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Optimized coupling of a submerged membrane electro-bioreactor with pre-anaerobic reactors containing anode electrodes for wastewater treatment and fouling reduction

Nader Taghipour, Mohammad Mosaferi, Mohammad Shakerkhatibi, Neemat Jaafarzadeh, Reza Dehghanzadeh and Hassan HosseinNia

ABSTRACT

In this paper, the performance of a submerged membrane electro-bioreactor with pre-anaerobic reactors containing anode electrodes (SMEBR$^+$) was compared with that of a membrane bioreactor (MBR) in municipal wastewater treatment. The new design idea of the SMEBR$^+$ was based on applications of direct current (DC) on the anode and cathode electrodes. The pilot study was divided into 2 stages and operated for 48 days. In Stage I, the MBR was continuously operated for 24 days without the application of electrodes. In Stage II, the SMEBR$^+$ was continuously operated for 24 days, while aluminum electrodes and an intermittent DC were working with an operational mode of 2 min ON/4 min OFF at a constant voltage of 1.4 V. The results indicated that membrane fouling was reduced by nearly 22.02% in the SMEBR$^+$ compared to the MBR. The results also showed that the SMEBR$^+$ increased the quality of effluent to the extent that high removals of NH_3^+-N, PO_4^{3-}-P, and chemical oxygen demand (COD) were 98%, 76%, and 90%, respectively. This system, in comparison with those proposed in other studies, showed a suitable improvement in biological treatments, considering the high removal of NH_3^+-N. Therefore, SMEBR$^+$ can be considered as a promising treatment alternative to the conventional MBR.

Key words | membrane fouling, pre-anaerobic reactor, submerged membrane electro-bioreactor, wastewater treatment

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INTRODUCTION

There is an increasing trend towards wastewater generation around the world. Wastewater treatment is essential to protect public health and prevent pollution. The effluent of wastewater treatment plants (WTPs) is a valuable water resource for reuse purposes due to many reasons such as water stress caused by climate change, etc. (Bani-Melhem & Smith 2012). Therefore, achievement of a good effluent quality and

improvement of the treatment plant efficiency and performance are important (Bani-Melhem & Elektorowicz 2011).

Numerous technologies and methods are used for wastewater treatment. Meanwhile, the membrane bioreactor (MBR) and electrocoagulation (EC) are considered as the processes to obtain good-quality effluents. Nevertheless, each of these processes, when separately operated, has some disadvantages (Hasan *et al.* 2014).

The use of MBRs in municipal and industrial wastewater treatment is widespread as it can supply high quality effluents and a high degree of automation, produce less sludge, improve process performance, and reduce the space

required for the facility installation (Liu *et al.* 2012b; Wang *et al.* 2013; Li *et al.* 2014; Zhang *et al.* 2014; Hawari *et al.* 2015).

The MBR technology is a combination of a conventional activated sludge process with membrane filtration units to maintain the microbial biomass (Akamatsu *et al.* 2010; Hasan *et al.* 2012; Dalmau *et al.* 2015). Despite the growing use of MBRs, membrane fouling is still the biggest problem and concern, reducing the membrane life-time and increasing operational costs, which should be resolved to ensure the sustainability of this technology (Trussell *et al.* 2006; Hasan *et al.* 2012; Ibeid *et al.* 2013).

The main reason for membrane fouling is the microorganisms active in the biomass, including the density of extracellular polymeric substances (EPS), which is a combination of elements such as proteins, polysaccharides, nucleic acids, lipids, humic acids, etc. Another reason is soluble microbial products (SMP), which play an important role in membrane fouling (Ni *et al.* 2011; Luna *et al.* 2014; Dalmau *et al.* 2015).

Drews (2010) categorized membrane fouling into reversible, irreversible that can be removed through maintenance cleaning, irreversible that can be removed through major chemical cleaning and irrecoverable fouling (Drews 2010; Ibeid *et al.* 2013; Hasan *et al.* 2014).

Many factors seem to contribute to membrane fouling, including deposition of negatively charged EPS and sludge, operational conditions, negative charged colloidal or submicron particles, sludge properties, and membrane characteristics (Trussell *et al.* 2006; Drews 2010; Hasan *et al.* 2012).

The membrane fouling phenomenon causes the permeate flux to decline, the trans-membrane pressure to increase, and the treatment process performance to decrease (Nagaoka *et al.* 1998; Hasan *et al.* 2012; Duan *et al.* 2013).

Several methods have been applied to mitigate membrane fouling such as optimizing process operating conditions (e.g. flux, solids retention time, etc.) (Schoeberl *et al.* 2005; Liu *et al.* 2012b), adding chemical coagulants (Lee *et al.* 2001; Wu & Huang 2008) or powdered activated carbons (Hu & Stuckey 2007; Tian *et al.* 2010), and improving system design such as aeration schemes (Bani-Melhem & Smith 2012; Liu *et al.* 2012b). Among

these methods, the aeration technique needs high energy to avoid membrane fouling and limits the application of MBRs for wastewater treatment (Akamatsu *et al.* 2010; Liu *et al.* 2013a).

Alternatively, the application of electrochemical techniques such as the EC process has been increased as a promising technology for wastewater treatment in order to directly control the properties of the membrane surface (Akamatsu *et al.* 2010; Bani-Melhem & Elektorowicz 2010). The use of an alternating electric field is proved to decrease membrane fouling (Akamatsu *et al.* 2010). This method is mainly attributed to the external electric field with the basic functions of electrophoresis and electrostatic repulsion of the charged particles (Liu *et al.* 2013b).

In recent years, applying an electric field within an MBR has induced electrophoresis away from the membrane surface thereby preventing deposition of negatively charged organic colloids such as planktonic microorganisms and EPS on the membrane surface (Liu *et al.* 2012b). On the other hand, EC reduces the cost and space for preparing coagulant agents, and produces larger flocs compared to those produced by chemical coagulation (Liu *et al.* 2012a). EC also requires less retention time (Kobya *et al.* 2006).

Meanwhile, SMP and EPS compounds in biological wastewater treatment systems are responsible for effluent quality and membrane fouling, and are released during normal biomass metabolism. Ni *et al.* (2011) found that these compounds could decay in an anaerobic reactor (Ni *et al.* 2011). Liu *et al.* (2013a) suggested that the aerobic condition in the cathode and the anaerobic condition in the anode would be suitable for enhancing effluent quality (Liu *et al.* 2013a).

In this study, we applied a pre-anaerobic reactor, containing aluminum anode electrodes, before the aerobic MBR, containing aluminum cathode electrodes with 1.4 V/cm electric current, in order for prevention of direct contact of the electrical current with the biomass in both the reactors, considering the fact that the optimal electric field helpful for microorganisms is 0.28–1.4 V/cm (Bani-Melhem & Elektorowicz 2010). Bani-Melhem & Elektorowicz (2011) illustrated that use of an electric field at the anode and cathode electrodes around the membrane

module in the aerobic MBR, leads to damage of nitrifying bacteria.

Consequently, the main differences of our study compared to other conducted studies are the positions where electrodes and the pre-anaerobic reactor are applied. We separately placed aluminum anode electrodes in the pre-anaerobic reactor and cathode electrodes in the aerobic reactor. However, in other studies, both anode and cathode electrodes were placed around the membrane module in the MBR.

In addition, an intermittently aerated MBR can cause a simultaneous nitrification and denitrification in the same reactor in accordance with time cycle of aeration and non-aeration (Radjenovic *et al.* 2008). Moreover, high shear aeration may breakdown the floes formation (Fu *et al.* 2012). Finally, the aeration mode (on and off) was considered in the aerobic MBR. The principal objective of this research was to investigate the performance of the submerged membrane electro-bioreactor (SMEBR) system where both the pre-anaerobic reactor, containing anode electrodes, and aerobic MBR with cathode electrodes operated in one hybrid unit. This combination resulted in effluents with excellent quality and reduced membrane fouling. The paper presents a comparison between two systems of MBR and SMEBR under the same conditions.

MATERIALS AND METHODS

Experimental setup

The experimental setup of this study consisted of two stages. In the first stage, a laboratory scale experimental setup (Figure 1) was used to continuously treat synthetic wastewater for 24 days.

This stage consisted of a polypropylene container with a capacity of 10 L which served as a synthetic wastewater feed tank. The synthetic wastewater was pumped from the feed tank to the MBR system via a feed pump.

The MBR system consisted of a bioreactor with flat sheet membranes (Sepro, USA) and its total and effective volumes were 3.74 L and 3 L, respectively. The membrane module with a diameter of 10 cm, pore size of 0.04 mm, and total surface of $0.0157 \, m^2$ was submerged in the middle of the reactor.

The effluent was suctioned out by a peristaltic pump (BT300-M, Shenchen pump Yz1515X, China) at a constant suction head using a flat sheet membrane module. The pump operated at 31 rpm with a flux of $7.05 \, L/m^2 \cdot h$.

An aeration system, which consisted of an air pump (AC-9603, China) with two air diffusers at the bottom of the aerobic tank, was placed on both sides of the membrane module to provide the necessary concentration of dissolved oxygen (DO) for bacterial growth and prevent the accumulation of activated sludge particles on the membrane surface until membrane fouling decreased. DO concentration in the MBR was controlled to be around 3.5–4.5 mg/L by adopted air flow meters. A timer (SZR M2, Turkey) was used for air recharging at a set operational mode (20 min ON/20 min OFF).

The second stage of the experimental setup shown in Figure 1 was used for the continuous treatment of the mentioned synthetic wastewater for 24 days. The procedures in the second stage were the same as those in the first stage.

In the second stage, a pre-anaerobic reactor containing aluminum anode electrodes was used and placed outside of the MBR. A blender was also used in this unit in order to keep the activated sludge particles suspended.

The aluminum cathode electrodes were applied in the MBR (SMEBR$^+$). The total and effective volumes of the SMEBR$^+$ were 3.74 L and 3 L, respectively. Moreover, the total and effective sizes of the pre-anaerobic reactor containing aluminum anode electrodes were 1.65 L and 1.56 L, respectively. The synthetic wastewater was pumped from the feed tank to the pre-anaerobic reactor via a feed pump, and flowed to the SMEBR$^+$ system by gravity. The effluent from the membrane module was withdrawn via a peristaltic pump (BT300-M, Shenchen pump Yz1515X, China) at a constant suction head.

In addition, Stage II of the pilot study included three pieces of aluminum plate with a thickness of 0.2 cm, width of 3 cm, and height of 18 cm. One aluminum plate was used as the anode and the other two were used as the cathode. Two electrodes were fixed as the cathode at a distance of 3 cm from each side of the membrane module. An Iranian (Micro, Iran) direct current (DC) supply system was used in this stage and the electrodes
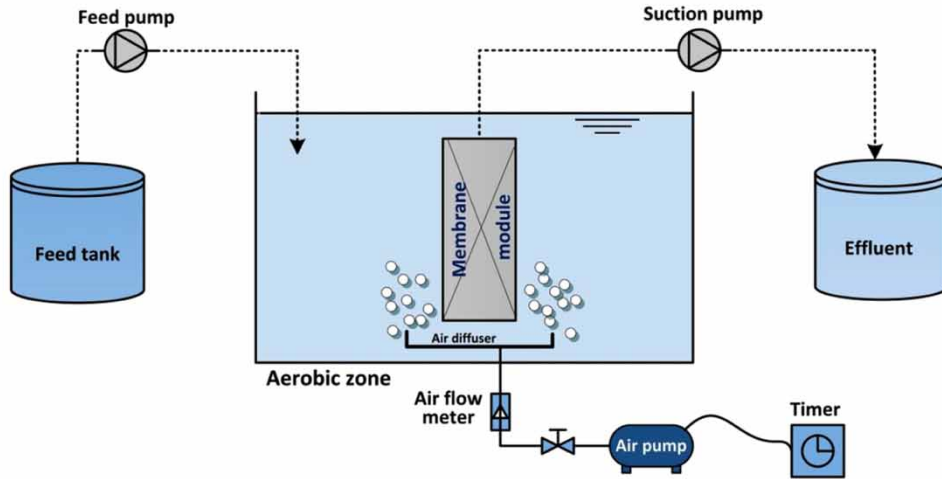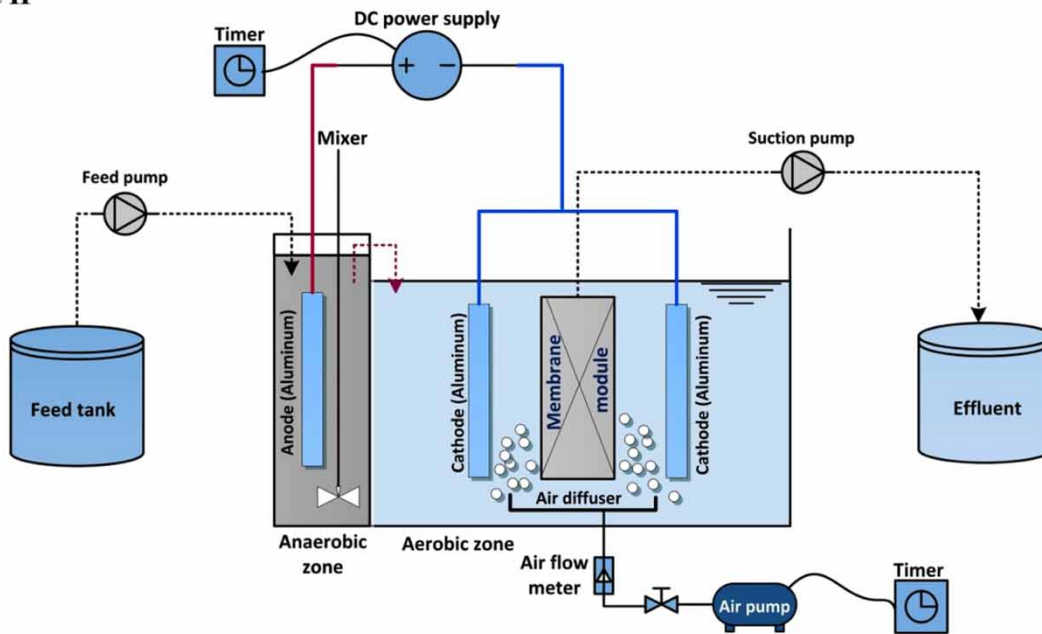
Figure 1 | Schematic of the experimental setup.

were connected to a digital DC power supply maintained at a constant 1.4 V. Based on some other studies, applying a DC field directly in the activated sludge may be harmful for microorganism activities (Bani-Melhem & Elektorowicz 2011; Bani-Melhem & Smith 2012). Further, Li *et al.* (2001) showed that when the electric current was high, the nitrifying bacteria metabolism was inhibited and the nitrification rate in the biofilm was reduced (Wei *et al.* 2011). Therefore,

the optimal electric field for microorganisms is 0.28–1.4 V/cm (Bani-Melhem & Elektorowicz 2010). Accordingly, we attempted to prevent the direct contact of the electric current of 1.4 V/cm with the microbial biomass, using the pre-anaerobic unit containing aluminum anode electrodes before the MBR unit with cathode electrodes. A time programmable switch (SZR M2, Turkey) was attached to a power supply to produce an electric field of sufficient

magnitude and provide an exposure mode of 2 min ON/ 4 min OFF.

Feed characteristics

To obtain consistency in the chemical and physical properties of the influent, both bioreactors were fed with synthetic wastewater.

The synthetic wastewater characteristics were as follows (in mg/L): glucose (465), KH_2PO_4 (19.7), NH_4HCO_3 (127.6), $MgSO_4.7H_2O$ (162), $CaCl_2.2H_2O$ (25.2), and 0.3 mL of trace solution per litre.

The trace elements were prepared by dissolving the following compounds in the synthetic solution (in mg/L): $FeCl_3.6H_2O$ (0.45), $CuSO_4.5H_2O$ (0.009), H_3BO_3 (0.045), KI (0.054), $MnCl_2$ (0.036), $Na_2 MoO_4.2H_2O$ (0.018), $ZnSO_4.7H_2O$ (0.036), $CoCl_2.6H_2O$ (0.045) (Ahmadi et al. 2015).

Table 1 shows the characteristics of the prepared synthetic wastewater in this study.

The activated sludge for inoculation was obtained from the WTP in Tabriz (The capital of East Azerbaijan Province, north-west of Iran). The sludge was acclimatized for 2 weeks prior to membrane filtration experiments.

Analytical methods

During the study, samples were taken from the influent and effluent wastewater on a daily basis and analyzed using the spectrophotometric method (OPTIZEN 2120 + MECASYS Company) for chemical oxygen demand (COD), ammonia nitrogen (NH_3^+-N), and orthophosphate (PO_4^{3-}-P).

DO and temperature (T) levels were measured using a DO meter and pH was measured using a pH meter (Microprocessor, RE 357, UK).

Table 1 | Variability of characteristics of synthetic wastewater used in this study

Item	Average value ± (standard deviation)
COD (mg/L)	440 ± (21.35)
NH_3^+-N (mg/L)	22.95 ± (1.56)
PO_4^{3-}-P (mg/L)	4.83 ± (0.21)
Temperature (°C)	24.38 ± (0.52)
pH	7.30 ± (0.04)

Mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were analyzed according to the Standard Methods (Part 2540 D) (2005 20th edn). A 50 mL sample of mixed liquor from the aerobic zone was taken every 2 days to measure MLSS. In addition, for the sludge volume index (SVI), a sample of the mixed liquor of the aerobic zone was measured and returned to the reactor soon after.

Experimental procedure

The MBR and SMEBR$^+$ systems were continuously operated at room temperature (24.38 ± 0.52 °C) for 48 days. The experimental period was divided into two consecutive stages and each stage was operated for 24 days. Prior to starting the stages, the activated sludge was obtained from the WTP for inoculation.

The sludge was acclimatized for 2 weeks prior to membrane filtration experiments. At the start of the experimental Stage I, initial MLSS in the bioreactor was nearly 2,100 mg/L.

During the implementation of the experimental Stage I, the MBR system was operated in an initial membrane flux. The fouling behavior was evaluated by measuring the decline of permeation flux with time. On a daily basis, the membrane flux was measured by a permeate volume through the membrane module. The membrane permeate flux was reduced to its initial value in order to compare the amount of membrane fouling in the MBR and SMEBR$^+$.

The quantitative determination of the permeation flux (J) in L/m^2·h was calculated based on Equation (1) (Bani-Melhem et al. 2015):

$$J = \frac{Q}{A_m} \tag{1}$$

where Q is the effluent flow rate (L/h) evaluated by measuring the accumulated effluent volume with time and A_m is the membrane surface area (m^2).

No backwashing of the membrane module was performed during the operation period; however, in order to enhance the recovery of the membrane permeability during the operating period in each stage, physical washing events of the membrane were conducted outside the reactor

for a few minutes. Therefore, when the membrane flux decreased over time, the membrane was removed from the aerobic zone and cake on the membrane surface was removed with tap water and then returned to the reactor. After that, the effluent in Stage I was suctioned out by a peristaltic pump.

Prior to the beginning of Stage II, the activated sludge was injected into the pre-anaerobic reactor containing aluminum anode electrodes, and then operation of the SMEBR$^+$ was commenced. Initial volumes of MLSS in the pre-anaerobic reactor and the SMEBR$^+$ were nearly 1,000 mg/L and 4,472 mg/L, respectively. In the second stage, the SMEBR$^+$ was operated by a DC with a mode of 2 min ON/4 min OFF at a constant voltage of 1.4 V. The main reactions which occur during the EC in the pre-anaerobic reactor are as follows (Bani-Melhem & Smith 2012).

- At the anode:

$$Al(s) \rightarrow Al^{3+} + 3e^- \qquad (2)$$

The main reactions which occur during the EC in the aerobic reactor are as follows (Bani-Melhem & Smith 2012).

- At the cathode:

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_2(g) + 3OH^- \qquad (3)$$
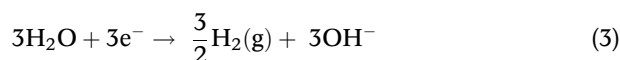
Then, the aluminum ions generated from the anode electrode enter into the aeration zone containing cathode electrodes. They then react with hydroxide ions generated by the cathode electrodes which produce aluminum hydroxide and cause fast nutrient absorption with a large surface area and therefore, decline membrane fouling.

- In the solution:

$$Al^{3+}(aq) + 3H_2O \rightarrow Al(OH)_3 + 3H^+(aq) \qquad (4)$$
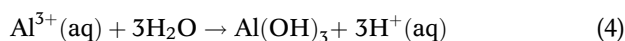
In part two of the experimental work shown in Figure 1, all the steps in Stage I are performed in Stage II.

Sludge retention time (SRT) is one of the most important performance parameters affecting MBR performance

(Grelier *et al.* 2006). The aerobic chamber SRT for each stage was about 24 days. Therefore, sludge discharge was only performed for measuring the MLSS during the reactor operation. The hydraulic retention time (HRT) was 12.5 h and 24 h, respectively in the anode and cathode chamber.

RESULT AND DISCUSSION

Effluent quality

Figure 2 shows the changes of COD, PO_4^{3-}-P, and NH_3^+-N concentration and turbidity within 48 days of the performance period of MBR and SMEBR$^+$.

Figure 2 also illustrates variations in the concentrations of COD, PO_4^{3-}-P, and NH_3^+-N in the SMEBR$^+$ effluent to be in the range of 3–23 mg/L, 0.9–1.6 mg/L, and 1.9–3 mg/L, respectively.

The results show that in Stage I as a reference stage, the percentage removals of COD, PO_4^{3-}-P, and NH_3^+-N were found to be 96%, 61% and 83%, respectively, which respectively increased to 98%, 76% and 90% in Stage II. The average amount of turbidity decreased to less than 1.07 ± 0.22 NTU (nephelometric turbidity units) in Stage II while in Stage I, this amount was 2.2 ± 0.64 NTU.

For the past few years, studies on the combination of MBR with EC have reported that it increases the effluent quality.

Hasan *et al.* (2014) applied an intermittent current density of 12 A m^{-2} (supplied as 5 min ON/10 min OFF) and the results demonstrated that average removals of ammonia (as NH_3^+-N), phosphorus (as PO_4^{3-}-P), and COD were 99%, 99% and 92%, respectively, and membrane fouling was reduced.

Bani-Melham & Elektorowicz (2011) applied an intermittent DC with an operation mode (1 V/cm, 15 min ON/ 45 min OFF). The results revealed that the use of an iron cathode and anode around the membrane module reduced both the nitrifier bacteria level of activity and nitrates and ammonia removal. Therefore, we attempted to apply anode and cathode electrodes in separate zones.

Advantages of the SMEBR$^+$ in municipal synthetic wastewater treatment were observed in reducing ammonia nitrogen. Moreover, the performance of NH_3^+-N removal was affected by the applied DC fields. Our results
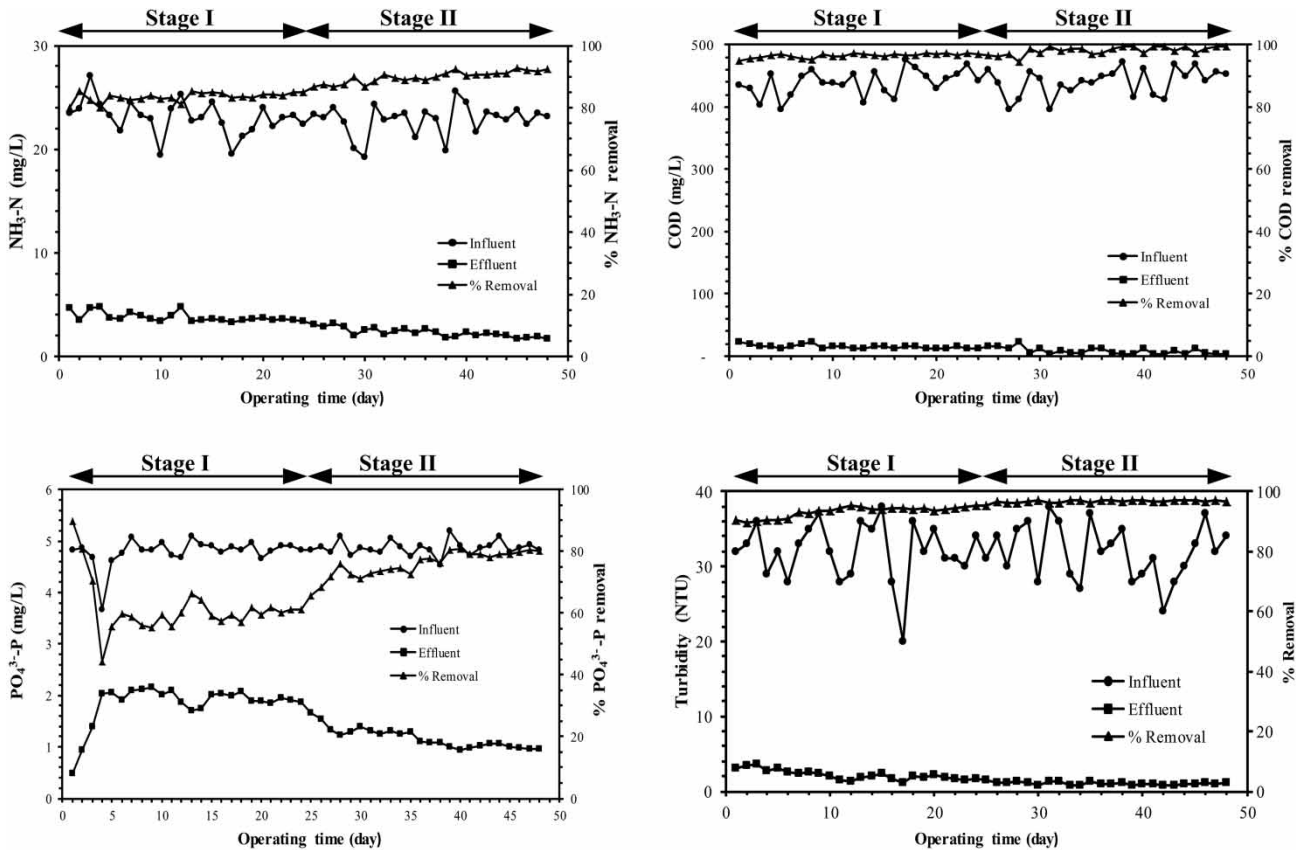
Figure 2 | Comparison of influent, effluent and removal rate of Stage I and Stage II.

showed that the SMEBR$^+$ can provide effective removal of NH$_3^+$-N. In Stage II, removal of ammonia nitrogen was increased to nearly 90%, as compared to 83% in Stage I. One reason to explain this may be the placement of a pre-anaerobic reactor containing anode electrodes which prevents the microbial biomass direct collision with the electrical current, leading to protection of microbial biomasses from electric shock. Another reason could be related to intermittent aeration.

In the case of intermittent aeration, denitrification and nitrification conditions were provided by 20 minutes of aeration on and 20 minutes off. When the air pump was on, the DO concentration in the wastewater reached an amount between 3.5 and 4.5 mg/L, while in the off-pump state, this amount decreased to 0.5 mg/L.

In this study, before starting the experimental stages, the acclimation process was carried out for 2 weeks. Then, the removal efficiency of NH$_3^+$-N at the first day started at

80% and was then stabilized at about 85% at the end of the 24th day of Stage I.

In the study of Bani-Melhem & Elektorowicz (2011), the NH$_3^+$-N removal rate decreased with the application of a DC electric field during the first days. In contrast to the findings obtained in the study of Bani-Melhem & Elektorowicz (2011), we observed that removal rates of NH$_3^+$-N increased over time in Stage II, and the effective removal rates fluctuated between 87 and 92% with an average value around 90% until the end of this stage. This improvement likely resulted from the interactions in the SMEBR process between aluminum ions and NH$_3^+$-N. This observation reveals that the removal of ammonia would be more significant with the application of DC fields than by other processes.

Figure 2 presents the removal efficiency of PO$_4^{3-}$-P in the MBR and SMEBR$^+$ systems. The influent phosphorus varied from 4.8 to 5.2 mg/L with an average of 4.87 ± 0.14 mg/L. The concentrations of PO$_4^{3-}$-P in the effluent fluctuated

between 0.9 and 1.6 mg/L in Stage II. Figure 2 also shows that the SMEBR$^+$ system had good PO$_4^{3-}$-P removal performance, which increased to over 76% on average after applying the DC, while in Stage I (MBR), the average removal was only 61%.

A quick decline in the PO$_4^{3-}$-P removal in Stage I (less than 45%) and rather lower removal of PO$_4^{3-}$-P in the effluent occurred at the 4th day, which could be attributed to the lower MLSS concentrations on that day. Then a few days later, the system again improved its efficiency by the enhancement of the MLSS concentration in the MBR (Stage I).

Beginning from Stage II (day 36 to day 48), after applying a DC field into the MLSS solution, improvement was observed in the PO$_4^{3-}$-P removal. This improvement can be attributed to the electrokinetic phenomenon in which PO$_4^{3-}$-P ions reacted with the generated aluminum ions from the anode electrode in the pre-anaerobic reactor.

The aluminum ions generated from the anode electrode enter into the aeration zone containing cathode electrodes. They then react with hydroxide ions generated by the cathode which produce aluminum hydroxide and cause fast phosphorus absorption with a large surface area. Generated Al^{3+} also react with PO$_4^{3-}$-P and generate AlPO$_4$(s) and thereby remove phosphorus.

Bani-Melhem & Smith (2012) described the mechanism of decreasing phosphorus by the EC process based on Equations (2)–(4). According to their study, the main reaction that may occur during the EC is as follows.

- In the solution:

$$Al^{3+} + PO_4^{3-} \rightarrow AlPO_4 \ (s) \tag{5}$$

In Stage I, where no EC process occurred, there was less chance for the existing phosphorous molecules to react with aluminum ions. Hence, the removal efficiency of phosphorus in Stage I was less than that in Stage II.

During the experimental period, the SMEBR$^+$ pilot reduced the average COD concentration in the influent from 441 ± 22 mg/L to an average output of 8 ± 5.58 mg/L (98% removal) using a pre-anaerobic reactor containing the anode electrode.

Figure 2 illustrates that both stages can often provide higher COD removal. The total COD removal efficiency of Stage I was preserved at a high level, surpassing 96% as a result of the efficient filtration of the membrane module. These data indicated that the membrane module played an important role in supplying reliable effluent quality during the MBR processes (Le-Clech *et al.* 2006).

The usage of a DC field within the MLSS solution improved the COD removal efficiency in Stage II, and the overall COD removal efficiency was kept at nearly 98%, up to the end of Stage II.

The removal efficiency of COD in the EC process depends on the quantity of generated aluminum, which can be attributed to the applied current during the electrolysis time. However, slight improvement in COD removal was observed in Stage II because of the application of the anode electrode in the pre-anaerobic reactor. Moreover, by applying an electric field, the electrochemical redox reactions occurred at the electrode surface (Thrash & Coates 2008).

In general, the mechanism of EC for wastewater treatment is very complex. The COD removal mechanism may be due to biodegradation, electrochemical oxidation, and absorption (Moreno-Casillas *et al.* 2007).

In this study, aluminum was used as the material for both the anode and cathode. When the DC field is employed, aluminum ions (Al^{3+}) can be released from the anode into the reactor, causing flocculation of colloidal particles (Bouamra *et al.* 2012). As time passes by, they are finally converted into the long chain of Al complexes. In the appropriate pH condition, Al(OH)$_3$ is produced based on Equation (4) (Bani-Melhem & Smith 2012).

Thus, the generated Al^{3+} are combined with negative ions in the wastewater and negatively charged colloids are combined and produce larger flocs that cause removal of COD. Defrance *et al.* (2000) reported that membrane fouling is caused not only by the microbial flocs, but also by the supernatant containing colloids and solutes.

Consequently, the use of a DC field in the mixed liquor can reduce the organic materials loading on the membrane surface and thus contribute to improvement in membrane permeability.

Variation of MLSS

Figure 3 shows a variety of MLSS concentrations during the entire operation of both systems. Development of biomass
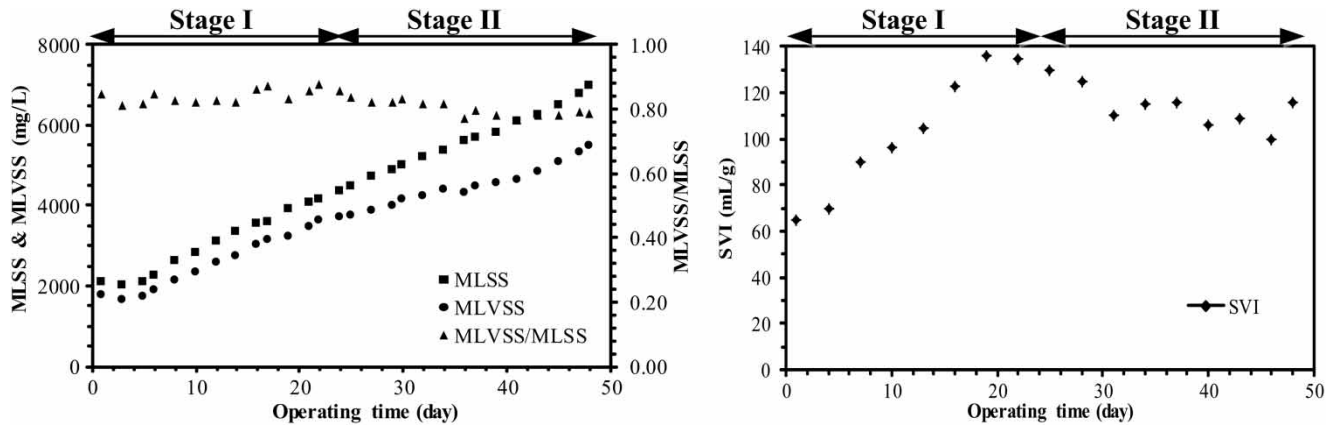
Figure 3 | Variation of SVI, MLSS, MLVSS and MLVSS/MLSS ratio during operation.

concentration in both stages was evaluated by determining the concentration of MLSS in the bioreactors. Since each of the two stages was consecutively operated under the complete SRT condition, MLSS concentration levels in stages I and II, were in the range of 2,000–4,332 mg/L and 4,472–6,980 mg/L, respectively.

At the end of Stage I, Stage II began with application of the electrodes and pre-anaerobic treatment. The EC fundamentally changed the properties of the sludge; the MLSS concentration in this stage was more than that in Stage I. As an important result, MLVSS/MLSS ratio in Stage II was less than that in Stage I. The increase in the MLSS concentration was related to the production of monomeric and polymeric materials in suspended sludge during the electrokinetic process (Hasan *et al.* 2012). However, in Stage I, the MLVSS/MLSS ratio of sludge was in the range of 81–87%, which was reduced to 76–83% during Stage II. Thus, a significant reduction of about 4–5% was observed in the ratio of MLVSS/MLSS. This could be explained by inorganic solids participation (such as $AlPO_4$, $Al(OH)_3$) as a result of EC/flocculation.

The SVI, defined as the volume (in mL) occupied by 1 g activated sludge after 30-min settling of an aerated suspension, was used to characterize the settleability of a specific sludge (Tafti *et al.* 2015). Accordingly, an activated sludge with a SVI below 120 mL/g was considered satisfactory, and that over 150 mL/g was considered bulking (Hasan 2011).

Figure 3 shows that the SVI in Stage I and Stage II respectively reached 102 ± 5.0 mL/g and 114 ± 11 mL/g, the latter demonstrating good settling properties in 24 d

SRT. This is in disagreement with Tian & Su (2012) who reported inverse relationship between SVI and SRT. Their results showed that the SVI value was 78 mL/g at SRT of 30 d, indicating poor settling properties at higher SRT. Therefore, electrokinetics improved the settleability of the sludge in the SMEBR$^+$ by producing dense flocs with good settling properties where SVI in Stage II at SRT of 24 d reached 114 ± 11 mL/g.

Membrane filtration performance

Figure 4 shows the change in the permeate flux (J/J_i) in the MBR (Stage I) and SMEBR$^+$ (Stage II). Since both the processes were performed at constant suction pressure, membrane fouling can often cause change in the permeate flux over time. Therefore, membrane fouling was determined by measuring the permeate flux change.
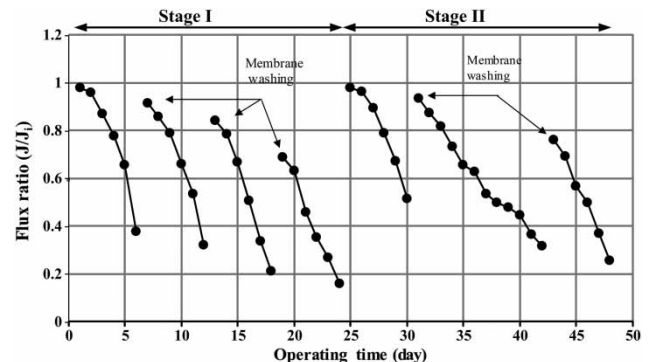
Figure 4 | Comparison of membrane permeability between MBR and SMEBR$^+$.

The corresponding percentage reduction in the permeate flux (PRPF) in each stage was used according to the following equation (Bani-Melhem & Smith 2012):

$$PRPF = \left(\frac{J_i - J}{J_i} \right) \times 100\% \tag{6}$$

where J_i is the initial permeate flux (7.05 L/m^2·h) measured during the first minute of each process and J is the permeate flux at any time.

Figure 4 shows that the permeate flux rapidly decreased during the first 6 days of the MBR system operation without application of electric field and the flux rate (J/J_i) was reduced to 0.377, which led to a rapid HRT increase in the MBR due to the decrease in the permeate flux.

The curve in Figure 4 is split into two stages: from day 1 to day 24 (Stage I), and from day 25 to day 48 (Stage II). The peaks appearing in the filtration performance curve (day 7, day 13, day 19, day 31, and day 43) represent the washing events of the membrane with tap water for a few minutes outside the reactor, when the membrane flux decreased over time. However, the flux decline was sharper. This could be attributed to the blocking of small particles within the membrane pores which could not be removed by washing the surface of the membrane alone.

In common, many factors contributed to membrane fouling such as: (1) membrane characteristics, including physical parameters likes pore size and configuration and chemical parameters such as hydrophobicity and membrane construction material; (2) feed-biomass characteristics that could be extracellular polymeric, SMP, floc characteristics, floe size, and nature of feed and concentration; (3) operating conditions such as aeration rate, solid retention time, HRT, and viscosity; and (4) operational mode like constant flux operation.

In Stage I and II, in order to control fouling caused by the deposition and thickening of sludge cake on the membrane surface as well as the increase in the permeate flux, the membrane module was taken out from the bioreactor and then returned to the reactor after a few minutes of physical removal of sludge cake by washing with tap water.

In Stage I, fewer repulsive forces between membrane surfaces and negatively charged particles cause particles to enter more quickly onto the membrane surface. During the 24-day operation in Stage I, the membrane surface required cleaning three times.

The results obtained in the second stage showed that reduction in membrane fouling and increase in permeate flux were improved by applying an electric field in the SMEBR$^+$. Repulsive forces between the membrane surface and the activated sludge particles with a negative charge were increased by applying an electric field which prevented their absorption on the membrane surface.

As shown in Figure 4, in the second stage of the reactor, the amount of flux (J/J_i) reached 0.515 within the first 6 days, indicating an improvement of 22.02% in membrane permeability compared to Stage I. Moreover, in Stage II, the number of times physical cleaning of the membrane surface was required was reduced in comparison with the first stage.

During the first operation in Stage I, the PRPF was 62.2% after 6 days (between days 1 to 6) of operation. At the end of Stage I, the PRPF reached 84% (day 24).

However, the PRPF in Stage II was 48.5% between days 24 to 30 of the operation. At the end of Stage II, the PRPF reached 74.4% (day 48).

The percentage improvement in the permeate flux was calculated by the following equation (based on 6 days of continuous operation in each stage) (Bani-Melhem & Smith 2012):

$$\%\eta = \left(\frac{PRPF_{stage\ I} - PRPF_{stage\ II}}{PRPF_{stage\ II}} \right) \times 100\% \tag{7}$$

The percentage improvement in the permeate flux is equivalent to 22.02% after applying a DC field to the MLSS solution.

This result is consistent with the studies of Bani-Melhem *et al.* conducted on synthetic wastewater treatment in 2011, which achieved 16.3% improvement in membrane permeability (Bani-Melhem & Elektorowicz 2011) and on grey water treatment in 2012, which achieved 13% reduction in membrane fouling (Bani-Melhem & Smith 2012).

In conclusion, our study showed that if an electric field is applied at the cathode electrodes around the membrane module and also at the anode electrode immersed in the pre-anaerobic reactor, the small colloids and particles become larger due to the electrokinetic process. Therefore,

their contribution to fouling resistance decreases, causing higher permeate flux.

CONCLUSION

In the present study, a SMEBR coupled with a pre-anaerobic reactor containing anode electrodes (SMEBR$^+$) was successfully developed to reduce the membrane fouling, maintain the permeate flux and also enhance the quality of the treated wastewater.

In the pilot-scale study, the reactor was fed with synthetic wastewater and the fouling rate decreased by about 22.02% in Stage II. This improvement is probably due to a number of factors, including the repulsive forces between the membrane surface and the negatively charged sludge particles when an intermittent electric field was applied, formation of large size sludge particles, and the decrease in the production of EPS in the pre-anaerobic reactor containing the anode electrode. Furthermore, removal of COD, NH_3^+-N, and PO_4^{3-}-P was greater in Stage II than in Stage I.

The application of intermittent aeration in the aerobic zone created conditions for simultaneous nitrification and denitrification and therefore, increased NH_3^+-N removal.

According to the results, this system can be considered as a promising alternative to conventional aeration treatment systems as it decreases the membrane fouling and also improves wastewater quality.

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