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Research article

Simultaneous electricity production and antibiotics removal by microbial fuel cells

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ABSTRACT

The removal of antibiotics is crucial for improvement of water quality in animal wastewater treatment. In this paper, the performance of microbial fuel cell (MFC) in terms of degradation of typical antibiotics was investigated. Electricity was successfully produced by using sludge supernatant mixtures and synthesized animal wastewater as inoculation in MFC. Results demonstrated that the stable voltage, the maximum power density and internal resistance of anaerobic self-electrolysis (ASE) −112 and ASE-116 without antibiotics addition were 0.574 V, 5.78 W m^{-3} and 28.06 Ω, and 0.565 V, 5.82 W m^{-3} and 29.38 Ω, respectively. Moreover, when adding aureomycin, sulfadimidine, roxithromycin and norfloxacin into the reactors, the performance of MFC was inhibited (0.51 V–0.41 V), while the output voltage was improved with the decreased concentration of antibiotics. However, the removal efficiency of ammonia nitrogen (NH_3-N) and total phosphorus (TP) were both obviously enhanced. Simultaneously, LC-MS analysis showed that the removal efficiency of aureomycin, roxithromycin and norfloxacin were all 100% and the removal efficiency of sulfadimidine also reached 99.9%. These results indicated that antibiotics displayed significantly inhibitions for electricity performance but improved the quality of water simultaneously.

1. Introduction

Antibiotics, one of the most important series of pharmaceuticals in human and veterinary medicine (Baran et al., 2011; Kemper, 2008), are widely utilized around the world as additives in the animal industry to promote the growth of animals. The amount of antibiotic consumption worldwide is significantly huge and still increasing due to global economic and population growths (Cheng et al., 2014), which is close to 100,000–200,000 tons per annum (Wise, 2002). The issues associated with the frequently adopted antibiotics are their potentially side-effect on human-beings. Liver damage (Jeon et al., 2008), yellowing teeth and gastrointestinal disturbance may be caused by antibiotics of tetracyclines, even at low concentrations (Bjornsson et al., 1997). Allergic reactions in hypersensitive individuals are produced as well. Additionally, their consumption for long periods resulted in drug resistance (Macheboeuf et al., 2006). Seriously, antibiotics which applied in the prevention and treatment of diseases (Kemper, 2008) have been frequently detected in multiple aquatic environments. Thus, aquatic environment as well as its related environmental issues and public health problems remain a serious environmental problem (Kuemmerer, 2009). Therefore, adopting appropriate measures to resolve antibiotics pollution issues especially in animal wastewater is urgent.

At present, the understanding of the pollution resources ranged from sanitary wastewater (Taebi and Droste, 2004), brewery wastewater (Feng et al., 2008) to industry wastewater (Hachem et al., 2001; Manohar et al., 2002) has been distinctly classified, yet the study on the antibiotics existed in the animal wastewater was limited. The animal wastewater which contains high strength and complex ingredients are difficult to be treated (Kim et al.,

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2008). Moreover, the existence of antibiotics undoubtedly poses a great challenge to treat such kinds of wastewater through traditional methods. Additionally, large amounts of energy for aerating are consumed in the way of aerobic treatment, on the other hand, anaerobic treatment is considered as a suitable way for high-strength wastewater streams especially produced by industry (Zhang et al., 2000).

Numerous methods such as biological treatment (Arikan, 2008), chlorination (Adams et al., 2002; Navalon et al., 2008), advanced oxidation technology (Dantas et al., 2008), electrochemical treatment (Hirose et al., 2005; Jara et al., 2007), adsorption (Ma et al., 2012), the membrane process (Koyuncu et al., 2008), and the ultrasonic cavitation effect method (Hou et al., 2012) are being used to remove antibiotics, but, the effects of these methods are still not ideal and they are difficult to apply in practical applications. Fortunately, microbial fuel cells (MFC), an emerging environmental-friendly technology (Logan et al., 2006), is capable to utilize microorganism to convert organic matter and generate electricity at the same time. Hao et al. (2016) invested that MFC could reduce V(V) and generate bioelectricity simultaneously. Wang et al. (2016) demonstrated that sulphamethoxazole could be effectively degraded by MFC and MFC may enhance the degradation of refractory pollutants in environment. With the development of MFC technology, the research of MFC has been gained much publicity involving the electrodes (Wei et al., 2011), structures and microorganisms (Luo et al., 2009; He and Angenent, 2009). But, there are limited information available regarding removing pollutants like antibiotics by MFC, which enriched varieties of microorganism and electricity.

As known, aureomycin and sulfonamide are difficult to be absorbed and decomposed by animals, and most of them are discharged into the environment (Spielmeyer et al., 2015). Roxithromycin has a long half-life, it is refractory in the environment, and half of the annual production of norfloxacin are generally used for breeding pigs in China. The above antibiotics containing wastewater can pollute the environment with the discharge of sewage (Kim et al., 2013). Therefore, the objective of this paper is to establish the behavior between the antibiotics removal and the performance of MFC. Such knowledge is expected to provide solution to complex contaminations and evaluate the efficiency of MFC. In order to determine the effect of extra pollution matter-antibiotics, in situ, we successfully initiated the MFC with wastewater, simulated with the animal wastewater as substrates when the bacterial biofilm was generated on the anode. After the optimum conditions were recognized, different approaches were adopted to explore the relationship between antibiotics and the performance of MFC. These included: (1) The contamination (chemical oxygen demand (COD), total phosphorus (TP), total nitrogen (TN), nitrate nitrogen (NO_3-N) and ammonia nitrogen (NH_3-N)) removal efficiency of whole MFC devices; (2) The antibiotic removal condition during the operation of MFC; and (3) The probable removal mechanism of antibiotic by MFC devices.

2. Materials and methods

2.1. Addition of antibiotics at different stages

2.1.1. Different kinds of antibiotics addition

MFC start-up and operation was provided as Supplementary material. Following by the constant operation of the MFC with simulated animal wastewater, the feed to the reactor was changed to a medium containing different kinds of antibiotics (aureomycin, sulfadimidine, roxithromycin and norfloxacin) in the form of overlying. Denoted A stage as simulated animal wastewater without antibiotics additives, B stages as aureomycin ($18\ \mu g\ L^{-1}$) sole additive, C stage as aureomycin ($15\ \mu g\ L^{-1}$) and sulfadimidine ($10\ \mu g\ L^{-1}$) additives, D stage as aureomycin ($3\ \mu g\ L^{-1}$), sulfadimidine ($2\ \mu g\ L^{-1}$) and roxithromycin ($1.2\ \mu g\ L^{-1}$) additives, and E stage as aureomycin ($3\ \mu g\ L^{-1}$), sulfadimidine ($2\ \mu g\ L^{-1}$), roxithromycin ($1.2\ \mu g\ L^{-1}$) and norfloxacin ($1.2\ \mu g\ L^{-1}$) additives.

2.1.2. Different concentrations of antibiotics addition

To determine continuous effect of different concentrations of antibiotics on MFC, the performances of the electrical as well as pollutants removal indexes in the presence of decreasing concentration were observed. F ~ H stage was classified as different concentration of antibiotics, which were F: aureomycin ($45\ \mu g\ L^{-1}$), sulfadimidine ($30\ \mu g\ L^{-1}$), roxithromycin ($18\ \mu g\ L^{-1}$) and norfloxacin ($18\ \mu g\ L^{-1}$), G: aureomycin ($15\ \mu g\ L^{-1}$), sulfadimidine ($10\ \mu g\ L^{-1}$), roxithromycin ($6\ \mu g\ L^{-1}$) and norfloxacin ($6\ \mu g\ L^{-1}$), and H: aureomycin ($3\ \mu g\ L^{-1}$), sulfadimidine ($2\ \mu g\ L^{-1}$), roxithromycin ($1.2\ \mu g\ L^{-1}$) and norfloxacin ($1.2\ \mu g\ L^{-1}$).

2.2. Calculation and analysis

2.2.1. Electricity analytical methods

The cell voltage (U) was recorded by using a data acquisition system (2700, Keithly). Current density ($A \cdot m^{-2}$) was calculated as $I = U/(RS)$, where S is the surface area of the anode ($6.28\ cm^2$) and R is the external resistance. Power density ($mW \cdot m^{-2}$) was calculated as $P = 1000U^2/(RS)$ (1000 is used for unit conversion). Polarization Curves were plotted by changing the external resistance over a range from $90000\ \Omega$ to $10\ \Omega$. The internal resistance (r) was determined by using steady state discharge method, which was measured by changing different external resistance.

2.2.2. Wastewater quality analytical methods

COD of anaerobic self-electrolysis (ASE) influents and effluents were measured according to the manufacture's instruction using COD digester (XJ-III, Shaoguan Tomorrow Environmental Protection Instrument Co., Ltd, China) and spectrophotometer (UV-759, Shanghai Jingke Scientific Instrument Co., Ltd, China). Nitrogen (NH_3-N, TN and NO_3-N) were taken regularly and analyzed according to the previously reported. pH was measured with a pH S-3C digital meter (Shanghai Leici Instruments Factory, China). All samples were analyzed in duplicate and mean values were presented.

2.2.3. Qualitative and quantitative for antibiotics analytical method

LC-MS was employed for the analysis of antibiotics including aureomycin, sulfadimidine, roxithromycin, norfloxacin. The LC-MS system consisted of an Agilent 1200 RRLC/6410B Triple Quard MS. The software Masslynx 4.1, also from Micromass (Manchester, UK), was employed to acquire the data and control the system. The analysis of the antibiotics was performed by using a Sunfire C18 column (150 × 2.1 mm i.d., 5.0 μm particle size) from Waters (MA, USA). For the pretreatment, the analytes were extracted with EDTA, acidified at dilute sulphuric acid, followed by enrichment extraction with cartridges before screening through 0.45 μm filter membrane. Afterwards, ultrapure water was used to rinse and the collected eluent was gathered through 0.22 μm filter membrane after nitrogen blowing instrument. Consequently, the final extract was injected to LC-MS for further detection.

3. Results and discussion

3.1. Power generation using artificial swine wastewater

In this research, the developed cylindrical MFC was qualified to continuously produce electricity from the degradable matter