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Performance of hybrid anaerobic membrane bioreactors (AnMBRs) augmented with activated carbon in treating palm oil mill effluent (POME)

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ABSTRACT

Anaerobic membrane bioreactor (AnMBR) is an effective technology for the treatment of wastewater. However, the cost of membrane and membrane fouling has limited its application in wastewater treatment on a commercial scale. Therefore, in this study, different particle's sizes of powder activated carbon (PAC) were added into four identical AnMBRs to investigate their effects on chemical oxygen demand (COD) and natural organic matters (NOM) removals, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) values, and membrane fouling control when treating of palm oil mill effluent. It was found AnMBRs with PAC performed better than the AnMBR without PAC. It was also found that the addition of a relatively smaller size of PAC (approximately 75 μm) enhanced the COD removal efficiency to 89.45 ± 2.48, while the concentration of MLSS and MLVSS were 21,420 and 16,452 mg/L, respectively, which was high enough to result in bigger floc size, lower NOM content and better membrane fouling control. Also, to investigate the performance of polyethersulfone membrane in fouling control, different concentrations of PAC had been incorporated into it. The results showed that integrated membrane with (5 wt.%) PAC was able to achieve up to 60% COD removal rate, compared to 38.6% without PAC. Besides, there was a trend where the membrane with a higher concentration of PAC integrated has a better performance in both membrane fouling control and pollutant removal ability.

Keywords: Palm oil mill effluent; AnMBR; Activated carbon; Particles size; Membrane fouling

1. Introduction

Anaerobic membrane bioreactor (AnMBR) has advantages such as producing a lower amount of biological waste and able to convert organic substances into valuable biogas compared with aerobic membrane bioreactor [1]. Its advantages had gained the interest of many parties to use this technology to treat municipal and industrial wastewater [1–5]. However, membrane fouling is still one of the main problems facing by the AnMBR technology, which if uncontrolled will contribute to flux reduction, increase in trans-membrane pressure (TMP), resulting in high energy consumption leading to increased operational costs [6–9].

The addition of powder activated carbon (PAC) into AnMBR had shown enhancement in flux and improvement on chemical oxygen demand (COD) removal [10–12]. The improvement in the performance of AnMBR by using PAC is due to the large surface area provided by PAC

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which can act as a supporting medium for the bacteria and provide bacteria with a suitable micro-living environment [13,14]. By adding PAC into an AnMBR, biological activated carbon (BAC) was formed and it could carry out two processes simultaneously named; adsorption and biodegradation [15,16].

Incorporated additives such as (i) crystalline silicotitanate and ferrihydrite [17], (ii) sulfated TiO_2 deposited on SiO_2 nanotubes [18], and (iii) nano-silica [19] onto membrane have also shown promising results in producing higher quality flux. Although there are a very limited number of studies on treating palm oil mill effluent (POME) wastewater using the PAC incorporated with the AnMBR technology, this unique study focuses on the effects of using different particle sizes of PAC on the resulted treatment. Therefore, in this study, different sizes of PAC had been added into the anaerobic bioreactor to study their effects on biological growth, pollutants removal efficiency, and membrane fouling control. In addition, the different dosage of PAC had been incorporated into the polyethersulfone (PES) membrane to study its effects on the membrane performance in terms of fouling control and its ability in producing a higher quality of treated water.

2. Material and methods

The study of AnMBR was carried out in two stages. In the first stage, four 1L anaerobic bioreactors, with and without PAC, namely R1, R2, R3, and R4 were set up and their performance was evaluated based on the removal efficiencies for COD, protein and polysaccharide and their membrane fouling control. In the second stage, the fabrication of a hybrid PES membrane integrated with PAC was carried out and tested for its treatment performance.

2.1. Materials

Granular activated carbon used in this study was grounded by using a conventional Panasonic food blender, (Japan) to grind. The particle size distribution is shown in Fig. 1. The D_{50} for each one of the three sizes from PAC in terms of volume and number is 265.80 ± 1.29 µm and 3.47 ± 0.66 µm (coarse), 152.69 ± 1.63 µm and 2.15 ± 0.46 µm (medium), 75.72 ± 1.52 µm and 2.19 ± 0.48 µm (fine) respectively.

The powdered activated carbon (PAC) used to incorporate into the PES membrane is supplied by GeneChem Company, (Canada). The anaerobic sludge and POME were

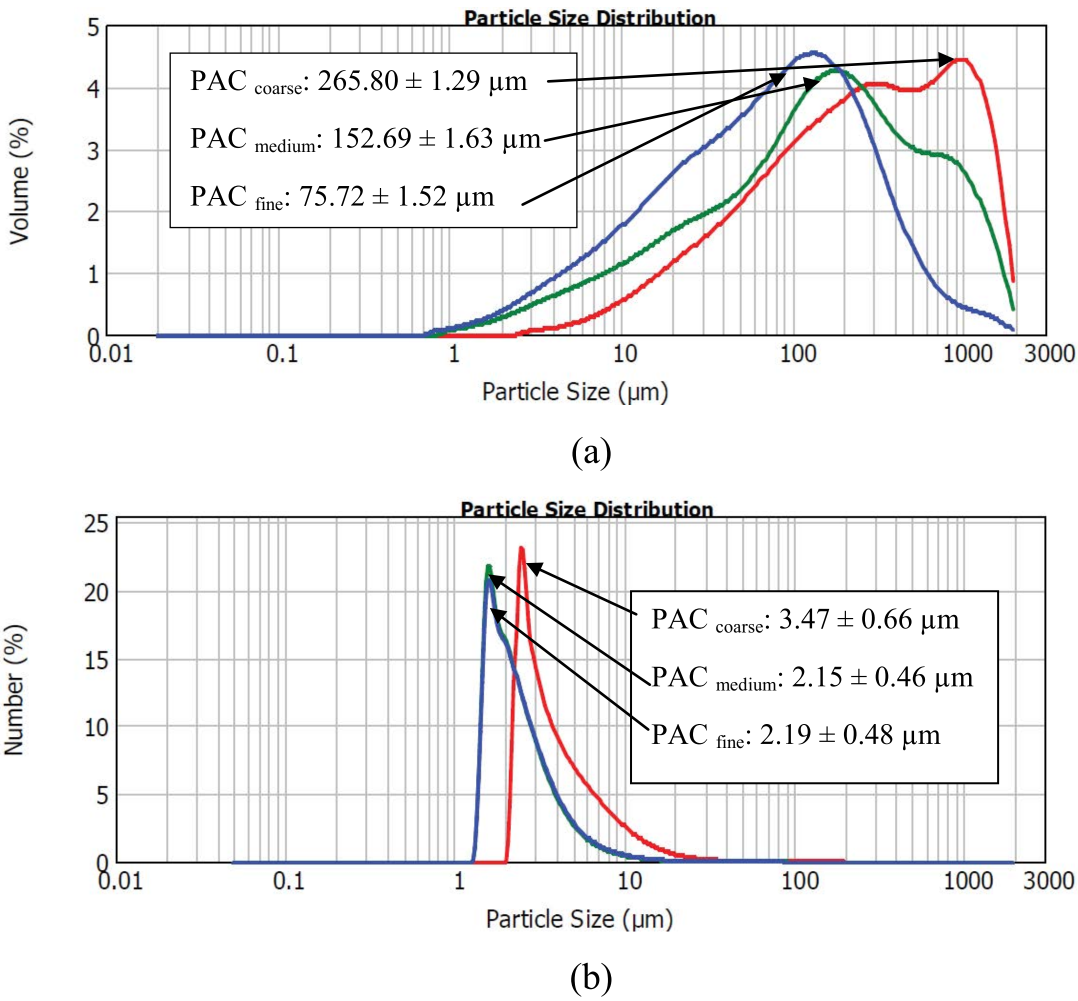

Fig. 1. (a) Particle size distribution of PAC in terms of volume and (b) number.

obtained from a POME treatment plant owned by Tian Siang Group and located in Perak, Malaysia. Tables 1 and 2 present the general characteristics of both the anaerobic sludge and POME, respectively. The dope used to cast the membrane consists of 1-methyl-2-pyrrolidone (NMP) supplied by Friedemann Schmidt Chemical (USA), and PES and both were mixed and heated at the temperature between 60°C to 70°C.

2.2. Operation of AnMBRs

Four 1 L AnMBRs kept in a water bath with a temperature of 45°C namely R1 (without PAC), R2 (with average PAC size of 265.80 μm), R3 (with average PAC size of 152.69 μm), and R4 (with average PAC size of 75.18 μm). All the bioreactors were added with 5 g/L of PAC except for R1. The sludge retention time and hydraulic retention time of the four AnMBRs were fixed at 30 and 12.5 d, respectively.

2.3. Fabrication of hybrid PES membrane

Dope with different ratio of PAC was prepared as shown in Table 3, the ratios PAC and PES were selected based on the recommendation by literature [21–23]. The dope was prepared by mixing NMP with PES with a ratio of 87:13 by weight (g) [23], under the temperature range from 60°C to 70°C. After that PAC has been added into the dope based on the percentage required and left in a sonicator bath for 8 h. The membrane was fabricated using the dry-wet phase technique with a membrane thickness of 15 μm. The fabricated membranes were left in a water bath for 24 h and immersed in methanol for 8 h for post-treatment purposes.

2.4. Analytical methods

Mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), and COD were analyzed by following the procedures from Standard Method. Polysaccharide concentrations were measured with the methods of phenol-sulfuric acid [24] and concentration of protein was measured by using Bradford reagent (England) with bovine serum albumin as standard [25]. The pH of the supernatant from AnMBRs was determined by using a pH meter (Hanna HI 2550, USA). Particle size distribution was determined by using the particle size analyzer (Malvern Mastersizer 2000, UK). The total biogas production in the AnMBRs was collected and measured by the water displacement method.

Table 1
Characteristics of palm oil mill effluent sludge

Parameters	Unit	Value
pH	–	7.40–7.44
Temperature	°C	26.1
Chemical oxygen demand, (COD)	mg/L	10,870–13,600
Ammoniacal nitrogen, (NH_3–N)	mg/L	161–190
Mixed liquor suspended solids, (MLSS)	mg/L	9,500–12,500
Mixed liquor volatile suspended solids, (MLVSS)	mg/L	8,350–9,700
MLVSS/MLSS	–	0.78–0.88
Oxidation reduction potential	mV	–30.5
Total suspended solids	mg/L	2,767–3,800
Color	Pt-Co	973
Turbidity	NTU	5,783–6,606
Electrical conductivity	mS	6.91

Table 2
Characteristics of raw POME

Parameters	Mean value	Range	Discharge limit [20]
pH*	4.2	3.4–5.2	5.0–9.0
Biochemical oxygen demand	25,000	10,250–43,750	100
Chemical oxygen demand, (COD)	51,000	15,000–100,000	400
Total solids	40,000	11,500–79,000	–
Suspended solids	18,000	5,000–54,000	400
Total volatile solids	34,000	9,000–72,000	–
Oil and grease	6,000	130–18,000	50
Ammoniacal nitrogen, (NH_3–N)	35	4–80	–
Total nitrogen	750	180–1,400	150

*Units in mg/L except pH

Table 3
Ingredients used in membrane dope preparation for polymer and hybrid membranes fabrication

Samples	PES (g)	NMP (g)	PAC (g)
0 wt.% PAC	13.00	87.00	0.00
1 wt.% PAC	12.87	87.00	0.13
5 wt.% PAC	12.35	87.00	0.65

3. Results and discussion

3.1. Treatment performance of AnMBRs

The performance of the four AnMBRs is shown in Table 4. Throughout the study, the pH of all four AnMBRs was maintained at the range of 7.8 to 7.9 and no noticeable fluctuation of pH was observed. The stability in pH indicated that there is a balance in both acidogenic and methanogenic activities in the bioreactors. The four AnMBRs, COD removal efficiency for R1 is the lowest compared to other AnMBRs which is only 54.09% ± 15.24%, Other AnMBRs have relatively higher COD removal efficiency. R4 shows the best results among all the AnMBRs with COD removal efficiency could reach 89%.

The addition of PAC would serve as a shelter for bacteria, with smaller PAC size it would adsorb more COD due to higher surface area and more COD would be biodegraded by the bacteria, resulting in higher COD removal efficiencies [26]. It can be clearly seen that relatively smaller particle size could remove more COD due to its higher surface area. However, as presented in Table 4, the AnMBR with larger PAC size (R2) has the highest amount of yielded biogas which was (148 ± 8 mL/h) than others with smaller PAC sizes or without PAC which gave the lowest which is supported by the conclusions of Xu et al. [27]. The polysaccharide can contribute to a higher tendency for fouling as it has a nature of large-size with gelling properties. The formation of thin impermeable gels on the membrane surface can significantly increase the filtration resistance [28]. The presence of natural organic matters (NOM) such as protein and polysaccharide in POME was the primary reason for membrane fouling [22,29]. As shown in Table 4, the removal rate of protein and polysaccharide increases with a decrease in PAC sizes. Identical to the COD removal rate, the smaller particle size of PAC provides a larger surface area which could adsorb more protein and polysaccharide [6,27].

3.2. Comparison of biomass concentration among AnMBRs

The effects of different sizes of PAC on the growth of MLVSS and MLSS of the AnMBRs were studied as shown in Table 5. The highest bacteria growth is in R4 with a concentration of 21,420 mg/L. AnMBRs with no PAC and with bigger PAC size show rather lower value in MLSS (13,004, 13,362 and 13,433 mg/L respectively). MLVSS indicates the volatile suspended solid which is more representative in terms of bacteria counts. The MLSS and MLVSS increased with the decrease in particle size because smaller particle size contributes to a larger surface area which promotes the growth of the attached bacteria [23]. Furthermore, the increase of the bacteria population would further enhance the removal rate of COD and NOM in POME, resulting in better supernatant quality [22,23,29].

3.3. Effect of protein and polysaccharide towards membrane fouling

Polymer membrane was used to carry out the filtration performance of the AnMBR. Fig. 2 shows the filtration performance of the AnMBR presented by the TMP values. It can be observed that R4 with the lowest concentration of fine pollutants (COD and protein); values have been presented in Table 4, had the best filtration result followed by R3 and R2. The worst performer is R1 which has no PAC inside its bioreactor, which reaffirms that the addition of PAC improves the AnMBR antifouling.

Table 4
Performance of AnMBRs augmented with different particle sizes of PAC

Parameter	R1	R2	R3	R4
Temperature, (°C)	45	45	45	45
pH	7.8 ± 0.1	7.8 ± 0.1	7.8 ± 0.1	7.8 ± 0.1
PAC dosage, (g/L)	NA	5	5	5
PAC particle size, (D_{50} volume)	NA	265.80 ± 1.29	152.69 ± 1.63	75.72 ± 1.52
PAC particle size, (D_{50} number)	NA	3.47 ± 0.66	2.15 ± 0.46	2.19 ± 0.48
Sludge retention time, (day)	30	30	30	30
Feed in COD, (mg/L)	16,449 ± 7,840	16,449 ± 7,840	16,449 ± 7,840	16,449 ± 7,840
Feed in protein, (mg/L)	5,599 ± 3,511	5,599 ± 3,511	5,599 ± 3,511	5,599 ± 3,511
Feed in polysaccharide, (mg/L)	139 ± 2	139 ± 2	139 ± 2	139 ± 2
Supernatant COD (mg/L)	7,551 ± 363	2,037 ± 270	1,918 ± 320	1,736 ± 302
COD removal efficiency in supernatant, (%)	54.09 ± 15.24	87.62 ± 3.30	88.34 ± 2.80	89.45 ± 2.48
Protein concentration, (mg/L)	1,875 ± 551	1,475 ± 615	1,315 ± 592	1,133 ± 552
Polysaccharide concentration, (mg/L)	85 ± 2	75 ± 3	74 ± 3	75 ± 3
Biogas yield, (mL/h)	129 ± 7	148 ± 8	140 ± 7	137 ± 7

Table 5
Comparison of MLSS and MLVSS concentrations in various AnMBRs with different PAC sizes

Parameter	R1	R2	R3	R4
MLSS, (mg/L)	13,004 ± 3,358	13,362 ± 1,192	13,433 ± 1,672	21,420 ± 2,604
MLVSS, (mg/L)	9,981 ± 2,455	10,741 ± 1,192	10,612 ± 1,672	16,452 ± 2,604

The presence of NOM plays a significant role in membrane fouling control [28,30]. As the amount of NOM increases, it is more likely that a membrane would be fouled more easily.

3.4. Effect of floc size towards membrane fouling

In addition to NOM, the floc size also plays an important role in fouling control. As shown in Fig. 3, the particle size distribution for activated sludge in R1 was smaller compared to other AnMBRs with PAC. These results show that by adding PAC into activated sludge, bigger particle size was produced. This indicates that PAC could act as an adsorbent which would attract bacteria to attach on its surface to transform the PAC to become BAC. As the size of PAC decreases, the floc size distribution shows higher value, and this phenomenon may be caused by the PAC with smaller size are more porous compared to PAC with a relatively larger size which results in forming larger floc more effectively [31].

Research shows that larger floc size would produce greater porosity and permeability filter cake which could reduce the TMP [31]. As shown in Figs. 2 and 3, floc size in R4 is 62.185 ± 1.45 μm and it had the best membrane fouling control performance which had indicated the filter cake formed was more porous compared to other AnMBRs referring to its longer time (min) required to reach the maximum TMP (1 psi). R1 performed worst and it has the smallest floc size among the AnMBRs.

3.5. Performance of the hybrid membranes incorporated with the different concentrations of PAC

In addition to the study about the performance of different AnMBRs added with different sizes of PAC, the performance of the hybrid membranes incorporated with different PAC content of 0% wt., 1% wt., 5% wt., was also carried out. As shown in Fig. 4, the performance of the hybrid membrane for the AnMBR (R4) increases as the PAC content increases, by incorporated 1% wt. of PAC into the polymer membrane would only show little improvement of membrane fouling control. However, when the concentration of PAC increased from 1 to 5%wt, the hybrid membrane had much better membrane fouling control as per Fig. 4. A dead-end filtration test was also being carried out to verify the above cross-flow filtration results and the

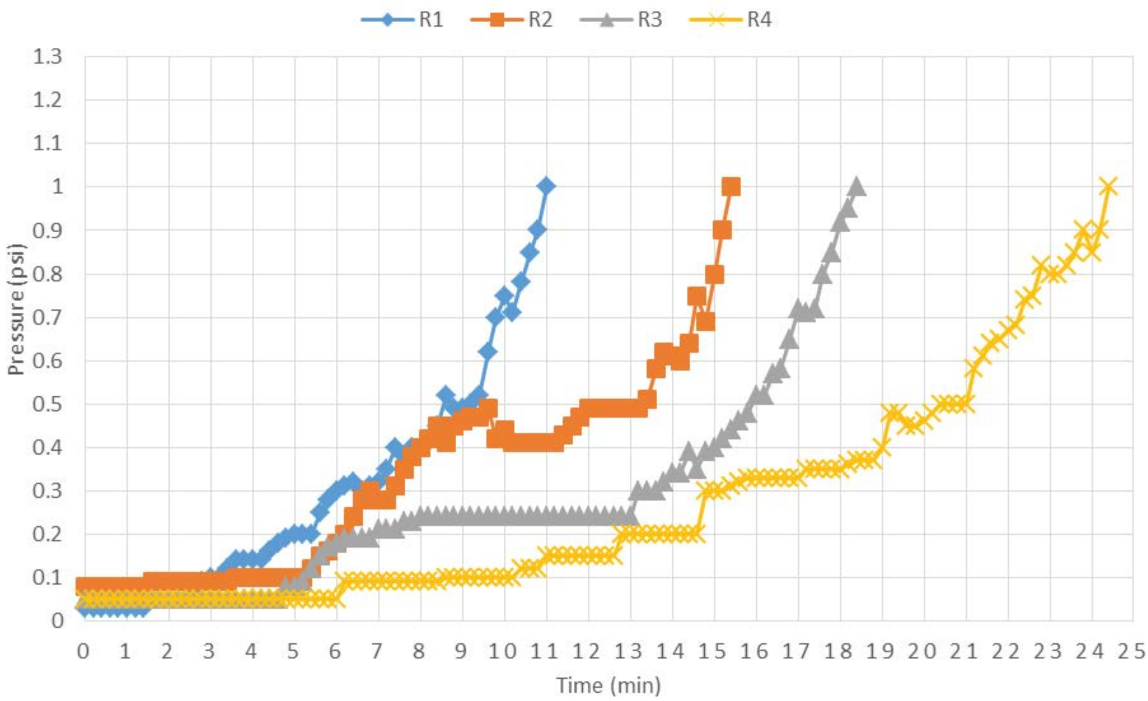

Fig. 2. Performance of membrane fouling control of different anaerobic membrane bioreactors added with different sizes of powdered activated carbon presented by the transmembrane pressure.

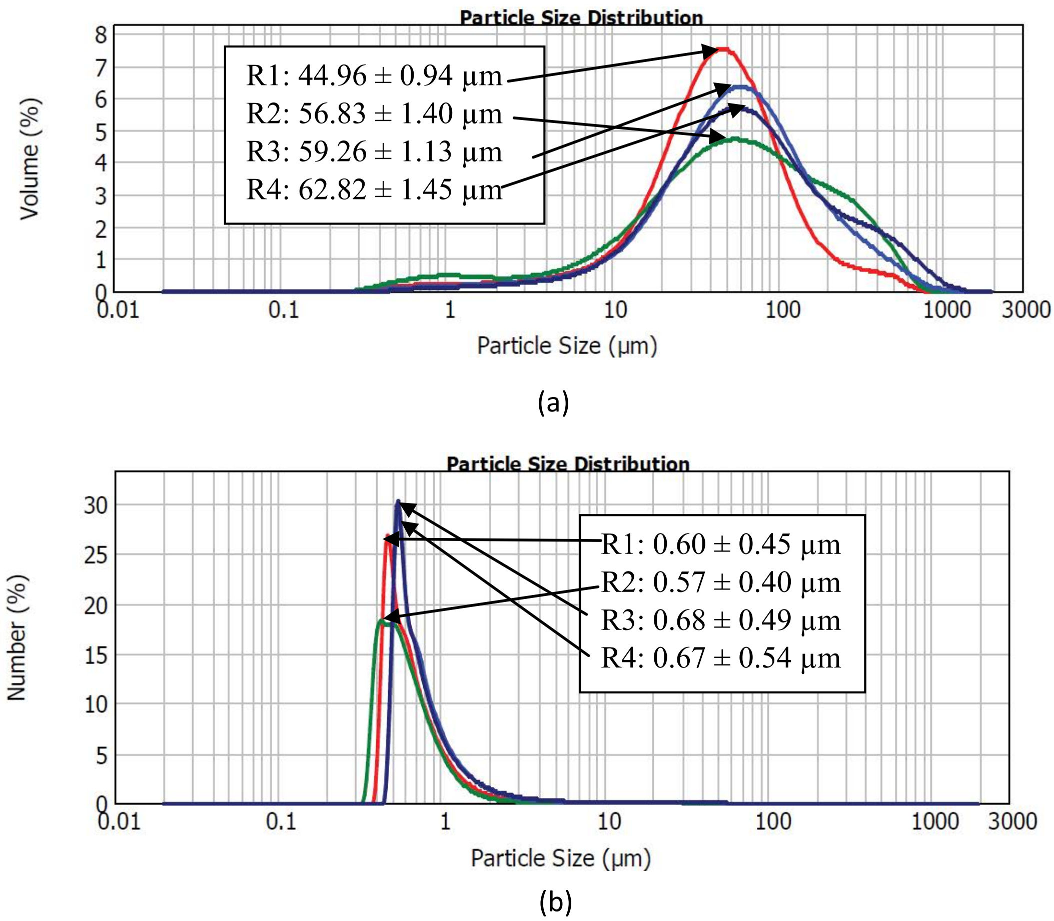

Fig. 3. (a) Microbial flocs size distribution of the different anaerobic membrane bioreactors added with different sizes of PAC in terms of volume and (b) number.

same trend was observed. As shown in Table 6, the volume of permeate collected increases as the PAC content in hybrid membrane increases. This indicates that incorporation of PAC into the polymer membrane help to increase the performance of the membrane fouling control and produce better permeate quality as per Table 6. However, it was noticed that all the membrane and hybrid membranes used for R4 had a good performance in pollutant removal as they could remove COD from the POME more than 90% as per Tables 4 and 6.

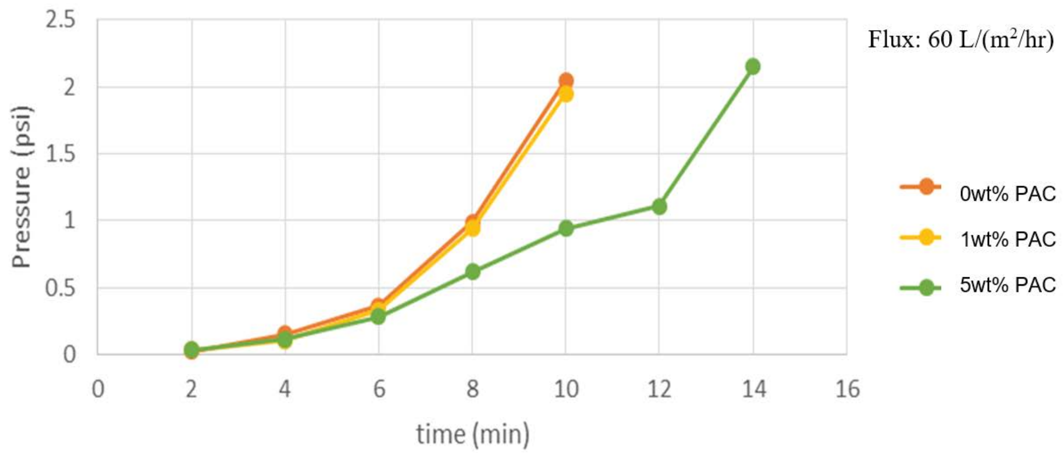

Fig. 4. Membrane fouling control performance of the different hybrid membranes incorporated with different PAC concentrations represented by the transmembrane pressure.

Table 6
Performance of different hybrid membranes incorporated with different concentrations of PAC in treating supernatant (COD = 1,736 ± 302) from R4

Parameter	0 wt.% PAC	1 wt.% PAC	5 wt.% PAC
Volume, (mL/h)	10 ± 5	10 ± 6	21 ± 5
Permeate COD, (g/L)	1,066 ± 33	740 ± 66	696 ± 274
COD removal efficiency, (%)	≈38.6	≈57.4	≈60

4. Conclusion

The addition of PAC in an AnMBR was proven to be effective in terms of COD and NOM removal. It was found that simultaneous processes of adsorption and biodegradation are the main elements that helped to enhance the performance of AnMBR added with PAC. AnMBR with relatively smaller PAC size (75.18 μm) could perform better compared to the AnMBR with relatively bigger PAC sizes or without PAC.

In addition, the reduction of NOM for the AnMBR added PAC also contributed to the better membrane fouling control. Bigger floc size was noticed for the AnMBR using smaller PAC size which helped to produce a more permeate filter cake and resulting in having better membrane fouling control.

It was found that adding PAC into the PES membrane to produce a hybrid membrane could improve the fouling resistance and enhanced fine pollutant removal rates. PES hybrid membrane performs best by having 5% wt. of PAC incorporated into it. PAC could act as an adsorbent for contaminants, preventing contaminants from direct contact with the membrane surface. Its effect would be better after being transformed into BAC which is equipped with the ability to do simultaneous adsorption and biodegradation processes.

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