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# Membrane fouling in a submerged membrane bioreactor: New method and its applications in interfacial interaction quantification

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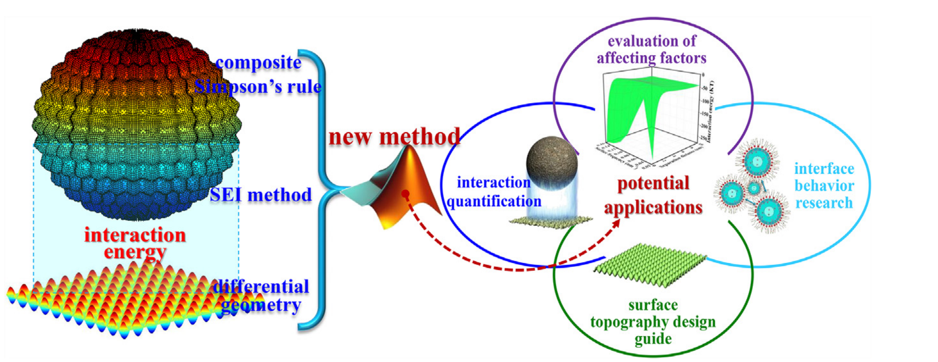
## HIGHLIGHTS

- Regularly rough membrane and rough particle surfaces were constructed.
- A new method to quantify interactions between two rough surfaces was proposed.
- Effects of rough surface topography on interfacial interaction were investigated.
- The new method had broad application fields including guiding surface design.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Quantification of interfacial interactions between two rough surfaces represents one of the most pressing requirements for membrane fouling prediction and control in membrane bioreactors (MBRs). This study firstly constructed regularly rough membrane and particle surfaces by using rigorous mathematical equations. Thereafter, a new method involving surface element integration (SEI) method, differential geometry and composite Simpson's rule was proposed to quantify the interfacial interactions between the two constructed rough surfaces. This new method were then applied to investigate interfacial interactions in a MBR with the data of surface properties of membrane and foulants experimentally measured. The feasibility of the new method was verified. It was found that asperity amplitude and period of the membrane surface exerted profound effects on the total interaction. The new method had broad potential application fields especially including guiding membrane surface design for membrane fouling mitigation.



## 1. Introduction

In the last past decades, membrane bioreactor (MBR) technology has been significantly developed since it features enhanced effluent quality, high microorganism concentration, reduced reactor footprint, and complete solid rejection over conventional activated sludge process for wastewater treatment (Lin et al., 2012; Meng et al., 2009; Yan et al., 2016). Membrane fouling, however, is still a major problem needed to be resolved (Chen et al., 2016; Guo et al., 2012; Wang et al., 2016; Zhang et al., 2015, 2017a). It is generally accepted that, in MBRs, membrane fouling mainly stems from adhesion or accumulation of sludge foulants on membrane surface (Deng et al., 2016; Lin et al., 2009; Wang and Li, 2008). Numerous experimental operations and theoretical studies have identified the decisive role of the short-ranged interfacial interactions between sludge foulants and membrane surface in foulant adhesion process (Brant and Childress, 2004; Chen et al., 2012; Lin et al., 2014b; Tang et al., 2011) because these

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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 particle and a planar surface (nm) |
| $dA$ | differential projected area of differential element on membrane surface ($m^2$) |
| $dr$ | differential ring radius (m) |
| $dS$ | differential projected area of differential circular arc on particle surface ($m^2$) |
| $d\theta$ | differential angle along $\theta$ coordinates (°) |
| $d\varphi$ | differential angle along $\varphi$ coordinates (°) |
| $h$ | separation distance between two planar surfaces (nm) |
| $\vec{i}$ | unit vector along positive $x$-direction |
| $n$ | frequency of the ripples |
| $\vec{n}$ | the unit outward normal to the surface |
| $\Delta G$ | interaction energy per unit area ($mJ \cdot m^{-2}$) |
| $p$ | scaled amplitude of rough surface asperities (nm) |
| $r$ | radius of sludge particle (nm) |
| $U$ | interaction energy between membrane surface and particle (kT) |
| $w$ | scaled frequency of rough surface asperities (nm) |
| $z$ | $z$-direction |

*Greek letters*

| | |
|---|---|
| $\alpha$ | particle radius when $\lambda$ is zero (nm) |
| $\gamma$ | surface tension parameter ($mJ \cdot m^{-2}$) |
| $\varepsilon_r\varepsilon_0$ | permittivity of the suspending liquid ($C \cdot V^{-1} \cdot m^{-1}$) |
| $\theta$ | angle coordinate in spherical coordinate system |
| $\kappa$ | reciprocal Debye screening length ($nm^{-1}$) |
| $\lambda$ | scaled amplitude of ripples |
| $\xi$ | zeta potential (mV) |
| $\phi$ | contact angle (°) |
| $\varphi$ | angle coordinate in spherical coordinate system |
| $\tau$ | decay length of AB interactions in water (0.6 nm) |

*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 |
| $s$ | solid |
| $w$ | water |
| $x$ | $x$-direction |
| $y$ | $y$-direction |

interactions are several orders of magnitude higher than other forces in short range distance within several nanometers (Brant and Childress, 2002; Van Oss, 1993; Zhao et al., 2016a). In this context, quantification of these interfacial interactions is the most pressing need for overcoming membrane fouling problem.

So far, the extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory provides the primary method to quantify the interfacial interactions between two smooth planar surfaces in water (Bhattacharjee and Elimelech, 1997; Brant and Childress, 2004; Van Oss, 1993). Despite the wide applications of this theory for interface behavior research, strictly speaking, it cannot be directly used to quantify the interactions in MBRs because real surfaces of both membrane and foulants in MBRs are far from smooth (Chen et al., 2012; Hoek and Agarwal, 2006; Zhao et al., 2016b). Therefore, there exists a great demand to develop new methods for quantification of interfacial interactions between two rough surfaces. Furthermore, recent development of three-dimensional (3D) printing and 3D engraving techniques have added this demand (Deger et al., 2016; Hashemi Sanatgar et al., 2017; Lee et al., 2008; Wang et al., 2017). These techniques could confer a solid surface with desirable regular topography in nanometer scale (Deger et al., 2016; Hashemi Sanatgar et al., 2017; Lee et al., 2008; Wang et al., 2017). If the relationships between membrane surface topography and interfacial interactions can be quantified by a new method, membrane fouling caused by adhesion process can be quantitatively predicted and controlled by designing membrane surface topography through these techniques, bringing significant interest to membrane fouling and interface behavior research.

The demand to develop a new quantitative method firstly requires properly constructing rough surfaces of two interacting entities (Bhattacharjee and Elimelech, 1997; Brant and Childress, 2004; Hoek and Agarwal, 2006; Lin et al., 2014a; Martines et al., 2008). It was reported that the natural surfaces of membrane and foulants were randomly rough (Brant and Childress, 2002; Chen et al., 2012; Mei et al., 2016). Basically, it is impractical to design randomly rough surface for membrane fouling control. However, it is practical to design a regular rough surface. Therefore, constructing regular rough surface has its inherent interests for interfacial interaction quantification. In the literature regarding interaction quantification, rough surfaces were generally constructed by placing a series of regular geometries (e.g. hemispheres, cylinders and cones) on an ideally smooth surface as protrusions or depressions (Bhattacharjee et al., 1998; Chen et al., 2012; Martines et al., 2008; Suresh and Walz, 1996). The surfaces constructed in these studies, however, cannot well represent the pertinent statistical characteristics of rough surface, nor be described by a rigorous mathematical equation, which are of fundamentals for interfacial interaction quantification. It is thus desirable to develop a new way to construct regularly rough surface in order to overcome the drawbacks of the previous method.

Above-mentioned demand also need develop a special method to quantify the interfacial interactions between two rough surfaces (Brant and Childress, 2004; Hoek and Agarwal, 2006). Since XDLVO theory enables to calculate the interfacial interactions between two smooth planar surfaces, the total interfacial interaction between two regularly rough surfaces should be quantified if it is regarded as sum of innumerable interfacial interactions between two opposing differential planar surface elements. This idea provide theoretical solution to quantify the interfacial interactions between two regularly rough surfaces, and to guide construction of membrane surface topography. Nevertheless, to our knowledge, none of study has tested this idea for quantification of the interfacial interactions between two regularly rough surfaces. It is expected that such a quantification would give significant implications for membrane fouling control and interface behavior research.

In this study, the rough surfaces of membrane and sludge foulant samples in a MBR were firstly constructed by the rigorous mathematical equations. Afterwards, the spatial relationships between the constructed surfaces were probed. These relationships