Identifying the major fluorescent components responsible for ultrafiltration membrane fouling in different water sources

Weiguang Sun[1,2], Jun Nan[1,*], Jia Xing[2], Jiayu Tian[1,*]

1. State Key Laboratory of Urban Water Resource and Environment, School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China
2. Heilongjiang Environmental Protection Academy of Science, Harbin 150056, China

ABSTRACT

Three-dimensional fluorescence excitation–emission matrix (EEM) coupled with parallel factor analysis (PARAFAC) was performed for a total of 18 water samples taken from three water sources (two lakes and one wastewater treatment plant (WWTP) secondary effluent), with the purpose of identifying the major ultrafiltration (UF) membrane foulants in different water sources. Three fluorescent components (C1, C2 and C3) were identified, which represented terrestrially derived humic-like substances (C1), microbially derived humic-like substances (C2), and protein-like substances (C3). The correlations between the different fluorescent components and UF membrane fouling were analyzed. It was shown that for the WWTP secondary effluent, all three components (C1, C2 and C3) made a considerable contribution to the irreversible and total fouling of the UF membrane. However, for the two lakes, only the C3 exhibited a strong correlation with membrane fouling, indicating that the protein-like substances were the major membrane foulants in the lake waters. Significant attachment of C1, C2 and C3 to the UF membrane was also confirmed by mass balance analyses for the WWTP secondary effluent; while the attachment of C1 and C2 was shown to be negligible for the two lakes. The results may provide basic formation for developing suitable fouling control strategies for sustainable UF processes.

© 2016 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

Introduction

Ultrafiltration technology is widely used in drinking water production and wastewater reclamation (Ke et al., 2013; Nakatsuka et al., 1996). Over the past 30 years, the ultrafiltration industry has grown steadily around the world due to the desire for high-efficiency removal of pollutants with low energy consumption, less maintenance, simple operation, and a large range of processing capacity (Ajmani et al., 2012; Laine et al., 2003). Despite the strong potential of ultrafiltration processes, membrane fouling is still one of the obstacles encountered in its application. Membrane fouling can increase the frequency of chemical cleaning (Tian et al., 2010; Wei et al., 2011), increase the energy consumption (Mamtani et al., 2014), reduce the service life of the membrane (Yu et al., 2014), and generally increase the cost of operation and maintenance (Huang et al., 2015).

The major challenge in control of membrane fouling is the identification of membrane foulants. Membrane foulants typically include particulate and organic matters. Particulate matters consist of inorganic particles and macromolecular colloidal organic matter, which primarily cause reversible membrane

---

fouling (Howe and Clark, 2002; Peiris et al., 2010). Organic matters are mainly composed of natural organic matter (NOM), which generally includes humic substances, proteins and polysaccharides (Her et al., 2002). NOM has been considered as the major UF membrane foulant in many studies (Lee et al., 2004; Peldszus et al., 2011). However, there are still some inconsistencies regarding the specific contribution of the different NOM subfractions to membrane fouling.

For example, Tian et al. (2013) found that the biopolymers, including both the polysaccharides and proteins, made the major contribution to UF membrane fouling, while the studies by Filloux et al. (2012) and Haberkamp et al. (2011) suggested the membrane fouling was mainly caused by protein-like substances. As for hydraulically irreversible fouling, Peiris et al. (2008) reported that both the humic-like and protein-like substances contributed to irreversible fouling. On the other hand, Peldszus et al. (2011) found that both the reversible and irreversible fouling on UF membranes can be solely attributed to protein-like substances, but not humic substances. Therefore, further studies are still required for a better understanding of the major NOM components that contribute to reversible and irreversible UF membrane fouling. In addition, further studies are also required to clarify if there is the difference in major membrane foulants among different water sources, such as lake water, river water, and secondary effluent from wastewater treatment plants, which is essential for developing suitable control strategies for sustainable UF operation in different applications.

Recently, three-dimensional fluorescence excitation–emission matrix (EEM) spectroscopy has been employed to identify the different fluorescent components in NOM due to the minimal need for sample pre-treatment and preparation, the ability to interpret NOM fluorescence properties, high instrumental sensitivity and the non-destructive nature of the technique (Chen et al., 2003; Peiris et al., 2008; Persson and Wedborg, 2001). However, fluorescence peak intensities cannot be used directly due to individual sample conditions. Thus, several data analysis tools, including principal component analysis (PCA) (Peldszus et al., 2011) and parallel factor analysis (PARAFAC) (Ishii and Boyer, 2012; Stedmon and Bro, 2008), have been developed to analyze the fluorescence matrices. The fluorescence EEM based methods can also been successfully used in the identification and quantification of major membrane foulants.

In this investigation, water samples were taken from different water sources, including two lakes and one waste water treatment plant (WWTP) secondary effluent. The UF membrane fouling behavior of different water samples was assessed, and the fluorescent components in the water samples were identified by using the EEM-PARAFAC method. The correlations between different fluorescent components and membrane fouling were analyzed, and the major foulants responsible for the reversible and irreversible UF membrane fouling were discussed.

1. Materials and methods

1.1. Raw water samples and UF experiments

The major membrane foulants for UF membranes were analyzed in three different waters: i) Jingbo Lake (JBL) and

Lianhua Lake (LHL), which belong to the Mudanjiang River basin; and ii) the secondary effluent from the Mudanjiang WWTP. A total of 6 water samples were collected for each of the three waters from May, 2014 to October, 2014. All the water samples were pre-filtered using a 0.45-μm cellulose ester membrane to remove particulate matter and larger colloids before the UF experiments, with the purpose of focusing on the fouling behavior of the organic substances. The samples were then stored at 4°C and used within 48 hr of collection. The water quality parameters of the raw water samples are listed in Table 1.

A dead-end ultrafiltration unit with an effective volume of 400 mL (Amicon 8400, Millipore, US) was used in the work; a schematic diagram of the system is shown in Fig. 1. UF membranes (OM100076, MWCO 100 kDa, America) with a cut-off molecular weight (MWCO) of 100 kDa were used in the UF membrane fouling experiments. The UF membrane was made of polyethersulfone (PES), with an effective membrane area of 41.8 cm². A fresh membrane was used in each of the UF experiments. The operating pressure was maintained at 0.1 MPa by a pressure controller. The permeate water was collected in a beaker on an electronic balance (SI-2002, Denver Instrument, China), which was connected to a personal computer equipped with a data acquisition system.

1.2. Reversible and irreversible fouling tests

In UF experiments, the pollutants in water samples tend to cause concentration polarization (CP) on the membrane surface, which also contributes to flux decline apart from membrane fouling. To avoid the interference of concentration polarization (CP) on the fouling resistance calculation, the following UF experimental protocols were adopted, as previously reported by Jermann et al. (2008) As shown in Fig. 2, three filtration cycles were performed for each water sample, and each cycle was divided into five steps: i) measuring the initial flux J_{100-1} using 100 mL of ultrapure water; ii) filtering 300 mL of the water samples; iii) filtering 100 mL of ultrapure water again to determine the end flux J_{100-2} of the first cycle; iv) backwashing the UF membrane with 50 mL of ultrapure water on the permeate side; and v) filtering 100 mL of ultrapure water through the membrane to determine the flux J_{100-3} after backwashing, which was used as the initial flux of the next cycle.

Table 1 – Raw water quality parameters from May to October 2014 (except June) and January in 2015.			
Parameters	WWTP	JBL	LHL
Temperature (°C)	11.5–25.6	13.6–20.8	12.1–19.2
pH	7.30–8.40	6.94–8.07	7.42–8.83
Turbidity (NTU)	4.76–20.48	45.6–188.3	10.8–43.6
DOC (mg/L)	4.76–15.13	3.927–9.326	3.933–8.767
Conductivity (ms/m)	48.6–213	17.9–109	7.33–103

WWTP: wastewater treatment plant; JBL: Jingbo Lake; LHL: Lianhua Lake.