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## Structure and flow calculation of cake layer on microfiltration membranes

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#### ABSTRACT

Submerged membrane bioreactors (SMBR) are widely used in wastewater treatment. The permeability of a membrane declines rapidly because of the formation of a cake layer on the membrane surface. In this paper, a multiple staining protocol was conducted to probe the four major foulants in the cake layer formed on a filtration membrane. Fluorescent images of the foulants were obtained using a confocal laser scanning microscope (CLSM). The three dimensional structure of the cake layer was reconstructed, and the internal flow was calculated using computational fluid dynamics (CFD). Simulation results agreed well with the experimental data on the permeability of the cake layer during filtration and showed better accuracy than the calculation by Kozeny–Carman method.  $\beta$ -D-Glucopyranose polysaccharides and proteins are the two main foulants with relatively large volume fractions, while  $\alpha$ -D-glucopyranose polysaccharides and nucleic acids have relatively large specific surface areas. The fast growth of  $\beta$ -D-glucopyranose polysaccharides in the volume fraction is mainly responsible for the increase in cake volume fraction and the decrease in permeability. The specific area, or the aggregation/dispersion of foulants, is less important to its permeability compared to its volume fraction.

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#### 43 Introduction

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Submerged membrane bioreactors (SMBRs) are widely used in 4546 wastewater treatment. The SMBR system has many advan-47 tages over conventional activated sludge methods, including 48 smaller space requirement, higher effluent quality, and lesser 49 sludge production (Judd, 2010). However, membrane fouling inevitably happens, which causes an increased flow resis-50tance and that severely declines the permeation flux (Zhao 51et al., 2000). The fouling problem causes high energy con-52sumption and frequent cleaning/replacement of the mem-53 brane, directly increasing the operational costs (Miura et al., 542007; Yang et al., 2006). For a fouled membrane, the total 55filtration resistance consists of three parts: the initial 56

resistance of a clean membrane (membrane resistance), the 57 pore blocking resistance (fouling resistance), and the cake 58 layer resistance caused by a cake formed by pollutant 59 particles on the membrane surface (Chang and Lee, 1998; 60 Cheryan, 1998; Guo et al., 2008). Among the three resistances, 61 cake layer resistance is considered the dominant one (Lee 62 et al., 2001; Chu and Li, 2006; Wang et al., 2007). 63

The cake layer consists of multiple foulants, such as 64 proteins and polysaccharides. The structure, composition, 65 and permeability of the cake layer change during the filtration 66 process (Yu et al., 2006). Chen et al. (2016) investigated the 67 mechanisms of membrane fouling caused by a gel layer and 68 found that filtration resistance linearly increased with gel 69 thickness. Studies have been conducted on the influences 70

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that cause the formation of cake layers, including sludge characteristics, operation conditions, and membrane materials (Le-Clech et al., 2006; Meng et al., 2009). However, understanding the fouling process without considering the complex structure of the cake layer remains difficult. Understanding the flow within the cake layer is helpful in revealing the fouling mechanism of SMBR.

Several approaches are used to observe the cake layer 78 79 structure, such as scanning electron microscopy (Lee et al., 2001; Fan and Huang, 2002) and atomic force microscopy 80 (Bowen et al., 1999; Yamamura et al., 2008). Gao et al. (2011) 81 characterized the cake layer structure by using various 82 analytical techniques and observed that the areal porosity 83 decreased from the top layer to the bottom layer. Recently, 84 fluorescent staining and confocal laser scanning microscopes 85 (CLSMs) have been frequently used to morphologically visual-86 ize cake layers (Neu and Lawrence, 1999; Strathmann et al., 87 2002; Yun et al., 2006). The main advantage of fluorescent 88 staining is that the different membrane foulants are shown 89 when corresponding fluorescence probes are used. Yoon Kim 90 et al. (2006) probed the cells, polysaccharides, and proteins 91 in a cake layer by using three stains: SYBR Green I, 92 fluorescent-labeled lectins and Hoechst 2495. Chen et al. 03 94 (2006a, 2006b) probed nucleic acids,  $\alpha$ -D-glucopyranose polysaccharides, β-D-glucopyranose polysaccharides, and proteins 95 96 using four stains: SYTO 63, Concanavalin A (Con A), Calcofluor 97 White, and Fluorescein Isothiocyanate (FITC). These compo-98 nents constitute the major biopolymers in a cake layer. In this study, a multiple staining protocol was conducted to probe the 99 four major foulants in the cake layer. 100

With the detailed component images acquired, the struc-101 tural parameters of the cake layer, such as porosity, can then 102be calculated (Chen et al., 2006a, 2006b; Hwang et al., 2007). 103 Computational fluid dynamics (CFD) can then be used to 104calculate 3D flow in the cake layer (Yang et al., 2012), and 105important flow parameters, such as permeability, can be 106 obtained. However, the CFD calculations are only accurate 107 when experimental validation is used. Thus far, very few 108 studies have been performed on this subject. 109

In this study, both experiment and CFD simulation were 110 performed on a submerged membrane filtration process. A 111 112multiple staining protocol was conducted to probe the four major foulants in the cake layer formed on the membrane, 113 and fluorescent images for each foulant were obtained. The 114 3D structure of the cake layer was reconstructed from the 115116 fluorescent images. Then, the flow in the cake layer was calculated using the CFD. The CFD method was compared 117 with the Kozeny-Carman model on permeability calculation. 118 The influence of volume fraction and specific surface area of 119 each foulant on its flow at different filtration times was 120121analyzed.

#### 123 1. Materials and methods

#### 124 **1.1. Experimental system**

125 A schematic of the submerged membrane filtration system is

shown in Fig. 1. The system consisted of a feed tank, a stirrer, amembrane chamber, a filtrate collection vessel, an electronic



Fig. 1 – Schematic diagram of the submerged membrane filtration system.

balance and a PC. The filter membrane was fixed on the 128 membrane chamber module with a filtration area of 1.77 cm<sup>2</sup>. 129 A water head of about 7.5 kPa (a 77 cm-high water column 130 below the membrane) supplied the filtration pressure. The 131 stirring rate is 200 r/min. The operating temperature was kept 132 at 20°C. The filtrate weight was measured using the electronic 133 balance and recorded by the PC. 134

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#### 1.2. Permeability from experiment

A mixed cellulose esters membrane (HAWG04700, Millipore) is 136 employed as the filtration membrane. It has a 0.45- $\mu$ m pore 137 size, 150-µm thickness and 79% porosity. The membrane is 138 hydrophilic and made from biologically inert materials of 139 cellulose acetate and cellulose nitrate. Prior to use, the 140 membrane was immersed in deionized water for 24 hr to 141 remove the soluble impurities and additives from the fabri- 142 cation process. Active waste sludge was acquired from a 143 wastewater treatment plant in Beijing. After 4-h gravitational 144 sedimentation, most suspended solids were removed from 145 the active sludge to show the contribution of colloidal 146 fractions in supernatant. The chemical oxygen demand for 147 the sludge and filtrate was 16,000 and 86.7 mg/L respectively. 148 The elemental composition of the dried samples was as 149 follows: C, 41.4%; H, 6.3%; and N, 5.6%. The sludge pH was 150 approximately 6.9. Membrane filtration experiments were 151 conducted under the same conditions, with different filtration 152 times of 300, 600, 900, 1200, 1500, and 1800 sec. The permeability 153 of the cake layer at different times was calculated using the 154 experiment method illustrated below. 155

In the filtration experiments, the permeate flux of fluid 156 across the membrane was expressed by Darcy's law with the 157 resistance-in-series model: 158

$$J_t = \frac{\Delta P_t}{\mu R_t} = \frac{\Delta P_t}{\mu (R_m + R_c + R_f)}$$
(1)

where  $J_t$  (m/s) is the membrane permeate flux,  $\triangle P_t$  (Pa) is 159 the transmembrane pressure,  $\mu$  (Pa·sec)is the fluid viscosity, 161  $R_t$  (m<sup>-1</sup>) is the total membrane filtration resistance,  $R_m$  (m<sup>-1</sup>) is 162

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