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Effects of hydrophilicity/hydrophobicity of membrane on membrane fouling in a submerged membrane bioreactor

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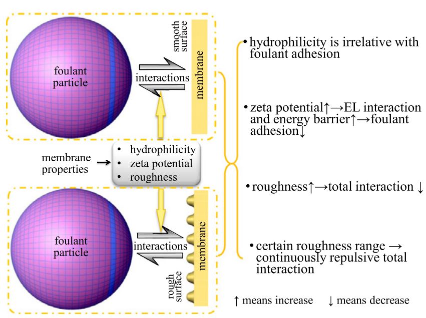
HIGHLIGHTS

- Membrane hydrophilicity is not directly relevant to interactions with flocs.
- Increasing surface charge highly raises electrostatic double layer interaction.
- Total interaction is continually repulsive when roughness is 300 nm.
- Strategy of improving membrane hydrophilicity cannot mitigate sludge adhesion.

GRAPHICAL ABSTRACT



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ABSTRACT

The interfacial interactions between sludge foulants and four different types of membranes were assessed based on a new combined calculation method. Effects of membrane surface hydrophilicity/hydrophobicity on the interfacial interactions were investigated. It was found that, membrane surface hydrophilicity/hydrophobicity was not directly relevant to the interfacial interactions with sludge particles. Increasing membrane surface zeta potential could significantly increase the strength of the electrostatic double layer (EL) interaction and the energy barrier. For membrane with a surface roughness of 300 nm, the total interaction was continuously repulsive in the separation distance coverage of 0–4 nm in this study. The results suggest that, under conditions in this study, designing membranes with a high zeta potential and certain roughness can significantly mitigate membrane fouling, whereas, the strategy of improving membrane surface hydrophilicity cannot alleviate sludge adhesion in the membrane bioreactor.

1. Introduction

Investigation of membrane fouling has been longstanding interest for the research community concerned with membrane bioreactor (MBR) technology since MBR was invented over four decades ago (Lesjean et al., 2011; Chen et al., 2012b; Wei et al., 2013). Numerous studies have indicated that sludge adhesion to form a cake layer is the main cause of membrane fouling in MBRs (Wang et al., 2007; Wang and Wu, 2009; Zhang et al., 2013). Therefore, considerable efforts have been made to understand and control sludge adhesion process in MBRs.

A number of factors, including hydrodynamic conditions, sludge properties and membrane properties, could affect the sludge adhesion process (Lin et al., 2014b). Hydrophilicity/hydrophobicity is a

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Nomenclature

D	closest distance between a particle and a planar surface (nm)
$f(r, \theta)$	local amplitude directly below the circular arc as a function of the position of the differential circular arc defined by r and θ
h	separation distance between two planar surfaces (nm)
e	electron charge (1.6×10^{-19} C)
k	Boltzmann's constant (1.38×10^{-23} J K^{-1})
ΔG	interaction energy per unit area (mJ m^{-2})
R	radius of foulant particle (μm)
r	radius of differential circular ring on particle surface (μm)
s	roughness of membrane surface (nm)
U	interaction energy between membrane surface and particle (kT)
Greek letters	
$\varepsilon_r\varepsilon_0$	permittivity of the suspending liquid (C V^{-1} m^{-1})
γ	surface tension parameter (mJ m^{-2})
κ	reciprocal Debye screening length (nm^{-1})
λ	decay length of AB interactions in water (0.6 nm)
ϕ	contact angle (°)
θ	angle of the circular arc in the circular ring
ξ	zeta potential (mV)
Superscripts	
AB	Lewis acid–base
EL	electrostatic double layer
LW	Lifshitz-van der Waals
tol	total
+	electron acceptor
−	electron donor
Subscripts	
f	foulant particle
h_0	minimum equilibrium cut-off distance (0.158 nm)
l	liquid
m	membrane
s	solid
w	water

primary membrane property. It is generally believed that hydrophilic membrane corresponds to lower membrane fouling potential than hydrophobic one (Kim et al., 2004; Weis et al., 2005; Santos and Judd, 2010). Based on this belief, improving membrane surface hydrophilicity was considered as an important strategy to mitigate membrane fouling in MBRs (Yu et al., 2005; Liu et al., 2013; Zhang et al., 2014a), although no solid theoretical base to support this belief. However, inconsistent results were also reported. For example, Choo and Lee (1996) found that the most hydrophobic polyvinylidene fluoride (PVDF) showed the smallest fouling tendency than the polysulfone (PSF) and cellulose acetate (CA) membranes. Chen et al. (2012a) reported that the flux decrease rate of the membranes followed the order CA > PVDF > polyether sulfones (PES) membranes, although CA membrane was most hydrophilic among them. The inconsistent results lead to the difficulty in understanding the exact roles of membrane hydrophilicity/hydrophobicity in sludge adhesion and membrane fouling. Literature analysis also showed that most of previous studies focused on the overall fouling behavior rather than the anatomized fouling steps such as sludge adhesion (Xiao et al., 2011).

Recent studies have reported that sludge adhesion to form a cake layer was mainly resulted from the interfacial interactions between sludge foulants and membrane surface (Feng et al., 2009; Hong et al., 2013; Su et al., 2013). While hydrodynamic forces forward the foulants close to membrane surface, it is the short-ranged interfacial interaction forces that are responsible for eventually binding the foulants to membrane surface (van Oss, 1995; Hong et al., 2013). The interfacial interactions comprised of Vander Waals (LW), electrostatic double layer (EL), and acid–base (AB) interaction could be generally described by the extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory (van Oss, 1995). By using XDLVO approach, Wang et al. (2013) and Zhang et al. (2014b) presented a plausible explanation for the membrane fouling behavior caused by soluble microbial products (SMPs) and pH, respectively. Therefore, assessment of these interfacial interactions through XDLVO theory may provide a route map to track the exact roles of membrane hydrophilicity/hydrophobicity in sludge adhesion. However, most previous studies simply assumed an infinite smooth surface for the studied membranes (Feng et al., 2009; Wang et al., 2013). In fact, most commercial membranes used in MBRs were significantly varied in surface morphology or roughness (Mahendran et al., 2011; Chen et al., 2012a). It is expected that the interfacial interactions between sludge foulants and rough membrane surface are more complicated than those regarding smooth membrane surface. Fortunately, a novel method developed by the authors of this study allows to quantitative calculation of the interfacial interactions between sludge foulants and rough membrane surface (Lin et al., 2014a). A quantitative assessment of these interfacial interactions would shed significant lights on sludge adhesion and membrane fouling. Nevertheless, neither study assessed the effects of membrane hydrophilicity/hydrophobicity on the interfacial interactions between sludge foulants and rough membrane surface in MBRs.

This study aims to theoretically and experimentally assess the effects of membrane hydrophilicity/hydrophobicity on the interfacial interactions and sludge adhesion process in an MBR. A submerged MBR (SMBR) setup operated at stable period was continuously run to supply the sludge samples. A series of membranes with known surface properties were adopted for the calculation of the interfacial interactions.

2. Methods

2.1. Experimental setup

A lab-scale submerged MBR (SMBR) treating synthetic municipal wastewater was continuously run for over 200 days. The SMBR contained a reactor with 60 L effective volume, where a flat sheet PVDF membrane model with 0.1 m^2 effective filtration area was vertically located. Air flow rate and membrane flux were about 180 $m^3_{air}/m^3_{permeate}$ and 30 L/(m^2 h), respectively. The sludge retention time (SRT) was approximately 45.5 d. The synthetic municipal wastewater used in this study possessed a composition as follows: 300 mg COD/L glucose plus the following mineral medium: $(NH_4)_2SO_4$ (27 mg N/L); KH_2PO_4 (7 mg P/L, 9 mg K/L); $NaHCO_3$ (23 mg Na/L, 50 mg CO_3/L); Na_2CO_3 (46 mg Na/L, 50 mg CO_3/L); $MgSO_4$ (7 mg Mg/L); $CaCl_2$ (6 mg Ca/L); $FeCl_3$ (4 mg Fe/L); $ZnCl_2$ (0.11 mg Zn/L); $MnSO_4$ (0.04 mg Mn/L); $CuSO_4$ (0.03 mg Cu/L); $CoCl_2$ (0.1 mg Co/L) and $NaMoO_4$ (0.02 mg Na/L, 0.07 mg Mo/L). In this study, activated sludge obtained from a sequencing batch reactor (SBR) for real municipal wastewater treatment was used as inoculum. The sludge samples obtained during stable operation period of SMBR were used for interfacial interaction assessment.