



Realization of quantifying interfacial interactions between a randomly rough membrane surface and a foulant particle



Jianrong Chen^a, Hongjun Lin^{a,*}, Liguo Shen^a, Yiming He^b, Meijia Zhang^{a,c}, Bao-Qiang Liao^c

^a College of Geography and Environmental Sciences, Zhejiang Normal University, Jinhua 321004, PR China

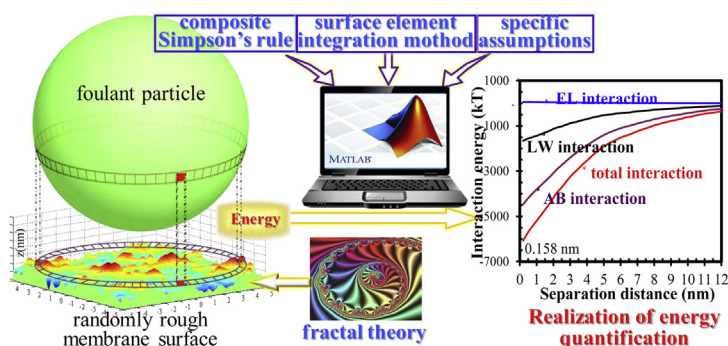
^b Institute of Physical Chemistry, Zhejiang Normal University, Jinhua 321004, PR China

^c Department of Chemical Engineering, Lakehead University, 955 Oliver Road, Thunder Bay, Ontario P7B 5E1, Canada

HIGHLIGHTS

- Randomly rough membrane surface can be well reconstructed by fractal geometry.
- A novel method for calculation of interactions with rough surface was developed.
- First realization of quantifying energy with randomly rough surface was provided.
- Membrane surface topography significantly affected interaction energy.

GRAPHICAL ABSTRACT



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ABSTRACT

Quantification of interfacial interaction with randomly rough surface is the prerequisite to quantitatively understand and control the interface behaviors such as adhesion, flocculation and membrane fouling. In this study, it was found that membrane surface was randomly rough with obvious fractal characteristics. The randomly rough surface of membrane could be well reconstructed by the fractal geometry represented by a modified Weierstrass-Mandelbrot function. A novel method, which combined composite Simpson's approach, surface element integration method and approximation by computer programming, was developed. By using this method, this study provided the first realization of quantifying interfacial energy between randomly rough surface of membrane and a foulant particle. The calculated interactions with randomly rough surface of membrane were significantly different from those with smooth surface of membrane, indicating the significant effect of surface topography on interactions. This proposed method could be also potentially used to investigate various natural interface environmental phenomena.

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1. Introduction

With more and more successful applications in the past decade (Lin et al., 2012; Huang and Lee, 2015), membrane bioreactor (MBR) has been considered as an established technology for

wastewater treatment and energy/resource recovery (Lin et al., 2013; Zhou et al., 2015). However, membrane fouling is still the major concern of this technology (Ng et al., 2006; Lin et al., 2014a; Wang et al., 2014). Quantitative identification of interaction energy between two entities is critically important to understand and control interfacial behaviors regarding deposition, coagulation and transport of particles (Chen et al., 2012; Shen et al., 2012; Lin et al., 2014a; Siegismund et al., 2014). In a MBR, interaction energy

* Corresponding author.

E-mail address: hjlin@zjnu.cn (H. Lin).

Nomenclature

A_H	Hamaker constant, equal to $-12\pi h_0^2 \Delta G_{h_0}^{LW}$
D_f	fractal dimension of a solid
dA	differential projected area of differential element on membrane surface (m^2)
dr	differential ring radius (m)
$d\theta$	differential angle of the differential circular arc ($^\circ$)
$f(r, \theta)$	local amplitude directly below the circular arc as a function of the position of the differential circular arc defined by r and $d\theta$
E	electron charge (1.6×10^{-19} C)
h	separation distance between two planar surfaces (nm)
k	Boltzmann's constant (1.38×10^{-23} J·K $^{-1}$)
L	sample length (m)
L_s	cutoff frequency (m)
M	number of superposed ridges
N	frequency number
G	fractal roughness (m)
ΔG	interaction energy per unit area ($mJ \cdot m^{-2}$)
R	radius of foulant particle (μm)
R	radius of differential circular ring on particle surface (μm)
S	closest distance between a particle and a planar surface (nm)
U	interaction energy between membrane surface and particle (kT)
Z	roughness of membrane surface (nm)

Greek letters

γ	surface tension parameter ($mJ \cdot m^{-2}$)
$\epsilon_r \epsilon_0$	permittivity of the suspending liquid ($C \cdot V^{-1} \cdot m^{-1}$)
η	parameter of frequency density
θ	angle of the circular arc in the circular ring
κ	reciprocal Debye screening length (nm^{-1})
λ	decay length of AB interactions in water (0.6 nm)
ξ	zeta potential (mV)
$\phi_{m,n}$	random phase
φ	contact angle ($^\circ$)

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
max	maximum value
s	solid
w	water

directly determines the adhesion of foulant particles and foulant layer formation (Ng et al., 2006; Tian et al., 2013; Wang et al., 2013; Shen et al., 2015), which is believed as the major cause of fouling (Hong et al., 2014; Zhou et al., 2015; Zhang et al., 2016a). It is traditional to depict this energy between two surfaces in aqueous phase by the theory of extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) (van Oss, 1993; Hoek and Agarwal, 2006; Lin et al., 2014b). While XDLVO theory is only applicable for ideally smooth planar surfaces, real surfaces of both foulant particles and membrane are significantly rough (Brant and Childress, 2004; Hoek and Agarwal, 2006; Chen et al., 2012). This problem significantly contributes to the discrepancies between experimental observations and theoretical predictions of interfacial behaviors regarding deposition, coagulation and transport of particles in the literature (Bhattacharjee et al., 1998; Brant and Childress, 2004).

Considerable efforts have been devoted into overcoming this problem. As a result, surface element integration (SEI) method was proposed (Bhattacharjee and Elimelech, 1997). This method has its unique niche in interaction energy quantification with rough surface because it integrates the differential interaction per unit area over whole surface, and thus enables to calculate the whole energy between two curved surfaces in theory (Bhattacharjee and Elimelech, 1997; Bhattacharjee et al., 1998). The major disadvantage of this method is the double integrals involved. Due to the tremendous calculation of the double integrals, utilization of this method was confined to regularly rough surfaces (Bhattacharjee et al., 1998; Hoek and Agarwal, 2006; Martinez et al., 2008; Chen et al., 2012).

Membrane surfaces have been conventionally modeled by randomly placing geometrically regular shapes such as hemispheres, cylinders and cones as asperities on a plate surface, or using periodic sinusoidal or Gaussian functions to describe the surface topography (Bhattacharjee et al., 1998; Chen et al., 2012; Hong et al., 2016). However, the surfaces modeled by these methods

seemed too regular, and were totally unlike the real surface topographies even judged by the intuition. Recent studies have shown that, natural surfaces including membrane surfaces are irregular and disorder, showing fractal property (Hong et al., 2016; Zhang et al., 2016b). One of the notable fractal properties is self-similarity, a property meaning that an object comprises sub-units statistically similar to the whole object structure (Majumdar and Bhushan, 1991; Meng et al., 2005). As for membrane surfaces, the fractal property is demonstrated in forms of the appearance of smaller scales of roughness when a surface section is magnified (Zhang et al., 2016b). Obviously, conventional techniques or Euclidean geometry cannot properly model fractal surfaces (Meng et al., 2005; Zhang et al., 2016b), which calls for an efficient approach for real membrane surface modeling. This demand has been satisfied by the new method proposed by our recent study (Hong et al., 2016). The new method enables to model randomly rough membrane surface by the modified two-variable Weierstrass-Mandelbrot (WM) function included in fractal theory, thus giving significant insights into rough surface topography (Hong et al., 2016).

The progress in rough membrane surface modeling raises the possibility to quantify the interaction energy with randomly rough membrane surface, though currently it is still a big challenge. This is not surprising, as the double integrals involved in SEI method causes tremendous computing workload, especially for randomly rough membrane surface (Hong et al., 2016). Meanwhile, obtaining the antiderivative of integrals in SEI method is basically impossible for rough surface (Zhao et al., 2016). Moreover, the unexpected random tiny protrusions on the real membrane surfaces may intersect with foulant particle surface when a particle gets close to the membrane surface, which would also prevent quantification of interaction energy. Therefore, effective methods have yet to be developed to solve these problems. If these problems were solved, it is expected that quantification of interaction energy between

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