

PERFORMANCE OF NEWLY CONFIGURED SUBMERGED MEMBRANE BIOREACTOR FOR AEROBIC INDUSTRIAL WASTEWATER TREATMENT

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Abstract

The application of membrane to replace secondary clarifier of conventional activated sludge, known as membrane bioreactor, has led to a small footprint size of treatment with excellent effluent quality. The use of MBR eliminates almost all disadvantages encountered in conventional wastewater treatment plant such as low biomass concentration and washout of fine suspended solids. However, fouling remains as a main drawback. To minimize membrane fouling, a new configuration of submerged membrane bioreactor for aerobic industrial wastewater treatment has been developed. For the new configuration, a bed of porous particle is applied to cover the submerged ends-free mounted ultrafiltration membrane. Membrane performance was assessed based on flux productivity and selectivity. By using tapioca wastewater containing high organic matter as feed solution, reasonably high and stable fluxes around 11 l/m².h were achieved with COD removal efficiency of more than 99%. The fouling analysis also shows that the newly configured ends-free membrane bioreactor exhibits lower irreversible resistance compared with the submerged one. In addition, the performance of pilot scale system, using a membrane module with 10 m² effective area and reactor tank with 120 L volume, was also assessed. The flux achieved from the pilot scale system around 8 l/m².h with COD removal of more than 99%. Hence, this study has demonstrated the feasibility of the newly configured submerged ends-free MBR at larger scale.

Key words: ends-free, flux, fouling, membrane bioreactor, wastewater

Abstrak

Aplikasi membran untuk menggantikan klarifier sekunder dalam sistem lumpur aktif konvensional membutuhkan tempat pengolahan yang kecil dengan kualitas luaran baik sekali. Sistem ini dikenal sebagai bioreaktor membran. Penggunaan bioreaktor membran dapat mengeliminasi hampir semua kelemahan yang dihadapi dalam pengolahan air limbah konvensional seperti konsentrasi biomassa rendah dan terbawanya padatan tersuspensi halus, namun fouling masih menjadi kendala utama. Untuk meminimasi fouling membran, suatu konfigurasi baru bioreaktor membran terendam untuk pengolahan air limbah industri secara aerobik telah dikembangkan. Untuk konfigurasi baru ini, membran ultrafiltrasi ends-free terendam ditutupi dengan suatu unggun partikel porous. Kinerja membran dievaluasi berdasarkan pada produktivitas fluks dan selektifitas. Dengan menggunakan air limbah tapioka yang mempunyai kandungan organik tinggi sebagai larutan umpan, diperoleh fluks sekitar 11 l/m².h dan efisiensi penyisihan COD lebih dari 99%. Analisis fouling juga menunjukkan bahwa konfigurasi baru ini memiliki tahanan irreversible lebih rendah daripada konfigurasi terendam lainnya. Selain itu, kinerja sistem skala pilot dengan menggunakan modul membran 10 m² dan tangki reaktor 120 L juga dievaluasi. Fluks yang dihasilkan dari sistem skala pilot ini adalah sekitar 8 l/m².h dan efisiensi penyisihan COD lebih dari 99%. Jelas bahwa studi ini telah menunjukkan kelayakan konfigurasi baru yang dikembangkan untuk skala lebih besar.

Kata kunci: ends-free, fluks, fouling, bioreaktor membran, air limbah

INTRODUCTION

The discharge of the improper treated wastewater directly to water body has led to a serious

impact to the environment. Membrane technology as a separation process appears as the best alternative to improve the performance of conventional wastewater

treatment. The application of membrane unit to replace secondary clarifier of conventional activated sludge has led to a small footprint size of treatment plant with excellent effluent quality. This combination of membrane technology with biological process is known as membrane bioreactor (MBR). Today there are more than 1000 installations of MBR all over the world.

The use of membrane bioreactor eliminates almost all disadvantages encountered in conventional wastewater treatment plant, such as low biomass concentration (3000-4000 mg/l), washout of fine suspended solids in the effluent (>20 mg/l), and no elimination of germs (Gunder and Krauth, 1999). The sub micron size of membrane pore allows membrane bioreactor to retain the biomass completely, including bacteria and viruses, therefore it also act as disinfections device. The ability of MBR to retain biomass completely also allows the operation of MBR under biomass concentration higher than 8,000 mg/l (Yamamoto *et al.*, 1989; Ueda *et al.*, 1997). In some cases, the biomass concentration can reach 50,000 mg/l (Muller *et al.*, 1995), far beyond the biomass concentration commonly found in conventional biological wastewater treatment, *i.e.*, 3,000-4,000 mg/l.

The configurations of MBR consist of external-MBR and submerged-MBR. In the external MBR, membrane module is placed outside the bioreactor, while in the submerged MBR, membrane module is placed directly inside the bioreactor. Nowadays, there

is a strong tendency to employ the submerged configuration compared with external one due to lower cost of fabrication and maintenance, and also lower energy consumption.

However, fouling remains as a main drawback for the spreading use of MBR application. The presence of fouling is usually characterized by the flux decline during the filtration process. Fouling in membrane process can be defined as irreversible, or not easily reversible, deposition of retained species onto or into the membrane (Fane and Cho, 2001). Fouling makes frequent membrane cleaning and consequently membrane replacement, which then increases maintenance and operating costs.

Numerous fouling reduction techniques have been studied to minimize membrane fouling particularly in submerged configuration as shown in Table 1. By far, aeration is the most common method that used extensively for fouling reduction considering its ability to create shear stress on the surface of the membrane. However, several drawbacks are reported as a result of the use of aeration such as the trapping of bubbles among the membrane fibers and the unevenly bubbles flow distribution. Bouhabila, *et al.* (1998) observed that an increase in the air flow rate partly stimulated the cake removal efficiency. However, there was a critical value beyond which the air-flow rate increase had virtually no effect on the cake-removing efficiency.

Table 1. Fouling reduction technique

Techniques	Reference
Aeration	Ueda, <i>et al.</i> , 1997; Bouhabila, <i>et al.</i> , 1998; Rosenberger, <i>et al.</i> , 2002; Espinosa-Bouchot & Cabassud, 2003; Behman, <i>et al.</i> , 2003; Rabie, <i>et al.</i> , 2004
Hybrid with activated carbon completely mixed in the bioreactor	Pirbazari, <i>et al.</i> , 1996; Kim, <i>et al.</i> , 1998; Dosoretz & Dodekker, 2004; Li, <i>et al.</i> , 2005; Khirani, <i>et al.</i> , 2006; Guo, <i>et al.</i> , 2005; Mohammadi & Esmaelifar, 2005; Guo, <i>et al.</i> , 2006; Fang, <i>et al.</i> , 2006
Zeolite addition	Lee, <i>et al.</i> , 2001
ELDE-MBR configuration (external loop dead-end MBR)	Espinosa-Bouchot & Cabassud, 2003
Utilization of riser and downcomer	Shim, <i>et al.</i> , 2002
Modification of reactor chamber	Kulick III, 2004; Del Vecchio, <i>et al.</i> , 2003
Intermittent filtration	Chiemchaisri, <i>et al.</i> , 1993; Futamura, <i>et al.</i> , 1994; Yeom, <i>et al.</i> , 1999; Lee, <i>et al.</i> , 2001; Choi, <i>et al.</i> , 2002; Albasi, <i>et al.</i> , 2002; Shim, <i>et al.</i> , 2002; Song, <i>et al.</i> , 2003; Ahn, <i>et al.</i> , 2003
Backflush	Cote, <i>et al.</i> , 1997; Rosenberger, <i>et al.</i> , 2002; Chang, <i>et al.</i> , 2003; Espinosa-Bouchot & Cabassud, 2003
Operating at critical flux	Bouhabila, <i>et al.</i> , 1998; Blocher, <i>et al.</i> , 2002; Li & Fane, <i>et al.</i> , 2003
Optimization of the distance between the membrane module and the wall of bioreactor	Ozaki & Yamamoto, 2001
Utilization of inclined-plate	Xing, <i>et al.</i> , 2005
Utilization of moving carrier (polyurethane cubes coated with activated carbon, plastic granules)	Lee, <i>et al.</i> , 2006; Artiga, <i>et al.</i> , 2005
Polymer addition	Yoon & Collins, 2006
Membrane surface modification	Yu, <i>et al.</i> , 2005; Yu, <i>et al.</i> , 2006a; Yu, <i>et al.</i> , 2006b; Asatekin, <i>et al.</i> , 2006
The use of aerobic biogranules activated sludge	Li, <i>et al.</i> , 2005; Li, <i>et al.</i> , 2007

All of the above mentioned methods still enable the direct contact of mixed-liquor of activated sludge with membrane surface, hence, promote membrane fouling. In this work, a newly configured submerged MBR has been developed. The submerged membrane is covered by porous particle bed in which the porous particle bed will act as a protector to minimize direct contact of mixed-liquor with membrane surface. Moreover, the porous particle bed gives a scouring effect as *in situ* mechanical cleaning to minimize membrane fouling and to extend the lifetime of the membrane. Furthermore, the growth of microbes on the porous structure of the porous particles apart from microbe grown in suspended form is also expected to give additional biodegradation effect on small organics responsible for membrane fouling. The new configuration of submerged ends-free MBR (eMBR) is schematically described in Figure 8. The difference of this configuration compared with conventional submerged MBR is the existence of porous particle bed on the surface of the membrane. The ends-free hollow fiber membrane unit was immersed in bioreactor tank and covered by porous particle, i.e., granular activated carbon or zeolite. The activated sludge was mixed and aerated by airflow supplied at the bottom of bioreactor, which act to fluidize the porous particle bed. The membrane unit was operated in outside-in mode.

EXPERIMENTAL
Feed Solution

The feed solution used in this experiment was a mixture of activated sludge and tapioca starch wastewater. The activated sludge used in this study was acclimatized with tapioca starch wastewater for almost 2 years. The tapioca starch wastewater had a COD concentration of 4000 – 8000 mg/l.

Membrane Bioreactor Equipment

The MBR used in this study consist of three configurations that operate in parallel, i.e., submerged MBR, newly configured submerged ends-free MBR (eMBR) with zeolite and activated carbon as the porous particle to cover the membrane module (Ze-eMBR, Ac-eMBR), respectively. The configuration of this newly configured e-MBR is schematically described in Figure 1. The difference of this configuration compare with conventional submerged MBR is the existence of porous particle bed on the surface of the membrane. The ends-free hollow fiber membrane unit was immersed in bioreactor tank and covered by porous particle, i.e., granular activated carbon and zeolite. Each of them has a diameter size between 1-3 mm. The activated sludge was mixed and aerated by airflow supplied at the bottom of bioreactor, which act to fluidize the porous particle bed. The membrane unit was operated in outside-in mode. The characteristics of the membrane are shown in Table 2. These membranes were supplied by GDP Filter, Indonesia. The working volume of the bioreactor was constantly maintained to be 6 L for lab-scale apparatus and 120 L for pilot-scale system.

Experimental Procedure

The experiments were carried out in room temperature (23-25°C), operating pressure of 0.1-0.3 bar, aeration rate of 6.75 l/min and 9 l/min and operated on batch and continuous mode. To evaluate fouling mechanisms, pure water fluxes were measured under following conditions: (1) for a new membrane, (2) for a membrane which had been fouled by reversible fouling, and (3) for a membrane which had been fouled by irreversible fouling (cannot be cleaned by water surface cleaning).

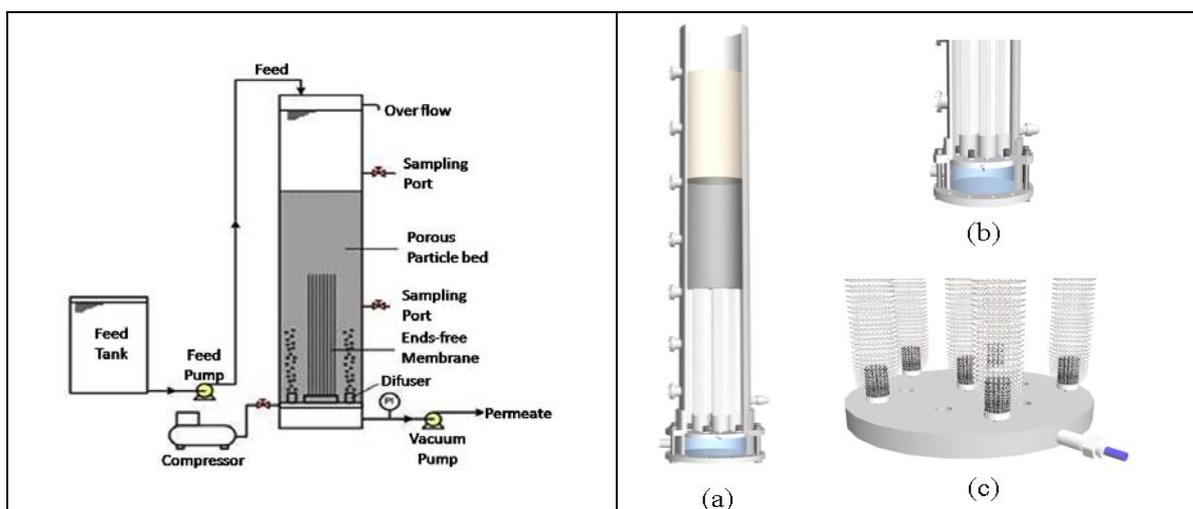


Figure 1. Schematic of newly configured submerged ends-free MBR (a: full-system; b: inner side of the MBR; c: air-flow pattern from diffuser located in the bottom of MBR)

Table 2. Membrane specification

Membrane process	Microfiltration
Membrane module	Ends-free hollow fiber
Membrane material	Polysulphone
Chemical surface	Hydrophilic
Pore size	0.1 μm
Outside diameter	1 mm/1.2 mm
Inside diameter	0.5 mm / 0.9 mm

Analytical Methods

The concentration of mixed liquor suspended solids and COD concentration were determined according to Standard Methods for Examination of Water and Wastewater [APHA, 1992].

RESULTS AND DISCUSSION

The COD removal characteristics of newly configured submerged ends-free MBR (eMBR) compared with conventional MBR and activated sludge is shown in Table 3. The activated sludge process contributes to 90% of COD removal. Meanwhile, by incorporating membrane, as in submerged MBR, an increase of COD removal at average of 97.65% is observed. In the meantime, both Ze-eMBR and Ac-eMBR show better COD removal compare with the submerged one, i.e. 97.68% and 99.36%, respectively. The residual COD found in the permeate represents the soluble non-biodegradable COD and/or soluble biodegradable COD of the treated water that is not degraded by microorganisms. The difference between the rejection characteristics of Ze-eMBR and Ac-eMBR also implies the importance of porous particle selection according to its effectiveness in adsorbing the soluble contaminants.

Table 3. COD removal comparison

Run	Rejection (%)			
	Activated sludge	Submerged MBR	e-MBR	
			Zeolite	Act. carbon
I	85.94	97.97	97.34	99.09
II	87.67	97.19	96.92	99.67
III	91.67	97.99	99.72	99.00
IV	90.71	96.85	99.11	99.79
V	90.15	97.65	97.68	99.36
Avg	90.15	97.65	97.68	99.36

During these trials under different TMPs tested for each configuration, the hydraulic resistance is rapidly increased at the beginning of the filtration period. However, the initial flux is reasonably high. This result indicate that fouling still occur in the e-MBR configuration but with different magnitude as can be seen from critical flux shown in Figure 2. Critical flux determination was done according to Le Clech *et al* (2003).

Figure 2 present critical flux of all configurations at TMP between 0.10 – 0.30 bar. It can

be seen that by increasing TMP up to 0.25 bars, higher critical flux is obtained. However, the increase of TMP to 0.30 bars gives a contrary result. According to Cheryan (1998) flux will be affected by the TMP under conditions where concentration polarization effects are minimal. These also emphasize that at TMP range of 0.10-0.25 bar, the filtration process is in the pressure-controlled region. Therefore, the optimum TMP for all configurations is achieved at TMP of 0.25 bars. Further, both Ze-eMBR and Ac-eMBR show higher flux compared with normal submerged configuration, in which the highest is achieved by Ac-eMBR. It is shown that fouling still occurs on eMBR configuration but the effect is less.

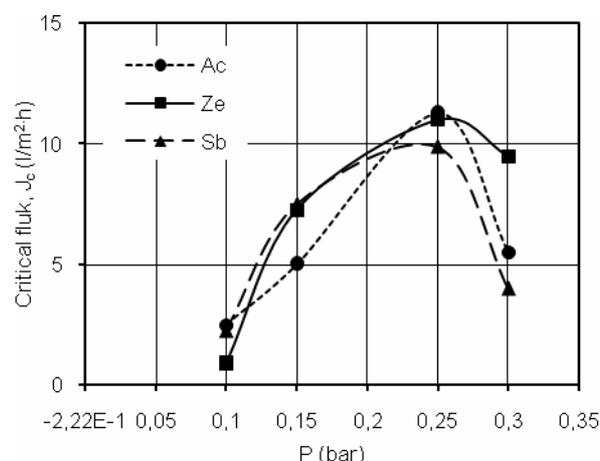


Figure 2. Effect of pressure on the critical flux for each MBR configuration

Fouling on membrane bioreactor is very complex phenomena because it involves a combination of physical, chemical, and biological aspects. Fouling can occur through three mechanisms, *i.e.*, particles deposition, adsorption (specific interaction between membrane materials and foulant), and pore blocking mechanism. The effects of fouling on membrane performance can be expressed in term of hydrodynamic resistances. At the beginning of filtration, initial flux will be dominantly influenced merely by the membrane resistance. However, with prolonged filtration time, the value of the total resistance will be changed because of the pore blocking and cake resistance formed on the membrane surface. In this experiment, the resistance-in-series model was applied to quantitatively evaluate the fouling resistance in each MBR configurations as shown in Figure 3.

Figure 3 shows series of resistances for each MBR configuration at TMP of 0.25 bars. R_t , R_{rf} , and R_{rif} respectively denote the value of total fouling resistance, reversible fouling resistance, and irreversible fouling resistance. The results clearly show that the total fouling resistance of submerged MBR is highest compared with the Ze-eMBR and the Ac-eMBR. This can be the reason for the lowest final flux obtained from submerged configuration in previous

discussion. Meanwhile, for the eMBR configuration, the use of activated carbon give lower total fouling resistance compared with zeolite.

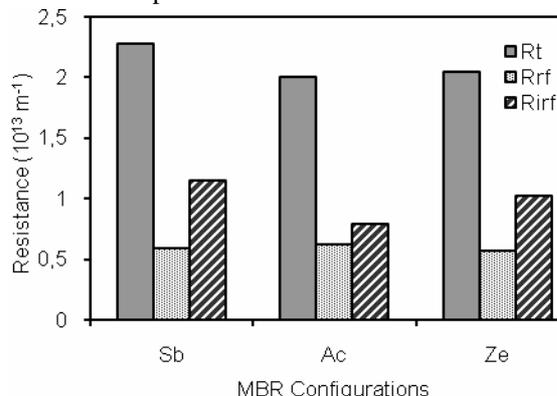


Figure 3. Series of resistance for each MBR configuration (Rt: total resistance; Rrf: reversible fouling; Rirf: irreversible fouling)

Figure 3 also demonstrates that in all configurations, the major resistance is contributed by irreversible fouling. Irreversible fouling is usually due to the adsorption of foulant on the membrane surface and the entrapment of foulant in the inner structure of the membrane. The highest irreversible fouling resistance is shown by submerged configuration. This implies that submerging the membrane directly into the mixed-liquor cause stronger tendency for the foulant to be adsorbed on the membrane and more chance for the solutes to be entrapped on the inner structure of the membrane. In the meantime, the Ac-eMBR shows lowest irreversible resistance. This may be due to the role of activated carbon bed hindering direct contact between foulant and membrane surface by the existence of scouring effect and adsorbing solutes which is responsible for the irreversible fouling.

Nevertheless, the reversible fouling resistance is also highest for Ac-eMBR. Reversible fouling is initiated by formation of cake layer on the membrane surface. From the above data, it can be seen that the eMBR configuration still allow the formation of cake layer on the membrane surface, though this can be easily removed by surface cleaning.

Figure 4 shows photograph of membrane after trial for submerged MBR (a) and Ac-eMBR (b). The eMBR one shows cleaner surface compared with the submerged one in which thick sludge deposit is observed. However, from the analysis on membrane fouling, the cake resistance of eMBR configuration was higher than the submerged one. It is assumed that in eMBR configuration, the cake layer resistances is partially attributed to the carbon active bed because, as seen on the picture, there are some activated carbon particles unevenly distributed on the membrane inter fiber. A significant scouring effect is also shown as there is no thick deposit cake layer observed on the membrane surface. Therefore, subsequent experiments were focused on Ac-eMBR configuration.



(a) submerged MBR (b) Ac-eMBR
Figure 4. Photos of membrane after trial

As can be concluded from the previous discussion, it is necessary to observe the effect of aeration and activated carbon bed thickness to further study fouling phenomena that occur in Ac-eMBR configuration. Figure 5 show relationship between flux of pure water at various TMP and aeration rate at different activated carbon bed thickness. Increased aeration rate is assumed to increase flux meanwhile increasing bed thickness increases resistance for mass transfer passing the membrane. In general, bed thickness of 8 cm and aeration rate of 9 l/min give highest flux compared with other variations at TMP between 0.2-0.3 bar. Meanwhile, for TMP less than 0.2 bar, highest flux is achieved by bed thickness/aeration rate of 8 cm/6.75 l/min and 15 cm/9 l/min, correspondingly. It can be seen that there is no clear pattern observed and the increase of flux is merely attributed to the increase of TMP. Hence, it is assumed that at a certain TMP, fouling resistance contributed by the difference thickness of activated carbon is not significant.

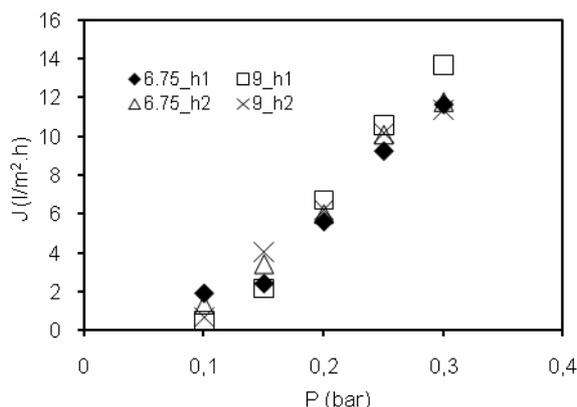


Figure 5. Profiles of stable flux of pure water at various bed thickness and aeration rate on TMP

In this experiment, the lab-scale Ac-eMBR was operated in continuous mode. The feed was continuously added to the Ac-eMBR and the permeate was continuously sucked and placed in permeate tank. In this experiment, the initial biomass concentration was ± 8.000 mg/l. Figure 6 shows relationship between flux and pressure in continuous mode. The flux and TMP are in the range of 5-15 l/m².h and 0.1-0.35 bar,

respectively. A reasonably high and relatively stable flux is obtained for more than 90 hours operation time. Kim, *et al.* (2001) found out that for MBR hybrid with completely mixed activated carbon, there is an optimum concentration of activated carbon before a cake layer is formed on membrane surface and increase the filtration resistance. Vigneswaran, *et al.* (2003) has also mentioned that the performance of membrane combined with adsorption process is influenced by reactor configuration, mode of operation, carbon dosage, adsorption, and influent characteristics.

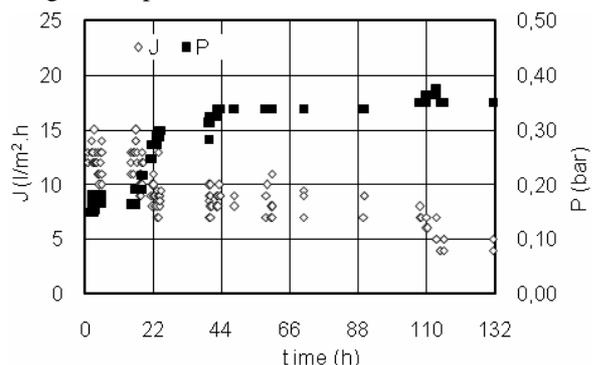


Figure 6. Relationship between flux and pressure for Ac-eMBR in continuous mode

Meanwhile, even though the resulted flux is dropped to 5 l/m².h, the rejection characteristic is excellent. The COD concentration in permeate are always below 100 mg/l at fluctuated feed concentration as shown in Figure 7. The average COD removal is approximately 98% with hydraulic retention time less than 24 hours.

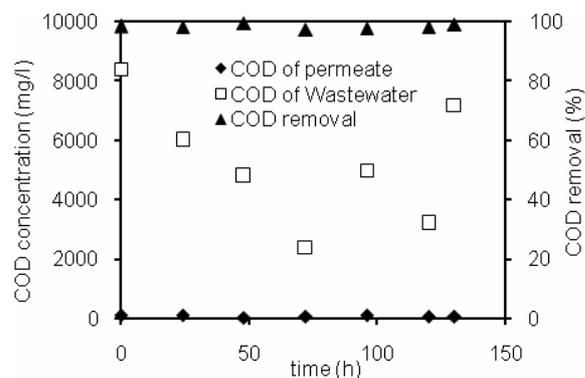


Figure 7. Rejection characteristics for Ac-eMBR in continuous mode

Pilot-plant test was also performed for more than 100 hours. During the pilot-plant test, the flux is relatively high ranging from 6-11 l/m².h, with operating pressure ranging from 0.03-0.17 bar as shown in Figure 8. It is likely that the ends-free module type and the presence of activated carbon can significantly reduce membrane fouling. The fluxes are stable and the operating pressures are only slightly increased for almost 140 hours operation time. According to Fang *et al.* (2006), the addition of

activated carbon may reduce the film resistance because of its capacity to absorb EPS as one of the main source of fouling in MBR. Guo *et al.* (2006), also has mentioned the role of activated carbon as pre-adsorption of dissolved organic substance which reduce the membrane fouling and maintain consistent permeate flux. Meanwhile, Li *et al.* (2005), stated that in long-term operation, the membrane fouling could be reduced effectively by adding PAC, and operating intervals could be extended about 1.8 times compared to the normal activated sludge system. The total resistance was also 40% lower than that of the activated sludge system. In the meantime, the COD is successfully removed in which the COD permeate were below 50 mg/l. These results demonstrated the feasibility of the Ac-eMBR at larger scale. The removal of small molecular weight compound is due to the biodegradation by bacteria grown on PAC particles and the slow formation of bio film on the membrane surface. Vigneswaran *et al.*, has shown that the addition of PAC could keep the organic removal efficiency constant without the need for chemically cleaning the membrane for a long time.

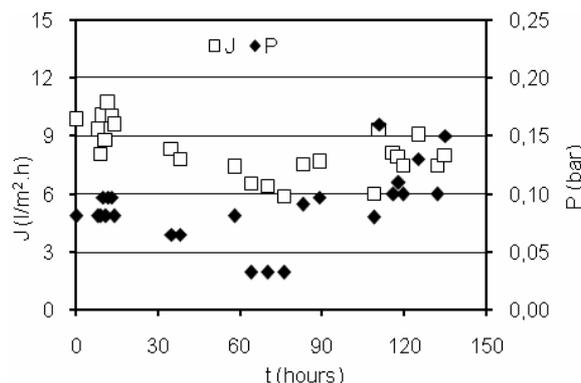


Figure 8. Pilot-scale performance of Ac-eMBR

CONCLUSIONS

A new configuration of submerged membrane bioreactor for aerobic industrial wastewater treatment has been developed. In this configuration, a bed of porous particle is applied to cover the submerged ends-free mounted ultrafiltration membrane into which a new configuration is made. Membrane performance was assessed based on flux productivity and selectivity. A reasonably high and stable flux around 11 l/m².h was achieved with COD removal efficiency of more than 99% from wastewater containing high organic matter. The fouling analysis shows that this newly configured ends-free membrane bioreactor exhibit lower irreversible resistance compared with the submerged one. The performance of pilot scale system, with 10 m² of membrane area in a 120 L tank volume, was also studied. The resulting flux from the pilot scale system was around 8 l/m².h with COD removal of more than 99%. Hence, this study has demonstrated the

feasibility of the newly configured submerged ends-free MBR at larger scale.

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