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Dynamics of biofouling development on the conditioned membrane and its relationship with membrane performance

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ABSTRACT

In this study, the impacts of a conditioning layer on the dynamics of biofouling development were investigated through batch biofouling experiments under the RO cross-flow filtration process. Fouled RO membranes extracted over the course of biofouling were prepared for measurements of extracellular polymeric substances (EPS) and total organic carbon (TOC) and confocal laser scanning microscopy (CLSM) observations. A close relationship between the biofilm dynamics and permeate flux and salt rejection were delineated. The patterns of membrane performance change demonstrated that the dominant mechanism governing the biofouling effects varied with the biofouling stage: concentration polarization dominated the early stage of biofouling, while fouling resistance was the dominant mechanism at the later stage. In addition, compared with the virgin membrane, the conditioning layer led to a severe deterioration in permeate flux and salt rejection. The results of biofilm dynamics showed the enhancement of bacterial initial attachment and biofilm maturation by the conditioning layer. Finally, there were increased biomass accumulation measured by TOC and the ratio of live to dead cells is higher on the bottom than that on the top of the biofilm on the conditioned membrane. In conclusion, conditioning layer had great influence on the counts of attached cells in the early stage of biofouling, but on the EPS concentration in the later stage of biofouling.

1. Introduction

As a new technology, reverse osmosis membranes have been widely applied in seawater desalination, drinking water treatment and wastewater reclamation [1]. However, fouling issues, including organic, inorganic, and colloidal fouling and biofouling, limit the widespread application of membrane technology, resulting in a deterioration in water quality and increased operating and maintenance costs [2]. Biofouling is recognized as the most destructive fouling and the most difficult to control because of the motion, secretion and multiplication characteristics of bacteria. Biofouling due to biofilm formation consists of layers of microorganisms embedded within extracellular polymeric substances (EPS) [3]. It has been reported that biofouling is still unavoidable even though 99% of bacteria are removed during pretreatment [4]. Consequently, a better understanding of biofouling development and determining effective control measures are of practical significance.

In general, there are four sequential steps of biofilm formation: (i) adsorption of nutrients and organic matter in the feed solution to the surface resulting in the formation of a so-called conditioning layer, (ii) initial reversible/irreversible adhesion of pioneer planktonic microorganisms onto conditioned surfaces, (iii) maturation of biofilms by the multiplication and metabolism of initial attachments, and (iv) detachment of the biofilm [5]. Initial bacterial adhesion is a critical stage in the overall process [6], and it is a prerequisite for the biofouling of membranes that is affected by various factors, such as the membrane surface properties and conditioning layers [7,8]. Conditioning layers are believed to have a significant impact on initial bacterial deposition, primarily through modifying the membrane surface properties, such as the hydrophobicity, charge and roughness of the membrane surface [9]. Furthermore, a conditioning layer not only changes the membrane properties but also provides a metabolically favorable environment for bacterial cells due to enhanced nutrient

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availability at that surface [10]. Baek et al. [11] reported that biofouling of the conditioned membrane finally caused a greater flux decline and higher polysaccharide concentration compared with the biofouling of a virgin membrane due to higher concentration of bacteria attached to the conditioned membrane. Generally, most researchers focus on the initial bacterial adhesion or only the total biomass concentration at the end of biofouling experiments in terms of the conditioning layers, ignoring the details and mechanisms governing the influence of such layers on the dynamics of biofilm development [12,13]. However, a good understanding of the dynamics of biofilm growth on the conditioned membrane is important for elucidating the mechanisms that are responsible for the effects of biofouling and for the development of biofouling control and cleaning strategies. Therefore, it is important to draw a well-supported conclusion regarding how the conditioning layer influences the overall biofouling formation process and subsequent membrane performance.

As is widely recognized, biofouling leads to a decline in membrane flux and salt rejection [14]. There are two well-acknowledged mechanisms that govern the loss in membrane performance [15,16]. On the one hand, microorganisms on the membrane surface hinder the back diffusion of salt, which leads to the "biofilm-enhanced concentration polarization" phenomenon and thus a decrease in permeate flux and salt rejection. On the other hand, EPS contributes to the decline in membrane permeate flux by increasing the hydraulic resistance to permeate flow. Dreszer et al. [17] revealed that performance loss in a membrane filtration system is caused by the biofilm resistance due to biofilm maturation and compaction and that biofilm thickness and EPS concentration are closely related to the fouling resistance. Moreover, it has also been reported that the decline in membrane performance is not predominantly caused by biofilm resistance for NF and RO systems and suggested that concentration polarization may play an important role [18]. These inconsistent results may be attributed to the different conditions for biofilm formation and the lack of studies on the dominant mechanism changes during biofouling. Therefore, the relationship between the membrane performance behaviors and the dynamics of biofilm development requires further investigation.

The objectives of this study are (1) to investigate the effects of a conditioning layer on the overall process of biofilm development and (2) to further explore the relationship between membrane performance behaviors (permeate flux and salt rejection) and the biofilm dynamics (biofilm thickness, live/dead cells, EPS and biomass concentration) with filtration time. Batch biofouling experiments with *Pseudomonas aeruginosa* terminated at different time points were conducted in an RO membrane system to investigate the biofilm dynamics on the conditioned membrane. Scanning electron microscopy (SEM) and CLSM images and EPS measurements provided visual and quantitative data regarding the evolution of the biofilm. The impact of the conditioning layer on the biofilm dynamics was elucidated through comparison with the biofouling of a virgin membrane. The biofilm dynamics were then related to the patterns of permeate flux and salt rejection variations, and the mechanisms of biofouling effects were delineated and discussed.

2. Materials and methods

2.1. RO membrane system, bacterial strains and growth media

To simplify the study and make it more controllable, we used synthetic wastewater as the feed water, and a representative species, i.e., *Pseudomonas aeruginosa*, to simulate biofouling experiments, which could be found elsewhere [16]. The synthetic wastewater was prepared freshly before every experiment and the composition was as follows: 1.16 mM $C_6H_5Na_3O_7 \cdot 2H_2O$, 0.94 mM NH_4Cl, 0.45 mM KH_2PO_4, 0.5 mM $CaCl_2 \cdot 2H_2O$, 0.5 mM $NaHCO_3$, 2.0 mM NaCl and 0.6 mM $MgSO_4 \cdot 7H_2O$. The final pH of this solution was 6.9. The model strain *Pseudomonas aeruginosa* (ATCC 27853) was incubated in 90 mL of LB broth overnight (optical density of 1 at 600 nm) to a final concentration of 10^9 cells/mL. Afterwards, 10 mL of *P. aeruginosa* was centrifuged for 10 min at 8000 rpm and then re-suspended in the synthetic water for three replicates. The washed *P. aeruginosa* was inoculated into the feed reservoir containing 1 L of synthetic wastewater to achieve an initial cell concentration of 10^7 cells/mL.

A cross-flow RO system using a flat sheet membrane (DOW 1812-50, PA) with an effective area of 24 cm^2 was used for the fouling experiments (Fig. S1). The constant operating conditions for all the fouling protocols including transmembrane pressure (TMP), cross-flow velocity, and temperature in lab-scale cross-flow RO system were 0.85 Mpa, 0.15 m/s, and 25 °C, respectively ($Re \approx 150$). The filtrate flux was calculated by weighing the permeate water using a digital balance connected to a personal computer. The permeate and retentate were circulated back to the feed tank to maintain a constant feed water quality. Samples from the feed reservoir and permeate were collected for the conductivity measurement during the filtration.

2.2. Biofouling experiment protocol

The experimental protocol for membrane fouling investigation is shown in Fig. 1. Firstly, before each experiment and prior to inserting the RO membrane into the cell, we disinfected the RO

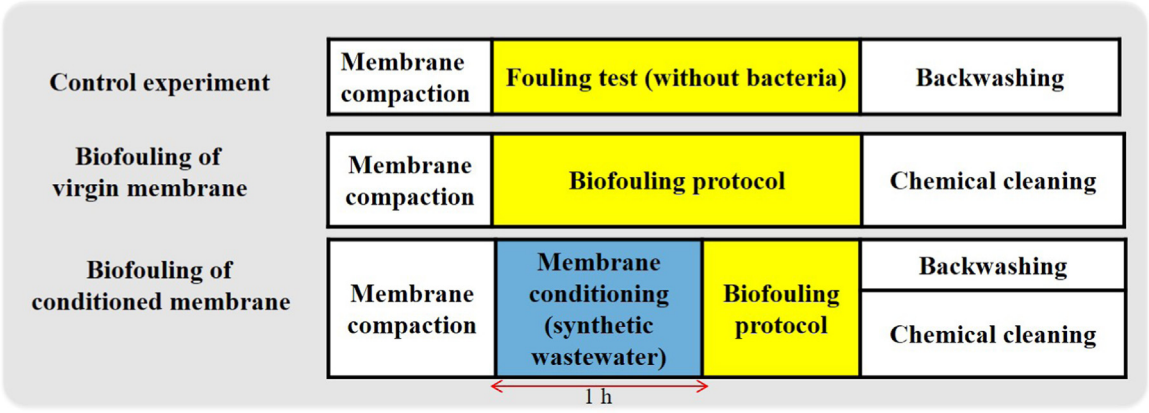

Fig. 1. Fouling experiment protocol.