



New insights into membrane fouling based on characterization of cake sludge and bulk sludge: An especial attention to sludge aggregation

Xinying Su^a, Yu Tian^{a,b,*}, Hui Li^a, Cuina Wang^a

^aSchool of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China

^bState Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology (SKLUWRE, HIT), Harbin 150090, China

HIGHLIGHTS

- ▶ Cake and bulk sludge aggregation in MBR was evaluated by extended DLVO theory.
- ▶ Cake sludge had higher DSI and more colloids, LB-EPS and negative charge.
- ▶ Cake sludge exhibited worse aggregation ability than bulk sludge.
- ▶ Worse sludge aggregates was more easily attached to membrane for cake formation.

ARTICLE INFO

Article history:

Received 17 June 2012

Received in revised form 31 October 2012

Accepted 1 November 2012

Available online 9 November 2012

Keywords:

Membrane bioreactor (MBR)

Membrane fouling

Sludge aggregation

Extended DLVO theory

ABSTRACT

In order to obtain a better understanding of the relationship between sludge characteristics and the cake formation in membrane bioreactors (MBRs), the characteristics of cake sludge and bulk sludge were investigated and compared. Based on the extended Derjaguin–Landau–Verwey–Overbeek (extended DLVO) theory, the aggregation abilities of cake sludge and bulk sludge were also evaluated. It is observed that cake sludge showed worse aggregation ability than bulk sludge. Further analysis indicated that small flocs, colloids, loosely bound extracellular polymeric substances (LB-EPS), hydrophobicity and negative charge played important role in cake formation and sludge aggregation. Cake sludge with worse aggregation had higher distribution spread index (DSI), more colloids and LB-EPS, higher hydrophobicity and more negative charge. The results indicated that sludge aggregation might reflect membrane fouling potential of sludge.

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1. Introduction

Membrane bioreactors (MBRs) offer many advantages over conventional activated sludge process, such as reduced footprint, superior effluent quality, higher biomass concentration and less sludge production (Meng et al., 2009; Zhang et al., 2011). However, membrane fouling results in severe flux decline or rapid transmembrane pressure (TMP) increase, high energy consumption, and frequent membrane cleaning or replacement, which directly leads to the increase in maintenance and operating costs (Wang et al., 2009).

Sludge cake formation on the membrane surface is viewed as the major cause of membrane fouling in MBRs (Khan et al., 2009; Lin et al., 2011; Meng et al., 2007). The sludge cake mainly originates from the biomass of bulk sludge. It is reasonable to think that bulk sludge play a major role in the formation of the cake layer on

the membrane surface (Le-Clech et al., 2006). Some studies have revealed that the fouling behaviors of bulk sludge and cake sludge were significantly different (Buyukkamaci, 2004; Wang et al., 2007). Therefore, a detailed comparison of cake sludge and bulk sludge will be helpful to understand the formation and development of cake layer.

To date, several attempts have been made to characterize cake sludge and bulk sludge. Wang et al. indicated that the accumulation of biopolymer clusters within the pores of the sludge cake was mostly responsible for the unusually high filtration resistance of cake sludge (Wang et al., 2007). Lin et al. argued that small flocs, bound extracellular polymeric substances (EPS) and inorganic materials played important role in cake formation process (Lin et al., 2011). These studies mainly focused on the effect of certain components in sludge suspension on cake layer formation. Recently, some researchers have confirmed that the aggregation ability of bulk sludge played a key role in cake formation during membrane filtration process. Sludge aggregation depends on the EPS, sludge structure, surface charge, flocculation, settling properties, dewatering properties and adsorption ability (Sheng et al.,

* Corresponding author at: School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China. Tel./fax: +86 451 8628 3077.
E-mail address: hittianyu@163.com (Y. Tian).

Nomenclature

Δp	the pressure applied (Pa)	HRT	hydraulic retention time
U_{121}^{AB}	AB interaction energy (KT)	LB-EPS	loosely bound EPS
U_{121}^{EL}	EL energy (KT)	LW	Lifshitz–van der Waals
U_{121}^{LW}	LW energy (KT)	MBR	membrane bioreactors
ΔG_{121}^{IF}	the interaction free energy per unit area (mJ m^{-2})	MF	microfiltration
U_{121}^{XDLVO}	total energy of the interaction (KT)	MLSS	mixed liquid suspended solids
A	the filtration area (m^2)	PVDF	polyvinylidene fluoride
AB	acid–base	R_c	the cake resistance formed by the cake layer deposited over the membrane surface (m^{-1})
b	the time-to-filtration ratio	SMP	soluble microbial products
C	the total suspended solids (kg m^{-3})	TB-EPS	tightly bound EPS
CST	capillary suction time (s)	TMP	transmembrane pressure
d_{10}	diameter corresponding to 10% of cumulative undersize		
d_{50}	median diameter	<i>Greek symbols</i>	
d_{90}	diameter corresponding to 90% of cumulative undersize	θ	contact angle ($^\circ$)
DO	dissolved oxygen	μ	the viscosity of the permeate (Pa s)
DSI	distribution spread index	α	the specific cake resistance (m kg^{-1})
EL	electric double layer	γ^+	electron acceptor components (mJ m^{-2})
EPS	extracellular polymeric substances	γ^-	electron donor components (mJ m^{-2})
extended DLVO	extended Derjaguin–Landau–Verwey–Overbeek		

2010), which could be the characteristics of the flocs as a whole. Poor sludge aggregation leads to an increase in single microbial cells, smaller or unstable aggregates. Meng et al. pointed out that smaller fractions of aggregation particles could be readily deposited on the membrane surface by permeation drag (Meng et al., 2007). Tian et al. found that poor stability of sludge aggregates was prone to produce serious membrane fouling (Tian and Su, 2012). The aggregation ability of sludge is primarily governed by the interactions between sludge cells, which could be described by the extended Derjaguin–Landau–Verwey–Overbeek (extended DLVO) theories (Liu et al., 2010). Thus, a comparative investigation on the aggregation abilities of cake sludge and bulk sludge based on the extended DLVO theory might provide new insights into membrane fouling in MBR systems.

The objectives of this study were (1) to explore and compare the aggregation abilities of cake sludge and bulk sludge based on the extended DLVO theory; (2) to identify the characteristics of cake sludge and bulk sludge such as the contents of colloids, soluble microbial products (SMP), loosely bound EPS (LB-EPS) and tightly bound EPS (TB-EPS), particles size distributions and surface characteristics; (3) to evaluate the relationship between sludge characteristics and the aggregation abilities. To the best of our knowledge, this might be the first attempt to correlate membrane fouling with sludge aggregation. The results obtained in this study will be helpful to elucidate fouling phenomena, and the improved understanding will facilitate to optimize appropriate operational parameters to control membrane fouling.

2. Methods

2.1. Operation of MBR

A lab-scale 8 L MBR was operated in this study at room temperature 22 ± 3 °C. The MBR was installed with a submerged hollow fiber microfiltration (MF) membrane module. The membrane module was made of polyvinylidene fluoride (PVDF) with a nominal pore size of 0.1 μm and an effective surface area of 0.1 m^2 (Motian, China). The MBR was fed with synthetic municipal wastewater (glucose 227 mg L^{-1} ; starch 227 mg L^{-1} ; NaHCO_3 254 mg L^{-1} ; urea 33 mg L^{-1} ; $(\text{NH}_4)_2\text{SO}_4$ 121 mg L^{-1} ; KH_2PO_4 15.4 mg L^{-1} ; K_2HPO_4 19.6 mg L^{-1} ; $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 51 mg L^{-1} ; CaCl_2 12 mg L^{-1} ; $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$

17.48 mg L^{-1} ; ZnCl_2 0.13 mg L^{-1} ; $\text{Pb}(\text{NO}_3)_2$ 0.27 mg L^{-1} and $\text{MnSO}_4 \cdot 4\text{H}_2\text{O}$ 0.13 mg L^{-1}). A liquid level control was used to control the water level in the bioreactor. The effluent was collected directly from the membrane module by a peristaltic pump. A vacuum gauge was fixed between the membrane module and the peristaltic pump to monitor the TMP. Aeration was provided continuously underneath the membrane module so as to control membrane fouling and supply air for the biomass. The dissolved oxygen (DO) was monitored with a portable on-line DO meter (WTW inoLab Oxi level 2) and aeration rate was adjusted through the air flow meter. The membrane flux was set at 10 $\text{L m}^{-2} \text{h}^{-1}$ with an intermittent suction of 8-min on and 2-min off. The hydraulic retention time (HRT) was 10 h and the mixed liquid suspended solids (MLSS) was maintained at $9000 \pm 500 \text{ mg L}^{-1}$ (see Fig. S1) by sludge discharge. Prior to the experiments, the MBR was operated for over 4 months. When the operation of the MBR was terminated, the resistance-in-series model, as shown in the Supporting Information, was applied to evaluate different filtration resistances.

2.2. Sludge samples preparation

Cake sludge and bulk sludge were sampled from the lab-scale MBR. At the end of operation, the fouled membrane module was taken out from the bioreactor. Then cake sludge samples were obtained by flushing the membrane surface with pure water. Meanwhile, bulk sludge samples were collected from the MBR.

2.3. Batch cell test for determining specific cake resistance

Specific filtration resistances of cake sludge and bulk sludge can be evaluated by batch filtration tests following the method by Wang et al. (2007). The test was conducted in a dead-ended filtration cell using a qualitative filter paper at a constant pressure. The production of filtrate under pressure was continuously recorded. The specific filtration resistances can be calculated by Eq. (1)

$$\alpha = \frac{2\Delta p A^2 b}{\mu C} \quad (1)$$

where α is the specific cake resistance (m kg^{-1}), C is the total suspended solids (kg m^{-3}), Δp is the pressure applied (Pa), A is the filtration area (m^2), b is the time-to-filtration ratio, which is the slope

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