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Procedia Engineering 148 (2016) 726 - 734

Procedia Engineering

www.elsevier.com/locate/procedia

4th International Conference on Process Engineering and Advanced Materials

Enhancement of Membrane Fouling Control in Hybrid Aerobic Membrane Bioreactor System for Domestic Waste Water Application: Effect of Alum Concentration

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Abstract

Additional of alum into the membrane bioreactor was found to be able to enhance the performance for the separation process. In this study, the influence of different alum concentrations on membrane fouling control and characteristics of activated sludge of membrane bioreactors were evaluated. Membrane bioreactor with low alum concentration of 1.0 g/L was found able to provide better filterability performance compared to MBRs without and with excessive alum addition (3.0 g/L and 5.0 g/L). This study indicates that agglomeration of flocs and biodegradation mechanisms which took place simultaneously in the MBR with 1.0 g/L of alum is able to reduce the total EPS concentration in the MBRs and subsequently contributing to better membrane fouling control.

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Keywords: Alum; membrane fouling control; activated sludge; membrane bioreactors

1. Introduction

Membrane bioreactor is a process which can be functioned to treat wastewater by integrating membrane together with activated sludge [1]. Application of membrane bioreactors have increased tremendously in industries because of their ability to produce good quality effluent [2], less sludge production due to relatively higher sludge retention time [3] and smaller footprint [4].

*Corresponding author. Tel.: +605-468 8888; fax: (+605)-466 7449. *E-mail address:* lywong@utar.edu.my Compared to the conventional municipal wastewater treatment systems, membrane bioreactor technologies have been recognized as a better alternative for wastewater treatment and water reclamation [5]. The main concern of membrane bioreactor is membrane fouling which is caused by pore clogging [6] and cake formation on the membrane surface [7]. The foulants can be in the forms of high viscosity mixed liquor suspended solids (MLSS) [8], soluble microbial products (SMP) [9], extracellular polymeric substances (EPS), colloids [10] and particle size of MLSS [11]. EPS refers to soluble organic macromolecules that are a major fraction of soluble microbial products due to the metabolism and cell. It comprises of polysaccharides, protein, nucleic acid and some polymeric compounds [12]. According to Frolund et al. (1996), concentrations of protein and polysaccharide are the two major components that would contribute to membrane fouling [13].

Recently, studies show that the fouling control of membrane bioreactor could be enhanced by adding coagulant into the system. Coagulant is commonly used to remove suspended solids, colour, turbidity and Chemical oxygen demand (COD) in industrial wastewater [14]. Sodium aluminate (NaAlO2), aluminium sulphate (alum) and polyaluminium chloride (PACl) are the common types of aluminium-based coagulants which are widely used in wastewater treatment in removing natural organic matter [15]. Three main mechanisms of coagulation are sweeping, bridging and charge neutralization [16]. Floc strength was arranged in the order of (1) sweep, (2) charge neutralization, and (3) bridging mechanism.

Furthermore, the coagulant added encourages the formation of larger flocs by agglomeration of colloids and smaller particles which are less likely to fit into the pore of the membrane surface [17]. Coagulants e.g. FeC1₃ and alum are reported to be effective in enhancing membrane filterability and limit membrane fouling. The data revealed that pre-added of coagulant into the wastewater should be done first instead of adding the coagulant directly into the bioreactor [17]. However, this would cause high accumulation of alum in the MBR, where high dosage of coagulant will lead to low pH, which is harmful to living cell and higher production of sludge. Therefore, some researchers suggested that the performance of membrane bioreactor is improved when the alum dosage is kept low (less than 20 mg/L)[10]. In this study, relatively higher alum concentrations at 1 g/L and more were added directly into the MBRs to investigate their performance in terms of membrane fouling control. The loss of the alum from the desludging process was top-up daily. This is to make sure the concentration of the alum in the MBRs can be maintained.

In summary, there are quite a number of studies which had clearly proved that the usage of coagulant can significantly improve the filterability of membrane. However, comparison of coagulant addition into MBRs at different concentrations in term of their functions and mechanisms has not been carried out so far. Therefore, this study is aimed to investigate the effect of different alum concentrations in membrane fouling control for MBR application.

2. Methods and Materials

Four bench-scales of submerged MBRs with different concentrations of coagulant were set-up under the same operating conditions. The working volume of each MBR was 2.5 L. The MBRs have aeration installed at the bottom of the reactor to provide oxygen to the biomass in the system. Alum $[Al_2(SO_4)^3]$ was selected as coagulant in this study due to its cost-effectiveness. 1 g/L, 3 g/L and 5 g/L of dosages were analysed in order to determine the optimum dosage of alum for MBRs system. The MBRs were namely MBR-C (conventional MBR without alum), MBR-1 (hybrid MBR with 1 g/L alum in the system), MBR-3 (hybrid MBR with 3 g/L alum in the system), and MBR-5 (hybrid MBR with 5 g/L alum in the system). Hollow fiber Polyethersulfone (PES) membrane with pore size range from 0.1-0.5 μ m was provided by Sakti Suria Sdn. Bhd. Set-up for the MBRs is shown in Figure 1.



Figure 1: Set-up of Conventional MBR and Hybrid MBRs in this study.

The sludge used was originally seeded from domestic wastewater activated sludge treatment plant, Kampar. Microbes were fed with synthetic wastewater daily [18]. The ingredients of the synthetic wastewater are shown in Table 1.

Table 1: Ingredients of synthetic wastewater

Formulations	Concentration (g/L)	Manufacturers	
Sodium acetate trihydrate $[C_2H_3NaO_2 \cdot 3(H_2O)]$	26.3	Grade AR produced by $QR\ddot{e}C^{TM}$	
Proteins	8.5	Dugro milk powder (Malaysia production)	
Glucose	30.0	Icing sugar (Malayan Sugar MFG.Co.BHD)	
Iron (II) sulfate (FeSO ₄)	2.0	Grade AR produced by $QR\ddot{e}C^{TM}$	
Monopotassium phosphate (KH ₂ PO ₄)	2.0	Grade AR produced by $QR\ddot{e}C^{TM}$	

All MBRs were operated at 24 hours hydraulic retention time (HRT) and sludge retention time (SRT) of 10 days; whereby 10% of sludge which equivalents to 250 ml of wastewater were removed daily. This action will decrease the alum concentration within the MBRs. In order to maintain the concentration of alum, a replenishment of alum will be done immediately after every de-sludge process. The operating conditions for all MBRs were kept constant along the experiment. The specification for each MBR is given in Table 2.

Type of MBR	Operation mode	Working Volume (L)	Type of MLSS	Organic loading rate (mg/L/d)	Type of microfiltration membrane
MBR-C	Batch	2.5	AS	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	Hollow fiber membrane
MBR-1	Batch	2.5	AS + 1 g/L alum	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	Hollow fiber membrane
MBR-3	Batch	2.5	AS + 3 g/L alum	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	Hollow fiber membrane
MBR-5	Batch	2.5	AS + 5 g/L alum	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	Hollow fiber membrane

Table 2: Operational parameters for MBRs

MLSS tests were carried out to indicate the concentration of activated sludge. The microbes are considered have reached acclimatization stage when the steady amount of MLSS concentration is achieved. In this study, the MLSS is determined using the following equation.

$$MLSS = [(M2 - M1)/V1] \times (1000mL/1L)$$
(2.1)

where,

MLSS = Mixed liquor suspended solid

M1 = Weight of the filter paper before filtration

M2 = Weight of the filter paper after drying in an oven for a day $(105^{\circ}C)$

V1 = Input volume for filtration (10mL)

Polysaccharide and protein are the major elements which can be found in EPS as the major foulants of membrane. In this study, concentration of polysaccharides and protein were analysed in accordance to the method of phenolsulfuric acid [19] and Bradford reagent for protein with bovine serum albumin (BSA) as standard [20] respectively. Meanwhile, particle size distribution of the suspended solid in sludge was determined by using Malvern Mastersizer 2000, Hydro2000 MU (A). Data for transmembrane pressure were obtained through transmembrane pressure transducers (Logit USA). Determination of COD was carried out using HACH method 8000 to investigate the effluent quality and the removal efficiency of MBRs system, which involved two equipment: COD heat reactor (model DRB 200) and spectrophotometer (model DR 6000).

3. Results and Discussion

In this study, the effectiveness of MBRs in membrane fouling control was tested after the MBRs had reached a steady state (after being cultivated for 90 days). According to the result as shown in Figure 2, MBR-1 had proved to be able to perform better than the MBR-C in membrane fouling control. This shows that the performance of the system in terms organic matter removal could be improved by adding 1 g/L of alum into the system.

However, further increment of the alum concentration in the system had caused adverse effects to the MBRs. MBR with the highest alum concentration (MBR-5) was found having the worst membrane fouling control among all the MBRs. To investigate the phenomenon, effects of alum on MLSS, particle size, pH, EPS and COD concentrations of the solute in MBRs were analysed and discussed respectively.



Figure 2: Transmembrane Pressure of MBRs with different alum dosages.

It was noticed that the concentration of MLSS in the MBR-5 was the highest, followed by MBR-3, MBR-1 and MBR-C as shown in Table 3. It shows that coagulant could increase MLSS production, where the same result was observed by [21]. By referring to the particle size, it shows that addition of alum in MBRs significantly reduced the particle size distribution of the activated sludge in terms of 'volume' but very less effect on 'number'. At first, it was expected the particle size of sludge would grow with the addition of alum due to the coagulation process. However in this study, the particle size was greatly reduced with the higher dosages of alum. According to Wang et al. (2010) and Zahid and El-Shafai (2012), this situation may cause by the effect of excessive alum which would retard the growth of filamentous bacteria that lead to a subsequent reduction in the floc size [21] [22].

Therefore, increment of the MLSS with comparatively smaller flocs in the system might be the reason that causing more attachment of smaller flocs to the pores of the membrane surface, and subsequently increase the membrane fouling rate for MBR-3 and MBR-5. Thus, optimum dosage of alum is critical in the system in order not to create excessive sludge production.

MBR Systems		Particle	size (µm)	
	MLSS (g/L)	MLVSS (g/L)	D ₅₀ (Volume)	D ₅₀ (Number)
MBR-C	7.50 ± 0.4	5.80 ± 0.3	0.883 ± 0.01	0.190 ± 0.01
MBR-1	7.90 ± 0.4	5.60 ± 0.3	0.841 ± 0.01	0.190 ± 0.01
MBR-3	8.70 ± 0.4	6.10 ± 0.3	0.669 ± 0.01	0.192 ± 0.01

Table 3: Concentration of MLSS, MLVSS and particle size (D50)

MBR-5 9.20 ± 0.4 6.60 ± 0.3 0.547 ± 0.01 $0.195 \pm$	0.01
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Figure 3 shows that mixed liquor of the four different MBRs which had become more acidic (pH decreased) as the dosage of coagulant increased. Initial pH in solutes of the MBR-3 and MBR-5 was 6.13 but dropped to below 6 when 3 g/L and 5 g/L of alum were added into their bioreactors respectively. According to Wu et al. (2005), as the pH of mixed liquor decreased below than 6.0, any additional of coagulants would then affect the activities of microorganisms [17]. Guo et al. (2007) also found out that, excessive inorganic coagulant residual of alum is one of the major inorganic foulants which would contribute to membrane fouling [23]. By referring to Figure 3, it shows that the optimal dosages of coagulant (alum) should be kept around 1.0 g/L or lower in order to control the pH above value of 6. That could explain why MBR-1 was able to perform better in membrane fouling control compared to the MBR-3 and MBR-5.



Figure 3: Variation of pH in MBRs with different alum dosage.

Polysaccharide and protein were also measured in this study and their total concentrations are known as total EPS. Concentrations of protein and polysaccharide in the MBRs are presented in Table 4. Highest polysaccharide concentration was found in MBR-5, followed by MBR-3, MBR-1, and MBR-C. Concentration of protein was found declined from the MBR-C to MBR-1, but increased greatly when the alum concentration in the system was increased to 3 g/L, and achieve the highest in the system with 5 g/L of alum.

Table 4: Concentrations of polysaccharides, protein and total EPS

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MBRs	Polysaccharides (mg/L)	Protein (mg/L)	Total EPS (mg/L)
MBR-C	76.14 ± 7	108.65 ± 8	184.79 ± 8
MBR-1	78.00 ± 6	101.26 ± 7	179.26 ± 6
MBR-3	78.05 ± 8	110.93 ± 7	188.98 ± 7
 MBR-5	93.00 ± 5	115.67 ± 8	208.67 ± 6

Total EPS concentration decreased when 1g/L of alum was added into the system but excessive dosage of inorganic coagulant (alum) would have adverse effect on MBR in terms of total EPS concentration. This finding was similar to the study results found by Ji et al. (2009) [24]. Relatively higher EPS concentration in their system may be the main reason led to the low membrane fouling control performance of the MBR-3 and MBR-5.

Table 5 shows the amount of organic loading rate as influent for each MBR per day and their performance in reducing COD concentration in their bioreactors. COD removal rate with the presence of 1 g/L alum is the best compared to other MBRs system. Song et al. (2007) also noticed that the COD removal rate with low concentration of alum in MBR would be improved as well its membrane fouling control [25]. The trend of COD removal rate is similar to total EPS removal rate. The improvement of the COD removal rate may be due to the coagulation effect of alum which involved both the adsorption and binding effects [17]. The binding ability of alum could provide simultaneous agglomeration and biodegradation effects that would improve the COD removal rate. However in this study, it was observed that excessive of alum would cause the adverse effect in the COD removal rate.

COD (mg/L)	Influent (mg/L)	Supernatant (mg/L)	Removal Rate by Bioreactor (%)	Permeate (mg/L)	Removal Rate by MBR (%)
MBR-C	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	$\begin{array}{c} 21000 \\ \pm 200 \end{array}$	22.79	8500 ± 100	68.75
MBR-1	$\begin{array}{c} 27200 \\ \pm \ 300 \end{array}$	$\begin{array}{c} 20700 \\ \pm 200 \end{array}$	22.91	$\begin{array}{c} 8300 \\ \pm 100 \end{array}$	69.49
MBR-3	$\begin{array}{c} 27200 \\ \pm \ 300 \end{array}$	$\begin{array}{c} 21500 \\ \pm 200 \end{array}$	20.96	9200 ± 100	66.18
MBR-5	$\begin{array}{c} 27200 \\ \pm 300 \end{array}$	$\begin{array}{c} 21700 \\ \pm 200 \end{array}$	20.22	$\begin{array}{c} 9100 \\ \pm \ 100 \end{array}$	67.50

Table 5: Chemical oxygen demand concentrations and removal rate of the MBRs

4. Conclusion:

Optimum concentration of alum plays an important role in enhancing performance of the MBRs system. The hydrolyzed coagulant could supply positive charges and long chain molecules for the formation of bigger flocs and colloids to enhance the charge neutrality and bridging which result in the removal or organic substances. The binding ability of alum could provide simultaneous agglomeration and biodegradation effects that improve the performance of membrane filterability. MBR with 1 g/L alum able to remove COD up to almost 70%. However, excessive addition of alum would lead to decrease in pH, particle size, increase of total EPS production, COD concentration and subsequently affect the membrane fouling control rate.

Acknowledgments:

The authors acknowledge financial support from research grants of Ministry of Science, Technology and Innovation (MOSTI) with project No. 03-02-11-SF0161. Sakti Suria (JB) Sdn Bhd is also acknowledged for their kindness to sponsor the membranes to carry out this study.

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