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A novel approach for mitigation of membrane fouling: Concomitant use of flocculant and magnetic powder

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HIGHLIGHTS

- Combined use of magnetic powder and poly dimethyl diallyl ammonium chloride.
- Reduction of SMPc and EPSc by adding the organic flocculant in MBR.
- Excellent performance in removing SMPc and EPSp by the magnetic powder.
- Enhanced dehydrogenase activity with the addition of magnetic powder.
- Improvement of membrane fouling mitigation.

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ABSTRACT

Membrane fouling alleviation by addition of poly dimethyl diallyl ammonium chloride (PDMDAAC) and magnetic powder (Fe_3O_4) was investigated. It was found that magnetic powder associated with PDMDAAC had a good performance on mitigation of membrane fouling, improvement in dehydrogenase activity and enhancement of biomass growth. The optimal dose of PDMDAAC was determined by using constant pressure dead-end filtration unit. Maximum permeate flux was attained at 400 mg/L of PDMDAAC addition. Continuous experiment was conducted in a submerged membrane bioreactor (MBR) system and biomass parameters such as soluble microbial products (SMP), extracellular polymeric substances (EPS), dehydrogenase activity, zeta potential, and capillary suction time (CST) were analyzed. Best results were obtained with a combination of 120 mg/L of magnetic powder and 400 mg/L of PDMDAAC. This study results demonstrated that PDMDAAC played a major role in SMPc and EPSc reduction, whereas magnetic powder had better performance in decreasing SMPc and EPSp.

1. Introduction

Membrane bioreactor (MBR) has been widely used for both municipal and industrial wastewater treatment process and it has been considered as an efficient way to effectively remove chemical, physical as well as microbiological contaminants from treated water before they are released into the environment (Huang and Lee, 2015; Rosenberger et al., 2002; Zhang et al., 2014). However, membrane fouling in MBR remains the bottleneck for its widespread application. According to previous reports, the characteristics of activated sludge mixed liquor had a significant impact on membrane fouling, including suspended solids, viscosity, particles (size distribution), soluble organic substances, extracellular polymeric substances (EPS: mainly contains carbohydrate (EPSc) and protein (EPSp)), soluble microbial products (SMP: mainly contains carbohydrate (SMPc) and protein (SMPp)), colloids and flocs (fractal dimension) (Iversen et al., 2009; Ji et al., 2008; Kim et al., 2001; Müller et al., 1995). The organic substances tended to deposit on the membrane surface, creating a gel-like structure followed by a cake layer (Jarusutthirak and Amy, 2006; Rosenberger et al., 2005). Consequently, the filterability of MBR is significantly weakened.

To alleviate membrane fouling, several approaches have been investigated by altering membrane material, changing hydraulic conditions, adding coagulants, adsorbents or flocculants (Hosseinzadeh et al., 2015; Jung et al., 2006; Matos et al., 2011; Wang et al., 2012; Wu et al., 2006). The organic flocculants addition in MBR resulted in the reduction of SMP and increase in mean floc size. Similarly, addition of inorganic flocculants prolonged the sustainable filtration time by reducing SMP and surface charge of wastewater sludge (Ji et al., 2010a). In comparison with inorganic

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flocculants, organic flocculants had the advantages of augmenting the average floc size, increasing the zeta potential, and improving the sludge dewatering performance (Wang et al., 2014). However, the addition of chemical flocculants is known to affect the microbial growth, biomass production and its activity. Further free H^+ produced by hydrolysis (of chemical flocculants) simulated the microorganisms to secrete more SMP (Barker and Stukey, 1999).

Magnetic particles could be applied as a foulant reducer without severe effect on biological floc or activity in MBR because of not only their high surface area for the adsorption of proteins, DNA, dyes, humic acids and heavy metals (Ai et al., 2011; Bromberg et al., 2009; Illes and Tombacz, 2003), but their inertness and biocompatibility. Moreover, the potential of magnetic particles can be exploited to enable their recovery, re-activation, and re-use in MBR on account of their responsiveness to magnetic field. The protein-induced fouling in MBR was inhibited through the porous micro-sized magnetite (Fe_3O_4) powder due to its significant adsorption for bovine serum albumin (BSA) (Semblante et al., 2013), whereas the impact of magnetic powder on sludge characteristics, and its corresponding effect on membrane fouling have not been investigated till date and still need to be verified.

In this work, the objective of this study was to explore the influence of magnetic powder associated with organic flocculant (PDMDAAC) on sludge characteristics and membrane fouling in a submerged membrane bioreactor. Possible mechanisms involved in membrane fouling reduction were also observed and examined.

2. Methods

2.1. Constant pressure dead-end filtration test

2.1.1. Activated sludge mixed liquor and flocculant

Activated sludge was collected from the secondary clarifier in the Qige sewage treatment plant, Hangzhou, China. The synthetic wastewater was used as a feed (in MBR), which consisted of sodium acetate (1292.9 mg/L), peptone (43.1 mg/L), NH_4Cl (98.7 mg/L), $MgSO_4$ (13.9 mg/L), KH_2PO_4 (22.8 mg/L), yeast extract (17.2 mg/L), $MnSO_4$ (1.6 mg/L) and $FeSO_4$ (0.3 mg/L). And its characteristics on average were as the following: pH value (7.3), COD (650 mg/L), TN (34.4 mg/L), TP (6.7 mg/L) and initial flux (50 mL/min). The mixed liquor suspended solids (MLSS) concentration was kept constant at around 8000 mg/L.

The cationic polymer, poly dimethyl diallyl ammonium chloride (PDMDAAC with the molecular formula of $(C_8H_{16}NCl)_n$) was used in the dead-end filtration experiment and the submerged MBR system. The flocculant was mixed with 200 mL of activated sludge mixed liquor with increasing concentrations (0–900 mg/L).

2.1.2. Experimental unit and operating conditions

The constant pressure dead-end filtration device used in this study was similar as described by Matos et al. (2011). The UF membrane with a pore size of 0.22 μm was used and the trans-membrane pressure (TMP) was kept constant at 0.05 MPa. A 200 mL of sample was fed into UF cup in each test and its permeate flux was recorded every 20 s by computer.

2.2. Submerged MBR system

2.2.1. Experimental unit and magnetic powder

The submerged MBR system was performed as per the optimal conditions mentioned by Wang et al. (2014), which mainly consisted of six parts, that was, five identical chambers of membrane bioreactors, feed tank, suction pumps, computer, air pumps and tubular diffusers (for hydraulic removal of particles from membrane surface). The membrane modules used in this study was made of polyvinylidene fluoride (PVDF) in flat-sheet type with a filtration area of 0.1 m^2 and a membrane pore size of 0.2 μm. And the effective volume of a MBR chamber was 21 L.

The characteristic of magnetic powder was described in Table 1.

2.2.2. Operating conditions

The characteristics of activated sludge and feed concentration of synthetic wastewater were the same as the one used in the dead-end filtration experiment. The well-mixed activated sludge in a larger feed tank was equally distributed into the five membrane bioreactors with MLSS concentration of 8000 mg/L each.

The submerged MBR system was regulated by an automatic control system. All suction pumps (operated at 27 rpm) were set to operate 9 min followed by 1 min idle. The air pumps were set to operate alternatively every 2 h and the trans-membrane pressure was recorded every 2 min by the computer.

The experiments were conducted in four rounds. Each round of experiment was performed with one control MBR (without any addition of flocculant or magnetic powder), one MBR was only supplemented with PDMDAAC, and the other three MBRs were operated with addition of both magnetic powder and PDMDAAC. The PDMDAAC concentrations were all same and the optimal dose was determined by constant pressure dead-end filtration experiment. The three reactors in each round were conducted with various concentration of magnetic powder until the optimal concentration was determined and the magnetic powder was supplemented after 30 min of flocculant addition. The five suction pumps began to work simultaneously 30 min after the flocculant and magnetic powder were all added, and the MBRs were run in parallel at the same operating conditions. Each round was completed once the TMP of all five MBRs reached to 50 kPa (maximum sensor level). And Γ_{50} was noted as the filtration time once the TMP reached to 50 kPa.

The membranes would be cleaned and replaced with the cleaned membrane once the TMP reached to 50 kPa. The cleaning of fouled membrane was conducted as follows: initially membranes were rinsed with tap water and then cleaned using chemical solutions (immersed in 0.5% sodium hypochlorite for 24 h and then 0.5% citric acid for another 24 h). Finally, the cleaned membranes were stored in 0.2% sodium hypochlorite.

2.3. Analytical methods

2.3.1. Dehydrogenase activity

The enzyme dehydrogenase plays an important role in biological redox reactions. Further, the dehydrogenase activity indicated the microbial biodegradation activity (Ji et al., 2010b; Łebkowska et al., 2011). To measure the dehydrogenase activity, 4 mL of activated sludge mixed liquor was centrifuged at 10,000 rpm for 8 min. The supernatant was discarded and the pellet settled in the centrifuge tube was re-suspended to 4 mL with distilled water, and this centrifugation step was repeated thrice. Finally, 2 mL of the re-suspended mixed liquor was extracted by addition of

Table 1
The characteristics and surface area of the magnetic powder used in this study.

Items	Parameters
The average pore diameter of adsorption (nm)	1.333
BET single-point ($P/P_0 = 0.2$) specific surface area (m^2/g)	21.313
BET multi-point specific surface area (m^2/g)	24.256
Langmuir specific surface area (m^2/g)	43.707
T-chart method micropore internal surface area (m^2/g)	21.197
T-chart method external specific surface area (m^2/g)	3.059
BJH cumulative adsorption pore internal surface area (m^2/g)	0.649
BJH cumulative desorption pore internal surface area (m^2/g)	226.408