An experimental study on the use of a sequencing-batch membrane bioreactor (SBMBR) for the treatment of mixed municipal wastewater

Yulan Gao, Jie Yang, Xinwei Song, Dongmei Shen, Wanfen Wang, Weimin Zhang and Jichao Jiang

ABSTRACT

Several water treatment techniques have been combined using the sequencing batch reactor with the membrane bioreactor for addressing water pollution. However, cleaning of the membrane is dependent on the approach involved as well as the operating conditions. In the present study, the sequencing-batch membrane bioreactor was used to treat real mixed municipal wastewater. The pollutant removal and membrane filtration performances were examined. The results show that the average removal rates of chemical oxygen demand (COD), total nitrogen, NH3-N, total phosphorus, and turbidity were 90.75, 63.52, 92.85, 87.58, and 99.48%, respectively, when the system was in continuous operation for 95 days. The membrane had a significant effect on COD and turbidity removal and provided stable performances for nitrogen and phosphorus removal. By observing the appearance of the membrane modules before and after the cleaning operation, it was concluded that the deposited sludge and granular sediment on the membrane surface can be effectively removed by hydraulic cleaning. In addition, recovery of membrane filtration performance to 60% of that of a new membrane can be achieved. Furthermore, we found that different sequences and duration of cleaning have different effects on the recovery of membrane filtration performance.

Key words | membrane cleaning, membrane fouling, mixed municipal wastewater, nitrogen and phosphorus removal, sequencing-batch membrane bioreactor

HIGHLIGHTS

- Sequencing-batch membrane bioreactor (SMBR) was used to treat municipal wastewater.
- We examined pollutant removal and membrane filtration performances over 95 days.
- Removal rates of COD, TN, NH3-N, TP, and turbidity varied from 63.52 to 99.48%.
- Recovery of membrane filtration performance to 60% was possible by hydraulic cleaning.
- Recovery increases to 97.6% if alkaline and acid washes follow hydraulic cleaning.

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INTRODUCTION

With the growing shortage of water resources and the progressive demand for energy conservation and emissions reductions on a global scale, advanced treatment and recycling of municipal wastewater are important measures in reducing environmental pollution and saving operating costs. Compared with the traditional activated sludge process, the membrane bioreactor (MBR) process has been widely implemented in sewage treatment plants in China due to its advantages of good effluent quality, small footprint, and low sludge output. However, membrane fouling is inevitable during the application of the MBR process, which increases the operating costs and has caused a global decline in the application of the MBR process. Only under the special conditions of extremely scarce land, very limited space, severe water shortage, and even water more expensive than oil does the MBR have a selective advantage (Bertanza et al. 2017; Hao et al. 2018). Currently, local and international studies on membrane fouling control mainly focus on: (a) optimisation of operating conditions (Paulen et al. 2015; Jelenský et al. 2016); (b) the modification of membrane material (Liu et al. 2015; Cheng et al. 2017); (c) improvement of the characteristics of the sludge mixture by adding fillers (Hu et al. 2012; Satyawali & Balakrishnan 2019; Yu et al. 2020); and (d) enhancement of hydrodynamic performance of the membrane surface (Braak et al. 2011; Culfaz et al. 2011; Mu et al. 2018).

With the rapid development of China’s economy and society, the nature of urban sewage has changed greatly. A large amount of industrial sewage is connected to the urban sewage network, resulting in large fluctuation of flow and load. Compared with general municipal sludge, the composition of this mixed municipal sewage is more complex, the total nitrogen and total phosphorus content is higher, and the biodegradability becomes lower and lower. The above-mentioned sewage is called mixed municipal sewage. In order to meet the more and more strict discharge standard and solve the problems of low carbon/nitrogen ratio, unstable denitrification effect and competition for carbon source between nitrogen removal and phosphorus removal, many studies (Guo et al. 2012; Li et al. 2014a; Wang et al. 2014; Liu et al. 2015; Lv & Liu 2019) combined the traditional nitrogen and phosphorus removal processes, such as the sequencing batch reactor (SBR), anaerobic-anoxic-oxic process, cyclic activated sludge technology, biological contact oxidation, and oxidation ditch technology, with the MBR process to strengthen the effect of nitrogen and phosphorus removal and improved the effluent water quality by studying the optimal operation strategy.

The SBR is an interesting alternative to the conventional (continuous) activated sludge process to accomplish the removal of nitrogen and phosphorus in small footprint treatment facilities. The SBR process has the advantages of small floor area, strong impact resistance, simple process, flexible operation, and easy to realise high automation. In order to improve the solid–liquid separation effect, the SBR is combined with the membrane separation process, which is
usually called sequencing batch membrane bioreactor (SBMBR) (Belli et al. 2017; Da-Costa et al. 2018). Due to the high flexibility in operation, the SBMBR can improve the hydraulic conditions of the membrane filtration process, change the characteristics of the mixture, and maintain a lower membrane fouling rate (Seo et al. 2000; Zhang et al. 2006; Yuan et al. 2011). Therefore, the application of improved treatment of urban and industrial wastewater has attracted extensive attention (Hasani Zonoozi et al. 2017), for instance, effect of the SBMBR on pollutant removal in municipal wastewater treatment (Belli et al. 2014; Belli et al. 2015; Belli et al. 2017), comparison of membrane pollution and microbial community characteristics in treating municipal wastewater (Yang et al. 2014; Da-Costa et al. 2018; Costa et al. 2019), effect of sludge retention time (SRT) and hydraulic retention time (HRT) on SBMBR membrane fouling (Hasani Zonoozi et al. 2015; Hasani Zonoozi et al. 2017), influence of Mg$^{2+}$ catalytic granular sludge on flux sustainability of the SBMBR (Sajjad & Kim 2015), and determination of SBMBR cycle time and process control (Krampe 2013; Souza et al. 2020) first evaluated the application of an intermittent current with low exposure time in an SBMBR system.

Membrane fouling is a major drawback of the membrane based bioprocesses. Deposition of sludge compounds on the membrane surface and/or within the membrane pores will either result in a higher transmembrane pressure for constant flux operation or lower flux over time in constant transmembrane pressure (TMP) filtration (Poorasgari et al. 2015). In addition, membrane fouling leads to increased frequency of membrane cleaning, shortening the service life and resulting in higher operating costs (Drews 2010; Rodríguez-Hernández et al. 2014). Therefore, it is of great significance for the long-term stable operation of the SBMBR to study the change trend, pollution characteristics and cleaning method of TMP in the intermittent filtration process.

Polyvinylidene fluoride (PVDF) hollow fibre membrane modules and an SBMBR were used in this study to treat real mixed municipal wastewater. The performance of pollutant removal was examined, and the filtration characteristics of the membrane were analysed. Furthermore, the effects of different cleaning sequences and time on the recovery of the filtration performance of the membrane are discussed. This study aims to evaluate the advantages of the traditional SBR and MBR, while enhancing the performance of nitrogen and phosphorus removal from mixed municipal wastewater. Through the use of suction filtration of the supernatant during the later stages of sedimentation in the SBR using the membrane module, the stability of the membrane filtration performance was better maintained, and membrane fouling was delayed. It provides a reference for upgrading and reconstruction of the sewage treatment subject to site constraints.

### EXPERIMENTAL SET-UP AND METHODOLOGY

#### Experimental set-up

An integrated MBR was used in this study and the experimental set-up is shown in Figure 1. The reactor with an effective volume of 150 L was separated by a baffle. A membrane module connected to a perforated aeration tube at the bottom was placed on one side of the baffle, and a mixer was placed on the other side. The membrane module used in the experiment consisted of a total of six PVDF hollow fibre microfiltration membranes with a pore size of 0.1 μm, and a single-membrane surface area of 0.60 m².

#### Experimental methodology

**Methodology of experiment operation**

The experiment was conducted in a sewage treatment plant in a development zone. The wastewater sample was taken from the collection tank in the sewage treatment plant. The main water quality parameters of the mixed wastewater are shown in Table 1. The retained sludge in the secondary sedimentation tank of the operating sewage treatment plant...
was used as an inoculum. The process operating steps are as follows: (1) instantaneous water inflow; (2) anaerobic mixing for 1.5 h; (3) aeration for 2.0 h; (4) sedimentation for 0.5 h; (5) membrane filtration for 1.0 h; and (6) idling for 1.0 h. The entire test system was automatically operated by a programmable logic controller. In order to slow down the pressure rise caused by membrane fouling, an intermittent suction method (3 min on/1 min off) was applied during membrane filtration. The system operating cycle lasted 6 h and four cycles were run per day. Dissolved oxygen (DO) was controlled between 2.0 and 3.0 mg·L⁻¹, and the sludge concentration was in the range of 3,000–4,000 mg·L⁻¹. The membrane flux was 20 L·m⁻²·h⁻¹. The drainage ratio was 1:3. The sludge age was 18 days.

**Evaluation of the performance of membrane fouling cleaning**

The membrane specific flux, defined as $K = J/P$ (which is the membrane flux value under unit filtration pressure difference), was used to characterise the membrane filtration performance. After cleaning, the normalised membrane specific flux, defined as $K/K_0$, was used to characterise the recovery of membrane permeability through a clean water flux test, where $K_0$ is the membrane specific flux of a new membrane (Huang & Mo 2003).

**Membrane cleaning**

After 85 d of system operation, the membrane specific flux dropped rapidly. After the system had operated for 95 days, it was no longer possible to meet the requirement of constant effluent output by adjusting the operating pressure. Membrane fouling had become severe, and system operation was halted to conduct the cleaning experiment on the membrane module. Due to the high acid resistance of the PVDF membrane, it can be cleaned with certain concentrations of hydrochloric acid, sulfuric acid, and citric acid. Moreover, the PVDF membrane does not have a strong resistance to NaOH but has a relatively strong resistance to oxidation. Thus, sodium hypochlorite can be used to sterilise or oxidise the organic matter under alkaline conditions (Li et al. 2014b). Hydraulic cleaning, alkaline cleaning using 5% NaClO solution, and acid cleaning using 5% H₂SO₄ solution were applied to clean the membrane modules. Four methods were used to clean the fouled membrane modules in order to investigate the effects of different cleaning sequences, chemical agents, and contact times with the membrane on the cleaning performance of the PVDF hollow fibre membrane. The specific methods are shown in Table 2.

**Parameters and methodology for analysis**

COD was measured by a Hach DRB200 digestion instrument and a Hach DR3900 spectrophotometer. Total nitrogen (TN) was determined by potassium persulfate oxidation–ultraviolet spectrophotometry. The volume of ammonia-nitrogen was determined by the salicylic acid method. Total phosphorus (TP) was measured by the alkaline potassium persulfate digestion–ascorbic acid method. Turbidity was measured by a WGZ-1 digital turbidity meter. Mixed liquor suspended solids was determined by the dry weight loss method. DO was measured by a Hach HQ50d portable DO meter, while pH was measured by a DELTA 320 pH meter.

**RESULTS AND DISCUSSION**

**Pollutant removal performance**

The status of pollutant removal after the 95 days continuous operation of the system is shown in Figure 2 and Table 3. Overall, the SBMBR process was effective in pollutant removal.
removal and the effluent quality remained stable. The average removal rates of COD, TN, NH₃-N, TP, and turbidity were 90.75, 63.52, 92.85, 87.58, and 99.48%, respectively. Additionally, the membrane contributed 6.10 and 14.62%, respectively, to the removal rate of COD and turbidity. Therefore, the process of membrane separation strengthened the performance of the system for COD and turbidity removal and improved the stability of effluent quality of the system, whose alternating anaerobic and aerobic operation provided a selective advantage for the metabolic growth of phosphorus accumulating organisms (Winkler et al. 2011; Xu et al. 2014), and the total phosphorus removal effect of the system was good. The TP concentration in the supernatant from the reactor was below 0.5 mg·L⁻¹ after 54 days of continuous system operation, which indicates that the phosphorus-accumulating bacteria in the reactor had already adapted to the alternating anaerobic-aerobic growth environment. In conclusion, the TP concentration in the supernatant from the reactor was below 0.5 mg·L⁻¹ after 54 days of continuous system operation, which indicates that the phosphorus-accumulating bacteria in the reactor had already adapted to the alternating anaerobic-aerobic growth environment.

Table 3 | Contaminant removal effect

<table>
<thead>
<tr>
<th>Item</th>
<th>COD (mg·L⁻¹)</th>
<th>pH</th>
<th>TN (mg·L⁻¹)</th>
<th>Turbidity (NTU)</th>
<th>NH₃-N (mg·L⁻¹)</th>
<th>TP (mg·L⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Class A standard limit</td>
<td>50</td>
<td>6–9</td>
<td>15</td>
<td>/</td>
<td>5</td>
<td>0.5</td>
</tr>
<tr>
<td>Sewage effluent</td>
<td>11.57–35.42</td>
<td>7–8</td>
<td>&lt;15</td>
<td>/</td>
<td>0.07–1.0&lt;</td>
<td>0.18–0.5</td>
</tr>
<tr>
<td>Supernatant</td>
<td>6–69</td>
<td>7.20–7.47</td>
<td>4.6–12.8</td>
<td>23.4–44.4</td>
<td>0.41–2.89</td>
<td>0.02–1.08</td>
</tr>
<tr>
<td>Membrane effluent</td>
<td>1–33</td>
<td>7.47–7.68</td>
<td>4.1–11.6</td>
<td>0.37–1.09</td>
<td>0–2.52</td>
<td>0.02–0.67</td>
</tr>
<tr>
<td>Average removal rate of system (%)</td>
<td>90.75</td>
<td>–</td>
<td>63.52</td>
<td>99.48</td>
<td>92.85</td>
<td>87.58</td>
</tr>
<tr>
<td>Average removal rate of membrane (%)</td>
<td>6.10</td>
<td>–</td>
<td>1.32</td>
<td>14.62</td>
<td>/</td>
<td>3.35</td>
</tr>
</tbody>
</table>

Figure 2 | Pollutant removal effect: (a) COD removal efficiency. (b) NH₃-N and TN removal efficiency. (c) TP removal efficiency. (d) Turbidity removal efficiency.
removal was significant, and the effluent output met the Class A discharge standard of the Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant (GB18918-2002) of China. The test process was simple, and the phosphorus removal performance was stable. Additionally, the TP concentration of the effluent was lower than that of the actual effluent of the secondary sedimentation tank in the sewage treatment plant. The method of timely addition of polyferric sulfate had been implemented by the sewage treatment plant to control the TP concentration of the final effluent to below 0.5 mg·L⁻¹.

Belli et al. (2015) used the SBMBR to treat urban sewage, and the total cycle time was 4 h. The results showed that the system had high removal efficiency of COD, NH₃-N and TN, with average removal rates of 97, 99 and 82% respectively. Meanwhile, the average removal efficiency of TP for the bio-membrane reactor was 48%. What is more, their results demonstrated that SRT reduction from 80 to 20 d had a negligible effect on COD removal and only a slight negative effect on nitrification. COD removal efficiency remained stable at 97%, whereas ammonium removal decreased from 99 to 97%. The TN removal efficiency was improved by SRT reduction, increasing from 80 to 86%. Although the TP removal was not significantly affected by the SRT reduction, ranging from 40 to 49%.

The SBMBR technique was used to treat real mixed municipal wastewater. When the system ran continuously for 95 days, the average removal rates for COD, TN, NH₃-N, TP, and turbidity were 90.75, 63.52, 92.85, 87.58, and 99.48%, respectively. The contribution of the membrane to the removal rates of COD and turbidity was 6.10 and 14.62% on average. The membrane separation process strengthened the capacity of the system to remove COD and turbidity, which improved the effluent quality. The effluent quality met the Class A discharge standard of the Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant (GB18918-2002) of China. The use of the SBMBR technique to treat real mixed municipal wastewater is a simple technological process and has a stable effect on nitrogen and phosphorus removal, which provides a reference for upgrading and reconstruction of sewage treatment subject to site constraints.

Separation characteristics of membrane filtration

The SBMBR was used to treat real mixed municipal wastewater without any membrane cleaning during the 95 days continuous operation. Figure 3 shows the trend curves of the TMP and K/K₀ during the operating timespan. It is apparent from Figure 3 that the TMP rose steadily from 12 to 21 kPa in the first 63 days, indicating that the growth rate of membrane fouling was relatively low at 0.142 kPa·d⁻¹. The reason for the latter is that, based on the characteristics of aeration, sedimentation and intermittent operation of the SBR, a hollow fibre microfiltration membrane was used in this test for the suction filtration of the supernatant during the sedimentation stage. Due to the low sludge content of the supernatant, membrane fouling during the separation process could be effectively reduced and the stability of membrane filtration performance could
be better maintained. Besides, a perforated aeration tube was installed under the membrane module, and a baffle was set in the middle of the reactor. During the aeration and reaction stage, an upward flow occurred on one side in the reactor, and a downward flow occurred on the other side, which boosted the mass transfer effect and produced a shearing effect on the membrane surface, decreasing the sedimentation of sludge on the surface of membrane filaments. As the operating time progressed, suspended particles in the reactor were deposited on the surface of membrane filaments and blocked the pores of the membrane, which accelerated the increase in the TMP at an average rate of 0.597 kPa·d⁻¹ during days 64 to 95. According to Figure 3, the value of K gradually decreased with increasing operating time. During days 85 to 95 of system operations, the TMP remained stable at 38–40 kPa. At that time, effluent output could no longer be ensured by increasing the operating pressure. By the end of the test, the K value had dropped from the initial value of 1.564 to 0.256 L·m⁻²·h⁻¹·kPa⁻¹, which is only 16.37% of the original filtration rate. This indicates that membrane fouling was relatively severe, and that it is therefore necessary to clean the membrane modules in order to improve their filtration performance.

Souza et al. (2020) evaluated the application of electric current with low exposure time in an SBMBR. The results indicate that the reactor performance regarding COD and NH₄-N removal was not influenced by the electric current application, exhibiting efficiencies above 99% in both experimental periods. On the other hand, the phosphorus concentration in the reactor effluent decreased as the electrocoagulation was applied, increasing its average removal efficiency from 62% (control period) to 89%. The use of electrocoagulation enhanced the flocculation process and sludge sedimentation, resulting consequently in a lower membrane fouling rate (1.19 kPa·d⁻¹) in comparison to the period without electrocoagulation (3.42 kPa·d⁻¹).

In Belli et al. (2015), the TMP for the first 50 days had been below 0.1 bar (10 kPa), between days 50 and 150, which increased slowly with fouling rate of 0.34 mbar·d⁻¹ (0.034 kPa·d⁻¹). On the other hand, from days 150 to 240, the TMP increased more intensely, presenting fouling rate of 1.2 mbar·d⁻¹ (0.12 kPa·d⁻¹), four times greater than that observed during the previous period.

In this study, a hollow fiber microfiltration membrane was used to filter the supernatant at the sedimentation stage of the SBR process, which reduced the adsorption and deposition of suspended solids and sludge particles on the membrane wire surface during the suction process. The membrane pollution developed slowly, and the TMP growth rate was 0.142 kPa·d⁻¹ (1–64 d) and 0.597 kPa·d⁻¹ (64–95 d), respectively, so the membrane filtration performance was relatively stable.

Surface characteristics of the fouled membrane

Membrane cleaning methods are classified as physical cleaning and chemical cleaning methods. As membrane fouling cannot be completely removed by physical cleaning, chemical cleaning is necessary to fully restore the membrane filtration performance. Generally, alkaline cleaning agents can effectively remove organic and biological pollutants, while the acidic cleaning agents are effective in removing minerals and inorganic pollutants. Acidic cleaning agents commonly used for removing membrane fouling primarily include HCl, HNO₃, and H₂SO₄, while alkaline cleaning agents mainly include NaOH and NaClO (Fu & Lin 2012; Zhou et al. 2019). Due to the different pollutants causing membrane fouling, the appropriate cleaning agent and method should be selected based on the properties of the membrane material and the characteristics of the pollutants (Li et al. 2014b). In addition, the cleaning efficiency is impacted by the operating conditions including temperature, pH value, concentration of chemical cleaning agents, and contact time of the chemical solution with the membrane (Mohammadi et al. 2002; Zhang et al. 2016).

The surface characteristics of the fouled membrane were observed for samples cleaned using cleaning method 2. After the membrane module was removed from the bioreactor, it was found that a large amount of sludge had accumulated on the surface of membrane filaments, as shown in Figure 4(a). This is because negative pressure suction was used to discharge effluent from the membrane bioreactor, which inevitably caused the suspended matter and other pollutants in the sludge mixture to become deposited on the surface and inside the pores of the membrane. As time progressed, the gel layer and later solid deposition layer on the membrane surface gradually became thicker and more compacted, which blocked the membrane pores and thereby affected the membrane flux.

By rinsing the fouled membrane module with clean water and wiping it with a sponge, most of the sludge deposited at the bottom and between filaments of the membrane module could be removed. The outer surfaces of the membrane filaments on which the sludge had been deposited were mostly brown and partially black. This is due to the alternating anaerobic–aerobic environment and the anaerobic respiration of the sludge inside the mud cake layer adhered to the outer surface of the membrane filaments.
during the operating process (Huang & Mo 2003; Li et al. 2014b), as shown in Figure 4(b). After immersing the membrane module into 5% NaClO solution for 12 h, it was found that the black sediments on the outer surface of membrane filaments were essentially removed, whereas a large amount of brown substances remained, as shown in Figure 4(c). An additional 12 h immersion in 5% H₂SO₄ solution, following the cleaning using 5% NaClO solution, cleaned most of surfaces of the membrane filaments by essentially removing the brown substances, and turning the filaments white, as shown in Figure 4(d).

**Comparison of cleaning performance**

The fouled hollow fibre membrane modules were cleaned according to the cleaning methods listed in Table 2. The resulting $K/K_0$ values after cleaning are compared in Figure 5, and it is apparent that approximately 60% of
be generated by the reaction between sulfuric acid and some pollutants on the surface and pores of the hollow fibre membrane, which block the pores of the membrane; after alkali washing, $K/K_0$ was recovered to 92.1% of the new membrane. All these possible mechanisms may therefore reduce the filtration performance of the hollow fibre membrane (Zhang et al. 2004). Further research is needed to determine the exact reasons for the differences observed in this system.

**CONCLUSIONS**

(1) By comparison, it is found that the characteristics of membrane fouling are different due to different sewage properties, operating conditions and membrane materials. In this experiment, precise quantitative prediction of membrane fouling behaviour is not made. In the later stage, the components of the pollutants can be analysed by three-dimensional fluorescence spectrum, infrared spectrum and energy spectrum analysis. The results provide a theoretical basis for optimising the operation mode, cleaning mode and effective control of membrane fouling.

(2) In this study, from observations of the surface of the fouled hollow fibre membrane and comparisons of the cleaning effect of the membrane module, we found that the hydraulic cleaning can effectively remove the sludge and granular sediments on the membrane surface, and the $K/K_0$ value can be restored to approximately 60% of that of a new membrane. The recovery of the $K/K_0$ value to 97.6% of that of a new membrane can be achieved with the immersion of membrane modules first in 5% NaClO for 12 h and then in 5% $\text{H}_2\text{SO}_4$ solution for another 12 h.

(3) The use of different cleaning sequences and times gave rise to variation in the recovery of filtration performance of the membrane. With the application of hydraulic cleaning as the first step, a better result was observed with first alkaline cleaning and then acid cleaning compared with the reverse sequence. In addition, if the contact time between the cleaning agent and the membrane module was insufficient, the membrane filtration performance could not be fully restored. However, if the contact time was too long, the surface structure of the membrane would be degraded, shortening the service life of the membrane.

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CONFLICTS OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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