



# Effective swine wastewater treatment by combining microbial fuel cells with flocculation



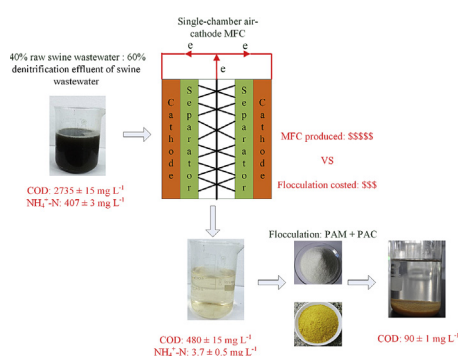
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## HIGHLIGHTS

- MFC and flocculation process were combined to treat the mixture of raw swine wastewater and its denitrification effluent.
- When the mixed ratio was 40:60, MFCs removed  $99.1 \pm 0.1\%$  of ammonia and produced a maximum power density of  $37.5 \text{ W m}^{-3}$ .
- MFC effluent was further treated by flocculation process with an overall COD removal efficiency of  $96.6 \pm 0.2\%$ .
- The economic benefit created by MFCs was sufficient to offset the cost of flocculation process.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Microbial fuel cells (MFCs) provide a cost-effective method for treating swine wastewater treatment and simultaneously producing electricity, yet they need to be combined with other wastewater treatment processes to improve the effluent water quality. In this paper, we constructed single-chamber air-cathode MFCs with a compact configuration for nitrogen and COD removal and high electricity production and combined them with a low-cost flocculation process to discharge higher quality wastewater. We show that MFCs could remove ammonia at a rate of  $269.2 \pm 0.5 \text{ g m}^{-3} \text{ d}^{-1}$  ( $99.1 \pm 0.1\%$  ammonia removal efficiency) with a maximum power density of  $37.5 \text{ W m}^{-3}$  and 21.6% of coulombic efficiency at a 40:60 ratio of raw swine wastewater, to denitrification effluent of swine wastewater. Up to  $82.5 \pm 0.5\%$  COD could be removed with MFCs, from  $2735 \pm 15 \text{ mg L}^{-1}$  to  $480 \pm 15 \text{ mg L}^{-1}$ , and flocculation further reduced levels to  $90 \pm 1 \text{ mg L}^{-1}$  for a  $96.6 \pm 0.2\%$  overall COD removal efficiency of the combination technology. Cost analysis of the combined MFC and flocculation process showed a net economic benefit of  $\$ 0.026 \text{ m}^{-3}$ . In summary, this novel combination wastewater treatment method provides an effective way to treat swine wastewater to low pollutant levels in the effluent at low cost (a net gain).

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## 1. Introduction

Swine wastewater is becoming a serious environmental concern due to increasingly large-scale and intensive operations in the pig farming industry. The main composition of swine wastewater are feces, urine, and washing water, which are characterized by high

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levels of suspended solids (SS), chemical oxygen demand (COD, 5000–30000 mg L<sup>-1</sup>), total nitrogen (TN, 800–6000 mg L<sup>-1</sup>), total phosphorus (TP, 100–1400 mg L<sup>-1</sup>) and pathogenic bacteria (Moral et al., 2005; Provolo and Martinezsuller, 2007; Moral et al., 2008; Blanesvidal et al., 2009; Villamar et al., 2011). Therefore, swine wastewater needs to be treated to meet emission standards before being released into the environment. Many biological treatment processes have been developed to remove inorganic and organic pollutants in swine wastewater including using activated sludge (Guo et al., 2013), aerobic sequencing batch reactors and reverse osmosis (Zhang et al., 2004) or wetland plants (Klomjek, 2016). However, all of these processes are associated with high operating costs.

Recovering energy in the form of hydrogen or methane gas from wastewater is an effective approach to offset the costs of swine wastewater treatment (Logan et al., 2002; Angenent et al., 2004; Wagner et al., 2009). Biological hydrogen production technology has hardly been applied to swine wastewater treatment because of its low energy recovery and high operating cost (Logan, 2004; Oh and Logan, 2005; Rabaey and Verstraete, 2005). In contrast, methanogenic anaerobic digestion in upflow anaerobic sludge blankets (UASB) (Lettinga et al., 1980), anaerobic migrating blanket reactors (AMBR) (Angenent et al., 2004), anaerobic sequencing batch reactors (ASBR) (Angenent et al., 2002) and microbial electrolysis cells (MECs) (Wagner et al., 2009) represent more feasible technologies for swine wastewater treatment thanks to relatively low investment and operating costs, high rates of organic matter removal and the capacity to recover energy in the form of methane gas (Caporgno et al., 2015). However, the effluent of anaerobic digestion still contains high concentrations of nitrogen, phosphorus and COD which need to be recycled and further treated (Caporgno et al., 2015).

Microbial fuel cells (MFCs) may represent a potential alternative to methanogenic anaerobic digesters for the treatment of swine wastewater as they can efficiently remove COD and nitrogen and produce electricity directly from swine wastewater (Min et al., 2005; Yan et al., 2012; Sun et al., 2016). MFCs have successfully been applied to the treatment of domestic (Cheng and Logan, 2011), food (Oh and Logan, 2005) and brewery wastewaters (Zhuang et al., 2012a) but little is known about their capacity to treat highly polluted wastewater such as swine wastewater.

In the past, swine wastewater often had to be diluted prior to be treated by MFCs because high levels of ammonia nitrogen or the volatile produced in the process of fermentation affect electricity generation (Nam et al., 2010). Up to 92% COD removal, 83% NH<sub>4</sub><sup>+</sup>-N removal and 11 W m<sup>-3</sup> were achieved in different MFC reactors with the diluted swine wastewater (Min et al., 2005; Ryu et al., 2013; Li et al., 2013; Zhuang et al., 2012b). These studies demonstrated that it was feasible to simultaneously remove pollutants and generate electricity directly from swine wastewater using MFCs. However, in order to offset operating costs, the power density outputs would need to be significantly increased, potentially by decreasing electrode spacing and increasing cathode surface area (Liu et al., 2005a; Cheng and Logan, 2011).

The relative high concentration of pollutants contained in the effluent of MFCs (800–1000 mg L<sup>-1</sup> COD, 115–250 mg L<sup>-1</sup> NH<sub>4</sub><sup>+</sup>-N) (Ryu et al., 2013) presented another challenge for swine wastewater treatment. The effluent of MFCs would need to be further processed with additional existing wastewater treatment methods to comply with discharging regulations (Li et al., 2014; Gude, 2016). Sequencing batch reactor (SBR) (Wang et al., 2014), activated sludge (Gajaraj and Hu, 2014), anaerobic-anoxic-oxic (Xie et al., 2011), membrane bioreactor (Wang et al., 2016) and wetland methods (Chen et al., 2012) have recently been combined with MFCs to enhance the removal of pollutants. However, the application of

these integrated technologies is hindered by increased treatment costs due to aeration or membrane requirements. Flocculation with polyaluminum chloride (PAC) and polyacrylamide (PAM) may be a better-suited method as its application to wastewater treatment is associated with lower cost, good disposal effects and relatively simple operations (Al-Mutairi et al., 2004).

In this study, single-chamber air-cathode MFCs with a compact configuration were constructed and tested for nitrogen and COD removal and simultaneous power generation from mixed swine wastewater. Residual COD contained in the MFC effluent was subsequently removed by flocculation to increase the quality of the discharging wastewater. In addition, the energy balance between the MFC and flocculation treatment processes was evaluated.

## 2. Materials and methods

### 2.1. Swine wastewater

The studied wastewater was a mixture of raw swine wastewater (RSW) and the effluent of denitrified swine wastewater (DEW) from a local swine farm (Hangzhou, China). The RSW to DEW ratios of the mixture ranged from 15:85, 20:80 and 30:70 to 40:60 (denoted as 15RSW, 20RSW, 30RSW and 40RSW, respectively). The wastewater mixture was filtered through a screen (300 mesh) to remove solid particles and stored in a refrigerator at 4 °C until use. The mixtures were fed as the substrate directly into MFCs without any modifications such as pH adjustment or nutrient additions. The parameters of the wastewater mixtures are shown in Table 1.

### 2.2. MFC configuration

Single chamber MFCs with two pieces of air cathode (200 × 100 mm) and nine carbon brush anodes (20 mm in diameter and 100 mm in length) were constructed with a cubic polyvinyl chloride (PVC) frame (200 × 100 × 20 mm) (Fig. 1). Carbon brushes were treated as precisely described (Feng et al., 2010). Carbon brushes were uniformly placed in the middle of the frame and connected outside of the frame by a copper wire (1.5 mm in diameter). Cathodes were composed of nickel foam current collector, activated carbon catalyst layer and polytetrafluoroethylene (PTFE) diffusion layer as described previously (Cheng and Wu, 2013). Two cathodes were placed on the opposite sides of anodes symmetrically with the catalyst layer facing the anode and the diffusion layer facing out. Glass fiber separators (1.4 mm thick) of the same size as the cathodes were placed between cathode and anode to prevent electrical short circuits and protect the cathode catalyst from biofouling (Zhang et al., 2011). The electrode spacing (the distance between the cathode and the core metal of the brush anode) was 12 mm. The solution volume of the MFC reactor was 340 mL.

### 2.3. MFC inoculation and operation

The reactors were initially inoculated with the effluent of an air-cathode MFC that had been running with sodium acetate for more than one year. Then, MFCs were operated with a nutrient solution containing 50 mM phosphate buffer solution (PBS), 1 g L<sup>-1</sup> sodium acetate, 12.5 mL L<sup>-1</sup> trace minerals and 5 mL L<sup>-1</sup> vitamin solution (Liu et al., 2005b) for 15 cycles. An external resistance of 100 Ω was used at both stages. After inoculation, MFCs were operated at a decreasing external resistance of 10 Ω to obtain high and stable performance. In order to enhance ammonia removal, slaughter wastewater was fed as the source of nitrifying bacteria until the nitrification process was well-established (Sotres et al., 2016). Subsequently, MFCs were switched to the swine wastewater

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