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Pretreatment of micro-polluted surface water with a biologically enhanced PAC–diatomite dynamic membrane reactor to produce drinking water

Huaqiang Chu[a,b], Yalei Zhang[b,*], Bingzhi Dong[b], Xuefei Zhou[b], Dawen Cao[c], Zhimin Qiang[d], Zhenxun Yu[a], Hongwu Wang[a]

[a]*School of Environmental Science and Engineering, Tongji University, Shanghai 200092, China*
[b]*State Key Laboratory of Pollution Control and Resource Reuse, Tongji University, Shanghai 200092, China*
Tel./Fax: +86 21 65985811; email: zhangyalei@tongji.edu.cn
[c]*National Engineering Research Centre for Urban Pollution Control, Shanghai 200092, China*
[d]*Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China*

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ABSTRACT

This study developed a biologically enhanced powder activated carbon (PAC) diatomite dynamic membrane reactor (BPDDMR) to pretreat micro-polluted surface water for drinking water production at lab-scale in continuous mode. In the start-up operation period, the BPDDMR required approximately 26 and 31 d to achieve stable removal efficiency of COD_{Mn} and NH_3–N, respectively. Turbidity was always below 0.5 NTU throughout the operation experiment in the permeate flux range of 21–54 $l\ m^{-2}\ h^{-1}$. The BPDDMR could effectively remove the hydrophilic portion of dissolved organic materials (DOM) present in the raw water. The temperature affected pollutant removal (especially COD_{Mn}), which was mainly ascribed to microbial degradation and was also enhanced by PAC and diatomite adsorption. During the precoating period, the stainless steel support mesh (aperture 74 μm) first intercepted the large PAC (50–100 μm), and then diatomite particles (5–20 μm) were intercepted to form a two-layer structure of the biologically enhanced PAC diatomite dynamic membrane (BPDDM). It was found that as the air pressure increased, the backwash efficiency improved and had less residual in the cake layer. Air backwash with a pressure of 200–250 kPa completely cleaned the BPDDM surface.

Keywords: Biologically enhanced PAC–diatomite dynamic membrane reactor; Micropolluted surface water; Filtration; Biodegradation; Air backwash; Drinking water treatment

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1. Introduction

To overcome the drawbacks of the traditional membrane bioreactor (MBR), such as high cost of membrane module, membrane fouling and high energy consumption [1], a new dynamic membrane technology has been developed in recent years. Specifically, the membrane can be created dynamically on the underlying support mesh while filtering a solution containing fine particles; thus, it is also called a formed-in-place membrane [2] or a secondary membrane [3]. Dynamic membrane technology has many advantages, including the low cost of the membrane module, reduced energy consumption, and high quality treated water; thus, it is considered a substitute for MBRs [4,5]. There are two types of dynamic membranes: precoated and self-forming [6].

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*Corresponding author.

The precoated dynamic membrane is created by passing a solution of one or more specific colloidal components over the surface of support mesh, while the self-forming dynamic membrane is created by the substances that exist in the mixed liquor to be filtered. The activated sludge self-forming dynamic membrane has attracted the attention of most environmental researchers [7–9].

Diatomite particles are good carriers of microorganisms [10]. Microbial colonies can form zoogloeas on diatomite particles through microbial capsules and surface mucus, and bio-diatomite is thus named. Recently, a bio-diatomite dynamic membrane reactor (BDDMR) has been developed for municipal wastewater treatment [11]. It was found that the BDDMR was highly effective in removing chemical oxygen demand (COD), NH_3–N and total nitrogen and also exhibited the advantages of good retention capacity for suspended solids, short precoating time, high filtration flux, and easy backwash.

Currently, the surface water is polluted to some extent in many parts of China. The primary pollutants include NH_3–N and dissolved organic materials (DOM), which are primarily composed of non-acidic hydrophilic fraction [12]. The conventional process of coagulation/sedimentation/sand filtration adopted by the majority of drinking water plants in China does not reduce NH_3–N and DOM effectively. In our previous work, the feasibility of treating micropolluted surface water with a BDDMR in the continuous-flow mode at lab-scale was investigated [13], and the results indicate that it effectively removed COD_{Mn}, dissolved organic carbon (DOC), UV_{254}, and NH_3–N and reduce the formation potential of trihalomethanes at a hydraulic retention time (HRT) of 3.5 h.

PAC, due to their porous structure and surface properties, are important materials in both water and wastewater treatment [14–16]. The addition of PAC to MBR can not only improve the removal of contaminants [17,18] but also effectively control the membrane fouling and flux decline [19,20]. In this study, to enhance the removal of pollutants and optimize the structure of dynamic membrane, PAC was added to the reactor and combined with diatomite to create a new biologically enhanced PAC–diatomite dynamic membrane reactor (BPDDMR) for drinking water production. The effectiveness of BPDDMR was assessed for pretreatment of micro-polluted surface water.

2. Materials and methods

2.1. Experimental setup

The schematic diagram of a lab-scale BPDDMR and its dynamic membrane (BPDDM) module is shown in Fig. 1. The BPDDMR consisted of two parts: an aerobic tank hosting the BPDDM and an automatic control system. The effective volume of the aerobic tank was 12 l. The support module using the stainless-steel mesh with an equivalent aperture of 74 µm was fixed in submerged mode with a double-sided effective filtration area of 0.051 m^2 (23 cm × 11 cm).

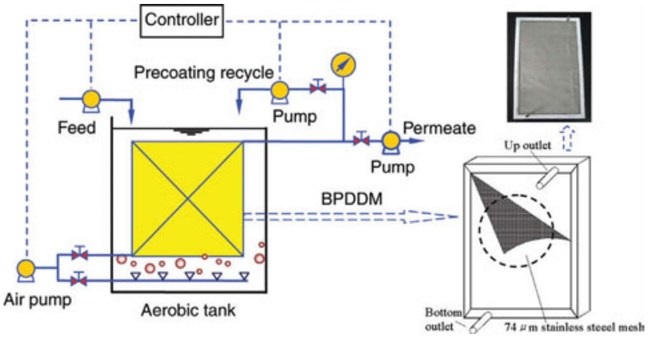

Fig. 1. Schematic diagram of (a) the BPDDMR and (b) the configuration of the BPDDM support module.

2.2. Experimental methods

The feed water was collected from the Sanhaowu stream on the Tongji campus, and its characteristics are summarized in Table 1. The continuous-flow natural biofilm mode was used to form the biologically enhanced PAC–diatomite mixed liquor in the start-up stage. PAC and diatomite particles with an equivalent diameter of 50–100 µm and 5–20 µm, respectively, were added to the bioreactor on the first day of the cultivation stage to reach a total solids concentration of about 14,000 mg l^{-1}, and then 1000 mg l^{-1} PAC–diatomite was added to the reactor daily for the following 6 d; the weight ratio of diatomite to PAC was 3:1. When the biologically enhanced PAC–diatomite mixture matured, the concentration of mixed liquor suspended solids (MLSS) in the reactor was maintained at 20,000 ± 800 mg l^{-1}. The bioreactor was operated continuously by feeding the raw

Table 1
Main quality parameters of feed water[1]

Quality parameters	Range
Turbidity (NTU)	4.81 ± 0.80
COD_{Mn} (mg l^{-1})	5.15 ± 0.52
UV_{254}	0.073 ± 0.010
NH_3–N (mg l^{-1})	1.21 ± 0.25
Temperature (°C)	7.2–30.6

[1] Values are given as mean concentration ± standard deviation; number of measurements: $n = 46$.

water with a peristaltic pump and withdrawing the permeate water with a frequency converter pump. During the experimental period, continuous aeration was provided at the bottom of the reactor to provide dissolved oxygen (i.e., 2–3 mg l^{-1}) for biologically enhanced PAC–diatomite mixed liquor, to suspend the mixed liquor suspended and to generate turbulence for the BPDDM. The aeration rate was controlled by a gas flowmeter at 2 l min^{-1}. The average sludge age was 40 d with 300 ml of excess biologically enhanced PAC–diatomite mixed liquor discharged daily. The feeding weight of PAC and diatomite was adjusted according to the excess biologically enhanced PAC–diatomite mixed liquor quantity and the real-time concentration of MLSS.

A high-precision vacuum pressure gauge was installed on the permeate pipe to measure the suction pressure (i.e., operation pressure) which was used to calculate the trans-membrane pressure (TMP). The operation period of the BPDDM involved three stages: precoating, filtration and backwash. In the precoating stage, the biologically enhanced PAC–diatomite mixed liquor was continuously recirculated by the frequency converter pump with flux about 100 l m^{-2} h^{-1} until the BPDDM formed on the surface of the stainless-steel support mesh with effluent less than 1 NTU, which took approximately 10 min. In the filtration stage, a constant flux was applied, and the BPDDM thickness continuously increased, which caused the filtration resistance to increase. The filtration was stopped once the operation pressure reached 40 kPa, and then backwash was started. On-line backwash was performed with pressurized air through the bottom outlet of the membrane module using an air pump (AC-002, Maple, China).

2.3. Analytical methods

Turbidity, COD_{Mn}, NH_3–N, NO_3–N and MLSS levels were analyzed according to the Standard Methods of the Chinese State Environmental Protection Agency [21]. Turbidity was measured by a turbidity meter (2100N, Hach, USA). NH_3–N was analyzed using the Nessler reagent method, and its absorbance was measured with a spectrometer (Gold Spectrumlab 53, Lingguang, China). UV absorbance at 254 nm (UV_{254}) was determined by a spectrophotometer (UV2550, Shimadzu, Japan) in a 1 cm quartz cell.

The membrane permeate flux was measured by the volumetric method with a graduated cylinder. The DOC concentrations of feed water and permeate were measured after filtration through a 0.45 µm membrane by a TOC analyzer (TOC-VCPH, Shimadzu, Japan).

The water samples was pre-filtered by a 0.45 µm membrane and then used to determine the molecular weight (MW) distribution. The MW distribution was measured with the gel permeation chromatography (GPC) method on a high performance liquid chromatography system (LC-10AD, Shimadzu, Japan) coupled with an SPD-20A UV detector and a TSK-GEL G3000PWXL column (7.8 mm × 300 mm). Anhydrous sodium sulfate (0.05 M) was used as the isocratic mobile phase. The separated compounds were detected by UV absorbance at 254 nm. The MW distribution pattern was derived by calibrating to the poly-styrene sulphonate MW standards of 14, 7.5, 4.3, 1.4, 0.7, 0.5 and 0.21 kDa. During the filtration process, a piece of BPDDM adhered to the support mesh and was cut off from the membrane module, whose thickness was measured by a ruler. After being dehydrated by natural evaporation for 48 h and coated with gold using a sputter coater, the sample was examined by scanning electron microscopy (SEM) (XL-30ESEM, Philips, Holland).

3. Results and discussion

3.1. Start-up operation period of BPDDMR

The removal efficiencies of turbidity, COD_{Mn}, NH_3–N and NO_3–N by the BPDDMR during the start-up operation period are shown in Fig. 2 at a permeate flux of 40 l m^{-2} h^{-1}, which corresponded to an HRT of 6 h in a temperature range of 19–30°C. Each data point in Fig. 2 represents an arithmetic mean calculated from three well-reproduced parallel measurements.

Fig. 2a shows that although an obvious fluctuation of turbidity was observed in the feed water, its concentration in permeate was consistently low (i.e., 0.28 ± 0.05 NTU), achieving a removal efficiency of 94 ± 1.3%. The permeate turbidity complied with the maximum contamination level (MCL) of 1.0 NTU according to the Chinese National Standards for Drinking Water Quality (CNSDWQ, GB5749-2006). In comparison to the particle rejection function of an ultrafiltration membrane, the permeate turbidity was controlled by the particle retention function of the BPDDM. The stainless-steel support mesh had little effect on the particle retention and only supported the dynamic membrane [22]. It was through the dynamic membrane that the BPDDM could obtain high solid-liquor separation efficiency.

During the initial 5 d, little NH_3–N removal was observed as the removal efficiency slightly declined from 9% to 4% (Fig. 2b). The NH_3–N removal efficiency gradually increased after 6 d, and was quite stable at approximately 90% after 31 d, when the permeate concentration of 0.08 ± 0.04 mg l^{-1} was achieved. At the beginning of the start-up period, the naked PAC–diatomite particles could only remove a small fraction of NH_3–N through adsorption, which agrees with the results reported by Wu et al. [23]. As microorganisms started to grow and accumulate on the surface of the PAC–diatomite

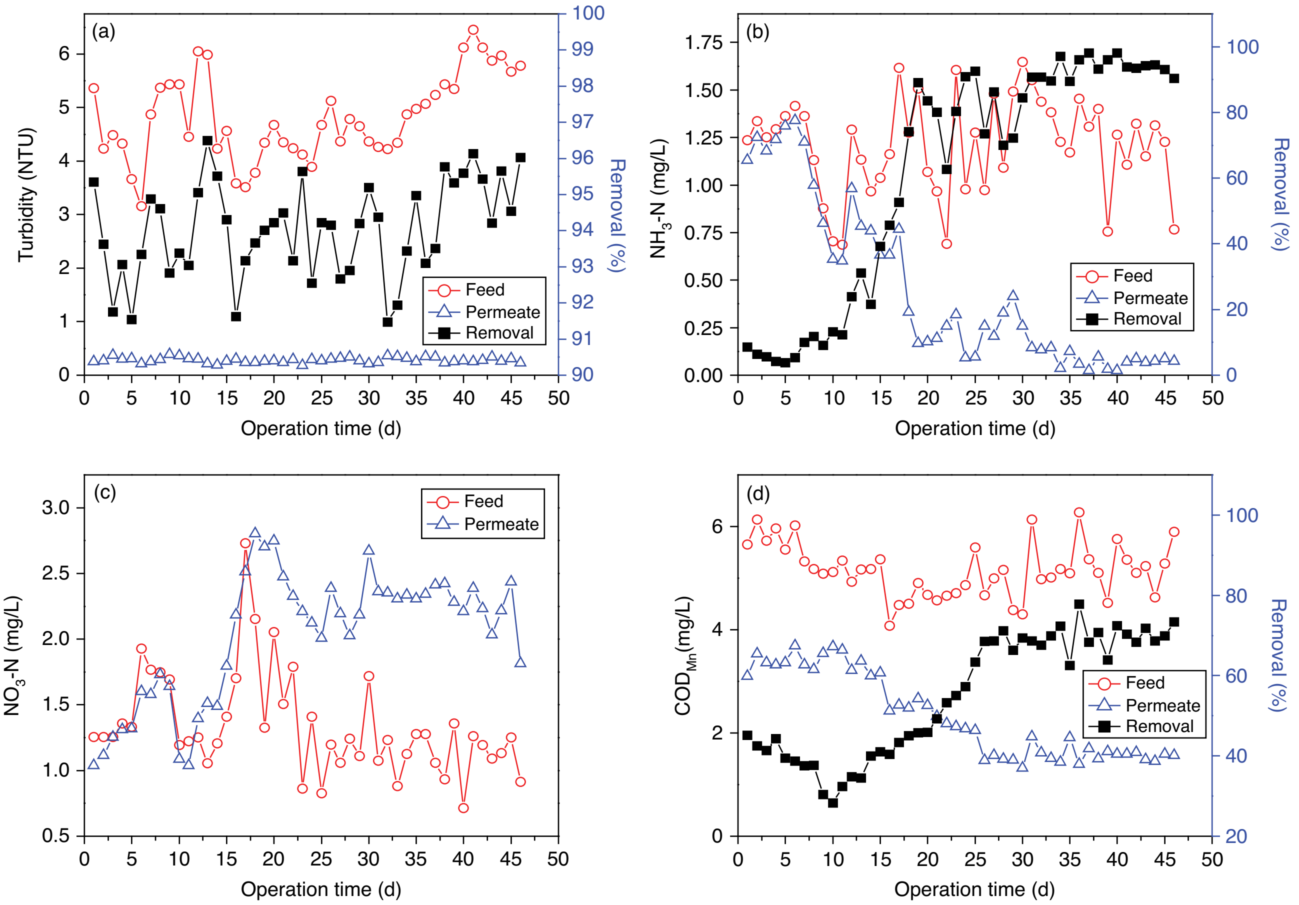

Fig. 2. Pollutants removal by BPDDMR in the start-up operation period.

particles, the NH_3–N removal efficiency showed a corresponding increase. It can also be seen from Fig. 2c that the nitrification process was accomplished slowly in the BPDDMR at the beginning of operation. The efficiency of the BPDDMR nitrification reaction was enhanced as the operation time increased because the amount of NH_3–N decreased and the amount of NO_3–N increased in permeate. As Fig. 2b and c show, it took about 31 d to cultivate biomass in the reactor for effective removal of NH_3–N.

The removal of COD_{Mn} during the start-up period is shown in Fig. 2d. In the beginning days of operation, the COD_{Mn} removal efficiency declined from 45% to 28% because the adsorption of PAC–diatomite was the only removal method. The adsorption capacity of PAC–diatomite for COD_{Mn} was much better than that for NH_3–N [23]. After 26 d of operation, the microbial decomposition of organic matter reached a steady level. The concentration of COD_{Mn} in permeate was 1.56 ± 0.15 mg l^{-1}, which corresponds to a removal efficiency of 70 ± 3.3%. The permeate COD_{Mn} concentration was much lower than the MCL of 3.0 mg l^{-1} regulated by the CNSDWQ (GB5749-2006).

Compared with the BDDMR in our previous study [13], the BPDDMR was more effective in removing turbidity, NH_3–N and COD_{Mn} of the same raw water. One fourth of diatomite was substituted by PAC, and the MLSS and MLVSS in this study were higher than those of the former study. PAC has a much larger average surface area (850 m^2 g^{-1}) than diatomite (59 m^2 g^{-1}); thus, it might adsorb more pollutants and provide more space for microorganism growth with the higher MLSS.

3.2. Determination of permeate flux

A series of fluxes (21, 43, 54 and 65 l m^{-2} h^{-1}) were adopted to determine the permeate flux of the BPDDM, which correspond to HRTs of 11.2, 5.5, 4.4 and 3.6 h, respectively, with an MLSS of 20,000 ± 800 mg l^{-1} in the lab-scale reactor. The change in TMP as a function of filtration time at various permeates fluxes is shown in Fig. 3a. Each data point represents an arithmetic mean calculated from six operation periods measured at the same time interval. The results indicate that the TMP increased quickly as the flux increased, and the filtration

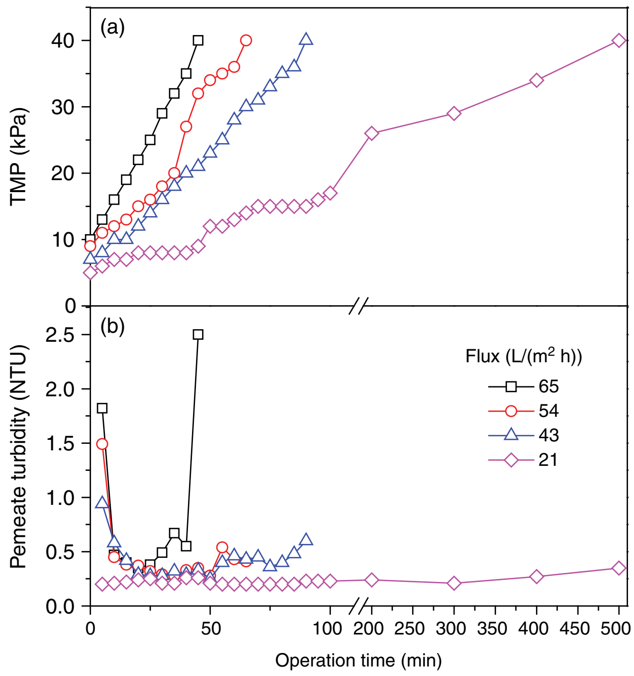

Fig. 3. Effect of flux on: (a) TMP and (b) permeate turbidity.

time to reach 40 kPa decreased. The permeate quality, taking turbidity as example, was notably affected by the flux (Fig. 3b). As the flux ranged from 21 to 54 l m^{-2} h^{-1}, the permeate turbidity was stable at 0.3–0.5 NTU. However, when the flux was raised to 65 l m^{-2} h^{-1}, the permeate turbidity rapidly increased to 2.5 NTU after 45 min of filtration. This significant change occurs because high flux can induce a strong shear force in the holes of dynamic membrane, which results in an increase in the permeate turbidity. The CNSDWQ (GB5749-2006) set an MCL of 1.0 NTU for water turbidity; therefore, the BPDDM flux should be controlled at 21–54 l m^{-2} h^{-1} during this lab-scale operation process. The BPDDM fluxes value adopted in this experiment are higher than or comparable to the microfiltration/ultrafiltration bioreactors for drinking water treatment but can be continuously operated under lower operation pressure, which is different from the intermittent on/off operation mode of a regular MBR [24–26].

3.3. MW distribution of removed DOM

When the start-up period ended, the changes in the MW distribution patterns of DOM in the feed water, mixed liquor and permeate of BPDDM were analyzed at a BPDDM permeate flux of 40 l m^{-2} h^{-1} by the GPC method, as shown in Fig. 4. Each peak cluster including MW 800–2500 Da peak cluster mainly represented hydrophilic substances, as reported in our previous work [13]. In the mixed liquor when both PAC–diatomite absorption and microbial decomposition were functioning, the peak cluster was decreased by about 40%. Comparing the two chromatograms of mixed liquor and permeate, it is noted that the BPDDM alone could remove the DOM with MW 800–2500 Da by about 30%. In contrast, the bio-diatomite dynamic membrane in the absence of PAC removed very little DOM [13]. PAC with high porosity and large specific area is an effective absorbent used in water purification. PAC has a weakly negative charged surface, and the negative charged moieties of DOM could complete the adsorption sites [27,28]. PAC adsorption could successfully reduce DOM with unsaturated carbon structures [29,30]. In the BPDDM retention process, the DOM with MW <2500 Da might be removed when both PAC–diatomite absorption and microorganisms biodegradation are functioning.

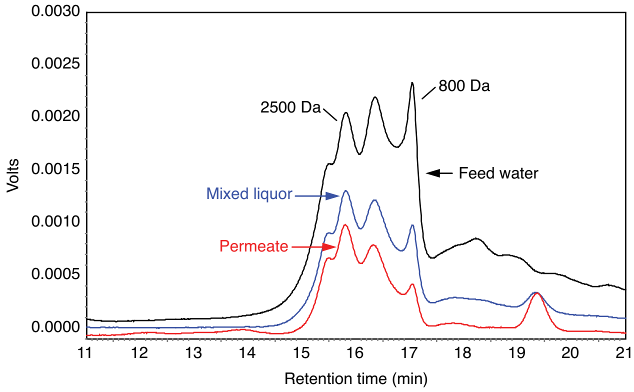

Fig. 4. Change of MW distribution during BPDDMR treatment.

3.4. Effect of temperature on pollutants removal

The BPDDMR was operated in a wide range of seasonal temperatures over a few months. The effect of temperature on pollutants removal (i.e., COD$_{Mn}$, NH$_3$–N and UV$_{254}$) by BPDDMR is shown in Fig. 5 at a permeate flux of 40 l m^{-2} h^{-1}. In Fig. 5, each data point represents an arithmetic mean calculated from three well-reproduced parallel measurements.

During the experimental period, the highest temperature was 33.9°C in the summer, and the lowest temperature was 5.4°C in winter. The results indicate that the performance of BPDDMR was positively related to water temperature, and the removal efficiencies of COD$_{Mn}$ and UV$_{254}$ appeared to be more sensitive to temperature variation than that of NH$_3$–N. In general, an increase in temperature affects pollutant removal in two opposite ways: (1) promoting microbial decomposition of pollutants [31,32]; and (2) reducing the adsorption capacity of PAC–diatomite for pollutants due to the exothermic nature of the adsorption process [33].

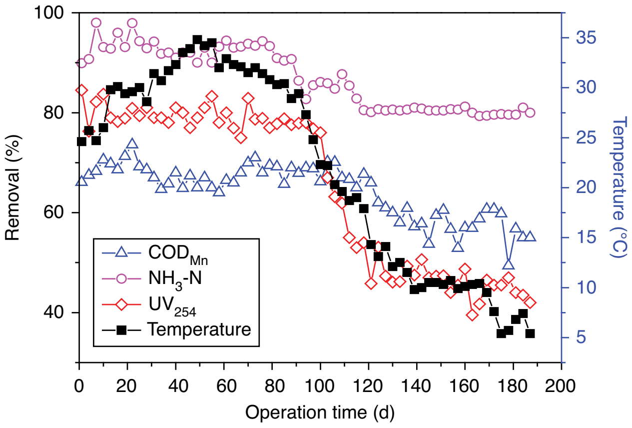

Fig. 5. Effect of temperature on pollutants removal.

The overall result indicates that the microbial decomposition overwhelmed the PAC–diatomite absorption in terms of pollutant removal. Although temperature had effect on the performance of BPDDMR, the permeate quality still complied with the requirements of CNS-DWQ (GB5749-2006), even in a cold weather (Fig. 5).

3.5. Mechanisms of pollutants removal

The mechanisms contributing to pollutant removal included microbial degradation, membrane interception, and PAC–diatomite adsorption. To determine which mechanism mainly accounted for pollutant removal, the BPDDM module was carefully taken out from the reactor after filtration for 2 h and then submerged into another tank that was only filled with the same feed water. Results from six repeated tests indicate that the turbidity, DOC, UV$_{254}$, NH$_3$–N and COD$_{Mn}$ had removal rates of 84 ± 1.1%, 9 ± 0.4%, 14 ± 0.3%, 7 ± 0.4% and 9 ± 0.3%, respectively. It is seen that the BPDDM alone was much less effective in removing pollutants, except turbidity. From the above discussion of Fig. 2, the PAC–diatomite particles without microorganisms only remove a small fraction of pollutants through adsorption. Therefore, the removal of pollutants was mainly ascribed to microbial degradation in the BPDDMR.

3.6. Morphological characteristics of BPDDM

The morphological characteristics of the BPDDM after filtration were examined by SEM, as shown in Fig. 6. It is clearly seen that the front surface of the dynamic membrane on the mixed liquor side was mainly composed of the biologically enhanced diatomite particles (Fig. 6a), while the back surface adhered to the stainless-steel support mesh was mainly composed of the biologically enhanced PAC particles (Fig. 6b). The figure shows that the BPDDM had a two-layer structure. The diatomite and PAC particles were in the ranges of 5–20 and 50–100 μm, respectively. During the precoating period, the stainless-steel support mesh (aperture 74 μm) first intercepted the large PAC particles to form an inner layer, and then the diatomite particles were retained by the PAC layer to form an outer layer. Some smaller particles could also fill the apertures present in the two layers.

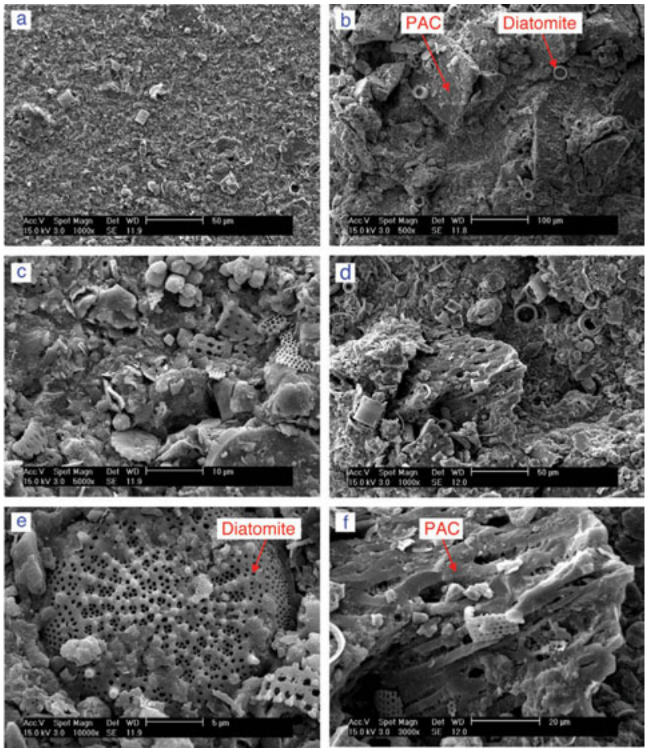

Fig. 6. SEM pictures of BPDDM: (a, c and e) the front surface on the mixed liquor side (1000×, 5000× and 10000×, respectively); (b, d and f) the back surface on the stainless-steel support mesh side (500×, 1000×, and 3000×, respectively).

The biologically enhanced PAC and diatomite particles were wrapped by microorganisms and extracellular polymer substances (Fig. 6c and d), and the BPDDM was bound tightly through microbial bridging. The diatomite and PAC particles could absorb both microorganisms and DOM to their surfaces and micropores (Fig. 6e and f). The presence of a large number of micropores in both diatomite and PAC particles facilitated microorganism growth.

3.7. Backwash of BPDDM

Air backwash was adopted in this study because it is highly effective in removing the cake layer on the support mesh [11,13]. Two permeate fluxes, that is, 38.5 and 54 l m^{-2} h^{-1}, were selected to test the air backwash efficiencies. As the TMP reached 40 kPa,

Table 2
Backwash parameters of BPDDM

Run #	Permeate flux ($\mathrm{l\ m^{-2}\ h^{-1}}$)	Operation pressure (kPa)	Backwash pressure (kPa)	Backwash flux ($\mathrm{l\ m^{-2}\ s^{-1}}$)	Backwash time (s)	Residual cake layer (%)
1	38.5	40	50	7.2	60	100
2	38.5	40	100	6.9	50	20
3	38.5	40	150	6.6	10	10
4	38.5	40	200	6.0	10	0
5	54	40	150	6.6	70	30
6	54	40	200	6.0	10	10
7	54	40	250	5.7	10	0

Fig. 7. Pictures of BPDDM: (a) before backwash; (b) after backwash; (c) cake layer right post-backwash.

the filtration process stopped and backwash started to recover the permeate flux. Table 2 lists various backwash conditions. The results indicate that at the same permeate flux (i.e., 38.5 $\mathrm{l\ m^{-2}\ h^{-1}}$), high air pressure 200 kPa with less backwash time 10 s improves the backwash efficiency with less residual cake layer. When the permeate flux increased to 54 $\mathrm{l\ m^{-2}\ h^{-1}}$), higher air pressure of 250 kPa was needed to completely remove the cake layer from the support mesh.

The pictures in Fig. 7 show the performance of air backwash while cleaning the BPDDM under the operational conditions of Run 7 (Table 2). It is seen that the cake layer had a smooth surface (Fig. 7a) and a thickness of about 2–3 mm (Fig. 7c). With sufficient air pressure, the cake layer was completely removed after backwash (Fig. 7b), and a clean support mesh surface was restored (Fig. 7b). The BPDDM remained unbroken when sloughed off the support mesh, which indicates that the diatomite and PAC particles were bound tightly through the microbial bridging force. Compared with the regular MBR (i.e., microfiltration/ultrafiltration MBR), the backwash of the BPDDM was much more convenient and did not require the addition of a chemical reagent [34,35].

4. Conclusions

1. A novel biologically enhanced PAC–diatomite dynamic membrane reactor was developed to pretreat micropolluted surface water for drinking water production. The BPDDMR effectively removed $\mathrm{COD_{Mn}}$, $\mathrm{NH_3}$–N and the hydrophilic portion of DOM present in the raw water. The temperature had effect on pollutant removal.
2. The removal of pollutants in the BPDDMR was mainly ascribed to microbial degradation and the BPDDM alone; PAC–diatomite absorption was much less effective in removing pollutants.
3. In the formation of the BPDDM, the stainless-steel support mesh (aperture 74 μm) first intercepted the large PAC (50–100 μm), and then diatomite particles (5–20 μm) were intercepted to form a two-layer structure.
4. To slough off the BPDDM at the end of the filtration stage, high air pressure improved the backwash efficiency with less residual cake layer. Air backwash with a pressure of 200–250 kPa completely cleaned the BPDDM surface.

Acknowledgements

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