Hybrid powdered activated carbon-activated sludge biofilm formation to mitigate biofouling in dynamic membrane bioreactor for wastewater treatment

Mohammad Reza Mehrnia, Fatemeh Nasiri, Fatemeh Pourasgharian Roudsari and Fatemeh Bahrami

School of Chemical Engineering, College of Engineering, University of Tehran, Tehran, Iran

ABSTRACT

Membrane costs and biofouling limit applications of membrane bioreactors (MBRs) for wastewater treatment. Here, powdered activated carbon (PAC) utilization in the formation and performance of a self-forming dynamic membrane consisting of activated sludge and PAC during hybrid wastewater treatment process was studied. Short-term agitation helped (non)biological particles to quickly uniformly settle on mesh filter, forming more uniform PAC-containing dynamic membranes (PAC-DMs). PAC adsorbed adhesive materials, resulting in an increase in average floc size and DM permeability while decreasing biofouling. The most efficient PAC concentration was 4 g L\(^{-1}\) considering techno-economics, i.e. the highest effluent quality (turbidity of 19.89 NTU) and the lowest biofouling (transmembrane pressure rise of 2.89 mbar). Short-term performance of hybrid PAC-DM bioreactor (PAC-DMBR) showed stability in effluent quality improvement including 92%, 95%, 83%, 84% and 98% reductions in turbidity, chemical oxygen demand, total dissolved solids, total nitrogen, and total phosphorous, respectively. Accordingly, adopting hybrid PAC-DMBR has potential to alleviate biofouling and capital cost.

GRAPHICAL ABSTRACT

ARTICLE HISTORY

Received 6 January 2022
Accepted 20 May 2022

KEYWORDS

Hybrid membrane bioreactor; self-forming dynamic membrane; powdered activated carbon; wastewater treatment; biofouling

Introduction

Global insufficiency of water resources besides the widespread pollution of existing water bodies has put the process of wastewater treatment and reuse among the most substantial issues in the environmental engineering area. As a modern paradigm shift, wastewater is now considered a renewable resource which is capable of producing clean water, useful nutrients and renewable energy (Aslam et al. 2022). Different wastewater treatment approaches have been proposed, including physical (Prathapar et al. 2006) and chemical (Pidou et al. 2008) techniques, advanced oxidation process (AOP) (Liu et al. 2021) and biological methods (Holloway et al. 2021). Among them, biological methods, independently or in combination with other techniques (Xiang et al. 2021), stand out because of being cost efficient and lack production of more dangerous intermediates (Ayed et al. 2017). Moreover, the hybrid techniques can overcome the
Combining solid-liquid separation and biological treatment (Lee et al. 2020), membrane bioreactors (MBRs) have been extensively adopted for municipal and industrial wastewater treatment over the last decades (Sokhandan et al. 2020). Some advantages of MBRs over conventional activated sludge processes including high concentrations of mixed liquor suspended solids (MLSS), high quality of permeate and substantially small footprint have made them broadly applicable in wastewater treatment (Ko 2018; Etemadi and Yegani 2019). Despite MBR advantages, fouling and the high cost of membranes are the main obstacles to the widespread use of MBRs in the industry (Liébana et al. 2015; Sari Erkan et al. 2020). The biofilm on the surface of a membrane reduces the operational flux or increases transmembrane pressure (TMP) depending on the operation mode (Mafirad et al. 2011). On the other hand, biofouling rejects finer particles such as viruses and inorganic ions (Najmi et al. 2020). Consequently, many scientific endeavours have been focusing on minimization of the fouling affecting long-term filtration of micro/ultra-polymeric membranes.

The utilization of a dynamic membrane bioreactor (DMBR) could be an effective approach to settling this bottleneck (Vergine et al. 2021). Among dynamic membrane (DM) types (i.e. self-forming dynamic membrane (SFDM) and pre-coated DM), SFDMs (also called biofilm (Mohan and Nagalakshmi 2020)) are formed gradually by deposition and adhesion of substances present in the wastewater, such as suspended solids (SS), colloids, microorganisms and their secreted extracellular polymeric substances (EPS) on the lying support media (Vergine et al. 2018). Since SFDM is a subset of DM, the latter is frequently used instead of the former. Therefore, DM is used in the following sections. Usually, cheaper materials, woven and non-woven fabric filters, such as polyester fabric filters, acrylate, nylons and mesh filters are utilized as the support of DM (Loderer et al. 2013; Hu et al. 2016). In a previous study (Rezvani et al. 2014), the process of DM formation on the mesh filter and its capability in treating municipal wastewater was investigated. It was found that the formation of DM on the surface of a mesh filter instead of using conventional polymeric membranes was a promising new approach which was able to enhance effluent quality and reduce costs simultaneously (Rezvani et al. 2014).

Some other researchers used chemical adsorbents to reduce fouling in MBR systems. As an example, Yuniarto et al. (2013) used three biofouling reducers (BFRs), powdered activated carbon (PAC), granular activated carbon (GAC), and fine-grained zeolite in an MBR and demonstrated their significant role in biofouling reduction. Damayanti et al. (2011) investigated the effectiveness of three BFRs such as PAC, zeolite and Moringa oleifera in MBR. They reported that PAC, at its optimum dosage of 8 g L⁻¹, provided above 70% reduction in fouling rates during the short-term filtration. Fang et al. (2006) concluded that virgin activated carbon reduced the film filtration resistance by 22%, whereas the reduction was only 14% by the activated carbon pre-sorbed with EPS and negligible by the inert diatomaceous earth. Yang et al. (2019) studied the impacts of PAC and GAC on treatment performance, mixed liquor and cake layer properties, as well as membrane fouling behaviors in anaerobic MBRs. In the past decade, several researchers have utilized PAC as an appropriate adsorbent for reducing the fouling and improving the effluent quality in MBR (Kulesha et al. 2018; Asif et al. 2020; Jiang et al. 2020; Kozak et al. 2021).

Few recent studies have coupled PAC addition and DM to enhance MBR performance. Some of them utilized time consuming formations of DMs by PAC pre-coating procedures (Chu et al. 2014; Poostchi et al. 2015) while others used aeration for sludge suspension during the formation stage of the DMs (Hu et al. 2017; Yu et al. 2019). However, in this study, agitation was introduced as a more suitable method for the quick-uniform formation of the PAC-containing DM. The idea of applying an agitator for DM formation was first proposed by Rezvani et al. (2014), and then examined by Poostchi et al. (2015). Rezvani et al. (2014) employed agitation as an appropriate method for uniform DM formation. Poostchi et al. (2015) utilized agitation for the formation of a pre-coated DM containing PAC. Nevertheless, very limited information can be gained from the literature stating that agitation enhanced the formation of PAC-DM and hybrid operation of PAC-DMBR.

In the present study, a mechanical agitator was used to provide just-complete suspension of the mixture of PAC and activated sludge throughout the volume of the bioreactor, named as formation stage (agitation stage) when the DM was formed. A low-cost monofilament mesh filter was used as a supporting medium to entrap the PAC-activated sludge mixture, forming a cake layer containing sludge flocs associated with PAC particles. At the formation stage, the effect of different concentrations of PAC on TMP and effluent quality, i.e. turbidity and chemical
oxygen demand (COD), were evaluated to determine the least fouling. After finding a locally optimum dosage of PAC, the performance of hybrid PAC-DMBR in wastewater treatment was evaluated in terms of TMP and effluent quality including turbidity, COD, total dissolved solids (TDS), total nitrogen (TN), and total phosphorous (TP) in short-term performance.

Materials and methods

Experimental set-up and operation conditions

As depicted in Figure S1 (Supplementary material), the experimental set-up used in this study is similar to Poostchi et al. (2015). In brief, the operating volume of the bioreactor was 7.5 L. A monofilament polyester mesh filter (average pore size of 39 µm and effective area of 156 cm²) was used to support SFDM. The mesh filter was properly fixed in the holder module and then submerged vertically in the reactor. A mechanical agitator was installed at a distance of 3 cm below the filter module. This agitator was used to provide an appropriate axial flow pattern and uniform deposition of activated sludge and PAC particles on the mesh filter. The in-situ visual observation showed that the rotational speed of 300 rpm of the axial-flow agitator was sufficient to perform just-complete suspension of the mixture of PAC particles and sludge flocs. Furthermore, to provide the required aeration for activated sludge during the aeration stage, five spargers were set on the bottom of the bioreactor. Regulation and control of influent and effluent, aeration rate and pressure were carried out similar to Poostchi et al. (2015).

The bioreactor was fed with synthetic wastewater containing 937.5 mg L⁻¹ of glucose, 193.13 mg L⁻¹ of ammonium sulfate and 42.58 mg L⁻¹ of ammonium phosphate with COD: N: P ratio of 100: 5: 1. All the aforementioned substances were purchased from Merck. The operating conditions are presented in Table 1. Also, PAC was supplied by the Research Institute of Petroleum Industry with characteristics presented in Table 2. Seed activated sludge was taken from a municipal wastewater treatment MBR plant and adapted with the feed for 105 days.

PAC-DM formation

Initially, the activated sludge was loaded into the MBR with an MLSS of 8 g L⁻¹, and the PAC particles were poured into the vessel to reach the final concentrations of 2, 4 and 5 g L⁻¹. To determine the PAC concentration, preliminary DM formation studies were conducted. While running experiments with a PAC concentration of 6 mg L⁻¹, some of the PAC particles were settled in the bottom of the bioreactor. Indeed, an agitator speed of 300 rpm was not enough to suspend all PAC particles at a concentration of 6 mg L⁻¹. To maintain the same operational conditions (i.e. mixing power), maximum PAC concentration which was completely suspended was selected. Therefore, the highest PAC concentration was chosen as 5 mg L⁻¹, which was well suspended at the applied mixing power. Afterwards, the mesh filter module was swamped in the middle of the bioreactor and it directly contacted the mixed liquor. DM formation commenced when the peristaltic pump started to suck the effluent out of the filter media. During the DM formation stage, the MBR effluent was recycled back to the bioreactor to keep the volume constant. In addition, permeate flux was adjusted to 150 LMH as reported by Rezvani et al. (2014).

Critical flux measurement

After DM formation, the axial-flow agitator was switched off. At this stage (aeration stage), critical flux was measured by stepping up the flux and monitoring the corresponding TMP according to Rezaei and Mehrnia (2014). Each run was started at a constant flux which was maintained for a step length of 20 min. When the TMP remained constant at the imposed flux, the flux was stepped up to an upper level by changing the revolution speed of the suction pump. The initial constant flux was 35 LMH and the step height of the imposed flux was set to 60 LMH. The procedure was repetitively performed till an obvious increase in TMP was observed.

Hybrid PAC-DM short-term performance

To investigate the performance of the formed hybrid PAC-DM and the stability of the bioreactor, short-
term wastewater treatment was carried out for 12 days. To start the short-term performance of the hybrid PAC-DMBR, the permeate flux was set to 60 LMH (lower than the measured critical flux). Aeration applied to the membrane was denoted as SADm (specific aeration demand in the volumetric rate of air per unit membrane area). In this study, SADm was adjusted to 5.625 m³m⁻²h⁻¹ to maintain the stability of the DM and supply sufficient oxygen as reported in an earlier study (Rezvani et al. 2014). To examine the performance of the hybrid PAC-DMBR, three different concentrations of PAC particles were used utilizing the same reactor in the 12-days operations so that each new experiment with a different PAC concentration was started in the reactor when the previous experiment had been completed.

**Analytical procedures**

To evaluate effluent quality, filtrate flow turbidity was determined at 750 nm (Sabaghian et al. 2018) using a spectrophotometer (Spectroquant Multy, Merck, Germany). In addition, COD, TN and TP concentrations, besides EPS and SMP values were analyzed using Merck reagents according to APHA standard methods (APHA 2005). TDS was evaluated with a TDS-meter (HM digital, America). Furthermore, to maintain constant operational conditions, dissolved oxygen (DO) and temperature were monitored continuously using a multi-meter (WTW 340 i, Germany). To minimize the experimental errors, each experiment was performed twice and the averages of the results were considered.

**Membrane morphology**

A scanning electron microscopy (S4160, Hitachi, Japan) was utilized to study cross-section structures of cake layers formed on the mesh filter. When preparing the DM samples, the high-pressure difference between the filter module and suction line can result in the backflow. Accordingly, to prevent the backflow, the suction pump was stopped and the permeate valve was normally closed at each sampling. The DM and its supporting filter were separated from the holding frame. The sample was fixed with 2% (v/v) glutaraldehyde in a 0.1 M phosphate buffer at pH 7.2 for 2 h and then washed for 10 min. It was again immersed for 1 h in the 0.1 M phosphate buffer and then dehydrated with ethanol. When drying was accomplished, the sample of DM was precisely obtained and accurately fixed to achieve appropriate scanning electron microscopy (SEM) images.

**Particle size analysis**

Particle size distribution (PSD) was analyzed by evaluating the settling velocity of particles and propagation of visible waves during the collision with flocs using a Shimadzu laser diffraction particle size analyzer (Shimadzu Co., Ltd, Kyoto, Japan). A mixed liquor suspension with a concentration of 0.8 g L⁻¹, recognizable to the analyzing device, was poured into a sampling container and placed in the device. After adjusting the device to settling mode, PSD and average particle diameter were obtained.

**Filtration resistance analysis**

To describe the filter fouling phenomena, the resistance-in-series model was employed (Mororó et al. 2018). This theoretical model uses:

\[ R_t = \frac{\Delta P}{\mu J} \]  
\[ R_t = R_m + R_c + R_f \]

where \( J \), \( \Delta P \), \( \mu \), \( R_m \), \( R_c \) and \( R_f \) represent flux (m³m⁻²s⁻¹), trans-filter pressure (Pa), permeate viscosity (Pa s⁻¹), total filtration resistance (m⁻¹), filter resistance (m⁻¹), cake resistance (m⁻¹), and fouling resistance (m⁻¹) and gel layer resistance (m⁻¹), respectively (Zhou et al. 2014; Poostchi et al. 2015). The above-mentioned resistances are calculated using the following equations:

\[ R_m = \frac{\Delta P_w}{\mu J} \]  
\[ R_f = \frac{\Delta P_{wf}}{\mu J} - R_m \]  
\[ R_c = \frac{\Delta P_{AS}}{\mu J} - R_m - R_f \]

where \( \Delta P_w \) is the initial value of TMP, \( \Delta P_{wf} \) is the final value of TMP after removing the cake layer by flushing with tap water, and \( \Delta P_{AS} \) is the trans-filter pressure of the activated sludge at the steady-state (Lee et al. 2001).

**Statistical analysis**

All experiments were performed at least twice. Results are expressed as mean ± standard deviation. Analysis of Variance (ANOVA) with a 95% confidence interval was used for statistical analysis of turbidity and COD.
results. Tukey multiple comparison tests with a 95% confidence interval were performed to validate the statistically significant difference between means.

Results and discussion

Effect of PAC on DM formation

The DM formation process and final characteristics were evaluated to investigate the effect of PAC concentration and mechanical agitation. The formation process was completed when no significant change in turbidity was observed (Rezvani et al. 2014). Figure 1a depicts a variation of turbidity with time for three different concentrations of PAC particles and a control DMBR system without PAC addition. As reported in Table 3, an increase in the concentration of PAC particles (from 0 to 2, 4 and 5 g L\(^{-1}\)) resulted in an increase in the time required for DM formation (from 20 min to 35, 47 and 55 min, respectively).

In addition, Figure 1a shows that as the concentration of PAC particles increased, a slower effluent turbidity change (slope) was obtained. Moreover, the EPS and SMP obtained for the mixed liquor in each DMBR showed that an increase in the PAC concentration led to considerable reductions in EPS and SMP after one hour of agitation (Table 3). It is

Table 3. A summary of the effect of PAC concentrations on the formation and characteristics of the formed DMs and the mixed liquor.

| PAC concentration (g L\(^{-1}\)) | Without PAC | 2 | 4 | 5 |
|----------------------------------|-------------|---|---|---|
| DM Formation Time (min)          | 20          | 35 | 47 | 55 |
| DM Thickness (μm)                | 258         | 220 | 198 | 192 |
| DM Compressibility Factor (kg SS m\(^{-3}\)) | 704.94 | 414.77 | 404.04 | 387.36 |
| EPS (mg L\(^{-1}\)) in the mixed liquor | 250.66 | 199.37 | 178.84 | 173.61 |
| SMP (mg L\(^{-1}\)) in the mixed liquor | 159.73 | 121.08 | 105.07 | 100.53 |

Figure 1. Effect of PAC concentration on the formation of the DMs in terms of (a) effluent turbidity, (b) DM cross-sectional thickness (SEM images), (c) effluent COD and (d) TMP changes.
known that PAC particles adsorb adhesive materials which act like glue for attaching particles (Lee et al. 2010). Thus, reducing the concentration of such substances in the mixed liquor. Iorhemen et al. (2016) and Kulesha et al. (2018) showed that the chemical-based reduction of adhesive materials results in slower biofilm formation. Therefore, it is deduced that an increase in PAC concentration led to reluctant adhesion of organic particles on the mesh filter resulting in slower DM formation.

PSD analysis revealed that at the beginning of the formation process, the mean sizes of sludge flocs and PAC particles were 43 \( \mu \text{m} \) and 50 \( \mu \text{m} \), respectively. Gradually, sludge flocs were attached to the surface of PAC particles within a matrix of adhesive substances. As a result, larger and more uniform flocs with an average size of 90.3 \( \mu \text{m} \) were formed. Yang et al. (2010) also reported sludge flocs enlargement in the presence of PAC particles. As seen in Figure 1a, during DM formation, the presence of PAC particles caused the turbidity to increase compared to the control DMBR system (without PAC). Considering the increase in the mean size of sludge particles in the presence of PAC, it is implied that larger flocs were deposited on the mesh filter and led to the formation of more permeable PAC-DMs. In general, the more permeable DM, the higher effluent turbidity due to the passage of more particles through the biofilm. Deposition of the enlarged flocs on the filter led to the formation of larger pores in the structure of the biofilms (DMs) which is visible in the field emission scanning electron microscopy (FESEM) images of the DMs for different concentrations of PAC (Figure 1b).

Moreover, uniform cross-sectional structures of the formed PAC-DMs and the DM without PAC are seen in FESEM images shown in Figure 1b. In addition, as reported in Table 3, thinner DMs with a lower compressibility index were achieved by PAC addition. In a previous study, it was shown that the axial-flow agitator homogeneously created an appropriate flow pattern in close proximity to the mesh surface (Rezvani et al. 2014). Thus, the use of a mechanical agitator expedited the DM formation and the homogenous mixture of PAC-mixed liquor in the DMBR progressively precipitated on the surface of the mesh filter and formed uniform DMs during the agitation stage.

Effluent CODs of control and each PAC-loaded DMBR over time of DM formation are displayed in Figure 1c. The trends of COD and turbidity variations were similar at different concentrations of PAC particles (Figure 1a and c). However, according to Figure 1c, a small increase in COD values after 20 min of agitation is observed in all tests. Although effluent COD increased after 20 min, no significant change in turbidity was observed (Figure 1a). As the soluble materials do not affect the permeate turbidity, it is inferred that increase in effluent COD is a result of the transfer of soluble materials across the DMs. In other words, during the short time of 20 min, particles did not attach to the mesh surface tightly; thus, some particles were detached from the DMs which were initially formed with loose spatial structures. Hence, the soluble materials much smaller than the pore size of the mesh filter were drawn by the drag force of the pump while other larger particles stably remained on the surface of the filter media.

It is believed that the best PAC-DM is the one which is formed in the shortest time, as well as the lowest possible TMP rise and holds both small soluble molecules and large particles on the membrane surface. Therefore, to find the most efficient PAC concentration (2, 4 or 5 g L\(^{-1}\)) for PAC-DM formation, three parameters including TMP, turbidity and COD removal were considered simultaneously as the criteria.

Figure 1d shows variations of TMP with time during DM formation for three concentrations of PAC particles. All of the profiles exhibited a similar small ascending trend due to the deposition of more sludge flocs and PAC particles with time. Accordingly, an increase in PAC concentration led to a reduction in TMP during agitation, because a higher PAC concentration could reduce fouling (Sagbo et al. 2008; Hu et al. 2014). These results along with other investigations on PAC addition to MBR (Sagbo et al. 2008; Hu et al. 2014) show that the addition of an efficient concentration of PAC particles leads to a decrease in TMP as a consequence of fouling reduction in PAC-(D)MBR systems. However, the decline in TMP is more remarkable with the increase in PAC concentration from 2 to 4 g L\(^{-1}\), while TMP change at a PAC concentration of 5 g L\(^{-1}\) was very similar to that of 4 g L\(^{-1}\). It can be concluded that 4 g L\(^{-1}\) PAC was enough to adsorb all the available absorbable materials, whereas an increase in PAC concentration above 4 g L\(^{-1}\) had no significant influence on TMP variation and quality of permeate.

As observed in Figure 1a and c, an increase in the PAC concentration caused the turbidity to increase although the COD removal was decreased compared to the control DMBR system (without PAC). On the other hand, at a 4 g L\(^{-1}\) PAC concentration, EPS and SMP content reduced up to 34% and 28%, respectively, in comparison to DMBR without PAC.
Statistical analysis indicated a significant difference \((p < 0.05)\) in effluent turbidity of the four DMs studied after formation. Therefore, it is demonstrated that the addition of PAC significantly affected DM formation. Tukey test with a 95% confidence interval revealed a significant difference between means of effluent turbidity obtained by 0, 2 and 4 g L\(^{-1}\) PAC concentrations. However, no significant difference was detected between means of effluent turbidity by 4 and 5 g L\(^{-1}\) PAC concentrations after the formation of DMs.

Similarly, a statistically significant difference was found in the COD of the four systems studied over the time of formation \((p < 0.05)\) using one-way ANOVA. Also, the Tukey comparison test revealed significant differences between CODs obtained for 0, 2 and 4 g L\(^{-1}\) PAC concentrations. However, no significant difference was detected between means of effluent turbidity by 4 and 5 g L\(^{-1}\) PAC concentrations after the formation of DMs. Accordingly, an increase in the PAC concentration from 2 to 4 g L\(^{-1}\) significantly affected the performance of PAC-DMBR in terms of COD removal. Thus, it is deduced that when the concentration of PAC increased, there were more active sites and surface area for adsorption of adhesive materials, leading to higher COD removal and better quality of permeate. However, the addition of the PAC concentration above 4 g L\(^{-1}\) did not enhance COD removal. Indeed, a 4 g L\(^{-1}\) PAC concentration was sufficient to adsorb EPS and SMP contents in the PAC-DMBR.

To recapitulate, according to Figure 1, PAC-DMBR operation during the formation of the PAC-DM was much more efficient at a 4 g L\(^{-1}\) PAC concentration than 2 g L\(^{-1}\), while very similar results (turbidity, TMP and COD) were obtained at a 5 g L\(^{-1}\) PAC concentration and no significant difference existed between 4 and 5 g L\(^{-1}\) PAC contents. So, there was no need to increase PAC concentration above 4 g L\(^{-1}\).

Hence, a PAC concentration of 4 g L\(^{-1}\) was the efficient concentration of PAC particles in the PAC-DMBR at the agitation stage.

**Critical flux of hybrid PAC-DMBR**

One convenient criterion to predict fouling is the critical flux (Field et al. 1995) defined as the flux below which fouling is absent or negligible (Yurtsever et al. 2021). Figure 2 shows the flux and TMP profiles obtained for hybrid (4 g L\(^{-1}\)) PAC-DMBR. As shown in Figure 2, during the first 260 min, with the imposed flux between 35 LMH and 107 LMH, the rate of TMP increase was low and TMP rose from 1 mbar to 5 mbar. However, in the subsequent 20 min, a sudden sensible TMP rise from 5 mbar to 7 mbar was observed; therefore, the imposed flux was considered as the critical flux which was about 107 LMH.

Results of the same experiments showed that the critical flux of DMBR enhanced from 107 LMH to 125 LMH and 137 LMH with an increase in PAC concentration from 2 to 4 and 5 g L\(^{-1}\), respectively. Previously, it was reported that the critical flux of the DMBR without PAC was 77 LMH (Poostchi et al. 2012) which was much lower than the critical flux obtained from the PAC-DMBR in the current study. Furthermore, Chu et al. (2014) and Remy et al. (2009) reported that the addition of PAC to a municipal wastewater treatment plant of MBR increased critical flux compared to MBR without PAC. Indeed, a major increase in critical flux is obtainable by adding PAC particles to MBRs due to PAC capability for adsorbing adhesive materials. The present results reveal that PAC particles successfully mitigated biofouling in the introduced PAC-DMBR. Hence, the critical flux increased. This preferred advantage makes the PAC-DMBR a promising system for industrial
applications where low cost and high critical flux are demanded.

**Performance of hybrid PAC-DMBR**

The most efficient concentration of PAC particles in this stage was obtained based on the hybrid PAC-DMBR ability to decline turbidity and TMP rise to investigate the stability of the bioreactor. Figure 3 shows turbidity changes for three different concentrations of PAC particles and control DMBR system without PAC addition. Statistical analysis demonstrated a significant effect of PAC concentrations and operation time ($p < 0.05$) on turbidity. Tukey pairwise comparison test showed that turbidity of each PAC-DMBR was significantly different from that of the other three concentrations on every day of the experiment. However, no significant difference was found between each pair of PAC concentrations throughout the whole operation time ($p = 0.998$), because the time considered for the formation of compressed layers on the DMs was enough. Hence, the adsorption capacity and concentration of PAC particles played minor roles in turbidity reduction. Nevertheless, it is worth noting that an increase in the PAC concentration from 2 to 4 g L$^{-1}$ led to a gentle reduction in turbidity. Therefore, a 4 g L$^{-1}$ concentration of PAC particles was the adequate concentration in the short-term performance.

**TMP and resistance variations in short-term performance**

After finding the efficient concentration of PAC particles in the two stages of agitation and aeration, the performance of the hybrid PAC-DMBR was investigated by monitoring TMP and effluent quality variations using the determined efficient concentration. Figure 4 displays TMP diagram during the short-term performance (aeration stage) and the corresponding total resistance of the hybrid PAC-DMBR on the second, sixth and twelfth day. As shown in Figure 4, the TMP diagram has an ascending trend during the 12 days of aeration, because the thickness and compressibility of DM increased over time due to the deposition of more flocs and particles on the mesh filter surface as well as the drag force created by the driving force of the peristaltic pump. On the first two days of operation, the total hydraulic filtration

![Figure 3. Effluent turbidity variation with time during the short-term performance of hybrid PAC-DMBRs.](image)

![Figure 4. TMP profile (left vertical axis) and fouling resistance ($R_f$) and cake resistance ($R_c$) at the corresponding times (right vertical axis) during the short-term performance of hybrid (4 g L$^{-1}$) PAC-DMBR.](image)
resistance increased to $3.14 \times 10^{11}$ m$^{-1}$, where contributions of cake resistance ($R_c$) and fouling resistance ($R_f$) were about 90% and 10%, respectively. A considerable enhancement in the total resistance value up to $5.01 \times 10^{11}$ m$^{-1}$ with the cake and gel layer constituting 92% and 8% of the total resistance, respectively, confirmed that DM became thicker and even more compressible on the sixth day of operation. During 12 days of operation, the total filtration resistance was enhanced to $2.7 \times 10^{12}$ m$^{-1}$, with 96% and less than 4% related to cake and gel, respectively. As a result, in a prolonged operation, enhancement in total resistance was achieved, thus the portion of the cake layer in hydraulic resistance became higher than that of the gel layer. After 12 days of operation, the thickness and compressibility of the DM were so high that the peristaltic pump was unable to suck the effluent, so the operation was stopped. In a previous study, resistance results of the same DMBR operated without PAC particles in the same conditions were reported (Rezvani et al. 2014). In the DMBR without PAC, the third stage of TMP started with a jump after 8 days of operation. However, according to Figure 4, the third stage of TMP occurred on the sixth day of hybrid PAC-DMBR operation. This difference is observed because of the higher operational flux imposed in the hybrid PAC-DMBR performance. Due to the higher critical flux of the PAC-DMBR compared to DMBR without PAC, the hybrid PAC-DMBR can operate at higher operational fluxes.

**Effluent quality variation in short-term performance**

To investigate hybrid PAC-DMBR short-term performance, variation of effluent quality was monitored during 12 days of operation using the efficient concentration (a PAC concentration of $4$ g L$^{-1}$). Figure 5a–d indicate variations of COD, TDS, TN and TP.
As displayed in Figure 3 and also Figure 5a–d, turbidity, COD, TDS, TN and TP values showed nearly similar decreasing trends with time. Reductions were more dramatic from the start of aeration until day one for two reasons: (1) DM was formed during this period, and (2) PAC possibly reached the equilibrium adsorption capacity (Poostchi et al. 2010). For these two reasons, the slope of the diagrams became much lower after the first day. Then, reductions continued until all of the parameters reached approximately constant levels after six days of operation. Afterwards, the variations of effluent COD, TDS, TN and TP were negligible. According to Figure 5b, the hybrid PAC-DMBR was able to remove TDS very efficiently while it was observed in our previous study that DMBR was not well-functioned in terms of TDS reduction (Rezvani et al. 2014). On the other hand, compared to a similar study by Ye et al. (2006) which used PAC as a pre-coating agent in DM formation, the hybrid PAC-DMBR investigated in the present study was able to reduce nitrogen and COD more efficiently in a shorter time, due to formation of a bio-gel layer on the DM with a high retention capacity.

According to Figure 5c, the hybrid PAC-DMBR was also able to remove TN. Generally, most of the soluble ammonium nitrogen \((\text{NH}_4^+ - N)\) in wastewater is removed and converted to nitrate nitrogen \((\text{NO}_3^- - N)\) by aerobic nitrifiers during aerated treatment. Subsequently, the remaining nitrate is removed by anoxic microorganisms (Chang et al. 2022). Considering 8% TN reduction in the present study, it seems that both anoxic and aerobic microorganisms may play a role in nitrogen removal in hybrid PAC-DMBR. In other words, it is possible that lack of aeration in the formation stage (agitation stage) followed by applying aeration in short-term performance (aeration stage) resulted in a stratification-structure biofilm formation on PAC particles (Jamal Khan et al. 2011). Furthermore, an increase in the size of suspended flocs in presence of PAC particles according to particle size analysis can be an indication of biofilm formation around PAC particles. Such biofilms, including an inner anoxic layer and an outer aerobic layer, can facilitate nitrification and denitrification reactions (Qureshi et al. 2005). Although an in-depth investigation regarding biofilm structure around PAC in DMBR needs to be conducted in further studies, the results of the present study show the ability of hybrid PAC-DMBR in removing TN.

Additionally, the results of this study were consistent with a similar study carried out by Chu et al. (2013) while being even more efficient in organic matter removal. Finally, it was verified that the PAC-DMBR containing a 4 g L\(^{-1}\) concentration of PAC particles was able to deplete pollutants very efficiently.

**Conclusions**

In this study, the suitability of PAC addition to DMBR was investigated during two stages of 1- DM formation (agitation), and 2- hybrid PAC-DMBR operation (aeration). In the first stage, since the axial-flow agitator uniformly created the appropriate flow pattern in close proximity to the mesh surface, the use of a mechanical agitator expedited the DM formation and improved the uniformity of the DM structure.

Results proved that PAC addition made the DM more permeable and enhanced the critical flux in PAC-DMBR as a result of biofouling mitigation by PAC particles. The efficient PAC concentration in both stages was 4 g L\(^{-1}\) since the highest organic matter removal and the least TMP rise were achieved at this dosage. The hybrid PAC-DMBR successfully reduced effluent turbidity, COD, TDS, TN and TP at the aeration stage. Particularly, the capability of the hybrid PAC-DMBR in removing TDS was enhanced significantly compared to DMBR systems without PAC. As a proof of concept, the hybrid PAC-DMBR introduced in this study possesses MBR qualifications in addition to resolving the drawbacks including membrane fouling and high costs.

**Disclosure statement**

The authors declare that they have no conflict of interest.

**Funding**

The author(s) reported there is no funding associated with the work featured in this article.

**References**

APHA. 2005. Standard methods for the examination of water and wastewater. 21st ed. Washington (DC): American Public Health Association.

Asif MB, Ren B, Li C, Maqbool T, Zhang X, Zhang Z. 2020. Powdered activated carbon – membrane bioreactor (PAC-MBR): impacts of high PAC concentration on micropollutant removal and microbial communities. Sci Total Environ. 745:141090. doi:10.1016/j.scitotenv.2020.141090

Aslam A, Khan SJ, Shahzad HMA. 2022. Anaerobic membrane bioreactors (AnMBRs) for municipal wastewater treatment- potential benefits, constraints, and future
Ayed, N., Asses, N., Chammem, N., Othman, B., & Hamdi, M. (2017). Advanced oxidation process and biological treatments for table olive processing wastewaters: constraints and a novel approach to integrated recycling process: a review. Biodegradation. 28:125–138. doi:10.1007/s10532-017-9782-0

Chu, H., Zhang, Y., Zhou, X., Dong, B. (2013). Bio-enhanced powder-activated carbon dynamic membrane reactor for municipal wastewater treatment. J Membr Sci. 433:126–134. doi:10.1016/j.memsci.2013.01.030

Chu, H., Zhang, Y., Zhou, X., Zhao, Y., Dong, B., Zhang, H. (2014). Dynamic membrane bioreactor for wastewater treatment: operation, critical flux, and dynamic membrane structure. J Membr Sci. 450:265–271. doi:10.1016/j.memsci.2013.08.045

Damayanti, A., Ujang, Z., Salim, M.R. (2011). The influence of PAC, zeolite, and Moringa oleifera as bioflooding reducer (BFR) on hybrid membrane bioreactor of palm oil mill effluent (POME). Bioresour Technol. 102:4341–4346. doi:10.1016/j.biortech.2010.12.061

Etemadi, H., Yegani, R. (2019). Effect of aeration rate on the anti-bioflooding properties of cellulose acetate nanocomposite membranes in a membrane bioreactor system for the treatment of pharmaceutical wastewater. Biofouling. 35:618–630. doi:10.1080/08927014.2019.1637858

Fang, H.H.P., Shi, X., Zhang, T. (2006). Effect of activated carbon fouling on fouling of activated sludge filtration. Desalination. 189:193–199. doi:10.1016/j.desal.2005.02.087

Field, R.W., Wu, D., Howell, J.A., Gupta, B.B. (1995). Critical flux concept for microfiltration fouling. J Membr Sci. 100:259–272. doi:10.1016/0376-7388(94)00265-Z

Holloway, T.G., Williams, J.B., Ouelhadj, D., Yang, G. (2021). Dynamic resilience for biological wastewater treatment processes: interpreting data for process management and the potential for knowledge discovery. J Water Process Eng. 42:102170. doi:10.1016/j.jwpe.2020.102170

Hu, J., Shang, R., Deng, H., Heijman, S.G.J., Rietveld, L.C. (2014). Effect of PAC dosage in a pilot-scale PAC-MBR treating micro-polluted surface water. Bioresour Technol. 154:290–296. doi:10.1016/j.biortech.2013.12.075

Hu, Y., Wang, X.C., Tian, W., Ngo, H.H., Chen, R. (2016). Towards stable operation of a dynamic membrane bioreactor (DMBR): operational process, behavior and retention effect of dynamic membrane. J Membr Sci. 498:20–29. doi:10.1016/j.memsci.2015.10.009

Hu, Y., Yang, Y., Wang, X.C., Hao, N.G., Sun, Q., Li, S., Tang, J., Yu, Z. (2017). Effects of powdered activated carbon addition on filtration performance and dynamic membrane layer properties in a hybrid DMBR process. Chem Eng J. 327:39–50. doi:10.1016/j.cej.2017.06.072

Iorhemen, O., Hamza, R., Tay, J. (2016). Membrane bioreactor (MBR) technology for wastewater treatment and reclamation: membrane fouling. Membranes. 6:33. doi:10.3390/membranes6020033

Jamal Khan, S., Ilyas, S., Javid, S., Visvanathan, C., Jegatheesan, V. (2011). Performance of suspended and attached growth MBR systems in treating high strength synthetic wastewater. Bioresour Technol. 102:5331–5336. doi:10.1016/j.biortech.2010.09.100

Jiang, Y., Liu, Y., Shi, D., Fu, W., Sun, P-F., Li, J., Shao, S. (2020). Membrane fouling in a powdered activated carbon – membrane bioreactor (PAC-MBR) for micro-polluted water purification: fouling characteristics and the roles of PAC. J Cleaner Prod. 277:122341. doi:10.1016/j.jclepro.2020.122341

Ko, D. (2018). Conceptual design optimization of an integrated membrane bioreactor system for wastewater treatment. Chem Eng Res Des. 132:385–398. doi:10.1016/j.cherd.2018.01.030

Kozak, M., Cirik, K., Bašak, S. (2021). Treatment of textile wastewater using combined anaerobic moving bed biofilm reactor and powdered activated carbon-aerobic membrane reactor. J Environ Chem Eng. 9:105596. doi:10.1016/j.jece.2021.105596

Kulesha, O., Maletskiy, Z., Ratnaweera, H. (2018). State-of-the-art of membrane flux enhancement in membrane bioreactor. Cogent Eng. 5:1489700. doi:10.1080/23311916.2018.1489700

Lee, J., Ahn, W-Y., Lee, C-H. (2001). Comparison of the filtration characteristics between attached and suspended growth microorganisms in submerged membrane bioreactor. Water Res. 35:2435–2445. doi:10.1016/S0043-1354(00)00524-8

Lee, K., Choo, K-H., Ng, H.Y., Lee, C-H. (2020). Preparation of a mesoporous silica quorum quenching medium for wastewater treatment using a membrane bioreactor. Biofouling. 36:369–377. doi:10.1080/08927014.2020.1749601

Lee, W-N., Yeon, K-M., Hwang, B-K., Lee, C-H., Chang, I-S. (2010). Effect of PAC addition on the physicochemical characteristics of bio-cake in a membrane bioreactor. Sep Sci Technol. 45:896–903. doi:10.1080/01496391003666999

Lièbana, R., Arregui, L., Belda, I., Gamella, L., Santos, A., Marquina, D., Serrano, S. (2015). Membrane bioreactor wastewater treatment plants reveal diverse yeast and protozoan communities of potential significance in biofouling. Biofouling. 31:71–82. doi:10.1080/08927014.2014.998206

Liu, L., Chen, Z., Zhang, J., Shan, D., Wu, Y., Bai, L., Wang, B. (2021). Treatment of industrial dye wastewater and pharmaceutical residue wastewater by advanced oxidation processes and its combination with nanocatalysts: a review. J Water Process Eng. 42:102122. doi:10.1016/j.jwpe.2021.102122

Loderer, C., Gahlenbauer, B., Steinbacher, K., Stelzer, C., Fuchs, W. (2013). Dynamic filtration – a novel approach for critical flux determination using different textiles. Sep Purif Technol. 120:410–414. doi:10.1016/j.seppur.2013.10.013

Mafrad, S., Mehrnia, M.R., Azami, H., Sarrafzadeh, M.H. (2011). Effects of biofilm formation on membrane performance in submerged membrane bioreactors. Biofouling. 27:477–485. doi:10.1080/08927014.2011.584619

Mohan, S.M., Nagalakshmi, S. (2020). A review on aerobic self-forming dynamic membrane bioreactor: formation, performance, fouling and cleaning. J Water Process Eng. 37:101541. doi:10.1016/j.jwpe.2020.101541

Mororó, R.R., Borges, C.P., Kronemberger, F.de A. (2018). New modules for membrane bioreactors: improving fouling control. Chem Eng Res Des. 136:295–303. doi:10.1016/j.cherd.2018.05.035

Najmi, M., Mehrnia, M.R., Tashauoei, H.R., Iranpoury, A., Alivand, M.S. (2020). Removal of personal care products perspectives: an updated review. Sci Total Environ. 802:149612. doi:10.1016/j.scitotenv.2021.149612
(PCPs) from greywater using a submerged membrane bioreactor (SMBR): the effect of hydraulic retention time. J Environ Chem Eng. 8:104432. doi:10.1016/j.jece.2020.104432

Pidon M, Avery L, Stephenson T, Jeffrey P, Parsons SA, Liu S, Memon FA, Jefferson B. 2008. Chemical solutions for greywater recycling. Chemosphere. 71:147–155. doi:10.1016/j.chemosphere.2007.10.046

Poostchi AA, Bayat M, Rezaei M, Amini E, Mehrnia MR. 2015. Formation of pre-coating dynamic membrane on mesh filter by cross-flow filtration of PAC-water suspension in a bioreactor: experimental and modeling. Desalin Water Treat. 55:17–27. doi:10.1080/19443994.2014.911118

Poostchi AA, Mehrnia MR, Rezvani F, Sarrafzadeh MH. 2012. Low-cost monofilament mesh filter used in membrane bioreactor process: filtration characteristics and resistance analysis. Desalinination. 286:429–435. doi:10.1016/j.desal.2011.12.002

Poostchi AA, Mehrnia MR, Sarrafzadeh MH. 2010. Removal of dissolved organic carbon by multi-walled carbon nanotubes, powdered activated carbon and granular activated carbon. Res J Chem Environ. 14:59–66.

Prathapar SA, Ahmed M, al Adawi S, al Sidiari S. 2006. Design, construction and evaluation of an abluton water treatment unit in Oman: a case study. Int J Environ Stud. 63:283–292. doi:10.1080/00207230600773257

Qureshi N, Annous BA, Ezeji TC, Karcher P, Maddox IS. 2013. Biofilm reactors for industrial bioconversion processes: employing potential of enhanced reaction rates. Microb Cell Fact. 4:24. doi:10.1186/1475-2859-4-24

Remy M, van der Marel P, Zwijnenburg A, Rulkens W, Temmink H. 2009. Low dose powdered activated carbon addition at high sludge retention times to reduce fouling in membrane bioreactors. Water Res. 43:345–350. doi:10.1016/j.watres.2008.10.033

Rezaei M, Mehrnia MR. 2014. The influence of zeolite (clino-nipolilolite) on the performance of a hybrid membrane bioreactor. Bioresour Technol. 158:25–31. doi:10.1016/j.biortech.2014.01.138

Rezvani F, Mehrnia MR, Poostchi AA. 2014. Optimal operating strategies of SFDM formation for MBR application. Sep Purif Technol. 124:124–133. doi:10.1016/j.seppur.2014.01.028

Sabaghian M, Mehrnia MR, Esmaeili M, Noormohammadi D. 2018. Formation and performance of self-forming dynamic membrane (SFDM) in membrane bioreactor (MBR) for treating low-strength wastewater. Water Sci Technol. 78:904–912. doi:10.2166/wst.2018.368

Sagbo O, Sun Y, Hao A, Gu P. 2008. Effect of PAC addition on MBR process for drinking water treatment. Sep Purif Technol. 58:320–327. doi:10.1016/j.seppur.2007.05.003

Sari Erkan H, Çağlak A, Soysaloglu A, Takatas B, Onkal Engin G. 2020. Performance evaluation of conventional membrane bioreactor and moving bed membrane bioreactor for synthetic textile wastewater treatment. J Water Process Eng. 38:101631. doi:10.1016/j.jwpe.2020.101631

Sokhandan F, Homayoonfal M, Davar F. 2020. Application of zinc oxide and sodium alginate for biofouling mitigation in a membrane bioreactor treating urban wastewater. Biofouling. 36:660–678. doi:10.1080/08927014.2020.1798934

Vergine P, Salerno C, Berardi G, Pollice A. 2018. Sludge cake and biofilm formation as valuable tools in wastewater treatment by coupling Integrated Fixed-film Activated Sludge (IFAS) with Self Forming Dynamic Membrane BioReactors (SFD-MBR). Bioresour Technol. 268:121–127. doi:10.1016/j.biortech.2018.07.120

Vergine P, Salerno C, Berardi G, Pollice A. 2021. Self-Forming Dynamic Membrane BioReactors (SFD MBR) for municipal wastewater treatment: relevance of solids retention time and biological process stability. Sep Purif Technol. 255:117735. doi:10.1016/j.seppur.2020.117735

Xiang S, Han Y, Jiang C, Li M, Wei L, Fu J, Zhu L. 2021. Composite biologically active filter (BAF) with zeolite, granular activated carbon, and suspended biological carrier for treating algae-laden raw water. J Water Process Eng. 42:102188. doi:10.1016/j.jwpe.2021.102188

Yang S, Zhang Q, Lei Z, Wen W, Huang X, Chen R. 2019. Comparing powdered and granular activated carbon addition on membrane fouling control through evaluating the impacts on mixed liquor and cake layer properties in anaerobic membrane bioreactors. Bioresour Technol. 294:122137. doi:10.1016/j.biortech.2019.122137

Yang W, Paetkau M, Cicek N. 2010. Effects of powdered activated carbon dosing on sludge characteristics and estrogen removal in membrane bioreactors. Water Sci Technol. 61:2193–2198. doi:10.2166/wst.2010.111

Ye M, Zhang H, Wei Q, Lei H, Yang F, Zhang X. 2006. Study on the suitable thickness of a PAC-precoated dynamic membrane coupled with a bioreactor for municipal wastewater treatment. Desalination. 194:108–120. doi:10.1016/j.desal.2005.11.005

Yu Z, Hu Y, Dzakpasu M, Wang XC, Ngo HH. 2019. Dynamic membrane bioreactor performance enhancement by powdered activated carbon addition: evaluation of sludge morphological, aggregative and microbial properties. J Environ Sci (China). 75:73–83. doi:10.1016/j.jes.2018.03.003

Yuniarto A, Noor ZZ, Ujang Z, Olsson G, Aris A, Hadihara T. 2013. Bio-fouling reducers for improving the performance of an aerobic submerged membrane bioreactor treating palm oil mill effluent. Desalination. 316:146–153. doi:10.1016/j.desal.2013.02.002

Yurtsever A, Basaran E, Ucar D, Sahinkaya E. 2021. Self-forming dynamic membrane bioreactor for textile industry wastewater treatment. Sci Total Environ. 751:141572. doi:10.1016/j.scitotenv.2020.141572

Zhou L, Zhang Z, Jiang W, Guo W, Ngo H-H, Meng X, Fan J, Zhao J, Xia S. 2014. Effects of low-concentration Cr(VI) on the performance and the membrane fouling of a submerged membrane bioreactor in the treatment of municipal wastewater. Biofouling. 30:105–114. doi:10.1080/08927014.2013.847925