



Fouling mechanisms of gel layer in a submerged membrane bioreactor



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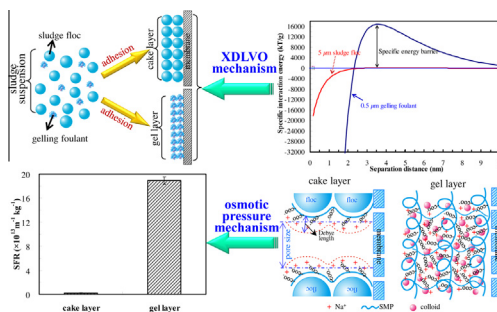
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HIGHLIGHTS

- Gel layer was more easily formed when SMP content was relatively high.
- Gel adhesion had higher contact interaction energy but had repulsive energy barrier.
- Gel layer had a SFR which was about 100 times higher than that of cake layer.
- The unusually high SFR of gel layer was caused by osmotic pressure mechanism.

GRAPHICAL ABSTRACT



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ABSTRACT

The fouling mechanisms underlying gel layer formation and its filtration resistance in a submerged membrane bioreactor (MBR) were investigated. It was found that gel layer rather than cake layer was more easily formed when soluble microbial products content in sludge suspension was relatively high. Thermodynamic analyses showed that gel layer formation process should overcome a higher energy barrier as compared with cake layer formation process. However, when separation distance <2.3 nm, attractive interaction energy of gelling foulant–membrane combination was remarkably higher than that of sludge floc–membrane combination. The combined effects were responsible for gel layer formation. Filtration tests showed that specific filtration resistance (SFR) of gel layer was almost 100 times higher than that of cake layer. The unusually high SFR of gel layer could be ascribed to the gelling propensity and osmotic pressure mechanism. These findings shed significant light on fouling mechanisms of gel layer in MBRs.

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1. Introduction

Membrane bioreactor (MBR) technology has been increasingly popular for various wastewater treatments during the last decades (Wang et al., 2008; Rajesh Banu et al., 2009; Banu et al., 2011; Lin et al., 2012). However, one primary drawback of this technology is membrane fouling, which highly limited its widespread applications (Zhang et al., 2006; Wang et al., 2008; Lin et al., 2011). Therefore, understanding and control of membrane fouling are essential

interest to the scientific and engineering community concerned with MBR technology.

Membrane fouling in MBRs is a general conception, which can actually be further distinguished as pore clogging, gel layer formation, cake layer formation and changes of cake layer. Among these processes, gel layer formation can highly reduce permeability of membrane, and has been considered as an important form of fouling in MBRs (Wang et al., 2008; Wang and Waite, 2009). Numerous studies have reported that gel layer mainly resulted from the gelation of the colloidal and dissolved matters (mainly soluble microbial products (SMPs) and biopolymer clusters (BPCs)) (note that these matters were termed as “gelling foulants” in this study) on membrane surface (Rosenberger et al., 2006; Liang

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et al., 2007; Wang and Waite, 2009; Yang et al., 2011; Xiao et al., 2013). It was reported that aeration could be effective in preventing long-term deposition of sludge particulates, whereas were difficult to remove gel layer (Wang et al., 2008; van den Brink et al., 2009, 2013; Wang and Waite, 2009). These studies indicate high interaction strength between gelling foulants and membrane surface, although sludge particulates (flocs) and gelling foulants are simultaneously present in mixed liquor in MBRs. Meanwhile, an apparent gel layer appears more easily formed under unfavorable operation conditions (Wang and Waite, 2009). While the formed gel layer is rather thin and porous, it usually has a high specific resistance (Wang and Waite, 2008a,b; Yang et al., 2011). From these studies, it can be seen that gelling foulants generally have high strength of interaction with membrane surface, and the formed gel layer possesses high specific resistance. To date, the causes of these phenomena are unclear, and have not been investigated exactly, although study has been conducted to characterize gel layer (Wang et al., 2008). Since gel layer highly affect membrane fouling, it is of primary significance to explore the fouling mechanisms of gel layer for better membrane fouling mitigation in MBRs.

It has been recently reported that adhesion of foulants to membrane surface in MBRs is a thermodynamic process, which can be depicted by extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory (Hong et al., 2013; Wang et al., 2013). Attachment of foulants to membrane surface highly depends on the interaction forces between foulants surface and membrane surface. Thus, the XDLVO theory may offer a novel way to track the fouling mechanisms of gelling foulants. As a general knowledge, SMP and BPC contained in gelling foulants typically carry negatively charged functional groups, such as carboxyl, hydroxyl and phosphoric groups. Presence of these groups may cause the change of water chemical potential in gel layer, which may in turn induce the change of filtration resistance (Keiding et al., 2001; Curvers et al., 2009; Chen et al., 2012). It was reported that presence of SMP and/or BPC in cake layer highly gave rise to cake resistance (Wang et al., 2007; Wang and Li, 2008; Lin et al., 2014). Therefore, analysis from chemical potential viewpoint may help to resolve the fouling mechanisms of gelling foulants (Wang et al., 2007). However, study neither experimentally investigated this issue based on chemical potential analysis.

Bearing the information above, the aim of this study was to investigate the fouling mechanisms of gel layer in a MBR. A laboratory scale MBR setup was continuously operated. Surface properties of foulant layers (including gel layer and cake layer) were compared. Based on these data, the interactions between foulants and membrane were analyzed. Thereafter, filtration resistance of both gel layer and cake layer were compared, and the underlying mechanism regarding the high specific filtration resistance (SFR) of gel layer was explored. This study would shed significant light on fouling mechanisms of gel layer.

2. Methods

2.1. Experimental setup and operation

A submerged MBR (SMBR) setup with 65 L effective volume was operated at the laboratory scale in this study (Fig. 1). The SMBR setup was composed of a biological tank where a membrane module was submerged. The membrane module with 0.1 m² effective filtration area was consisted of 5 vertical flat-sheet membrane elements. The membrane (SINAP Co. Ltd., China) was made of polyvinylidene fluoride (PVDF), and had a normalized pore size of 0.3 μm. Through the air diffuser underneath the membrane module, an air flow rate of about 180 m³_{air}/m³_{permeate} was supplied to provide shearing stress and oxygen. The SMBR was feed with

synthetic municipal wastewater, which had composition same to that used in previous studies (Hong et al., 2013; Zhang et al., 2013). Filtration was achieved by intermittent suction (4-min-on and 1-min-off) with a peristaltic pump. The membrane flux was maintained at approximately 30 L m⁻² h⁻¹ (LMH) with two calibrations made each day. The hydraulic retention time (HRT) and sludge retention time (SRT) were about 5.5 d and 45.5 d, respectively. Mixed liquid suspended solids (MLSS) concentration was ranged from 10 g L⁻¹ to 15 g L⁻¹ in the stable operation period.

2.2. Analytical methods

Sludge suspension samples were subjected to centrifuging at 2500×g for 5 min (GTR16-2, Beijing Era Beili Co., Ltd., China), and thereafter, the supernatants were collected as sludge “supernatant” samples. Meanwhile, sludge suspension samples were subjected to filtration in a stirred cell (model 8200, Amicon) with the same membrane used in MBR, and thereafter, the retained sludge were collected as cake layer samples. The gel layer samples were obtained by scraping off gel layer on membrane surface. These samples were then further treated for the following analyses.

Considering the easy degradability of influent organics, the organics in supernatant were believed to be mainly SMP. The supernatant SMP was normalized as the sum of proteins and polysaccharides. Another sample was prepared by dissolving a little amount of gel layer in deionized water. Proteins content in these samples was colorimetrically measured by Folin method, and polysaccharides content was colorimetrically measured by phenol/sulfuric acid method.

Zeta potentials of cake layer and gel layer samples were measured by using a Zetasizer Nano ZS (Malvern Instruments Ltd., UK) based on electrophoretic mobility method. A Zeta 90 Plus Zeta Potential Analyzer (Brookhaven Instruments, UK) was used to analyze zeta potential of membrane surface. Each prepared sample was at least measured in triplicate.

In order to analyze contact angles of three materials (virgin PVDF membrane, cake layer and gel layer), these samples were further treated. The virgin PVDF membrane was firstly immersed in ultrapure water for 48 h, and then cut into small pieces (2 cm × 4 cm). The small membrane pieces were mounted on a slide, and then dried in a desiccator for 24 h to remove surplus water. Above-mentioned prepared cake layer and gel layer samples were pressed to form a relatively flat surface by using two slides. Thereafter, these samples together with the pressing slides were dried in a desiccator for 24 h to remove surplus water. Three probe liquids (ultrapure water, glycerol and diiodomethane) were used for contact angle measurement. Static contact angles of the probe liquids on these prepared samples were measured by using a contact angle meter (Kino industry Co., Ltd., USA) based on the sessile drop method.

As for X-ray photoelectron spectrometer (XPS) analysis, above-mentioned prepared cake layer and gel layer samples were first placed in an alumina dish in a freeze-dryer for freeze-drying for 6 days (until freeze-dried). A Kratos Axis ULTRA XPS system was used to determine surface content of primary elements on sludge surface. This system uses a focused monochromatic Al K α X-ray (1486.7 eV) excitation source and a spherical section analyzer equipped with a 16-element multi-channel detector.

Water quality parameters including MLSS and chemical oxygen demand (COD) were measured according to the standard methods (APHA, 2005).

2.3. XDLVO approach

XDLVO theory can be used to describe the floc-membrane interfacial interactions in water (Hong et al., 2013). Accordingly,

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