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Aerobic granular sludge inoculated microbial fuel cells for enhanced epoxy reactive diluent wastewater treatment

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HIGHLIGHTS

- Aerobic granular sludge was incorporated into MFCs to form a hybrid system.
- AGS-MFCs shortened the startup time from 13 d to 7 d compared to MFCs without AGS.
- Superior performance for epoxy reactive diluent wastewater treatment was achieved.
- The high efficiency is possibly due to the synergic effect of both MFC and AGS.

GRAPHICAL ABSTRACT

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ABSTRACT

Microbial consortiums aggregated on the anode surface of microbial fuel cells (MFCs) are critical factors for electricity generation as well as biodegradation efficiencies of organic compounds. Here in this study, aerobic granular sludge (AGS) was assembled on the surface of the MFC anode to form an AGS-MFC system with superior performance on epoxy reactive diluent (ERD) wastewater treatment. AGS-MFCs successfully shortened the startup time from 13 d to 7 d compared to the ones inoculated with domestic wastewater. Enhanced toxicity tolerance as well as higher COD removal (77.8% vs. 63.6%) were achieved. The higher ERD wastewater treatment efficiency of AGS-MFC is possibly attributed to the diverse microbial population on MFC biofilm, as well as the synergic degradation of contaminants by both the MFC anode biofilm and AGS granules.

1. Introduction

The release of industrial wastewater containing recalcitrant organics could generate severe adverse impacts on both surface and ground water (Luo et al., 2009; Liu and Li, 2014; Abbasi et al., 2016; An et al., 2016; Wang et al., 2016), posing severe threat to both public health and ecological systems (Habibul et al., 2016). Current wastewater treatment plants for domestic wastewater treatment are mostly constructed based on the well-established activated sludge process. However, they are not suitable for the treatment of hazardous wastewater due to the susceptibility of microorganisms to the toxic components (Sahinkaya and Dilek, 2005). Microbial fuel cells have been intensively studied as novel platforms for both wastewater treatment and energy harvest (Liu et al., 2004; Ogugbue et al., 2015; Sonawane et al., 2014), demonstrating capabilities of combusting versatile organics from glucose

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(Chaudhuri and Lovley, 2003), acetate (Pant et al., 2010a,b), sucrose (Kim et al., 2010) to domestic and brewery wastewater that contain more complicated mixed compounds (Kim et al., 2015; Wen et al., 2009). Furthermore, both organic and inorganic wastes originated from vegetable oil industries, glass and marble industries and chemical industries were also treated by MFCs, with 85–90% of COD removals achieved (Abbasi et al., 2016). MFCs have also demonstrated their conceptual usages in recalcitrant contaminants removal using phenol as a typical treatment target (Pant et al., 2010a,b). However, phenol degradation efficiency with closed circuit MFC was only 8–14% higher than that worked at opened circuit condition. Most of the removed phenol (92.4%) was attributed to anaerobic digestion but not exoelectrogenic microbial conversion. Thus, the advantages of using MFCs as a treatment strategy of phenol are very limited (Luo et al., 2009). In addition, a prolonged startup time for microbial biofilm acclimation is usually inevitable and a low degradation efficiency as well as unstable performance are frequently observed.

Degradation efficiency of a specific contaminant in an MFC is highly dependent on the microbial consortiums aggregated on the anode surface, which constitute electrochemical active bacteria that directly convert organic compounds into electricity, as well as non-electrochemical active bacteria that are also critical for electricity generation through synergetic cooperation. Source of inoculum and carbon source types are two critical parameters to determine the constitution of microbial consortiums of the electrogenic microbial biofilm. Metal reducing bacteria such as *Geobacter* spp., *Shewanella* spp. are typical microorganisms that are uncovered from MFC anode biofilm when natural river water or domestic wastewater is used as inoculum (Zhi et al., 2014; Hou et al., 2009). It is found that *Desulfovibrio* (18.4%) constitutes the key genus level content when brewery waste was used as the inoculum and azo dye as the carbon source (Miran et al., 2015). The microbial groups on anode biofilm were highly diverse, with 62.3% of all sequences unclassified. Feeding the MFC reactor with acetic acid decreased the diversity and richness of biofilm species, especially the richness of *Geobacter sulfurreducens* (Kiely et al., 2011).

Diverse microbial communities in MFCs are desired to enhance electrogenic capability of biofilm for simultaneous carbon digestion and bioenergy generation (Kiely et al., 2011), as well as to build up *anti*-toxic property enabling efficient reproduction of the microbial population. Previous study which used suspended sludge as microbial inoculum of MFCs was found to enhance power generation when treating feces wastewater (Zhi et al., 2008). Aerobic granular sludge (AGS) is considered to be a complicated system formed by large varieties of aerobic and anaerobic respiring microbes that colonized in different granule layers (Wen et al., 2009). They are densely-packed microbial aggregates with rich and strong microbial structure, high biomass retention, excellent biological efficiency and toxicity tolerance. Aerobic sludge granules have been successfully applied for the treatment of high strength organic and toxic industrial wastewater like phenol, 4-chlorophenol, *etc.* Aerobic granules played a promising role in adsorption of toxic chemicals and further degradation through co-metabolism of assorted microbial communities colonized in different layers of granules (Gao et al., 2011). The cultivation of aerobic granular sludge in MFC cathode chamber has been studied and the granule's physiochemical properties were also investigated (Chen, 2014). However, strategies to enhance the efficiency of MFCs for recalcitrant wastewater treatment were very limited, and combining the advantages of both AGS and MFC technologies to simultaneously realize efficient recalcitrant wastewater treatment as well as enhancing bioenergy harvest has not been studied.

Here in this study, the addition of AGS as the inoculum of MFCs was studied for the treatment of recalcitrant wastewater from epoxy reactive diluent (ERD) production. The startup time was evaluated compared to MFCs without AGS. Besides, toxicity tolerance to the influent industrial wastewater was discussed. Both COD removal and power generation were obtained and compared to domestic wastewater inoculated MFCs. Mechanisms of the synergic effects of anodic biofilm and AGS on ERD wastewater treatment were also proposed.

2. Materials and methods

2.1. Aerobic granular sludge cultivation

Mature aerobic granular sludge assembled in microbial fuel cells had been previously cultivated in a column sequencing batch reactor (SBR) with a working volume of 9.9 L (diameter of 8.4 cm, height of 200 cm, H/D ratio of 21.4), a superficial gas velocity of 2.0 cm/s and a water exchange ratio of 61.8% at the end of each cycle (6 h). Activated sludge (70%) and previously cultivated aerobic granules (30%) were mixed and added into the SBR reactor to initiate the granulation. Synthesized wastewater consisted of sodium acetate, ammonia nitrogen, phosphates and minerals was fed. Other operation parameters were used as previously reported (Long et al., 2014).

2.2. MFC configuration and operation

Carbon cloth anode (3.8 cm in diameter) was pretreated as follows: It was firstly cleaned by soaking in acetone, ethanol and deionized water sequentially with each for 15 min to remove possible organic contaminants. The cleaned carbon cloth was then heat-treated in a muffle furnace at 450 °C for 30 min. It was rinsed with deionized water three times before used in an MFC (Feng et al., 2010). To fabricate the air-cathode (3 cm in diameter), 100 mg activated carbon powder,10 mg carbon black and 1 mL 10% (w/v) PVDF solution were mixed together, then the mixture was uniformly smeared on a stainless steel mesh and dipped in deionized water with the carbon side facing down for 15 min (Yang et al., 2014).

Single-chamber air-cathode MFC reactor with a volume of 28 mL operated under batch mode as previously described was used (Liu and Logan, 2004). Aerobic granular sludge was weighed for 3 g and loaded on the surface of anode to form a layer covering the anode. Stainless steel mesh was gently pressed against the aerobic granular sludge to hold the AGS granules in place to prevent them from dropping off the anode surface while not breaking the granule structures (Fig. 1 and Fig. S1).

MFC devices were fed with medium solution containing sodium acetate (1 g/L), 50 mL phosphate buffer solution (PBS), vitamins (5 mL/L) and minerals (12.5 mL/L) as previously described (Lovley and Phillips, 1988). During the start-up, AGS inoculated MFCs (AGS-MFCs) were fed with 28 mL medium solution, while MFCs without AGS were fed with 14 mL medium and 14 mL domestic municipal wastewater (DW-MFCs) from Tangxun Lake municipal wastewater treatment plant in Wuhan, China. Anode and cathode were connected with a 1000 Ω external resistor during the startup process. For ERD wastewater treatment, different proportions of ERD wastewater was mixed with 1 g/L sodium acetate synthetic wastewater to achieve ERD concentrations from 10% to 100% and added into both types of MFC sequentially.

The influent (sodium acetate) in startup period was changed whenever the voltage output dropped below 50 mV. In the steady power generation phase, sodium acetate substrate was exchanged every 2 days for both AGS-MFCs and DW-MFCs. For ERD wastewater treatment, a retention time of 36 h was used for each cycle. Substrate was exchanged using a 10 mL syringe. All experiments were carried out in duplicates at room temperature (23 ± 1 °C).