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Impacts of hydraulic retention time on a continuous flow mode dual-chamber microbial fuel cell for recovering nutrients from municipal wastewater

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Abstract

Nutrients recovery has become a meaningful solution to address shortage in the fertilizer production which is the key issue of nations' food security. The concept of municipal wastewater is based on its ability to be a major potential source for recovered nutrients because of its vast quantity and nutrient-rich base. Microbial fuel cell (MFC) has emerged as a sustainable technology, which is able to recover nutrients and simultaneously generate electricity. In this study a two-chambered MFC was constructed, and operated in a continuous flow mode employing artificial municipal wastewater as a substrate. The effects of hydraulic retention time (HRT) on the recovery of nutrients by MFC were studied. The COD removal rates were insignificantly influenced by varying HRT from 0.35 to 0.69 d, that were over 92%. Furthermore, the recovery rate of nutrients was insignificantly affected while increasing the HRT, which fluctuates from 80% to 90%. In contrast, the maximum power generation declined when HRT increased and the lowest one was 510.3 mV at the HRT of 0.35 d. These results demonstrate that the lab-scale double chamber MFC using municipal wastewater as the substrate can provide a highly effective removal strategy for organic matter, nutrients recovery and electricity output when operating at a specific HRT.

Keywords: Microbial fuel cell; Electricity generation; Municipal wastewater; Nutrient recovery; Hydraulic retention time.

1. Introduction

Nitrogen (N) and phosphorus (P) are un-substitutable micronutrients for the growth of all living organisms (Guo et al., 2020; Wang et al., 2020). Such nutrients are commonly employed as fertilizers for crops and plants in agriculture (Hunter & Deshusses, 2020). However, the supply of N and P is recently becoming more challenging, especially given the projected increases in food production to satisfy the growing global population and the relevant global fertilizer production, which is required to reach approximately 263 million tons in 2050 (Alexandratos & Bruinsma, 2012). In industry, the Haber Bosch process utilizes the N_2 gas from the atmosphere to produce ammonia for fertilizer production under high pressure and temperature (Khan et al., 2020; Ye et al., 2018). Obviously, this generation process demands a large input of energy and costs; this results in the emission of toxic gases into the environment. On the other hand, Cordell et al. (2011) reported that nonrenewable phosphate-based rocks serving as the main source of P can only serve the industrial demand in the coming 50–100 years.

Apart from this, applications of nutrients (i.e., N and P) in a full-scale context may eventually cause their accumulation in streams, and a high content of nutrients in the aquatic environment may trigger environmental damage such as eutrophication (Zhou et al., 2020). Besides, human activity has made possible the mobilization of phosphate from phosphate-bearing rocks to the hydrosphere over the last 50 years in the global P cycle, and this amount was approximately 500 million tons of P (Cordell et al., 2011). Thus, it is critically important to combine the removal of nutrients with their recovery in waste treatment, which: 1) reduces the need for fertilizer production; 2) curtails the impact of fertilizer production and transportation on climate change; and 3) contributes to a more healthier ecology, and sustainable development (Ajmal et al., 2020; Luo et al., 2018; Sobhi et al., 2019; Ye et al., 2017).

Wastewater management has received increased attention for its efficiency and sustainability in recent years (Walker et al., 2014). It is generally accepted that retrieving nutrients (mainly N and P) from wastewater for use in agriculture could contribute to sustainable and better wastewater management (Hermassi et al., 2017; Yan et al., 2018; Ye et al., 2018). Due to containing rich ammonium and phosphate ions, such a recovery strategy in the treatment system of municipal wastewater is attractive (Kataki et al., 2016; Li et al., 2020). Reportedly, the recovery of phosphate in the sewage treatment process could theoretically supplement 15–20% of the worldwide phosphate demand (Xia et al., 2016).

In the domestic wastewater treatment, chemical precipitation and adsorption are considered as the most common methods for recovering nutrients (Ye et al., 2017). However, the nutrients recovery through adsorption needs additional desorption process, thus increasing the operational complexity and overall costs. This is despite the fact that the adsorption process for nutrients recovery is simple to design and operate. Therefore, chemical precipitation is currently acted as a promising method for recovering nutrients and the common final products in this process include hydroxyapatite and struvite, which could be applied in agriculture as a slow-release fertilizer (Yan et al., 2018; Ye et al., 2017). It should be noted here that struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) precipitation could simultaneously recycle ammonium and phosphate. For this reason, the nutrient recovery through chemical precipitation in domestic wastewater treatment has been widely investigated, in order to evaluate its feasibility from laboratory-scale to full-scale (De Vrieze et al., 2016; Ye et al., 2017). To increase the economic feasibility of the nutrients recovery system, some membrane processes such as forward osmosis, membrane distillation and electrodialysis have been integrated with chemical precipitation to realize this objective (Yan et al., 2018; Ye et al., 2018). This combination can enhance the enrichment of nutrients within the reactor, but the membrane fouling is still a big challenge. Apart from this, the large-scale industrial

application for recovering nutrients by chemical precipitation is challenged by greatly additional chemicals that are associated with high costs (Romero-Güiza et al., 2015a; Romero-Güiza et al., 2015b). In particular, the addition of alkaline reagents for increasing the pH solution is essential in the chemical precipitation process, which contributes a great deal to the overall costs in the process, and greatly influences the economic feasibility of the nutrient recovery system during wastewater treatment (Melia et al., 2017; Wang et al., 2019). Considering this, it is challenging for implementing the proposed objective of nutrient recovery in current conventional systems.

Using microbial fuel cell (MFC) system for recovering nutrients from wastewater has attracted attention of researchers (Kelly & He, 2014; Pepè Sciarria et al., 2019; Ye et al., 2019b) as this method could achieve the pH elevation without the addition of alkaline chemicals. Thus, it could greatly enhance the economic feasibility of the chemical nutrient recovery system. Initially, the MFC technology was used to transfer the energy stored in the organics into the electrical energy in wastewater treatment. In this scenario, the bio-catalytic capabilities of microbes are utilized to generate a current field (Do et al., 2020; Xu et al., 2018a), which applies to a range of organic fuel sources in the absence of metal catalysts. In a typical double-chamber MFC, anaerobic bacteria at the anode chamber are employed to oxidize biodegradable substrates, and electrons and protons are thus generated through the anaerobic respiration process. After that, electrons generated transfer to the surface of anode electrode and then to the cathode electrode through the external circuit. Furthermore, the generated protons transport from the anode compartment to the cathode compartment through a cation exchange membrane (CEM), after which they react with an electron acceptor (e.g., oxygen) and electrons to produce water (or other reduced compounds) (Hou et al., 2019; Oliveira et al., 2013; Palanisamy et al., 2019). During the cathode reaction, hydroxyl ions localized the cathode electrodes being generated, so the high pH zone of the cathode offers an

opportunity for applying chemical precipitation to the recovery of nutrients. Moreover, the energy balance of the nutrient recovery system is improved because of electricity generated in the MFC reactor. Also, the membrane fouling in the MFC reactor could be reduced since the sludge on the membrane surface is electrostatically repulsed by the current field that is formed by the reactor (Wang et al., 2013). Both of this indicates the sustainability of MFC system for recovering nutrients from sewage.

In general, the MFC performance is governed by many factors such as configurations, electrode materials, inoculum, operation conditions and oxygen supply (Mardanpour et al., 2017; Xu et al., 2018b). At the beginning of these MFC studies, the reactors always operate in batch mode, but have now been changed to the continuous mode. The possible reason for this is that the MFC systems are expected to simultaneously generate electricity and purify wastewater, and continuous mode facilitates the further scaling up of MFC reactors.

Hydraulic retention time (HRT) plays an important part in designing and operating MFC reactors, and greatly influences the energy requirements, which in turn directly affects the MFC's performance. It is reported that HRT exerts an important impact on the power output of continuous-flow MFCs because the variations in HRT have a direct effect on the type and quantity of bacteria in the bioreactor (Sharma & Li, 2010; Sobieszuk et al., 2017). Sharma and Li (2010) believed that the optimum HRT is the time that is consistent with the time taken to generate living microbes. Apart from this, the HRT determines the value of shear stress which directly affects the formation of biofilm on a surface (Lecuyer et al., 2011). Thus, the value of HRT should be correctly determined when utilizing MFC systems (Sobieszuk et al., 2017). However, there are few studies with reference to the HRT effects on recovering nutrients via MFC systems. In addition, our group has developed a new double-compartment MFC which successfully retrieved nutrients from municipal wastewater (Ye et al., 2019b). This work mainly focused on the investigation of the role of HRT with the aspects of the

efficiency of power output and recovery of nutrients while applying the developed new dual compartment MFC reactor to recover nutrients in the urban wastewater treatment.

2. Materials and methods

2.1 MFC preparation

The anaerobic sludge used as inoculum in the anode compartment of the MFC reactor was provided by an Australian wastewater treatment plant. The sludge was firstly cultivated in a sequencing batch MFC under room temperature (22 ± 2 °C), during which the synthetic municipal wastewater – it had a fixed composition (see Table 1) - was fed to the anode compartment. This process aimed to make the anaerobic microbes adapt to the new environment, where the MFC reactor operated until the voltage stabilized. After that, the double-chamber MFC system was continuously run and fed with solution (i.e., synthetic municipal wastewater). The pH of feed solution was regulated to be approximately 7 through the addition of HCl and NaCl solutions whilst the influent COD concentrations were measured, which depend on the initial concentration of glucose. Apart from this, the catholyte initially contained the distilled (DI) water in the cathode chamber.

Table 1. Composition of artificial municipal wastewater

Parameter	Average \pm Std.	Parameter	Average \pm Std.
pH	7.00 ± 0.02	COD	300 ± 15 mg/L
NH ₄ Cl	20 ± 1.0 mg/L	KH ₂ PO ₄	4.6 ± 0.5 mg/L
MgSO ₄ ·7H ₂ O	5.4 ± 0.5 mg/L	CaCl ₂ ·2H ₂ O	0.4 ± 0.01 mg/L
Yeast	32 ± 1.0 mg/L	Trace nutrients	0.61 mL

2.2 MFC design and operation

The design of the double-chamber MFC applied in this work was on the basis of the information in a recently published study (Ye et al., 2019a). The MFC reactor consisted of two square chambers which were made by plexiglass plates with the thickness being at 5 cm. The two chambers with an active volume of 350 mL were separated by a CEM membrane (CMI7000, Membranes International Inc., USA). Furthermore the anode electrode was made of cylinder-shaped graphite felt (6mm thickness, 30 mm in diameter) whereas the cathode electrode was a carbon-fiber brush (30 mm long, 30 mm in diameter), in which both electrodes were provided by Sanye Carbon Co. Ltd. in China. An external resistor (1000 Ω) was employed to complete the electric circuit, which was connected to the electrodes via the copper wire. Besides, the hydraulic connection of these two chambers made the anode effluent be used as the cathode influent. A peristaltic pump (Model 77202-60, Masterflex) was utilized to transfer the feed solution to the anode chamber. To achieve different HRTs, the flow control of this pump allowed for different flow rates to occur as required. Notably, internal pH control was not needed throughout these experiments.

The dual-chamber MFC operated at four different times for a period of 120 days (see Table 2). In the present experiments, the volumetric flow rate was changed by the pump (0.35, 0.47, 0.59 and 0.70 mL/min), resulting in different HRTs (0.69, 0.52, 0.41 and 0.35 d). When operating the MFC, electricity generation is the most important parameter for the reactor. The MFC's working condition would be considered as steady when the value of voltage generated remains constant or changes in a negligible range. In each experimental period, the MFC reactor continuously operated until reaching the steady state at the new HRT.

Table 2. Operational parameters of the double-compartment MFC

Parameters	Experimental period			
	i	ii	iii	iv
Days	1-30	31-60	61-90	91-120
Flow rate (mL/min)	0.35	0.47	0.59	0.70
HRT (d)	0.69	0.52	0.41	0.35
Influent COD concentration (mg/L)	300	300	300	300
OLR (mgCOD/L·d)	435	580	725	870

2.3 Calculations

In the present study, equation (1) was exploited to calculate the power density (P_A , mV/m²).

$$P_A = \frac{U^2}{RA} \quad (1)$$

where R (Ω) is the resistance at 1000; U (mV) is the voltage generated by the MFC reactor; A (m²) is the anode electrode's surface area in both sides.

Apart from this, coulombic efficiency (CE, %) presents electrons' recovery in the MFC reactor and be achieved by the following equation (Logan, 2008).

$$C_E = \frac{8 \int_0^t I dt}{F \Delta COD V} \times 100\% \quad (2)$$

where I (mA) is the current across the resistor in the MFC reactor at operation time (t , d); V (mL) is the anode chamber's active volume; ΔCOD (mg/L) is achieved through the subtraction of influent COD concentration to effluent COD concentration; F (96485 C/mol) is the Faraday's constant.

The recovery efficiency of nutrients was calculated by the following equation.

$$R_E = \frac{C_{anode-effluent} - C_{cathode-effluent}}{C_{anode-effluent}} \times 100\% \quad (3)$$

where $C_{anode-effluent}$ represents the concentration of NH_4^+ -N or PO_4^{3-} -P in the anode effluent; and $C_{cathode-effluent}$ represents the concentration of NH_4^+ -N or PO_4^{3-} -P in the cathode effluent.

2.4 Analysis

The effluents from the anode chamber and cathode chamber were daily analyzed for the measurement of the pH solution and concentration of COD and nutrients to determine the MFC's performance. Prior to analysis, all the liquid samples were firstly centrifuged and then filtered by 0.20 μm -filters (Merck Millipore, Burlington, USA). The concentrations of COD and nutrients were measured by different test kits provided by Hanna Instruments Australia and Merck Millipore, respectively. For monitoring the solution pH, a pH meter (HI9025, Hanna Instruments) was utilized. Regarding electricity generation, a universal digital meter purchased from Shenzhen City Station Win Technology Co. Ltd. was employed to monitor the changes in the voltage generated across the resistor in the MFC.

3. Results and discussion

3.1 Electricity generation

The impacts of HRT on the power output in the MFC reactor were evaluated, and Figure 1 shows the variations of voltage generated by the MFC reactor during the operation period. In the anode chamber, the electrically active bacteria (EBA) are responsible for the power output, and their activity and concentration directly affects the energy production. From this figure, at the beginning of the first period (HRT = 0.69 d), the generated voltage increased rapidly, probably due to the change in operation mode from batch to continuous. Subsequently, the voltage became stabilized within nine days and the maximum voltage generated in this period was 598.9 mV. When the HRT was 0.52 d (period II), the maximum voltage fell to 577.6 mV. A further reduction in the HRT resulted in reducing the maximum generated voltage to 564.5 mV at the HRT of 0.41 d (period III). As the HRT declined to 0.35 d (period V), the maximum voltage was approximately 10% lower than the value obtained at

the HRT of 0.41 d.

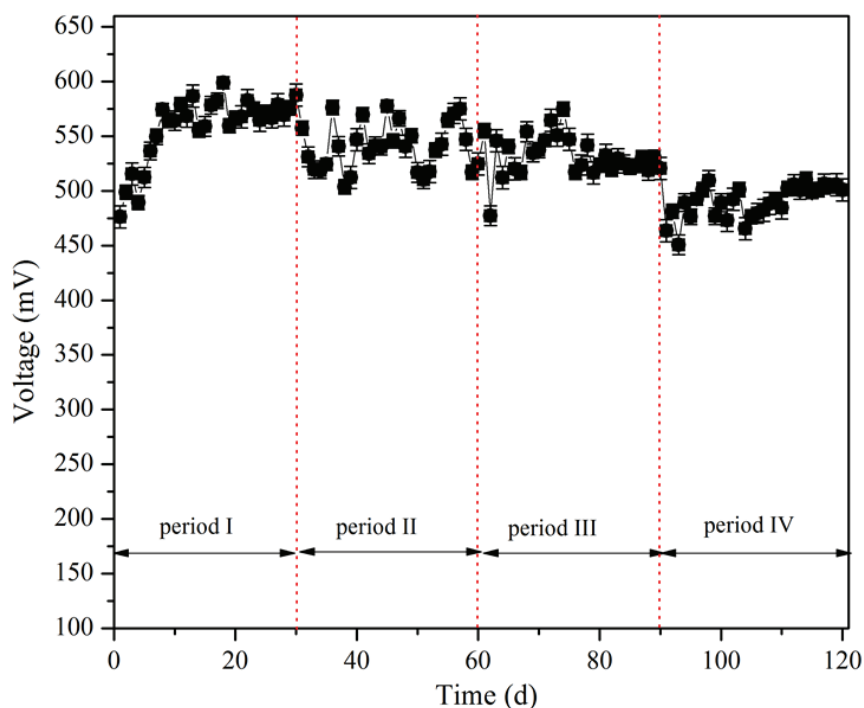


Figure 1. Power output vs operation time at different HRTs (0.35-0.69 d) in the double-compartment MFC.

Overall, the electricity generation of the MFC reactor revealed a downturn in HRT during the experiments. A possible reason for this is that decreasing HRT may result in: firstly, high flow rates of feed solution which may worsen the mixing conditions (Sobieszuk et al., 2017); and secondly, the loss of anaerobic microbes to some extent in the anode chamber. These two outcomes may greatly influence the concentration of EBA and seriously compromise the growth of the EBA. Moreover, higher HRT enhanced the residence time of the substrate within the MFC reactor and thereby facilitated the degradation of substrates via EBA. As a result of this, more electrons would be generated and transferred to the surface of the anode electrode (Sharma & Li, 2010). Consequently, the power output could be improved. Santos et al. (2017) found that the EBA's activity could be enhanced by increasing HRT, which contributes to the EBA's degradation and transforms the substrate into electricity.

Another possible explanation for this is that lower HRT may result in an increase in the OLR correspondingly, and sufficient organics were thus retained in the anode chamber (Ma et al., 2016). In this scenario, the organic biodegradation was enhanced as well as the formation of biofilm attached to the anode electrode. Consequently, the thickness of the biofilm on the anode surface was increased as well as the mass transfer resistance, which inhibited the generation of power. In their analysis, Wei et al. (2012) indicated a dynamic equilibrium existing in the anode chamber, which demonstrated a constant proliferation rate of EBA. For this reason, the voltage would generally become stabilized.

The present results have also been observed in other studies (Li et al., 2013; Rahimnejad et al., 2011; Srikanth et al., 2016). In contrast, a few MFC analyses reported an opposite conclusion in that increasing HRT may adversely influence the voltage generation in MFCs (Akman et al., 2013; Ge et al., 2013; Juang et al., 2012; Wei et al., 2012). This contradiction may be attributed to the MFC configuration and other factors which influence the power output in the MFC reactors. For example, Ge et al. (2013) explored the impacts of HRT on the power output in an osmotic microbial fuel cell fed with municipal wastewater. In their study, decreasing the HRT from 24 h to 6 h caused electricity generation to increase significantly. Apart from this, some researchers thought high HRT may reduce the concentrations of substrate and the cell metabolism at the anode chamber (Liu et al., 2008), which inhibited the MFC reactor's production of energy. A reduced HRT results in more degradation of the substrate used for generating electricity at a unit time in the MFC system, thereby enabling biomass to convert energy (Luo et al., 2017). However, this scenario may require a higher concentration of EBA or stronger biofilm, which could oxidize more organics in a shorter time.

In addition, it is possible that there is an optimum HRT for the electricity generation of each continuously operated MFC reactor, but these optimum values vary from MFC

configurations to another (Rahimnejad et al., 2011). Apart from this, the maximum power output (598.9 mV) in the present study was observed at the HRT of 0.69 d, which was also optimal for this MFC configuration. Akman et al. (2013) reported that it was about 40% higher with reference to the maximum voltage generated at a HRT of 0.5 d. By contrast, this value was around 30% lower than that achieved at the HRT of 0.28 d by Juang et al. (2012) who analyzed a continuous MFC. This may be attributed to the use of neutral red in the anode chamber, which was served as an electron transfer mediator and thereby improved electricity generation.

For generating electricity in the MFC system, how to increase the output power is hugely important. For this reason, the power density is a critical constant in MFC reactors as well as coulombic efficiency. Figure 2 depicts the maximum voltage generated and corresponding power density at each HRT. The maximum coulombic efficiency and power density were 25.01% and 253.84 mW/m², respectively, at the HRT of 0.69 d. In this work, optimizing HRT to generate electricity in the dual chambered continuous MFC was one of the main objectives. For this reason, the HRT of 0.69 d could satisfy this purpose. However, the HRT is expected to be minimized when the effective removal of COD is the main objective of MFCs for wastewater treatment because this could increase the economic feasibility of the MFC system (Chang et al., 2004).

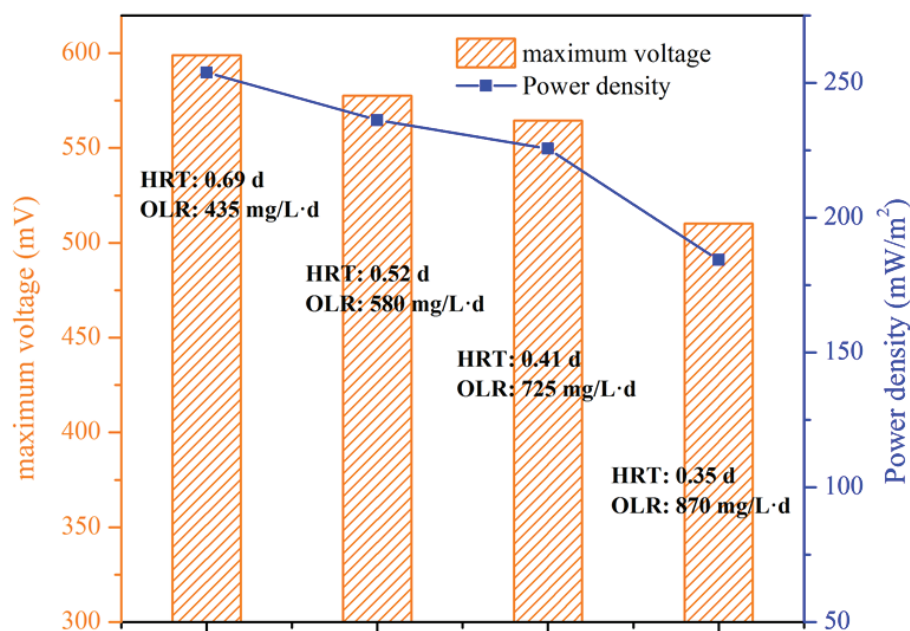


Figure 2. Maximum voltage generated and corresponding power density at different HRTs (0.35-0.69 d) in the double-chamber MFC.

Apart from this, the present study's maximum power density achieved was greater compared to the values reported by other studies. For example, Song et al. (2018) operated an up-flow MFC and achieved the maximum power density of 15.41 mW/m^2 at the HRT of 3.0 d. Recently, Yang et al. (2019) found that the modification of the cathode surfaces could accelerate the electron consumption, and thereby improve the power density of a MFC. In their study, the MFC with the mesoporous carbon-modified cathodes could have higher power density compared to that equipped with bare carbon electrodes, and the maximum power density could reach $1.179 \pm 0.031 \text{ W/m}^2$. The possible explanation for this is that the mesoporous carbon-modified electrode had approximately double the porosity utilization compared to the bare carbon electrode. Similarly, in the study by Luo and He (2016), their results showed that maximum power density of tubular MFCs could rise from 4.29 to 6.50 W/m^3 when the carbon fiber cathode was coated with nickel. It is certain that the power density could be improved by optimizing the MFC through changing the operational

parameters (e.g., influent COD concentration and solution conductivity).

3.2 COD removal

The performance of this double-chamber MFC with reference to wastewater treatment efficiency was evaluated by the COD removal efficiency. The COD reduction was tested while increasing the HRT from 0.35 to 0.69 d. As shown in Figure 3, the double-chamber MFC reactor effectively removed COD in the HRT range of 0.35-69 d. This may well have been due to the lowest HRT (i.e., 0.35 d), which was still sufficient for the microorganism in the anode chamber to efficiently degrade organic material. Similar results were reported elsewhere (Song et al., 2018; Xie et al., 2018). For instance, Song et al. (2018) observed this phenomenon when they utilized an up-flow MFC coupled constructed wetlands for the removal of sulfadiazine. In this scenario, the removal efficiencies of COD were in the range of 92.4%-94.4% under different HRT conditions.

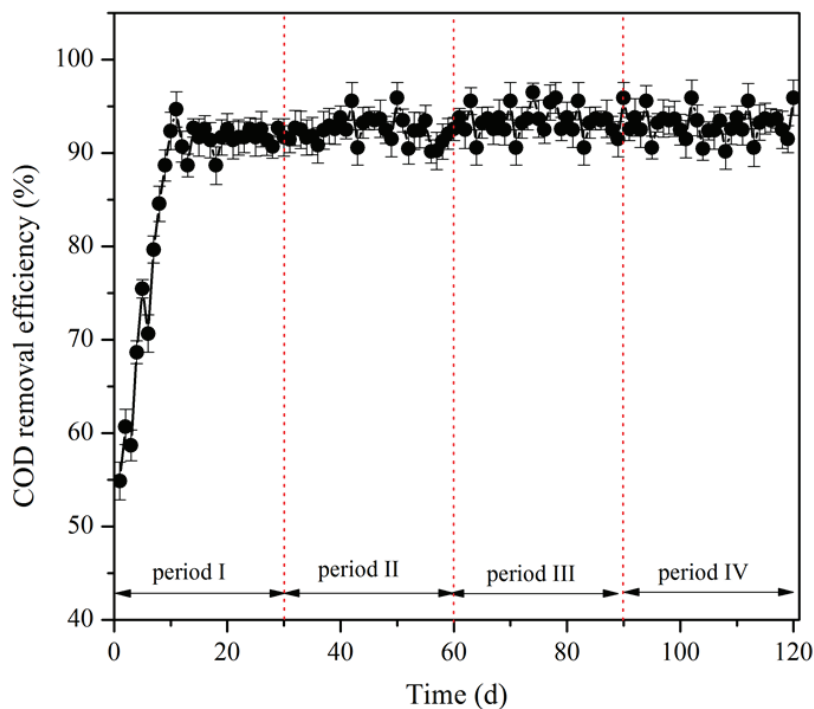


Figure 3. COD reduction efficiencies vs operation time at different HRTs (0.35-0.69 d) in the double-compartment MFC.

The content of all microbial species in the anode chamber of MFC reactors highly determine the removal of COD as well as the balance between the different species of bacteria. Many researchers reported that higher HRT favorably influences the COD degradation (Fazli et al., 2018; Kim et al., 2016; Kim et al., 2015). A commonly stated explanation for this is that higher HRT results in longer residence time of substrate in the anode chamber, and subsequently anaerobic microbes are allowed to have more contact time to digest the organics, which leads to higher removal efficiencies of COD and better effluent quality (Li et al., 2013). Luo et al. (2017) found that longer HRT could even cause full oxidization of organics in domestic wastewater.

In the study by Ge et al. (2013), they found that the removal efficiencies of COD were approximately 40–60% and 70–80% at the HRT of 10 h and 24 h, respectively, whilst continuously operating an osmotic MFC for domestic wastewater treatment. Similarly, Haavisto et al. (2017) employed a two-chamber up-flow MFC to produce electricity from xylose, in which decreasing the HRT from 3.5 d to 0.17 d resulted in reduced removal of COD from 95% to 78%. In contrast, Akman et al. (2013) reported that increasing HRT from 0.5 to 1.5 d may lead to an increase in the removal of dissolved organic carbon from 38% to 45% in the dual-chamber MFC. Similarly, Li et al. (2013) found that the activity of EBA and organic matter degradation was not inhibited in animal carcass wastewater treatment. This difference in findings may result from the MFC's configuration (Li et al., 2013). Moreover, lowering HRT could increase the quantity of substrate entering the anode compartment, so the total amount of substrate degraded by the microbes (i.e., COD removal efficiency) may be increased (Juang et al., 2012).

3.3 Recovery of nutrients

The effects of HRT ranging from 0.35 to 0.69 d on recovering nutrients in the dual-compartment MFC were assessed and the results are shown in Figure 4. At the anode

chamber, the nutrients were mainly removed by microbial activity for their growth. Besides, some ammonium ions may transport from the anode to the cathode compartments through the CEM to realize its removal at the anode compartment.

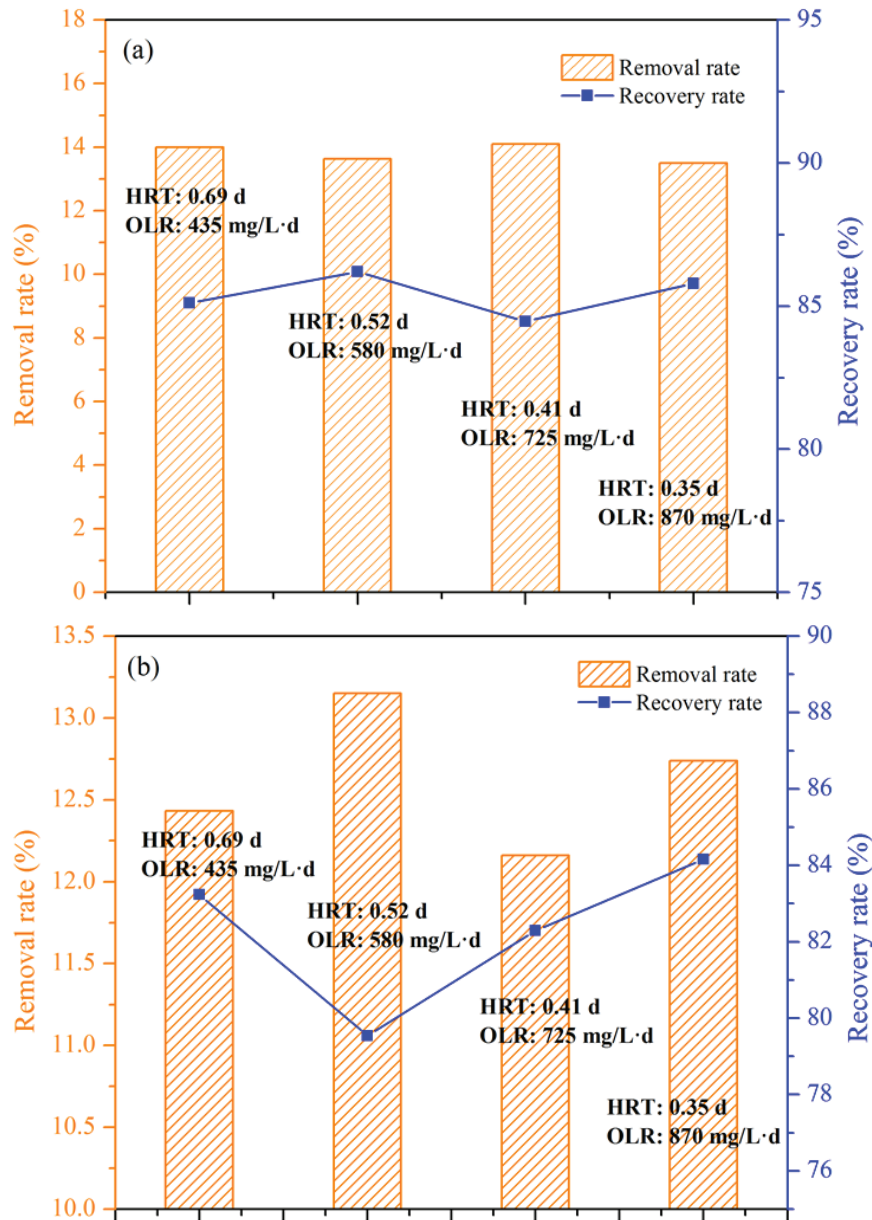


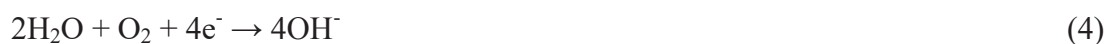
Figure 4. Average removal and recovery efficiency of (a) NH_4^+ -N and (b) PO_4^{3-} -P in the anode chamber and cathode chamber at different HRTs (0.35-0.69 d) in the double-compartment MFC.

Reportedly, lower HRT resulted in higher OLR, which may cause more bio-consumption

of nutrients (Xu et al., 2014; Ye et al., 2019a). However, the analysis of the removal of NH_4^+ -N and PO_4^{3-} -P undertaken in the present study shows that the average removal efficiencies of NH_4^+ -N and PO_4^{3-} -P at the anode chamber were in the 13%-15% and 12%-14% ranges, respectively. The removals also occurred at different HRTs, which indicated that such HRT ranges wielded an insignificant impact on the removal of ammonium and phosphate (see Fig. 4). One possible reason for this is that the enhanced bio-consumption of nutrients at higher OLR resulted from a higher concentration of COD at a unit time whereas changes in the HRT in this work did not vary the influent COD concentration. Again, the lowest HRT could still offer sufficient contact time for the anaerobic bacteria to take up NH_4^+ -N and PO_4^{3-} -P.

Isma et al. (2014) reported that the microbial growth could be improved by decreasing the HRT which leads to a higher F/M ratio, and thus provides more nutrients to the biomass. However, the employment of synthetic domestic wastewater as a substrate in the present study is readily available with sufficient nutrients for the bacterial growth even at low HRT. Apart from this, in the acidic wastewater treatment, Liu et al. (2016) designed a photobioreactor system and documented the significant impacts of HRT on the nutrients removal in this system. This may be attributed to the different functional microorganisms organized in the photobioreactor system and MFC system for the nutrients removal.

On the other hand, at the cathode chamber, the generated electrons and electron acceptor (i.e., O_2) may have the following reaction.



The pH elevation may result in the nutrients recovered through chemical precipitation and the shrinking concentrations of nutrients within the cathode compartment being confirmed. Furthermore, partial NH_4^+ -N ions were removed through air stripping in this scenario, in which the transformation of ammonium into volatile ammonia occurred (Ye et al.,

2019b). The changes in the average recovery efficiency of $\text{NH}_4^+\text{-N}$ and $\text{PO}_4^{3-}\text{-P}$ at the cathode chamber were negligible.

Furthermore, the experimental results indicated that the average recovery efficiencies of nutrients were approximately 85% of $\text{NH}_4^+\text{-N}$ and 83% of $\text{PO}_4^{3-}\text{-P}$ at different HRT conditions. As discussed above, lower HRT could still result in efficient COD removal and effective recovery of nutrients despite the slight inhibition of electricity generation. For this reason, the HRT could be somewhat reduced to enhance the recovery system's economic feasibility, especially if the system's main objective is to recover nutrients and reduce COD concentrations.

4. Conclusion

The impacts of HRT (0.35-0.69d) on the dual-chamber MFC's performance were investigated in this research study. Results demonstrated that the maximum voltage generated was 598.9 mV with a corresponding power density of 253.84 mW/m^2 at HRT = 0.69 d. The effective removal of COD was achieved coupled with the recovery of $\text{NH}_4^+\text{-N}$ (85%) and $\text{PO}_4^{3-}\text{-P}$ (83%) proving successful at different HRT conditions. Overall, the dual-chamber MFC was feasible for producing power and recovering nutrients with simultaneous COD reduction in sewage treatment. This is unique compared to the previous nutrient recovery systems as it provided a high pH zone and essential chemicals such as magnesium ions for recovering nutrients through struvite precipitation while others introduced additional chemicals for the pH increase and chemical precipitation. Nevertheless, the HRT could be reasonably reduced if the MFC's application to the recovery of nutrients is the major priority. Conclusively, the study provides theoretical and technical supports for the commercial application of nutrient recovery via the MFC system from municipal wastewater.

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References

1. Ajmal, Z., Muhmood, A., Dong, R., Wu, S., 2020. Probing the efficiency of magnetically modified biomass-derived biochar for effective phosphate removal. *J. Environ. Manag.* **253**, 109730.
2. Akman, D., Cirik, K., Ozdemir, S., Ozkaya, B., Cinar, O., 2013. Bioelectricity generation in continuously-fed microbial fuel cell: effects of anode electrode material and hydraulic retention time. *Bioresour. Technol.* **149**, 459-464.
3. Alexandratos, N., Bruinsma, J. 2012. World agriculture towards 2030/2050: the 2012 revision. ESA Working paper FAO, Rome.
4. Chang, I.S., Jang, J.K., Gil, G.C., Kim, M., Kim, H.J., Cho, B.W., Kim, B.H., 2004. Continuous determination of biochemical oxygen demand using microbial fuel cell type biosensor. *Biosens. Bioelectron.* **19**(6), 607-613.
5. Cordell, D., Rosemarin, A., Schröder, J.J., Smit, A., 2011. Towards global phosphorus security: A systems framework for phosphorus recovery and reuse options. *Chemosphere* **84**(6), 747-758.
6. De Vrieze, J., Smet, D., Klok, J., Colsen, J., Angenent, L.T., Vlaeminck, S.E., 2016. Thermophilic sludge digestion improves energy balance and nutrient recovery potential in full-scale municipal wastewater treatment plants. *Bioresour. Technol.* **218**, 1237-1245.
7. Do, M.H., Ngo, H.H., Guo, W., Chang, S.W., Nguyen, D.D., Liu, Y., Varjani, S., Kumar, M., 2020. Microbial fuel cell-based biosensor for online monitoring wastewater quality: A critical review. *Sci. Total Environ.* **712**, 135612.
8. Fazli, N., Mutamim, N.S.A., Jafri, N.M.A., Ramli, N.A.M., 2018. Microbial Fuel Cell (MFC) in treating spent caustic wastewater: Varies in Hydraulic Retention Time (HRT) and Mixed Liquor Suspended Solid (MLSS). *J. Environ. Chem. Eng.* **6**(4),

- 4339-4346.
9. Ge, Z., Ping, Q., Xiao, L., He, Z., 2013. Reducing effluent discharge and recovering bioenergy in an osmotic microbial fuel cell treating domestic wastewater. *Desalination* **312**, 52-59.
 10. Guo, X., Cui, X., Li, H., 2020. Effects of fillers combined with biosorbents on nutrient and heavy metal removal from biogas slurry in constructed wetlands. *Sci. Total Environ.* **703**, 134788.
 11. Haavisto, J.M., Kokko, M.E., Lay, C.-H., Puhakka, J.A., 2017. Effect of hydraulic retention time on continuous electricity production from xylose in up-flow microbial fuel cell. *Int. J. Hydrogen Energy* **42**(45), 27494-27501.
 12. Hermassi, M., Valderrama, C., Gibert, O., Moreno, N., Querol, X., Batis, N.H., Cortina, J.L., 2017. Recovery of nutrients (N-P-K) from potassium-rich sludge anaerobic digestion side-streams by integration of a hybrid sorption-membrane ultrafiltration process: Use of powder reactive sorbents as nutrient carriers. *Sci. Total Environ.* **599-600**, 422-430.
 13. Hou, Q., Yang, Z., Chen, S., Pei, H., 2019. Using an anaerobic digestion tank as the anodic chamber of an algae-assisted microbial fuel cell to improve energy production from food waste. *Water Res.*, 115305.
 14. Hunter, B., Deshusses, M.A., 2020. Resources recovery from high-strength human waste anaerobic digestate using simple nitrification and denitrification filters. *Sci. Total Environ.* **712**, 135509.
 15. Isma, M.A., Idris, A., Omar, R., Razreena, A.P., 2014. Effects of SRT and HRT on treatment performance of MBR and membrane fouling. *Int. J. Chem. Mol. Nucl. Mater. Metall. Eng.* **8**, 485-489.
 16. Juang, D., Yang, P., Kuo, T., 2012. Effects of flow rate and chemical oxygen demand

- removal characteristics on power generation performance of microbial fuel cells. *Int. J. Environ. Sci. Technol.* **9**(2), 267-280.
17. Katakai, S., West, H., Clarke, M., Baruah, D.C., 2016. Phosphorus recovery as struvite from farm, municipal and industrial waste: feedstock suitability, methods and pre-treatments. *Waste Manag.* **49**, 437-454.
18. Kelly, P.T., He, Z., 2014. Nutrients removal and recovery in bioelectrochemical systems: a review. *Bioresour. Technol.* **153**, 351-360.
19. Khan, M.S., Koizumi, N., Olds, J.L., 2020. Biofixation of atmospheric nitrogen in the context of world staple crop production: Policy perspectives. *Sci. Total Environ.* **701**, 134945.
20. Kim, K.-Y., Yang, W., Evans, P.J., Logan, B.E., 2016. Continuous treatment of high strength wastewaters using air-cathode microbial fuel cells. *Bioresour. Technol.* **221**, 96-101.
21. Kim, K.-Y., Yang, W., Logan, B.E., 2015. Impact of electrode configurations on retention time and domestic wastewater treatment efficiency using microbial fuel cells. *Water Res.* **80**, 41-46.
22. Lecuyer, S., Rusconi, R., Shen, Y., Forsyth, A., Vlamakis, H., Kolter, R., Stone, H.A., 2011. Shear stress increases the residence time of adhesion of *Pseudomonas aeruginosa*. *Biophys. J.* **100**(2), 341-350.
23. Li, N., Wan, Y., Wang, X., 2020. Nutrient conversion and recovery from wastewater using electroactive bacteria. *Sci. Total Environ.* **706**, 135690.
24. Li, X., Zhu, N., Wang, Y., Li, P., Wu, P., Wu, J., 2013. Animal carcass wastewater treatment and bioelectricity generation in up-flow tubular microbial fuel cells: effects of HRT and non-precious metallic catalyst. *Bioresour. Technol.* **128**, 454-460.
25. Liu, H., Cheng, S., Huang, L., Logan, B.E., 2008. Scale-up of membrane-free

- single-chamber microbial fuel cells. *J. Power Sources* **179**(1), 274-279.
26. Liu, S., Zhang, G., Zhang, J., Li, X., Li, J., 2016. Performance, carotenoids yield and microbial population dynamics in a photobioreactor system treating acidic wastewater: Effect of hydraulic retention time (HRT) and organic loading rate (OLR). *Bioresour. Technol.* **200**, 245-252.
27. Logan, B.E. 2008. *Microbial fuel cells*. John Wiley & Sons.
28. Luo, N., Yan-Mei, G., Xiao-Jia, L., Xie, T., Guo, X., 2017. Effect of Hydraulic Retention Time on MFC-A₂/O Process for Domestic Wastewater Treatment. *DEStech Transactions on Environment, Energy and Earth Sciences*(icesee).
29. Luo, S., He, Z., 2016. Ni-coated carbon fiber as an alternative cathode electrode material to improve cost efficiency of microbial fuel cells. *Electrochim. Acta* **222**, 338-346.
30. Luo, Y., Li, H., Huang, Y.-R., Zhao, T.-L., Yao, Q.-Z., Fu, S.-Q., Zhou, G.-T., 2018. Bacterial mineralization of struvite using MgO as magnesium source and its potential for nutrient recovery. *Chem. Eng. J.* **351**, 195-202.
31. Ma, D., Jiang, Z.-H., Lay, C.-H., Zhou, D., 2016. Electricity generation from swine wastewater in microbial fuel cell: Hydraulic reaction time effect. *Int. J. Hydrogen Energy* **41**(46), 21820-21826.
32. Mardanpour, M.M., Yaghmaei, S., Kalantar, M., 2017. Modeling of microfluidic microbial fuel cells using quantitative bacterial transport parameters. *J. Power Sources* **342**, 1017-1031.
33. Melia, P.M., Cundy, A.B., Sohi, S.P., Hooda, P.S., Busquets, R., 2017. Trends in the recovery of phosphorus in bioavailable forms from wastewater. *Chemosphere* **186**, 381-395.
34. Oliveira, V., Simões, M., Melo, L., Pinto, A., 2013. Overview on the developments of microbial fuel cells. *Biochem. Eng. J.* **73**, 53-64.

35. Palanisamy, G., Jung, H.-Y., Sadhasivam, T., Kurkuri, M.D., Kim, S.C., Roh, S.-H., 2019. A comprehensive review on microbial fuel cell technologies: Processes, utilization, and advanced developments in electrodes and membranes. *J. Clean. Prod.* **221**, 598-621.
36. Pepè Sciarria, T., Vacca, G., Tambone, F., Trombino, L., Adani, F., 2019. Nutrient recovery and energy production from digestate using microbial electrochemical technologies (METs). *J. Clean. Prod.* **208**, 1022-1029.
37. Rahimnejad, M., Ghoreyshi, A.A., Najafpour, G., Jafary, T., 2011. Power generation from organic substrate in batch and continuous flow microbial fuel cell operations. *Appl. Energy* **88**(11), 3999-4004.
38. Romero-Güiza, M., Astals, S., Mata-Alvarez, J., Chimenos, J.M., 2015a. Feasibility of coupling anaerobic digestion and struvite precipitation in the same reactor: evaluation of different magnesium sources. *Chem. Eng. J.* **270**, 542-548.
39. Romero-Güiza, M., Tait, S., Astals, S., del Valle-Zermeño, R., Martínez, M., Mata-Alvarez, J., Chimenos, J., 2015b. Reagent use efficiency with removal of nitrogen from pig slurry via struvite: a study on magnesium oxide and related by-products. *Water Res.* **84**, 286-294.
40. Santos, J.B.C., de Barros, V.V.S., Linares, J.J., 2017. The Hydraulic Retention Time as a Key Parameter for the Performance of a Cyclically Fed Glycerol-Based Microbial Fuel Cell from Biodiesel. *J. Electrochem. Soc.* **164**(3), H3001-H3006.
41. Sharma, Y., Li, B., 2010. Optimizing energy harvest in wastewater treatment by combining anaerobic hydrogen producing biofermentor (HPB) and microbial fuel cell (MFC). *Int. J. Hydrogen Energy* **35**(8), 3789-3797.
42. Sobhi, M., Guo, J., Cui, X., Sun, H., Li, B., Aboagye, D., Shah, G.M., Dong, R., 2019. A promising strategy for nutrient recovery using heterotrophic indigenous microflora

- from liquid biogas digestate. *Sci. Total Environ.* **690**, 492-501.
43. Sobieszuk, P., Zamojska-Jaroszewicz, A., Makowski, L., 2017. Influence of the operational parameters on bioelectricity generation in continuous microbial fuel cell, experimental and computational fluid dynamics modelling. *J. Power Sources* **371**, 178-187.
44. Song, H.-L., Li, H., Zhang, S., Yang, Y.-L., Zhang, L.-M., Xu, H., 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.
45. Srikanth, S., Kumar, M., Singh, D., Singh, M., Das, B., 2016. Electro-biocatalytic treatment of petroleum refinery wastewater using microbial fuel cell (MFC) in continuous mode operation. *Bioresour. Technol.* **221**, 70-77.
46. Walker, R.V., Beck, M.B., Hall, J.W., Dawson, R.J., Heidrich, O., 2014. The energy-water-food nexus: Strategic analysis of technologies for transforming the urban metabolism. *J. Environ. Manag.* **141**, 104-115.
47. Wang, F., Wei, J., Zou, X., Fu, R., Li, J., Wu, D., Lv, H., Zhu, G., Wu, X., Chen, H., 2019. Enhanced electrochemical phosphate recovery from livestock wastewater by adjusting pH with plant ash. *J. Environ. Manag.* **250**, 109473.
48. Wang, M., Zhang, S.-C., Tang, Q., Shi, L.-D., Tao, X.-M., Tian, G.-M., 2020. Organic degrading bacteria and nitrifying bacteria stimulate the nutrient removal and biomass accumulation in microalgae-based system from piggery digestate. *Sci. Total Environ.* **707**, 134442.
49. Wang, Y.-K., Li, W.-W., Sheng, G.-P., Shi, B.-J., Yu, H.-Q., 2013. In-situ utilization of generated electricity in an electrochemical membrane bioreactor to mitigate membrane fouling. *Water Res.* **47**(15), 5794-5800.

50. Wei, L., Yuan, Z., Cui, M., Han, H., Shen, J., 2012. Study on electricity-generation characteristic of two-chambered microbial fuel cell in continuous flow mode. *Int. J. Hydrogen Energy* **37**(1), 1067-1073.
51. Xia, P., Wang, X., Wang, X., Zhang, J., Wang, H., Song, J., Ma, R., Wang, J., Zhao, J., 2016. Synthesis and characterization of MgO modified diatomite for phosphorus recovery in eutrophic water. *J. Chem. Eng. Data* **62**(1), 226-235.
52. Xie, T., Jing, Z., Hu, J., Yuan, P., Liu, Y., Cao, S., 2018. Degradation of nitrobenzene-containing wastewater by a microbial-fuel-cell-coupled constructed wetland. *Ecol. Eng.* **112**, 65-71.
53. Xu, L., Zhao, Y., Tang, C., Doherty, L., 2018a. Influence of glass wool as separator on bioelectricity generation in a constructed wetland-microbial fuel cell. *J. Environ. Manag.* **207**, 116-123.
54. Xu, S., Wu, D., Hu, Z., 2014. Impact of hydraulic retention time on organic and nutrient removal in a membrane coupled sequencing batch reactor. *Water Res.* **55**, 12-20.
55. Xu, T., Wang, Q., Wu, S., Fu, B., Liang, P., Huang, X., Zhang, X., 2018b. A novel filtration composite anode configuration of microbial fuel cell for efficient wastewater treatment and enhanced power generation. *J. Clean. Prod.* **178**, 305-313.
56. Yan, T., Ye, Y., Ma, H., Zhang, Y., Guo, W., Du, B., Wei, Q., Wei, D., Ngo, H.H., 2018. A critical review on membrane hybrid system for nutrient recovery from wastewater. *Chem. Eng. J.* **348**, 143-156.
57. Yang, Y., Choi, C., Xie, G., Park, J.-D., Ke, S., Yu, J.-S., Zhou, J., Lim, B., 2019. Electron transfer interpretation of the biofilm-coated anode of a microbial fuel cell and the cathode modification effects on its power. *Bioelectrochemistry* **127**, 94-103.
58. Ye, Y., Ngo, H.H., Guo, W., Chang, S.W., Nguyen, D.D., Liu, Y., Nghiem, L.D., Zhang, X., Wang, J., 2019a. Effect of organic loading rate on the recovery of nutrients and

- energy in a dual-chamber microbial fuel cell. *Bioresour. Technol.* **281**, 367-373.
59. Ye, Y., Ngo, H.H., Guo, W., Liu, Y., Chang, S.W., Nguyen, D.D., Liang, H., Wang, J., 2018. A critical review on ammonium recovery from wastewater for sustainable wastewater management. *Bioresour. Technol.* **268**, 749-758.
60. Ye, Y., Ngo, H.H., Guo, W., Liu, Y., Chang, S.W., Nguyen, D.D., Ren, J., Liu, Y., Zhang, X., 2019b. Feasibility study on a double chamber microbial fuel cell for nutrient recovery from municipal wastewater. *Chem. Eng. J.* **358**, 236-242.
61. Ye, Y., Ngo, H.H., Guo, W., Liu, Y., Li, J., Liu, Y., Zhang, X., Jia, H., 2017. Insight into chemical phosphate recovery from municipal wastewater. *Sci. Total Environ.* **576**, 159-171.
62. Zhou, Y., Wang, L., Zhou, Y., Mao, X.-z., 2020. Eutrophication control strategies for highly anthropogenic influenced coastal waters. *Sci. Total Environ.* **705**, 135760.