



# Influences of acid–base property of membrane on interfacial interactions related with membrane fouling in a membrane bioreactor based on thermodynamic assessment



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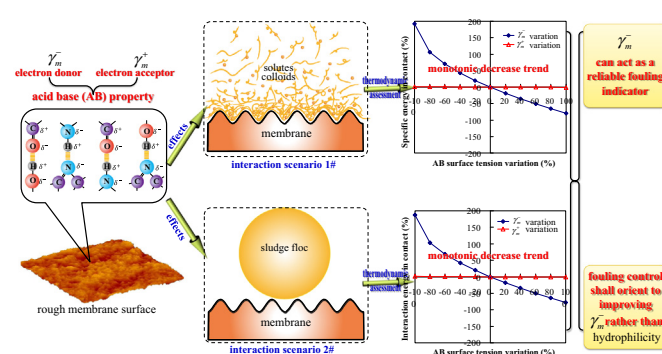
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## HIGHLIGHTS

- Total interaction energy monotonically decreases with membrane electron donor.
- Total interaction energy is marginally affected by membrane electron acceptor.
- Membrane electron donor is a reliable indicator predicting membrane fouling.
- Fouling control shall improve membrane electron donor rather than hydrophilicity.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Failure of membrane hydrophobicity in predicting membrane fouling requires a more reliable indicator. In this study, influences of membrane acid base (AB) property on interfacial interactions in two different interaction scenarios in a submerged membrane bioreactor (MBR) were studied according to thermodynamic approaches. It was found that both the polyvinylidene fluoride (PVDF) membrane and foulant samples in the MBR had relatively high electron donor ( $\gamma^-$ ) component and low electron acceptor ( $\gamma^+$ ) component. For both of interaction scenarios, AB interaction was the major component of the total interaction. The results showed that, the total interaction monotonically decreased with membrane  $\gamma^-$ , while was marginally affected by membrane  $\gamma^+$ , suggesting that  $\gamma^-$  could act as a reliable indicator for membrane fouling prediction. This study suggested that membrane modification for fouling mitigation should orient to improving membrane surface  $\gamma^-$  component rather than hydrophilicity.

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## 1. Introduction

While membrane bioreactor (MBR) technology has been regarded as one of most promising technologies for wastewater treatment (Meng et al., 2009; Wang et al., 2014), and has been

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## Nomenclature

$D$	closest distance between a sphere and a planar surface (nm)	$\lambda$	decay length of AB interaction in water (0.6 nm)
$h$	separation distance between two planar surfaces (nm)	$\phi$	contact angle ( $^{\circ}$ )
$e$	electron charge ( $1.6 \times 10^{-19}$ C)	$\theta$	angle of the circular arc in the circular ring
$\Delta G$	interaction energy per unit area ( $\text{mJ}\cdot\text{m}^{-2}$ )	$\xi$	zeta potential (mV)
$k$	Boltzmann's constant ( $1.38 \times 10^{-23}$ J $\cdot\text{K}^{-1}$ )	<i>Superscripts</i>	
$p$	roughness of membrane surface (nm)	AB	Lewis acid–base
$R$	radius of sludge floc ( $\mu\text{m}$ )	EL	electrostatic double layer
$r$	radius of differential circular ring on particle surface ( $\mu\text{m}$ )	LW	Lifshitz–van der Waals
$U$	interaction energy between membrane surface and particle (kT)	tol	total
$z(r, \theta)$	local amplitude directly below the circular arc as a function of the position of the differential circular arc defined by $r$ and $\theta$	+	electron acceptor
<i>Greek letters</i>		–	electron donor
$\epsilon_r \epsilon_0$	permittivity of the suspending liquid ( $\text{C}\cdot\text{V}^{-1}\cdot\text{m}^{-1}$ )	<i>Subscripts</i>	
$\gamma$	surface tension parameter ( $\text{mJ}\cdot\text{m}^{-2}$ )	$f$	foulant
$\kappa$	reciprocal Debye screening length ( $\text{nm}^{-1}$ )	$h_0$	minimum equilibrium cut-off distance (0.158 nm)
		$l$	liquid
		$m$	membrane
		$s$	solid
		$w$	water

extensively studied for more than 40 years, investigating factors, mechanisms and control strategies of membrane fouling is still the largest issue for this technology (Meng et al., 2009; Lin et al., 2014; Yan et al., 2016).

Foulant adhesion has been considered as the prominent cause of membrane fouling in MBRs (Chen et al., 2012; Wang et al., 2013; Lin et al., 2014). Adhesion of sludge foulants on membrane surface is determined by a complicated interplay of forces including the Lifshitz–van der Waals forces (LW), electrostatic double layer (EL) interactions, and acid–base (AB) interactions. The dependence of these interactions on separation distance is generally incorporated in the so-called extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) approach (van Oss, 1995). The AB interaction is based on electron donor ( $\gamma^-$ )/electron acceptor ( $\gamma^+$ ) interactions between polar moieties in aqueous medium (van Oss, 2003). It was reported that AB interaction between biopolymers and substratum surfaces occurring in water is quantitatively more dominant than either LW or EL interactions (van Oss, 2003; Nguyen et al., 2011), as they are up to 10–100 times higher than LW or EL interactions (van Oss, 2003). Therefore, to reduce the adhesive fouling in MBRs, it is essential to understand the properties and effects of AB interaction on membrane fouling.

Hitherto, XDLVO approach has been applied to interpret and predict membrane fouling phenomena in MBRs (Chen et al., 2012; Hong et al., 2013; Wang et al., 2013). In this approach, the AB property of a substance could be experimentally determined based on contact angle measurement (CAM) combined with the equations developed by Van Oss et al. (1988). Consequently, XDLVO approach may provide a useful tool to explore the exact roles of AB interaction in membrane fouling. However, XDLVO approach is only valid for the interactions between two smooth planar surfaces (van Oss, 1993), in contrast, the real membrane surface is significantly rough as reported in lots of previous studies (Hoek and Agarwal, 2006; Mahendran et al., 2011; Chen et al., 2012). Meanwhile, the prevailed foulants in MBRs can be generally classified into three categories: sludge flocs, colloids and solutes (mainly soluble microbial products (SMPs) (Meng et al., 2009; Wu et al., 2012)). These foulants have different chemical and morphological properties. The interactions between rough membrane surfaces and varies of foulants should differ from each other, and should be significantly different from those between two smooth

planar surfaces. This situation suggested the inapplicability of XDLVO approach for these interaction scenarios, and also called for a proper approach to assess effects of AB interaction. Recently, the authors of this study have developed a novel approach which allows modeling rough membrane surface and quantitatively assessing interaction energy between sludge flocs and rough membrane surfaces (Zhao et al., 2016). The new developed approach, combining the XDLVO approach may provide a methodological background for tracking the exact roles of AB property of membranes in membrane fouling in different interaction scenarios.

Moreover, current fouling control strategy based on membrane modification mainly oriented to increasing membrane surface hydrophilicity. In XDLVO theory, surface hydrophilicity is quantitatively defined by the free energy of interaction between two identical surfaces immersed in water ( $\Delta G_{\text{sws}}$ ) (van Oss, 1995). However, it has been frequently reported that surface hydrophilicity failed to well predict membrane fouling (Choo and Lee, 1996; Chen et al., 2012; Subhi et al., 2012; Zhang et al., 2015). The limitations of membrane hydrophobicity require a more reliable indicator for membrane fouling prediction and mitigation. Considering the strength of AB interaction, AB property of membrane may be able to serve for this requirement. However, to our knowledge, there is no specific study investigating this issue based on thermodynamic analysis.

This study, therefore, aimed to investigate influences of AB property of membrane on membrane fouling in a submerged MBR (SMBR). Surface properties of membrane and foulants sampled from the SMBR apparatus were characterized. The interfacial interactions in different interaction scenarios were calculated. Influences of membrane AB property on interaction energies and membrane fouling were assessed by sensitivity analyses. Finally, implications for membrane fouling control were discussed.

## 2. Material and methods

### 2.1. Experimental MBR setup

A lab-scale SMBR apparatus with 60 L effective volume was continuously run. The simulated municipal wastewater was pumped to the reactor as influent. The influent was composed with 300 mg COD/L glucose plus the mineral medium. A flat sheet

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