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Membrane bioreactor performance improvement by adding adsorbent and coagulant: a comparative study

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ABSTRACT

Membrane bioreactors (MBRs) have become popular in recent years due to its excellent organic pollutants removal efficiency. However, its popularity has been restricted by a major constraint which is high maintenance cost incurred by membrane fouling in the system. Currently, hybrid MBRs with additives were found efficient in controlling membrane fouling. In this study, the performance of two additives consist of powdered activated carbon (PAC) and alum in enhancing membrane fouling control in MBRs were determined and compared. Three 6L laboratory-scale submerged MBRs known as conventional MBR (without additive), MBR-PAC (added with PAC), and MBR-Coagulant (added with alum) with SRT 30 were set up. It was observed that the MBRs with additive could enhance MLSS concentrations by about 7–13% and reduce the protein concentration (one of the main foulants) by 43–70%, respectively. The results revealed that by adding alum and PAC into the MBRs, membrane fouling control of MBRs could be improved. MBR-PAC performed best in this study as it could be operated steadily without any sign of transmembrane pressure "jump" as compared to the other two MBRs even it was operated under "stress" condition by treating high strength wastewater during the experimental filtration period.

Keywords: Membrane bioreactors; Additives; Powdered activated carbon; Alum; Fouling control

1. Introduction

Membrane bioreactor (MBR) is a combination process of biological with membrane in a reactor which could replace the clarifiers and tertiary processes in the conventional water treatment plants. MBRs are mostly employed to treat industrial and domestic wastewater. The use of MBR technology is increasing worldwide due to the following: (1) it is able to produce relatively higher effluent quality, (2) its smaller footprint, (3) it could treat relatively higher organic loading rate, and (4) it has relatively lower surplus sludge production [1].

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However, the advantages of the MBR technology have been offset by some constraints where membrane fouling is the major concerns. Membrane fouling has caused a significant drawback on the use of the MBRs commercially as it incurs high maintenance costs [2]. One of the foremost reasons that initiate the fouling of a membrane is the pore blockage during filtration process [3]. This is mainly due to the size of the suspended pollutants in the MBRs [4] and the accumulation of the activated sludge on the membrane surface [2]. Solutions to the problems include (1) prevention of the activated sludge accumulation on the membrane surface and (2) reduction of fine pollutants in suspension in the MBRs using adsorbent or coagulant process [5].

In order to improve the MBRs performance, various studies were carried out recently by incorporating additives into the system. One of the popular additives is coagulant, which has been used for decades in water treatment system to control the water quality throughout the process. In MBR application, chemical coagulation has been used to enhance sludge characteristic and control membrane fouling [6].

Inorganic coagulant, e.g. alum and iron has positive effect in lowering the fouling rate for biofilm MBR even in relatively lower dosage [7]. When appear in the system, alum could significantly remove the submicron organic contents. Polymeric coagulant (ferric sulfate) is able to enhance the removal of organic matter in supernatant and enlarge the sludge floc size. It could enhance membrane fouling control and thus prolong the membrane filtration process [8]. Research work which had been done by Song et al. also indicated that alum as coagulant is able to enhance membrane fouling control by coagulating fine particles into larger particles. The permeate flux of a MBR system increased by twofolds after it was added with 500 mg/L of alum [6]. Waleed and Saber also confirmed the advantages of using alum as a coagulant as it could inhibit the growth of filamentous bacteria in the MBRs [4].

Even though the benefits of using coagulant as additive in MBRs had been reported, but its limitation and mechanism in helping membrane fouling control are still unclear. One of the limitations reported is that concentration of coagulant needed to bring beneficial effect to MBRs would be higher than that of using it for traditional water and wastewater treatment [9]. Moreover, a relatively higher dosage might bring down the pH of wastewater in the bioreactor, which would be harmful for living cells and might cause higher sludge production rate [8].

Due to those limitations, PAC has become a new attraction to the researchers as additive to enhance the

MBRs performance [10]. Studies found that PAC could turn into "biologically activated carbon" (BAC) once it is mixed with activated sludge and able to improve fine pollutants removal [3]. BAC was proven to be capable in removing color [10], organic carbon [11], micropollutants [5], and trace organic [12] in wastewater treatment. Simultaneous processes of adsorption and biodegradation may be the reason for BAC having good performance where microorganisms in the biofilm of BAC help to biodegrade the pollutants previously absorbed by the PAC [3].

Furthermore, PAC was found to be able to reduce EPSs in the floc [13] and the bulk liquid [14] and other fine foulants such as total organic carbon (TOC) [15], fine colloids [16], soluble metabolic products (SMPs) (3), and chemical oxygen demand (COD) [17] in the supernatant due to its adsorption effect. According to Seo et al., adsorption and biodegradation were able to eliminate most of the substances with molecular weight cutoff less than 1,000, and those with higher than 1,000 were gradually degraded by microorganisms of BAC during extended contact [15].

In addition, the scouring effect of PAC in the MBR could also enhance the membrane flux [18]. It was reported that PAC in a MBR could depolarize and remove fine particles accumulated on the membrane surface through scouring effects or enhanced fluid turbulence in the presence of bubbling with appropriate condition [16]. The good performance of MBR with PAC may also be due to its simultaneous adsorption and biodegradable effects [19,20]. It was reported that MBRs with PAC is able to improve fouling control up to 3.5 times compared to the MBRs without PAC [20].

To date, there are several studies being done on the effects of coagulant and PAC in MBRs. However, those studies focused only on either coagulant or PAC addition, and at different operation conditions. Comparison of performances among the additives of alum and PAC under the same conditions is still lacking. Therefore, this study aims to evaluate the effectiveness of both the additives in enhancing membrane fouling control in MBR simultaneously under the fair conditions.

2. Materials and methods

2.1. Materials

The membrane used was cellulose acetate membrane. The synthetic feed for the MBRs consisted of 26.3 g/L of sodium acetate trihydrate [C₂H₃NaO₂·3 (H₂O)] grade AR produced by QRëCTM, 17.1 g/L of Dugro milk powder produced in Malaysia, 52.6 g/L of icing sugar produced by Malayan Sugar MFG. Co. Bhd, 2.0 g/L of iron (II) sulfate (FeSO₄) grade AR produced by QRëCTM, and 2.0 g/L of monopotassium phosphate (KH₂PO₄) grade AR produced by QRëCTM.

2.2. Experimental set-up

The study was carried out by setting up three 6L MBRs: Conventional MBR (without additive), MBR-PAC (added with PAC), and MBR-Coagulant (added with alum) as per Fig. 1. The MBRs were cultivated using activated sludge from the local municipal wastewater treatment plant in Kampar, Perak, Malaysia. The Mixed Liquor Suspended Solid (MLSS) of the MBRs were cultivated to reach the concentration above 20 g/L. High MLSS concentration might stress the systems to produce relatively higher concentration of fine pollutants, so the beneficial of using PAC and coagulant in membrane fouling control could be evidenced.

In this study, 1.0 g/L of PAC and 1.0 g/L of aluminum sulfate were added into the MBR-PAC and MBR-Coagulant, respectively. The sludge retention time (SRT) for the MBRs was 30 d. A 200 mL of sludge was disposed and refilled with 200 mL of fresh synthetics wastewater on a daily basis. Disposed additives were refilled daily to compensate the losses and maintain the concentration in the respective MBRs. After cultivating the MBRs for 120 d, the performances of PAC and the coagulant in membrane fouling control were compared and tested using crossflow (Fig. 2) and dead-end filtration systems (Fig. 3).

2.3. Analytical methods

Concentration of polysaccharides (mg/L) in the MBRs was measured using the method of phenol–sulfuric acid [21] and Bradford reagent with bovine serum albumin (BSA) as standard was used for protein measurement (mg/L) [22]. COD (mg/L) concentration

was measured using Hach DR 6000 Spectrophotometer at 620 nm wavelength. MLSS concentration was measured using the following formula:

$$MLSS = [(M_2 - M_1)/V_1] x (1,000 \text{ mL/L})$$
(1)

where MLSS = Mixed liquor suspended solid; M_1 = Weight of the filter paper before filtration; M_2 = Weight of the filter paper after drying in oven; V_1 = Input volume for filtration (10 mL).

3. Results and discussion

Three MBRs with the volume of 6 L each were setup in this study, namely the conventional MBR (without additives), MBR-Coagulant (with alum), and MBR-PAC (with powdered activated carbon). A total of 1.0 g/L alum and 1.0 g/L PAC were added into the MBR-Coagulant and MBR-PAC, respectively, from the beginning of cultivation stage. The MBRs were operated at SRT of 30 d where 200 mL of sludge was discharged daily from all MBRs for SRT control. A total of 3.3% of the PAC and alum from MBRs was lost daily due to desludging. In order to maintain the concentration of additives in the MBRs, PAC and alum were replenished for the respective MBRs accordingly. MBRs were cultivated for 120 d before they were analyzed based on their performance in terms of membrane fouling control, polysaccharides and protein concentration, COD concentration, MLSS, and particle size distribution.

3.1. Membrane fouling control efficiency of the different MBRs

The result of the membrane fouling control of the MBRs is as shown in Fig. 4. It revealed that addition of additives into the MBRs could successfully improve the performance of MBRs in membrane fouling





Fig. 2. Crossflow filtration system (A: Sludge tank; B: Water pump; C: Inlet pressure gauge; D: Membrane holder; E: Outlet pressure gauge; F: Computer).



Fig. 3. Dead-end filtration system.



Fig. 4. Fouling control performance for conventional MBR, MBR-PAC, and MBR-coagulant.

control. Both of the MBRs with additives performed better compared to the conventional MBR. The membrane used for conventional MBR had seriously fouled just after it was operated for 30 min at the flux of 10 $L/m^2/h$. By adding alum in the MBR-Coagulant, the filtration operation could be prolonged to about 70 min using the same operation flux. MBR-PAC performed best with no sign of serious membrane fouling even it was operated for about 120 min. This proved that PAC is better than alum in terms of membrane fouling control.

To verify the findings, the used membranes from the MBRs were tested for their irreversible fouling control with the dead-end filtration system as per Fig. 3. The result obtained is shown as per Table 1. Throughout the filtration process, the used membrane from the conventional MBR needed the longest time to filter out 100 mL distilled water, followed by the used membranes from the MBR-Coagulant and MBR-PAC. This indicates that the membrane used in conventional MBR had suffered relatively serious irreversible fouling compared to the other membranes

Types of membrane	Duration (sec)	Excess (sec)	
New membrane	17.81	0	
Used membrane in conventional MBR	58.10	40.29	
Used membrane in MBR-Coagulant	37.69	19.88	
Used membrane in MBR-PAC	19.60	1.79	

Table 1

Duration needed for 100 mL of distilled water to pass through used membranes

used in MBR-Coagulant and MBR-PAC. Addition of additives into the system did prevent fine pollutants from clogging the pores of the membranes. It is quite a surprise to observe that the used membrane from MBR-PAC could still provide almost the same performance as the brand new membrane, even it had been used to filter the high strength wastewater from the MBR-PAC for about 120 min.

3.2. Polysaccharides and protein concentration in the different MBRs

Polysaccharides and protein (indicators of EPSs concentration) have been identified as the major foulants to cause membrane fouling during filtration [23]. In this study, the concentrations of polysaccharides and protein in respective MBRs were tested and the results are as shown in Table 2. The conventional MBR had the highest concentration of EPSs, followed by the MBR-Coagulant and the MBR-PAC which contained the least EPSs concentration. This may be the reason to explain the good performance of MBR-PAC in membrane fouling control and higher flux production compared to the conventional MBR and MBR-Coagulant. Chang and Lee also reported that, when the concentration of EPSs inside the bioreactor increased, the performance of membrane fouling control would decrease [24].

3.3. COD and MLSS concentrations in the different MBRs

Table 3 shows the information about the COD concentrations in influent, supernatant, and effluent of the three different MBRs. The results revealed that the three MBRs were able to treat the high strength wastewater effectively. However, the MBRs added with alum and PAC performed slighly better than the conventional MBR by having relatively better quality of effluent.

Table 4 shows the average values of MLSS concentrations of the MBRs which they had been cultivated for 120 d. The finding shows that the addition of activated carbon and coagulant into the MBRs could increase the MLSS concentration. The improved growth of the MLSS in both the MBR-Coagulant and the MBR-PAC may due to the attached growth of the bacteria. The binding property of the coagulant would encourage the formation of a bigger and heavier floc in the MBR-Coagulant. Higher MLSS was found in the MBR-PAC, this may be due to its activated carbon has relatively higher surface area which may improve the adsorption of pollutants and indirectly encourage the attached growth of the bacteria.

Previous findings reported by Cicek et al. indicated that higher MLSS would lead to higher membrane fouling rate in MBR [25]. However, this study shows that additional additives into the MBR had significantly improve the membrane fouling control where the MBR-PAC and MBR-Coagulant had relatively higher MLSS concentration compared to the conventional MBR. One of the major reasons contributing to such a phenomenon may be due to scouring effect created by the flocs that forms by additives in the MBRs. The flocs, in the presence of air bubble, scour and removed the foulants that clogging the membrane surface when they contacted with the membranes in the MBRs. Bigger floc was found to be more effective in removing the fine foulant from the membrane surface

Table 2 Concentration of total EPS in the different MBRs

Types of MBR	Polysaccharides concentration (mg/L)	Protein concentration (mg/L)	Total EPS (mg/L)	Flux (L/m²/h)
Conventional MBR	242.47 ± 0.78	170.00 ± 0.42	412.47	10.91
MBR-Coagulant	234.58 ± 0.54	96.38 ± 0.14	330.96	18.71
MBR-PAC	233.93 ± 0.24	51.58 ± 0.62	285.51	36.38

COD (mg/L)	Influent	Supernatant	Permeate	Removal rate (%)
Conventional MBR	$13,217 \pm 30$	$2,700 \pm 30$	$2,354 \pm 12$	82.18
MBR (Coagulant)	$13,217 \pm 30$	$2,640 \pm 24$	$2,178 \pm 60$	83.52
MBR (PAC)	$13,217 \pm 30$	$2,563 \pm 12$	$1,952 \pm 0.78$	85.23

Chemical oxygen demand (COD) concentrations and removal rate of the MBRs

Table 4 MLSS Concentration in various respective MBRs

Types of MBR	MLSS Concentration (g/L)	Excess (%)	
Conventional MBR	28.61 ± 1.16	0.00	
MBR (Coagulant)	30.83 ± 0.98	7.76	
MBR (PAČ)	32.57 ± 0.59	13.84	

compared with the smaller flocs [20]. This indicates that PAC and coagulant able to form bigger flocs, and creating a condition which can depolarize and remove fine particles accumulated on the membrane surface in the presence of bubbling [16].

In addition to the scouring effect, the adsorption effect from the biological activated carbon (BAC) was also able to reduce the total amount of EPSs in the MBR-PAC [19] as per Table 2 which would contribute to the better membrane fouling control. The MBR-PAC which had performed best in membrane fouling control is also had the lowest total amount of EPSs compared to the other MBRs.

3.4. Fine particle size distribution of the different MBRs

It is known that the tiny particles have relatively higher tendency to foul the membrane. Based on the



Fig. 5. Particle size distribution in conventional MBR, MBR-PAC, and MBR-coagulant in number.

particle size distribution as shown in Fig. 5, it was observed that the conventional MBR on average is consistently having relatively higher percentage of tiny particles compared to both the MBR-Coagulant and the MBR-PAC. This indicates that the conventional MBR would be more easily to be fouled compared to both MBR-PAC and MBR-Coagulant as supported by the result obtained in Fig. 4.

4. Conclusions

The effects of PAC and alum toward the performance of MBRs were successfully investigated. Compared with the conventional MBR, both the hybrid MBRs performed better in terms of membrane fouling control. The findings show that MBR-PAC and MBRcoagulant were able to remove EPSs more effectively. Even having higher MLSS in the systems, MBRs with additive were able to perform more efficiently in decreasing the pollutant compared to the conventional MBR. By comparing between MBR and PAC and MBR-Coagulant, the former has achieved better performance in both the membrane fouling control and pollutant removal rate which may be due to its simultaneous adsorption, biodegradation, and scouring effects.

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Table 3

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