



Evaluation of fouling in nanofiltration for desalination using a resistance-in-series model and optical coherence tomography

Jongkwan Park^a, Sungyun Lee^b, Jeongyeop You^a, Sanghun Park^a, Yujin Ahn^c,
Woonggyu Jung^c, Kyung Hwa Cho^{a,*}

^a School of Urban and Environmental Engineering, Ulsan National Institute of Science and Technology, UNIST-gil 50, Ulsan 44919, Republic of Korea

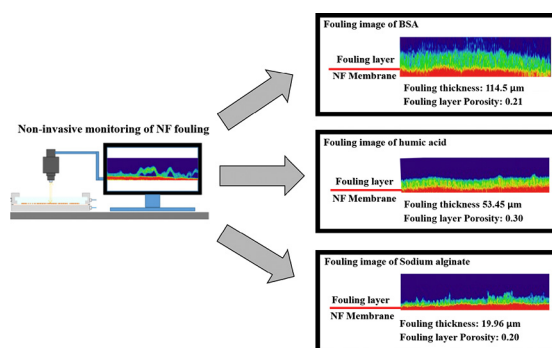
^b Department of Environmental Machinery, Korea Institute of Machinery and Materials, Daejeon 34103, Republic of Korea

^c School of Biomedical Engineering, Ulsan National Institute of Science and Technology, UNIST-gil 50, Ulsan 44919, Republic of Korea

HIGHLIGHTS

- Property of fouling layer was investigated by the membrane resistance value and fouling thickness.
- Humic acid, sodium alginate, and BSA fouling layers in brackish water were visualized via OCT.
- Gel/cake layer and adsorbed/pore blocking fouling types were distinguished by 2D OCT images.

GRAPHICAL ABSTRACT



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ABSTRACT

Resistance-in-series models have been applied to investigate fouling behavior. However, it is difficult to model the influence of morphology on fouling behavior because resistance is indirectly calculated from the water flux and transmembrane pressure. In this study, optical coherence tomography (OCT) was applied to evaluate the resistance of the fouling layer based on fouling morphology. Sodium alginate, humic acid, and bovine serum albumin (BSA) with high salts concentrations (conductivity: 23 mS/cm) were used as model foulants. At the same total fouling resistance, BSA showed the highest cake layer thickness (BSA (114.5 μm) > humic acid (53.5 μm) > sodium alginate (20.0 μm)). However, a different order was found for the cake layer resistance (BSA > sodium alginate > humic acid). This indicates that fouling thickness is not correlated with cake layer resistance. According to the Carman–Kozeny equation, fouling layer porosity decreased in the following order: humic acid (0.30) > BSA (0.21) > sodium alginate (0.20). In addition, we provided a specific value that was calculated using the ratio between the fouling thickness and cake layer resistance. The results show that alginic acid induced a stronger cake layer resistance, despite its thin fouling layer, whereas BSA showed a relatively low potential for inducing cake layer resistance. The results obtained in this study could be used for estimating and predicting fouling behavior.

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* Corresponding author.

E-mail address: khcho@unist.ac.kr (K.H. Cho).

1. Introduction

Nanofiltration (NF) consumes less energy than a reverse osmosis (RO), and achieves higher solute rejection than microfiltration (MF) and ultrafiltration (UF); Therefore, it has been used in various water treatment fields such as brackish water desalination, removal of micro-pollutants, and wastewater reuse (Fersi and Dhahbi, 2008; Haddada et al., 2004; Lee et al., 2008; Yu et al., 2001). In particular, NF membranes have been increasingly applied to pretreat water for desalination, because they can remove approximately 90% of divalent ions, which cause scaling (Park et al., 2016). However, membrane fouling is also an inevitable issue in NF that increases the operating cost and decreases the permeate flux. The different physicochemical properties of feed water produce various fouling types on these membranes such as inorganic, organic, and biofouling. Brackish water can include foulants that can develop into all of these types (i.e., salts and divalent ion, natural organic matter, and bacteria). It was also reported that the interaction between the various foulants enhances the concentration polarization and flux decline (Hoek and Elimelech, 2003; Lee et al., 2005). Thus, the fouling process is more complex for brackish water than it is for fresh water.

Various membrane fouling models such as gel layer model, osmotic pressure model, and resistance-in-series model have been developed to describe fouling behavior (Shirazi et al., 2010). In particular, the resistance-in-series model has been applied in many studies (Cho et al., 1999; Kaya et al., 2011; Rai et al., 2006) to describe resistances that cause flux decline. The total resistance is divided by two sub-resistances, namely the clean membrane resistance and the fouled membrane resistance. The clean membrane resistance is constant, whereas the fouled membrane resistance varies, and is related to the feed water quality and the membrane operating conditions. Specifically, the fouled membrane resistance is a factor of concentration polarization, cake/gel layer thickness, absorption, and pore blockage. The calculated resistances are used to quantify membrane fouling and to reveal a significant fouling mechanism in the filtration process.

To simulate the fouling process, various foulants are used, such as sludge, colloid, humic acid, bovine serum albumin (BSA), and dextran (Ang and Elimelech, 2007; Astaraee et al., 2015; Lim and Bai, 2003). Kaya et al. (2011) measured the resistances to describe membrane fouling in the treatment of cleaning-in-place wastewater originating from the production of liquid dishwashing detergent. They reported that the resistance of the gel layer was more important in reducing the permeate flux than that of concentration polarization or internal pore fouling. Ho and Zydney (2000) developed a new mathematical model for the permeate flux regarding to pore blockage and the growth of a protein cake layer. In this paper, BSA was used to model the protein foulant and the cake layer thickness was estimated by a resistance. Hong and Elimelech (1997) investigated the role of chemical and physical interactions in organic fouling of an NF membrane by using humic acid. Their experimental results indicated that the organic fouling thickness increased at high ionic strength because the presence of coiled humic macromolecules gives rise to a thick deposit layer.

Previous studies have reported that membrane resistance can be used as a practical indicator of fouling thickness. However, this has a drawback, in that the resistance value takes into account not only fouling thickness but also pore blockage and foulant adsorption. It is possible that the thickness is lower than the estimated value when pore blocking and foulant adsorption strongly affect the flux decline. In addition, it cannot show the specific fouling height with unit (e.g. μm).

Conventionally, the membrane fouling layer has been examined by conducting a membrane autopsy. This method provides sufficient samples for characterizing the membrane performance and the foulant through physical, chemical, microbiological, and microscopic examination (Dudley and Darton, 1996). However, membrane autopsy requires several sample preparation steps prior to analysis, such as cutting, coating, and drying. These steps can damage and alter the sample. To overcome this problem, non-destructive fouling characterization

techniques, such as electrical impedance spectroscopy (EIS), ultrasonic time domain reflectometry (UTDR), magnetic resonance imaging (MRI), X-ray microimaging, and direct observation through membrane (DOTM), have been developed (Gao et al., 2014; Li et al., 2017; Valladares Linares et al., 2016).

Optical coherence tomography (OCT) is a type of DOTM technique that has been applied to obtain high-resolution cross-sectional images of transparent samples such as tissues in biological studies (Huang et al., 1991). The practical aspects of OCT extend its application range to physics, optics, materials and environmental science (Tomlins and Wang, 2005; Valladares Linares et al., 2016; Wagner and Horn, 2017). Recently, OCT has been applied to investigate membrane fouling. The growth of the fouling layer, biofouling dynamics including formation, structure and detachment, and fouling quantification have been studied (Dreszer et al., 2014; Li et al., 2016; Park et al., 2018).

In this study, fouling in an NF membrane used for the desalination of brackish water was evaluated by a resistance-in-series model and OCT. Humic acid, sodium alginate, and BSA were used as the representative foulants in brackish water. Flux decline experiments were conducted with the synthesized brackish water containing different foulants, and 2D images of membrane fouling were obtained by using OCT at the same membrane resistances. The dependence of the different fouling mechanisms on the foulants' properties was investigated through comparisons of membrane resistance value and fouling thickness.

2. Materials and methods

2.1. Preparation and analysis of feed water solutions

Humic acid (Sigma-Aldrich, USA), sodium alginate from brown algae (Sigma-Aldrich, USA), and BSA (Sigma-Aldrich, USA) were used as model foulants. Table 1 shows the characteristics of the model foulants in terms of their typical shape, molecular weight, zeta potential value, dissolved organic carbon (DOC) concentration, and specific UV absorbance (SUVA). Each of the foulants were dissolved in synthetic brackish water (SBW) (conductivity: 23 mS/cm), which was prepared by dissolving 15 g/L of a commercial sea salt (Reef Plus marine salt, Aqua ocean, China) into deionized (DI) water. Because a typical NF membrane system uses the UF or MF membrane for pretreatment to remove particulate matters, the feed water was filtered using a 0.45 μm membrane (Advantec, Japan) prior to NF membrane fouling tests. The foulant concentration of DOC in the feed water was 90 mgC/L. For all feed water solutions, the pH values were adjusted to 7.7 by the addition of HCl or NaOH as needed. Ion chromatography (ICS-90, Dionex, CA, USA) using an AS14 (250 \times 4 mm, Dionex, CA, USA) and a CS12A column (250 \times 4 mm, Dionex, CA, USA) was performed to measure the cation and anion concentrations in the SBW. The DOC concentration was measured using a total organic carbon analyzer (TOC-V CPH, Shimadzu, Japan). The result was applied to the SUVA value using the UV absorbance value at 254 nm obtained with an ultraviolet-visible spectrometer (UV-1601, Shimadzu, Japan). pH and conductivity were measured using Orion 250A (USA) and Orion 115 (USA) instruments.

2.2. NF fouling experiments

A thin-film composite NF membrane (MWCO: 210 Da, NE90, Toray, Japan) was used to evaluate the membrane resistances. A rectangular filtration cell was constructed from stainless steel and had a Pyrex window to facilitate OCT scanning. Its dimensions were 150 mm \times 250 mm \times 74 mm, and the window dimensions were 140 mm \times 42 mm. A flat membrane sheet was installed, and the channel dimensions were 8 cm \times 18 cm \times 0.3 cm. The active area of the cell was 144 cm² (8 cm \times 18 cm). The applied transmembrane pressure (TMP) and feed flow rate were 20 bar and 0.5 L/min, respectively. The temperature of the feed was kept at 20 °C. The fouling experiment was operated in the recycling mode that the permeate water was returned to the feed

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