Evaluation of a continuous flow microbial fuel cell for treating synthetic swine wastewater containing antibiotics

Dongle Cheng[a], Huu Hao Ngo[a,b,,*], Wenshan Guo[a], Soon Woong Chang[c], Dinh Duc Nguyen[c,d], Yiwen Liu[a] ,Yi Liu[e], Lijuan Deng[a], Zhuo Chen[f]
[a]Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Sydney, NWS 2007, Australia
[b]NTT Institute of Hi-Technology, Nguyen Tat Thanh University, Ho Chi Minh City, Viet Nam
[c]Department of Environmental Energy Engineering, Kyonggi University, 442-760, Republic of Korea
[d]Institution of Research and Development, Duy Tan University, Da Nang, Vietnam
[e]Department of Environmental Science and Engineering, Fudan University, Shanghai 200438, China
[f] Environmental Simulation and Pollution Control State Key Joint Laboratory, State Environmental Protection Key Laboratory of Microorganism Application and Risk Control (SMARC), School of Environment, Tsinghua University, Beijing 100084, PR China
***** Corresponding author: *E–mail address: H. H. Ngo; E–mail address: ngohuuhao121@gmail.com*

Abstract

Microbial fuel cell (MFC) systems are promising technologies for wastewater treatment and renewable energy generation simultaneously. Performance of a double-chamber microbial fuel cell (MFC) to treat synthetic swine wastewater containing sulfonamide antibiotics (SMs) was evaluated in this study. The MFC was operated in continuous modes at different conditions. Results indicated that the current was successfully generated during the operation. The performance of MFC under the sequential anode-cathode operating mode is better than that under the single continuous running mode. Specifically, higher removal efficiency of chemical oxygen demand (>90%) was achieved under the sequential anode-cathode operating mode in comparison with that in the single continuous mode (>80%). Nutrients were also be removed in the MFC's cathode chamber with the maximum removal efficiency of 66.6 ± 1.4% for NH_4^+-N and 32.1 ± 2.8% for PO_4^{3-}-P. Meanwhile, SMs were partly removed in the sequential anode-cathode operating with the value in a range of 49.4% - 59.4 % for sulfamethoxazole, 16.8% - 19.5% for sulfamethazine and 14.0% - 16.3% for sulfadiazine, respectively. SMs' inhibition to remove other pollutants in both electrodes of MFC was observed after SMs exposure, suggesting that SMs exert toxic effects on the microorganisms. A positive correlation was found between the higher NH_4^+-N concentration used in this study and the removal efficiency of SMs in the cathode chamber. In short, although the continuous

flow MFC is feasible for treating swine wastewater containing antibiotics, its removal efficiency of antibiotics requires to be further improved.

Keywords: Swine wastewater, microbial fuel cell, antibiotics, nutrients, continuous flow

1. Introduction

Small-scale pig farms have been gradually converted into large-scale and intensive pig industries to increase the pig production. As reported earlier, the world pig production increased by 456.6% from 1890 to 2014 (Roser, 2017). A consequence of the mass production of pigs was the discharge of large amounts of swine wastewater from intensive pig industries (Jin et al., 2020). Moreover, veterinary antibiotics are commonly used in swine industries as feed additives to prevent infectious diseases and enhance growth rates of pigs (Cheng et al., 2018b). Lou et al. (2018) reported that the annual antibiotics consumption in China’s pig production industry rose by 97000 tons. However, the low digestibility of antibiotics in animals’ guts produced high concentrations of antibiotics released into animal wastewater through animal faeces and urine (Cheng et al., 2018b). Zhang et al. (2015) indicated that around 50000 tons of antibiotics were released by animal wastes every year. Therefore, swine wastewater not only contains high concentrations of organic matter and nutrients, but also has been recognized as the major source of antibiotics in the environment (Cheng et al., 2018b). Consequently, the direct discharge or irrigation of swine wastewater can pose a high threat to the environment due to: firstly, the toxic effects of antibiotics on environmental biology; and secondly, the development of antibiotic resistant genes (ARGs) (Yang et al., 20202). Sulfonamide antibiotics (SMs) have been reported as the most widely used antibiotic classes in the swine industry and the dominant antibiotic components detected in swine wastewater (Han et al., 2020). According to the review paper by Cheng et al. (2018b), the detected concentration of SMs in swine wastewater was mainly in the 0.4 to 324.4 μg/L range. Hence, the effective treatment of swine wastewater containing antibiotics will significantly eliminate its dangerous effects on the environment and human health.

Anaerobic treatment technologies are commonly recommended for the treatment of swine wastewater, due to their cost effectiveness and high concentrations of organic matter in swine wastewater (Zeng et al., 2019). Nevertheless, the widely used anaerobic processes in current wastewater treatment plants are mainly devised to remove general pollutants, yet have been poor in their antibiotics' removal efficiency (Cheng et al., 2018b). As well, nutrients contained in swine wastewater could not be effectively removed by single anaerobic processes (Cao et al., 2019). Bioelectrochemical systems (BESs) are promising technologies for treating wastewater containing antibiotics with combined processes of microbial metabolism and electrochemical redox reactions (Hassan et al., 2020). For example, microbial fuel cell (MFC) has emerged as a 'green' technology for improving the degradation of refractory pollutants from wastewater and generating electricity simultaneously through microorganisms on the anode (Wu et al., 2020; Zhou et al., 2018). The effective removal of SMs from wastewater by MFCs has been reported in recent studies, mainly because of the enhanced microbial metabolism in the presence of anode (Wang et al., 2016; Wu et al., 2020). For instance, our previous study found higher removal efficiencies of SMs from swine wastewater in a batch running MFC reactor than those in a conventional anaerobic reactor (Cheng et al., 2020b). Wang et al. (2016) and Wu et al. (2020) demonstrated that sulfamethoxazole (SMX) can be rapidly and completely degraded by MFC and the power generation was enhanced due to the presence of SMX.

Another sulfonamide antibiotic, sulfadiazine (SDZ), can also be degraded into 2-aminopyrimidine, 2-amino-4-hydroxypyrimidine and benzenesulfinic acid under the behavior of microorganisms in MFCs (Wang et al., 2018). The effective removal or recovery of nutrients from wastewater by MFCs also has been reported previously (Ye et al., 2019a). However, most previous studies only focused on the removal of individual antibiotics by

MFCs, while different antibiotics are usually present in swine wastewater simultaneously (Zhi et al., 2020). It is still unclear about the removal of multiple antibiotics in MFCs and the influence of mixed antibiotics on the performance of MFCs. Furthermore, the removal of antibiotics by MFCs was mainly done in the batch mode so far, which is not effective and realistic in practical applications in the future. Moreover, less attention was paid to the feasibility of applying MFC processes to the treatment of swine wastewater containing antibiotics.

Therefore, a continuous flow operation of MFC was conducted in this study for simultaneously removing sulfamethoxazole (SMX), sulfamethazine (SMZ) and sulfadiazine (SDZ) from synthetic swine wastewater. The removal efficiency of COD, nutrients and the selected antibiotics was examined in single continuous and sequential anode – cathode continuous mode of MFC, respectively. The effect of higher ammonium and phosphorus centration in swine wastewater on the performance of the MFC was also investigated in this research. Furthermore, the generation of electricity was also investigated.

2. Materials and methods

2.1 Experimental design and set up

A double-chamber MFC reactor was made of plexiglass material, with the effective volume of 350 ml for each chamber. A cation exchange membrane (CEM) (CMI7000, Membranes International Inc., USA) was used to separate the two electrodes. The anode and cathode consisted of a cylindrical graphite felt (3 cm in diameter and 6 mm thickness) and a carbon-fiber brush (3 cm diameter and 3 cm length), respectively. Electrodes used in this study were bought from Sanye Carbon Co. Ltd. in China. The two electrodes were connected by copper wire, and the external resistance was 1000 Ω. Anaerobic sludge collected from a pilot-scale anaerobic digester (sewage sludge used as feedstock, HRT=15d, volume=25L) was used to inoculate the anode chamber by mixing it with synthetic swine wastewater to make a concentration of mixed liquor suspended solids (MLSS) of 5000 mg/L. The synthetic swine wastewater mainly contained glucose (3000 mg/L of COD), NH_4Cl (223 and 446 mg/L), KH_2PO_4 (66 and 132 mg/L), $MgSO_4 \cdot 7H_2O$ (54 mg/L) and $CaCl_2 \cdot 2H_2O$ (4 mg/L). Major contaminants of the synthetic swine wastewater were based on components of swine wastewater reported previously (Huang et al., 2015). After voltage stabilization, SMX, SMZ and SDZ were added to the synthetic swine wastewater with the initial concentration of 100 μg/L. The feeding solution was purged with nitrogen gas for 15 min and then adjusted to pH 7.5 ± 0.15 by employing $NaHCO_3$ (1 mol/L) and H_2SO_4 (0.1 mol/L) solutions prior to pumping into the anode chamber.

2.2 Experimental operation

In this experiment, the double-chamber MFC was operated under single continuous mode and sequential anode-cathode mode, respectively, as shown in Fig.1. Throughout the experiment, the synthetic swine wastewater was continuously pumped from the bottom of the anode chamber via a peristaltic pump (Model 77202-60, Masterflex, Illinois, United States). In the single continual operating mode (mode 1), water flowed out continuously from the anode outlet. Under this operating mode, deionized water was supplied in the cathode compartment as catholyte. To further purify effluents of the anode chamber, the sequential anode-cathode mode was conducted (mode 2). Under this operating mode, effluent from the anode chamber served as a continuous influent of the cathode chamber to be further treated under aerobic conditions. Considering the large nutrients content in swine wastewater, the concentrations of NH_4Cl and KH_2PO_4 were increased from 223 to 446 mg/L and 66 to 132 mg/L, respectively, in the later operating mode (mode 3), to study their effect on antibiotics' removal efficiency and MFC performance. The whole experiment was conducted at the HRT of 24 h and at room temperature (~25 °C).

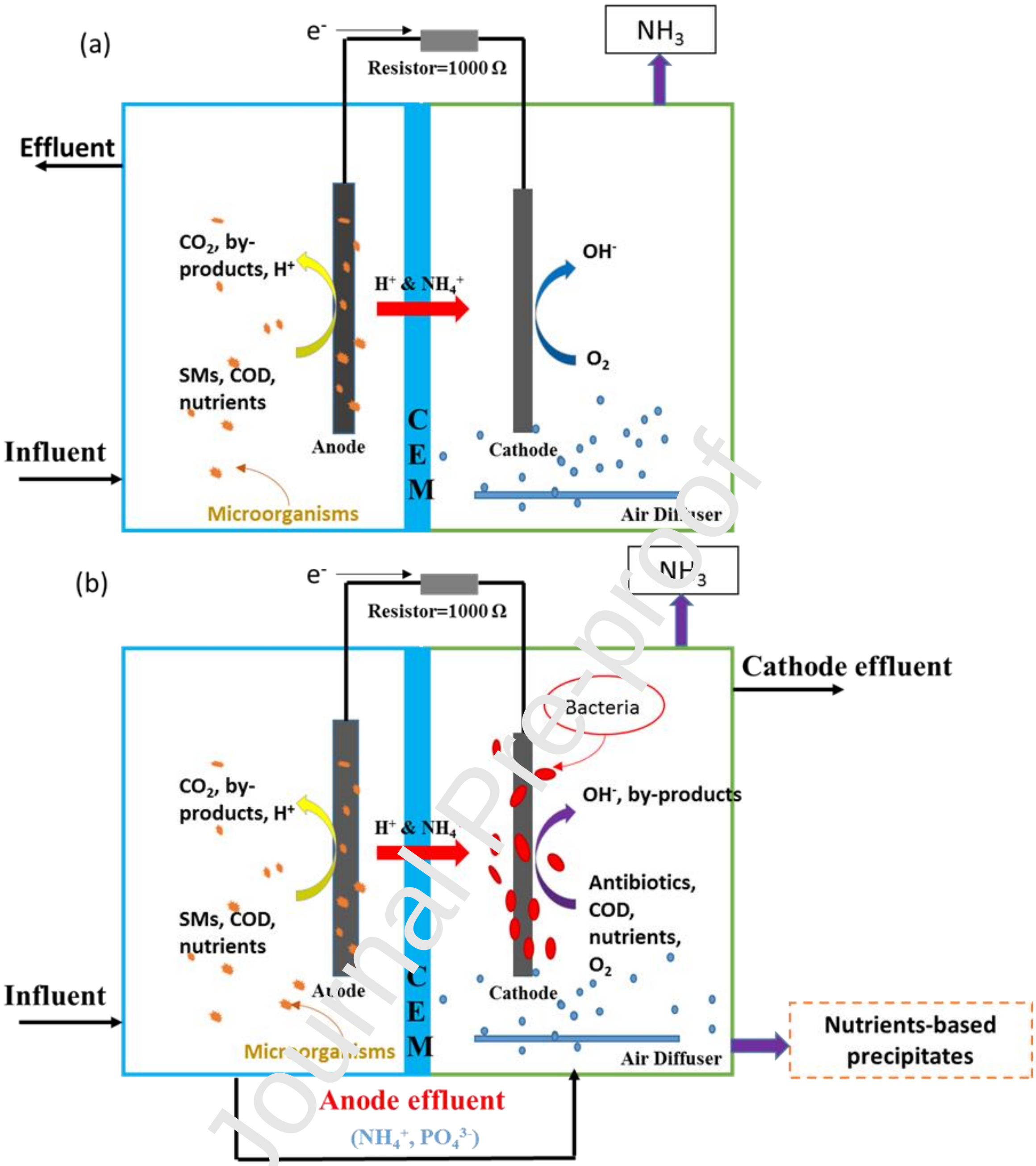

Fig. 1 Schematic diagram of the MFC operated under: (a) single continuous mode and (b) sequential anode-cathode mode

2.3 Analytical methods

The concentrations of SMX, SMZ and SDZ in the effluent samples collected from the MFC's anode and cathode chamber were detected using triple quadrupole liquid chromatograph mass spectrometer (LCMS-8060, Shimadzu). The separation of antibiotics was performed by a Phenomenex C18 column (Luna, 3.0 × 100 mm, 3 μm) with a mobile phase of water and acetonitrile with 0.1% (V/V) formic acid at a constant temperature of 28 °C. The concentrations of COD, NH_4^+-N and PO_4^{3-}-P concentration in the effluent sample were determined based on the Standard Methods (Federation and Association, 2005), by using the test kit HI93754B-25 (Hanna Instruments Australia, Melbourne, Australia) for COD, 100683 and114848 (Merck Millipore, Burlington, USA) for NH_4^+-N and PO_4^{3-}-P, respectively. Before being sent for analysis, the collected samples were filtered by 0.20 μm filters (Merck Millipore, Burlington, USA). A DO meter (OM-51, Horiba, Tokyo, Japan) was employed to check the DO concentration in the cathode chamber (maintained at 6 mg/L of DO during the operation). The cell voltage (U) generated during the experiment periods was detected by a universal digital meter (VC86E, Shenzhen City Station Win Technology Co. Ltd., Shenzhen, China). The value of the voltage generated was recorded every two hours from 8:00 am to 8:00 pm every day during the operating period, and the daily average value was reported here. Duplicate samples were collected and analyzed in this study and mean values were presented.

3. Results and discussions

3.1 SMs removal in continuous flow MFC systems

The simultaneous removals of SMX, SMZ and SDZ in different operation modes of the continuous flow MFC are displayed in Fig. 2. In mode 1 (single continuous mode), removal efficiencies of SMs in the anode compartment of MFC and the open-circuit control were compared. From Fig. 2, it is observed that higher removal efficiencies of SMs were achieved in the MFC than those in its open-circuit control system. Similar results have been concluded by earlier studies for the removal of individual SMX and SDZ in MFC systems (Miran et al., 2018; Song et al., 2018). Under this single continuous mode, the average removal efficiency of SMX, SMZ and SDZ in the anode chamber of MFC was 48.6%, 10.3% and 15.4%, respectively. This result is comparable with previous studies about the removal of SMs in conventional anaerobic digesters (Chen et al., 2012; Tang et al., 2019). For instance, the removal efficiency of SMX and SDZ was 31% and 8.3%, respectively, in the anaerobic process for swine wastewater treatment (Chen et al., 2012). The resistance removal of SDZ during anaerobic digester was also found by the study of Tang et al. (2019). Accordingly, the removal of SMs in both closed and open-circuit MFC systems was due to the role of biodegradation in comparison to adsorption (Cheng et al., 2020a; Harnisch et al., 2013). In the MFC system, degradation products of SMX and SDZ, their degradation mechanisms, as well as the functional microorganism that contributed to their removal have been successfully identified by previous researchers (Wang et al., 2018; Xue et al., 2019). The relatively higher efficiency in removing SMs in the MFC system was mainly possible because the enhanced metabolic rate of bacteria in the anode chamber that caused by the electron transfer to the cathode chamber via the external circuit. This could also result in faster oxidation of the co-substrate to produce more electrons for SMs degradation leading to accelerated degradation of SMX (Aghababaie et al., 2015).

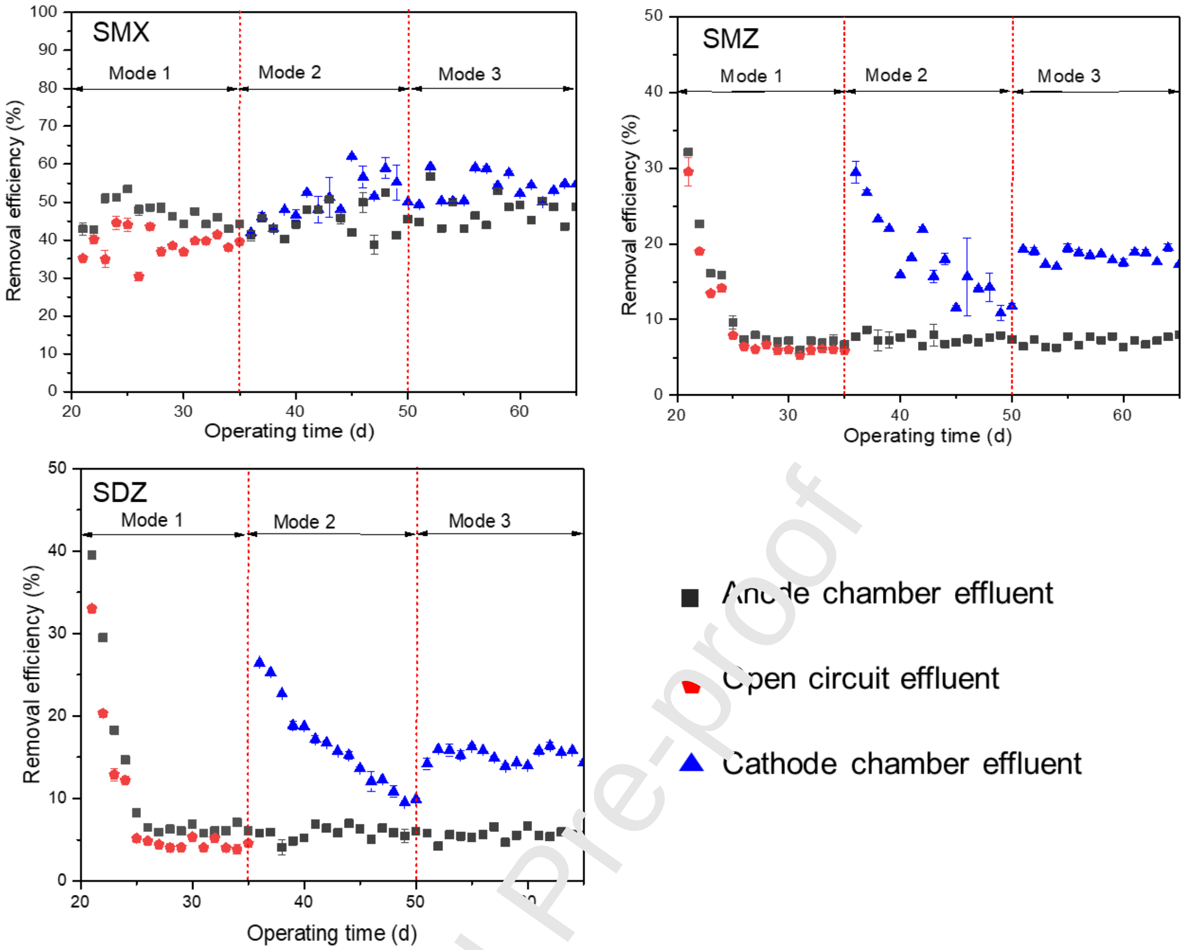

Fig. 2 Removal efficiencies of SMs during the continuous operation of MFC under different scenarios: (a) SMX removal efficiency; (b) SMZ removal efficiency; (c) SDZ removal efficiency.

Compared with the high removal efficiency (>80%) of individual SMX and SDZ in MFC systems under self-loop batch operating conditions (Miran et al., 2018; Wang et al., 2018; Xue et al., 2019), the efficiency in the simultaneous removal of SMX, SMZ and SDZ was quite low under the single continuous operation mode of MFC in this study. As shown in Fig. 2, the removal efficiency of SMX, SDZ and SMZ in the mode 1 was in the 42.6 ± 0.1% - 53.4 ± 0.4%, 6.4 ± 0.2% - 32.1 ± 0.1% and 5.9 ± 0.3% -35.2 ± 0.5% range, respectively. SMX showed higher and more stable removal than SDZ and SMZ in this study, this result is consistent with one of our previous study (Cheng et al., 2020b). Cheng et al. (2020b) investigated the degradation of SMX, SDZ and SMZ in a batch operating of MFC and indicated that the biodegradation of SMX was easier than that of SDZ and SMZ with a much higher degradation rate constant and less time for the degradation of 50% SMs. A similar result was concluded by Harnisch et al. (2013), and these authors found SMX was completely removed but only some SMZ was removed in the batch operating MFC in 7 days. Moreover, a decline was observed for the removal efficiency of SMs in the first few days after continuous pumping of SMs into the MFC, which reflected the toxic effect of SMs and/or their degradation intermediates on the microorganisms responsible for this degradation. In addition to the different operating conditions in comparison to a previous study, the poor removal efficiency of SMs in this study may also result from the increased toxicity of the coexistence of SMX, SDZ and SMZ to the microorganisms in MFC. The review paper by Cheng et al. (2018a) has indicated that combined antibiotics revealed more inhibition than the individual antibiotic. It is clear that effluent from the anode chamber of the MFC still contains a high concentration of SMs, so further treatment is necessary to reduce their emission into the environment.

To increase the removal efficiency of SMs in the continuous operation of MFC, the sequential anode–cathode mode (mode 2) was conducted in the present study. Under this mode, the effluent from the anode compartment was used as a continuous feed for the cathode chamber, which enabled aerobic bacteria to grow in the cathode part (Freguia et al., 2008). Therefore, the double-chamber MFC can be operated as a combination of the anaerobic (anode) and aerobic (cathode) processes, in which the pollutants in the anode chamber effluent may be further degraded by aerobic bacteria activities. An obvious improvement was observed for the removal of SMZ and SDZ in the cathode effluent compared to their removal in the effluent from the anode chamber, although their removal also decreased gradually as this operation progressed (see Fig. 2).

By contrast, the removal efficiency of SMX experienced only a slight improvement in this mode although the biodegradability of SMX under anaerobic and aerobic conditions has been extensively concluded by earlier studies (Müller et al., 2013; Wang and Wang, 2018a). The possible reason might be the relatively high efficiency in removing SMX in the anode chamber may limit its biodegradation rate in the following cathode chamber, possibly due to the low bioavailability of SMX when its concentration was too small (Wang and Wang, 2018a; Wang and Wang, 2018b). In contrast, the high residual concentration of SDZ and SMZ in the anode chamber effluent may make them much more bioavailable in the cathode chamber. Collectively, the sequential anode–cathode operating mode of MFC can enhance the removal efficiency of SMs from swine wastewater.

Considering the large ammonium concentration in swine wastewater and the influence of ammonium on the microorganisms in MFC, the NH_4Cl concentration in the synthetic swine wastewater was increased from 223 mg/L to 446 mg/L in mode 3 of this experiment. This was done to investigate its effect on the removal of SMs and the performance of MFC. As observed in Fig. 2, the removal efficiency of SMs was increased in the cathode effluent when the NH_4^+-N concentration was doubled in the feeding wastewater. Based on previous studies, the increased removal of SMs may be linked to the improved activities of ammonia-oxidizing bacteria (AOB) in the cathode chamber of MFC by increasing the NH_4^+-N concentration. The AOB can use NH_4^+-N as the growth substrate, which increased largely through increasing NH_4^+-N concentrations. A positive relationship between micropollutants elimination and AOB activity has been reported in other analyses (Kumwimba and Meng, 2019; Xu et al., 2016). The biodegradation of micropollutants by AOB was mainly due to its non-specific enzyme ammonia monooxygenase (AMO), which is able to degrade various kinds of micropollutants through cometabolic biodegradation (Helbling et al., 2012; Roh et al., 2009; Xu et al., 2016). Hence, an inherent connection can be found between the removal of SMs and the concentration of NH_4^+-N in MFC (Delgadillo-Mirquez et al., 2011; Tran et al., 2016; Tran et al., 2018). It can be assumed that the increase in the growth substrate NH_4^+-N to some extent can improve the cometabolic degradation of SMs, which has been confirmed by the experimental results of the present study.

Overall, the final removal efficiencies of SMX, SMZ and SDZ in this sequential anode-cathode double-chamber MFC were around 49.4 ± 0.2% - 59.4 ± 0.6%, 16.8 ± 0.1% - 19.5 ± 0.5% and 14.0 ± 0.2% - 16.3 ± 0.4%, respectively. Based on the above discussion, such poor removal efficiency of SMs in this MFC system was mainly due to the toxic effect of SMs and or degradation byproducts on the microbial community in both the anode and cathode electrodes. However, this study also discovered that the activity of the microorganism that was responsible for degrading the SMs in the MFC could be recovered gradually after stopping exposure to SMs. After 5 days stopping the addition of SMs to the influent of MFC, the SMs removal efficiencies reached 78.2 ± 0.7% for SMX, 51.9 ± 0.4% for SMZ and 49.5 ± 0.6% for SDZ. Therefore, further research is required to reduce the toxic effect of antibiotics on microorganisms to improve the removal efficiency.

3.2 Electricity generation

Electricity generation was evaluated by monitoring the voltage production in the present study. Fig. 3 describes the variations in voltage production at different operating modes of MFC for treating swine wastewater. In the acclimatization process, the wastewater was continuously pumped into the anode compartment of the double-chamber MFC, and the MFC was conducted under open circuit mode for 20 days to form the biofilm on the anode electrode's surface. Thereafter, the closed circuit mode was conducted until the stable operation of the MFC with the voltage generation ranged from 630 mV to 720 mV, indicating the electrochemically active biofilms were formed on the anode of MFC (Zhou et al., 2018). Little change was observed for the voltage generation after the injection of SMs (see Fig. 3).

This is consistent with the study by Harnisch et al. (2013), who found that the power output remained constant after exposure to SMs in the MFC.

Interestingly, some researchers even detected an increase in power generation after the addition of a single antibiotic (SMX or oxytetracycline) (Sun et al., 2019; Wu et al., 2020). Wu et al. (2020) explained that the increased abundance of exoelectrogens in MFC was due to the decline in the competitive population in the anode chamber caused by the presence of SMX. Zhou et al. (2018) demonstrated that although the MFC's output voltage could be inhibited by adding antibiotics into the reactor, it subsequently improved when the concentration of antibiotics shrunk. The present result of this study may explain why the inhibition of SMs (100 μg/L) to exoelectrogenic bacteria was less when compared to other competitive organisms in the MFC's anode. In addition, the degradation products of SMX, SMZ and SDZ may serve as mediators to facilitate the electron transfer from bacteria to the anode, thereby compensating for their inhibition (Sun et al., 2019).

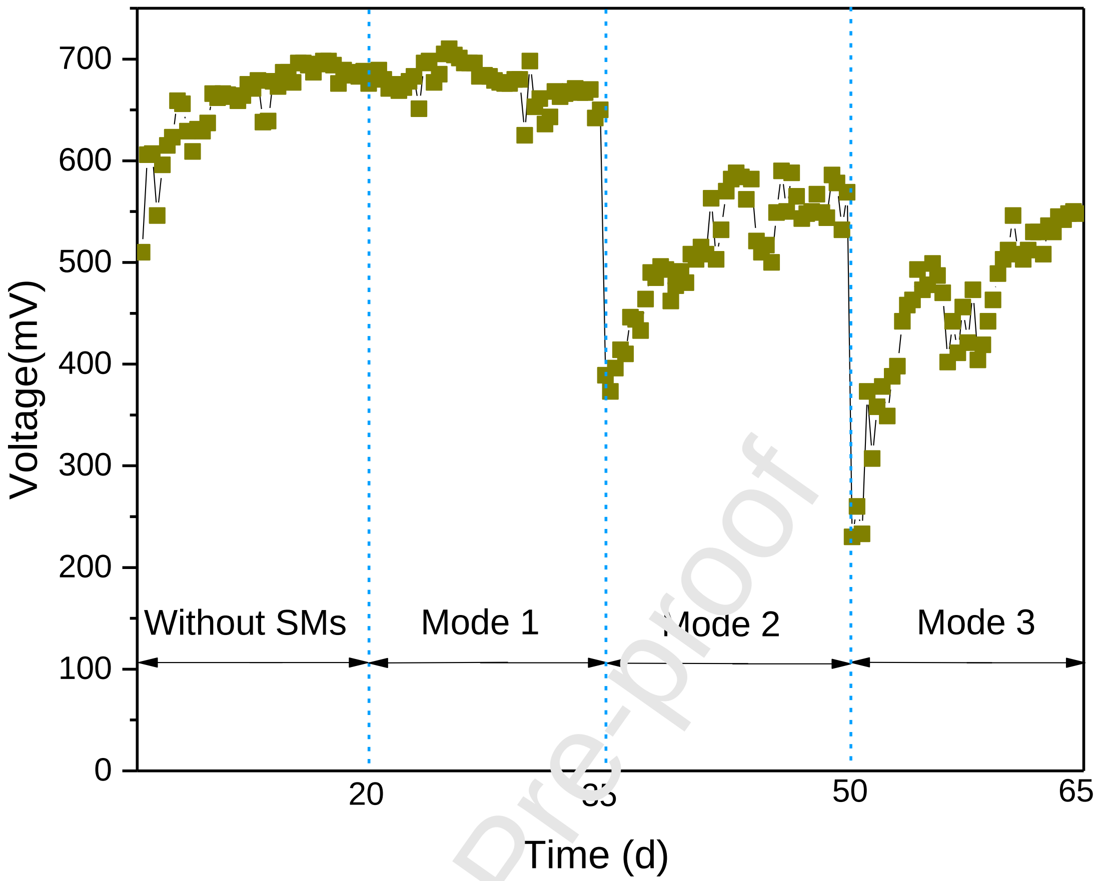

Fig. 3 Voltage generation when the MFC is operating under different modes with and without SMs addition.

It can be seen that the voltage generation decreased in the sequential anode–cathode mode. Generally, the generation of electricity in the MFC mainly depends on: 1) the oxidation of organic substrates in anode, 2) the transfer of electrons from microbes to anode, 3) transfer of protons through the membrane, and 4) the concentration of oxygen in the cathode chamber (Amari et al., 2015; Gil et al., 2003). Compared with the single continuous mode, the catholyte in this mode was changed from deionized water to the continuous supply of anode effluent. The residual organic matter in the anode chamber effluent enhances the growth of aerobic heterotrophs in the cathode compart, leading to the reduction of oxygen concentration in the cathode (Freguia et al., 2008). This has been confirmed by the further removal of COD in the cathode section. In this study, oxygen served as the final electron accepter in the cathode portion, and the competitive consumption of oxygen can limit the voltage generation (Amari et al., 2015; Najafgholi et al., 2015). The other possible reason is due to the permeation of oxygen from the anode to the cathode chamber through what connects them. This may limit the activity of some exoelectrorogenic bacteria in the anode chamber and decrease the amount of power generation (Kim et al., 2007; Najafgholi et al., 2015). Facultative bacteria could use oxygen as electron accepter in the anode chamber without transferring electrons to the anode, thereby reducing current generation (Oh et al., 2009). The investigation by Oh et al. (2009) found that the power generation of a double-chamber MFC decreased by increasing the oxygen concentration in the anode chamber.

The voltage generation fell to around 230 mV after the NH_4^+-N concentration in the influent doubled suddenly (Fig. 3). The negative effect of ammonium on electricity generation in MFC systems was also reported by other researchers (Hiegemann et al., 2018; Kim et al., 2011; Tice and Kim, 2014; Ye et al., 2019b). For instance, Ye et al. (2019b) investigated the impact of ammonium on power generation in a double-chamber MFC and indicated that the voltage generation decreased gradually by increasing the NH_4^+-N concentration from 5 to 40 mg/L. This may result from the inhibition of NH_4^+-N on the activity of exoelectrogenic bacteria in microbial fuel cells (MFCs) (Liu et al., 2017). Conversely as can be seen from Fig. 3, the power generation recovered gradually with the operating time, suggesting the adaptation of bacteria to the double ammonium concentration (117.8 mg/L) used in the last mode through continuous exposure (Kim et al., 2011). Therefore, the electricity generated by the MFC was affected by the sudden increase of ammonium concentration in the influent, whereas, it could be recovered gradually under long-term continuous flow operation.

3.3 COD removal

The efficiency of the MFC in this study for removing COD from swine wastewater was investigated under different operating modes (Fig. 4). As can be seen from Fig. 4, COD can be effectively removed (90.9 ± 0.1% - 93.5 ± 0.2%) in the anode chamber of the MFC when operations remain stable. The COD removal efficiency in the MFC was better than that in its open-circuit control system (85.0 ± 0.1% - 88.3 ± 0.6%) under mode 1. This finding confirmed that the exoelectrogenic bacteria activities in the MFC anode were also responsible for COD removal compared to the open circuit system with no exoelectrogenic bacteria. The study by Zhang et al. (2015) and Cheng et al. (2020b) also proved that the current production in MFC could accelerate the COD removal rate and the final removal efficiency in comparison with the open circuit control system. A slight fall in the COD removal efficiency (80.2 ± 0.6% - 91.2 ± 0.6%) did occur after adding SMs into the feeding wastewater. Comparatively, a greater reduction in COD removal efficiency was observed in the open-circuit system (average value of 12.3%) than that in the MFC system (average value of 7.5%) after the injection of SMs. This indicated that the MFC system had better buffering capacity and adaptability to SMs than the conventional anaerobic reactor. According to the review by Cheng et al. (2018a), the reduced removal of COD might due to the stimulation of antibiotics to fermentative or acid-forming bacteria and the inhibition of antibiotics to volatile fatty acids (VFAs) degrading bacteria and acetoclastic methanogens. Wang et al. (2018) also indicated that the biotoxicity of SMs might be effectively eliminated in the MFC.

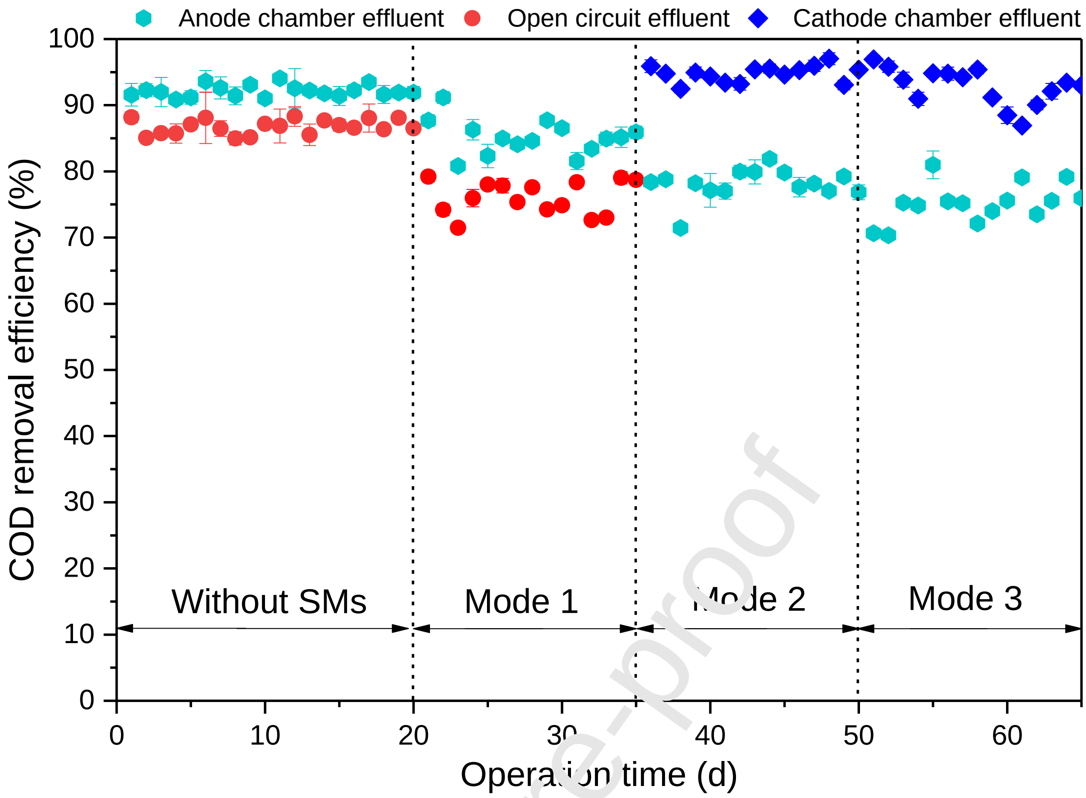

Fig. 4 COD removal from swine wastewater as MFC operates under different modes with and without SMs present.

In the sequential anode–cathode scenario, COD removal efficiencies in the anode chamber (71.46 ± 0.1% - 81.85 ± 0.2%) were relatively low in comparison with the previous mode, but their further increase was evident in the following cathode chamber (92.5 ± 0.4% - 97.0 ± 0.6%). The slight decrease in COD removal in the anode chamber might be attributed to the intrusion of oxygen into the anode chamber, which restricted the activity of anaerobic microorganisms. The overall large removal of COD in the sequential anode–cathode configuration of the MFC was consistent with what previous studies have reported. They stated that COD could be further removed at the cathode compartment by heterotrophic bacteria (Freguia et al., 2008; Wen et al., 2010). Hence, the enhanced removal of COD in the cathode chamber of MFC in this study confirmed the presence of aerobic heterotrophs.

Although electricity generation was influenced by the sudden increase in NH_4^+-N concentration in the feeding wastewater, the COD removal efficiency was only minimally affected. It maintained a similar range to the last operating mode in both the anode and cathode electrodes (see Fig. 4). Research by other groups has found similar results. For instance, Ye et al. (2019b) did not observe any reduction in COD removal efficiency in the MFC because the influent concentration of NH_4^+-N increased from 5 to 40 mg/L. Liu et al. (2017) also found little change in COD removal efficiency (from 95.8% to 92.2%) when the NH_4^+-N concentration rose from 45 mg/L to 600 mg/L. It is understood that the effect of NH_4^+-N on COD removal efficiency mainly depends on the concentration of NH_4^+-N, COD/NH_4^+-N (C/N) ratio and other operating parameters (Koraturk and Erguder, 2016; Li et al., 2016; Tice and Kim, 2014; Yenigun and Demirel, 2013). As reported by Tice and Kim (2014), COD removal efficiency only started to decline for MFCs when the NH_4^+-N concentration exceeded 1000 mg/L. Yadu et al. (2018) maintained that a substantial removal efficiency of COD (>90%) could be achieved with a high C/N ratio (7-30). Therefore, considering the relatively low concentration of NH_4^+-N (150 mg/L) and high C/N (15:1) ratio in the synthetic swine wastewater used in this study, it is reasonable to observe that only a small fluctuation in COD removal efficiency occurred.

3.4 Nutrients removal

Fig. 5 illustrates the removal of NH_4^+-N and PO_4^{3-}-P in the double-chamber MFC for different operating conditions. Before the injection of SMs into the feeding wastewater, the removal efficiency of NH_4^+-N in the anode chamber of MFC was around 35.1 ± 0.8% - 37.1 ± 0.7% with the initial concentration of 75 mg/L. Previous reports have demonstrated that the potential mechanism for the removal of NH_4^+-N in the anode chamber of MFC was the joint action of biological processes and diffusion of NH_4^+ from the anode chamber to cathode chamber through CEM membrane (Kim et al., 2008; Min et al., 2005). It is obvious that one aspect of ammonium reduction was to synthesize new biomass in the anode chamber.

Meanwhile, NH_4^+ in the anode chamber can transfer to the cathode chamber through a cation exchange membrane (CEM), which was further converted into NH_3 under alkaline pH and air-breathing conditions in the cathode chamber (Zhou et al., 2015). The NH_3 removed by stripping can be considered for recovery by sulfuric acid absorption. As reported earlier, the removal of NH_4^+-N in the anode compartment might also occur through nitrification/denitrification and/or the *Anammox* process (He et al., 2009; Kim et al., 2016). However, NO_3^--N or NO_2^--N was not detected in the anode chamber of this study, suggesting that the reaction of nitrification/denitrification or Anammox was impossible due to the anaerobic conditions. This result agrees with the previous report by Tao et al. (2014), who indicated that no nitrifying or anammox bacteria existed in the anode chamber.

The removal of PO_4^{3-}-P in this mode was in the range of 37.7 ± 0.6% - 44.2 ± 0.8% with the initial concentration of 15 mg/L, which was mainly through absorption by the microbial organisms. After the presence of SMs in the anode chamber was observed, a decline in NH_4^+-N and PO_4^{3-}-P removal efficiency in the anode chamber became evident (Fig. 5). Similar to the effect of SMs on COD removal, the inhibition of SMs to the microorganisms could reduce their uptake to these nutrients. It is notable that the constant voltage generation following the addition of SMs could confirm the stable movement of NH_4^+ between the two electrodes. Therefore, the toxic effect of SMs on the microorganism was the main explanation for the reduced removal of both NH_4^+-N and PO_4^{3-}-P in the anode chamber.

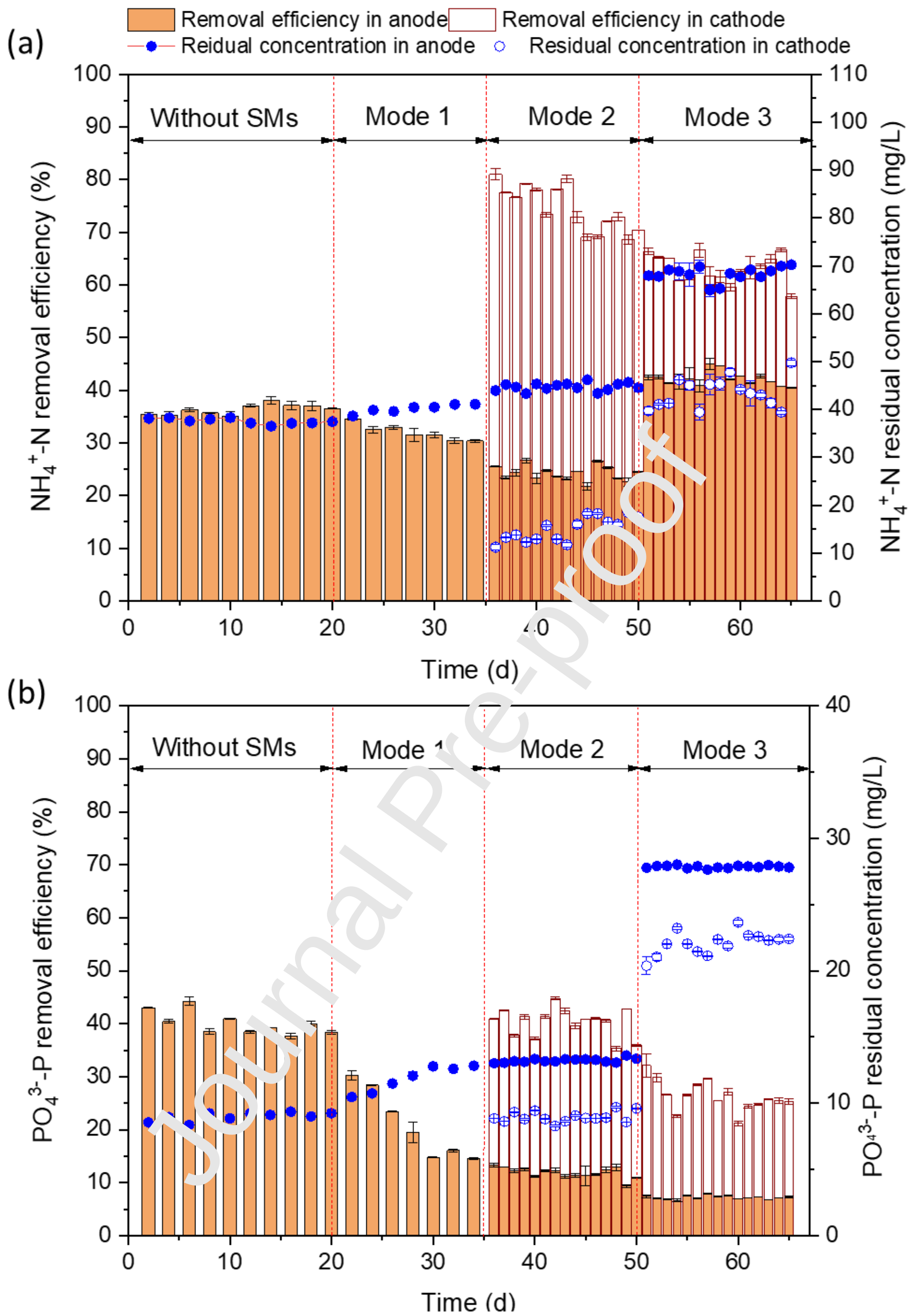

Fig. 5 Nutrients removal efficiencies as the MFC operated under different modes with and without SMs addition: (a) NH_4^+-N removal efficiency; (b) PO_4^{3-}-P removal efficiency.

A decreasing trend was observed for the removal efficiency of NH_4^+-N (21.7 ± 0.7% - 26.6 ± 0.5%) and PO_4^{3-}-P (9.4 ± 0.3% - 13.3 ± 0.4%) in the anode chamber under the sequential anode-cathode running mode. According to the above discussion, this may result from the relatively low microbial activities in the anode compartment in comparison with the previous mode. Additionally, the NH_4^+ concentration gradient between the anode and cathode electrodes was lower than the last mode due to the residual NH_4^+ in the anode effluent, which limited the ammonium diffusion to the cathode compartment (Ye et al., 2019b). It can be seen from Fig. 5 that the removal efficiencies of NH_4^+-N and PO_4^{3-}-P in the following cathode chamber further rose to 68.6% - 81.0% and 35.3% - 44.8%, respectively.

It is clear that part of NH_4^+-N and PO_4^{3-}-P could be used as a substrate to maintain the activity of heterotrophic bacteria in the cathode chamber. As well, NH_4^+-N can be oxidized by the activity of nitrifiers and AOB, which was suggested by the detected NO^{3-} (1.0 - 1.6 mg/L) and NO^{2-} (0.06 - 0.1 mg/L) in the cathode effluent. In addition, the reaction between oxygen and the electrons can form hydroxyl ions, resulting in an increase in the pH value in the cathode compartment. Previous reports have documented that NH_4^+-N and PO_4^{3-}-P can be recovered from swine wastewater by precipitation when the pH rises to between 8 and 10 (Kim et al., 2017; Suzuki et al., 2007). As monitored in this study, the pH value in the cathode chamber ranged from 8.2 to 8.6, which facilitated the removal/recovery of NH_4^+-N and PO_4^{3-}-P through precipitation with magnesium and/or calcium ions in the wastewater [14]. Air stripping also contributed to the higher removal efficiency of NH_4^+-N when the aeration condition and high pH value in the cathode chamber were taken into consideration (Ye et al., 2019a).

Interestingly, the removal efficiency of NH_4^+-N in the anode chamber increased to >40% when the initial concentration also increased (see Fig. 5). This outcome agrees with the study by Ye et al. (2019b), who also found the increased amount of NH_4^+-N being removed was made possible by increasing the NH_4^+-N concentration. The more NH_4^+ there was in the anode chamber increased the ammonium concentration gradient between the two electrodes, which enhanced its diffusion to the cathode chamber. In contrast, the average PO_4^{3}-P concentration reduction in the anode chamber was 1.8 mg/L and 2.2 mg/L, respectively, with the initial concentration of 15 mg/L and 30 mg/L. Therefore, although the PO_4^{3-}-P removal efficiency in the anode compartment decreased by increasing its initial concentration to 30 mg/L, a slight increase was also observed for the capacity of bacteria in the anode chamber to absorb phosphorus. As for their removal in the cathode compartment, the simultaneous reduction for the removal of ammonium and phosphate was mainly possible due to the deficiency of magnesium and/or calcium ions in the synthetic swine wastewater, which limited their recovery by precipitation (Kim et al., 2017).

4. Conclusions

A continuous flow double-chamber MFC was used for simultaneously removing antibiotics, organic matter and nutrients from swine wastewater. Results indicated that the sequential anode-cathode operation mode of the MFC performed better than the single continuous mode in terms of the removal of organic matter, nutrients and SMs from swine wastewater. Electricity could be successfully generated by pumping synthetic swine wastewater to the anode chamber continuously. In this study, negative effects of SMs on the removal of the above-mentioned pollutants in MFC was observed in the first few days after their exposure, whereas little effect was found on the voltage generation. Results also indicated that a higher NH_4^+-N concentration could improve the removal of SMs in the cathode chamber. Overall, this study successfully convinces that the continuous flow MFC is feasible for treating swine wastewater that contains antibiotics, but the challenge is to further improve the removal efficiency of SMs. Further investigation of this proposed MFC using real swine wastewater is necessary to ensure its practical application.

Acknowledgement

This research was supported by the Centre for Technology in Water and Wastewater, University of Technology Sydney, Australia (UTS, RIA NGO) and the Korea Institute of Energy Technology Evaluation and Planning (KETEP) and the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea (Grant No. 20173010092470).

References

1. Aghababaie, M., Farhadian, M., Jeihanipour, A. and Biria, D. 2015. Effective factors on the performance of microbial fuel cells in wastewater treatment–a review. Environ. Technol. Rev. 4 (1), 71-89.

2. Amari, S., Vahdati, M. and Ebadi, T. 2015. Investigation into effects of cathode aeration on output current characteristics in a tubular microbial fuel cell. Int. J. Environ. Sci. Technol. (Tehran) 12 (12), 4037-4042.

3. Cao, L., Wang, J., Xiang, S., Huang, Z., Ruan, R. and Liu, Y., 2019. Nutrient removal from digested swine wastewater by combining ammonia stripping with struvite precipitation. Environ. Sci. Pollut. Res. 26 (7), 6725-6734.

4. Chen, Y., Zhang, H., Luo, Y. and Song, J., 2012. Occurrence and dissipation of veterinary antibiotics in two typical swine wastewater treatment systems in east China. Environ. Monit. Assess.184 (4), 2205-2217.

5. Cheng, D., Ngo, H., Guo, W., Chang, S., Nguyen, D., Kumar, S.M., Du, B., Wei, Q. and Wei, D. 2018a. Problematic effects of antibiotics on anaerobic treatment of swine wastewater. Bioresour. Technol. 263, 642-653.

6. Cheng, D., Ngo, H., Guo, W., Liu, Y., Zhou, J., Chang, S., Nguyen, D., Bui, X. and Zhang, X. 2018b. Bioprocessing for elimination antibiotics and hormones from swine wastewater. Sci. Total Environ. 621, 1664-1682.

7. Cheng, D., Ngo, H.H., Guo, W., Chang, S.W., Nguyen, D.D., Liu, Y., Shan, X., Nghiem, L.D. and Nguyen, L.N. 2020a. Removal process of antibiotics during anaerobic treatment of swine wastewater. Bioresour. Technol. 300, 122707.

8. Cheng, D., Ngo, H.H., Guo, W., Lee, D., Nghiem, D.L., Zhang, J., Liang, S., Varjani, S. and Wang, J., 2020b. Performance of microbial fuel cell for treating swine wastewater containing sulfonamide antibiotics. Bioresour. Technol. p.123588.

9. Delgadillo-Mirquez, L., Lardon, L., Steyer, J.-P. and Patureau, D. 2011. A new dynamic model for bioavailability and cometabolism of micropollutants during anaerobic digestion. Water Res. 45 (15), 4511-4521.

10. Federation, W.E. and Association, A.P.H. 2005. Standard methods for the examination of water and wastewater. American Public Health Association (APHA): Washington, DC, USA.

11. Freguia, S., Rabaey, K., Yuan, Z. and Keller, J. 2008. Sequential anode–cathode configuration improves cathodic oxygen reduction and effluent quality of microbial fuel cells. Water Res. 42 (6-7), 1387-1396.

12. Gil, G.C., Chang, I.S., Kim, B.H., Kim, M., Jang, J.K., Park, H.S. and Kim, H.J. 2003. Operational parameters affecting the performannce of a mediator-less microbial fuel cell. Biosensors Bioelectron. 18 (4), 327-334.

13. Han, Y., Yang, L., Chen, X., Cai, Y., Zhang, X., Qian, M., Chen, X., Zhao, H., Sheng, M. and Cao, G. 2020. Removal of veterinary antibiotics from swine wastewater using anaerobic and aerobic biodegradation. Sci. Total Environ. 709, 136094.

14. Harnisch, F., Gimkiewicz, C., Bogunovic, B., Kreuzig, R. and Schröder, U. 2013. On the removal of sulfonamides using microbial bioelectrochemical systems. Electrochem. Commun. 26, 77-80.

15. Hassan, M., Zhu, G., LU, Y.Z., AL-Falahi, A.H., LU, Y., Huang, S. and Wan, Z., 2020. Removal of antibiotics from wastewater and its problematic effects on microbial communities by bioelectrochemical Technology: Current knowledge and future perspectives. Environ. Eng. Res., 26 (1), p.190405.

16. He, Z., Kan, J., Wang, Y., Huang, Y., Mansfeld, F. and Nealson, K.H. 2009. Electricity Production Coupled to Ammonium in a Microbial Fuel Cell. Environ. Sci. Technol. 43 (9), 3391-3397.

17. Helbling, D.E., Johnson, D.R., Honti, M. and Fenner, K. 2012. Micropollutant biotransformation kinetics associate with WWTP process parameters and microbial community characteristics. Environ. Sci. Technol. 46 (19), 10579-10588.

18. Hiegemann, H., Lubken, M., Schulte, P., Schmelz, K.G., Gredigk-Hoffmann, S. and Wichern, M. 2018. Inhibition of microbial fuel cell operation for municipal wastewater treatment by impact loads of free ammonia in bench- and 45 L-scale. Sci. Total Environ. 624, 34-39.

19. Huang, H., Xiao, D., Liu, J., Hou, L. and Ding, L., 2015. Recovery and removal of nutrients from swine wastewater by using a novel integrated reactor for struvite decomposition and recycling. Sci. Rep. 5, p.10183.

20. Jin, E., Cao, L., Xiang, S., Zhou, W., Ruan, R. and Liu, Y., 2020. Feasibility of using pretreated swine wastewater for production of water spinach (Ipomoea aquatic Forsk.) in a hydroponic system. Agric. Water Manag. 228, p.105856.

21. Kim, B.H., Chang, I.S. and Gadd, G.M., 2007. Challenges in microbial fuel cell development and operation. Appl. Microbiol. Biotechnol. 76 (3), 485-494.

22. Kim, D., Min, K.J., Lee, K., Yu, M.S. and Park, K.Y. 2017. Effects of pH, molar ratios and pre-treatment on phosphorus recovery through struvite crystallization from effluent of anaerobically digested swine wastewater. Environ. Eng. Res. 22 (1), 12-18.

23. Kim, H.W., Nam, J.Y. and Shin, H.S. 2011. Ammonia inhibition and microbial adaptation in continuous single-chamber microbial fuel cells. J. Power Sources 196 (15), 6210-6213.

24. Kim, J.R., Zuo, Y., Regan, J.M. and Logan, B.E. 2008. Analysis of ammonia loss

mechanisms in microbial fuel cells treating animal wastewater. Biotechnol. Bioeng. 99 (5), 1120-1127.

25. Kim, T., An, J., Lee, H., Jang, J.K. and Chang, I.S. 2016. pH-dependent ammonia removal pathways in microbial fuel cell system. Bioresour. Technol. 215, 290-295.

26. Kocaturk, I. and Erguder, T.H. 2016. Influent COD/TAN ratio affects the carbon and nitrogen removal efficiency and stability of aerobic granules. Ecol. Eng. 90, 12-24.

27. Kumwimba, M.N. and Meng, F. 2019. Roles of ammonia-oxidizing bacteria in improving metabolism and cometabolism of trace organic chemicals in biological wastewater treatment processes: A review. Sci. Total Environ. 659, 419-441.

28. Li, J.Z., Meng, J., Li, J.L., Wang, C., Deng, K.W., Sun, K. and Buelna, G. 2016. The effect and biological mechanism of COD/TN ratio on nitrogen removal in a novel upflow microaerobic sludge reactor treating manure-free piggery wastewater. Bioresour. Technol. 209, 360-368.

29. Liu, S.X., Li, L., Li, H.Q., Wang, H. and Yang, P. 2017. Study on ammonium and organics removal combined with electricity generation in a continuous flow microbial fuel cell. Bioresour. Technol. 243, 1087-1096.

30. Lou, Y., Ye, Z.-L., Chen, S., Ye, X., Deng, Y. and Zhang, J. 2018. Sorption behavior of tetracyclines on suspended organic matters originating from swine wastewater. J. Environ. Sci. 65, 144-152.

31. Min, B., Kim, J.R., Oh, S.E., Regan, J.M. and Logan, B.E. 2005. Electricity generation from swine wastewater using microbial fuel cells. Water Res. 39 (20), 4961-4968.

32. Miran, W., Jang, J., Nawaz, M., Shahzad, A. and Lee, D.S. 2018. Biodegradation of the sulfonamide antibiotic sulfamethoxazole by sulfamethoxazole acclimatized cultures in microbial fuel cells. Sci. Total Environ. 627, 1058-1065.

33. Müller, E., Schüssler, W., Horn, H. and Lemmer, H. 2013. Aerobic biodegradation

of the sulfonamide antibiotic sulfamethoxazole by activated sludge applied as co-substrate and sole carbon and nitrogen source. Chemosphere 92 (8), 969-978.

34. Najafgholi, Z., Rahimnejad, M. and Najafpour, G. 2015. Effect of Electrolyte Conductivity and Aeration on Performance of Sediment Microbial Fuel Cell. J. Renew. Energy Environ. 2(1), 43-48.

35. Oh, S.E., Kim, J.R., Joo, J.H. and Logan, B.E., 2009. Effects of applied voltages and dissolved oxygen on sustained power generation by microbial fuel cells. Water Sci. Technol. 60 (5), 1311-1317.

36. Roh, H., Subramanya, N., Zhao, F., Yu, C.-P., Sandt, J. and Chu, K.-H. 2009. Biodegradation potential of wastewater micropollutants by ammonia-oxidizing bacteria. Chemosphere 77 (8), 1084-1089.

37. Roser, H.R.a.M. 2017 Meat and Seafood Production & Consumption, Our World in Data, https://ourworldindata.org/meat-and-seafood-production-consumption.

38. Song, H.L., Li, H., Zhang, S., Yang, Y.L., Zhang, L.M., Xu, H. and Yang, X.L. 2018. Fate of sulfadiazine and its corresponding resistance genes in up-flow microbial fuel cell coupled constructed wetlands: Effects of circuit operation mode and hydraulic retention time. Chem. Eng. J. 350, 920-929.

39. Sun, J., Xu, W., Yang, P., Li, N., Yuan, Y., Zhang, H., Wang, Y., Ning, X., Zhang, Y. and Chang, K. 2019. Enhanced oxytetracycline removal coupling with increased power generation using a self-sustained photo-bioelectrochemical fuel cell. Chemosphere 221, 21-29.

40. Suzuki, K., Tanaka, Y., Kuroda, K., Hanajima, D., Fukumoto, Y., Yasuda, T. and Waki, M. 2007. Removal and recovery of phosphorous from swine wastewater by demonstration crystallization reactor and struvite accumulation device. Bioresour. Technol. 98 (8), 1573-1578.

41. Tang, R., Yuan, S., Chen, F., Zhan, X., Wang, W. and Hu, Z., 2019. Effects of roxarsone and sulfadiazine on biogas production and their degradation during anaerobic digestion. Int. Biodeterior. 140, 113-118.

42. Tao, Q.Q., Luo, J.J., Zhou, J., Zhou, S.Q., Liu, G.L. and Zhang, R.D. 2014. Effect of dissolved oxygen on nitrogen and phosphorus removal and electricity production in microbial fuel cell. Bioresour. Technol. 164, 402-407.

43. Tice, R.C. and Kim, Y. 2014. Influence of substrate concentration and feed frequency on ammonia inhibition in microbial fuel cells. J. Power Sources 271, 360-365.

44. Tran, N.H., Chen, H., Reinhard, M., Mao, F. and Gin, K.Y.-H. 2016. Occurrence and removal of multiple classes of antibiotics and antimicrobial agents in biological wastewater treatment processes. Water Res. 104, 461-472.

45. Tran, N.H., Reinhard, M. and Gin, K.Y.H. 2018. Occurrence and fate of emerging contaminants in municipal wastewater treatment plants from different geographical regions-a review. Water Res. 133, 182-207.

46. Wang, J. and Wang, S. 2018a. Microbial degradation of sulfamethoxazole in the environment. Appl. Microbiol. Biotechnol. 102 (8), 3573-3582.

47. Wang, L., Liu, Y., Ma, J. and Zhao, F. 2016. Rapid degradation of sulphamethoxazole and the further transformation of 3-amino-5-methylisoxazole in a microbial fuel cell. Water Res. 88, 322-328.

48. Wang, L., You, L., Zhang, J., Yang, T., Zhang, W., Zhang, Z., Liu, P., Wu, S., Zhao, F. and Ma, J. 2018. Biodegradation of sulfadiazine in microbial fuel cells: reaction mechanism, biotoxicity removal and the correlation with reactor microbes. J. Hazard. Mater. 360, 402-411.

49. Wang, S. and Wang, J. 2018b. Biodegradation and metabolic pathway of sulfamethoxazole by a novel strain Acinetobacter sp. Appl. Microbiol. Biotechnol. 102 (1),

425-432.

50. Wen, Q., Wu, Y., Wang, G.L., Zhao, L.X., Sun, Q. and Kong, F.Y. 2010. Microbial Fuel Cell Using Sequential Anode-cathode to Treat Brewery Wastewater. Chem. J. Chinese U. 6.

51. Wu, D., Sun, F., Chua, F.J.D. and Zhou, Y. 2020. Enhanced power generation in microbial fuel cell by an agonist of electroactive biofilm–Sulfamethoxazole. Chem. Eng. J. 384, 123238.

52. Wu, Q., Jiao, S., Ma, M. and Peng, S., 2020. Microbial fuel cell system: a promising technology for pollutant removal and environmental remediation. Environ. Sci. Pollut. Res. 1-16.

53. Xu, Y., Yuan, Z. and Ni, B.-J. 2016. Biotransformation of pharmaceuticals by ammonia oxidizing bacteria in wastewater treatment processes. Sci. Total Environ. 566, 796-805.

54. Xue, W., Li, F. and Zhou, Q. 2019. Degradation mechanisms of sulfamethoxazole and its induction of bacterial community changes and antibiotic resistance genes in a microbial fuel cell. Bioresour. Technol. 289, 121632.

55. Yadu, A., Sahariah, B.P. and Anandkumar, J. 2018. Influence of COD/ammonia ratio on simultaneous removal of NH4+-N and COD in surface water using moving bed batch reactor. J. Water Process Eng. 22, 66-72.

56. Yang, F., Han, B., Gu, Y. and Zhang, K., 2020. Swine liquid manure: a hotspot of mobile genetic elements and antibiotic resistance genes. Sci. Rep. 10, 1-10.

57. Ye, L. and Zhang, T. 2011. Ammonia-oxidizing bacteria dominates over ammonia-oxidizing archaea in a saline nitrification reactor under low DO and high nitrogen loading. Biotechnol. Bioeng. 108 (11), 2544-2552.

58. Ye, Y., Ngo, H.H., Guo, W., Liu, Y., Chang, S.W., Nguyen, D.D., Ren, J., Liu, Y. and

Zhang, X. 2019a. Feasibility study on a double chamber microbial fuel cell for nutrient recovery from municipal wastewater. Chem. Eng. J. 358, 236-242.

59. Ye, Y.Y., Ngo, H.H., Guo, W.S., Chang, S.W., Nguyen, D.D., Liu, Y.W., Ni, B.J. and Zhang, X.B. 2019b. Microbial fuel cell for nutrient recovery and electricity generation from municipal wastewater under different ammonium concentrations. Bioresour. Technol. 292, p.121992.

60. Yenigun, O. and Demirel, B. 2013. Ammonia inhibition in anaerobic digestion: A review. Process Biochem. 48 (5-6), 901-911.

61. Zeng, Z., Zhang, M., Kang, D., Li, Y., Yu, T., Li, W., Xu, D., Zhang, W., Shan, S. and Zheng, P., 2019. Enhanced anaerobic treatment of swine wastewater with exogenous granular sludge: Performance and mechanism. Sci. Total Environ. 697, p.134180.

62. Zhang, Q.Q., Ying, G.G., Pan, C.G., Liu, Y.S. and Zhao, J.L. 2015. Comprehensive evaluation of antibiotics emission and fate in the river basins of China: source analysis, multimedia modeling, and linkage to bacterial resistance. Environ. Sci. Technol. 49 (11), 6772-6782.

63. Zhi, S., Zhou, J., Liu, H., Wu, H., Zhang, Z., Ding, Y. and Zhang, K., 2020. Simultaneous extraction and determination of 45 veterinary antibiotics in swine manure by liquid chromatography-tandem mass spectrometry. J. Chromatogr. B 1154, p.122286.

64. Zhang, X., He, W., Ren, L., Stager, J., Evans, P.J. and Logan, B.E., 2015. COD removal characteristics in air-cathode microbial fuel cells. Bioresour. Technol. 176, 23-31.

65. Zhou, X., Qu, Y., Kim, B.H., Du, Y., Wang, H., Li, H., Dong, Y., He, W., Liu, J. and Feng, Y. 2015. Simultaneous current generation and ammonia recovery from real urine using nitrogen-purged bioelectrochemical systems. RSC Advances 5 (86), 70371-70378.

66. Zhou, Y., Zhu, N., Guo, W., Wang, Y., Huang, X., Wu, P., Dang, Z., Zhang, X. and Xian, J. 2018. Simultaneous electricity production and antibiotics removal by microbial fuel

cells. J. Environ. Manage. 217, 565-572.