

## Regular Article

# Novel indicators for thermodynamic prediction of interfacial interactions related with adhesive fouling in a membrane bioreactor



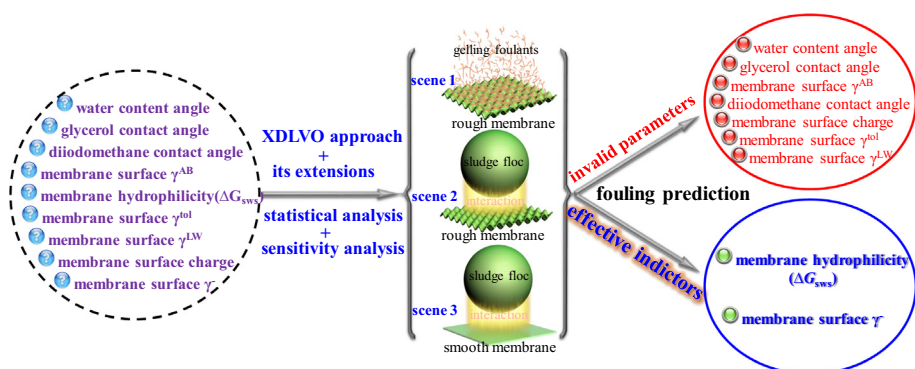
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## GRAPHICAL ABSTRACT



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## ABSTRACT

This study focused on developing indicators to predict adhesive membrane fouling in a membrane bioreactor (MBR). Thermodynamic interactions between membrane surface and foulants in various interaction scenes were comprehensively evaluated. It was revealed that, the total interaction energy in contact could be considered as a critical value affecting adhesion of foulants. Surface hydrophilicity cannot be simply represented by water contact angle. Statistical analysis showed that membrane acid-based (AB) surface tension, Lifshitz-Van der waals (LW) surface tension, total tension, zeta potential and water contact angle had no apparent correlation with adhesive fouling, suggesting the infeasibility of these parameters as fouling predictors. It was found that, interaction between two identical membrane surface in water ( $\Delta G_{sws}$ ) and membrane surface electron donor tension ( $\gamma^-$ ) strongly correlated with adhesive fouling, and could be reliable indicators to predict adhesive fouling. This study identified the relationships of series membrane surface properties with adhesive fouling in MBRs.

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## 1. Introduction

While membrane bioreactor (MBR) technology was deemed to a promising technology for wastewater treatment and reclaim, its wide-spread application has been significantly impeded by membrane fouling [1–3]. Therefore, great attention has been paid to

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**Nomenclature**

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

understand and alleviate membrane fouling in the last two decades.

It is generally accepted that, the predominant fouling form in MBRs is the adhesive fouling in terms of foulant adhesion [4–6], which is governed by the complex interplays between membrane and foulants in MBRs [7–9]. These interplays include hydrodynamic forces and thermodynamic interactions. It has been revealed that, hydrodynamic forces are responsible for bringing foulants closer to surface of membrane, while short-ranged thermodynamic interactions take roles of finally binding foulants to surface of membrane [10,11]. The thermodynamic interactions between two flat surfaces could be generally described by XDLVO approach which comprises three interaction components: electrostatic double layer (EL) interaction, acid-based (AB) interaction and Lifshitz-Van der waals (LW) interaction [7,8].

It is of essential interest to probe effective indicators among many membrane surface properties for fouling prediction and mitigation [12]. The thermodynamic interaction energies directly stem from surface properties of membrane for a given MBR. These surface properties provide candidates of indicator enable to predict and control membrane fouling in MBRs. Among various membrane surface properties, hydrophilicity/hydrophobicity of membrane has drawn special attention due to its important roles in membrane fouling. In numerous previous studies [13–15], membrane hydrophilicity was usually considered to be equal to water contact angle. Based on this consideration, although many studies reported the feasibility to reduce membrane fouling by increasing membrane hydrophilicity [15–17], inconsistent or even contradictory phenomena have been also observed. For instance, Choo and Lee [18] concluded that surface hydrophilicity itself was not enough to evaluate adhesive fouling. Brant and Childress [8] reported that membrane hydrophobicity estimated by water contact angle was not account precisely for the non-electrostatic interactions. The contradictory conclusions regarding effects of membrane hydrophilicity may be not surprising, as considering that the thermodynamic relationship between membrane hydrophilicity and water contact angle has not been revealed. Nevertheless, these studies demonstrated the infeasibility of membrane hydrophilicity quantified by water contact angle as an indicator to predict adhesive fouling, and also called for more reliable alternative indicators.

It is evident that the eventual adhesion of foulants to surface of membrane is governed by thermodynamic interaction energies between membrane surface and foulants [10,19]. Therefore, XDLVO approach and its extensions used to quantify these thermodynamic interactions provide methodological possibilities to explore novel fouling indicators. Among the three kind of thermodynamic interactions, AB interaction is generally much more predominant (account for over 90% of total interaction) than either EL or LW interaction [20]. AB interaction directly depends on AB properties (namely, electron donor ( $\gamma^-$ )/electron acceptor ( $\gamma^+$ ) component) of membrane. Recent studies have indicated that membrane surface electron donor ( $\gamma^-$ ) component critically affected the total interaction between membrane and foulants [21,22]. However, the thermodynamic relationship between AB properties of membrane and adhesive fouling has not been well explored. Therefore, it is quite desirable to screen various candidates, and then propose reliable indicators enable to predict adhesive fouling in MBRs.

The objective of this study is to explore the thermodynamic relationships between membrane surface properties and adhesive fouling. Accordingly, characterization of typical properties of the foulants in a MBR system and the virgin membrane was performed, and the thermodynamic interaction energies between foulants and membrane in different interaction scenes were assessed. Effects of AB properties and membrane hydrophilicity on the thermodynamic interactions were analyzed. Based on the thermodynamic analysis, novel indicators for adhesive fouling prediction in MBRs was proposed.

## 2. Materials and methods

### 2.1. Experimental apparatus and operation

A MBR apparatus at lab-scale, configured with a reactor and five flat-sheet membrane modules, was used in this investigation (Fig. 1). The reactor contained 65 L effective volume with dimensions of 0.54 m height  $\times$  0.30 m length  $\times$  0.40 m width. The membrane module had two-side flat-sheet membrane laminated by polyvinylidene fluoride (PVDF) (Fig. 1a). The normalized pore

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