Electricity production and sludge reduction by integrating microbial fuel cells in anoxic-oxic process

Benyi Xiao^{a,b}, Meng Luo^{a,b}, Xiao Wang^c, Zuoxing Li^{a,c}, Hong Chen^d, Junxin Liu^{a,b}, Xuesong Guo^{a,b,*}

^a Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

^b University of Chinese Academy of Sciences, Beijing 100049, China

^c School of Environment Science and Spatial Informatics, China University of Mining and Technology, Xuzhou 221116, China

^d School of Hydraulic Engineering, Changsha University of Science and Technology, Changsha, Hunan 410004, China

ARTICLE INFO

Article history:

Received 10 October 2016

Revised 22 June 2017

Accepted 25 June 2017

Available online 1 August 2017

Keywords:

Anoxic-oxic process

Electricity production

Microbial fuel cell

Municipal wastewater

Sludge reduction

ABSTRACT

To produce energy and reduce sludge production from the treatment of municipal wastewater, four identical microbial fuel cells (MFCs) were constructed in an anoxic-oxic (A/O) process (MFCs-A/O system). Experimental results indicated that this system enhance the removals of chemical oxygen demand (COD) and total nitrogen (TN). The electricity produced by each MFC were ranged from 0.371 to 0.477 V (voltage) and from 138 to 227 mW/m³ (power density) at the stable stage, when the external resistance was fixed at 1000 Ω. The coulombic efficiency of the MFCs-A/O system ranged from 0.31% to 1.68% (mean = 0.72%) at the stable stage, respectively. The removals of COD and TN in the MFCs-A/O system were slightly higher than those in the control system. Compared with the control system, the MFCs-A/O system can reduce waste activated sludge production and sludge yield by 24.0% and 24.2%, respectively. The experimental results indicated that the MFC constructed in A/O system improves wastewater treatment and the MFCs-A/O system can produce electricity while reducing sludge production and increasing wastewater treatment efficiency.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Fossil fuels currently account for over 80% of the primary energy consumed worldwide (Bardhan et al., 2015) and their extensive utilization has led to serious energy crisis and numerous global environmental problems (Kumar et al., 2016; Qi et al., 2016). The need to mitigate these ongoing issues is urgent; therefore, new, renewable, and environmentally friendly sources of energy need to be developed (Kumar et al., 2016; Qi et al., 2016). Electricity production using a microbial fuel cell (MFC) is one promising method (Logan and Rabaey, 2012; Mohan et al., 2014; Moqsud et al., 2013). MFC is a biochemical-catalyzed electrochemical system that converts chemical energy to electrical energy by oxidizing biodegradable organic matter via the catalytic reaction of microorganisms (Logan and Rabaey, 2012; Mohan et al., 2014; Moqsud et al., 2013). MFC provides a new opportunity for direct electricity generation from renewable and biodegradable materials using active microorganisms. Furthermore, it is considered as a promising sustainable technology to meet increasing energy needs and uses the

organic contaminants in wastewater as substrates (Logan and Rabaey, 2012; Mohan et al., 2014; Li et al., 2014; Moqsud et al., 2013). MFC is regarded as a highly adaptable technology for sustainable wastewater treatment for several reasons: (1) electric energy is directly produced and value-added products are generated; (2) the process produces good quality effluent with a low environmental footprint; (3) the process provides real-time monitoring and control; and (4) there is operation stability (Li et al., 2014).

However, to make it suitable for real-world applications, the MFC has to be combined with various wastewater treatment processes or systems (Li et al., 2014). Some wastewater treatment processes have been combined with MFC including activated sludge process (Li et al., 2014), anaerobic digestion (Li et al., 2014), membrane bioreactor (Li et al., 2014), anoxic-oxic (A/O) process (Chang et al., 2014; You et al., 2010), anaerobic-anoxic-oxic (A²O) process (Xie et al., 2014, 2016). The A/O process is a common wastewater treatment process, which has been applied to treating a wide range of municipal and industrial wastewaters by removing chemical oxygen demand (COD), ammonium (NH₄⁺-N), total nitrogen (TN), and suspended solids (SS) (Chan et al., 2009; Chang et al., 2014; Rasool et al., 2014). The A/O process has two stages, the anoxic stage and oxic stage, which are separated and distinguished by

* Corresponding author at: Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China.

E-mail address: gxs0122@rcees.ac.cn (X. Guo).

different dissolved oxygen (DO) levels (Chan et al., 2009). The DO in the anoxic stage is lower than 0.2 mg/L and is 2–4 mg/L in the oxic stage. In a two-chamber MFC, a typical type of MFC, the anode chamber is an anaerobic, or anoxic environment while the cathode chamber is an aerobic (oxic) environment. The two chambers are separated by a proton exchange membrane (PEM) or other types of membranes (Madani et al., 2015). Therefore, the environmental conditions of a two-chamber MFC are similar to those of the A/O process and it can be integrated in the A/O process. Researchers have been studying the combined MFC and A/O process system (MFC-A/O system). For example, Chang et al. (2014) studied the treatment of pharmaceutical and personal care product (PPCP)-containing sewage with a pilot-scale MFC-A/O system, and You et al. (2010) studied the treatment of saline seafood wastewater and electricity generation in a U-tube MFC-A/O system.

Sludge is an important by-product of the activated sludge process, which is the process applied in the oxic stage of A/O. The sludge yield in normal activated sludge process is approximately 0.3–0.5 g VSS (volatile suspended solids)/g COD or 0.5–0.8 g SS/g COD (SS/VSS = 0.6) (Metcalf and Eddy, 2003). The improper treatment and disposal of sludge will lead to some environmental issues, such as greenhouse gas emissions and secondary pollution (Li et al., 2015). Although sludge can be used to produce energy materials such as methane (Xiao et al., 2014), hydrogen (Sun et al., 2014), and electricity (Xiao et al., 2014), its treatment and disposal costs are still high (Guo et al., 2013). Because of its mass-yield, sludge treatment and disposal has become an important issue for many wastewater treatment plants, and reducing sludge production in the wastewater treatment is very significant (Guo et al., 2013). Sludge production is expected to be low in the wastewater treatment using MFCs because part of organic matter energy is converted into electricity by electricity-producing microorganisms (electricigens). The cell yield of electricigen (0.07–0.16 g VSS/g COD) is much less than that of normal activated sludge microorganisms (Huggins et al., 2013; Logan and Rabaey, 2012; Logan et al., 2015). Research has been conducted on reducing sludge production in wastewater treatment using MFCs (Gajaraj and Hu, 2014; Su et al., 2013). For instance, Su et al. (2013) reported the sludge production could be reduced by approximately 5.1% in a system combining a MFC and a membrane bioreactor. Later, Gajaraj and Hu (2014) noted that the combination of activated sludge processes with MFCs could reduce sludge production by approximately 6–11%. To date, however, the reduction of sludge production in wastewater treatment systems integrated with MFCs is still low, and no studies have been conducted so far on sludge production in the MFC-A/O process.

It is possible to simultaneously produce electricity, treat wastewater and reduce sludge production by integrating MFCs in the A/O process. Meanwhile, it is very important to produce electricity with practical meaning sludge reduction for applying MFC in practical wastewater treatment. The novelty of this study is to obtain practical meaning sludge reduction in wastewater treatment by enhancing the conversion of organic matters to electricity of integrating MFC. Therefore, the aims of this study are to investigate electricity production from practical municipal wastewater using the MFC-A/O process, as well as reduction of sludge production and wastewater treatment efficiencies of this process.

2. Material and methods

2.1. Municipal wastewater

The municipal wastewater, used in this study, was collected from a residential quarter of Beijing, China. The characteristics of this wastewater are summarized in Table 1.

Table 1

The characteristics of municipal wastewater used in the test.

Index	Range	Average
COD (mg/L)	108–371	203
pH	7.4–8.4	7.8
SS (mg/L)	24–451	165
NH ₄ ⁺ -N (mg/L)	71.7–90.8	79.8
TN (mg/L)	73.8–93.3	82.8
C/N	1.3–4.7	2.5

2.2. MFC-A/O system

Two A/O process apparatuses, which were made of polyvinyl chloride, were used in the test. One A/O process apparatus was constructed with four identical MFCs with external resistances (MFCs-A/O), while the other, used as control system, was constructed with four MFCs without external resistances (i.e., open MFC). The work volumes of the two A/O process tanks were 1 L (10 × 10 × 10 cm, anoxic tank) and 4 L (25 × 20 × 10 cm less 10 × 10 × 10 cm, oxic tank). To fully use the working volume of A/O process, the anoxic tank was placed in the oxic tank, and the four identical MFCs were constructed in the A/O process as follows (Fig. 1). Four carbon felt brushes were placed in the anoxic tank and used as the MFC anodes. Four carbon felt brushes, were placed in the oxic tank and used as the cathodes. Four PEMs (Ø = 5 cm) were installed in each of the four walls of anoxic tank. Then copper wires were used to connect the four external circuits with four fixed external resistances (1000 Ω) in the MFCs-A/O system. The anode carbon felt brushes were made of four carbon felt pieces (6 × 2 × 1 cm), which were fixed on a graphite rod (0.8 cm in diameter by 12.0 cm in length). The cathode carbon felt brushes were made of four carbon felt pieces (6 × 1 × 1 cm) which also fixed on a graphite rod (0.8 cm in diameter by 12.0 cm in length). The two electrodes were placed 2 cm apart for each of the four MFCs.

2.3. Wastewater treatment and electricity production

The wastewater was continuously pumped into the anoxic tank of systems with a flow velocity of approximately 9.26 ml/min using a peristaltic pump (Lange Co., China). The anoxic tank was mixed with a slow-speed stirrer to maintain low DO levels between 0.2 and 0.5 mg/L. To maintain an aerobic environment, the oxic tank was aerated with an aerator to maintain approximately 2.0 mg/L of DO. The settled sludge from the settlement tank was returned to both the anoxic and oxic tanks. The sludge return rates for the two tanks were 25%, with a flow velocity of about 2.31 ml/min, in the anoxic tank, and 75%, with a flow velocity of about 6.94 ml/min, in the oxic tank. The wastewater hydraulic retention times in the anoxic and oxic tanks were 1.8 h and 7.2 h, respectively. The activated sludge in the oxic tanks of two systems was periodically discharged as waste activated sludge (WAS) to maintain a stable sludge concentration (2.0–3.0 g SS/L).

2.4. Analytical methods and calculation of electrical parameters

The soluble COD (SCOD) and total COD (TCOD) of the influent and effluent for the two systems were quantified using a COD measurement system (HACH DR2800, USA) and COD kit (HACH 20–1500 mg COD/L, USA). The samples were filtered using 0.45 µm membranes before determining their SCOD. The pH of the wastewater was measured with a pH meter (Sartorius PB-10, Germany). The sludge concentrations (SS and VSS) were measured using the weight method, and NH₄⁺-N and TN were measured using the spectrophotometric method (APHA, 1998). Wastewater TC was

Fig. 1. Schematic of the MFCs-A/O system and a photograph of the experimental apparatus. A: Plan view; B: Front view; C: Photograph.

measured with a TOC meter (Shimadzu TOC-V CPH, Japan). The production, reduction, and yield of sludge were calculated using previously described methods (Guo et al., 2014). The sludge activities in the oxic tanks were assessed on the basis of specific oxygen uptake rate (SOUR). The DO of activated sludge was measured with a DO meter (WTW Oxi3310, Germany). Oxygen uptake rate (OUR) was determined using the slope of the linear portion of the DO versus time curve. The specific oxygen uptake rate (SOUR) was calculated by dividing the OUR by the VSS concentration (APHA, 1998).

The output voltages of MFCs were recorded using a precision digital multimeter and a data acquisition system (Beijing Ruibohua Co. China) connected to a computer. The polarization curves of MFC were determined by lowering the external resistances from 50,000 Ω to 0 Ω at an interval of two minutes to stabilize the voltages when the MFC performance was consistent (Gajjaraj and Hu, 2014). Open circuit voltage, internal resistance, and maximal power density were obtained from analyzing the polarization curves. The coulombic efficiency (CE) of MFCs was estimated by integrating the measured current relative to the theoretical current on the basis of consumed COD (More and Ghangrekar, 2010),

$$E = \frac{C_E}{C_T} \times 100$$

The theoretical current production, C_T , was estimated as

$$C_T = \frac{F \times n \times w}{M}$$

where F = Faraday constant (96,485 C/mol), n = number of moles of electrons produced per mole of substrate, $n = 4$ for wastewater COD, w = daily COD load removed, in grams, M = molecular weight of the

substrate, in grams. The actual current production, C_E , was integrated as

$$C_E = I \times t$$

where I = current (A) and t = time (s).

3. Results and discussion

3.1. Electricity production and MFC characteristics

The electricity outputs of the four MFCs in the A/O system during the 54-day test were similar, and the results are summarized in Fig. 2.

In the first stage (0–15 d), the voltage and power density for each MFC were approximately 0.345–0.444 V (mean = 0.400 V) and 119–197 mW/m³ (mean = 160 mW/m³), respectively. After sludge microorganisms acclimated to the growth environment, the system entered a stable stage at 15 d. In this stage, the electricity production of each MFC increased to 0.371–0.477 V (mean = 0.439 V) and 138–227 mW/m³ (mean = 193 mW/m³) due to the electricigens enrichment. Previous studies (Kim and Logan, 2004; Zhang et al., 2008) have reported similar results, namely an increase in electricity production. For example, Zhang et al. (2008) found that the voltage from MFCs continuously increased in their study. The electricity produced by each MFC in the current study was higher than that reported in previous studies (Gajjaraj and Hu, 2014; Sun et al., 2014; Xie et al., 2014), but lower than the results in You et al. (2010). Xie et al. (2014) only obtained an average voltage of 0.169 $\pm$ 0.008 V and an average power density of 14.3 $\pm$ 1.4 mW/m³, with an external resistance of 2000 Ω , when studying the MFC incorporated into an anaerobic–anoxic–oxic

Fig. 2. The electricity production of the four MFCs in the test.

wastewater treatment process with real sewage as the influent. You et al. (2010) obtained a peak voltage of 0.42 V and 8900 mW/m³–16,200 mW/m³ with an external resistance of 500 Ω while studying the treatment of saline seafood wastewater and electricity generation with an MFC-A/O system. The difference in electricity outputs between the current study and previous studies may be attributed to the dissimilar MFC structures and the location of the integrated MFC in the wastewater treatment process (Li et al., 2014; Mohan et al., 2014). For instance, Xie et al. (2014) embedded anode and cathode into the bottom of a 1 L anaerobic tank and 2 L anoxic tank, respectively, without membrane; and You et al. (2010) used a U-tube MFC with a work volume of 452 ml. In this study, the anodes and cathodes were placed in the 1 L anoxic tank and 4 L oxic tanks, respectively. The power density of the MFCs-A/O system, which is the sum of the densities of four MFCs, ranged from 561 mW/m³ to 910 mW/m³ (mean = 740 mW/m³) during the stable stage. The reported power density of the MFCs-A/O system was also higher than those reported in similar studies involving municipal wastewater (Jiang et al., 2011; Zhang et al., 2013), but remained lower than those in similar studies conducted with synthetic (Dekker et al., 2009) and industrial wastewater (You et al., 2010; Zhuang et al., 2012).

The electricity outputs of the four MFCs were similar, therefore, the polarization curves of one MFC were determined at 10 d and 40 d, which presented two stages of electricity production (Fig. 3). Three important MFC parameters can be derived from these curves (Table 2) according to previous studies (Nanchaiah et al., 2016; Kumar et al., 2017). The open circuit voltage and maximal power density of MFC increased from 0.596 V and 482.9 mW/m³ to 0.736 V and 855.4 mW/m³, respectively, when the operation time increased from 10 d to 40 d. Mean-

Fig. 3. The polarization curves of one MFC in the MFCs-A/O system. U-10 d, U-40 d: Voltage-current curves in 10 d, 40 d; P-10 d, P-40 d: Power density-current curves in 10 d, 40 d.

Table 2

Three parameters of one MFC during the electricity generating process.

Operation time (d)	10 d	40 d
Open circuit voltage (V)	0.596	0.736
Internal resistance (Ω)	199	99
Maximal power density (W/m ³)	482.9	855.4

while, the internal resistance of MFC decreased from 199 Ω to 99 Ω , which was consistent with the changes in maximal power density. The increase in open circuit voltage and maximal power density, and decrease in internal resistance suggested that the MFC performance improved with operation. The internal resistance of MFC in this study was lower than that reported by Xie et al. (2014) (6000 Ω) and higher than that obtained by Cha et al. (2010) (17 Ω). These differences may have been caused by the differences in the MFC structure and wastewater characteristics, as mentioned previously.

3.2. Wastewater treatment of the two systems

The primary purpose of the MFCs-A/O system was to serve as a wastewater treatment system, so the wastewater treatment efficiencies of the system were evaluated. Fig. 4 summarizes the changes of COD, NH₄⁺-N and TN in the influents and effluents, as well as their removal, in the test.

Fig. 4A shows that the COD of the influents varied from 108 mg/L to 371 mg/L and that of the effluents for the two systems was in the range of 28–76 mg/L (MFCs-A/O system) and 32–81 mg/L (control system). The COD removal of the MFCs-A/O system was varied between 48.1 and 87.5% (mean = 75.6%), which was higher than those for the control system (39.8–84.9% [mean = 72.2%]). Although the COD removal in two systems was slightly lower than in previous studies (Wang and Chen, 2016; Zhang et al., 2016), the COD of the effluent at the end of experiment was approximately 40 mg/L, which meets the Chinese Discharge Standard (GB18918-2002) (China State EPA, 2002). The NH₄⁺-N removal was another important function for A/O process (Chan et al., 2009; Rasool et al., 2014) and the results of this study are summarized in Fig. 4B. The NH₄⁺-N of the influents varied from 71.7 to 90.8 mg/L and that of the effluents ranged from 2.4 to 6.9 mg/L (MFCs-A/O system) and 3.1–7.0 mg/L (control system). The NH₄⁺-N removal was 92.4–96.7% (mean = 94.4%) (MFCs-A/O system) and 92.2–95.9% (mean = 94.2%) (control system). The TN removal for the two systems was also measured (Fig. 4C). The effluents TN ranged from 39.7 to 51.1 mg/L (MFCs-A/O system) and 37.0–48.2 mg/L

Fig. 4. The wastewater treatment efficiencies of two systems. A: COD; B: NH₄⁺-N; C: TN. Inf: Influent; Eff: Effluent; Rem-MFCs-A/O: Removal of MFCs-A/O system; Rem-Con: Removal of control system.

(control system). The TN removal for MFCs-A/O system was 46.6–51.4% (mean = 49.4%), which was higher than that for control system (43.3–48.9% [mean = 45.8%]). Generally, the MFCs-A/O system had higher COD and TN removals than the control system and similar NH₄⁺-N removal. This suggested that integrating the MFCs in A/O process enhances COD and TN removals, while do not affect NH₄⁺-N removal. These results are in agreement with previous studies (Gajaraj and Hu, 2014; Nancharaiyah et al., 2016; Xie et al., 2014, 2016). For example, Xie et al. (2014) found that integrating the MFC in an A2O process enhanced COD and TN removal by 15.9% and 9.3%, respectively. The similar NH₄⁺-N removal for two systems might be due to the quantity of DO in the oxic tanks of the two systems, which was higher than 2.0 mg/L (Spellman, 2003). Additionally, the TN removal levels for the two systems were lower, which might be ascribed to an inappropriate C/N concentration (Metcalf and Eddy, 2003; Spellman, 2003). In the test, the C/N of the influent shifted within the range of 1.3–4.7 (mean = 2.5) (Table 1), which is

Table 3
Change in sludge SOUR in the oxic tanks.

SOUR	10 d	30 d	45 d
MFCs-A/O system (mg O ₂ /g MLVSS h)	12.18 ± 0.11	13.27 ± 0.08	14.45 ± 0.13
Control system (mg O ₂ /g MLVSS h)	12.22 ± 0.09	13.23 ± 0.10	14.42 ± 0.12

slightly lower than the theoretical C/N ratio for complete biological nitrogen removal (about 2.86) (Bernat et al., 2008) and far lower than the actual C/N ratio (>5.0) needed for nitrogen removal (Zhao et al., 2014).

The SOURs of the activated sludge in the oxic tanks were measured in both systems, and the results are summarized in Table 3. They were in the range of 12.18–14.45 mg O₂/g MLVSS h (MFCs-A/O system) and 12.22–14.42 mg O₂/g MLVSS h (control system), which were in the normal SOUR range of activated sludge (Metcalf and Eddy, 2003). For example Van Aken et al. (2015) found the sludge SOUR were in the range of 2–16 mg O₂/g MLVSS h. The result indicated that the SOURs of sludge were similar in the two systems, which suggests that integrating MFC in the A/O process did not affect the sludge activity in the oxic tank. Because the SOUR index provides a measure of sludge microbial metabolic activity (Metcalf and Eddy, 2003), the similar values in the two systems indicated that the sludge microbial metabolic activity was also similar.

3.3. Coulombic efficiency of the MFCs-A/O process

Fig. 5 depicts the change in coulombic efficiencies for the four MFCs in the test, which indicates their general similarity. The coulombic efficiencies of the four MFCs ranged from 0.08% to 0.42% (mean = 0.18%), and that of the MFCs-A/O system (Total MFC) reached 0.31–1.68% (mean = 0.72%), which decreased with system operation in the first stage (before 27 d) and then stabilized after 27 d. The low coulombic efficiency may have been due the removal of organic matters primarily by normal microorganisms in the activated sludge, rather than having been converted by electricigens. Furthermore, in comparing the MFC electricity outputs and coulombic efficiencies with the COD removal in the experimental MFCs-A/O system (Figs. 2, 4 and 5), the MFC coulombic efficiencies did not increase with the increase of electricity outputs of MFCs and COD removal. In other words, the changes in these parameters were inconsistent. This inconsistency might be due to the simultaneous removal of wastewater COD by both normal sludge microorganisms (Spellman, 2003) and electricigens, which increased with the operation time, and the former organisms had a higher removal rate than the latter. The coulombic efficiencies of the MFCs-A/O system in this study were higher than those reported by Gajaraj and Hu (2014) (0.05%) and lower than those indicated by Su et al. (2013) (5.9%), You et al. (2010) (2.11–15.2%) and Chang et al. (2014) (2.77–25.20%). The results suggests that the coulombic efficiency of MFC-A/O system in this study needs to be further increased although it has been enhanced by the integrating method.

3.4. Sludge production in the two systems

The accumulative WAS productions for the two systems are shown in Fig. 6A, reaching 32.7 g SS (MFCs-A/O system) and 43.0 g SS (control system) at the end of the test. The reduction of accumulative WAS for the MFCs-A/O system was approximately 24.0% at the end of the test (Fig. 6A). The sludge yields of the two systems were also calculated (Fig. 6B). The sludge yield from the control system was stable, approximately 0.40–0.44 g SS/

Fig. 5. Changes in the coulombic efficiencies of the MFCs in the test. Total MFC: Combination of MFCs 1, 2, 3 and 4, where the value is the sum of all four MFCs.

Fig. 6. Accumulative WAS production (A) and sludge yield (B) of the two A/O systems. CON: Control system; MFC: MFC-A/O system.

g COD removed (mean = 0.42 g SS/g COD removed), which was similar to previous studies (Feng et al., 2014; Guo et al., 2014). The sludge yield of the MFCs-A/O system was 0.34–0.41 g SS/g COD removed (mean = 0.37 g SS/g COD removed) at 0–15 d stage and 0.30–0.33 g SS/g COD removed (mean = 0.32 g SS/g COD removed) at 15–54 d stage. In the overall test, the mean sludge yield of the MFCs-A/O system was 0.33 g SS/g COD removed. The reduction of sludge yield for the MFCs-A/O system was 19.5–28.6% (mean = 24.2%) for the 15–54 d stable stage. Electricigens convert part of the energy in organic matter to electricity and the electricigens cell yield is much less than that of the activated sludge

(Huggins et al., 2013; Logan and Rabaey, 2012; Logan et al., 2015). Therefore, it is reasonable that the sludge production in the MFCs-A/O system was lower than in control system. The sludge reduction in this study was higher than that found in previous similar studies of wastewater treatment combined with an MFC (Gajaraj and Hu, 2014) and similar to some sludge reduction processes, such as chemical uncoupling (Chen et al., 2002; Guo et al., 2013, 2014). For example, Gajaraj and Hu (2014) obtained a 6–11% sludge reduction in a combined MFC activated sludge process. Chen et al. (2002) obtained a sludge reduction of 22–44% in an activated sludge culture with 0.5–1.0 mg/L 3,3',4',5-tetrachloro salicylanilide and Guo et al. (2014) obtained a sludge reduction of 22.4% in an anaerobic-anoxic-oxic process with 1.08–1.86 ml/m³ tetrakis (hydroxymethyl) phosphonium sulphate. The results suggested the sludge reduction was increased by enhancing the coulombic efficiency of the integrating MFC. In summary, these prior results suggest that the sludge reduction from the MFCs-A/O system in this study was sufficient to make it a potentially important and practical wastewater treatment method.

4. Conclusions

The electricity outputs of each MFC were within the range of 0.371–0.477 V (voltage) and 138–227 mW/m³ (power density) in the stable stage with a fixed external resistance of 1000 Ω. The power densities of the MFCs-A/O system were within the range of 561–910 mW/m³. MFCs integrated in the A/O process enhanced the removal of COD and TN by 3.4% and 3.6%, respectively, and did not affect the removal of NH₄⁺-N. Moreover, the sludge production was reduced and the reductions of accumulative WAS and sludge yield were respectively, 24.0% and 24.2% at the stable stage.

Acknowledgements

The authors gratefully acknowledge the financial support from the Major Science and Technology Program for Water Pollution Control and Treatment of China (Nos. 2014ZX07101-012 and 2012ZX07203-001) and the National Natural Science Foundation of China (Nos. 51378492 and 51308068). Additionally, the authors thank Editage for its linguistic assistance during the preparation of this manuscript.

References

- APHA, 1998. Standard Methods for the Examination of Water and Wastewater. American Public Health Association, Washington DC, USA.
- Bardhan, S.K., Gupta, S., Gorman, M.E., Haider, M.A., 2015. Biorenewable chemicals: feedstocks, technologies and the conflict with food production. *Renew. Sust. Energy Rev.* 51, 506–520.
- Bernat, K., Wojnowska-Baryła, I., Dobrzyńska, A., 2008. Denitrification with endogenous carbon source at low C/N and its effect on P (3HB) accumulation. *Biores. Technol.* 99, 2410–2418.
- Cha, J., Choi, S., Yu, H., Kim, H., Kim, C., 2010. Directly applicable microbial fuel cells in aeration tank for wastewater treatment. *Bioelectrochemistry* 78, 72–79.
- China State EPA, 2002. The National Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant (GB18918-2002), Beijing, China (in Chinese).
- Chan, Y.J., Chong, M.F., Law, C.L., Hassell, D.G., 2009. A review on anaerobic-aerobic treatment of industrial and municipal wastewater. *Chem. Eng. J.* 155, 1–18.
- Chang, Y.-T., Yang, C.-W., Chang, Y.-J., Chang, T.-C., Wei, D.-J., 2014. The treatment of PPCP-containing sewage in an anoxic/aerobic reactor coupled with a novel design of solid plain graphite-plates microbial fuel cell. *Biomed. Res. Int.* 2014, 765652.
- Chen, G.H., Mo, H.K., Liu, Y., 2002. Utilization of a metabolic uncoupler, 3,3',4',5-tetrachlorosalicylanilide (TCS) to reduce sludge growth in activated sludge culture. *Water Res.* 36, 2077–2083.
- Dekker, A., Heijne, A.T., Saakes, M., Hamelers, H.V.M., Buisman, C.J.N., 2009. Analysis and improvement of a scaled-up and stacked microbial fuel cell. *Environ. Sci. Technol.* 43, 9038–9042.
- Feng, X.-C., Guo, W.-Q., Yang, S.-S., Zheng, H.-S., Du, J.-S., Wu, Q.-L., Ren, N.-Q., 2014. Possible causes of excess sludge reduction adding metabolic uncoupler, 3,3',4',5-tetrachlorosalicylanilide (TCS), in sequence batch reactors. *Biores. Technol.* 173, 96–103.

- Gajaraj, S., Hu, Z., 2014. Integration of microbial fuel cell techniques into activated sludge wastewater treatment processes to improve nitrogen removal and reduce sludge production. *Chemosphere* 17, 151–157.
- Guo, S.X., Yang, J.M., Liang, Y., Liu, J.X., Xiao, B.Y., 2014. Evaluation of sludge reduction by an environmentally friendly chemical uncoupler in a pilot-scale anaerobic/anoxic/oxic process. *Bioprocess Eng.* 37, 553–560.
- Guo, W.Q., Yang, S.S., Xiang, W.S., Wang, X.J., Ren, N.Q., 2013. Minimization of excess sludge production by in-situ activated sludge treatment processes - a comprehensive review. *Biotechnol. Adv.* 31, 1386–1396.
- Huggins, T., Fallgren, P.H., Jin, S., Ren, Z.Y.J., 2013. Energy and performance comparison of microbial fuel cell and conventional aeration treating of wastewater. *J. Microb. Biochem. Technol.* 5, 6, 002.
- Jiang, D., Curtis, M., Troop, E., Scheible, K., McGrath, J., Hu, B., Suib, S., Raymond, D., Li, B., 2011. A pilot-scale study on utilizing multi-anode/cathode microbial fuel cells (MAC/MFCs) to enhance the power production in wastewater treatment. *Int. J. Hydrogen Energy* 36, 876–884.
- Kim, B., Logan, B.E., 2004. Continuous electricity generation from domestic wastewater and organic substrates in a flat plate microbial fuel cell. *Environ. Sci. Technol.* 38, 5809–5814.
- Kumar, Y., Ringenberg, J., Depuru, S.S., Devabhaktuni, V.K., Lee, J.W., Nikolaidis, E., Andersedn, B., Adjei, A., 2016. Wind energy: trends and enabling technologies. *Renew. Sust. Energy Rev.* 53, 209–224.
- Kumar, S.S., Basu, S., Bishnoi, N.R., 2017. Effect of cathode environment on bioelectricity generation using a novel consortium in anode side of a microbial fuel cell. *Biochem. Eng. J.* 121, 17–24.
- Li, M.Y., Xiao, B.Y., Wang, X., Liu, J.X., 2015. Consequences of sludge composition on combustion performance derived from thermogravimetry analysis. *Waste Manage.* 35, 141–147.
- Li, W.W., Yu, H.Q., He, Z., 2014. Towards sustainable wastewater treatment by using microbial fuel cells-centered technologies. *Energy Environ. Sci.* 7, 911–924.
- Logan, B.E., Wallack, M.J., Kim, K.-Y., He, W.H., Feng, Y.J., Saikaly, P.E., 2015. Assessment of microbial fuel cell configurations and power densities. *Environ. Sci. Technol. Lett.* 2, 206–214.
- Logan, B.E., Rabaey, K., 2012. Conversion of wastes into bioelectricity and chemicals by using microbial electrochemical technologies. *Science* 337, 686–690.
- Madani, S., Gheshlaghi, R., Mahdavi, M.A., Sobhani, M., Elkamel, A., 2015. Optimization of the performance of a double-chamber microbial fuel cell through factorial design of experiments and response surface methodology. *Fuel* 150, 434–440.
- Metcalf and Eddy, Inc., Burton, F., Tchobanoglous, G., Tsuchihashi, R., Stensel, H.D., 2003. *Wastewater Engineering: Treatment and Reuse*. McGraw-Hill, New York.
- Mohan, S.V., Velvizhi, G., Modestra, J.A., Srikanth, S., 2014. Microbial fuel cell: critical factors regulating bio-catalyzed electrochemical process and recent advancements. *Renew. Sust. Energy Rev.* 40, 779–797.
- Moqsud, M.A., Omine, K., Yasufuku, N., Hyodo, M., Nakata, Y., 2013. Microbial fuel cell (MFC) for bioelectricity generation from organic wastes. *Waste Manage.* 33, 2465–2469.
- Nancharaiyah, Y.V., Mohan, V.S., Lens, P.N.L., 2016. Recent advances in nutrient removal and recovery in biological and bioelectrochemical systems. *Biores. Technol.* 215, 173–185.
- Qi, J.J., Zhu, F.F., Wei, X., Zhao, L.Y., Xiong, Y.Q., Wu, X.M., Yan, F.W., 2016. Comparison of biodiesel production from sewage sludge obtained from the A²/O and MBR processes by in situ transesterification. *Waste Manage.* 49, 212–220.
- Rasool, K., Ahn, D.H., Lee, D.S., 2014. Simultaneous organic carbon and nitrogen removal in an anoxic-oxic activated sludge system under various operating conditions. *Biores. Technol.* 162, 373–378.
- Spellman, F.R., 2003. *Handbook of Water and Wastewater Treatment Plant Operations*. Lewis Publishers, USA.
- Su, X., Tian, Y., Sun, Z., Lu, Y., Li, Z., 2013. Performance of a combined system of microbial fuel cell and membrane bioreactor: wastewater treatment, sludge reduction, energy recovery and membrane fouling. *Biosens. Bioelectron.* 49, 92–98.
- Sun, R., Xing, D., Jia, J., Ren, N., 2014. Optimization of high-solid waste activated sludge concentration for hydrogen production in microbial electrolysis cells and microbial community diversity analysis. *Int. J. Hydrogen Energy* 39, 19912–19920.
- Van Aken, P., Van den Broeck, R., Degrève, J., Dewil, R., 2015. The effect of ozonation on the toxicity and biodegradability of 2,4-dichlorophenol-containing wastewater. *Chem. Eng. J.* 280, 728–736.
- Wang, Q.B., Chen, Q.W., 2016. Simultaneous denitrification and denitrifying phosphorus removal in a full-scale anoxic-oxic process without internal recycle treating low strength wastewater. *J. Environ. Sci. China* 39, 175–183.
- Xiao, B.Y., Han, Y.P., Liu, X., Liu, J.X., 2014. Relationship of methane and electricity production in two-chamber microbial fuel cell using sewage sludge as substrate. *Int. J. Hydrogen Energy* 39, 16419–16425.
- Xie, B.Z., Dong, W.B., Liu, B.J., Liu, H., 2014. Enhancement of pollutants removal from real sewage by embedding microbial fuel cell in anaerobic-anoxic-oxic wastewater treatment process. *J. Chem. Technol. Biotechnol.* 89, 448–454.
- Xie, B.Z., Liu, B.J., Yue, Y., Yang, L.G., Liang, D.W., Zhu, Y., Liu, H., 2016. Microbiological mechanism of the improved nitrogen and phosphorus removal by embedding microbial fuel cell in anaerobic-anoxic-oxic wastewater treatment process. *Biores. Technol.* 207, 109–117.
- You, S.-J., Zhang, J.-N., Yuan, Y.-X., Ren, N.-Q., Wang, X.H., 2010. Development of microbial fuel cell with anoxic/oxic design for treatment of saline seafood wastewater and biological electricity generation. *J. Chem. Technol. Biotechnol.* 85, 1077–1083.
- Zhang, F., Ge, Z., Grimaud, J., Hurst, J., He, Z., 2013. In situ investigation of tubular microbial fuel cells deployed in an aeration tank at a municipal wastewater treatment plant. *Biores. Technol.* 136, 316–321.
- Zhang, G.D., Lee, D.-J., Cheng, F.Q., 2016. Treatment of domestic sewage with anoxic/oxic membrane-less microbial fuel cell with intermittent aeration. *Biores. Technol.* 218, 680–686.
- Zhang, J.N., Zhao, Q.L., You, S.J., Jiang, J.Q., Ren, N.Q., 2008. Continuous electricity production from leachate in a novel upflow air-cathode membrane-free microbial fuel cell. *Water Sci. Technol.* 2008, 1017–1021.
- Zhao, Y., Zhang, Y., Ge, Z., Hu, C., Zhang, H., 2014. Effects of influent C/N ratios on wastewater nutrient removal and simultaneous greenhouse gas emission from the combinations of vertical subsurface flow constructed wetlands and earthworm eco-filters for treating synthetic wastewater. *Environ. Sci. Process. Impacts* 16, 567–575.
- Zhuang, L., Yuan, Y., Wang, Y., Zhou, S., 2012. Long-term evaluation of a 10-liter serpentine-type microbial fuel cell stack treating brewery wastewater. *Biores. Technol.* 123, 406–412.