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Short Communication

New insights into membrane fouling in submerged MBR under sub-critical flux condition

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HIGHLIGHTS

- Membrane fouling under subcritical flux condition is a three-stage process.
- Bound EPS plays a significant role in fouling development at early stage.
- SMPs appear to be the major contributor to the self-accelerating fouling phenomena.
- Fouling is likely determined by floc characteristics when a cake of flocs formed.

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ABSTRACT

This study investigated the membrane fouling in MBRs under sub-critical flux condition. Results showed membrane fouling at subcritical flux evolved through a three-stage process: a slow linear increase in transmembrane pressure (TMP) (stage I), followed by an exponential increase in TMP (stage II), and finally a rapid linear TMP rise was observed at stage III. It was found that bound EPS would play a significant role in fouling development at stage I, while SMPs appeared to be the major contributor to self-accelerating fouling phenomena observed at stage II. At stage III, the entire membrane was covered by a cake layer of flocs, as the result, the fouling rate was likely determined by floc characteristics. This study offers new insights into the fouling development under sub-critical flux condition.

1. Introduction

Nowadays, membrane fouling remains a big challenge for widespread applications of membrane bioreactors (MBRs). It has been suggested that submerged MBR should be operated at a flux below the "critical flux", so-called subcritical flux, to maintain a sustainable permeability (Guglielmi et al., 2007). However, membrane fouling has been observed even at subcritical flux in MBR regardless of the system characteristics (Cho and Fane, 2002; Zhang et al., 2006; Hwang et al., 2008). The development of subcritical flux fouling over time has been generally described by a two-step process: a very small TMP rise followed by a TMP jump with a self-accelerating nature. The subcritical flux fouling behaviours have been studied in terms of extracellular polymeric substances (EPS) and other dissolved matters (Hwang et al., 2008; Patsios and Karabelas, 2011). However, the reason behind still remains unclear.

Therefore, this study aimed to investigate the subcritical flux fouling in two lab-scale MBRs for the treatment of synthetic wastewater at different SRTs. The fouling resistance and cake resistance were measured periodically, while analyses pertaining sludge characteristics, microscopic observations, and molecular weight distribution were performed in relation to subcritical flux fouling in the two MBRs. It is expected that this study can provide a better understanding of the subcritical flux fouling, especially TMP self-accelerating phenomena in MBRs.

2. Methods

2.1. Experimental set-up and operation conditions

Two identical MBRs (MBR-A and MBR-B) were operated in parallel at the same hydraulic retention time (HRT) of 12 h, aeration

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rate of 3 L/min, flux of 10 L/m^{2}h and a room temperature of 25 °C. However, MBR-A was operated at sludge retention time (SRT) of 30 days, while 80 days in MBR-B. The two MBRs were operated at the flux lower than the respective critical fluxes of 20–22 L/m^2 h (MBR-A) and 16–18 L/m^2 h (MBR-B). In each MBR, two flat sheet membrane modules were directly submerged in the aerobic compartment with a total effective membrane area of 0.1 m^2. The flat sheet membrane used in this work is hydrophilic PVDF membrane with a pore size of 0.22 μm (Millipore, GVWP). Permeate was obtained by a suction pump operated according to the 8 min on and 2 min off mode. Transmembrane pressure (TMP) in the MBR was monitored by the pressure transducers connected to a data logging system.

The seed sludge taken from a local municipal wastewater treatment plant was first acclimatized in a lab-scale MBR for more than one year. Each MBR was seeded with 5 g/L of the acclimatized activated sludge and fed with the synthetic wastewater consisting of 300 mg/L of glucose, 250 mg/L of sodium acetate, 160 mg/L of NH_4Cl, 40 mg/L of peptone, 30 mg/L of meat extract, 7.5 mg/L of KH_2PO_4, 7.5 mg/L of $FeSO_4$ and 2.5 mg/L of $MgSO_4$. In order to ensure the identical condition of the membrane in all the experiments, the used membrane was replaced with a new membrane at steady state when the TMP reached 55 kPa. MBR-A was operated for 60 days, while 160 days for MBR-B to reach steady state.

2.2. Analytical methods

Mixed liquor suspended solids (MLSS) and Mixed liquor volatile suspended solids (MLVSS) were measured according to standard methods (APHA, 1998). SMPs were separated from MLSS by centrifugation at 4000 rpm for 10 min (Universal 32R, Hettich-Zentrifugen, Germany), and the recovered supernatant was then filtered through 0.45 μm glass fiber filter.

In this study, bound EPS (bEPS) located at the outside surface of cells was extracted by formaldehyde–NaOH solutions as described by Li et al. (2012). Protein and polysaccharide concentrations were determined by UV absorbance on an UV/visible spectrophotometer (JASCO V-550, JAPAN) using bovine serum albumin (SIGMA, US) and glucose standards (SIGMA, US) as the reference materials respectively. Proteins were determined by Bradford method and polysaccharides were determined by phenol/sulphuric acid method.

Membrane resistance (R_m), fouling resistance caused by adsorption of dissolved matter and/or colloidal pore blockage (R_f) and cake layer resistance (R_c) were calculated from the resistance-in-series model (Hwang et al., 2008). Specific cake resistance (α_c) for activated sludge was determined according to the method by Ji et al. (2008). Mastersizer 2000 (Malven, UK) was used to measure the mean floc size (d_a) in the bulk solution as well as to calculate the fractal dimension (d_f) by means of the analysis on forward scattered light (Guan et al., 1998; Li et al., 2008). The fouled and unfouled membrane surfaces were examined by a scanning electron microscope (SEM) (JSM-5310LV, JEOL Japan). Analysis of molecular weight distribution of supernatant and permeate was conducted by means of Gel Permeation Chromatography (Waters, USA) equipped with an ultrahydrogel linear column (MW ranges: 103–107, Waters, USA) and a refractive index detector (RID, Waters 2414, USA).

3. Results and discussion

3.1. Trans-membrane pressure profiles and membrane fouling rate at different operation of the MBRs

Fig. 1 shows the time course of transmembrane pressures (TMP) in the steady state MBRs. Changes in TMP in the two MBRs followed a similar pattern, i.e. an initially slow linear TMP rise (Stage I) followed by an exponential TMP rise (Stage II). More interestingly, a rapid linear TMP rise (Stage III) was also observed in Fig. 1. However, it appeared that the time length of each stage would be related to SRTs. For example, in MBR-A (Run 1 and Run 2), the time span of stage I was 476–486 h, whereas 373–383 h was observed in MBR-B (Run 3 and Run 4). It was found that Stage I and II obviously lasted longer in MBR-A than in MBR-B, while Stage III was slightly shorter in MBR-B than MBR-A.

The experimental results at different stages were fitted into a curve, and the most fitted curves were linear, exponential and linear for stage I, stage II and stage III, respectively. The fouling rate in terms of $d\Delta P/dt$ at stage I in MBR-A was 0.0049 kPa/h (average), which was slightly higher than in MBR-B (0.0036 kPa/h). However, at stage II, the fouling rate in MBR-B was nearly 2.5-time higher than that in MBR-A. It was also observed that at stage III, fouling rate of MBR-B (1.55 kPa/h) was slightly higher than that in MBR-A (1.33 kPa/h).

3.2. Effects of biomass characteristics on membrane fouling rate at different stages

In this study, the fouling behavior was directly related to sludge characteristics because feed water characteristics, membrane material and hydrodynamic conditions for both MBR-A and MBR-B were identical.

Table 1 shows the sludge characteristics of the two MBRs operated at different SRTs. The different biomass characteristics were mainly caused by the different SRTs. For example, the higher bEPS in MBR-A was probably correlated to the shorter SRT, which is in agreement with the studies reported previously, i.e. bEPS in sludge flocs decreased as SRT increased (Li et al., 2008). SMPs were higher in MBR-B than in MBR-A, as presented in Table 1. Giving the synthetic feed solution used in this study, there would be little substrate residuals from the feed water in the supernatant, and the less biodegradable SMPs induced by cell lysis or cell release would probably account for most of the SMPs (Wu et al., 2011). Due to the lower F/M ratio in MBR-B (1.1 kg COD/kg MLSS d) compared to MBR-A (2.0 kg COD/kg MLSS d), it is believed that the higher SMPs in MBR-B was mainly caused by the accumulation of these less biodegradable substances. The different particle size (d_a) and fractal dimension (d_f) presented in the MBRs were believed to be closely associated with microbial community, and which was ultimately affected by the sludge retention time (Wu et al., 2011; Li et al., 2012).

It is known that critical flux represents the boundary between fouling by the dissolved/colloidal components and suspended matter of the biomass (Cho and Fane, 2002; Ognier et al., 2004; Zhang et al., 2006). Due to the subcritical flux condition and the intermittent relaxation (2 min after every 8 min of filtration) in this study, the sludge flocs could be removed from the membrane surface at stage I. Therefore, the pore blocking and/or surface fouling caused by favorable foulant-membrane attractions, in this case EPS and SMP, would dominate the fouling process. The fouling resistance caused by adsorption of dissolved matter and/or colloidal pore blockage within the membrane (R_f), was found to be pre-dominant at stage I (Fig. 2). This further confirmed that the foulant over membrane pores and/or surface at this stage was mainly consist of dissolved matter and/or colloidal. However, the fouling rate in MBR-B is lower compared to MBR-A despite SMP concentration was higher in MBR-B than in MBR-A. This indicated that fouling rate at stage I was probably correlated to bEPS instead of SMP (Table 1). In addition, compared to cake resistance, bEPS resistance would not be reduced effectively by increasing the aeration intensity (Khalili-Garakani et al., 2011). Therefore, the fouling at this stage reflects a more natural process colonization and/or biofilm