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Influence of surface hydrophilicity on polytetrafluoroethylene flat sheet membrane fouling in a submerged membrane bioreactor using two activated sludges with different characteristics

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ABSTRACT

This study focused on how membrane surface hydrophilicity affects fouling in a membrane bioreactor. A lab-scale membrane bioreactor incorporating immersed polytetrafluoroethylene flat-sheet membrane modules with different contact angles (approximately 70° and 130°) was operated using the inocula activated sludge taken from a lab reactor and from a municipal wastewater treatment plant (WWTP). The reactor also contained a polyvinylidene difluoride flat-sheet membrane module as a reference. The hydrophobicity of the WWTP sludge was higher than that of the lab-reactor sludge. Results showed that when lab-reactor sludge was used, increases in transmembrane pressure (TMP) for the hydrophobic membrane with a high contact angle occurred more rapidly than for the hydrophilic membrane with a low contact angle. In contrast, when the WWTP sludge was used, TMP increases for the hydrophilic membrane occurred more rapidly than for the hydrophobic membrane. These results indicate that the effects of surface hydrophilicity on membrane fouling depend on the hydrophobicity of the activated sludge. When the WWTP sludge was used, the hydrophobic membrane exhibited much higher cake layer resistance than did the hydrophilic membrane. Formation of a dense cake layer on hydrophobic polytetrafluoroethylene membranes may deter adhesion of foulants on membrane surfaces.

1. Introduction

Membrane bioreactors (MBRs) have been increasingly used in wastewater treatment to minimize solid–liquid phase separations often encountered in conventional activated sludge clarifiers [1]. MBR systems have the advantages of operating at high concentrations of mixed liquid suspended solids (MLSS), generating less excess sludge, and allowing treated water to be reused more readily [2]. In addition, the biological nutrient removal (BNR) processes using MBRs can be attractive because the plant footprint is reduced by the absence of settling tanks [1]. However, in MBR systems, membrane fouling is a major problem that negatively affects the permeability of the membrane and increases operating and maintenance costs—effects that pose major obstacles to widespread MBR applications [3,4]. Despite the extensive efforts taken to understand the causes of membrane fouling, considerable confusion still exists because of the complexity and variability of membrane foulant responses to foulant–membrane and foulant–foulant interactions [5–7].

Major membrane foulants are believed to be extracellular polymeric substances (EPSs) that originate from bacterial cell lysis, microbial metabolites, and unmetabolized wastewater components [8]. Foulants consist of several classes of organic macromolecules (>300 kDa) and micromolecules (<1 kDa) [9,10], including polysaccharides, proteins, nucleic acids, (phospho) lipids, and other polymers [8]. The EPS in activated sludge encapsulates bacterial cells and can be extracted and categorized as extractable EPS [3]. EPSs are also released from microbial aggregates into the water phase; these are referred to as soluble microbial products (SMP) [11]. Proteins and carbohydrates are considered to be the two major components of EPS, and interest has focused on determining the relationship between fouling and the ratio of EPS proteins and carbohydrates (*P*/*C*). According to the literature, the *P*/*C* value is usually greater than unity when fouling

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occurs [12–16]. However, no clear relationship between *P* and *C* in reactors and membrane fouling in MBRs has been found in pilot-scale tests [17].

Membrane characteristics such as construction material, pore size and distribution, roughness, and hydrophilicity are considered to be important factors that affect interactions between membranes and foulants [3]. Furthermore, membrane fouling is believed to be more severe in hydrophobic than in hydrophilic membranes because of hydrophobic interactions between solutes, microbial cells, and membrane material [18–21]. In MBR processes, hydrophobic flocs lead to a high propensity for flocculation and weak interactions with hydrophilic membranes [3]. However, only a few studies [22] have focused on the relationship between membrane hydrophilicity and the hydrophobicity of seed-activated sludge in MBRs. Further, few experiments have been conducted, particularly in submerged MBRs, to understand how sludge hydrophobicity and membrane hydrophilicity might affect membrane fouling.

The objective of this study was to develop a better understanding of how membrane surface hydrophilicity/hydrophobicity and feed biomass characteristics, including hydrophobicity, can affect filtration performance. In this study, we operated a lab-scale MBR that incorporated immersed polytetrafluoroethylene (PTFE) flat-sheet membranes with different hydrophilicities and a polyvinylidene difluoride (PVDF) flat-sheet membrane with hydrophilicity similar to that of a PTFE membrane. The inocula of activated sludge were taken from a lab reactor and from a municipal wastewater treatment plant (WWTP). Particular attention was given to the carbohydrate and protein levels in the water phase (i.e., carbohydrate and protein in SMP), which were monitored over time before and after permeation of the immersed membrane.

2. Materials and methods

2.1. Membrane characteristics and experimental MBR setup

Table 1 lists characteristics of the symmetrical hydrophobic and hydrophilic PTFE flat-sheet membranes (Nippon Valqua Industries, Ltd., Tokyo, Japan) and asymmetrical hydrophilic PVDF flat-sheet membranes (Toray Industries, Inc., Tokyo, Japan). All PTFE flat-sheet membranes were laminated with polyethylene terephthalate (PET) and then attached to both sides of membrane modules; the membranes were separated from the modules by spacers (Figs. 1a and b). The PVDF flat-sheet membrane, which is widely used in MBRs, was also attached to both sides of the same membrane modules as the PTFE membranes. The total surface area of a membrane module was 0.029 m^2 (0.12 m × 0.12 m × 2 sides). A lab-scale MBR was configured with a reactor tank having a 21-L working volume and incorporating three immersed flat-sheet membrane modules (Figs. 1c and d). The filtrate was recovered using a roller pump with “8-min-on” and “2-min-off” suction modes. Constant filtrate flux was maintained by adjusting the pump rotation rate. To investigate the influence of membrane surface characteristics on membrane fouling, the transmembrane pressure (TMP) generated by filtration was measured periodically with pressure gages (AP-51A, Keyence, Osaka, Japan). Air for washing membrane surfaces was continuously supplied at 1 L min^{-1} from each of three diffusers located directly below the membrane modules.

2.2. Synthetic wastewater and operating conditions

Concentrated synthetic wastewater (SWW) was prepared with the following contents (all values in g L^{-1}): glucose, 8.81; bacto peptone, 4.28; and KH_2PO_4, 0.396. The concentrated SWW was made with milli-Q water and sterilized by autoclaving (121 °C, 20 min). A level sensor, which was connected to a roller pump to provide tap water and a microtube pump MP-3 (Eyela, Tokyo, Japan) to provide the SWW, was used to maintain a constant water level (21 L) and a constant substrate feed rate (glucose, 2.94 g day^{-1}; bacto peptone, 1.43 g day^{-1}; and KH_2PO_4, 0.132 g day^{-1}). The MBR was operated twice with different inocula: once for 70 days (operation 1) and a second time for 17 days (operation 2). The inoculum for operation 1 was obtained from a 40-L lab-scale batch reactor, fed with the same SWW as described above for more than 10 years. For operation 2, the inoculum was taken from a return sludge tank at a conventional municipal wastewater treatment plant in Yokohama (Japan).

Processing was interrupted twice in operation 1 (days 37 and 58) and once in operation 2 (day 12), when the modules were removed from the MBR and washed. Thus, the operational period was divided as follows for operation 1: Run 1 (days 0–37), Run 2 (days 37–58), and Run 3 (days 58–70); for operation 2: Run 1 (days 0–12), Run 2 (days 12–17). The MBR was operated at room temperature (operation 1, 19.2–24.8 °C in Run 1, 16.7–19.2 °C in Run 2, and 15.2–16.7 °C in Run 3; operation 2, 18.1–20.6 °C). To preserve the original characteristics of the inocula, the pH was not controlled but monitored with a pH meter (HM-21P with GST-2729C probe, TOA-DKK, Tokyo, Japan). Values of the pH were as follows: for operation 1, 7.1–7.4 in Run 1, 7.1–7.4 in Run 2, and 7.3–7.5 in Run 3; for operation 2, 6.9–7.0. Filtration fluxes and HRTs were controlled at 26.0 L m^{-2} h^{-1} (LMH) and 27.8 h (days 0–13 in operation 1), and 34.7 LMH and 20.9 h (days 13–70 in operation 1 and days 0–17 in operation 2), respectively. The SRT was uncontrolled with no biomass wasted, so the MLSS level increased from 6800 to 8300 mg L^{-1} in operation 1 and from 5560 to 6680 mg L^{-1} in operation 2.

2.3. Monitoring supernatant of mixed liquor and membrane permeate

During both operations, carbohydrate and protein levels in the supernatant of mixed liquor and the membrane permeate were monitored. The mixed liquor and the membrane permeate were collected from the reactor tank and each membrane module once

Table 1
Properties of membranes used in this study.

Membrane	Nominal pore size		Surface porosity (%)	Surface roughness (nm)	Membrane thickness (μm)	Pore morphology
Material	Specs (μm)	Bubble point (μm)				
PVDF	0.08	n.a.	7[a]	7.4 × 10	320[a]	Asymmetric
PTFE (hydrophobic)	0.3	0.30	85	1.3 × 10^2	52[b]	Symmetric
PTFE (hydrophilic)	0.3	0.18	n.a.	1.3 × 10^2	50[b]	Symmetric

n.a.: Not available.

[a] The value was reported by van der Marel et al. [31].

[b] Total membrane thickness including laminated PET.