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A novel approach for quantitative evaluation of the physicochemical interactions between rough membrane surface and sludge foulants in a submerged membrane bioreactor

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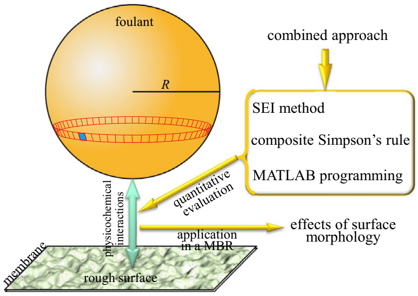
HIGHLIGHTS

- A new method to quantitatively assess physicochemical interactions was proposed.
- Membrane surface morphology highly affected the interaction energies with foulant.
- The method could serve as a primary tool for membrane fouling study in MBRs.

GRAPHICAL ABSTRACT



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ABSTRACT

This study proposed a novel approach for quantitative evaluation of the physicochemical interactions between a particle and rough surface. The approach adopts the composite Simpson's rule to numerically calculate the double integrals in the surface element integration of these physicochemical interactions. The calculation could be achieved by a MATLAB program based on this approach. This approach was then applied to assess the physicochemical interactions between rough membrane surface and sludge foulants in a submerged membrane bioreactor (MBR). The results showed that, as compared with smooth membrane surface, rough membrane surface had a much lower strength of interactions with sludge foulants. Meanwhile, membrane surface morphology significantly affected the strength and properties of the interactions. This study showed that the newly developed approach was feasible, and could serve as a primary tool for investigating membrane fouling in MBRs.

1. Introduction

With 4 decades development, membrane bioreactor (MBR) has been actively employed in various wastewater treatment and reuse due to its outstanding advantages over conventional activated sludge (CAS) system (Robles et al., 2012; Wu and He, 2012; Abdollahzadeh Sharghi et al., 2014). Nevertheless, membrane fouling, which will rise operating cost and reduce membrane life span, still remains one of the most serious challenges for application of MBR technology (Khan et al., 2009; Lin et al., 2011). Therefore, there have been longstanding interests to study the factors, mechanisms and control strategies of membrane fouling in MBRs.

Membrane fouling is directly related with adhesion of sludge matters on membrane surface in MBRs (Yeo et al., 2007; Lin et al., 2014). In disturbed systems like MBRs, hydrodynamic forces resulted from aeration could forward sludge matters nearby

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membrane surface. However, the eventual adhesion of these matters to form a foulant layer on membrane surface is determined by the physicochemical interactions between sludge matters and membrane (van Oss, 1994; Hong et al., 2013; Wang et al., 2013). It is therefore of primary importance to quantitatively assess these physicochemical interactions for a given MBR.

Generally, physicochemical interactions between two solid surfaces in aqueous medium could be characterized within the framework of the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory, which accounts for Vander Waals (LW), electrostatic double layer (EL), and acid–base (AB) interaction energies (van Oss, 1993). The XDLVO approach allows to quantitatively calculate these three types of interaction energies between two infinite parallel flat plates (van Oss, 1994). However, sludge foulant is generally assumed to be a global particle. In order to calculate the physicochemical interactions between a sphere and an infinite flat plate, Derjaguin approximation (DA) method, which treats sphere surface as a series of concentric rings, was applied (Derjaguin, 1934). While emerging as a convenient calculation approach, this method has several limitations. For example, DA is simply an approximation and is not valid in a strictly mathematical sense (Dantchev and Valchev, 2012). Moreover, this method does not include the effect of geometry of interacting surfaces (Bhattacharjee and Elimelech, 1997; Dantchev and Valchev, 2012). Actually, the surface of membranes used in MBRs was very rough as illustrated by atomic force microcopy (AFM) observation (Hoek et al., 2003; Brant and Childress, 2004; Mahendran et al., 2011; Chen et al., 2012). The limitations of DA method, together with the rough property of membrane surface, give significant impetus to the development of surface element integration (SEI) method (Bhattacharjee and Elimelech, 1997). The SEI method, which integrates the interaction energy per unit area between opposing differential planar elements over the entire surfaces, can circumvent the limitations of DA method (Bhattacharjee and Elimelech, 1997; Dantchev and Valchev, 2012), and thus is expected to enable to quantitatively assess the physicochemical interactions between a sludge foulant and a rough membrane surface. Unfortunately, due to the complicated morphology of membrane surface, it is basically impossible in practice to obtain the antiderivative of integrals in SEI method (Hoek et al., 2003; Hoek and Agarwal, 2006; Chen et al., 2012). This problem highly limits its real application. Particularly, to the best of our knowledge, there has been no specific study investigating the application of SEI method in MBRs. Therefore, it is quite desirable to develop an effective approach to quantitatively assess the physicochemical interactions between a sludge foulant and a rough membrane surface.

The primary goal of this study is to develop a combined method which enables to quantitatively compute the physicochemical interactions between sludge foulants and rough membrane surface. In this study, a novel method which combined SEI method with composite Simpson's rule was firstly established. In order to improve the computational efficiency and accuracy, MATLAB software was applied to complete the mass data computation. Thereafter, this method was applied to calculate the interactions between sludge foulants and membranes with different surface morphologies in a lab-scale MBR. Effects of roughness on the physicochemical interactions were also briefly discussed.

2. Methods

2.1. Analytical methods

Sludge samples were taken from sludge suspension in a stably-running lab-scale submerged MBR treating synthetic municipal wastewater. The details regarding the MBR system and the wastewater composition have been reported in a previous study (Zhang et al., 2014). The membrane (0.3 μm normalized pore size) used in the MBR was made of PVDF material, and provided by Shanghai SINAP Co., Ltd.

Zeta potentials of sludge foulants and membrane were measured by a Zetasizer Nano ZS (Malvern Instruments Ltd., UK) and A Zeta 90 Plus Zeta Potential Analyzer (Brookhaven Instruments, UK), respectively. Three measurements were performed for each sample. Samples including virgin PVDF membrane and sludge foulants were first treated according to the methods described in Wang et al. (2014). Thereafter, a contact angle meter (Kino industry Co., Ltd., USA) based on the sessile drop method was adopted to determine static contact angles of three probe liquids (ultra-pure water, glycerol and diiodomethane) on these prepared samples. The morphology of the virgin membrane surface was observed by AFM (NT-MDT). Particle size distribution (PSD) of sludge suspension samples was measured through a Malvern Mastersizer 2000 instrument. Calculation of double integrals based on composite Simpson's rule was executed using MATLAB v5.3.

2.2. XDLVO approach

Thermodynamic interactions including LW, EL and AB interaction between sludge foulant and membrane in water can be described by XDLVO theory (van Oss, 1994). The individual XDLVO interaction energy per unit area ($\Delta G^{LW}(h)$, $\Delta G^{AB}(h)$ and $\Delta G^{EL}(h)$) between two infinite planar surfaces is given by:

$$\Delta G^{LW}(h) = -\frac{A_H}{12\pi h^2} \tag{1}$$

$$\Delta G^{AB}(h) = \Delta G^{AB}_{D_0} \exp\left(\frac{h_0 - h}{\lambda}\right) \tag{2}$$

$$\Delta G^{EL}(h) = \varepsilon_r \varepsilon_0 \kappa \zeta_f \zeta_m \left(\frac{\zeta_f^2 + \zeta_m^2}{2\zeta_f \zeta_m}(1 - \coth \kappa h) + \frac{1}{\sinh \kappa h}\right) \tag{3}$$

where h is the separation distance between two planar surfaces; A_H $(= -12\pi h_0^2 \Delta G^{LW}_{h_0})$ is Hamaker constant; Contact of two planar surfaces is assumed to occur at a hypothetical minimum equilibrium cut-off distance (minimum separation distance (h_0), assigned to be 0.158 nm) (Meinders et al., 1995); λ (=0.6 nm) is the decay length of AB energy in water; $\varepsilon_r\varepsilon_0$ is the permittivity of sludge suspension; ζ_f and ζ_m are the surface zeta potential of foulant and membrane, respectively; κ is the reciprocal Debye length; $\Delta G^{LW}_{h_0}$, $\Delta G^{AB}_{h_0}$ and $\Delta G^{EL}_{h_0}$ are the LW, AB and EL interaction energy per unit area between two infinite planar surfaces at the minimum separation distance, respectively, which are given by Eqs. 4–6, respectively:

$$\Delta G^{LW}_{h_0} = -2\left(\sqrt{\gamma_m^{LW}} - \sqrt{\gamma_w^{LW}}\right)\left(\sqrt{\gamma_f^{LW}} - \sqrt{\gamma_w^{LW}}\right) \tag{4}$$

$$\begin{aligned}\Delta G^{AB}_{h_0} = 2\Big[&\sqrt{\gamma_w^+}\left(\sqrt{\gamma_f^-} + \sqrt{\gamma_m^-} - \sqrt{\gamma_w^-}\right) + \sqrt{\gamma_w^-}\left(\sqrt{\gamma_f^+} + \sqrt{\gamma_m^+} - \sqrt{\gamma_w^+}\right)\\ &-\sqrt{\gamma_f^- \gamma_m^+} - \sqrt{\gamma_f^+ \gamma_m^-}\Big]\end{aligned} \tag{5}$$

$$\Delta G^{EL}_{h_0} = \frac{\varepsilon_0 \varepsilon_r \kappa}{2}(\xi_f^2 + \xi_m^2)\left[1 - \coth(\kappa h_0) + \frac{2\xi_f \xi_m}{\xi_f^2 + \xi_m^2} csch(\kappa h_0)\right] \tag{6}$$

where γ^{LW}, γ^+ and γ^- are the surface tension parameters of foulant (subscript f), water (subscript w) and membrane (subscript m). The surface tension parameters of a solid material can be calculated by solving a set of three Young's equations (van Oss, 1994), provided that the contact angle (θ) data of three probe liquids (their surface tension parameters are known) on the solid material are measured.