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Simultaneous removal of sulfanilamide and bioelectricity generation in two-chambered microbial fuel cells

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ABSTRACT

Sulfanilamide antibiotics show the highest detection frequencies and concentrations in surface water of China. In this work, we aim to investigate the feasibility of sulfanilamide removal in the anode of two-chambered MFCs and to examine the influence of sulfanilamide on the electricity generation in MFCs. Results show that sulfanilamide can be efficiently removed in the anode of MFCs and exerts exciting positive effect on electricity generation at the studied concentration range (10–30 mg L^{-1}). In comparison with MFCs using glucose as the sole substrate, the peak voltage output is increased by 8.5, 13.9, and 15.7% for addition of 10, 20, and 30 mg L^{-1} sulfanilamide, respectively. The anode and the cathode polarization curves reveal that the lower anode overpotential with addition of sulfanilamide is responsible for the promoted electric power output of MFCs. Four groups of comparison experiments are conducted. In normal MFCs, sulfanilamide removal efficiency reaches 90% in 96 h, while the removal efficiencies are 58, 10.8, and 7.5% in open-circuit, no co-substrate and abiotic MFCs, respectively. The results of comparison experiments indicate that sulfanilamide removed in MFCs is mainly stemmed from the biocatalytic co-metabolism degradation other than physical adsorption and the current generated in MFCs plays an active role in accelerating the removal efficiency. These experimental results offer theoretical basis for the possibility of the MFCs application on the industrial wastewater treatment containing sulfanilamide and simultaneous electricity generation.

Keywords: Microbial fuel cells; Sulfanilamide; Removal; Electricity generation

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1. Introduction

Recently, researchers have found that bioelectrochemical processes can enhance the removal rate of contaminants [1,2]. Microbial fuel cell (MFC) is the most popular bioelectrochemical system (BES), which exploits micro-organisms to catalyze electrochemical reactions and to generate electricity from various substrates [3,4]. In MFC, the anode could serve as a sufficient anaerobic terminal electron acceptor for microbes to enhance the degradation of organic contaminants under anaerobic conditions [5–7]. And in contrast to planktonic cells, anodic microbial biofilms in BES show less susceptibility to toxins (such as heavy metals and antibiotics) [8–10]. MFC is becoming a promising candidate for the removal of environmental contaminants and renewable energy generation in the past decade.

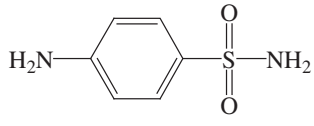
Recently, studies have been reported on the degradation of recalcitrant contaminants in MFCs. Huang et al. studied the degradation of pentachlorophenol (PCP) in a MFC with fermentable and non-fermentable co-substrates. The results indicated that PCP degradation rates and power densities were affected by current generation and the type of electron donor [11]. Liu et al. demonstrated that the MFC displayed a maximum power density of 1.778 mW m^{-2} and a maximum PNP degradation rate of 64.69% when PNP was used as a sole substrate [12]. Wang et al. adopted 26 trace organic compounds (TOrCs) with broad physicochemical properties to spike in synthetic wastewater, and studied the removal and fate of these TOrCs, and confirmed that TOrCs removal process involved both sorption and biodegradation [13]. However, the study of recalcitrant contaminants removal in MFCs is still in its infancy and tremendous effort is required to extend the application fields of MFCs before practical application can be realized.

As a type of antimicrobial used in the treatment and prevention of bacterial infection, large amounts of antibiotics are produced and consumed in controlling diseases in humans and animals. Recently, with the development of analysis technology, antibiotics have been detected in various aquatic environments [14]. Therefore, antibiotics have received growing global attentions as emerging contaminants threatening human health and aquatic ecosystems.

So far, a few reports could be tracked on the research of antibiotics removal and simultaneous electricity generation in MFCs. Wen et al. constructed air-cathode MFCs to generate electricity with glucose-ceftriaxone sodium or glucose-penicillin mixtures. In comparison to 1 g L^{-1} glucose (19 W m^{-3}), the maximum power density is increased by 495% for 50 mg L^{-1} ceftriaxone sodium + 1 g L^{-1} glucose (113 W m^{-3}). The maximum current density with 50 mg L^{-1} penicillin (10.73 A m^{-2}) was 3.5-fold compared with that without penicillin (3.03 A m^{-2}) [15,16]. Wu and colleagues studied the impact of tobramycin on the performance of MFC and experimental results demonstrated that the inhibition ratio of the MFC increased exponentially with the tobramycin concentrations in the range of 0.1–1.9 g L^{-1} [10]. Sun et al. and Liang et al. studied the reductive degradation of CAP in the cathode chamber of BES with applied external voltage of 0.5 V [17,18]. Sun et al. reported that CAP reduction potential of the biocathode was 0.3 V higher than that of the abiotic cathode. It indicated that the overpotential of CAP reduction with the biocathode was decreased, and which would in turn lower the energy barrier for CAP reduction. Liang et al. focused their attention on the degradation pathway of CAP, providing detailed information about the degradation mechanism. Harnisch et al. studied the removal of the sulfonamides from artificial wastewater using anodic microbial biofilms. They reported that the removal process did not affect the microbial bioelectrocatalytic performance. But they did not report the simultaneous electricity generation during the removal process [9].

As we know, MFC is gaining an increasing attention for simultaneous wastewater treatment and biomass-based electricity production. Since the explorations about the simultaneous electricity generation during the process of antibiotics removal using MFC are still scarce. Therefore, we are interested in whether antibiotics (such as Sulfanilamide) could be removed in the anode of MFC and whether their degradation process would have any positive or negative impact on the power output of MFC.

Sulfanilamide falls into the category of sulfonamide antibacterial. It is frequently used in the form of a topical cream or powder to treat surface infections, as well as a pill for internal infections. Chemically, it is an organic compound consisting of an aniline derivitized with a sulfonamide group and the chemical structure is:



Considering sulfanilamide antibiotics showed the highest detection frequencies and levels in surface water of China [19]. In this work, sulfanilamide, which possesses the basic structure of sulfanilamides antibiotics, is selected as a representative antibiotic to investigate the feasibility of sulfanilamide removal in MFCs and to explore the influence of sulfanilamide on the

electric power generation of MFCs. These results could provide basic data of the simultaneous sulfanilamide removal and bioelectricity generation in MFCs.

2. Materials and methods

2.1. Construction and operation of MFCs

Two-chambered MFC, as Fig. 1 showed, was consisted of two equal rectangular perspex frames (served as an anode chamber and a cathode chamber) with an operating volume of 140 mL (7 cm × 5 cm × 4 cm) as previously reported [20,21]. The two frames were held together by an external metal screw and CMI-7000 cation exchange membrane was sandwiched between the two frames. Rubber gaskets were used to secure a seal between perspex walls and the membrane. Carbon papers (without waterproofing or catalyst) with a projected surface area of 16 cm^2 were used as electrodes. Prior to use, they were soaked in acetone for a period of 4 h and then soaked in 1 M hydrochloric acid and 1 M sodium hydroxide, respectively, for 24 h. Titanium wires were used to connect the circuit with an external resistance of 1,000 ohms (except for the polarization experiments) and all leaks were sealed to maintain anaerobic microenvironment in the anode chamber.

Anaerobic sludge collected from a local wastewater treatment plant was used for original inoculum for MFC and was controlled as 25% (volume) of the whole anode chamber. During the start-up and acclimation stage, the anode chamber was filled with anodic growth medium, which was composed of basic nutrient medium (50 mM PBS, 12.5 mL L^{-1} vitamins, and 12.5 mL L^{-1} mineral) and organic carbon source (glucose, 1,000 mg L^{-1}) as our previously reported [20,21]. Once the start-up and acclimation stage accomplished, different concentration of sulfanilamide (10, 20, and 30 mg L^{-1}, respectively) was added into the above anodic growth medium to further examine the sulfanilamide removal and simultaneous bioelectricity generation. For all MFC experiments: 100 mM $K_3[Fe(CN)_6]$ in 50 mM (phosphate buffer solution (PBS)) (PBS, pH 7.0) was used as the highly efficient electron acceptors [22,23]; the anodic growth medium was replaced by fresh cultivation solution when the voltage decreased below 50 mV; N_2 gas was flushed continuously for 15 min to remove dissolved oxygen in the anodic chamber before each batch test. MFC experiments are affected by the biological fluctuation of the anaerobic sludge and other experimental conditions; therefore, parallel groups of experiments are carried out in our work.

Four groups of comparison experiments, including normal MFCs, open-circuit MFCs, no co-substrate MFCs, and abiotic MFCs (as shown in Table 1), were conducted to test the impact of electron transfer and the effect of physical adsorption on the removal of sulfanilamide in MFCs. All the comparison experiments were operated in batch-fed mode with the same experimental parameters, the same sampling method, and the same pretreatment.

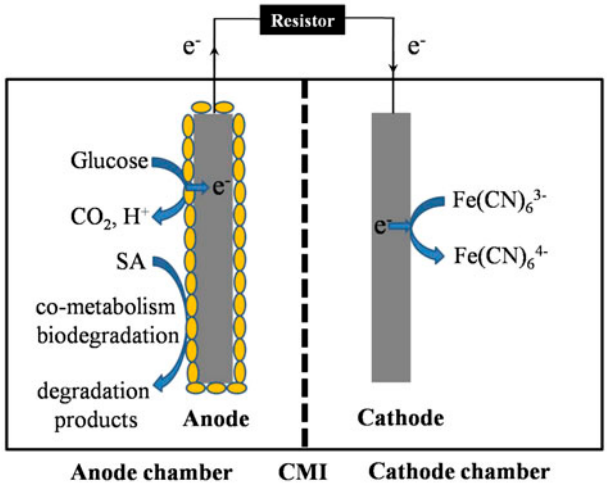


Fig. 1. Configuration of the two-chembered MFC (SA is short for sulfanilamide).

2.2. Analysis and calculations

The voltage across an external resistor was recorded with a digital multimeter every 30 min. Polarization and power density curves of the anode, the cathode, and the whole cell were obtained by changing external circuit resistance from 10,000 down to 50 Ω. The current density, I_A (A m^{-2}) and the power density, P_A (W m^{-2}), of the system were calculated using the formula: $I_A = V/(R\,A)$ and $P_A = V^2/(R\,A)$, where V (V) is the cell voltage, R (Ω) is the external resistances, and A (m^2) is the projected area of the anode.

A UV–vis spectrometer (Beijing Purkinje General Instrument Co., Ltd, China) was used for absorbance measurements. The spectra of Sulfanilamide displayed the maximum absorption band at 258 nm in this work. The UV–vis absorbance of the characteristic band decreased gradually with the increase in reaction time reflecting the successful removal of Sulfanilamide in the anode solution. So, removal of Sulfanilamide was determined by monitoring the decrease in the absorbance at a wavelength of 258 nm. Samples were withdrawn from the anode chamber at a time interval of 2 h during every cycle with different concentrations of

Table 1
Operation mode for comparison experiments

	External circuit	Co-substrate	Inoculums
Normal MFCs	Connected	Added	Anaerobic sludge
Open-circuit MFCs	Disconnected	Added	Anaerobic sludge
No co-substrate MFCs	Connected	Not added	Anaerobic sludge
Abiotic MFCs	Connected	Added	Autoclaved anaerobic sludge

sulfanilamide. In order to remove impurities and suspended biomass from the anode solution, samples were firstly centrifuged at 4,000 rpm for 15 min and then filtered through a 0.22 μm-pore-size syringe filter unit. Finally, sample solutions with higher concentration of sulfanilamide were diluted prior to measurement of absorbance. The anode solution taken from the anode of MFCs without sulfanilamide was used as the blank solution and was pretreated with the same procedures as that of the samples. Sulfanilamide removal efficiency is calculated as the following:

$$\text{Removal efficiency}\,(\%) = \frac{A_0 - A_t}{A_0} \times 100\% \quad (1)$$

where A_0 is the absorbance of the initial solution taken from the anode chamber, and A_t is the absorbance of solution taken from the anode chamber at certain reaction time t (min).

Chemical oxygen demand (COD) was measured according to the standard dichromate titration method. Glucose was analyzed using the anthrone method. The COD removal efficiency of the anode solution is measured by the decrease in COD of the anode solution and is estimated by the following expression:

$$\text{COD removal efficiency}\,(\%) = \frac{\text{COD}_0 - \text{COD}_t}{\text{COD}_0} \times 100\% \quad (2)$$

where COD_0 is the COD of the initial solution taken from the anode chamber, and COD_t is the COD of the solution taken from the anode chamber at certain reaction time t (min).

Glucose degradation efficiency was calculated as the following:

$$\text{Glucose degradation efficiency}\,(\%) = \frac{G_0 - G_t}{G_0} \times 100\% \quad (3)$$

where G_0 is the glucose concentration of the initial solution taken from the anode chamber, and G_t is the glucose concentration of solution taken from the anode chamber at certain reaction time t (min).

3. Results and discussion

3.1. Effects of sulfanilamide on the performance of MFCs

Prior to the experiments, MFCs should be started up with 1 g L^{-1} glucose as the sole organic carbon source and anaerobic sludge as mixed culture bacterial source. During the acclimation period, the maximum voltage gradually ascends as the time increases and yields a stable maximum output voltage of 570 ± 10 mV after nearly 500 h, indicating the exoelectrogenic biofilm formation and marking the ending of the start-up stage.

The electricity generating function has always been a distinguishing feature of the MFC technology, so it is interesting to explore the effect of sulfanilamide on the electric power output of MFCs. Distinct concentrations of sulfanilamide (10, 20, and 30 mg L^{-1}) are added into the anode of the MFCs. One of the representative cycles is presented in Fig. 2, exciting voltage changing trend is shown at these tested concentration levels. The maximum output voltages are not only

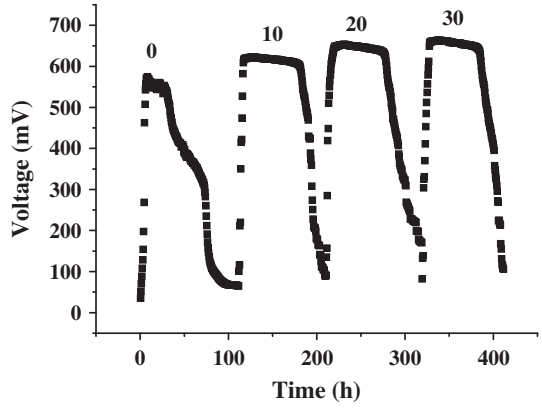


Fig. 2. Voltage generation from MFCs with 1 g L^{-1} glucose and different concentration of sulfanilamide (numbers indicated the concentration of sulfanilamide, mg L^{-1}) under external resistance of 1,000 Ω.

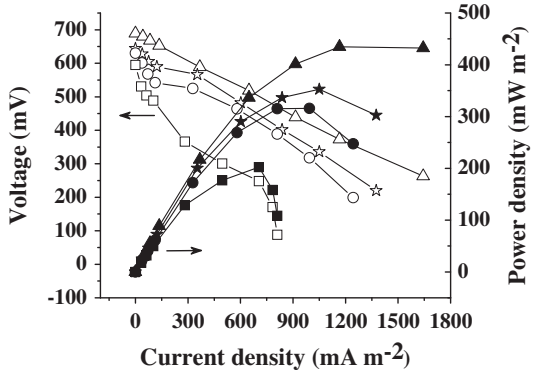


Fig. 3. Polarization (hollow symbol) and power density (solid symbol) curves of MFCs with 1 g L^{-1} glucose and different concentrations of sulfanilamide (0■, 10●, 20★, and 30▲ mg L^{-1}).

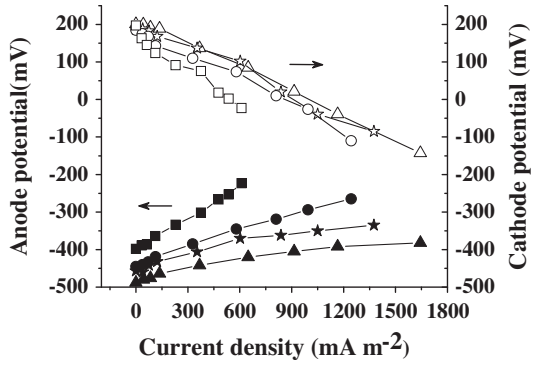


Fig. 4. Anode (solid symbol) and cathode (hollow symbol) polarization curves of MFCs with 1 g L^{-1} glucose and different concentrations of sulfanilamide (0■, 10●, 20★, and 30▲ mg L^{-1}).

restrained by the added sulfanilamide, but also are promoted to reach the higher voltages. In comparison with MFCs using glucose as the sole substrate, the peak voltage output is increased by 8.5% for addition of 10 mg L^{-1} sulfanilamide, 13.9 and 15.7% for 20, and 30 mg L^{-1}, respectively.

Fig. 3 shows the polarization curves and power density curves of MFCs with different concentration of sulfanilamide. The MFC generates a maximum power density of 202 mW m^{-2} using glucose as the sole substrate. Addition of 10 mg L^{-1} of sulfanilamide improves the maximum power density by 56% to 316 mW m^{-2}. When 20 mg L^{-1} of sulfanilamide is added, the maximum power density reaches 352 mW m^{-2}, which is 74% higher than that obtained without sulfanilamide. Increasing the sulfanilamide concentration to 30 mg L^{-1}, further increased the maximum power density to 434 mW m^{-2}, which is two times larger than that obtained without sulfanilamide. The polarization curves further verify that sulfanilamide added in the anode chamber indeed exerts positive effect on electricity generation at the studied concentration level with the improvement of the power output.

3.2. Electrode polarization analysis

As we know, the output voltage of the whole cell is determined by the anode potential and the cathode potential. In order to make it clear that whether the anode or the cathode plays the more important role in promoting the performance of the MFCs, individual electrode polarization behaviors are investigated in the present or absent of sulfanilamide. Results are depicted in Fig. 4, as 1 g L^{-1} glucose mixed with sulfanilamide of 0, 10, 20, and 30 mg L^{-1}, the anode open-circuit potential (OCPa, vs. SCE) are –398, –445, –457, and –488 mV. The OCPa moves toward more negative values from –398 mV (without sulfanilamide) to –488 mV (with addition of 30 mg L^{-1} sulfanilamide). Furthermore, decreased slope of the anode polarization curves can be found in MFCs with addition of sulfanilamide compared to those with no sulfanilamide added. With the increase in the current density from 0 to 600 mA m^{-2}, the anode potential of MFC without sulfanilamide increases by 24% from –398 to –263 mV, indicating that a larger overpotential would be required for the bioelectrochemical reaction at higher currents [24], while the anode potentials of the MFCs with 10, 20, and 30 mg L^{-1} sulfanilamide increase by 22, 19, and 14%, respectively. The lower anode overpotential with sulfanilamide reflects the positive role plays by sulfanilamide in enhancing the bioelectroactivity of the anode [25]. At the same time, the cathode open-circuit potentials and the cathode potential changing trends with the increase in the current density exhibit almost the same features. The results reflected by the cathode polarization curves are understandable because the cathodes and the cathodic solutions are exactly the same as each other. These results of the electrode polarization behaviors further reveal that the differences in the overall power output of MFCs originate from the anode rather than the cathode.

In MFCs, the anode potential is controlled by the kinetics of electron transfer from the micro-organisms to the anode. We speculate that the increase in voltage output or the lower anode overpotential with sulfanilamide could be attributed to the following factors. Firstly, sulfanilamide is likely to play an active role in accelerating the electron transfer from

electricity-generating bacteria to the anode, leading to a decreased anodic charge transfer resistance and then promoting the voltage output of the MFC. Researchers have reported that azo dye and its decolorization products could act as electron mediator for conveniently electrons' transfer from bacteria to the anode in MFCs [26,27]. It's well known that the decolorization products of azo dye are usually aromatic amine products resulted from the cleavage of the –N=N– bond under anaerobic conditions [28]. Sulfanilamide just possesses the aromatic amine structure. Therefore, we suppose that the addition of sulfanilamide probably results in the decreased electron transfer resistance from micro-organisms to the anode and then promoted the electricity generation. Secondly, the electricity-generating bacteria acclimatized in the biofilm on the anode probably present a high tolerance to sulfanilamide at these relatively low concentrations and the electrochemical activity of these bacteria may be not seriously inhibited.

Furthermore, the power output or the maximum voltage outputs are gradually increased with the increase in sulfanilamide concentration. Based on the above discussion, it is not difficult to deduce that more sulfanilamide is added in the anode chamber, the electrons could more conveniently transfer from bacteria to the anode, resulting in higher output voltage. But the maximum output voltage does not increase in exact proportion to the concentration of sulfanilamide. It is probably due to the accumulated toxicity of sulfanilamide which would produce an inhibition on electrochemical active bacteria and consequently lead to negative impact on electricity generation. We deduce that the observed voltage changing trend is probably the result of the joint effect of the two opposite effects.

3.3. Synthetic sulfanilamide wastewater treatment performance in MFCs

In order to find out the rudimentary removal mechanism and the essential factors of sulfanilamide removal in MFCs, four groups of comparison experiments, including normal MFCs, open-circuit MFCs, no co-substrate MFCs, and abiotic MFCs, are conducted at an initial sulfanilamide concentration of 10, 20, and 30 mg L^{-1}. As sulfanilamide concentrations are 10, 20, and 30 mg L^{-1}, the removal efficiencies of the comparison experiments show the similar characteristics and changing trends. Therefore, only the curves at sulfanilamide concentrations of 30 mg L^{-1} are plotted.

As is clearly shown in Fig. 5, in normal MFCs, sulfanilamide removal efficiency reaches 83% after 48 h and 90% is achieved as prolonging the reaction time to 96 h, while the removal efficiencies are 58, 10.8, and 7.5% in open-circuit, no co-substrate, and abiotic MFCs, respectively. Firstly, the low removal efficiencies in no co-substrate and abiotic MFCs indicate that sulfanilamide removal originated from physical adsorption, including the adsorption onto the microbial cells, the anaerobic sludge, and the components of MFCs reactors, may be very limited. Most of the removed sulfanilamide is probably stemmed from the biocatalytic co-metabolism degradation with co-substrate and micro-organisms as the two essential elements. Secondly, compared with sulfanilamide removal in open-circuit control, the accelerated removal in normal MFC reveals that the current between the anode and the cathode is favorable to the sulfanilamide removal in MFCs. The continuous electron transfer from the anode to the cathode leads to the more efficient and faster substrate metabolism than that in open-circuit control, which is apparently beneficial to sulfanilamide removal. The results are consistent with the results reported in literatures [29,30].

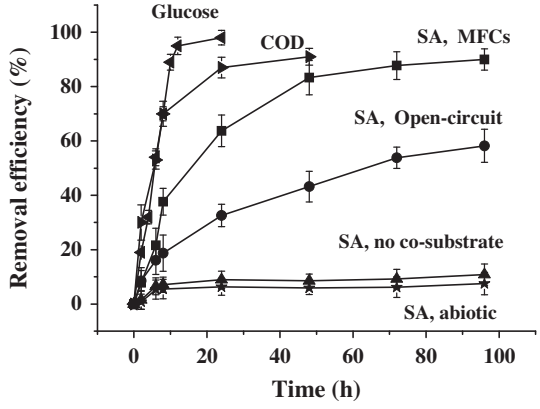


Fig. 5. Removal performances of sulfanilamide (30 mg L^{-1}) in normal MFCs, open-circuit MFCs, no co-substrate MFCs, and abiotic MFCs (SA is short for sulfanilamide); glucose and COD removal performance in normal MFCs.

The synthetic sulfanilamide wastewater treatment performance is also evaluated by the removal of COD of the anodic solution with 30 mg L^{-1} sulfanilamide and 1 g L^{-1} glucose as mixed substrates. As is clearly shown in Fig. 5, COD of the anode solution is efficiently removed 87% during 24 h, and slowly ascends to 90% in 48 h. The easily degradation co-substrate glucose can be quickly and almost completely consumed during each cycle and the removal efficiency achieves 95% during 12 h. Another noticeable phenomenon is that the changing trends of removal curve of COD and that of glucose are very similar in 8 h during each cycle test.

This phenomenon suggests that glucose degradation may mainly contribute to the COD removal we tested. It is probably due to that the concentrations of sulfanilamide added in the anode solution (the maximum concentration is 30 mg L^{-1}) are much lower than that of glucose (1 g L^{-1}), so the COD of the anode solution is predominantly composed by the co-substrate glucose. Thus, the degradation efficiency of glucose determines the COD removal efficiency of the whole anode solution. While the residual COD may be due to both the intermediates resulting from the co-metabolism degradation of sulfanilamide under anaerobic condition and the sulfanilamide not removed. Therefore, growing attention need to be paid to understand the degradation mechanism and to realize the complete mineralization of sulfanilamide in MFCs in our future work.

4. Conclusions

It is feasible to realize the simultaneous sulfanilamide removal and bioelectricity generation in MFCs. Sulfanilamide shows the positive effects on the electrical energy generation of MFCs. In comparison with MFCs using 1 g L^{-1} glucose as the sole substrate, the peak output voltage is increased by 15.7% for 1 g L^{-1} glucose and 30 mg L^{-1} sulfanilamide as mixed substrates. The electrode polarization behaviors further reveal that the differences in the overall power output of MFCs originate from the anode rather than the cathode. Compared with sulfanilamide removal efficiencies in open-circuit (58%), no co-substrate (10.8%), and abiotic MFCs (7.5%), accelerated sulfanilamide removal is achieved in normal MFCs, reaching 90% in 96 h. In MFCs, sulfanilamide removal is probably stemmed from the biocatalytic co-metabolism degradation and the current generation between the anode and the cathode plays a favorable role. But the residual COD of the anodic solution points out that growing attention needs to be paid to understand the degradation mechanism and to realize the complete mineralization of sulfanilamide in our future work.

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