Effects of powdered activated carbon addition on filtration performance and dynamic membrane layer properties in a hybrid DMBR process

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Abstract

A powdered activated carbon-dynamic membrane bioreactor (PAC-DMBR) was developed and used to treat domestic wastewater by dosing with 3 g/L PAC. The experimental results were compared with those of a control DMBR to investigate the filtration performance and various properties of the dynamic membrane (DM) layer. One flat-sheet DM module made of nylon mesh (pore size 75 μm) was used for effluent production at a high stable flux (50–100 L/m^2 h) under a 10 cm water head by gravity flow, resulting in continuous operation cycles of 60–120 h. During the operation period, the PAC-DMBR showed enhanced removal efficiency of pollutants, higher stable membrane flux (10 L/m^2 h more), lower filtration resistance ($6.0–8.0 \times 10^{10}$ m^{-1}), quicker formation of the DM layer (within 5 min), and better DM layer regeneration after air backwashing. The DM layer in the PAC-DMBR showed a more porous and incompressible structure, because less extracellular polymeric substance and a portion of the biological PAC were incorporated into the DM layer formed as verified by the analytical results. Using high-throughput pyrosequencing technology, it was revealed that at the genus level the diversity of bacterial communities increased from 18 to 23 genera, while several genera that were favored in the PAC-assisted environment or were responsible for degrading complex organics were enriched. Moreover, the abundance of phylum *Proteobacteria*, which served as pioneer surface colonizers, was reduced in the PAC-DMBR. It was concluded that PAC addition could modify various aspects of the activated sludge and the DM layer properties, which affected the filtration behavior of the DM layer in the PAC-DMBR.

1. Introduction

Dynamic membrane (DM) technology is regarded as a promising and cost-effective approach for wastewater treatment, as it uses low-cost materials (such as meshes, woven and non-woven fabrics) to support the formation of a DM layer, a ‘‘secondary” membrane prior to the support material when filtering suspended solid particles such as activated sludge [1,2]. The dynamic membrane bioreactor (DMBR) process is a combination of the biological treatment process with the DM technology for treating wastewater. It shows several advantages (lower membrane cost, higher flux and easier membrane cleaning) over conventional MBRs using ultrafiltration or microfiltration (UF/MF) membranes [3].

However, a main drawback to using a self-forming dynamic membrane is that the DM layer is not as good as the conventional UF/MF membranes. This can be attributed to the following possible reasons. First, a certain formation time is commonly needed for the attachment and maturation of the DM layer for first-time usage of the DM module and for the subsequent physical cleaning. During this stage, poor effluent quality could be expected [4]. In addition, the DM layer formed could be adversely affected by several factors such as aeration intensity, water head and sludge properties, and thus a sudden reduction in effluent quality could occur. Last, although physical cleaning for DM regeneration could almost completely recover the permeate flux to the initial state, certain residual foulants were still observed in some cases [5,6], to which more attention should be paid during long-term continuous operational cycles.

Therefore, several studies were attempted to improve the performance and stable operation of DMBRs using various strategies, such as the optimization of operational parameters, the selection of proper supporting material and membrane module configurations, and the modification of sludge properties using various additives [7–9]. For example, it was shown that the formation time of DMs could be shortened in the case of a sludge suspension containing more particles with larger mean diameter and higher relative hydrophobicity (RH). The fouling propensity of fresh DMs was found to be affected by another four sludge properties, including apparent viscosity, extracellular polymeric substances (EPS), carbohydrate content in the EPS, and the specific oxygen uptake rate (SOUR) [7]. Another work identified the effects of different mesh openings (25–140 μm) and different tubular module configurations (inside-outside and outside-inside) in the DMBR on process performance [8]. It was noted that the configuration of the filter module had little impact on effluent quality and that the mesh pore size influenced the effluent turbidity. Therefore, for a given DMBR system, modification of the sludge properties could be more practical and suitable for enhancing the filtration ability of the DM layer.

Hybrid DMBRs developed by adding various additives were verified to be effective in improving treatment performance, modifying sludge properties and enhancing the permeability of the DM layer [10–14]. A previous DMBR study reported that sludge properties, including flocculating, settling and dewatering abilities, were enhanced after PAC addition, and were accompanied by better DM layer permeability [10]. In a bio-diatomite enhanced DMBR, good permeability of the DM layer was attributed to the homogeneous, semi-compressibility, and highly porous structure of the cake layer [11]. In a biologically enhanced PAC-diatomite DMBR system, stable removal efficiencies of routine pollutants were achieved, and the DM layer showed a two-layer structure due to successive deposition of larger PAC and smaller diatomite particles [12]. In addition, morphology analysis of the cake layer in a bio-diatomite DMBR showed a smooth surface and a thickness of 2–3 mm [13]. When PAC-DMBRs were applied for treating industrial wastewater from an alcohol distillery, it was found that the microbial activity, sludge particle size and total EPS concentration were not affected but sludge dewaterability was improved and the EPS composition was altered [14]. The reported results indicated that modification of sludge properties could affect the sludge filterability, and more importantly, play a role in the structure and permeability of the formed DM layer.

Another important issue was that the added PAC would serve as a bio-carrier facilitating the adsorption of organics and the growth of microorganisms to form a complex matrix. This would assure that PAC-assisted biological processes contain many micro ecosystems. Such attached-growth microorganisms could contribute to the removal of some slowly biodegradable substances, and also affect the microbial community structure in the activated sludge and the cake layer [15–17]. So far, investigations to detect microbial properties using modern techniques, to understand the potential microbial effects of PAC addition in a hybrid DMBR system, are quite limited.

In all, although some useful information on the characterization of the DM layer in hybrid DMBRs was obtained, systematic investigation (such as the structure, and physicochemical and microbial properties of the DM layer) was still necessary to improve our understanding of the filtration behavior of the DM layer in relation to its physicochemical and biological characteristics. Therefore, in this study, a series of analytical techniques, including particle size distribution (PSD) analysis, three-dimensional excitation emission matrix (EEM) fluorescence spectroscopy, gel filtration chromatography (GFC), attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy, scanning electron microscopy (SEM), energy-diffusive X-ray (EDX) analysis, and 454 high-throughput pyrosequencing technology were applied to investigate the filtration performance and various properties of the DM layer in the PAC-DMBR. The findings were compared against the corresponding results from the control DMBR. The results obtained would provide useful information on the behavior of the DM layer, and also advance the practical application of the PAC-DMBR hybrid process.

2. Materials and methods

2.1. Experimental setup and operation

Two identical rectangle DMBRs with an effective working volume of 15 L (width × length × height = 18 × 21 × 46 cm) were developed and run in parallel for domestic wastewater treatment (shown in Fig. 1). The reactors are located at a local wastewater treatment plant (WWTP) in Xi’an, China, and are hereafter referred to as the C-DMBR (without PAC) and PAC-DMBR (with PAC addition). During the startup period, 3 g/L of commercial PAC was added to the PAC-DMBR. The PAC was analytical grade provided by Tianjin Fangzheng Reagent Corporation, with a mean particle size of 29.4 μm and a specific surface area of approximate 1100 m^2/g. In each bioreactor, one flat-sheet DM module was immersed vertically with a double-sided effective filtration area of 0.04 m^2. The DM module was made of PVC plates, an inner support layer (commercial stainless steel mesh with 10 mm pore size of knitted steel wire with a diameter of 0.8 mm), and an outer layer of support material (nylon mesh with 75 μm pore size). The inoculation sludge was obtained from the local WWTP. After two weeks acclimation, the sludge was put into the two DMBRs for continuous operation. At the same time, PAC cleaned with deionized water was dosed into the PAC-DMBR as the additive. After a short time for startup, the DMBRs achieved stable operation.

Four identical air diffusers (made of quartz sand, height and diameter 3 and 2.2 cm, respectively) were installed at the bottom of the four corners in each reactor, causing visible micro-bubbles with a diameter of several millimeters. Air pumps were used to continuously supply the oxygen demand for biomass and to mix

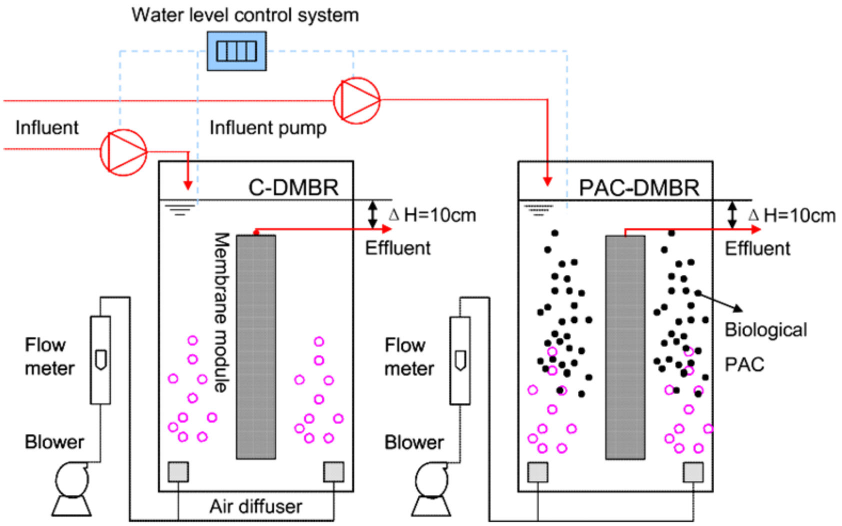

Fig. 1. Schematic diagram of the experimental setup.

the activated sludge in the bioreactor, which caused the dissolved oxygen (DO) concentration in the DMBRs to fall within the range 2.0–6.0 mg/L. Each day, 50 mL of sludge was sampled for analysis of sludge properties, without further sludge discharge. The MLSS concentrations averaged at 2500 mg/L (C-DMBR) and 5000 mg/L (PAC-DMBR), respectively. Real domestic wastewater was fed into the PAC-DMBRs by submersible pumps. The effluent was withdrawn continuously by maintaining a 10 cm water level difference between the bioreactor and the effluent port until the flux dropped to a predetermined value. As such, the hydraulic retention time varied in the range 4–10 h. Then air backwashing with a flow rate of 72 L/min for 2 min was applied for DM regeneration to start the next operation cycle [6]. The characteristics of the influent and effluent from the C-DMBR and PAC-DMBR are given in Table 1. It was detected that although the influent water quality fluctuated widely, the removal of pollutants was generally stable and satisfactory. Moreover, the enhancement of treatment performance by PAC addition was clear in the comparison of the effluent quality of the two DMBRs.

Table 1
Influent and effluent water quality of the C-DMBR and PAC-DMBR.

Parameters	Influent	Effluent	
		C-DMBR	PAC-DMBR
COD (mg/L)	110.6–204.4	21.8	19.0
UV_{254} (cm^{-1})	0.10–0.23	0.10	0.07
NH_3-N (mg/L)	16.7–31.4	0.64	0.55
TP (mg/L)	1.9–3.8	2.41	2.38
Color (c.u.)	97.0–181.0	17.5	9.2

2.2. Analytical methods

2.2.1. Sludge sample collection and pretreatment

DM layer formed on the membrane modules was scraped off by a plastic sheet, which was operated according to the previous studies [18,19]. In detail, firstly the collected sludge was diluted with deionized water to a similar MLSS concentration as that of the activated sludge in the bioreactor, and then the diluted sample was placed on a magnetic blender and gently mixed. Finally the pretreated samples were subjected to the following measurements.

2.2.2. EPS analysis

EPS could be divided into soluble EPS (SEPS) and bound EPS (BEPS), respectively, according to their different locations to sludge flocs. SEPS and BEPS were extracted from both the activated sludge and cake sludge samples according to the reported method [20]. The analysis of the extracted EPS samples was carried out for proteins using the modified Lowry method [21] with bovine serum albumin (BSA) as the standard, and for polysaccharides using the phenol-sulfuric acid method [22] with glucose as the standard.

2.2.3. PSD analysis

PSD of the sludge samples was measured using a laser granularity distribution analyzer (LS 230/SVM+, Beckman Coulter Corporation, USA), which had a detection range of 0.4–2000 μm and showed a good accuracy and reproducibility. In this study the typical PSD curves were reported.

2.2.4. SEM-EDX analysis

To detect the morphology and inorganic elements of DM layers and nylon mesh, SEM (VEGA 3LMH, Tescan Corporation, Czech) and EDX analyzer (Oxford INCA Energy 350, UK) were applied. The pretreatment for the DM layers and nylon mesh was consistent with the described methods [6]. In brief, the nylon meshes covered by DM layer were cut from the middle of the DM modules. Then the samples were fixed with 2.0% glutaraldehyde for 8 h, followed by dehydrated with ethanol and coated with aurum-platinum alloy. The dehydrated samples were used for SEM-EDX analysis, while new mesh was directly adopted for SEM-EDX analysis without any pretreatment.

2.2.5. ATR-FTIR analysis

ATR-FTIR spectrometer (Nicolet iS50, Thermo Electron Corporation, USA) was used to characterize the major functional groups of different samples. The sludge and mesh samples were collected and dried naturally, then the direct measurement was conducted without further sample pretreatment. The wave number was determined over the range of 4000–400 cm^{-1}.

2.2.6. 3D-EEM analysis

The 3D-EEM fluorescence spectra of the dissolved organics in various samples were measured using an FP-6500 spectroflurometer (Jasco Corporation, Japan). During the measurement, excitation wavelengths increased from 220 nm to 450 nm with a step of 5 nm. For each excitation wavelength, the emission was detected from 220 nm to 550 nm with a step of 5 nm. The scan speed was set at 12,000 nm/min. EEM spectra as the elliptical shape of contours was plotted using the software Origin Pro 8.0 (Origin Lab Corporation, USA).

2.2.7. GFC analysis

The molecular weight distributions of samples were determined using a GFC analyzer (LC-2010A, Shimadzu Corporation, Japan) installed with a Zenix sEC-100 type gel column (Sepax Technologies Corporation, USA) and a UV detector (SPD-10, Shimadzu Corporation, Japan) at 40 °C. 150 mM sodium phosphate buffer (including Na_2HPO_4 and NaH_2PO_4) was adopted as the eluent at a flow rate of 1.0 mL/min. The dissolved organic matter in samples was obtained by filtering a 0.22 μm prior to the injection, while the injection volume was 50 μL for effluent and SEPS, and lower (5 μL) for BEPS.

2.2.8. Microbial property analysis

To understand the microbial community structure in the DMBRs during the stable operation period, activated sludge and DM layer samples were analyzed by 454 high-throughput pyrosequencing method according to the previously reported references [23–26]. In brief, the procedures included DNA extraction, PCR amplification, 454 high-throughput 16S rRNA gene pyrosequencing, and then biodiversity analysis and phylogenetic classification. To date, the extracted DNA was amplified by PCR using the primer 8F (5′-AGAGTTTGATCCTGGCTCAG-3′) and 533R (5′-TTACCGCGGC TGCTGGCAC-3′) for the V1-V3 region of the 16S rRNA gene. Pyrosequencing was conducted using a Roche 454 GS FLX Titanium sequencer.

2.2.9. Other analysis

Measurements of chemical oxygen demand (COD), UV_{254}, ammonia (NH_3-N), total phosphorus (TP), color, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) in the bioreactor were according to the standard methods [27]. Microscopy observation of the sludge samples was captured by a digital camera (N90i, Nikon Corporation, Japan) attached to a microscope. The photography of the membrane modules was taken by an SLR camera (EPM2, Olympus Corporation, Japan). Turbidity was measured with a turbidity meter (ET266020, Lovibond Corporation, Germany), and the filtration flux of the DM with the volumetric method. The total filtration resistance (R_t) was calculated based on Darcy law using the varied flux during filtration cycles and constant water head or trans-membrane pressure (TMP) according to the previous study [10].

3. Results and discussion

3.1. Overall performance of the DMBRs

3.1.1. Filtration performance

The two DMBRs were stably operated for about two months. The treatment performance of the DMBRs can be found in Table 1 and Supporting Information (Fig. S1). The results showed that the removal of COD, UV_{254}, NH_3, and color was quite successful because the oxic conditions (DO 2–6 mg/L) maintained in the bioreactors were beneficial for the growth of heterotrophic bacteria and nitrifiers. Obviously, compared to the C-DMBR, lower pollutant concentrations in the effluent and better treatment performance were continuously found for the PAC-DMBR. However, no significant difference in the removal of total phosphorus (TP) was detected in the two DMBRs. This was because the removal efficiencies were both quite low (20–30%) due to the absence of anaerobic/anoxic conditions for efficient TP removal. It has been recognized that biodegradation is the main mechanism for pollutant removal and that the retention effect of the DM layer could enhance their removal [5,6]. In this study, more efficient pollutant removal (especially of organic substances) in the PAC-DMBR was attributed to improvement in the physicochemical and biological effects by PAC due to enhanced adsorption, biodegradation, and also the retention effect of the DM layer, as noted in previous studies conducted on DMBRs as well as MBRs [6,10,28,29].

The typical variations of the membrane flux and total filtration resistance (R_t) with operation time in the DMBRs are demonstrated in Fig. 2, while the decline of effluent turbidity can be found in Fig. S2 in Supporting Information. In the DMBRs, the filtration was carried out by the gravity at a constant water head (average 10 cm), so the fluxes decreased with time under the constant pressure operation mode. As documented, the operation cycles could be divided into three stages by detecting the changes of flux and effluent turbidity, which included a quick formation and maturation stage, a stable operation stage, and a backwashing stage [6,30].

In this study, it was found that formation time was about 5–20 min and less than 5 min in the C-DMBR and PAC-DMBR, respectively. Because the flux halved and effluent turbidity decreased below 1NTU during the DM formation stage, shorter DM formation time in the PAC-DMBR was needed after PAC addition. Then, the flux tended to stabilize within 4 h, with a tendency to gradual decline, but kept quite constant afterwards at a negligible rate of flux decrease that lasted for about 60–120 h. Similar evolution of the flux in the DMBRs was detected, however higher stable flux (about 10 L/m^2h) was noted in the PAC-DMBR, with values in the range 50–70 L/m^2h. Furthermore, R_t showed an increasing tendency evidenced by a sharp increase followed by a gradual rise. Higher filtration resistance was constantly found in the C-DMBR, with the final values in the range $8.0–10.0 \times 10^{10}\ m^{-1}$ compared to those in the PAC-DMBR ($6.0–8.0 \times 10^{10}\ m^{-1}$); however, these values were still one to two orders of magnitude lower than those commonly observed in con- ventional MBRs [31]. The differences in filtration performance in PAC-DMBR and C-DMBR were considered to be determined by modifications in the properties of the activated sludge and the formed DM layer, which will be discussed further in the subsequent sections.

3.1.2. Air backwashing for DM regeneration

From the above analysis, it was also noted that at the end of one operational cycle, a physical cleaning method like air backwashing was adopted for DM layer regeneration. After this, the flux could almost be recovered to the initial condition during successive operational cycles. At the end of the experiment, the membrane modules were taken out of the bioreactors for further analysis.

Fig. 3 shows photographs of the different membrane modules. From Fig. 3 (A)–(C), compared to the new mesh, the 2–3 mm thick DM layer could easily be observed on the surface of the mesh from the two DMBRs, and was evenly distributed on the mesh surface. However, the DM layer in the PAC-DMBR was black due to the attachment of biological PAC sludge, which was different from the yellow color found in the C-DMBR. As seen in Fig. 3 (D) and (E), no obvious DM layer or foulants could be found on the mesh surface after air backwashing, and this was verified by the ATR-FTIR analysis showing that the spectra of backwashed meshes were almost the same as that of the new mesh (Fig. S3 in Supporting Information).

Further observation was also carried out using SEM measurements (shown in Fig. 3). From Fig. 3 (a)–(e), it was indicated that the DM layers were quite different because a gel-like and less-porous structure was found in the C-DMBR while for the PAC-DMBR a more porous and less dense structure existed. Moreover, after backwashing, some residuals still remained on the mesh surface and the intersections of the mesh fibers, which could be

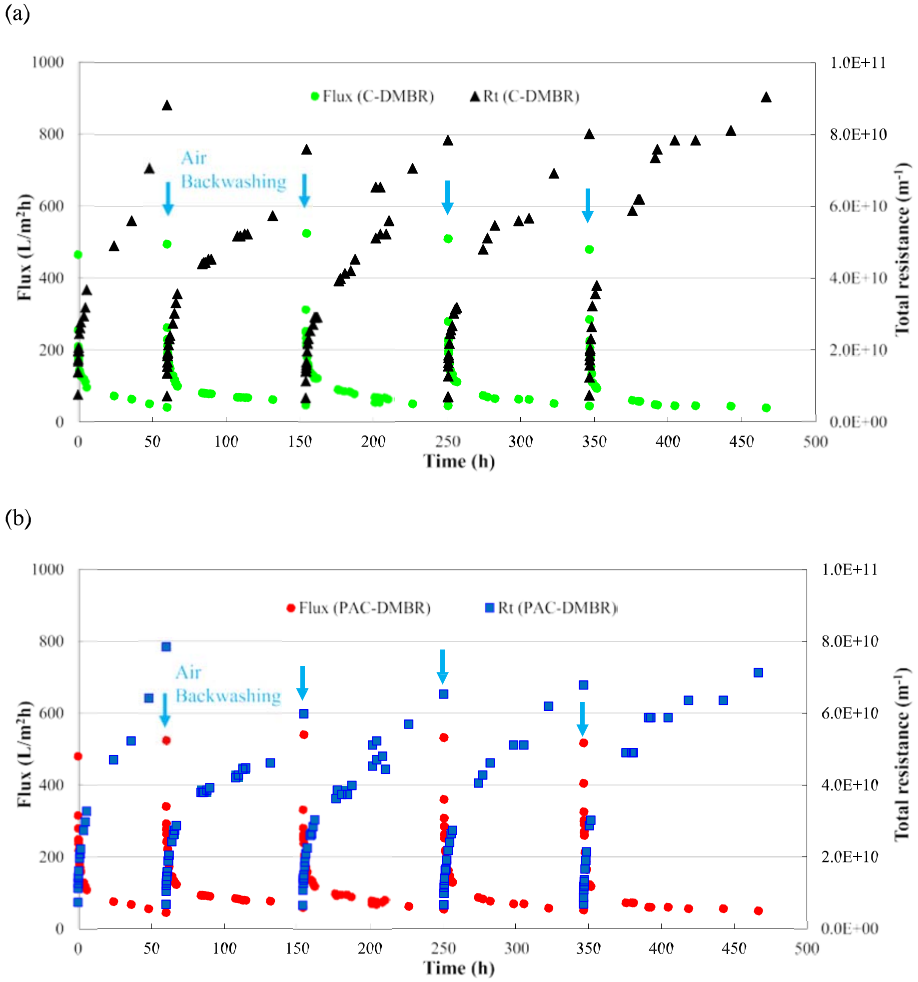

Fig. 2. Filtration performance of (a) C-DMBRs and (b) PAC-DMBR in terms of flux and total filtration resistance.

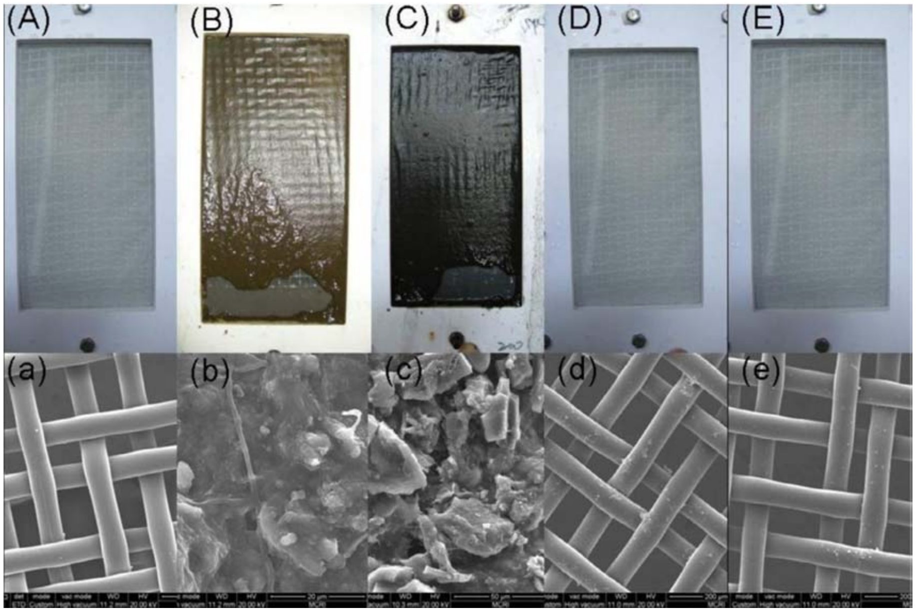

Fig. 3. Observation of membrane modules: (A) new membrane; (B) DM layer on the C-DMBR; (C) DM layer on the PAC-DMBR; (D) backwashed membrane in C-DMBR; (E) backwashed membrane in PAC-DMBR; (a) SEM of new membrane (x500); (b) SEM of DM layer on the C-DMBR (x5000); (c) SEM of DM layer on the PAC-DMBR (x2000); (d) SEM of backwashed membrane in C-DMBR (x500) and (e) SEM of backwashed membrane in PAC-DMBR (x500).

ignored for short term tests; but which however, would cause detrimental effects during long-term operation, as pointed out by other researchers [6,13]. A careful comparison indicated that less residual material existed on the mesh surface from the PAC-DMBR, and the potential reasons were as follows: (1) The concentrations of potential foulants such as EPS and SMP in the activated sludge were reduced due to the effects of the added PAC. (2) The PAC in the DM layer prevented contact between potential foulants and the mesh surface, which is discussed further later. Moreover, it was noted that the regeneration of the DM could be effectively implemented by physical cleaning (such as air back-washing) without using chemical reagents, which is in agreement with the results from previous DMBR studies [2,6]. Thus, easy cleaning of the DM layer by physical means for repeated cycles of regeneration and filtration could be one of the major advantages of the DMBR over conventional MBRs.

3.2. Analysis of the physicochemical properties

3.2.1. Morphological analysis

Observations by microscopy and PSD measurements were conducted to compare the differences in morphology between activated sludge and the DM layer in the DMBRs. Analyzing the images of sludge samples obtained by microscopy (shown in Fig. S4 in Supporting Information), it was found that the activated sludge and DM layer in the C-DMBR was smaller in particle size and also more loosely distributed. In contrast, the sludge samples from the PAC-DMBR had more compact structure and larger size, with the PAC particles being incorporated into or distributed around the sludge flocs. This verified the interaction among the PAC particles and sludge flocs, and also the formation of biological PAC sludge.

Moreover, the PSD was measured and is demonstrated in Fig. S5 in Supporting Information. The volume-based mean particle size of the activated sludge in the DMBRs was 50.7 μm and 52.8 μm, with more large particles existing in the PAC-DMBRs. However, the volume-based mean particle size of the DM layer in the two DMBRs was 62.6 μm and 73.5 μm, which were larger than their corresponding activated sludge particles. This is in accordance with previous results [6,7]. The potential physicochemical and biological interactions among the accumulated components and the compressibility of the DM layer caused the observed difference [18,31]. This is because the DM layer was a matrix containing various microorganisms, as well as organic and inorganic substances generated from the activated sludge. Therefore, the results indicated that PAC addition obviously modified the morphology of both the activated sludge and the DM layer.

Except for the sludge morphology and PSD, other properties (such as sludge flocculation, settling, and dewatering abilities) were also found to be enhanced to some extent, as reported in other references and in our previous study [7,10,13]. Moreover, the characteristics of biopolymers such as EPS and soluble microbial products (e.g., their quantity and quality related to the production, biodegradation, and accumulation process in the bioreactor) was an important concern affecting both effluent quality and DM layer filterability.

3.2.2. 3D-EEM and GPC analysis

As the fouling substances, EPS have attracted widespread attention both in MBR and DMBR studies. In this study, SEPS and BEPS in the activated sludge and in the DM layer from the two DMBRs were extracted and characterized by chemical analysis, EEM spectra, and the GPC method. The EPS concentrations are given in Table 2; generally BEPS concentrations were higher than those of SEPS, and proteins were the dominate components rather than polysaccharides. Furthermore, it was found that for SEPS samples, the concentrations ranked in the following order: sludge (C-DMBR) > sludge (PAC-DMBR) > DM layer (C-DMBR) > DM layer (C-DMBR). BEPS in different samples also showed a similar tendency. From Table 2, it is worth noting that lower content of proteins and polysaccharides in the SEPS samples from the DM layer than from the activated sludge, was observed for both bioreactors. This was due to the low retention effects of SEPS by the DM layer, and also to the degradation of part of the biodegradable organics in the DM layer under a substrate deficient condition [6]. A similar trend was also noted for the BEPS, and lower BEPS concentrations were detected in sludge samples from the PAC-DMBR than from the C-DMBR. This would be beneficial for DM filtration performance, because one recent study indicated that BEPS, and the polysaccharides in BEPS, had a significant impact on the fouling propensity of the DM layer [7].

In addition, previous studies reported that the EPS concentrations were lower after PAC addition, which was considered to be related to the adsorption and biodegradation effects of the biological PAC [10,16]. Moreover, researchers claimed that the two mechanisms played different roles in organics adsorption related to the adsorption capacity of PAC. Because during the initial stage after PAC addition adsorption was the main reason for organics reduction, while the period after the biological PAC formation, biodegradation and regeneration seemed to be more important (than adsorption) for organics removal [28,32]. Therefore, it should be noted that during long-term operation of PAC-DMBR system without continuous PAC addition, adsorption and biodegradation effects would both contribute to SEPS reduction, although different behaviors and significance of the abovementioned mechanisms could be expected.

EEM spectroscopy was used to characterize the fluorescent organic substances in the EPS samples from the DMBRs. As shown in Fig. 4, three fluorescence peaks: namely peak A (230 nm/310–350 nm), peak B (275–290 nm/335–370 nm), and peak C (310–350 nm/405–440 nm), respectively, were observed in the BEPS samples, while peak C was negligible in the SEPS samples. Peak A represented aromatic protein-like substances, peak B reflected tryptophan protein-like substances, and peak C showed the existence of humic acid-like substances [33]. The SEPS in the DM layer mainly stemmed from the biopolymers retained during the sludge filtration process and was not closely coupled within the DM layer; thus, organics with higher molecular weight could be expected in SEPS. On the other hand, BEPS were derived from the attached fine flocs, colloids, biopolymers and other organics, which were tightly bound within the DM layer by the complex interactions between various foulants (such as microorganisms, organics and inorganics) as documented by many researchers [5,18]. Moreover, certain amounts of humic-acid substances were noted in BEPS rather than in SEPS, which could be largely related to their different original sources. BEPS had a broader origin, including the decay and lysis of the incorporated microorganisms and the metabolism products during substrate utilization, which would surely produce biomass-stemmed humic-acids [34].

In addition, through GPC analysis (shown in Fig. 5) of SEPS samples in the DM layer in the DMBRs, MWD profiles were revealed with similar elute time (7–17 min). However, these were different from those of the BEPS samples, which showed a broader distribution range (7–25 min) covering the micro, high, intermediate, and low MW organics [10]. Surely, the retention effect of the DM layer on various potential foulants such as fine particles, colloids, SEPS, and so on that exist in activated sludge, could increase the concentrations of large MW organics in BEPS. At the same time, the interactions between the aforementioned foulants and inorganic foulants (multivalent metal ions) would further increase the molecular weight of accumulated organics [18]. The results pro-

Table 2
EPS concentration in the sludge samples.

Sludge samples	SEPS (mg/gMLSS)			BEPS (mg/gMLSS)		
	PS	PN	Total	PS	PN	Total
Sludge (C-DMBR)	1.14	6.08	7.22	8.27	13.31	21.58
Sludge (PAC-DMBR)	0.58	3.81	4.39	4.41	9.86	14.27
DM layer (C-DMBR)	0.56	1.82	2.38	3.09	14.03	17.12
DM layer (PAC-DMBR)	0.59	0.53	1.12	1.02	6.99	8.01

Note: PS and PN mean polysaccharides and proteins; the number of measurements: n = 5.

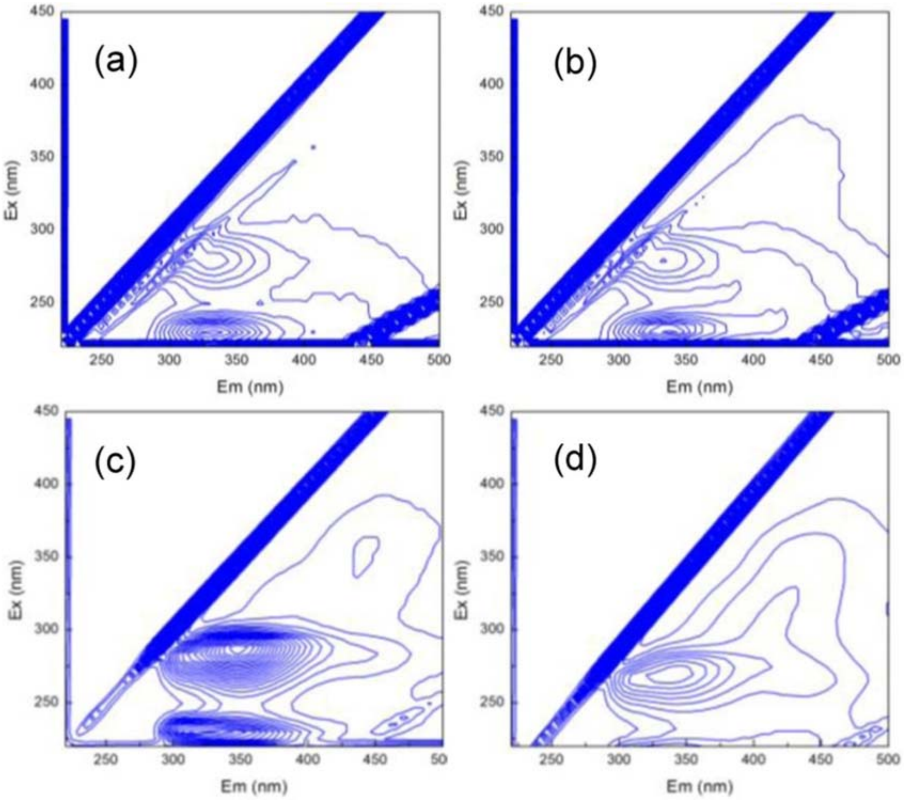

Fig. 4. EEM fluorescence spectra of EPS samples extracted from the DM layer in DMBRs: (a) SEPS (C-DMBR); (b) SEPS (PAC-DMBR); (c) BEPS (C-DMBR) and (d) BEPS (C-DMBR).

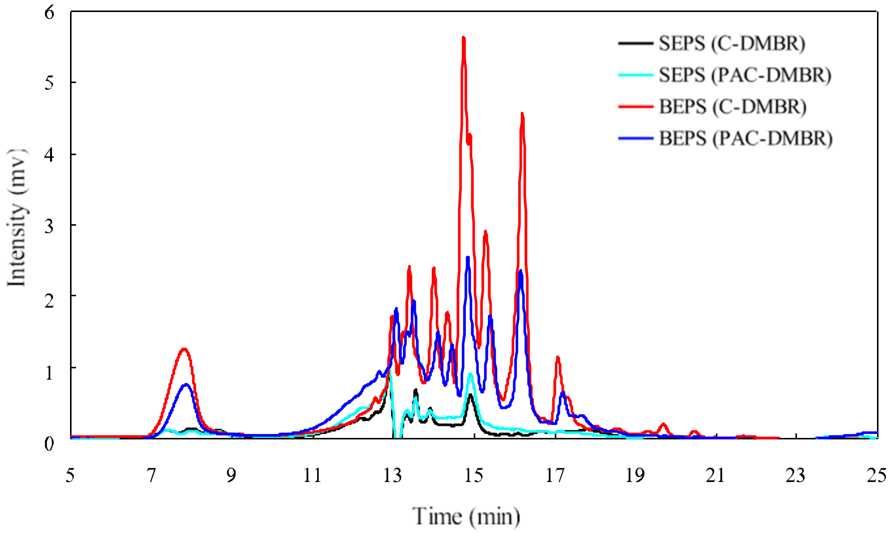

Fig. 5. MWD curves of EPS samples extracted from the DM layer in DMBRs.

vided useful information on EPS properties in the DM layer after PAC addition regarding their sources, accumulation behavior, and potential interaction mechanisms.

3.2.3. ATR-FTIR and EDX analysis

ATR-FTIR and EDX analysis were carried out to characterize the organic and inorganic components in the DM layers from the DMBRs. First, Fig. 6 presents the FTIR spectra of the samples, and it was noted that the profiles are similar. In detail, the peaks at (3240, 2900 and 986) cm^{-1} are attributed to the presence of polysaccharides or polysaccharide-like substances in MBRs [35]. The characteristic peaks for proteins included those at (1624, 1521 and 1420) cm^{-1} [36]. The other organic substances identified included aliphatic substances (peaks near 2900 cm^{-1}), fat and/or cellulose (peaks near 1411 and 1239 cm^{-1}) [37]. Based on the analysis, the major organics in the DM layer were identified as proteins and polysaccharides. The results confirmed the existence of biopolymers (such as EPS) in the DM layers, as recorded in the above EPS extraction and contents analysis.

Fig. 7 illustrates the main inorganic elements in the DM layer revealed by EDX analysis. As shown, the following elemental composition was detected in the DM layer from the C-DMBR system, and the relative weight percentages were as follows: 33.4% C, 13.6% N, 42.7% O, 0.5% Na, 0.8% Mg, 1.0% Al, 1.7% Fe, 2.2% Ca, 2.4% Si, 1.0% P, and 0.8% S. There were some differences found in the DM layer from PAC-DMBR: 56.2% C, 33.87% O, 0.7% Na, 0.2% K, 0.7% Mg, 0.6% Al, 2.3% Fe, 2.1% Ca, 1.93% Si, and 1.37% S. Obviously, the relative content of C was much higher in the DM layer from the PAC-DMBR (56.2%) than from the C-DMBR (33.4%), indicating the contribution of PAC to the DM layer formation in the PAC-DMBR. In addition, as reported, although the content of the accumulated elements (especially for Mg, Al, Fe, and Ca) were less than that of other foulants (fine particles and biopolymers), they would enhance the cake layer formation through charge neutralization and the bridging effect due to synergistic interactions among various foulants [18,31]. FTIR and EDX analysis indicated that organic and inorganic substances existed in the DM layers, while PAC addition resulted in a high carbon-content DM layer that might further affect the filterability of the DM layer in PAC-DMBR for the following two reasons: (1) the difference in the DM layer structure due to the biological PAC formation and attachment; (2) the difference in the biomass surviving in the PAC sludge and activated sludge from the PAC-DMBR and C-DMBR, and their different abilities to adapt to the DM layer environment (attached growth). Obviously, more effort is still needed to investigate further the interactions among various foulants and their importance in DM layer formation and regeneration. More importantly, the addition of PAC might alter the complicated interactions.

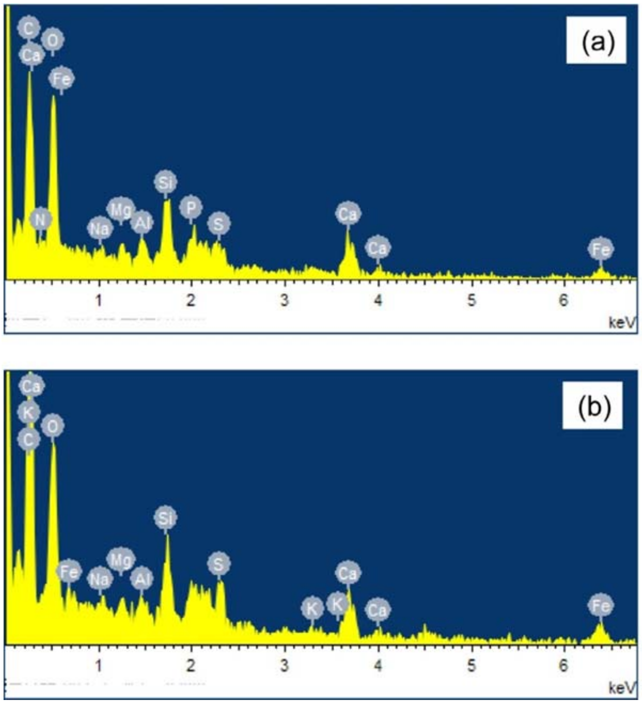

Fig. 7. EDX analysis of DM layer samples from: (a) C-DMBR and (b) PAC-DMBR.

3.3. Microbial properties analysis

As known, the added PAC in the bioreactor could function as an adsorbent for organics (such as biopolymers and slowly biodegradable compounds) and serve as a fixed surface for the attachment of

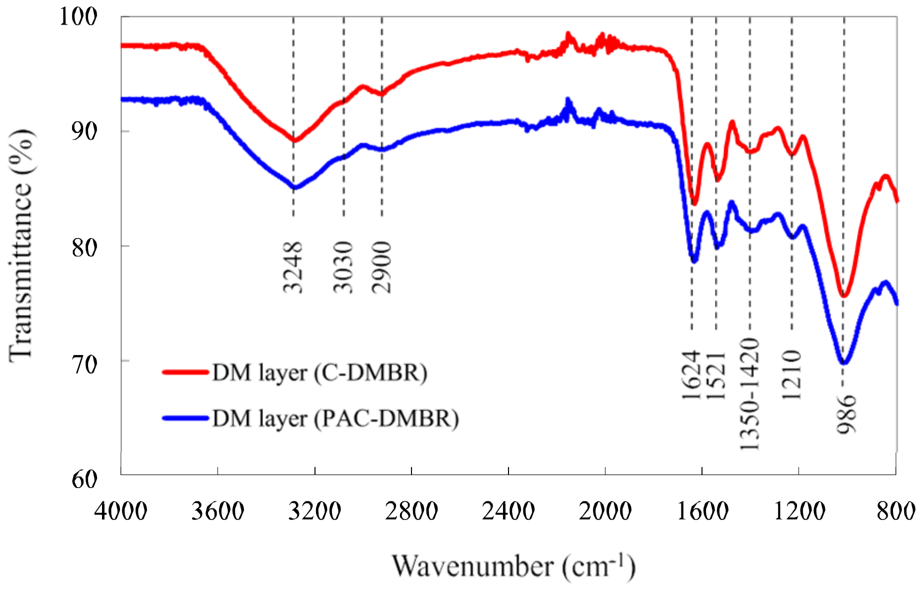

Fig. 6. ATR-FTIR spectra of DM layer samples.

growing biomass to form biological PAC. It could also contribute much to the formation of the DM layer in DMBRs. In this case, the microbial properties of the activated sludge and DM layer were expected to be altered after PAC addition; so it was meaningful to investigate the microbial properties using advanced analytical techniques (such as the high-throughput pyrosequencing technology).

To better understand the structures of the microbial communities in the two DMBRs, the relative abundances at the phylum and genus level for sludge samples was calculated, and are shown in Fig. 8. From Fig. 8(a), at the phylum level the community composition showed high diversity and certain similarity in the sludge samples: ten different phyla were generally observed in this study. The most abundant phylum in all the samples was Proteobacteria, which was followed by Planctomycetes, Bacteroidetes, Chloroflexi, Verrucomicrobia, Firmicutes, Actinobacteria, and other phyla. However, the percentages (relative abundance) obtained for these phyla were quite different. In the activated sludge from the C-DMBR, Proteobacteria, Planctomycetes, Bacteroidetes Chloroflexi, and Firmicutes accounted for (74.7, 11.4, 4.6, 2.2, and 1.6)%, respectively. For the same phyla, the percentages were (71.0, 11.7, 8.6, 2.2, and 1.0)% for the DM layer from the C-DMBR; (68.6, 7.1, 9.5, 2.3, and 8.1)% for the activated sludge from the PAC-DMBR; and (64.3, 8.2, 14.3, 2.5, and 5.4)% for the DM layer from the PAC-DMBR. These results were consistent with results of previous studies that claimed Proteobacteria was the dominant phylum in activated sludge and biofilms, and also that Bacteroidetes, Chloroflexi, and Firmicutes were commonly detected phyla [24,25].

In addition, it was reported that members of Proteobacteria contributed to the cake layer formation and functioned as pioneers in the surface colonization of membranes in the MBR system. Members of Bacteroidetes were involved in the degradation of protein N-acetylglucosamine and chitin, and were proficient in degrading part of the high molecular mass fraction of the DOM. Members of Chloroflexi were responsible for the degradation of soluble microbial products; and Firmicutes was related to metabolism of the complex organic matrix [23,24,38,39]. Therefore, the reduction of Proteobacteria and increase of Bacteroidetes, Chloroflexi and Firmicutes in the PAC-DMBR system might benefit the mitigation of several fouling issues. On the other hand, the enhancement of organic pollutant removal and biopolymer degradation in the PAC-DMBR could also be expected, although their performance in the treatment of wastewater containing complicated organics (such as industrial wastewater) needs further investigation and verification.

Further analysis was carried out at the genus level to compare bacterial communities present in the DMBRs. As shown in Fig. 8 (b), bacterial diversities were similar in the activated sludge and the DM layer collected from the same bioreactor, showing that the microbial communities present in the DM layers originated from the bulk sludge. Although the composition of the bacterial communities was similar, the abundances of each genus were dif-

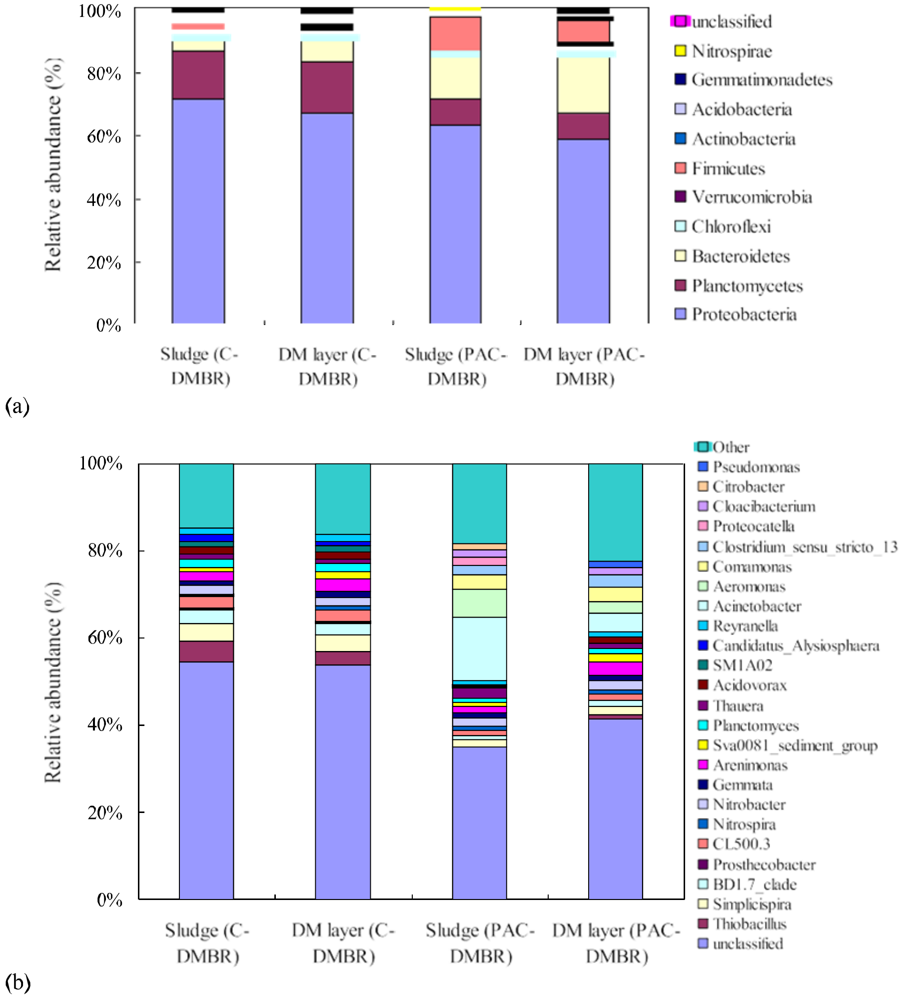

Fig. 8. Microbial community analysis of sludge samples in the DMBRs at the (a) phylum level and (b) genus level.

ferent in the activated sludge and the DM layer. This was related to differences in the morphology and structure of the two types of sludge, resulting in different living environments for the bacteria. Moreover, by further comparison between the sludge samples from C-DMBR and PAC-DMBR, it was noted that after PAC addition, the abundance of bacterial communities increased: 18 genera were detected in the C-DMBR and 23 genera were detected in the PAC-DMBR. Three genera (including Prosthecobacter, SM1A02, and Candidatus Alysiosphaera) detected in the C-DMBR were absent from the sludge samples of the PAC-DMBR; however, another eight genera (Acinetobacter, Aeromonas, Comamonas, Clostridium_sensu_stricto_13, Proteocatella, Cloacibacterium, Citrobacter, and Pseudomonas) were only found in the sludge samples from the PAC-DMBR.

It was interesting to find out that Acinetobacter was a genus belonging to the wider class of Gammaproteobacteria, which are non-motile and which accounted for about 15.6% and 4.2% in the activated sludge and DM layer in the PAC-DMBR, but were not detected in the C-DMBR samples. This indicated that PAC could interact with the biomass and provide fixed surfaces for microbial growth as a bio-carrier. Moreover, Acinetobacter was a genus useful for degradation of hydrocarbons (especially of aromatic compounds) [40], which would enhance the removal of relatively complex organics from the PAC-DMBR system. This was detected in a similar hybrid DMBR system treating industrial wastewater [16]. Moreover, some genera (such as Aeromonas, Clostridium_sensu_stricto_13, Proteocatella, Cloacibacterium, and Citrobacter) detected in the PAC-DMBR were types commonly found to be facultative anaerobic or anaerobic, which means that in the biological PAC particles, anaerobic conditions were created due to limitations of oxygen and substrate.

The results showed that the diversity of bacterial communities increased after PAC addition, while the composition of the bacterial communities obviously shifted due to adaption of the bacteria to the PAC-assisted environment. From this perspective, the 454 high-throughput pyrosequencing method was useful in revealing the significant effects of PAC addition on the microbial community structure and on the microbial composition in the DMBRs, and could potentially explain the differences in performance aforementioned, between the C-DMBR and PAC-DMBR.

3.4. Practical applicability of the hybrid DMBR process

In this study, the effects from PAC addition on the filtration performance and various DM layer properties in a PAC-DMBR were systematically investigated. It was found that during the operation period with a high stable flux (50–100 L/m^2 h) from gravity-driven filtration, the PAC-DMBR showed enhanced pollutant removal and filtration performance, quicker DM formation, and better DM layer regeneration after physical cleaning. The preliminary views of the underlying enhancement mechanisms of PAC addition were discussed. The added PAC seemed first to modify the activated sludge properties by adsorption and biodegradation of organics and by formation of biological PAC, and then also changed the microbial community diversity in the activated sludge. The variation in sludge properties resulted in the DM layer from PAC-DMBR showing a different structure (more porous and incompressible) compared with that in the C-DMBR. It was thought to be related to the observed changes in filtration behaviors of the two DMBRs. Based on the experimental results and analysis, a schematic diagram of the formation of the DM layers in the DMBRs is presented in Fig. 9. It is worth noting that the filtration process of the DM layer was actually a dynamic process involving several stages (dynamic formation, stable operation, and cleaning for regeneration); thus the properties of the DM layer (such as the morphology and structure) would also be expected to change with time and be affected by the operation period (such as startup or stable operation period). Fig. 9 is useful for better understanding the properties of the formed DM layer during steady operation period. However, to well present the temporal and spatial variations of the DM layer, more investigation will be needed in the future.

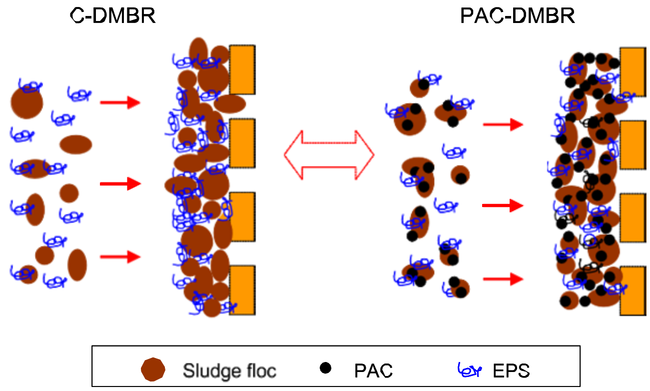

Fig. 9. Schematic diagram representing the DM layer formation in the DMBRs.

On the other hand, the practical applicability of the hybrid DMBR process is an important issue of concerned to researchers, engineers, industrial producers and so on. Therefore, there is a need for further discussion to clarify the limitations, solutions, and prospects of hybrid DMBRs. First, considering the technical aspects, the main concerns relate to pollutant removal, filtration ability, and DM cleaning strategies. The removal of various pollutants in the hybrid DMBRs was nearly the same as for the MBR process during stable operation periods; however, during the initial DM formation stage, the effluent quality was commonly poor due to the passage of sludge flocs into the permeate. It is widely recognized that in hybrid DMBRs (such as the PAC-DMBR), pollutant removal could be enhanced to some extent due to the adsorption and biodegradation effects of added adsorbents/bio-carriers. The treatment objects included industrial wastewater, municipal wastewater, and slightly polluted surface water [2,6,14]. As for the filtration performance, previous results indicated that the flux in DMBRs was generally higher than that in UF/MF in conventional MBRs, in constant flux or constant pressure modes [2,5,13]. However, the integrity of the supporting mesh in DMBRs might be not as high as that of the hollow fiber/flat-sheet UF/MF membranes, so the affordable gravity water head or TMP would be limited to a relative low value. In terms of the cleaning strategy, it was indicated that physical cleaning or low frequency chemical cleaning was sufficient for recovery of DM permeability. Thus, the above analysis demonstrated that the hybrid DMBR process even showed some technical advantages over the MBR process, although how to stabilize the effluent quality still needs more investigation.

Second, from the economic aspect, attention should be paid to the cost of the DM module, various additives, the energy demand for aeration, and permeate production. In DMBRs, the DM module cost could be much reduced by using coarse-pore supporting meshes, although the selection of high integrity materials would increase the cost. The demand for aeration in the DMBR was less than that in MBRs, because less shear force was needed to scour the DM surface to maintain a relatively stable DM layer for the solid-liquid separation [6]. For various additives like PAC, one time addition could be used for a long time before the next replenishment although the gradual loss of additives indeed occurred, thus selecting appropriate additives (low-cost and highly durable) would make a negligible cost increase during the life cycle of the hybrid DMBRs. Based on the literature results, it was easy to find out that gravity-driven and pump-driven approaches were both commonly used for permeate production [2]; however, to obtain the same flux, the water head or TMP in DMBRs was much lower than that in MBRs due to the smaller intrinsic mesh resistance and cake layer filtration resistance.

Last, regarding the status of current research and application, it should be noted that more effort is still needed to resolve the following two problems. One thing is that recent work has mainly been conducted in lab-scale DMBRs, while studies implemented in pilot-scale DMBRs are quite limited [2]. For this reason, the aforementioned technical and economic analysis should be further verified and evaluated in large-scale or full-scale DMBRs. The other problem is that, some limitations still exist in the design, operation, and modeling of hybrid DMBRs. These are listed and discussed below. It is needed to ensure the intensity of the DM module by optimizing the selection of supporting material and design of the DM module configuration. It is needed to realize quick formation of the DM layer and to prolong the stable operation period through control of the operation parameters and the sludge properties related to the microbial community. It is needed to develop an efficient modeling tool involving the cake layer filtration properties of DMBRs to guide the design and operation of DMBRs. For the absorbent-assisted hybrid DMBRs, the saturation of the adsorption capacity and its loss in permeate after DM cleaning should be properly resolved by periodical replenishment of absorbent and by other methods.

In all, based on the great research efforts and abundant achievements regarding the DMBR process, it is concluded that the hybrid DMBR process is technically and economically feasible although more work is still needed to optimize the design, operation and modeling of hybrid DMBRs to fulfill the large-scale practical application.

4. Conclusions

A PAC-DMBR was developed and compared with a C-DMBR to investigate the filtration performance and characteristics of the DM layer using various analytical methods. During stable operation, the PAC-DMBR showed better pollutant removal (COD, UV_{254}, NH_3, and color) and filtration performance (higher flux and lower filtration resistance). PAC addition first modified the sludge properties (morphological, component, and microbial properties) to facilitate biological PAC formation, and then resulted in a DM layer with more porous and incompressible structure in the PAC-DMBR. This resulted in the better filtration behavior of the DM layer. The pyrosequencing results further indicated that the performance enhancement could be attributed to enriching some specific microbial genera able to biodegrade complex organics and reducing the abundance of several microbial genera regarded as pioneers in the colonization of the membrane surface. Evaluation of its practical applicability shows that the hybrid DMBR is a promising process for large-scale wastewater treatment although more effort is still needed to address the encountered limitations.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cej.2017.06.072.

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