



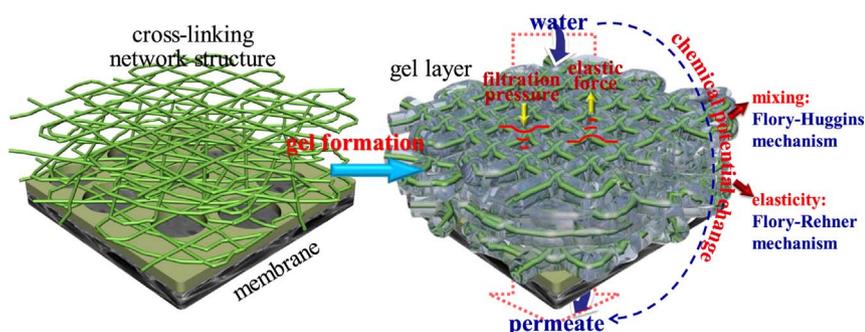
Mechanism analyses of high specific filtration resistance of gel and roles of gel elasticity related with membrane fouling in a membrane bioreactor

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GRAPHICAL ABSTRACT



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ABSTRACT

In this study, mechanisms and roles of gel elasticity in extremely high specific filtration resistance (SFR) were investigated. It was found that, as compared with cake layer in a membrane bioreactor (MBR), real gel layer in the MBR and agar gel possessed extremely high SFR. Foulant characterization showed that foulants were easy to bind water, and agar gel possessed a network structure. Mechanisms based on Flory-Huggins and Flory-Rehner models were deduced to describe the high SFR of agar gel. Model simulation showed that sum of SFR induced by the mixing chemical potential and the elastic chemical potential change is close to that of the agar gel, suggesting feasibility of the deduced models. Gel elasticity accounted for about 13% of total SFR of agar gel under conditions in this study. This study satisfactorily explained the extremely high SFR of gel, and addressed roles of gel elasticity in gel SFR.

1. Introduction

Membrane bioreactor (MBR) technology has been extensively researched and applied for wastewater treatment and reclaim in recent years mostly because of its tremendous advantages over conventional activated sludge process (CASP), steadily falling membrane price,

intensive demand for water reclamation, and stricter emission standard (Cai et al., 2017, Lin et al., 2012, Meng et al., 2017, Shen et al., 2015). In spite of the promising future of this technology, membrane fouling is still the major technical roadblock limiting its development (Chen et al., 2017, Shen et al., 2017, Zhao et al., 2017).

So far, intense aeration and subcritical flux operation appear the

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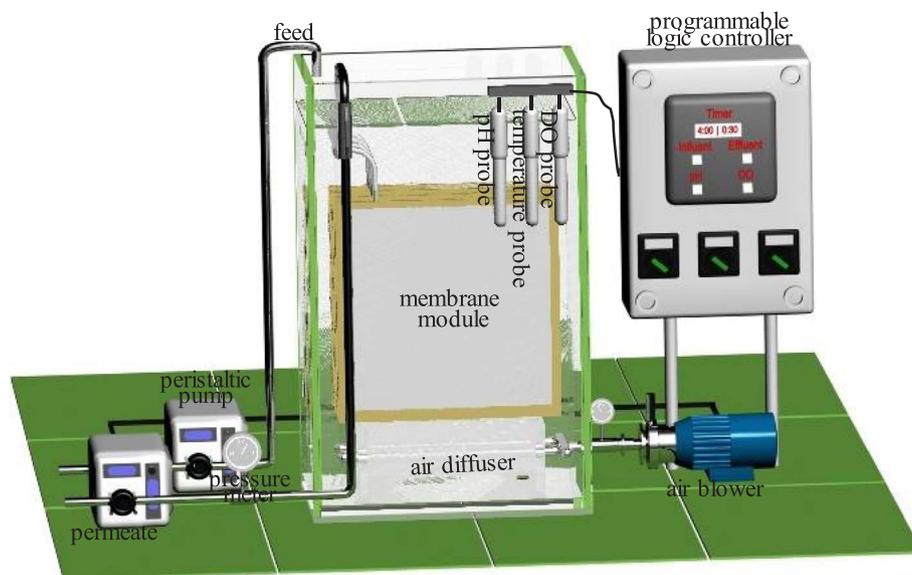


Fig. 1. Schematic of the lab-scale SMBR system.

most common strategies to mitigate foulant adhesion and membrane fouling in MBRs (Lin et al., 2013, Lin et al., 2014b, Wang and Waite, 2009). While efficient to resist long-term adhesion of sludge flocs and other particulates on membrane surface, these strategies appear cut no ice with gel layer formation in the long run of MBRs. For example, Wang et al. (2008) found that gel layer preferentially formed under subcritical flux operation with intense aeration in MBRs. Moreover, although the formed gel layer is generally porous and thin, it possesses extremely higher filtration resistance than other porous media. Specific filtration resistance (SFR) of cake layers has been reported to be usually at level of 10^{13} – 10^{14} m \cdot kg $^{-1}$ (Ping Chu and Li, 2005, Zhang et al., 2014). However, SFR of gel layer could reach dozens or even thousands of times of those of cake layers (Hong et al., 2014, Lin et al., 2009, Zhang et al., 2014). These phenomena cannot be explained by Carman-Kozeny equation, a representative equation of the traditional filtration theory (Hong et al., 2014, Yarnpakdee et al., 2015), puzzling the research community for a long time. Recently, a new mechanism deemed to be osmotic pressure effect underlying filtration through gel layer has been reported (Chen et al., 2016, Zhang et al., 2017, Zhang et al., 2013). It is believed that, water in gel layer has lower chemical potential than water in permeate due to its lower purity in gel layer (Zhang et al., 2018). As a result, there needs an additional force to pull water from gel layer to permeate in order to overcome this chemical potential gap. Such a chemical potential gap is sometimes called as osmotic pressure effect as osmotic pressure directly stems from chemical potential gap (Zhang et al., 2013). Existence of this mechanism has been well verified in the literature (Lin et al., 2014a).

These studies improved fundamental understanding of membrane fouling, and offered a general mechanical framework for studying membrane fouling related with gel. However, there are still several concerns remained unaddressed. Previous studies considered that the chemical potential gap was either resulted from two aspects: 1) retention of counter-ions in gel layer due to the abundant negatively charged functional groups carried by gel polymers (Hong et al., 2014, Zhang et al., 2013), and 2) mixing between polymers in gel and water (Chen et al., 2016, Lei et al., 2016). However, it appeared that these two aspects were not sufficient to explain all the related phenomena of gel filtration. It had been evident that gel layer typical possessed a key characteristic of honeycomb-type (network) structure (Wang and Waite, 2008a, Yarnpakdee et al., 2015). Obviously, above-mentioned two aspects did not involve the roles of this network structure. Meanwhile, macroscopic observation showed that a relatively dense gel layer

possessed a certain elasticity, and enabled to undergo elastic deformation under an applied load (Nitta et al., 2014). Moreover, for a gel layer modeled by agar, although the simulated SFR induced by the proposed osmotic pressure effect caused by mixing chemical potential mechanism was close to measured SFR of agar gel, it was still 10–20% lower than the experimental SFR value (Chen et al., 2016, Wang et al., 1997). So far, the lost 10–20% SFR value of gel layer, as well as above-mentioned structure and elasticity of gel layer remain unexplained. Fouling behaviors of activated sludge and real gel in MBRs are very complex and involve various processes and/or interactions, which hinders to explore the exact mechanisms responsible for aforementioned phenomena. In the literature, agar has been frequently used as a model gelling agent due to its availability, cost effectiveness, and the high ability to form gel (Yarnpakdee et al., 2015). Investigating filtration behaviors of agar gel may be helpful to reveal the universal fouling mechanisms of gel filtration.

Therefore, the aim of this study is to explore the universal mechanisms underlying the extremely high SFR of gel formed in MBRs. Special attention would be paid on the roles of honeycomb-type (network) structure and elasticity of gel layer. In practice, a model gelling agent, agar was utilized to form agar gel, which was subjected to filtration experiments. Based on the experimental results, new mechanisms and models responsible for the SFR of gel layer were proposed. This study would identify the contribution of elastic free energy to the gel filtration resistance, and reveal the root cause of extremely high SFR of general gels.

2. Material and methods

2.1. Experimental setup and operation

A laboratory scale submerged MBR (SMBR) device possessing 26 L effective volume with dimension (0.45 × 0.36 × 0.16 m height × length × width) was used for treatment of synthetic wastewater in this study. The schematic of the lab-scale SMBR system is shown in Fig. 1. Four flat-sheet modules of polyvinylidene fluoride (PVDF) membrane (normalized pore size: 0.1 μ m, effective membrane area: 0.4 m 2) were vertically mounted in a frame in the reactor. During operation, flux of membrane was maintained at about 20 L \cdot m $^{-2}$ ·h $^{-1}$ (LMH), and mixed liquid suspended solids (MLSS) concentration was in range of 10–15 g \cdot L $^{-1}$. Permeate was intermittently sucked (4 min-on and 1 min-off) through a peristaltic pump. Sludge suspension samples

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