



# Quantitative evaluation of the interfacial interactions between a randomly rough sludge floc and membrane surface in a membrane bioreactor based on fractal geometry



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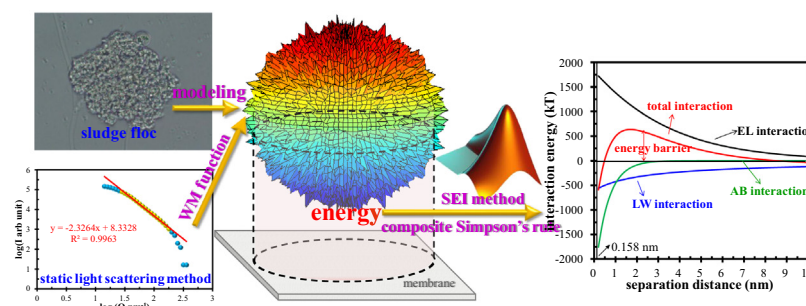
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## HIGHLIGHTS

- Randomly rough sludge surface can be well modeled by fractal geometry.
- A novel method to quantify interactions with randomly rough floc was developed.
- The proposed method was verified to be correct and feasible.
- The method had broad application prospect in membrane fouling research.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this paper, a new method for quantification of interfacial interactions between a randomly rough particle and membrane surface was proposed. It was found that sludge flocs in a membrane bioreactor were of apparent fractal characteristics, and could be modeled by the modified two-variable Weierstrass-Mandelbrot (WM) function. By combining the surface element integration (SEI) method, differential geometry and composite Simpson's rule, the quantitation method for calculating such interfacial interactions was further developed. The correctness and feasibility of the new method were verified. This method was then applied to evaluate the interfacial interactions between a randomly rough particle and membrane surface. It was found that, randomly rough particle possesses stronger interaction strength than regularly rough particle but weaker strength than smooth particle with membrane surface, indicating significant effects of surface morphology and roughness. The proposed method in this study has broad application prospect in membrane fouling study.

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## 1. Introduction

While membrane bioreactor (MBR) is regarded as a well-established technology for wastewater treatment and reuse,

membrane fouling remains a serious barrier affecting the economy and practicability of this technology (Lin et al., 2014b; Meng et al., 2009; Wang et al., 2012; Zhang et al., 2017). It is generally believed that adhesion of foulants on membrane surface (foulant layer formation) is the main form of membrane fouling (Chen et al., 2016; Hong et al., 2014; Shen et al., 2015; Su et al., 2014), which is directly controlled by the thermodynamic interactions between

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## Nomenclature

$A_H$	Hamaker constant, equal to $-12\pi h_0^2 \Delta G_{h_0}^{LW}$		
$D$	closest distance between a floc and a planar surface (nm)		
$dA$	differential projected area of differential element on membrane surface ( $m^2$ )		
$dS$	differential projected area of differential circular arc on floc surface ( $m^2$ )		
$d\theta$	differential angle along $\theta$ coordinates ( $^\circ$ )		
$d\varphi$	differential angle along $\varphi$ coordinates ( $^\circ$ )		
$h_-$	separation distance between two planar surfaces (nm)		
$\underline{d}R$	the position vector		
$\underline{i}$	unit vector along positive $x$ -direction		
$\underline{j}$	unit vector along positive $y$ -direction		
$\underline{k}$	unit vector along positive $z$ -direction		
$R$	radius of the modeled sludge floc (nm)		
$\underline{n}$	the unit outward normal to the surface		
$\Delta G$	interaction energy per unit area ( $mJ \cdot m^{-2}$ )		
$\Delta r$	the increment from the overlapping fractal surface		
$r$	radius of sludge floc (nm) without increment		
$U$	interaction energy between membrane surface and particle (kT)		
$I$	light intensity		
$Q$	scatter vector		
$D_f$	fractal dimension of a solid		
$L$	sample length (m)		
$G$	fractal roughness (m)		
$M$	number of superposed ridges		
$n_{max}$	highest frequency		
$L_s$	cutoff frequency (nm)		
$R_a$	average roughness of the semisphere		
$n$	frequency of the ripples		
		<i>Greek letters</i>	
		$\gamma$	surface tension parameter ( $mJ \cdot m^{-2}$ )
		$\epsilon_r \epsilon_0$	permittivity of the suspending liquid ( $C \cdot V^{-1} \cdot m^{-1}$ )
		$\theta$	angle coordinate in spherical coordinate system
		$\varphi$	angle coordinate in spherical coordinate system
		$\kappa$	reciprocal Debye screening length ( $nm^{-1}$ )
		$\xi$	zeta potential (mV)
		$\phi$	contact angle ( $^\circ$ )
		$\lambda$	decay length of AB interactions in water (0.6 nm)
		$\eta$	parameter of frequency density
		$\phi_{m,n}$	random phase
		$\vartheta$	scaled amplitude of ripples
		<i>Superscripts</i>	
		AB	Lewis acid-base
		EL	electrostatic double layer
		LW	Lifshitz-van der Waals
		tol	total
		+	electron acceptor
		–	electron donor
		<i>Subscripts</i>	
		$f$	foulant particle
		$h_0$	minimum equilibrium cut-off distance (0.158 nm)
		$l$	liquid
		$m$	membrane
		$s$	solid
		$w$	water

foulants and membrane surface (Chen et al., 2015; Su et al., 2014; Wang et al., 2013; Zhang et al., 2008). Therefore, quantitation of the interfacial interactions between foulants and membrane surface is of primary significance for better understanding and mitigation of membrane fouling.

The extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory (van Oss, 1993, 1994), which consists of Vander waals (LW), acid-base (AB) and electrostatic double layer (EL) interaction energies, is the most extensively accepted method for quantitative calculation of interfacial interactions between two entities in aqueous media. In the frame of XDLVO theory, Derjaguin approximation (DA) approach provides a solution to calculate the interfacial interactions between a particle and membrane surface. However, XDLVO theory as well as the DA approach was proposed based on the assumption that the two interaction surfaces are ideally smooth (Derjaguin, 1934; van Oss, 1993, 1994). In reality, at a small enough scale, the surfaces of sludge foulants (such as flocs and colloids) in MBR systems are significantly rough (Hua et al., 2015; Mei et al., 2016; Semblante et al., 2013; Zhou et al., 2016). Therefore, it is apparent that DA approach is not valid for quantitative calculation of the interfacial interactions between membrane and rough sludge particles in MBRs (Mei et al., 2016; Zhou et al., 2016). In order to solve this problem, the surface element integration (SEI) method, which integrates the interaction energy per unit area between two opposing differential planar elements over the whole surfaces, was further developed (Bhattacharjee and Elimelech, 1997). This approach can overcome the limitations of DA method, and therefore allows to quantify interfacial interactions between two curved surfaces (Bhattacharjee and Elimelech,

1997; Hong et al., 2016; Lin et al., 2014a; Siegismund et al., 2014). Nevertheless, the calculation results by this method are highly depending on the surface morphology of the investigated subjects. Consequently, modeling a sludge floc with a randomly rough surface similar to the reality is primarily important for the quantification of interfacial interactions between membrane and sludge flocs.

During the past several decades, a lot of efforts have been devoted into modeling natural rough surface. Pursuing literature shows that the conventional methods for surface modeling can be classified into two groups. One is randomly placing regular geometric asperities (such as hemispheres, cylinders and cones) on a smooth surface (Bhattacharjee et al., 1998b; Chen et al., 2012; Hoek and Agarwal, 2006; Martines et al., 2008), and the other is defining the surface roughness as a periodic function (like sinusoidal function) (Bhattacharjee et al., 1998b; Cai et al., 2017; Lenhoff, 1994; Zhao et al., 2016). Although these methods deepened our insights into rough surface modeling, the modeled morphologies were too regular, and could not reflect the real situation accurately. Actually, the surface of sludge flocs is non-stationary, and possesses typical fractal characteristics (Mei et al., 2016). Accordingly, Weierstrass-Mandelbrot (WM) function, a typical fractal function for three dimension isotropic surface construction (Chen et al., 2017; Liu et al., 2015; Peng and Guo, 2007; Yan and Komvopoulos, 1998), seems a suitable method for the model of sludge surface morphology, although WM function itself cannot be directly used to model spherical surface. Recently, by combining the modified two-variable WM function and coordinate transformation, Mei et al. (2016) have successfully modeled the rough

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