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Simultaneous Pollutant Removal and Electricity Generation in a Combined ABR-MFC-MEC System Treating Fecal Wastewater

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Abstract Simultaneous power generation and fecal wastewater treatment were investigated using a combined ABR-MFC-MEC system (anaerobic baffled reactor-microbial fuel cell-microbial electrolysis cell). The installation of multi-stage baffles can benefit retaining the suspended solids in the system and help separate the hydrolysis-acidification and the methanogen processes. The efficiencies of the nitrification-denitrification process were improved because of the weak current generation by coupling the microbial electrochemical device (MFC-MEC) with the ABR unit. Maximum removal rates for chemical oxygen demand (COD) and ammonia nitrogen (NH_4^+-N) were 1.35 ± 0.05 kg COD/m^3/day and 85.0 ± 0.4 g NH_4^+-N/m^3/day, respectively, while 45% of methane (CH_4), 9% of carbon dioxide (CO_2), and 45% of nitrogen gas (N_2) contents in volume ratio were found in the collected gas phase. An average surplus output voltage of 452.5 ± 10.5 mV could be achieved from the combined system, when the initial COD concentration was 1500.0 ± 20.0 mg/L and the initial NH_4^+-N concentration was 110.0 ± 5.0 mg/L, while the effluent COD could reach 50.0 mg/L with an HRT of 48 h. The combined process has the potential to treat fecal wastewater efficiently with nearly zero energy input and a fair bio-fuel production.

Keywords Fecal wastewater · Anaerobic baffled reactor (ABR) · Microbial fuel cell (MFC) · Microbial electrolysis cell (MEC) · Electrodes

1 Introduction

Worldwide, there are over 2.5 billion people without access to safe sanitation, where black water is the major pollution (Glorina P. Orozco 2012). Fecal contamination is a widespread and serious issue influencing urbanized areas, rural zones, and wild regions (Biache et al. 2015). The commonly used on-site treatment for human waste is the water-flushing toilet with a septic tank and a drainage system (Quitzau 2007; Xuan et al. 2011). Composting-based technologies have been developed to reduce pollution from toilets. Closed systems are used for local reuse of black water, including composting toilet technology, urine diversion dehydration toilets, separation system with vacuum toilets, source separating vermin composting toilet, and the urine diverting vermin composting toilet (Anand and Apul 2014; Hill and Baldwin 2012; Lalander et al. 2013; Oarga Mulec et al. 2016; Skjelhaugen 1999; Uddin et al. 2014). However, the digestive efficiency is relatively low in most processes, which requires large hydraulic retention

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time (HRT) demand and low volumetric loading. From the perspective of water conservation and safety, new technologies for the treatment of fecal wastewater are urgently needed (Yatmo and Atmodiwirjo 2012).

The microbial fuel cell (MFC) technology can be utilized to recover ammonium from urine and produce electricity from feces simultaneously (Kuntke et al. 2012). In a MFC system, the substrate is oxidized by the microorganisms providing electrons at the anode, which flow to the cathode where they are combined with oxygen and protons to form water (Wang et al. 2012). In a microbial electrolysis cell (MEC) system, the local micro-aerobic condition can be generated by oxygen evolution in the region of the anode (Wu et al. 2013). Hydrogen production in the cathode is able to promote activities of methanogens and increase methane yield (Cheng and Logan 2011). Nitrogen removal can be significantly enhanced when the MEC unit is assisted by MFC, while higher short-circuit current of MFC is essential for developing the coupled system (Shaoan Cheng and Logan 2011; Yang Li et al. 2012). MFC can be connected in series to supply power and provide the reaction power for MEC using an anaerobic baffled reactor (ABR) process (Pirsaheb et al. 2015), while MEC can transform various types of nitrogen sources into nitrogen gas (Cheng and Logan 2011). Ammonia, for example, is converted into nitrite by microbes at the anode while nitrogen is evolved at the cathode (Jia et al. 2010; Li et al. 2010; Liu and Li 2007; Mohan et al. 2008; Sudarsan et al. 2014; Wu et al. 2013). The reactions occurring at the MFC and MEC electrodes can be demonstrated with the following equations:

$$\text{Anode MFC}: CH_3COO^- + 4H_2O \rightarrow 2HCO_3^- + 9H^+ + 8e^- \quad (1)$$

$$\text{Cathode MFC}: 2NO_3^- + 10e^- + 12H^+ \rightarrow N_2 + 6H_2O \quad (2)$$

$$\text{Anode MEC}: 2NH_4^+ + 2H_2O \rightarrow NO_2^- + 6e^- + 8H^+ \quad (3)$$

$$\text{Cathode MEC}: 2NO_2^- + 6e^- + 8H^+ \rightarrow N_2 + 4H_2O \quad (4)$$

The hydrolysis of the solid organic matter is promoted and the amount of sludge is minimized while the activity of the anaerobic methanogen, nitrification, and de-nitrification reaction of ammonia are enhanced by the coupling of microorganism and electrochemical effect in a MFC-MEC system (Chang et al. 2005). The production of hydrogen is also possible in a coupled MFC-MEC process (Kiely et al. 2011; Min et al. 2008). Multiple benefits can be achieved by introduction of microbial electrochemical device (MFC-MEC) into ABR: (1) the hydrolysis of solid organic matters is enhanced under the stimulation of electrochemical reactions; (2) the activities of methanogenic bacteria are improved by coupling microbial electrochemical reactions, and the generation of hydrogen sulfide odor is suppressed; (3) the combination of MFC and MEC can also promote the nitrification-denitrification process so as to improve the water quality of effluent; (4) power output from MFC is sufficient to support the operation of MEC, so that the internal energy supply-demand balance can be realized.

This study presents a novel combined "ABR-MFC-MEC" process treating fecal wastewater, where a coupled MFC-MEC unit is introduced into the ABR process and operated in an energy self-sufficient mode. The MFC is configured based on the redox of influent fecal wastewater in the ABR, while the surplus electricity energy is contributing to the formation of MEC and stimulating the microorganism activity in the ABR unit. The MFC and MEC were started separately during ramp up of the system and then the combined ABR-MFC-MEC system was investigated and optimized.

2 Materials and Methods

2.1 Set-Up of Experiments

The experimental set-up for the single chamber MEC is shown in Fig. 1, where the dual chamber set-up can be found in the electronic supplement as Fig. S1. The reactors are made of plexiglas (5 mm thick). The configuration is composed of three parts namely an ABR_{1-4} unit, a MFC (cathode-anode) unit, and a MEC (cathode-anode) unit. The ABR unit is composed of an upper-

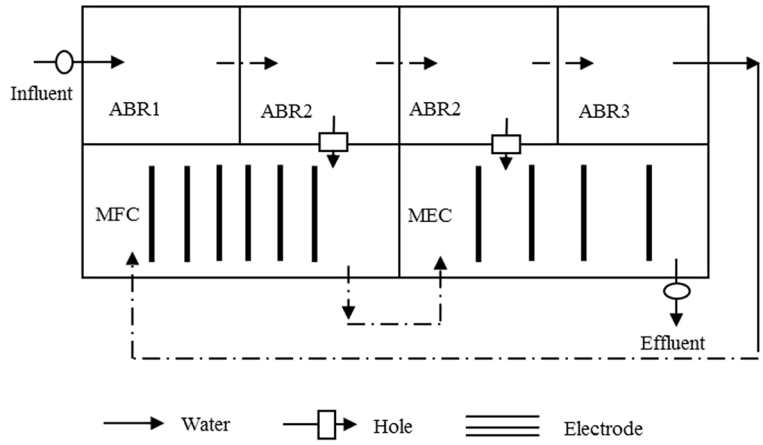

Fig. 1 Schematic diagram of the experimental set-up (single-chamber MEC)

flow-module with an angle of 45° to the baffled plate and a lower-flow chamber consisting of two or four compartments, respectively. The size of the main compartment is 640 mm × 180 mm × 250 mm with an effective volume of 28 L, and the effective volume of MFC chamber equals to the MEC chamber, i.e., 9.6 L.

A schematic diagram of the experimental set-up for the double-chamber configuration is shown in supplementary materials Fig. S1. For the single chamber system, the second and the third compartment of ABR connected with MFC and MEC, respectively, using a membrane-less strategy, while the layout of MFC and MEC is in parallel. For the double-chamber system, the anode compartment of MFC and the cathode compartment of MEC were connected in series using the proton exchange membrane Nafion117.

Graphite carbon felt was chosen as anode specifically due to its large specific surface area and conductivity, while the stainless steel wire mesh metal was used as cathode, which can accelerate the electron transfer. The MFC unit is made up of three pieces of carbon felts (8 cm × 10 cm) and three pieces of steel meshes (8 cm × 10 cm) (abbreviated as MFCc cathode and MFCa anode, respectively). Titanium wires of 0.2 mm are used as collecting electrodes in the external circuit, while the additional external resistance is fixed to 1200 Ω.

The MEC unit is formed by the same electrodes (abbreviated as MECc and MECa, respectively), which are both made of carbon felt (8 cm × 10 cm). Between the electrodes, an external voltage of 0.10–0.40 V is fed by a DC electronic power supply (IT8800 series) during the ramp up stage and then by the MFC after the start-up is finished. The electrodes are connected into the circuit with titanium wires and 1 Ω resistance, while gases are collected by the air trap above the reactors. The thermal maintenance of the system is achieved in a 30 °C thermostatic chamber with a batch-fed mode (Hengjing Yan 2013). Biogas production, HRT, and aeration rate were studied to investigate simultaneous pollutant removal and electricity generation. A diode is connected between two electrodes to control the direction of current from power supplier MFC to electricity user MEC.

2.2 Wastewater

Both real fecal wastewater and synthetic fecal wastewater are used in the study; characteristics of the synthetic and the real fecal wastewater are listed in Table 1. The real fecal wastewater is taken from a small storage tank for dormitory toilet waste from University of Shanghai for Science and Technology. The synthetic fecal water consists of hydrous glucose, sodium acetate, monopotassium phosphate, ammonium chloride, and other

Table 1 Characteristics of the synthetic and real fecal wastewater

	pH	COD (mg/L)	NH_4^+-N (mg/L)	TN (mg/L)	SS (mg/L)	TP (mg/L)	NO_3-N (mg/L)
Real fecal wastewater	7.3–8.6	255–1570	18–130	49.2–190.7	46–161	67–95	4–20
Synthetic fecal wastewater	7.4	1500	120	150	150	90	15

nutrient solution. These chemicals provide the source of carbon, nitrogen, and phosphorus, respectively. Trace element solution of 1 mL/L with the following components is also used for the synthetic fecal wastewater: 0.900 mg/L $FeCl_3$, 0.120 mg/L $ZnSO_4 \cdot 7H_2O$, 0.060 mg/L $Na_2MoSO_4 \cdot 2H_2O$, 0.180 mg/L KI, 0.03 mg/L $CuSO_4 \cdot 5H_2O$, 0.150 mg/L $CaCl_2 \cdot 6H_2O$, and 0.150 mg/L H_3BO_3. The initial pH of the influent synthetic fecal wastewater is adjusted to 7.2–7.5 with 1 M HCl and 1 M NaOH, and then, 0.1 M phosphate buffer is also necessary. The inoculated activated sludge is taken from the municipal sewage treatment plant Dongqu, Shanghai China, and is added into each cell compartment of the MFC-MEC-ABR system with a biomass/sludge of 4330.0 ± 410.0 mg COD/L (Sharma and Kundu 2010). The start-up is carried out with a step increasing volume loading scheme from 0.2 g COD/(L/d) to 1.5 g COD/(L/d). In the later period of each experiment, the real fecal wastewater was used instead of the synthetic fecal wastewater.

2.3 Analyses

COD, NH_4-N, and total phosphorus (TP) of wastewater were determined by standard methods (American Public Health Association (APHA) 1998). The produced voltage was measured automatically by a data acquisition system (PICOLOG-ADC-24) at an interval of 30 s. Total organic carbon (TOC) was tested by a Multi N/C 3100 analyzer, and detection of biogas concentration was analyzed by a GC-TCD.

3 Results and Discussion

3.1 Removal of Pollutants and Production of Biogas

Effluent COD and NH_4-N are important indicators to evaluate the performance of the ABR-MFC-MEC system treating fecal wastewater. First, a continuous synthetic influent with a constant COD concentration of 1500 mg/L was employed in the study. As shown in Fig. 2, a micro-aeration rate of 50 mL/min was employed to form the air-cathode during the start-up stage (Cheng et al. 2011; Diak et al. 2013; Feng et al. 2010; Logan and Regan 2006). The effluent COD kept relatively stable at the end of the start-up stage within a range of 50–150 mg/L while effluent NH_4-N could reach 10 mg/L. The COD removal rate was about 93

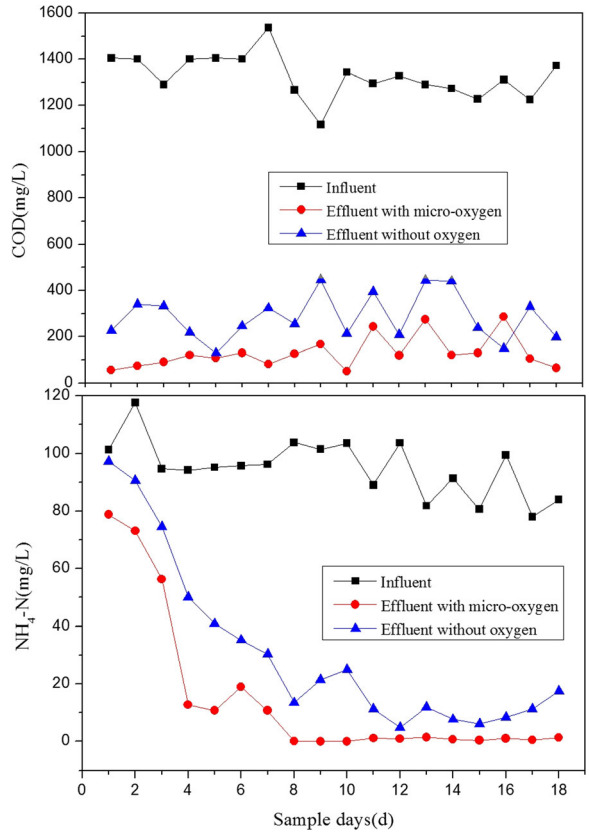

Fig. 2 Removal of COD and NH_4^+-N by the ABR-MFC-MEC system

%. When the system reached a steady state, the aeration intensity was slowly reduced to zero. Effluent COD and NH_4-N concentrations were about 150–300 and 25–30 mg/L, respectively, at the later stage of the operation when no aeration was implemented. Most COD and nitrogen were removed when the cathodic dissolved oxygen (DO) was at low levels (less than 0.2 mg/L), where oxygen was used as the electron acceptor to promote chemical reactions; MFC was used to remove organic matter and MEC to remove ammonia nitrogen (Tao et al. 2014).The electron donor and acceptor were different in the chambers. In a mixture containing, respectively, nitrate as catholyte and ammonium as anolyte, ammonia nitrogen could be used as electron donor and reactants between the cathode and the anode to provide energy for microorganisms in the process of bio-electro-chemical anaerobic ammonia oxidation, while the high potential of anode in MEC prompts nitrification rate, even with limited dissolved oxygen.

Components of the collected biogas were also analyzed with results shown in Fig. 3. The major content of the bio-

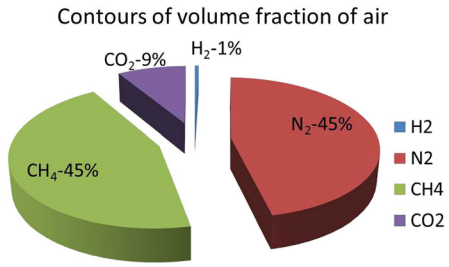

Fig. 3 Components of the collected biogas

gas is N_2 and CH_4, both accounted for nearly 45% of the total gas volume. The gas composition is slightly lower with respect to the CH_4 content but much higher for N_2 in comparison to a general anaerobic reactor (CH_4 55–65%, CO_2 30–45%), mainly due to the micro-aeration which brings N_2 into the system. Another reason is that nitrogen compounds are reduced to nitrogen with the de-nitrification occurrence in the system. The major adverse effect of pH is its inhibition on methanogenic activity of the ABR unit (Kim et al. 2004).

3.2 Electricity Production

The stimulating effect of the microbes in a coupled MFC-MEC process played an important and positive role in the quick start-up of the system. A low voltage from MFC is applied to the end chambers to build MEC, which has the ability for the hydrolysis of the solid organic matter and the nitrogen removal on both the local anodes and cathodes of MEC.

Power generation of both the single-chamber and the double-chamber configurations of the combined ABR-MFC-MEC system and/or the single MFC system are compared in Fig. 4a. Power generated reached 350.0 ± 100.0 and 450.0 ± 5.6 mV in the double-chamber and the single-chamber systems, respectively. The MFC was located at the fourth chamber of the treatment process for the double-chamber system, where less organic matter was left in the wastewater. Fewer microorganisms were attached to electrodes, and the capacity of output voltage would lessen and cause instability in the process. A part of the generated power was also consumed in the MEC chamber (Rahimnejad et al. 2011). When the feed water was changed, the environment for electrogenesis microorganism was influenced and the community composition would change. Along with the adaptability of electricigens, the production capacity was improved.

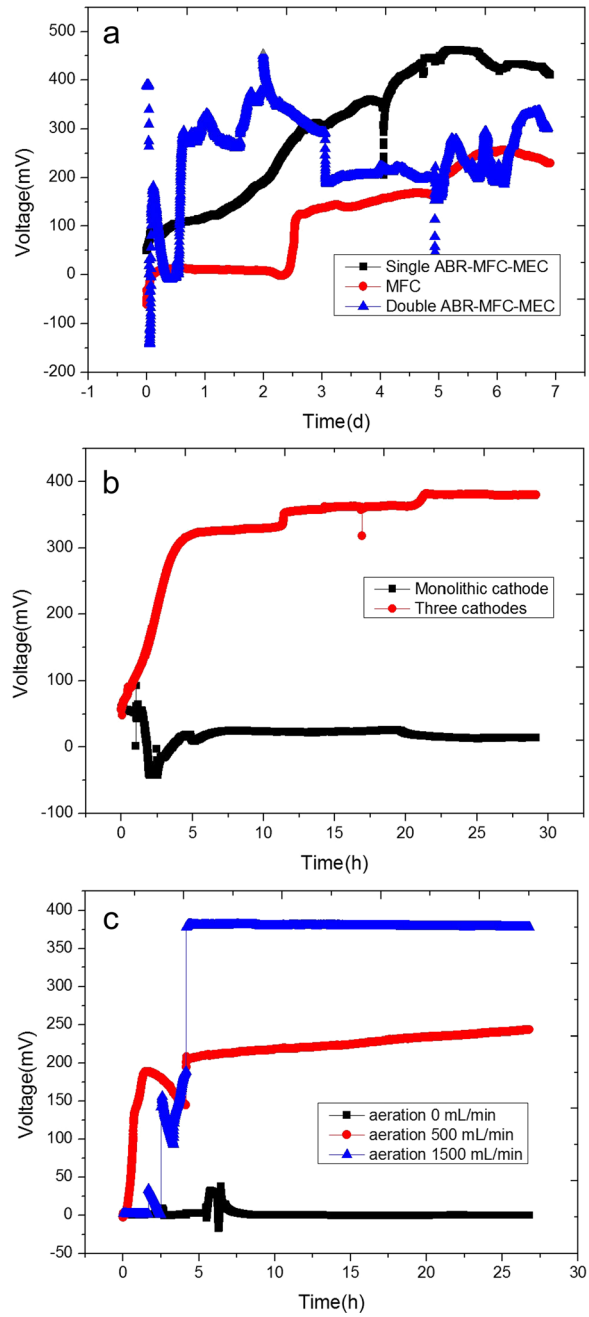

Fig. 4 Electricity production of the studied process. **a** Electricity production comparison of the ABR-MFC-MEC process and the MFC process. **b** Electricity productions with different sizes of anode. **c** Electricity production under different aeration rates

Electrode density (surface area/sewage volume) was 28.6 cm^2/L for the anode, and increase of the electrode density at the cathode would improve the output voltage. When the electrode density of three electrodes

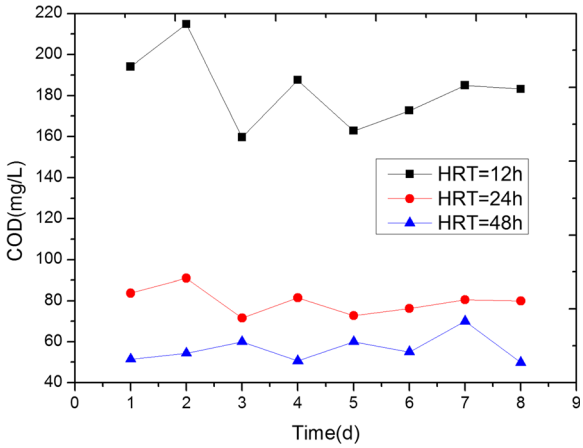

Fig. 5 COD removal under different HRTs

increased to 85.8 cm^2/L, the voltage maintained at 410 mV (Fig. 4b).

Size and surface of electrodes played an important role in electricity generation, and thus, the performance of the MFC with different numbers of electrodes was investigated (Sakdaronnarong et al. 2013). The voltage generation was associated with the MFC biomass and the electrode. The influence of the electrode surface on the voltage generation in the study system is presented in Fig. 4b.When the influent COD concentration was kept constant as 1200 mg/L, potential of MFC anode decreased in the continuous experiments, which resulted from the instantaneous potential difference at the beginning of the operation. Electrons released from the microbial metabolism were attached to the anode first and then used the anode as an electron acceptor. The number and size of electrodes determined the content of microbial, so that it could decide the output voltage indirectly. The voltage rose up rapidly and kept at around 400 mV when the electrodes were increased to three units of its numbers. MFCs had the tendency to be stable after 200 min in either one or three electrodes. Finally, with the depletion of the substrate and accumulation of metabolites, the bacterial activity was reduced and the corresponding output voltage decreased. Timely replacement of the substrate is necessary to maintain the reaction.

3.3 Influence of HRT and Aeration Rate

The influence of HRT on the COD removal for real fecal wastewater is illustrated by Fig. 5. The performance of COD removal was relatively sensitive to changes of

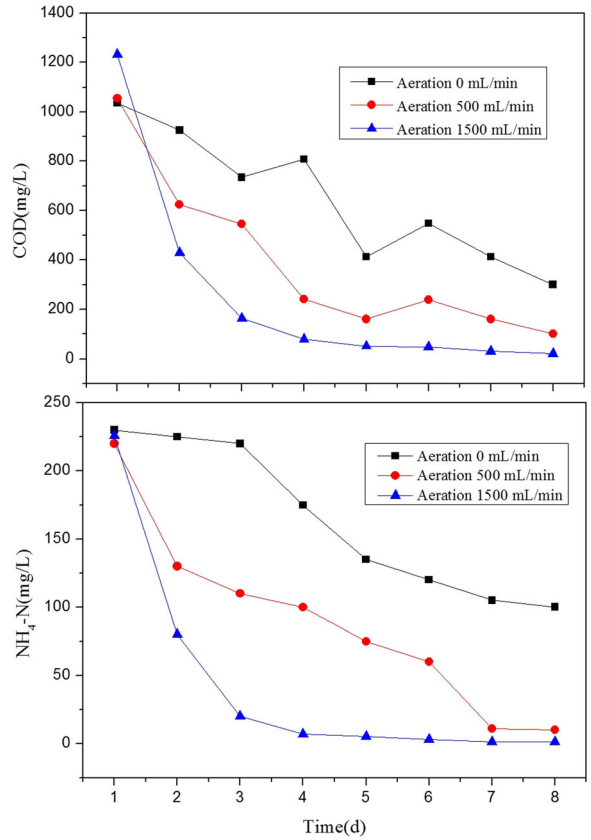

Fig. 6 COD and $NH4^+$-N removal under different aeration rates

HRT. Effluent COD remained at 150–200 mg/L when the HRT was 12 h; however, the effluent COD was improved to 80–100 mg/L when the HRT increased to 24 h, and further improved to 50–60 mg/L when the HRT was 48 h. The effluent COD of the combined system was much better than of each single unit under the same HRTs (Escapa et al. 2012).

The influence of aeration rate on electricity production of the combined ABR-MFC-MEC is illustrated in Fig. 4c. The provision of cathode electron acceptors and the oxidation reduction potential was 1.229 V, which made the cathode reduction reaction easier. The more air bubbled in, the quicker the reaction was, which boosted the output voltage (Sakdaronnarong et al. 2013). Results indicate that the aeration rate has a significant influence on power generation. The voltage was relatively small without aeration but increased to 200 mV when the aeration rate was 500 mL/min. The voltage was increased to 400 mV, when the aeration rate was 1500 mL/min.

The COD concentration decreased with the input of external electron acceptor. Figure 6 indicates that the COD removal rate was 60.1% without oxygen input and increased to 77.2% with an aeration rate of 500 mL/min, which was further increased to 95.9% when the aeration rate reached 1500 mL/min. The NH_4^+-N concentration decreased dramatically in the cathode chamber when the aeration was limited in the cathode and reduced from 219.1 to 10.9 mg/L with the increase of aeration.

4 Conclusions

A combined ABR-MFC-MEC system was applied to treat fecal wastewater. The effluent COD remains as low as 50 mg/L using the same HRT as in conventional septic tanks, even when the influent COD was higher than 1500 mg/L and ammonium was 110 mg/L. The NH_4^+-N concentration decreased dramatically in the cathode chamber when the aeration was limited in the cathode and reduced from 219.1 to 10.9 mg/L with the increase of aeration. The de-carbonizing and de-nitrification occurrence are responsible for the slightly lower CH_4 content and much higher N_2 content in the connected biogas. Furthermore, a surplus average output voltage of 452.5 ± 10.5 mV could be harvested from the combined system.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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