

# Nitrogen recovery from wastewater using microbial fuel cells

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**Abstract** Nitrogen is one of major contaminants in wastewater; however, nitrogen, as bio-elements for crop growth, is the indispensable fertilizer in agriculture. In this study, two-chamber microbial fuel cells (MFCs) were first operated with microorganisms in anode chamber and potassium ferricyanide as catholyte. After being successfully startup, the two-chamber MFCs were re-constructed to three-chamber MFCs which were used to recover the  $\text{NO}_3^- - \text{N}$  and  $\text{NH}_4^+ - \text{N}$  of synthetic wastewater into value-added nitrogenous fertilizer from cathode chamber and anode chamber, respectively. Ferric nitrate was used as the sole electron acceptor in cathode, which also was used to evaluate the  $\text{NO}_3^- - \text{N}$  recover efficiency in the case major anion of  $\text{NO}_3^-$  in cathode. The output voltage of these MFCs was about 600–700 mV at an external load of 500  $\Omega$ . About 47%  $\text{NH}_4^+ - \text{N}$  in anode chamber and 83%  $\text{NO}_3^- - \text{N}$  in cathode chamber could be recovered. Higher current density can selectively improve the recovery efficiency of both  $\text{NH}_4^+ - \text{N}$  and  $\text{NO}_3^- - \text{N}$ . The study demonstrated a nitrogen recovery process from synthetic wastewater using three-chamber MFCs.

**Keywords** nitrogen recovery, microbial fuel cells (MFCs), electromigration, wastewater treatment

## 1 Introduction

Nitrogen element plays a crucial role for life because it is a component of amino acids and nucleic acids. Although Earth's atmosphere is an abundant source of nitrogen gas, most plants cannot directly use it for growth. Therefore, nitrogen needs to be artificially converted to nitrogen compounds, usually ammonia, to be used by crops. In 2011, the nitrogenous fertilizer (nitrogen) consumption of the world is about 107.5 million tons [1]. The production

of nitrogenous fertilizer also will consume lots of energy and yield many pollutants.

On the other hand, nitrogen in domestic and industrial wastewater is usually considered as contaminant with the main form of ammonium ( $\text{NH}_4^+$ ) and nitrate ( $\text{NO}_3^-$ ) [2], due to the severe impacts on regulating the eutrophic level of water bodies and on human and animal health [3]. For example, the concentration of total nitrogen (mainly  $\text{NH}_4^+$ ) in swine wastewater is higher than 6000  $\text{mg} \cdot \text{L}^{-1}$  [4]. Therefore, the treatment of nitrogen contaminated wastewater is attracting increasing attention, and many modifications and novel processes, such as anaerobic ammonium oxidation [5] and single reactor system for high activity ammonium removal over nitrite [6], have been developed and implemented for nitrogen removal from wastewater.

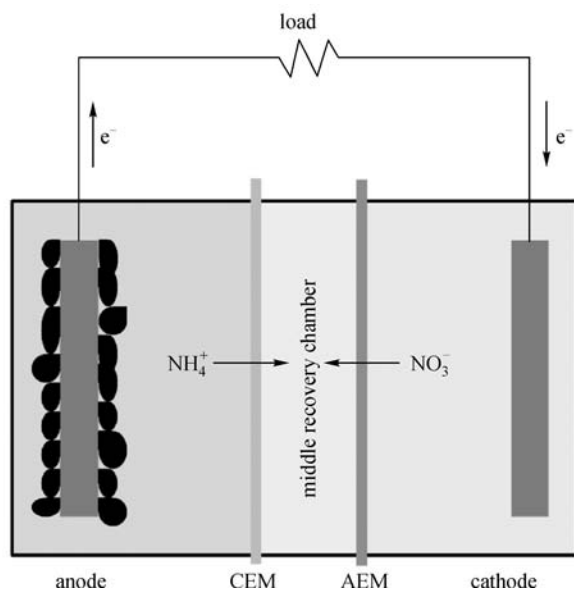
Microbial fuel cells (MFCs) can simultaneously treat domestic or industrial wastewater and produce electricity [7–17]. An MFC usually consists of an anode chamber and a cathode chamber, separated using a cation exchange membrane (CEM) or an anion exchange membrane (AEM). Electrons are produced at the anode and transferred through an external circuit to the cathode, while a charge balancing number of cations and/or anions are transferred between anode and cathode via the CEM or the AEM. The movement of ions across membranes during current generation provides a method for removing ions from saline waste [11] or recovering useful substances from wastewater [18,19]. Though some studies have employed membrane technology to recover nitrogen from wastewater, external energy usually should be supplied [18,19]. Up to now, few studies have employed MFCs technology to recover nitrogen from wastewater as fertilizer without energy input.

In this study, we propose MFCs technology for simultaneously removing and recovering nitrogen contaminants from synthetic wastewater. The results demonstrated that it exerts potential to simultaneously remove and recover most of ammonia and nitrate from wastewater without external energy supply.

## 2 Materials and methods

### 2.1 MFC construction

Triplicated MFCs, i.e. MFC1, MFC2 and MFC3, were used in this study. The anode was inoculated with 5 mL of activated sludge. During the startup process, two-chamber MFCs were constructed and operated under  $-0.05$  MPa for 3 cycles as our previous report described [20], in the purpose that speeding up the startup of anodic electrogenic microbes. After 3 stable operated cycles of the two-chamber MFCs, three-chamber MFCs, consisting of three chambers (anode, middle recovery and cathode chamber, respectively) separated by CEM and AEM membranes (QQ-YLM, Qianqiu Group, China) (Fig. 1), were constructed. Two pieces of carbon felt ( $3\text{ cm} \times 3\text{ cm}$  each) were used as electrode material in the anode and cathode chambers, and the external load was a  $500\ \Omega$  resistor. The electrodes were connected by titanium wire. The net volume of anode, cathode and middle recovery chambers were 100, 100 and 15 mL, respectively. The distance between the anode and cathode was about 2 cm.



**Fig. 1** Schematic diagram of three-chamber MFC used for nitrogen recovery

### 2.2 MFCs operation

The two-chamber MFCs and the three-chamber MFCs were operated in batch mode. The two-chamber MFCs were operated following our previous study [20]. In each cycle, anode medium of 80 mL and catholyte of 100 mL were replaced by fresh solution. The MFCs were treated with negative pressure for 2 cycles to enhance their output power [20].

After the anodes of two-chamber MFCs have been able to provide steady power output, the MFCs were reconstructed into three-chamber MFCs. To investigate the impact of reconstruction on MFC performance, the anode and cathode chambers were filled with the same solution as that in two-chamber MFCs, and the middle recovery chamber was filled with deionized water of 15 mL in the first two batch cycle.

For the three-chamber MFCs recovering nitrogen, the anode was first fed with 100 mL medium (pH 7.0) containing  $10\text{ mmol}\cdot\text{L}^{-1}$   $\text{CH}_3\text{COONH}_4$ ,  $7.2\text{ mmol}\cdot\text{L}^{-1}$   $\text{NaH}_2\text{PO}_4$  and  $12.8\text{ mmol}\cdot\text{L}^{-1}$   $\text{Na}_2\text{HPO}_4$ , the middle recovery chamber was filled with deionized water of 15 mL, and the cathode was filled with 100 mL solution containing  $20\text{ mmol}\cdot\text{L}^{-1}$   $\text{Fe}(\text{NO}_3)_3$  (pH 2–3). The  $\text{CH}_3\text{COONH}_4$  in anode chamber was used both as carbon and nitrogen sources, and  $\text{Na}^+$  was used to evaluate the effect of coexisting cation on  $\text{NH}_4^+$  recovery. The  $\text{Fe}(\text{NO}_3)_3$  in cathode chamber was used as electron acceptor (reducing  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$ ) and nitrogen source, and the major anion of  $\text{NO}_3^-$  in cathode was used to evaluate the nitrogen recover efficiency without other ion. In each cycle, outflow of 80 mL in anode was replaced with inflow of 80 mL containing  $12.5\text{ mmol}\cdot\text{L}^{-1}$   $\text{CH}_3\text{COONH}_4$ ,  $7.2\text{ mmol}\cdot\text{L}^{-1}$   $\text{NaH}_2\text{PO}_4$  and  $12.8\text{ mmol}\cdot\text{L}^{-1}$   $\text{Na}_2\text{HPO}_4$  (pH 7.0), and the 15 mL solution in middle recovery chamber and the 100 mL catholyte were replaced by deionized water of 15 mL and  $20\text{ mmol}\cdot\text{L}^{-1}$   $\text{Fe}(\text{NO}_3)_3$  of 100 mL, respectively. The three-chamber MFCs which was added the same solution as that in experiment group except being operated open circuit were served as control tests. All MFCs were operated at  $28^\circ\text{C}$ .

### 2.3 Analyses and calculations

Chemical oxygen demand (COD) measurement for anode medium was based on digestion with  $\text{K}_2\text{Cr}_2\text{O}_7$  in concentrated  $\text{H}_2\text{SO}_4$ .  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were determined according to the standard methods issued by the Ministry of Environmental Protection of China. Concentration of  $\text{Na}^+$  was determined with ion chromatograph (ICS3000, Dionex, USA). All analyses were performed in triplicate.

The voltage ( $E$ ) across the resistor ( $R_e$ ) was recorded by an Integra 2700 series equipped with 7700 multiplexer (Keithley Instruments, USA), and the current ( $I$ ) through the electrical circuit was calculated by  $I = E/R_e$ . The theoretical amount of electrons ( $Q_{th}$ ), expressed in coulombs (C), from the oxidation of acetate were calculated by  $Q_{th} = \Delta\text{COD}\cdot F/8(\text{g}\cdot\text{mol}^{-1})$ , where  $\Delta\text{COD}$  is the reduced COD (g) during and  $F$  is Faraday's constant ( $96485\text{ C}\cdot\text{mol}^{-1}$ ). Electrons harvested through the electrical circuit ( $Q$ ) was determined by integrating current over time ( $Q = \int I dt$ ). The coulombic efficiency ( $\eta_{ce}$ ) of anode was calculated by  $\eta_{ce} = Q/Q_{th}$ .

The mol ratio ( $R_{mol}$ ) of  $\text{NH}_4^+$  to cations electrochemi-

cally migrated from the anode chamber to the middle recovery chamber was calculated as  $R_{mol} = (M_{mir} - M_{dif}) / (M_{ele} - M_{nitrate})$ , where  $M_{mir}$  was the sum of  $\text{NH}_4^+$  in the cathode and the middle recovery chambers,  $M_{dif}$  was  $\text{NH}_4^+$  diffusing into the middle recovery chamber in control tests,  $M_{ele}$  was the mol of electron harvested through the electrical circuit, which was calculated from  $M_{ele} = Q/F$ , and  $M_{nitrate}$  was the  $\text{NO}_3^-$  transmitted to the anode chamber.

### 3 Results and discussion

#### 3.1 MFCs performance

Using potassium ferricyanide as electron acceptor, the two-chamber MFCs outputted a stable voltage of about 600 mV in three reproducible cycles (Appendix 1), suggesting that the anode performance have reached a steady-state for next operation. Two-chamber MFCs of 1, 2 and 3 had average coulombic efficiencies of 27.6%, 31.7% and 23.8% (Appendix 2), respectively, and their maximum output power were 1.5 to 2 mW.

Though an AEM was added into the MFCs and the distance between the anode and cathode increased from about 1.5 cm to about 2.0 cm, the reconstruction of three-chamber MFC seemed to have little negative impact on the electricity generation (Appendix 3). The potential of 60  $\text{mmol}\cdot\text{L}^{-1}$  ferric nitrate was about 600 mV, higher than about 230 mV of potassium ferricyanide. The open circuit voltage of the three-chamber MFCs was as high as 1200 mV (Appendix 4). However, the stable output voltage of the three-chamber MFCs across an external resistor of 500  $\Omega$  was at the range of 600–700 mV (Fig. 2), only a little higher than that of the two-chamber MFCs. The MFC3 had higher output voltage than that of MFC1 and MFC2. However, the three MFCs of 1–3 output similar

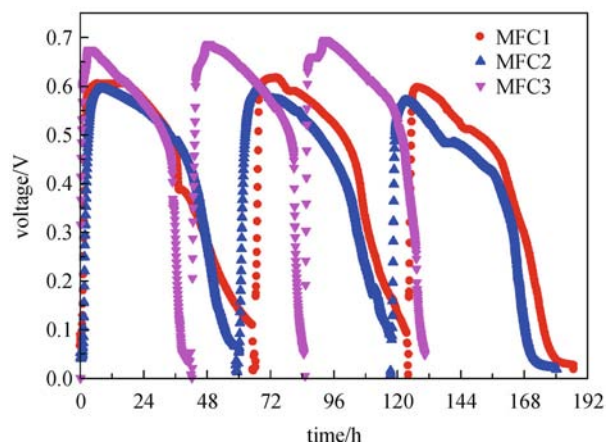


Fig. 2 Output voltage of the three-chamber MFCs for nitrogen recovery

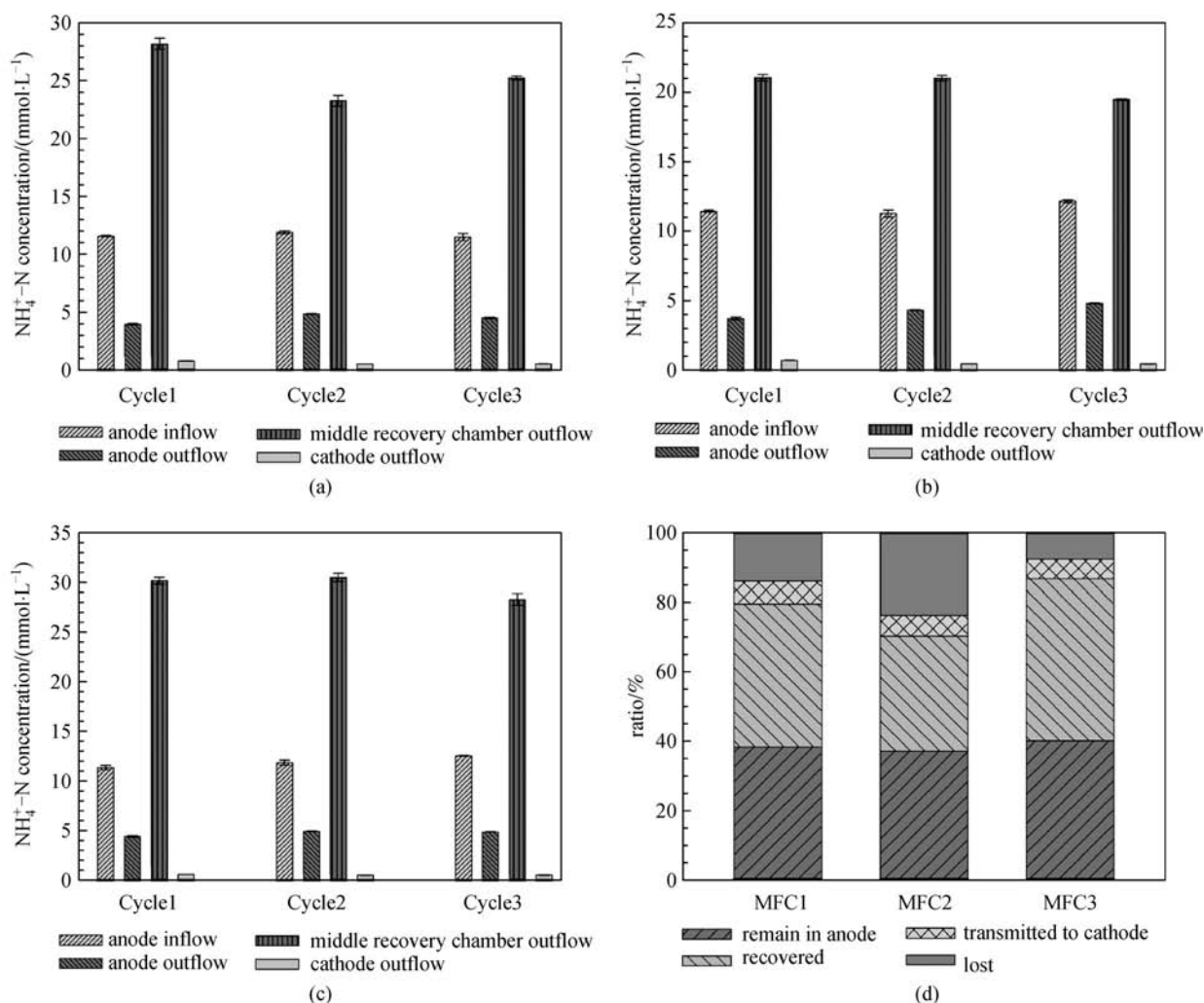
amount of electron and had similar coulombic efficiencies of about 20% in each single cycle (Appendix 5), which was 10% to 50% lower than that of the three-chamber MFCs.

#### 3.2 $\text{NH}_4^+$ -N recovery and mass balance

The  $\text{NH}_4^+$ -N concentration and its distribution in the three chambers of each MFC was shown in Fig. 3. No  $\text{NH}_4^+$ -N was detected in the inflow of cathode and middle recovery chambers. The detected  $\text{NH}_4^+$ -N concentration in the anode inflow (80 mL in each MFC) of the three MFCs ranged from 11.4 to 12.5  $\text{m mol}\cdot\text{L}^{-1}$ , which was well corresponded with the theoretical value of 12.5  $\text{m mol}\cdot\text{L}^{-1}$ . The  $\text{NH}_4^+$ -N concentration in anode outflow of the three MFCs was stabilized at 3.5–5.0  $\text{m mol}\cdot\text{L}^{-1}$ , which meant that about 60% of  $\text{NH}_4^+$ -N in the anode inflow was removed from anode chamber (Fig. 3(d)). As shown in Fig. 1, cation of  $\text{NH}_4^+$  can migrate to the middle recovery chamber via the CEM between the anode and middle recovery chambers and in theory will be retained in the middle recovery chamber by the AEM between the cathode and the middle recovery chambers. In a single cycle,  $30.5 \pm 0.4 \text{ m mol}\cdot\text{L}^{-1}$  of  $\text{NH}_4^+$ -N, 7 times higher than that in the anode outflow, was accumulated in the middle recovery chamber. Due to  $\text{NH}_4^+$  accumulation in the middle recovery chamber, about 41%, 33% and 47% of  $\text{NH}_4^+$ -N in the anode inflow or 67%, 53% and 78% of removed  $\text{NH}_4^+$ -N in the anode inflow were recovered in MFCs 1, 2 and 3, respectively. The results showed that more than half of the removed  $\text{NH}_4^+$ -N in the anode inflow could be successfully recovered and MFC3 seemed to possess the highest recovery efficiency among the three MFCs.

In theory, the AEM can stop the migration of cation between cathode and middle recovery chamber. But in the cathode outflow, we still detected 0.3–0.7  $\text{m mol}\cdot\text{L}^{-1}$  of  $\text{NH}_4^+$ -N (Fig. 3(d)). The cation of  $\text{NH}_4^+$  should be from the middle recovery chamber as our control test showed that no  $\text{NO}_3^-$  was electrochemically reduced to  $\text{NH}_4^+$  in cathode. Excluding the  $\text{NH}_4^+$ -N recovered in the middle recovery chamber and transmitted to the cathode chamber, about 14%, 24% and 8% of  $\text{NH}_4^+$ -N in the anode inflow was missing from MFCs 1, 2 and 3, respectively.  $\text{NH}_4^+$ -N is an easy-to-use nitrogen source for microorganism growth, and some studies also have reported that  $\text{NH}_4^+$  could be used by microbes for direct electricity generation in MFCs [21]. However, in the control tests, only 2% of  $\text{NH}_4^+$ -N in the anode inflow was recovered in the middle recovery chamber but as high as 12%–17% of  $\text{NH}_4^+$ -N in the anode inflow was lost in the MFCs (Appendix 6). The results indicated that electromigration was the major process for the  $\text{NH}_4^+$ -N recovery and the  $\text{NH}_4^+$ -N loss in the MFCs should be mainly caused by microbial growth.

In this study, the MFC technology was used to recover



**Fig. 3**  $\text{NH}_4^+-\text{N}$  concentration in the inflow and outflow of the anode chambers,  $\text{NH}_4^+-\text{N}$  concentration in the middle recovery chambers and  $\text{NH}_4^+-\text{N}$  concentration transported to the cathode chamber in MFC1 (a), MFC2 (b) and MFC3 (c), respectively. The error bar is for  $n = 3$  repeated measurements. d: Distribution of  $\text{NH}_4^+-\text{N}$  from the anode inflow within three cycles of each MFC

nitrogen contained ions. Electromigration and diffusion were the two processes for ions flux in MFCs. In the control tests i.e. at open circuit condition, only 3.5–4.5  $\text{mol}\cdot\text{L}^{-1}\text{NH}_4^+-\text{N}$  was detected within middle recovery chamber outflow (Appendix 7). The open circuit control tests showed that, employing diffusion as the main process, the  $\text{NH}_4^+-\text{N}$  recovery efficiencies for the three MFCs was only 5.5%–6.5%. Comparing with the  $\text{NH}_4^+-\text{N}$  recovery efficiencies of 41%, 33% and 47% in MFCs with 500  $\Omega$  external resistors, the results indicated that only a little  $\text{NH}_4^+-\text{N}$  recovery was caused by the diffusion process and electromigration must be the major process for  $\text{NH}_4^+-\text{N}$  recovery in the three-chamber MFCs.

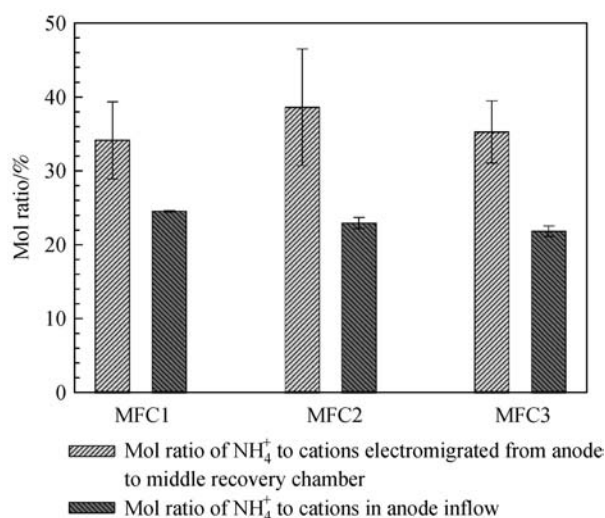
As for electromigration process, many factors, e.g. pH, current density, and ion type and concentration [22], may influence the nitrogen recovery efficiency. In the present study, pH was maintained at 6–7 in the anode chambers,

where the low concentration of proton in neutral pH solution should have not significantly influenced the migration of other cations (mainly  $\text{Na}^+$  and  $\text{NH}_4^+$ ). Therefore, the influences of current density and other ion type on nitrogen recovery efficiencies in MFCs were investigated by taking  $\text{NH}_4^+-\text{N}$  recovery as an example.

The existence of other cations e.g.  $\text{Na}^+$  might decrease the electromigration of  $\text{NH}_4^+$  from the anode chamber to the middle recovery chamber, which sequentially reduced the recovery efficiency of  $\text{NH}_4^+-\text{N}$ . Therefore, the impact of other cations on  $\text{NH}_4^+-\text{N}$  recovery efficiency was investigated by adding  $\text{Na}^+$  (a common ion in wastewater) to the anode chamber. In the three MFCs, 21%–25% of cations in the anode inflow were detected as  $\text{NH}_4^+$  (Fig. 4). However, the actual  $R_{mol}$  of  $\text{NH}_4^+$  in MFCs 1, 2 and 3 were  $34.2\%\pm 5.2\%$ ,  $38.7\%\pm 7.9\%$  and  $35.3\%\pm 4.2\%$ , respectively. The result demonstrated that the CEM might



selectively transfer  $\text{NH}_4^+$  from the anode chamber to the middle recovery chamber, which was in accord with a previous report [22]. The  $R_{mol}$  was similar to each other in MFCs 1, 2 and 3, although the current density in MFC3 was about 16% higher than that in MFC1 and MFC2. This result seemed to indicate that current density did not have any influence on the membrane selectivity to  $\text{NH}_4^+$ . However, the current density still showed a positive influence on the recovery efficiency of  $\text{NH}_4^+$ -N because MFC3 had the highest  $\text{NH}_4^+$ -N recovery efficiency of 47% within the three MFCs.



**Fig. 4** Mol ratio of  $\text{NH}_4^+$  to cations in the anode inflow and cations electromigrated from the anode chamber to the middle recovery chamber. The error bar is for  $n = 3$  repeated cycles

### 3.3 $\text{NO}_3^-$ -N recovery and mass balance

The  $\text{NO}_3^-$ -N concentration in the three-chamber MFCs was shown in Fig. 5, and no  $\text{NO}_3^-$ -N was detected in the inflow of the anode and the middle recovery chambers. The  $\text{NO}_3^-$ -N concentration in the inflow of cathode chamber ranged from 58.5 to 60.5  $\text{mmol}\cdot\text{L}^{-1}$ , which was well corresponded with the theoretical concentration of 60  $\text{mmol}\cdot\text{L}^{-1}$ . After each single cycle operation, the  $\text{NO}_3^-$ -N concentration in the outflow of the anode and the middle recovery chambers was 38–44  $\text{mmol}\cdot\text{L}^{-1}$  and 75–110  $\text{mmol}\cdot\text{L}^{-1}$ , respectively, and a high concentration of  $\text{NO}_3^-$ -N obviously was accumulated in the middle recovery chambers.

In theory, once electrons were transferred from the anode to the cathode through the external load, there must be cations and anions with equal charge passing through the CEM between the anode and the middle recovery chambers and the AEM between the cathode and the middle recovery chambers, respectively. In the cathode chamber, the pH was lower than 3 and most of the anions

were  $\text{NO}_3^-$ . Therefore,  $\text{NO}_3^-$  could be thought as the sole anion passing through the AEM, and the theoretical concentration of  $\text{NO}_3^-$ -N in the outflow of the middle recovery chambers could be calculated according to the collected electrons shown in Appendix 5. However, the actual concentration of  $\text{NO}_3^-$ -N in outflow of the middle recovery chambers was lower than the theoretical values (Figs. 5(a)–5(c)). The discrepancy could be attributable to two reasons: 1) there were some cations e.g.,  $\text{Na}^+$  and  $\text{NH}_4^+$  which were able to pass through the AEM and transported from the middle recovery chamber to the cathode chamber, and the transport of cations reduced the transportation efficiency of anion from the cathode chamber to the middle recovery chamber; 2) some  $\text{NO}_3^-$  should be transported from the middle recovery chamber to the anode chamber through the CEM since 5%–15% of  $\text{NO}_3^-$ -N in the cathode inflow was detected as loss in closed circuit tests (Fig. 5(d)) but as low as 1% of  $\text{NO}_3^-$ -N in the cathode inflow was lost in control tests (Appendix 8). Since  $\text{NO}_3^-$  could be easily reduced to dinitrogen gas at the presence of organic substance, we did not detect any  $\text{NO}_3^-$ -N in the anode outflow.

65.5%–67.5% of  $\text{NO}_3^-$ -N in the cathode inflow remained in outflow, and about 23%, 20% and 27% of the total  $\text{NO}_3^-$ -N in MFC1-3 was recovered in the middle recovery chamber, respectively (Fig. 5(d)). The remaining  $\text{NO}_3^-$ -N was lost. Consequently, the average  $\text{NO}_3^-$ -N recovery efficiencies for MFCs 1, 2 and 3 were 71%, 57% and 83%, respectively. Since there was no organism in the cathode chamber, the lost  $\text{NO}_3^-$ -N might be mainly transported to the anode chamber and was sequentially reduced to nitrogen gas as no  $\text{NO}_3^-$ -N was detected in the anode chamber. Denitrification process usually consumes organic substances (theoretically reduce 1.0 g of  $\text{NO}_3^-$ -N to nitrogen gas consumes about 1.4 g of COD), which might cause the decrease of coulombic efficiency in the three-chamber MFCs. Due to the highest transportation efficiency (about 15%) of  $\text{NO}_3^-$  from the middle recovery chamber to the anode chamber in MFC2, this MFC showed a 50% decrease in coulombic efficiency following the reconstruction from two-chamber MFC to three-chamber MFC (Appendix 2 and Appendix 5).

In the control tests,  $\text{NO}_3^-$ -N in the outflow of the middle recovery chambers was about 7  $\text{mmol}\cdot\text{L}^{-1}$  (Appendix 9), which was only 6%–9% of the recovered  $\text{NO}_3^-$ -N in the middle recovery chambers with a 500  $\Omega$  resistor. The results demonstrated that electromigration must be the major process for  $\text{NO}_3^-$ -N recovery in the middle recovery chamber.

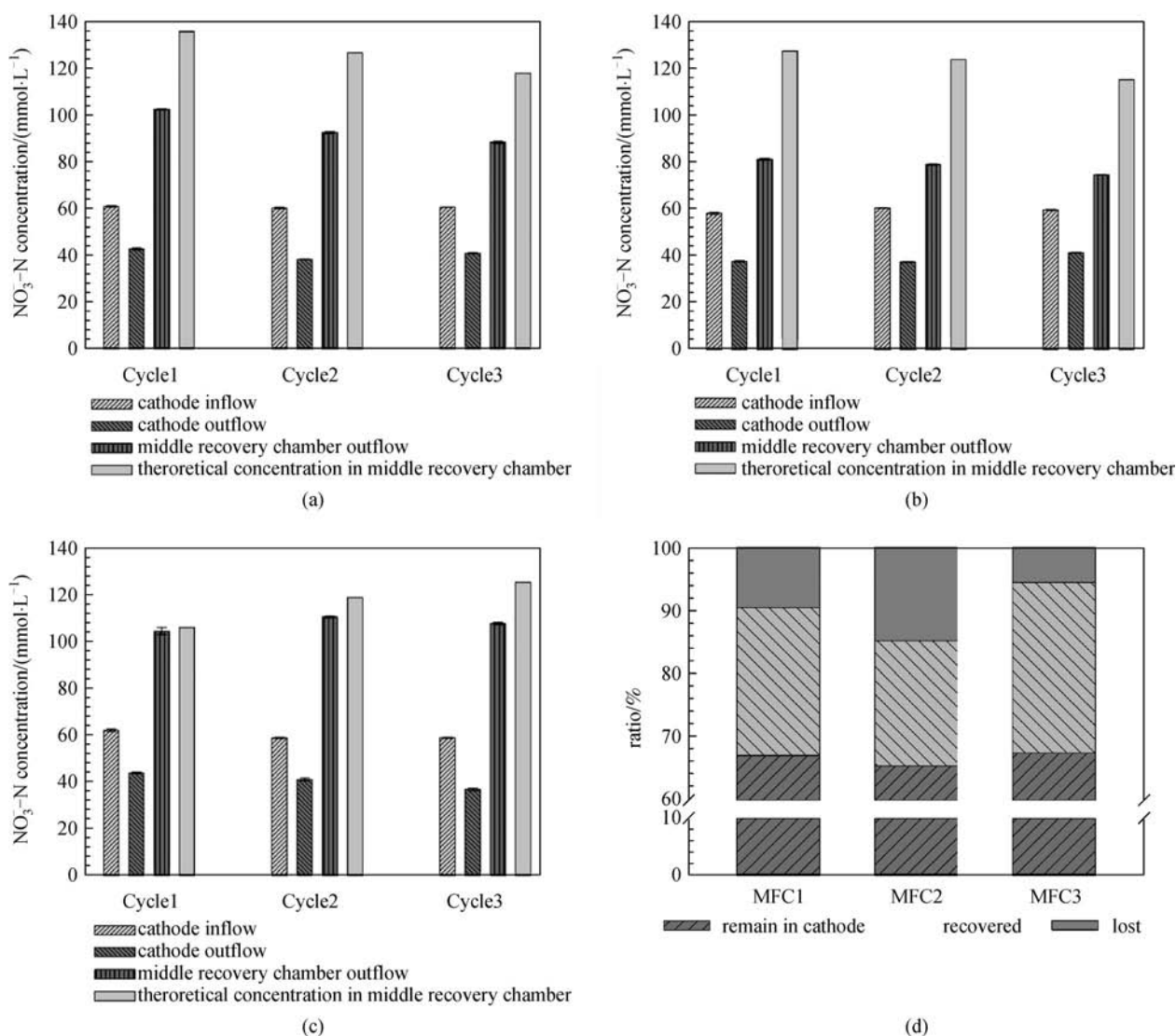
As discussed above, the major anion in cathode was  $\text{NO}_3^-$ , and the three MFCs had the same construction. Therefore, the impact of current density on nitrogen recovery could be evaluated with the electromigration of  $\text{NO}_3^-$ . Higher current density meant a higher rate for electromigration of  $\text{NO}_3^-$ , and the mass of electrochemi-

cally migrated ion of  $\text{NO}_3^-$  was related to charge of electrons. Similar charge of electron was collected in MFCs 1, 2 and 3 within the three cycles, which meant that the mass of electrochemically migrated  $\text{NO}_3^-$  in the three MFCs should be similar to each other. However, MFC3, which had the highest current density in these MFCs (Fig. 2), showed a higher  $\text{NO}_3^-$ -N recovery efficiency than that of MFC1 or MFC2. It's interesting that, when the voltage in MFC1 was only slightly higher (averaging 15%) than that in MFC2, the  $\text{NO}_3^-$ -N recovery efficiency in MFC1 was 25% higher than that in MFC2. Furthermore, from the results showed in Fig. 5(d), it seemed that higher voltage considerably decreased the transportation of  $\text{NO}_3^-$  from the middle recovery chamber to the anode chamber. The results showed a positive correlation between current

density and  $\text{NO}_3^-$ -N recovery efficiency in three-chamber MFCs.

## 4 Conclusions

In this study, we propose a proof-of-concept of the nitrogen recovery process by modifying MFCs using three chambers, and MFC technology shows a great potential to selectively recover nitrogen from synthetic wastewater. Higher current density promises higher recovery efficiency of both  $\text{NH}_4^+$ -N and  $\text{NO}_3^-$ -N. The study demonstrates that, without external energy supply, it is possible to recover most of nitrogen from wastewater e.g. swine wastewater.



**Fig. 5**  $\text{NO}_3^-$ -N concentration in the inflow and outflow of the cathode chambers, and actual and theoretical  $\text{NO}_3^-$ -N concentration in the outflow of the middle recovery chambers in MFC1 (a), MFC2 (b) and MFC3 (c), respectively. The error bar is for  $n = 3$  repeated tests. (d): Distribution of  $\text{NO}_3^-$ -N from the anode inflow within three cycles

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**Appendix** is available in the online version of this article at <http://dx.doi.org/10.1007/s11783-014-0730-5> and is accessible for authorized users.

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