



# Fractal reconstruction of rough membrane surface related with membrane fouling in a membrane bioreactor



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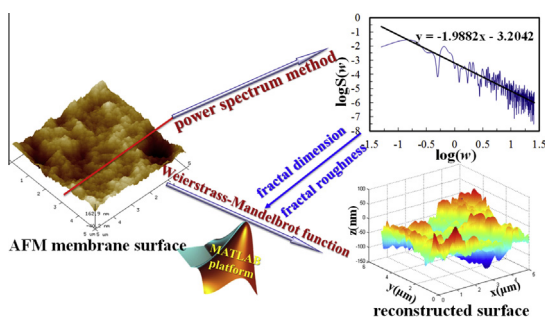
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## HIGHLIGHTS

- Power spectrum method was used to obtain fractal parameters of membrane.
- Membrane surface was successfully reconstructed by fractal geometry method.
- Good match of morphology between the reconstructed and real surface was obtained.
- The reconstructed membrane surface can be used for interaction energy evaluation.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 19 April 2016

Received in revised form 5 June 2016

Accepted 9 June 2016

Available online 11 June 2016

### Keywords:

Fractal geometry  
Surface reconstruction  
Rough surface  
Membrane fouling

## ABSTRACT

In this paper, fractal reconstruction of rough membrane surface with a modified Weierstrass-Mandelbrot (WM) function was conducted. The topography of rough membrane surface was measured by an atomic force microscopy (AFM), and the results showed that the membrane surface was isotropous. Accordingly, the fractal dimension and roughness of membrane surface were calculated by the power spectrum method. The rough membrane surface was reconstructed on the MATLAB platform with the parameter values acquired from raw AFM data. The reconstructed membrane was much similar to the real membrane morphology measured by AFM. The parameters (including average roughness and root mean square (RMS) roughness) associated with membrane morphology for the model and real membrane were calculated, and a good match of roughness parameters between the reconstructed surface and real membrane was found, indicating the feasibility of the new developed method. The reconstructed membrane surface can be potentially used for interaction energy evaluation.

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## 1. Introduction

Membrane bioreactor (MBR) technology possesses several distinct advantages (including higher effluent quality, reduced footprint and easier manipulation) over the conventional activated sludge process (CAS), and has been widely used for wastewater treatment and reuse (Le-Clech et al., 2006; Meng et al., 2009). However, membrane fouling, which would decrease treatment

performance and increase operational cost, has been regarded as the key barrier for the universal application of MBR (Le-Clech et al., 2006; Lin et al., 2014; Meng et al., 2009). Thus, during the past decade, considerable efforts have been devoted into exploring mechanisms and control of membrane fouling (Lin et al., 2014; Meng et al., 2009). The foulants in MBRs can be generally classified into sludge flocs and gelling foulants (Lin et al., 2014; Meng et al., 2009). It is well accepted that adhesion of foulants on membrane surface is the main cause of membrane fouling in MBRs (Lin et al., 2014; Wang and Li, 2008), and is generally considered as a result of complex physicochemical interactions between

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sludge foulants and membrane surface (Hong et al., 2013; Wang et al., 2013). Therefore, it is of primary importance to obtain an accurate knowledge regarding these physicochemical interactions.

The extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory, which contains Lifshitz-van der Waals (LW), electrostatic double layer (EL), and acid-base (AB) interaction energies, has been generally used to depict physicochemical interactions between two surfaces (van Oss, 1993). However, XDLVO approach is only valid for the physicochemical interactions between two smooth flat surfaces, while is inapplicable for the real interaction scenarios involving morphological heterogeneity of membrane and foulant surfaces (van Oss, 1993). In order to overcome this limitation, the surface element integration (SEI) method, which integrates the interaction energy per unit area between opposing differential planar elements over the entire surfaces, was further developed (Bhattacharjee and Elimelech, 1997; Dantchev and Valchev, 2012). SEI method allows quantitative assessment of the physicochemical interaction energies between two curved surfaces, provided that a rigid mathematical model of surface morphology is involved. Considering that membrane surface morphology is a vital factor affecting membrane fouling (Chen et al., 2012; Fu et al., 2008; Zhao et al., 2015), reconstruction of membrane surface morphology is a primary interest in membrane fouling study. In the previous studies, natural surface morphology was depicted as a stationary random process (Morag and Etsion, 2007; Nayak, 1973). Accordingly, Bhattacharjee et al. (1998) and Hoek et al. (2003) constructed the rough membrane surface by setting a series of hemispherical protrusions and depressions on a smooth planar surface. Similarly, cylinders and cones were adopted by Martines et al. (2008) and Suresh and Walz (1996) to simulate the protruding and hollow asperities, respectively. Recently, Zhao et al. (2015) proposed a more ideal approach, which describes surface morphology as sine functions of  $x$ - and  $y$ -directions in a plane coordinate system. Unfortunately, all of these efforts failed to reconstruct a surface highly close to the real membrane surface morphology. In fact, membrane surface morphology is a non-stationary process (Sayles and Thomas, 1978). A general phenomenon related with such a process is that, when a section of the rough surface is magnified, smaller scales of roughness appear. Previous conventional approaches apparently cannot reflect the non-stationary property of membrane surface. Therefore, it is quite desirable to develop an effective approach which could reconstruct a surface much close to the real surface.

Fractal geometry, which originally meant irregular and broken objects, was first conceived and developed by Mandelbrot (1967), and has been extensively applied in various fields of science and technology to denote the complicated pictures and processes (Perfect, 1997; Perfect and Kay, 1995). Self-affinity is an important characteristic of fractal surface, and means the profile of surface remaining similar to the original form when scale changes (Mandelbrot, 1967). It has been found that most material surfaces in nature exhibit disorder properties on the micro-scale and satisfy the self-affinity (Martan et al., 2005; Morag and Etsion, 2007). Many studies have verified that the non-stationary and self-affinity properties of rough surface could be represented by fractal geometry (Majumdar and Bhushan, 1991; Peng and Guo, 2007). Therefore, it is envisaged that fractal geometry could serve as an effective approach for reconstruction of membrane surface morphology. Nevertheless, no study has explored this issue up to date.

This study focused on membrane surface reconstruction. The membrane surface was firstly measured by atomic force microscopy (AFM), and its fractal characteristics were analyzed. Thereafter, Weierstrass-Mandelbrot (WM) function involving the fractal geometry was adopted to reconstruct the membrane surface morphology. The parameters (including  $R_a$  and  $R_q$ ) associated with the modeled and real membrane morphologies were calcu-

lated and compared. Finally, the reconstructed membrane surface was applied in calculation of physicochemical interactions between sludge foulants and membrane. This study will throw significant light on reconstruction of membrane surface topography.

## 2. Methods

### 2.1. Analytical methods

A submerged MBR system with 65 L effective volume was operated for the treatment of simulated municipal wastewater. The membrane unit made by polyvinylidene fluoride (PVDF) had effective area of 1.0 m<sup>2</sup> and a normalized pore size of 0.1 μm. Peristaltic pump was utilized to achieve the effluent through an intermittent suction mode of 4-min-on and 1-min-off. Membrane flux was maintained at approximately 30 L m<sup>-2</sup> h<sup>-1</sup>, corresponding to a hydraulic retention time (HRT) of 5.5 h.

Membrane surface morphology was characterized by an atomic force microscopy (AFM) (NT-MDT). The membrane samples were pretreated with the following procedures: virgin membrane pieces with an area of 2 cm × 2 cm were firstly immersed in deionized water for 12 h to remove impurities on the membrane, and then transferred to a desiccator for 24 h to remove excess water. The Nanoscope software was adopted for the roughness and fractal dimension analysis of membrane acquisition (5 μm × 5 μm areas). The calculated fractal dimension was employed for the reconstruction of the membrane surface. Roughness parameters, including average roughness ( $R_a$ ) and root-mean-square roughness ( $R_q$ ) were used to rate the feasibility of the reconstruction method. Each membrane sample was conducted in triplicate measurements.

### 2.2. Fractal dimension calculation

Fractal dimension is considered as a helpful parameter for the complexity characterization of membrane surface. There are various methods, such as the box counting, the variation and the power spectrum method (Thielen et al., 2016), were developed to calculate profile fractal dimension. In this article, the power spectrum method incorporating with the AFM image data was adopted to compute the fractal dimension.

In fractal geography, a rough surface can be described by the WM function as follows (Gagnepain and Roques-Carnes, 1986):

$$z(x) = L \left( \frac{G}{L} \right)^{D-1} \sum_{n=0}^{\infty} \frac{\cos(2\pi\eta^n \frac{x}{L})}{\eta^{(2-D)n}} \quad (1)$$

where  $D$  is the fractal dimension of two dimensional profile ( $1 < D < 2$ ),  $\eta$  is the parameter that determines the frequency spectrum of the profile,  $G$  is the height scaling parameter independent of frequency (fractal roughness), and  $L$  is the sample length.

The multi-scale WM function contains a series of roughness spectrum, which can be characterized by spectral density function as below:

$$S(\omega) = \frac{G^{2(D-1)}}{2 \ln \eta} \frac{1}{\omega^{(5-2D)}} \quad (2)$$

where  $S(\omega)$  is the power of the spectrum, and  $\omega$  is the frequency of the surface roughness.

In a double-log coordinate, if the relationship between  $S(\omega)$  and  $\omega$  is approximate to a straight line, and the level of slope is between  $-3$  and  $-1$ , then the profile is considered of fractal characteristic. Using fast fourier transform (FFT), the power spectrum of profile can be calculated and then be plotted verses the frequency on a log-log scale. The inclination ( $k_p$ ) and intercept ( $B$ ) can be calculated via regression analysis. Thereafter,  $D$  and  $G$  can be obtained by Eqs. (3) and (4), respectively.

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