



Thermodynamic analysis of membrane fouling in a submerged membrane bioreactor and its implications



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HIGHLIGHTS

- AB interactions predominated in the total interactions of membrane-foulants combination.
- Proteins play an essential role in adhesion process.
- Sludge adhesion to membrane encountered a repulsive interaction energy barrier.
- Small flocs possessed higher attractive interaction energy per unit mass.
- Initial sludge adhesion reduced energy barrier, facilitating the following adhesion.

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ABSTRACT

The thermodynamic interactions between membrane and sludge flocs in a submerged membrane bioreactor (MBR) were investigated. It was found that Lewis acid–base (AB) interaction predominated in the total interactions. The interaction energy composition of membrane–sludge flocs combination was quite similar to that of membrane–bovine serum albumin (BSA) combination, indicating the critical role of proteins in adhesion process. Detailed analysis revealed the existence of a repulsive energy barrier in membrane–foulants interaction. Calculation results demonstrated that small flocs possessed higher attractive interaction energy per unit mass, and therefore adhered to membrane surface more easily as compared to large flocs. Meanwhile, initial sludge adhesion would facilitate the following adhesion due to the reduced repulsive energy barrier. Membrane with high electron donor surface tension component was a favor option for membrane fouling abatement. These findings offered new insights into membrane fouling, and also provided significant implications for fouling control in MBRs.

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1. Introduction

Membrane bioreactors (MBRs) is increasingly becoming an important innovation in biological wastewater treatment due to its distinct advantages over conventional activated sludge (CAS) systems, including higher biomass concentration, reduced footprint, less sludge production, and highly-improved effluent quality (Le-Clech et al., 2006; Zhang et al., 2006; Hasar, 2009; Lin et al., 2009; Meng et al., 2009; Santos and Judd, 2010; Wu and He, 2012). However, membrane fouling, which would increase operational and maintenance costs, highly limited the universal application of MBRs (Le-Clech et al., 2006; Zhang et al., 2006; Wang and Li, 2008; Meng et al., 2009; Lin et al., 2011; Wu and He, 2012).

It is generally believed that membrane fouling in MBRs is initiated by the adhesion of one or more bacteria and/or flocs to the membrane surface, followed by the growth and multiplication of the sessile flocs, which can eventually form a cake layer on the membrane surface (Le-Clech et al., 2006; Meng et al., 2009). Cake layer formation was considered as the main form of membrane fouling in MBRs (Wang and Li, 2008; Lin et al., 2009). Efforts were therefore extensively devoted into identification of the foulants affecting cake layer formation. Biomass sludge was initially believed as the major factor determining cake layer formation on membrane surface. Thereafter, inorganic foulants, such as struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$), were reported to play a significant role in the formation of the strongly attached cake layer in MBRs, especially in anaerobic MBRs (Choo and Lee, 1996). Later, extracellular polymeric substances (EPS) surrounding sludge cells were considered to be mainly responsible for sludge attachment on membrane

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Nomenclature

a_f	flocs radius (μm)
d	separation distance (nm)
e	electron charge ($1.6 \times 10^{-19} \text{ C}$)
k	Boltzmann's constant ($1.38 \times 10^{-23} \text{ J K}^{-1}$)
n_i	number concentration of ion i in solution
ΔG	interaction energy per unit area (mJ m^{-2})
T	temperature (298.15 K)
U	interaction energy between separation surface and particle (kT)
z_i	valence of ion i

Greek letters

$\epsilon_r \epsilon_0$	permittivity of the suspending liquid ($\text{C V}^{-1} \text{ m}^{-1}$)
γ	surface tension parameter (mJ m^{-2})
κ	reciprocal Debye screening length (nm^{-1})
λ	decay length of AB interactions in water (nm)
θ	Contact angle ($^\circ$)
ζ	zeta potential (mV)

Superscripts

AB	Lewis acid–base
EL	electrostatic double layer
LW	Lifshitz–van der Waals
Tol	Total
XDLVO	extended Derjaguin–Landau–Verwey–Overbeek
+	electron acceptor
–	electron donor

Subscripts

d_0	minimum equilibrium cut-off distance (0.158 nm)
f	foulant
l	liquid
m	membrane
s	solid
w	water

surface. In the last decade, much attention has been paid on soluble microbial products (SMP) and other forms of organic matter in the liquid phase (Zhang et al., 2006; Liang et al., 2007; Meng et al., 2009). Studies have indicated that these substances may act as a “glue”, facilitating formation of an apparent slime layer (Liang et al., 2007; Lin et al., 2010). In recent years, Wang and Li (2008) found that biopolymer clusters (BPC) was largely responsible for the high filtration resistance of the cake layer. While these studies largely extended understanding of membrane fouling, the underlying mechanisms regarding why and how these foulants affect cake layer formation appeared not well reported. Meanwhile, attempts have been made to characterize the cake layer in MBRs (Meng et al., 2007; Hwang et al., 2008). It has been frequently observed that resuspended cake layer had smaller particle size distribution than that of sludge suspension, indicating that small flocs or colloids had a strong tendency to deposit on membrane surface (Meng et al., 2007; Lin et al., 2011; Su et al., 2013). This phenomenon has not yet been exactly explained.

It has been suggested that the formation of cake layer on membrane surface in MBRs is a dynamic process (Meng et al., 2007; Lin et al., 2010). In general, the adhesion of foulants on membrane surface depends on both the hydrodynamic and thermodynamic properties of the foulants in the MBRs. There are two opposite forces that control the motion of foulants towards membrane surface: permeation drag, generated by permeate flux, and back transport, consisted of Brownian diffusion, inertial lift and shear induced diffusion (Belfort et al., 1994). While hydrodynamic drag forces bring the foulants close to the membrane surface, it is the thermodynamic forces (short-range physic–chemical interactions) that cause binding of the foulants to the membrane, and eventually form a cake layer. For this regarding, assessment of these short-range interactions between the foulants and membrane surface would be very conducive to better understanding of membrane fouling. However, previous studies regarding membrane fouling appear to mostly focus on foulants identification or characterization other than assessing these short-range interactions in MBRs.

Interfacial interactions in aquatic media are traditionally described through the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory, which accounts for van der Waals (LW), electrostatic double layer (EL) and Lewis acid–base (AB) interactions (van Oss et al., 1999). XDLVO theory has been used to depict static adhesion of model colloids (silica, aluminum oxide colloids, and polystyrene microspheres) (Brant and Childress, 2002), special

microbial strains (*Campylobacter jejuni*, *Klebsiella oxytoca*) (Feng et al., 2009; Nguyen et al., 2011) on microfiltration (MF), ultrafiltration (UF) or reverse osmosis (RO) membranes. Results have shown that these interactions between them could be well qualitatively and quantitatively predicted. Although aeration and complex bulk sludge properties make the cake layer formation in MBR system much more complicate, the adhesion process of foulants would be generally similar to those reported in above studies. Thus, XDLVO analysis may offer a new approach helping recognize and resolve membrane fouling in MBRs.

Bearing the information above, this study aims to thermodynamically assess membrane fouling in a MBR based on XDLVO approach. In this study, a lab-scale MBR was operated for over 200 days. The membrane fouling behavior as well as surface properties of bulk sludge, model foulants and membrane was investigated. The thermodynamic interactions between membrane and foulants were comprehensively analyzed. This study would give new insights into membrane fouling, and, more importantly, provide significant implications for membrane fouling control in MBRs.

2. Methods

2.1. Experimental setup and operation

A submerged MBR (SMBR) setup with 100 L total volume ($0.30 \times 0.40 \times 0.85 \text{ m}$ length \times width \times height) and 65 L effective volume ($0.30 \times 0.40 \times 0.54 \text{ m}$ length \times width \times height) (Fig. 1) was used to treat synthetic municipal wastewater. The MBR reactor was divided into a riser zone and a down-comer zone by a baffle frame. A flat sheet membrane module composed of an array of polyvinylidene fluoride (PVDF) membrane (effective filtration area was 0.5 m^2) with 140 kDa molecular weight cut-off (MWCO) was submerged in the riser zone. The membrane module was supplied by Shanghai SINAP Co. Ltd. PVDF appeared one of the most popular membrane materials used in MBR applications due to its excellent chemical resistance and thermal–mechanical properties (Hester et al., 1999). It has been reported that PVDF together with polyethersulfone (PES) membranes accounted for around 75% of the total products in the market including 9 out of the 11 most commercially important products (Santos and Judd, 2010). Usage of PVDF membrane was therefore a representative selection in

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