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Development of a novel two-stage powdered activated carbon-dynamic membrane filtration (PAC-DMF) system for direct physicochemical wastewater treatment

Sha Li[a], Yuan Yang[a], Yisong Hu[a,b,*], Jialing Tang[a], Xiaochang C. Wang[a,b,c,*]

[a]*Key Lab of Northwest Water Resource, Environment and Ecology, MOE, Xi'an University of Architecture and Technology, Xi'an 710055, China, Tel. +8602982205652; emails: yshu86@163.com (Y. Hu), lililimomos@163.com (S. Li), 383842205@qq.com (Y. Yang), 921221788@qq.com (J. Tang)*
[b]*International Science & Technology Cooperation Center for Urban Alternative Water Resources Development, Xi'an 710055, China, Tel. +8602982205841; email: xcwang@xauat.edu.cn (X.C. Wang)*
[c]*Key Lab of Environmental Engineering, Xi'an University of Architecture and Technology, Shaanxi Province, Xi'an 710055, China*

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ABSTRACT

Due to the issues of high energy consumption, high carbon emissions, and low resource and energy recovery, there is an urgent need to develop new paradigms for simultaneous wastewater treatment and resource utilization using physicochemical processes rather than conventional biological means. Recently, direct membrane filtration of municipal wastewater using ultrafiltration/microfiltration technology was proposed and showed satisfactory performance, with high membrane costs and fast membrane fouling as the main limitations. In this study, a novel two-stage powdered activated carbon-dynamic membrane filtration (PAC-DMF) system was developed using PAC as a carrier to enhance DM formation and organic pollutant retention. The particle size and dosage of PAC, strategy for operating the continuous reactor in the first stage, and selection of the PAC particle size in the second stage were investigated. The effluent from the first stage was found to have a stable water quality, with turbidity in the range of 50–70 NTU. Efficient retention of organic materials was also achieved with a retention rate of 74%–82%, and the COD content in the concentrate was above 3,000 mg/L in the cycle filtration experiment. In the second stage, further enhancement of water quality and organics retention were observed, as evidenced by the low effluent COD (<70 mg/L) and turbidity (5–20 NTU). Meanwhile, the effluent that was rich in nitrogen and phosphorus was suitable for agricultural irrigation or gardening. The results indicated that the developed two-stage PAC-DMF system could be a highly promising alternative to the conventional wastewater treatment process.

Keywords: Powdered activated carbon; Dynamic membrane; Wastewater direct filtration; Resource recovery; Membrane fouling

1. Introduction

Currently, biological wastewater treatment technologies (such as conventional activated sludge processes and various biological nutrient treatment processes) are intensively applied to provide effluent that meets the increasingly stringent discharge standards. However, these processes have several disadvantages, including high energy consumption and carbon emissions, which are mainly caused by aeration and the large-scale footprint [1], and low resource and energy recovery, due to the mineralization of organic matter and the conversion and elimination of nutrients (nitrogen and phosphorus) with low environmental capacities [2]. Thus, it is urgent to develop novel and sustainable strategies

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* Corresponding author.

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that can simultaneously provide wastewater treatment and resource utilization using short-cut technologies (such as the physicochemical process) rather than conventional biological means [3] to help combat the global energy crisis and climate change.

Recent attention in the area of wastewater treatment has focused on minimizing energy consumption or achieving net energy production; recovering nutrients, such as phosphorus and nitrogen; and reclaiming treated water using various methods [4,5]. Organic recovery as a pre-step was proposed to enhance energy recovery. The highly concentrated organics from the wastewater could then be used for energy production, such as biogas generation through anaerobic digestion (AD) or bio-electrochemical techniques for electricity generation and storage [6–9]. The remaining nitrogen and phosphorus in the wastewater could be recovered through energy efficient processes to produce value added fertilizer [10].

Therefore, a pre-step that could enhance the concentration of organic materials and facilitate nutrient recovery was deemed appropriate. Direct membrane filtration of municipal wastewater using various membrane filtration technologies (such as ultrafiltration [UF]/microfiltration [MF]) was proposed and investigated by several researchers [11,12]. Direct membrane filtration in a dead-end process with chemically enhanced backwash (CEB) showed satisfactory performance for pre-concentration of the organic material, with a final COD of 7,000 mg/L in the concentrate after 200 h of operation [11], while frequent CEB adopted for the MF membrane fouling control caused organic matter loss by chemical oxidation. In another work, a coagulation/adsorption assisted direct sewage MF membrane filtration system showed efficient pre-concentration of organics (COD in the concentrate 7,000 mg/L after 6 d of operation) by using continuous coagulant addition and one-time adsorbent addition for fouling mitigation [12]. Recent works focused on the optimization of the UF/MF membrane-based organics pre-concentration system by optimizing and coupling the following operation strategies, including the coagulation process, intermittent aeration and air backwash [4,6,10,12], for effective fouling control and performance enhancement. However, high membrane costs and serious membrane fouling were still the main technical bottlenecks that limit the widespread application of such technologies.

The emerging dynamic membrane (DM) filtration technology might be useful in solving the aforementioned issues. DM, also called the secondary membrane, is formed on an underlying support material (such as coarse pore mesh and filter cloth) when the filtered solution contains suspended solid particles, such as microbial cells and flocs [13]. Organic materials and colloidal particles, which normally result in fouling of the membrane, will be trapped in the DM layer, thus the mesh pore blocking is negligible. Additionally, the low membrane module cost, high flux and easy backwash made DM technology an attractive alternative compared with the membrane bioreactor (MBR) in biological wastewater treatment, both aerobically and anaerobically (the so called dynamic membrane bioreactor, DMBR) [14–17]. To date, quite limited work has been conducted using DM technology for direct wastewater filtration without using biological degradation, which was the main mechanism for the removal of pollutants in the DMBRs. Only one study using a double-layer membrane module (50 μm Dacron cloth as the inner layer and 1 μm propene polymer cloth as the outer layer) reported that COD could be concentrated to 4,500 mg/L in 70 h, but that the trans-membrane pressure (TMP) quickly increased to 80 kPa in less than 10 h at a flux of 2–10 L/m^2 h [18]. Other previous studies focused on salt rejection and industrial wastewater treatment (such as oily and textile wastewater) using microfiltration, ultrafiltration, reverse osmosis, and nanofiltration membranes as supporting materials [13], which is different from the abovementioned low-cost DM technology. Therefore, the above analysis indicates that much effort is still needed to design, optimize, and apply the DM-based wastewater direct filtration process.

Granular carriers (such as kaolin, diatomite, and powdered activated carbon [PAC]) have also been recognized to enhance the dynamic membrane formation process and shorten its formation time, and also to modify the physicochemical structure and lower the filtration resistance [15]. On the other hand, some granular carriers those have large specific surface areas, such as PAC, showed superior adsorption effects on dissolved and colloidal substances [19]. Thus, PAC was used as a potential DM formation granular and adsorbent and coupled with dynamic membrane filtration technology to develop a two-stage powdered activated carbon-dynamic membrane filtration (PAC-DMF) system for direct wastewater treatment using physicochemical means. The objective of this study was to optimize various operation factors (including particle size and dosage of PAC, DM backwashing, and mixed liquor sieving) to achieve the stable operation of the two-stage PAC-DMF system for municipal wastewater treatment and resource recovery.

2. Materials and methods

2.1. Characteristics of PAC, wastewater, and membrane module

The raw wastewater was obtained before the coarse screen in a local domestic wastewater treatment plant in Xi'an, and then was screened by a 5-mm sieve. The characteristics of the wastewater are listed in Table 1. PAC (Aladdin Biological Technology Co., Ltd., Shanghai, China; CAS No.: 7440-44-0) with a particle size of 20–50 mesh and density of 0.25–0.60 kg/L was ground and sieved to obtain different particle sizes: the large (diameter of 200–500 μm), the medium

Table 1
Characteristics of the domestic wastewater

Parameters	Value
Temperature (°C)	18.9 ± 0.2
Turbidity (NTU)	120 ± 30
pH	8.1 ± 0.2
COD (mg/L)	269.8 ± 83.3
UV_{254} (cm^{-1})	0.24 ± 0.08
NH_4^+–N (mg/L)	24.7 ± 11.6
TN (mg/L)	44.1 ± 14.1
TP (mg/L)	4.7 ± 1.8
Chroma (c.u.)	333 ± 106

(diameter of 100–200 μm), and the small (diameter less than 100 μm) samples. A flat-sheet dynamic membrane module was made of nylon mesh (pore size of 30 μm or 50 μm) with a double-side filtration area of 0.02 m^2, and the support frame of the dynamic membrane was made of plexiglass.

2.2. Experimental setup and operating conditions

The schematic diagram of the two-stage PAC-DMF system is shown in Fig. 1. Two identical reactors were used in the first and second stages of the PAC-DMF system. The reactor was rectangular (length × width × height = 11 cm × 6 cm × 38 cm) and made of plexiglass, with a working volume of 1.1 L. Magnetic stirring devices were used to blend the mixed liquor evenly at a speed of 150–200 rpm. The dynamic membrane modules were submerged into the reactors. An influent pump connected to a liquid level sensor was used to maintain a constant water level in each reactor during the experiment. The membrane-filtered effluent was extracted by a peristaltic pump (BT-100 Longer, USA). The initial flux was set to 50–60 L/m^2 h in both reactors. A pressure gauge and a paperless recorder were connected to monitor the TMP value. During the operation period, although the constant flux mode was chosen for the DM operation in the two-stage reactors, a slight decrease in the filtration flux (reduction less than 20% of the initial flux) was noted, accompanied by an increase in the TMP.

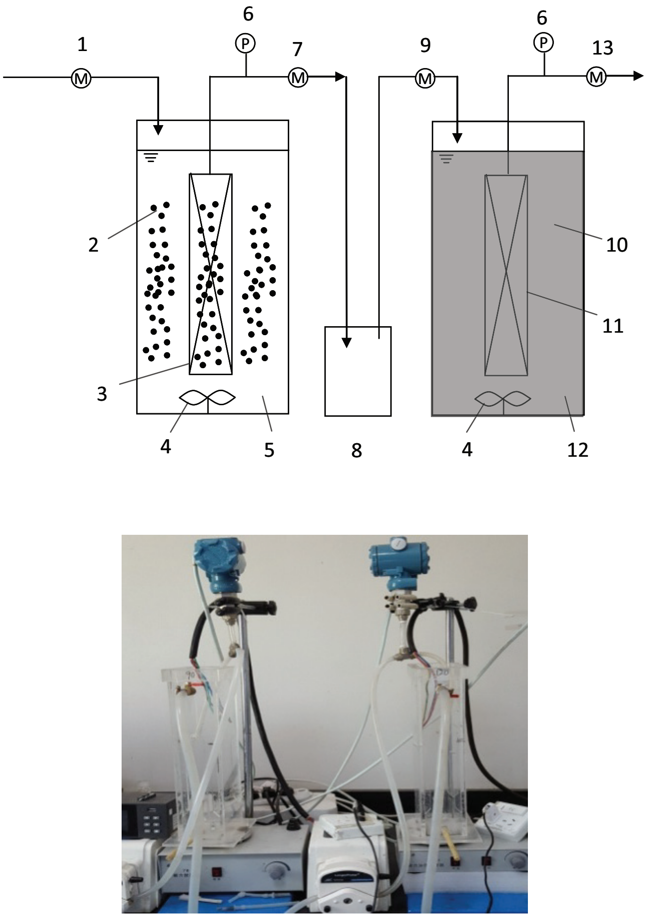

Fig. 1. Schematic diagram of the two-stage PAC-DMF reactor system (left) and picture of lab-scale reactor (right). (1. Influent pump; 2. large size PAC; 3. membrane module; 4. magnetic stirrer; 5. first stage reactor; 6. pressure gauge; 7. effluent pump; 8. effluent tank; 9. influent pump; 10. small size PAC; 11. membrane module; 12. second stage reactor; 13. effluent pump.)

The indicators to stop one operation cycle were set as follows: (1) the flux decreases to 80% of the initial flux; (2) the TMP exceeds 15 kPa; (3) the turbidity of the effluent increases rapidly. All the above items indicate that the permeability of the dynamic membrane has been reduced, and stable operation cannot be maintained. Thus, air backwash was used to regenerate the DM for 2 min with a gas flow rate of 70 L/min through the effluent pipe, but in a reverse direction, resulting in the air passing the DM module in an inside-outside mode. Then, the nylon mesh was manually cleaned with a soft brush. The same backwash method was applied in the two-stage reactors for DM regeneration.

2.3. Experimental design

2.3.1. Optimization of the first stage PAC-DMF reactor

2.3.1.1. Selection of PAC dosage Three identical reactors were operated simultaneously for selection of the PAC dosage. A nylon mesh with a pore size of 50 μm was chosen as the DM supporting material, and the dosages of PAC (particle size of 100–200 μm) were tested at 1, 2, and 3 g/L. Single cycle filtration tests were performed to determine an appropriate PAC dosage based on the process performance.

2.3.1.2. Selection of PAC particle size Three identical reactors were running in parallel. The pore size of the nylon mesh was 50 μm. The PAC dosage was set to 2 g/L. PAC with particle sizes less than 100 μm, 100–200 μm, and 200–500 μm were used in single cycle tests to determine the optimal PAC particle size.

2.3.1.3. Backwashing and strategy for long-term operation During the filtration experiments, after backwashing, the DM layer was not easily re-formed due to a large amount of retained substances that were re-suspended in the reactor, which caused high turbidity of the effluent and a quick increase of TMP, resulting in a great challenge for the continuous multi-cycle operation. Thus, an operation strategy was used to investigate the feasibility for long-term system operation. In detail, the concentrated liquor after backwashing was obtained and screened manually using a commercial stainless steel sieve of 100 μm to recover the retained PAC for the next cycle of operation. After DM cleaning, the concentrate and the sieved liquid were sampled for subsequent analysis. In the tests, a nylon mesh with a pore size of 50 μm was used, and the PAC dosage (100–200 μm) was set to 2 g/L.

2.3.2. Optimization of the second stage PAC-DMF reactor

Two identical reactors treating the effluent from the first stage reactor were operated simultaneously to select an appropriate PAC particle size (less than 100 μm and

100–200 μm). A nylon mesh with pore size of 30 μm was chosen as the supporting material, and the PAC dosage was set at 1.5 g/L according to our previous results.

2.3.3. Performance of the two-stage PAC-DMF system during long-term operation

After optimizing the operation parameters of the two-stage PAC-DMF system, long-term operation was conducted to investigate the system performance. The optimized parameters were determined as follows: at the first stage, the pore size of the nylon mesh was 50 μm, and the particle size of the PAC was 100–200 μm at a dosage of 2 g/L, air backwashing was used to regenerate the membrane, then, the concentrate was sieved and the retained PAC was reused; for the second stage, the nylon mesh had a pore size of 30 μm and the particle size of the PAC was less than 100 μm at a dosage of 1.5 g/L.

2.4. Analytical methods

Water quality parameters, including COD, NH_4^+–N, total nitrogen (TN), total phosphate (TP), chroma and UV_{254}, were determined according to the Standard Methods [20]. pH was measured by a pH meter (sensION1, HACH, USA). Turbidity was measured using a turbidity meter (2100Q, HACH, USA).

3. Results and discussion

3.1. Factors affecting the operation of the first stage PAC-DMF reactor

3.1.1. Particle size of the PAC

The effects of PAC with different particle sizes on the TMP and effluent turbidity in the first stage reactor were investigated (Fig. 2). During the operation of the DM cycle, three PAC types with different particle sizes (<100 μm, 100–200 μm, and 200–500 μm) could effectively promote the formation of a dynamic membrane on the mesh surface, which was evidenced by the fact that the effluent turbidity decreased to 30 NTU in a short time and remained relatively stable afterwards. However, when adding the PAC with particle sizes less than 100 μm, the TMP increase rate was significantly higher than that when adding other two types of PAC. The reason could be that PAC particles with size less than 100 μm showing a mean particle size of 23.9 μm, which was close or even smaller than the membrane pore size (50 μm), thus large amount of smaller PAC particles would be lost during the initial DM layer formation stage by retaining larger PAC particles; however the DM layer formed by PAC with particle sizes less than 100 μm would be denser with less porosity compared with other two types of PAC, resulting in the quick TMP increase [21]. This was consistent with the Carman-Kozeny equation and previous studies, which reported that particles with smaller size would more easily attach to the membrane surface, resulting in less porosity, a denser structure, and higher filtration resistance of the fouling layer [22,23].

In addition, when adding larger PAC (200–500 μm), the effluent turbidity was slightly lower than that of adding the smaller PAC (100–200 μm). However, the long-term operation of the first stage reactor with air backwash of the DM cleaning indicated that the performance of the dynamic membrane regeneration was better when adding smaller PAC (100–200 μm). During the second cycle operation after DM cleaning, when the smaller PAC (100–200 μm) was added, the growth rate of the TMP was slower than that of adding larger PAC (200–500 μm), and the turbidity of effluent decreased rapidly and remained stable afterwards. This occurs because the large particle size PAC (200–500 μm) could not easily adhere to the membrane after the first cycle operation, and the time it took to form the DM would be prolonged [24]. Previous studies stated that the movement of particles towards the membrane surface was determined mainly by the permeation drag force and the shear force induced by cross-flow velocity, meanwhile particles with larger sizes encountered higher shear force compared with the smaller particles, thus larger particles would be more easily detached from the membrane surface [23,25]. In this case, smaller particle in the mixed liquid would form the DM layer prior to the large size PAC, resulting in the rapid growth of TMP [24,26]. Therefore, by comparison, it was noted that the first stage reactor showed better stability by addition of PAC with particle sizes of 100–200 μm in long-term operation rather than the two others. Therefore, in the subsequent experiment, PAC with particle sizes of 100–200 μm was selected for use in the first stage reactor.

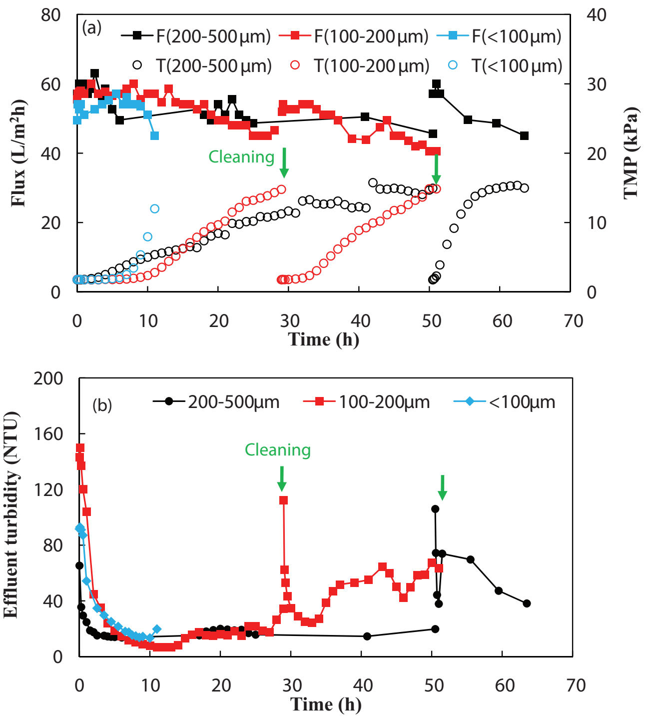

Fig. 2. (a) Profile of the flux and TMP and (b) evolution of the effluent turbidity under different PAC particle size in the first stage PAC-DMF system.

3.1.2. Dosage of the PAC

The effect of PAC dosage (with PAC size of 100–200 μm) on the formation and operation of the DM was further investigated (Fig. 3). With the three PAC dosages (1, 2, and 3 g/L),

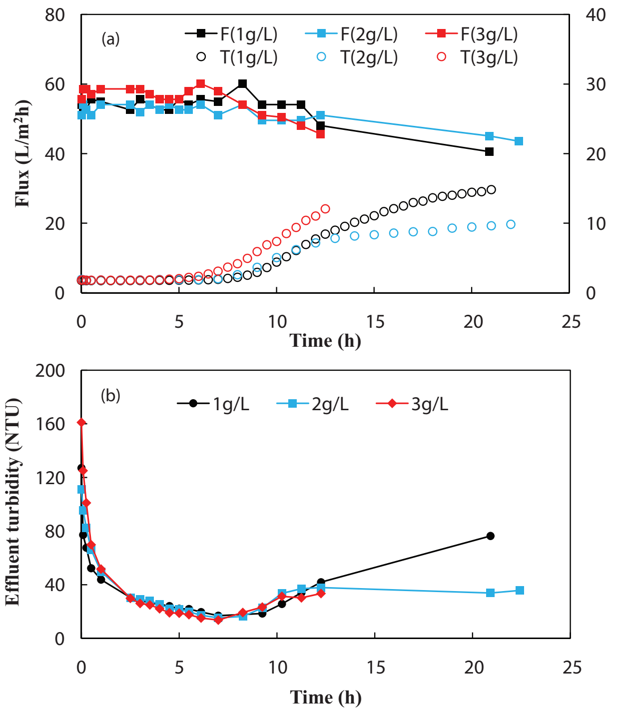

Fig. 3. (a) Profile of the flux and TMP and (b) evolution of the effluent turbidity under different PAC dosage in the first stage PAC-DMF system.

the formation time of the DM was similar (<2 h), and the TMP remained stable (<2 kPa), but the effluent turbidity rapidly decreased from 100 NTU to less than 40 NTU. At the very beginning, the interception effect was mainly achieved by the nylon mesh itself so that the retention of particles was not satisfied, the rapid formation of the dynamic membrane was promoted by adding PAC; then TMP and the effluent turbidity became relatively stable, showing that the DM layer was formed and a good particle retention effect was obtained; after 7 h, the TMP increased rapidly, and the effluent turbidity also began to increase slowly, this might be due to the gradually thickening of the DM layer caused by the continuous deposition of PAC and particulate substances in wastewater. This caused the quick increase in the filtration resistance, and simultaneously some of the fine particles in DM layer were extruded into the effluent through the pores of the DM layer due to extremely high local filtration flux [27]. By careful comparison, it was found that when the dosage of PAC was 2 g/L, the TMP gradually stabilized after the rapid growth period, indicating that the dynamic membrane structure tended to be stable at this dosage, and it was favorable for the long-term operation of the first stage reactor.

3.1.3. Strategies after DM cleaning for the continuous operation

3.1.3.1 Direct filtration after DM cleaning At the end of one single cycle operation, direct filtration after air backwashing was performed to investigate the feasibility of continuous operation. However, as presented in Fig. 4, in the next cycle, the initial effluent turbidity was very high (600 NTU), and a quick increase in TMP was also observed. After backwashing, the DM layer detached from the mesh surface into the concentrate, in which large particles and even the agglomeration and a large amount of retained small particles and colloidal substances from the wastewater existed.

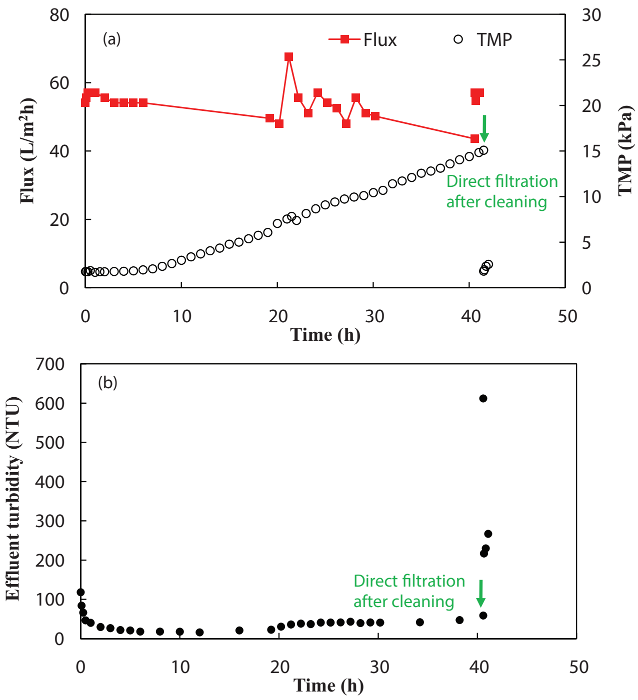

Fig. 4. Direct filtration after cleaning for continuous operation of the first stage reactor: (a) profiles of the flux and TMP and (b) evolution of the effluent turbidity.

The smaller particles would attach to the mesh surface faster and more easily than larger ones in the next DM formation process, which would lead to more small particles passing through the mesh pores, increasing the effluent turbidity and losing the substances retained in the reactor [28]. Certainly, once the small particles attach on the mesh, a denser DM layer would be established, which undoubtedly causes a rapid increase in TMP [23]. Thus, based on the analysis, the strategy of direct filtration after air backwashing was not suitable for the long-term operation of the first stage reactor.

3.1.3.2 Sieving after DM cleaning Based on the above results, another operation strategy using the sieving method after DM cleaning was adopted by using a 100-μm sieve to separate the small particles from the large particles (containing PAC) in the mixed liquor. As such, the concentrated liquid was screened to recover the large particulates, such as PAC, after DM backwashing for reuse in the next operation cycle (Fig. 5). After backwashing, the membrane flux basically recovered to the initial efficiency. The reactor could operate stably after screening the concentrated liquid, which was evidenced by the fact that the growth rate of TMP was stable during the DM formation process, and the effluent turbidity dropped to 30 NTU after 1 h and remained stable until the end of the cycle. The analysis indicated that sieving after DM cleaning was an effective strategy to sustain the stable operation of the first stage reactor.

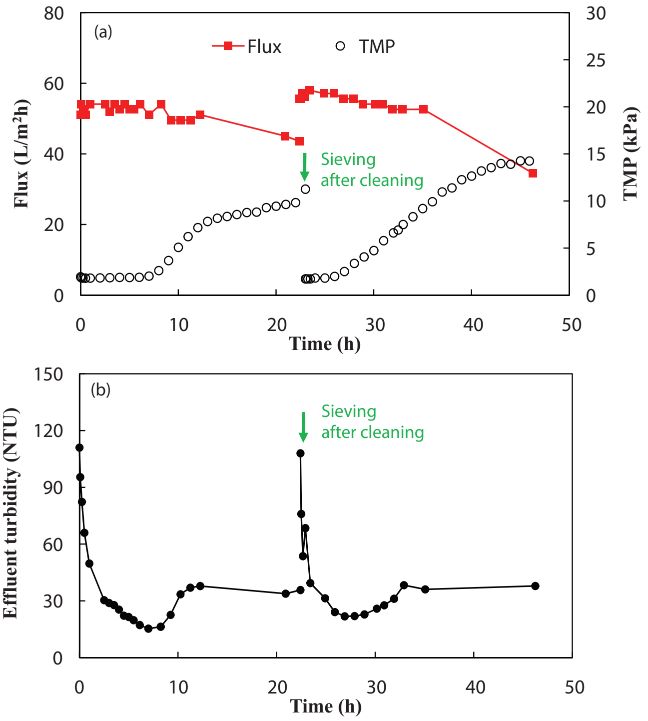

Fig. 5. Sieving after cleaning for continuous operation of the first stage reactor.

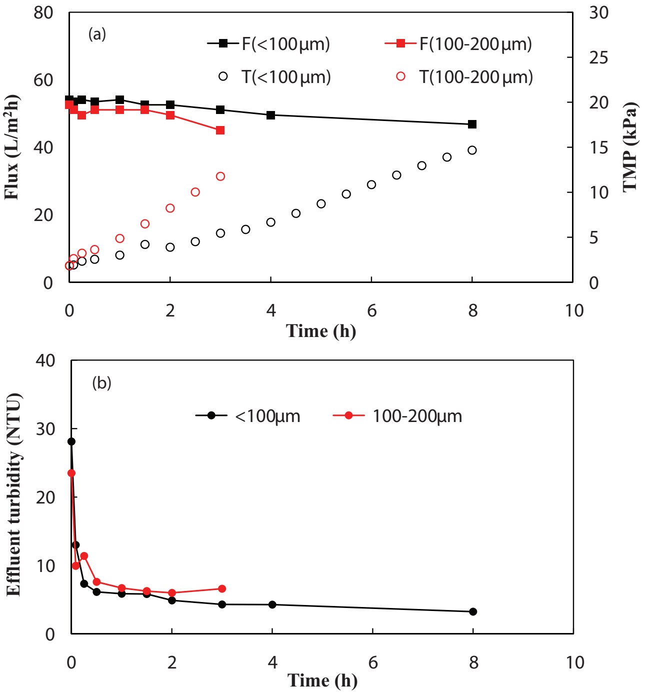

Fig. 6. (a) Profile of the flux and TMP, (b) evolution of the effluent turbidity at the second stage PAC-DMF system under different particle size of PAC at initial flux of 60 L/(m^{2}h).

3.2. Effect of PAC particle size on the operation of the second stage PAC-DMF reactor

To further improve the water quality of the first stage effluent and to enhance the organic matter retention, the particle size of the PAC, which is an important factor affecting the operation of the second stage PAC-DMF reactor, was studied. Based on the previous bath filtration test (data not shown), a nylon mesh with a pore diameter of 30 μm was selected as the DM supporting material due to the smaller particle size of the first stage effluent, which was used as the influent for the second stage reactor. The changes in the TMP and effluent turbidity with the addition of PAC with different particle sizes (less than 100 μm and 100–200 μm) are shown in Fig. 6.

Obviously, the effluent turbidity decreased rapidly below 10 NTU within 30 min and remained stable afterwards. Although a similar trend for the effluent turbidity was observed, the TMP growth was relatively slower with the addition of PAC with particle sizes less than 100 μm, and could be stabilized for an extended time. If the particle size of the PAC was much larger than the pore diameter of the supporting mesh (30 μm in this study), like the PAC with the 100–200 μm pore size, the formation of a DM was difficult, or the formation rate was slow because the large-diameter PAC could not easily adhere to the membrane. Under this condition, small particles in wastewater can easily attach to the surface of membrane which would result in the fast increase of the filtration resistance [24]. Thus, PAC with particle sizes smaller than 100 μm was selected to enhance DM formation in the second stage reactor.

3.3. Operation performance of the two-stage PAC-DMF system

3.3.1. Filtration performance

The long-term operation results of the first stage reactor are shown in Fig. 7. The DM can be effectively regenerated after air backwashing and the flux was able to be recovered to the initial value. During the first three cycles, the growth rates of the TMP were similar, and only in the fourth cycle, after 80 h, did the TMP growth rate increase. The increase was due to the fluctuation of the wastewater quality as the influent COD in the fourth cycle (343.1 mg/L) was higher than that in previous three cycles (233.2 mg/L). After backwashing in each cycle, the effluent turbidity decreased rapidly and reached a stable value after 1 h (<50 NTU). However, compared with the overall effluent quality of each cycle, the turbidity of the effluent gradually increased after backwashing. The purposes of the first stage reactor were (1) to provide relatively stable effluent to avoid great fluctuations in the raw wastewater; (2) and to concentrate organics (mainly particulate) for resource recovery by using the AD process. Therefore, satisfactory filtration performance was achieved in terms of effluent stability, TMP increase, and operation cycle length. One previous single stage DMF study using a double-layer membrane module (50 μm Dacron cloth as inner layer and 1 μm propene polymer cloth outer layer) without the addition of PAC reported that COD could be concentrated to 4,500 mg/L within 70 h, but the TMP quickly increased to 80 kPa in less than 10 h at a flux of 2–10 L/m^{2}h [18]. Thus, the low flux and high TMP made the mentioned process less cost-effective, further indicating the advantages of the present study.

Taking the first stage effluent as the influent of the second stage reactor, the long-term operation performance of the second stage reactor was investigated (Fig. 8). During the first cycle, the growth rate of the TMP was fast in the dynamic membrane formation stage. This is due to the low influent turbidity of the second stage. In addition, small particles and dissolved organics were the main substances that mixed with the PAC to form a denser DM layer, resulting in quicker TMP increase. When the membrane formed again after backwashing, although the influent turbidity increased, the TMP increase was slower during the operation cycle. Moreover, it was also found that the effluent turbidity in the first cycle decreased to 10 NTU and became stable after 3 h. However, during the second cycle, the effluent turbidity dropped to 10 NTU within 1 h.

Additionally, Fig. 9 shows images of the new membrane module and those used in the first and second stage reactor, which confirmed the DM layer formation in the two-stage PAC-DMF system. Additionally, the continuous operation test assured the applicability of the selected operation parameters and strategies. However, more tests were needed to verify the feasibility of using the concentrated liquid for bioenergy production through AD, as well as the reuse potential of PAC particles (affected by abrasion and adsorption saturation) during long-term operation.

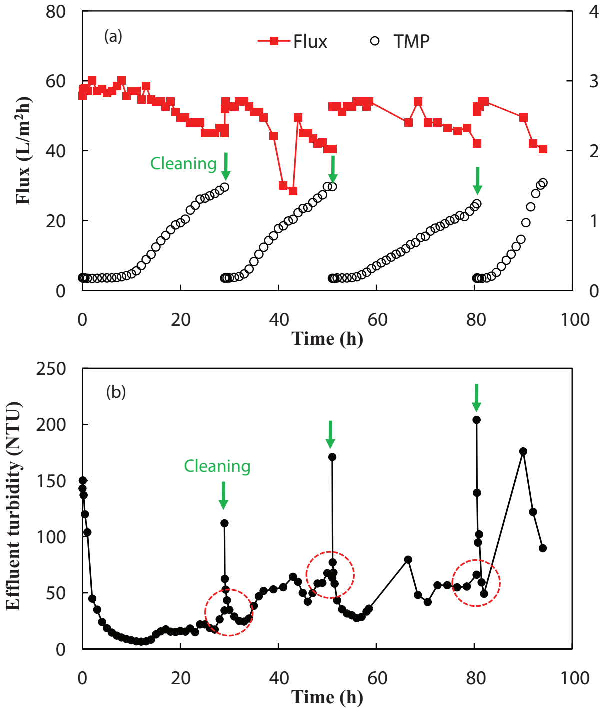

Fig. 7. (a) Flux and TMP profiles and (b) evolution of the turbidity of the first stage reactor in long-term operation.

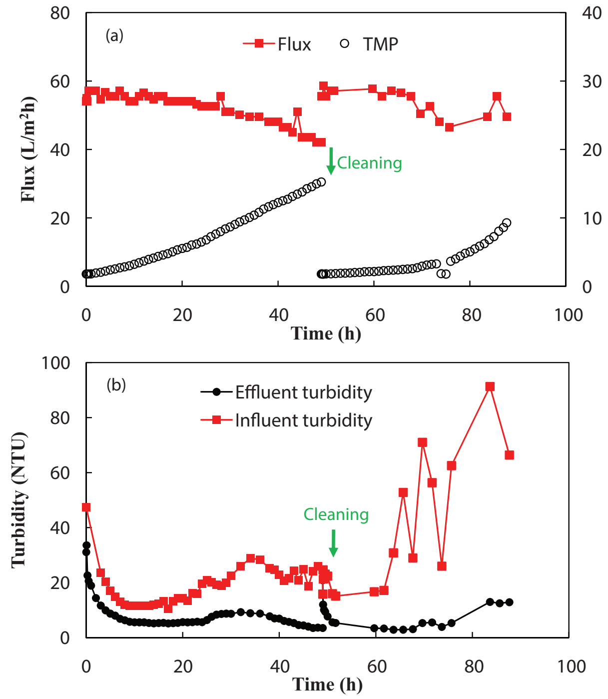

Fig. 8. (a) Flux and TMP profiles and (b) evolution of the turbidity of the second stage reactor in long-term operation.

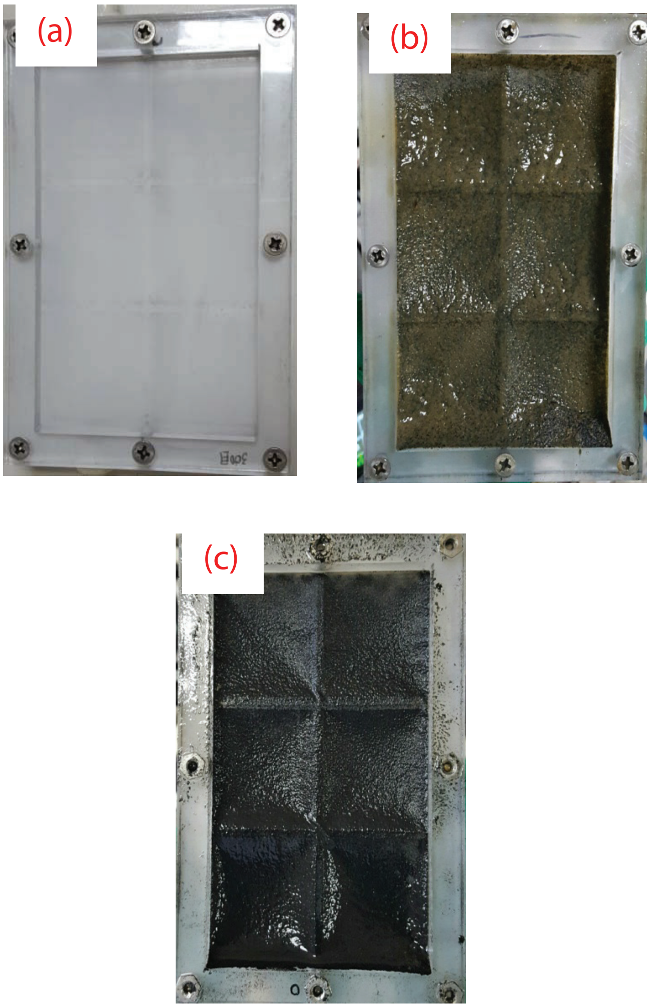

Fig. 9. Pictures of membrane modules: (a) new one; (b) first stage, and (c) second stage.

3.3.2. COD removal and organics concentrating performance

The variations in the total COD (TCOD) and soluble COD (SCOD) in a single cycle of the long-term operation of the two-stage PAC-DMF system were investigated. As shown in Fig. 10, the TCOD in the effluent after the first 2 h was higher and decreased continuously, indicating that the dynamic membrane was not completely formed, and the interception effect was not acceptable. After the DM was established (approximately 4 h), the TCOD of the effluent was stable (50–70 mg/L) with a removal rate of 74.1%–81.5%.

The effluent TCOD of the second stage fluctuated in the range of 30–70 mg/L at 2 h. Due to the rapid formation of the dynamic membrane, a large amount of organic materials were intercepted, which was evidenced by a rapid drop in TCOD and a stable TCOD tendency in the effluent. After 28 h, with the gradual compaction of the DM layer, small particles and soluble substances penetrated the DM layer and entered the effluent, and the TCOD content (mainly SCOD) in the effluent began to rise. This indicated that the particulate organics were largely trapped and low effluent turbidity was achieved at the second stage reactor.

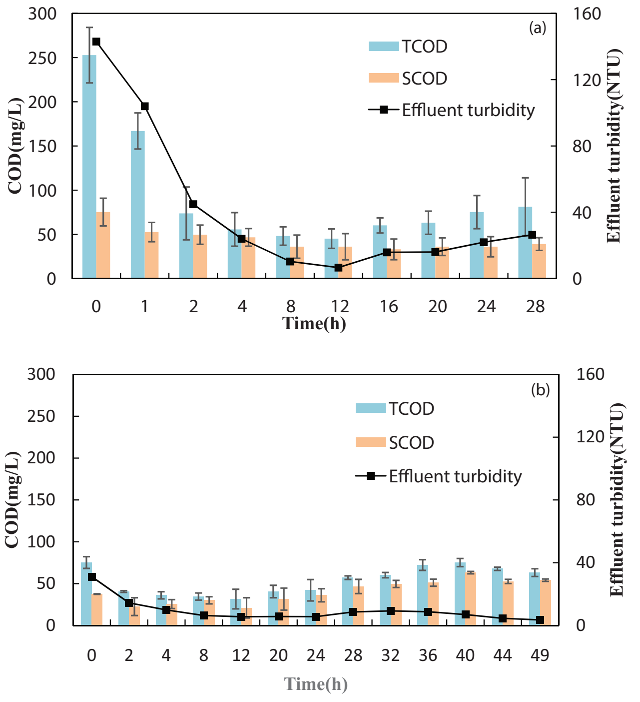

Fig. 10. Effluent organics and turbidity in the two-stage PAC-DMF system: (a) first stage and (b) second stage.

At the end of the single cycle operation of the first stage PAC-DMF, the concentrate was screened (100 μm) to recover the large particulates, such as PAC, for the next cycle operation, and the filtrate was used to recover the organic material. As shown in Table 2, the TCOD of the concentrated liquid was 3,175 ± 166 mg/L before sieving, and 1,637 ± 158 mg/L after sieving. The SCOD was much lower (less than 200 mg/L), indicating that particulate organic materials were the dominant components, and further explained the high interception rate of the DM. The much higher COD content in the concentrate than that in the wastewater indicated that it was feasible to concentrate organic matter using the PAC-DMF system.

3.3.3. Other pollutants removal performance

The removal performance of other pollutants from the wastewater by the two-stage PAC-DMF system is shown in Table 3. Each stage of the two-stage PAC-DMF system demonstrated certain removal effects for TN, TP, and NH_4^+–N. The second stage reactor further improved the effluent quality with the total removal rates of NH_4^+–N and TN, TP, UV_{254}, and chroma of approximately 9.7%, 24.8%, 41.1%, 48.8%, and 88.5%, respectively. The high removal rate of UV_{254} and chroma was largely due to the adsorption effect of the PAC used in the system [19].

For nitrogen and phosphorus removal, the retention effect of the DM layer was more important, but quite different removal rates were due to their existing forms (soluble or particulate). TN was known to mainly exist in the soluble state (such as ammonia nitrogen), while a considerable portion of the TP existed in the particulate form [29], and therefore different retention effects were observed. Moreover, the main goals of this study were: to retain and recover organic substances in the wastewater as more as possible from the two-stage reactors, facilitating further bio-energy production

Table 2
Concentrating effect of organics in first stage reactor (mg/L)

Filtration cycles	Before sieving (100 μm mesh)		After sieving (100 μm mesh)	
	TCOD	SCOD	TCOD	SCOD
First cycle	3,401	93.3	1,730.5	79.8
Second cycle	3,198	36.1	1,791	30.1
Third cycle	3,032.5	161	1,587.5	147.5
Fourth cycle	3,070	164	1,437	153.5

Table 3
Pollutants removal by the two-stage PAC-DMF system

Parameters	Influent	First stage effluent	Second stage effluent	Averaged removal rate (%)	
				First stage	Second stage
NH_4^+–N (mg/L)	24.7 ± 11.6	23.6 ± 11.2	22.3 ± 11.0	4.7	9.7
TN (mg/L)	44.2 ± 14.1	35.8 ± 15.4	33.2 ± 16.3	18.9	24.8
TP (mg/L)	4.8 ± 1.8	3.9 ± 1.8	2.8 ± 1.3	19.1	41.1
UV_{254} (cm^{-1})	0.24 ± 0.08	0.19 ± 0.07	0.12 ± 0.05	18.3	48.8
Chroma (c.u.)	333 ± 106	157 ± 75	38 ± 8	52.9	88.5

Table 4
Reclaimed water quality standards for agricultural irrigation and gardening

Parameters	China	USA	Europe
COD	200	–	100
TKN	30	–	15–25
TP	10	–	2–5
SS	100	30-NS^a	10–20^a
NH_4^+–N	–	10	2–20

Note: ^{a}TSS; NS means not specified.

using AD; (2) and to reuse the final nitrogen and phosphorus-rich effluent for agricultural purposes. Thus, an averaged removal rate of TN and TP less than 50% was acceptable to enrich nutrients as much as possible, certainly within the reclaimed water reuse standards for agricultural irrigation [30–32], which are shown in Table 4. Based on the results, it was noted that the effluent was rich in nutrients (nitrogen and phosphorus) with low contents of organics, turbidity, and chroma, which indicated that the two-stage PAC-DMF system was promising for producing reclaimed water for agricultural irrigation, gardening, etc.

4. Conclusions

In this study, a novel PAC-DMF system was developed and investigated for domestic wastewater treatment. By adding PAC with particle sizes of 100–200 μm and a dosage of 2 g/L in the first stage reactor, the effluent demonstrated a stable water quality. PAC with particle sizes less than 100 μm at a dosage of 1.5 g/L was used in the second stage reactor, which further enhanced the water quality and organic material retention, which was evidenced by the low effluent turbidity (5–20 NTU) and COD (<70 mg/L). The strategy of sieving the concentrated liquid by using a 100 μm sieve was adopted for the long-term operation, which retained the PAC for reuse and separated the smaller sized substances for organic material recovery. The long-term operation results demonstrated that the COD content in the concentrate could reach above 3,000 mg/L in the single cycle filtration experiment at the first stage reactor. The effluent of the second stage showed a low organic material content, but was rich in nutrients suitable for agricultural irrigation or gardening.

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