Pilot trial study of a compact macro-filtration membrane bioreactor process for saline wastewater treatment

Dao Guan, W. C. Fung, Frankie Lau, Chao Deng, Anthony Leung, Ji Dai and G. H. Chen

ABSTRACT

Conventional membrane bioreactor (MBR) systems have increasingly been studied in recent decades. However, their applications have been limited due to their drawbacks such as low flux, membrane fouling, and high operating cost. In this study, a compact macro-filtration MBR (MfMBR) process was developed by using a large pore size membrane to mitigate the membrane fouling problem. A pilot trial of MfMBR process was set up and operated to treat 10 m^3/day of saline wastewater within 4 h. The system was operated under an average permeate flux of 13.1 m^3/(m^2·day) for 74 days. The average total suspended solids, total chemical oxygen demand, biological oxygen demand, total Kjeldahl nitrogen, and total nitrogen removal efficiencies achieved were 94.3, 83.1, 98.0, 93.1, and 63.3%, respectively, during steady-state operation. The confocal laser scanning microscopy image indicated that the backwash could effectively remove the bio-cake and dead bacteria. Thus, the results showed that the MfMBR process, which is essentially a primary wastewater treatment process, had the potential to yield the same high quality effluent standards as the secondary treatment process; thereby suggesting that it could be used as an option when the economic budget and/or land space is limited.

Key words | marco-filtration membrane bioreactor, organic and nitrogen removal, pilot trial, saline wastewater

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INTRODUCTION

In recent decades, membrane bioreactor (MBR) technologies have been developed and applied to water and wastewater treatment (Judd & Judd 2006). Compared with the conventional activated sludge process, MBR technology has many advantages including small footprint, low excess sludge production, and good effluent quality (Melin *et al.* 2006). Additionally, due to its long sludge retention time (SRT) and complete separation between hydraulic retention time (HRT) and SRT, the MBR process can achieve high ammonium removal efficiency at a short HRT (Henze *et al.* 2008). However, its application has mainly been limited to high-strength industrial wastewater or domestic wastewater treatment for effluent reuse (Gander *et al.* 2000). Membrane fouling control and low permeate flux are the major constraints (Le-Clech *et al.* 2006), which result in relatively high energy consumption and high operating cost (Zhang *et al.* 2006; Judd 2008). Moreover, membrane replacement further increases the maintenance cost (Meng *et al.* 2009).

In order to ease these constraints, a high-flux macro-filtration MBR (MfMBR), with a mean pore size >5 μm, has been developed in Hong Kong (Chen & Pang 2006). This MfMBR process achieved effective permeate flux of 6 m^3/(m^2·day) under the relatively low air-to-water ratio of 15 (Bai *et al.* 2010). A pilot trial of this process for saline sewage treatment demonstrated a non-fouling operation period of up to 370 days at such high permeate flux (Bai *et al.* 2011). This paper reports on the pilot trial of an optimized MfMBR targeting conditions of higher flux and shorter HRT than presented in most literature. These conditions may be suitable for the upgrade of existing chemically enhanced primary treatment (CEPT) plants towards secondary treatment with minimized land space and sludge disposal requirements. The MfMBR process

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presented in this study integrates a sludge hydrolysis tank under oxic–anaerobic conditions that are similar to those of the oxic–settling–anoxic (OSA) process developed by Chen *et al.* (2003). Hence, this tank is named OSA tank, whose functions are to stabilize the MfMBR sludge, reduce its sludge production and generate secondary substrate (mainly volatile fatty acids, VFAs) to enhance post-denitrification. The whole process is named the membrane enhanced primary treatment (MEPT®) process because its total HRT of 4 h is comparable to that of the CEPT process.

MATERIALS AND METHODS

MEPT pilot plant

A MEPT pilot plant was installed and constructed at Kwai Chung industrial wastewater pumping station, which receives about 10 m^3/day of wastewater (Figure 1). The process consisted of two tanks: an aerobic MfMBR tank and an OSA tank. The MfMBR tank was a cuboid tank having a membrane set and aeration diffusers; the volume of the tank was 0.833 m^3. The membrane set consisted of 10 membrane modules, which provided a total effective membrane surface area of 0.184 m^2. The membrane material was nylon woven fabric mesh whose mean pore size was approximately 55 μm. The OSA tank was a 0.833 m^3 anoxic cuboid tank with a propeller mixer. Raw wastewater was fed into the membrane tank after passing through a 2 mm fine screen, and subsequently flowed through an equalization tank having a short HRT of 14 min. After 2 h of aerobic reaction in the MfMBR tank, the mixed liquor entered the OSA tank for another 2 h of anoxic–anaerobic reaction. The flow recirculation ratio between these two tanks was set at 1. The dissolved oxygen (DO) concentration in the aerobic tank was kept higher than 2 mg/L and that in the OSA tank was below 0.5 mg/L (data not shown). The effluent was continuously drawn from the MfMBR tank under an operational permeate flux of 13.1 $m^3/(m^2 \cdot day)$. Daily membrane backwash with the effluent of the system was programmed to last 1 min every 24 h. Additionally, there was no chemical cleaning during the entire trial. The operation of the entire plant was controlled via an automatic programmable logic controller equipped with water level sensors. The seeding sludge was taken from the Shatin sewage treatment works of Hong Kong, which uses the conventional activated sludge process for the treatment of mainly domestic wastewater. The mixed liquor suspended solids (MLSS) concentration in both tanks was maintained at around 3.5 g/L. Throughout the whole trial period no sludge withdrawal was purposefully done.

Sampling and sample analysis

From March to May 2012, 24 h composite samples of the influent, effluent, and mixed liquor of the MBR and OSA tanks were collected every 2 days by using a peristaltic pump with automatic control. All samples were stored at 4 °C. The 24 h composite samples for total chemical oxygen demand (TCOD) and soluble chemical oxygen demand (SCOD) were analyzed on site, while the 24 h

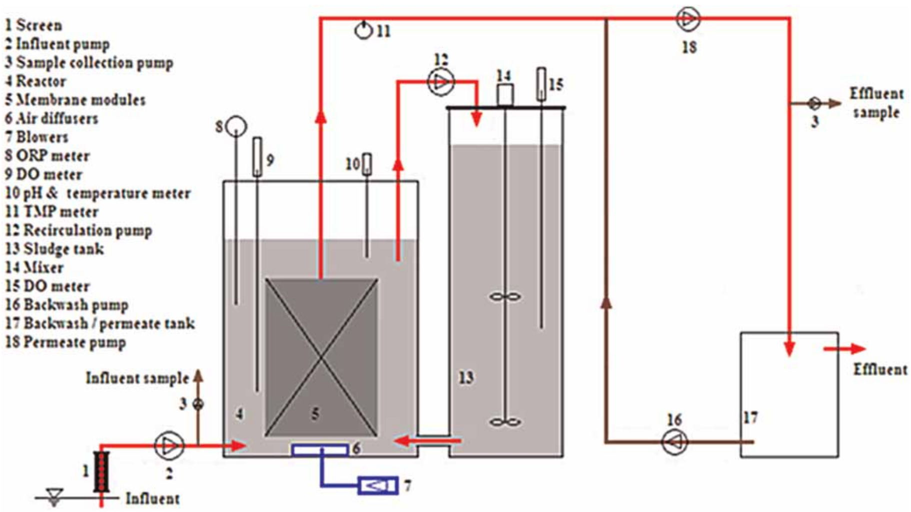

Figure 1 | Schematic of the MEPT pilot plant.

composite samples for biological oxygen demand (BOD_5) total suspended solids (TSS), ammonium (NH_4^+), total Kjeldahl nitrogen (TKN), and total nitrogen (TN) were measured in the laboratory within 12 h of sampling; so also were the grab samples for MLSS. At the end of the trial several pieces of membrane were cut off for bio-cake analysis, and subsequently preserved at 4 °C. The membrane pieces were then transported to the laboratory, and dyed and then examined by confocal laser scanning microscopy (CLSM) within 2 h.

Analytical methods

The determination of BOD_5 followed the British Standard BS EN 1899-1:1998 (BSI 1998), according to which nitrification is inhibited by allylthiourea (ATU). The procedures for measuring MLSS, TSS, TCOD, SCOD, TKN, ammonium, nitrate (NO_3^-), nitrite (NO_2^-), chloride (Cl^-), sulfate (SO_4^{2-}), DO concentrations, pH, and temperature followed *Standard Methods for the Examination of Water and Wastewater* (APHA 2005). Prior to the measurement of ammonium, nitrate and nitrite, samples were filtered through 0.45 μm filter membrane and then concentrations measured with a flow injection analyzer (FIA) (QuickChem FIA + 8000Series, Lachat, USA). The TKN samples were digested without filtration, before subjecting them to the FIA analysis. The TN concentration was determined from the TKN, nitrate, and nitrite concentrations. Chloride and sulfate concentrations were detected using ion chromatography (LC-20AD SP, Shimadzu, Japan) with a 7 μm pore Allsep™ A-2 anion column (Alltech, USA). The DO concentration was measured with a DO meter (YSI 52, USA). The TCOD and SCOD concentrations were determined with a COD reactor (HACH) coupled with a direct reading spectrophotometer (DR/2000, HACH, USA).

In order to investigate the fouling conditions, when a membrane module was removed from the MfMBR tank, several pieces of the membrane were cut off for bio-cake examination, including application of the LIVE/DEAD® BacLight™ Bacterial Viability and Counting Kit (L34856, Life Technologies, USA), to visualize the live and dead bacteria cells, in which green-fluorescent SYTO®9 and red-fluorescent propidium iodide were applied as dyes in sequence. After 15 min of incubation in the dark, samples were examined using CLSM (Zeiss, LSM7 DUO [710 + LIVE], Germany). More than nine images at different locations of each sample were taken to ensure the representativeness.

RESULTS AND DISCUSSION

Influent condition

The pilot plant suffered from high salinity in the influent due to the application of seawater toilet flushing in Hong Kong. The average Cl^- and SO_4^{2-} concentrations during the trial period were 6,473.84 ± 1,514.65 and 133.77 ± 71.49 mg/L, respectively. The detailed characterization of the influent is shown in Table 1. The high standard deviations of nitrate and nitrite (NO_x-N) and TN concentrations of 29 ± 83 and 84 ± 93 mg/L, respectively, were interpreted as being due to the presence of a fraction of industrial wastewater in the influent, which was not expected since the MfMBR was designed for domestic wastewater treatment. However, the constant volatile suspended solids to TSS ratio, stable TSS concentration, and high ammonium and TN removal efficiencies indicated that the influent had no apparent toxic effect on the sludge; this will be discussed later.

TSS removal

TSS concentrations in the influent and effluent and TSS removal efficiency are shown in Figure 2. Except in the 28 days of start-up operation, the effluent TSS was always below 30 mg/L despite great variation in the influent TSS from 500 to 1,000 mg/L. The overall TSS removal efficiency was 94.3%, suggesting that the MEPT® process could achieve good TSS removal.

The pore size of the membrane used was larger than 50 μm, which was in the range of the activated sludge particle size distribution. Comparing with conventional micro-filtration (MF) and ultra-filtration (UF) membranes, the bio-cake formed on the membrane surface in the MfMBR system plays a key role on filtering the activated

Table 1 | Average influent and effluent quality as well as removal efficiencies of the MEPT® process during 46 days of steady-state operation

Parameter	Influent	Effluent	Removal efficiency (%)
TSS (mg/L)	586 ± 187	28.8 ± 17.5	94.3 ± 3.1
SCOD (mg/L)	336 ± 193	54.1 ± 33.6	80.3 ± 10.6
TCOD (mg/L)	593 ± 288	86.0 ± 37.0	83.1 ± 8.4
BOD_5 (mg/L)	318 ± 177	4.6 ± 2.9	98.2 ± 1.4
NH_3-N (mg/L)	31.6 ± 13.7	1.38 ± 1.49	95.3 ± 5.3
NO_x-N (mg/L)	29 ± 83	13.9 ± 9.8	–
TKN (mg/L)	44.6 ± 18.8	2.69 ± 1.62	93.1 ± 4.6
TN (mg/L)	84 ± 93	20.2 ± 9.0	63.3 ± 23.1

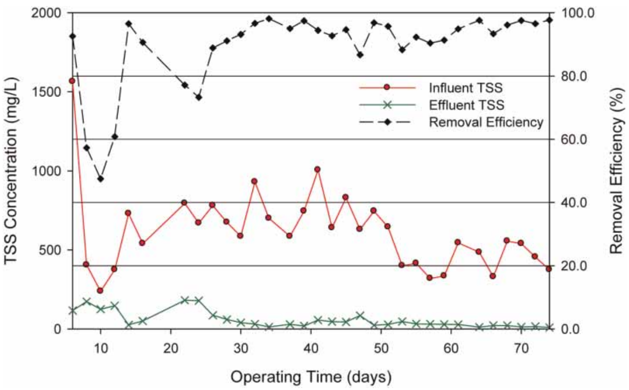

Figure 2 | TSS removal performance in MEPT pilot trial.

sludge – an observation also made by Fan & Huang (2002) who indicated that during the operation of large pore size membrane, a formation stage would be necessary for the cake layer to form on the supporting material. The high effluent TSS in the start-up period was due to incomplete formation of the bio-cake on the membrane surface. Therefore, a relatively long period was necessary for the build-up of the bio-cake layer. This indicated that a *start-up* period might be a requirement for the MfMBR process although its duration could be reduced depending on operational strategy such as the provision of sufficient seeding sludge.

Organic carbon removal

Figure 3 shows the TCOD and BOD_5 concentrations in the influent and effluent and respective removal efficiencies. Despite great TCOD fluctuation in the first 28 days, the effluent TCOD became stable at around 86 mg/L after this period. Similar to TCOD, the effluent BOD_5 was stable at 3–4 mg/L. The good removal of TCOD was attributed to stable bio-cakes having formed, thus providing good solids separation. The average removal efficiencies of TCOD and BOD_5 during the steady-state period were 83.1 and 98.0%, respectively. The results suggested that the process could remove almost all biodegradable COD in wastewater. Therefore, the relatively low TCOD removal efficiency and relatively high effluent TCOD concentrations were attributed to the high portion of slowly biodegradable and non-biodegradable carbon in the raw wastewater from an industrial area, which could not be removed during the short HRT of 4 h.

Nitrogen removal

Figure 4 shows the TKN and TN removal of the MEPT pilot trial. During the start-up period, the drop in the effluent TKN, which was as a result of improved TKN removal efficiency, was expected as more nitrifiers accumulated in the system owing to well developed bio-cakes. The TN removal efficiency during the steady-state period was 63.3% even when the influent nitrate was above 200 mg/L, which is much higher than typical municipal wastewater. These results indicated that the OSA tank in the MEPT process satisfactorily removed TN. The low BOD_5 level in the effluent of the MfMBR tank indicated that readily biodegradable organics, fed through the influent, were probably consumed in the aerobic tank (see Figure 3(a)). Consequently, the carbon source available for denitrification in the OSA tank became insufficient unless extra carbon was provided through the hydrolysis of sludge. Previous studies (Saby *et al.* 2003; Yuan *et al.* 2011) reported that sludge decay could take place in the OSA tank, thus generating VFAs, which became the carbon source for denitrification.

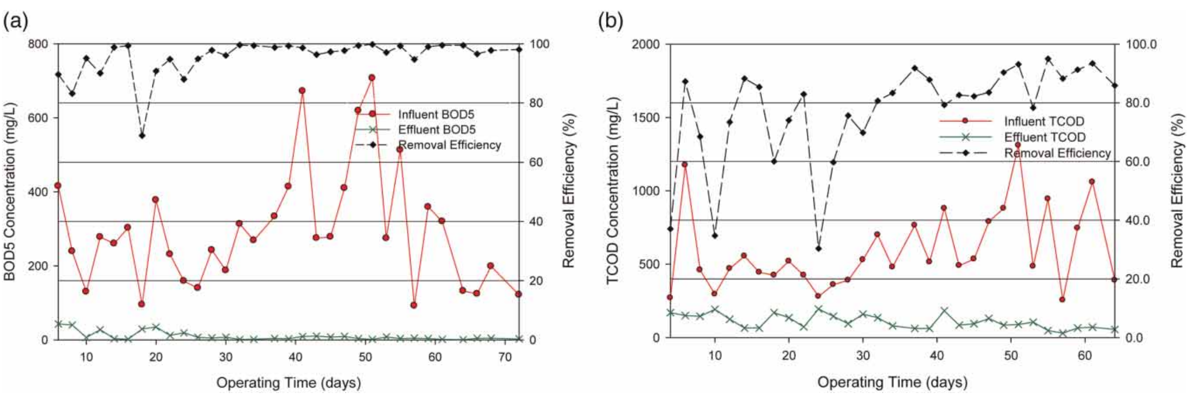

Figure 3 | (a) BOD_5 and (b) TCOD removal performance in MEPT pilot trial.

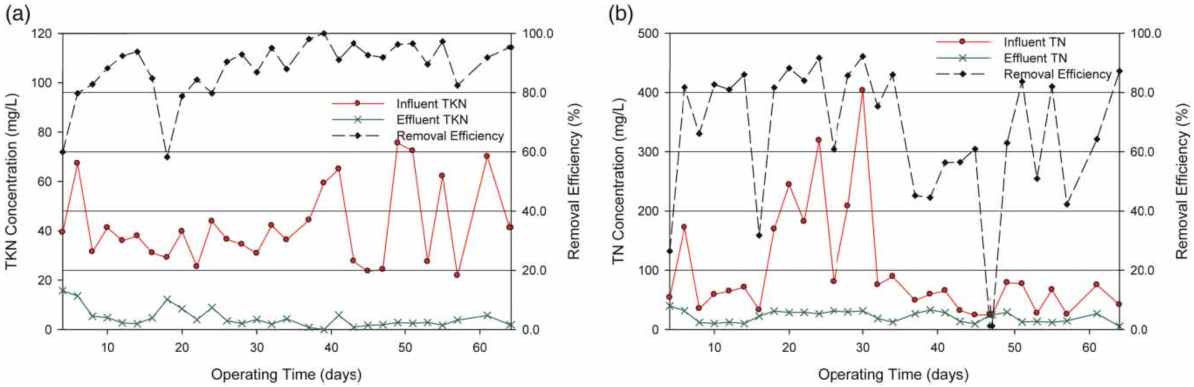

Figure 4 | (a) TKN and (b) TN removal performance in MEPT pilot trial.

General performance of MEPT® process

In general, the MEPT® process performed well during the steady-state operation. A summary of the quality of influent and effluent as well as the removal efficiencies is given in Table 1. The rather large standard deviation indicated that the quality of the influent raw wastewater varied significantly, especially its NO_x-N concentration. However, the effluent quality and removal efficiency were both relatively stable during the 46 days of steady-state operation. In summary, the overall performance of the MEPT® process was satisfactory in terms of membrane filtration, biological conversion, organics, and nitrogen removal. The effluent quality was consistently good and could fulfill the effluent discharge standards in Hong Kong (EPD 1997). The removal efficiencies in terms of TSS, TCOD, BOD_5, ammonia, TKN, and TN were high. Although the effluent SCOD concentration was not low, the removal of biodegradable carbon in the MEPT® process was high as indicated by the high BOD_5 removal. This showed that the bulk of the effluent SCOD might be in the form of unbiodegradable organics.

The average MLSS concentrations in the MfMBR and OSA tank were 3,214 and 3,188 mg/L, respectively. The calculated SRT of the MfMBR tank was 18 days. The observed overall sludge yield of the plant (including the solids loss via the effluent) was approximately 0.1 gMLSS/gCOD, which is much lower than that of the conventional anoxic/oxic process (i.e. around 0.35 gMLSS/gCOD when the SRT is 20 days) (Muller *et al.* 2003).

Membrane condition

During the 74 days of operation, the MfMBR system maintained a high permeate flux with an average value of 13.1 $m^3/(m^2 \cdot day)$, while the backwash frequency was maintained at 1 min every 24 h. The operational flux in this study was significantly higher than other studies

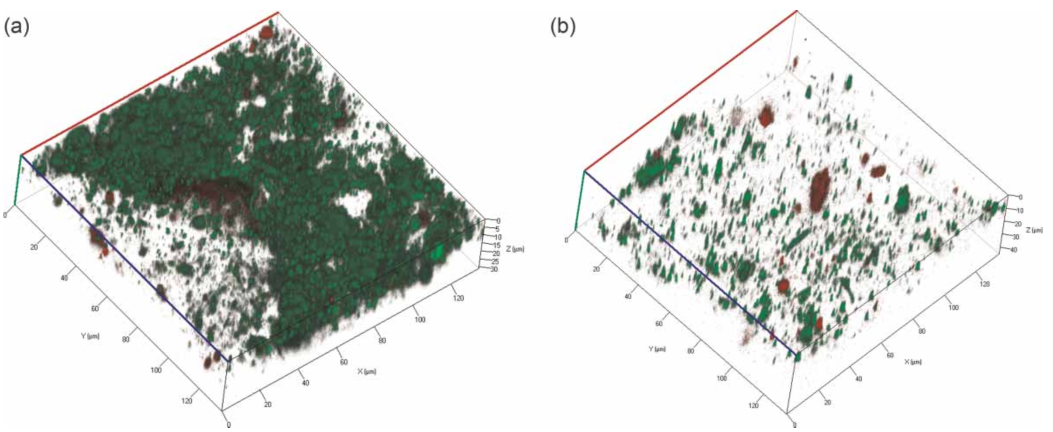

Figure 5 | CLSM image analysis on the bio-cake formed on the membrane before (a) and after (b) backwash at the end of the trial. Grey (green in online version) indicates live bacteria. Dead bacteria appear as black (red in online version) color. The full color version of this figure is available online at http://www.iwaponline.com/wst/toc.htm.

whereas the membrane pore size was in the same range (0.4–5 $m^3/(m^2 \cdot day)$) (Ersahin *et al.* 2012). Nonetheless, membrane fouling was under control as indicated by the low operational transmembrane pressure (TMP) value (<0.2 bars). At the end of the trial, live/dead double-staining combined with the CLSM image analysis was conducted to examine the characteristics of the bio-cake on the membrane surface. Figure 5(a) shows that the bio-cake developed well on the membrane surface, thus suggesting that a well-formed bio-cake could act as a filter to achieve the good effluent quality obtained in this study. Moreover, dead bacteria accumulation on the membrane surface was negligible, as indicated by the few black (red in online version) dots. The accumulation of dead bacteria has the potential to increase TMP as well as membrane fouling (Hwang *et al.* 2008). However, the fact that no apparent membrane fouling occurred in this study suggested that no or only few dead bacteria accumulated on the membrane. After membrane backwash, the bio-cake had significantly been removed, as shown in Figure 5(b), which suggested that backwash was still effective at the end of the trial, hence further substantiating the non-fouling condition of the membranes.

CONCLUSIONS

In this study, the performance of the MEPT® process was closely monitored and investigated under conditions of high flux and short SRT. The major findings are as follows.

(1) The pilot plant can remove TSS effectively and generate effluent with high quality comparable to that of the secondary wastewater treatment process. Compared with traditional MF or UF membrane, the MfMBR can provide an alternative solution when the effluent discharge standard (EPD 1997) is not rigorous and the budget is limited.
(2) Under a high flux (13.09 $m^3/(m^2 \cdot day)$), low backwash frequency (1 min every 24 h) and no chemical cleaning operation, the membranes operated continuously for 74 days without membrane fouling.
(3) The MEPT® process can achieve remarkable TKN removal, high COD and BOD removal, and a good TN removal efficiency.
(4) The OSA tank produced enough carbon source, in the form of VFAs, from sludge hydrolysis under well-controlled conditions to enhance TN removal.

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