al., 2014).

Moreover, there is still considerable disagreement over the extent to which the particles contribute to membrane fouling. In some studies, the synergistic fouling effect of particulate matter and natural organic matter (NOM) on UF membrane fouling was emphasized, due to modification of the colloidal surface properties and the formation of a combined fouling layer (Jermann et al., 2008; Peldszus et al., 2011; Philippe and Schaumann, 2014). However, some other studies reported that particulates have little impact on hydraulically irreversible fouling (Howe and Clark, 2002; Peiris et al., 2013; Tian et al., 2013). These inconsistent results were likely attributable to the diversity of NOM (Yuan and Zydney, 2000; Zularisam et al., 2007), the discrepancy in membranes (Lee et al., 2004) and variation of the solution chemistry (Jones and O'Melia, 2001; Sutzkover-Gutman et al., 2010). In view of these inconsistent results, the contribution of FNOM fractions to UF fouling should be evaluated to determine which NOM constituent should be primarily monitored and removed first under particulate-free conditions.

Coagulation has proven to be one of the most successful pre-treatment methods for full-scale applications for UF fouling control, due to the removal of particles, colloids and NOM (Huang et al., 2009). However, membrane fouling may not be completely ameliorated by coagulation because not all contaminants are reactive or adsorptive to the hydrolytic species formed by coagulants. For this reason, powdered activated carbon (PAC) adsorption has been combined with pre-coagulation to improve

the removal of organic contaminants and alleviate membrane fouling. Because different kinds of PAC exert different selective affinity to NOM components (Stoquart et al., 2012), several types of commercial PAC adsorbents, coupled with coagulation, were employed to modify the matrix of FNOM in this study. Accordingly, variation of the FNOM matrix could render insight into the fouling mechanisms of various FNOM components. Furthermore, the combined process could also limit the overlapping effects of the FNOM constituents in the UF feedwater, which affects the distinctiveness and stability of the fluorescence signature (Henderson et al., 2009).

Bench-scale studies have often been conducted to evaluate membrane fouling potential by providing a special given feed. An important suggestion was made that a few hours of initial filtration testing would not reveal the long-term membrane performance; therefore, a few days of operation was recommended (Nguyen et al., 2011). This is another reason for the inconsistent results mentioned above. Some previous reports indicated that the fouling mechanisms evolved from initial pore blockage to cake/gel filtration during the filtration of natural surface water (Yamamura et al., 2007) and domestic wastewater (Zheng et al., 2010), anaerobically treated wastewater (Ding et al., 2015). A cake layer might not develop after only a few liters of feedwater or in a filtration experiment only a few hours long. Therefore, filtration tests over a few hours and over a few days should be compared to describe initial and long-term FNOM fouling behavior, respectively, which has not been explored in detail in previous literature. Significantly, fouling behavior was dominated by a concentration polarization effect during ultrafiltration of model foulant bovine serum albumin (BSA) (Kwon et al., 2008). It was also reported that the concentration polarization of humic acids (Aldrich) led to increased HA transmission (Shao et al., 2016), and caused flux decline during filtration (Yuan and Zydney, 2000). However, the effect on membrane fouling of concentration polarization caused by fluorescent proteins and HS in natural water has still not been examined systematically.

The aim of this study was to investigate the roles of different fractions of FNOM in UF membrane fouling, including concentration polarization fouling and hydraulically irreversible fouling, during long term operation of UF membrane systems. The FNOM matrix in pre-coagulated surface water was modified using eight different types of commercial PAC. A bench-scale hollow-fiber UF system was adopted for use in twelve-hour and six-day filtration tests to assess filtration resistance to initial fouling and long-term fouling, respectively. Three-dimensional (3-D) Excitation emission matrix (EEM) fluorescence spectroscopy was employed to characterize the FNOM of the feedwater, the permeate, the concentrate, the backwash water, and the cleaning water during long-term filtration of the modified NOM matrix. Hence, the fate of FNOM during the whole filtration process was elaborated to better understand the fouling behavior of different fluorescent components. Correlations of FNOM fractions with initial fouling and long-term fouling were established to provide insight into the fouling potential of the FNOM components. The results provided a comprehensive understanding of membrane fouling mechanisms and suggested practical suggestions for monitoring fouling-responsible FNOM.

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