



Performance of a microbial fuel cell-based biosensor for online monitoring in an integrated system combining microbial fuel cell and upflow anaerobic sludge bed reactor



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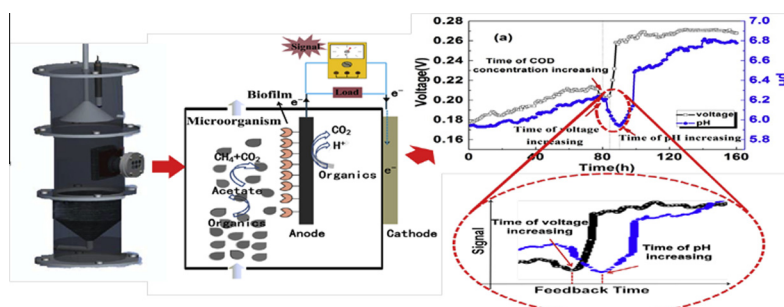
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HIGHLIGHTS

- A novel MFC-UASB system was established for real-time online monitoring rapidly and steady.
- Better signal feedback sensitivity and reproducibility were achieved when COD concentration changed.
- The MFC-based biosensor through the electric signal feedback on the concentration was more sensitivity than by pH.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 10 May 2016

Received in revised form 15 June 2016

Accepted 16 June 2016

Available online 18 June 2016

Keywords:

Upflow anaerobic sludge blanket

Microbial fuel cell

Biosensor

Signal feedback

Sensitivity

ABSTRACT

A hybrid system integrating a microbial fuel cell (MFC)-based biosensor with upflow anaerobic sludge blanket (UASB) was investigated for real-time online monitoring of the internal operation of the UASB reactor. The features concerned were its rapidity and steadiness with a constant operation condition. In addition, the signal feedback mechanism was examined by the relationship between voltage and time point of changed COD concentration. The sensitivity of different concentrations was explored by comparing the signal feedback time point between the voltage and pH. Results showed that the electrical signal feedback was more sensitive than pH and the thresholds of sensitivity were $S = 3 \times 10^{-5} \text{ V}/(\text{mg/L})$ and $S = 8 \times 10^{-5} \text{ V}/(\text{mg/L})$ in different concentration ranges, respectively. Although only 0.94% of the influent COD was translated into electricity and applied for biosensing, this integrated system indicated great potential without additional COD consumption for real-time monitoring.

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1. Introduction

Due to obvious advantages in removing pollutants and recovering energy, upflow anaerobic sludge blanket (UASB) reactors have become widely used for wastewater treatment in the last decade

(Pant et al., 2010). The UASB reactor has some special characteristics in treating wastewater. Previous studies found that its ability to remove COD was superior to other biological treatment processes and the advantage of lower cost and less sludge production (Jing et al., 2013; Lu et al., 2015, 2016). So far, numerous studies have focused on the applicability of the UASB process to various industrial and municipal wastewaters (Onodera et al., 2014). However, the anaerobic microorganism acted as the core of operating. It was both complex and harsh for environmental

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conditions and sensitive to the changes of water quality and/or organic loading shock. Therefore, the absence of reliable dynamic information and complete online monitoring methods for anaerobic digestion are the current obstacles in applying the UASB process. Although the sensors are critical to provide stable and effective precaution strategies for the anaerobic reaction process, the biosensor performance is affected as the extension of running time, which cause inherent problems, such as equipment corrosion, signal fuzzy, and deviation for date analysis. To date, only very limited work has been undertaken to assess the UASB system's operating conditions.

Microbial fuel cell (MFC) is a type of biological electrochemical device subject to intensive investigation, and the last three decades have witnessed significant developments in the environmental and energy field (Li et al., 2011). MFCs are able to convert chemical energy into electricity directly via bioprocesses catalyzed by exoelectrogenic microorganisms (Logan et al., 2006a,b; Lovley, 2006; Srikanth and Venkata Mohan, 2012). Therefore, due to the possibility of wastewater treatment and utilization of the electrical signals for biological sensing simultaneously. MFCs are considered as prosperous applications for facilitating sustainability. Since voltage can be monitored easily online, MFC has the ability to function as an inexpensive on-line biosensor that operates in a steady way and is constructed with low-cost material (In Seop et al., 2005; Kim et al., 2003) where microorganisms in the anode compartment act as biological recognition element whereas electrodes and proton exchange membrane (option) serve as a transducer (Kumlanghan et al., 2007). As a consequence such an on-line MFC-based biosensor measured an electrical voltage as the signal, does not need any mediation to convert it into a signal (Feng et al., 2013). Compared to other kinds of biosensors, the main advantages of MFC are miniaturization, portability and real-time monitoring (Di Lorenzo et al., 2009).

A number of researchers have used biosensor as a converter utilized by weak voltage for monitoring water quality in real time (Xu et al., 2015) and a comparative table (Table S1) showed the significance of the achievements in the light of already published literature. For instance, Dual-chamber microbial fuel cells were employed for energy valorization of an untested substrate, the liquid fraction of pressed municipal solid waste (LPW). It can be a useful, direct tool to access the impact of process disturbances though the time-profile of bioelectricity production monitoring in bioelectrochemical systems (Kook et al., 2016). Zhang and Angelidaki (2012) developed a submersible microbial fuel cell (SBMFC) as a biosensor for real-time monitoring of dissolved oxygen (DO) in water based on the electricity principle of MFC. Di Lorenzo et al. (2009) developed a Biochemical Oxygen Demand (BOD₅) biosensor fueled with artificial water. A linear relationship between MFC power output and BOD₅ concentration was established, indicating the applicability of real-time monitoring for BOD_{5b} (Di Lorenzo et al., 2009). A new wall-jet MFC sensor was designed by Liu et al. (2011) for the monitoring of anaerobic digestion process by the real-time detection of acetate based intermediates (Liu et al., 2011). A simple compact membrane MFC (MMFC) sensor by stacking two flat filter membranes without the proton exchange membrane (PEM) and paper reservoir was developed (Xu et al., 2015). The unique flat structure of MMFCs makes the direct installation on wastewater facilities and serves as an "on line sticker sensor" for in-situ real time wastewater quality monitoring with sensitivity and stability. Quek et al. (2015) developed an online low assimilable organic carbon (AOC) detection system for oxygen-saturated seawater by combining a suitably designed electrochemical oxygen removal cell with a MFC-biosensor. The coupling of an electrochemical cell with a MFC-biosensor can be effectively used as an online, rapid and inexpensive measure of AOC concentrations and as an indicator for biofouling potential of seawater (Quek et al., 2015). The subjects of variation of the

current reflecting the toxic compounds in substrates were studied by other MFC-based biosensor studies (Jiang et al., 2015; Shen et al., 2013). However, most analyses of MFC-based biosensor did not discuss the performance of biosensor in detail.

Based on the above issues, this research aimed to develop a novel monitoring system for the MFC-UASB process, which consisted of a single-chamber MFC where the UASB reactor worked as an anode chamber and the carbon cloth worked as an air-cathode. The objective of the series tests in this research is to investigate the performance of the MFC-based biosensor between different COD concentrations. Furthermore, optimum conditions are applied for studying the performance of the biosensor at various hydraulic retention times (HRTs) and external resistance to achieve a suitable condition for sensing. The development of a MFC-UASB system will provide a feasible method for UASB monitoring.

2. Material and methods

2.1. Integrated MFC-UASB system

The MFC-UASB was made of a plexiglass cylinder that consisted of an anode and cathode (Fig. S1). The UASB reactor worked as an anode chamber (internal diameter of 20 cm and height of 100 cm) with a working volume of 10 L. At the top of the reactor. A flow meter (LMF-1, Changchun Lvqingqi Co., Ltd., China) was connected to the gas outlet of the three-phase separator in the UASB. The MFC's structure was a single chamber without a proton exchange membrane (PEM) in which a piece of carbon felt that had an effective surface area of 64 cm² (8 cm × 8 cm) and 5-mm thick (Beijing Fengxiang Co., Ltd., China) served as an anode electrode and carbon cloth that had an effective surface area of 50.24 cm² with diameter was 8 cm and 1.5-mm thick (HCP330N, Shanghai Hesens Co., Ltd., China) prepared as an air cathode electrode, respectively. Air was continuously sparged inside the cathode and functioned as the final electron acceptor. The MFC-UASB was operated by feeding the wastewater through the bottom of the UASB using a peristaltic pump (YZ1515, Tianjin Xieda Tech Co., Ltd., China). The electrode assembly was then submerged in the UASB reactor. The electrical circuit connected between the anode and cathode was fabricated by copper wires with a resistance box of 0–9999 Ω and the voltage across resistor was collected by a multimeter.

The cathode was prepared according to the procedure that is briefly described below. Carbon cloth was placed in 5% polytetrafluoroethylene (PTFE) solution soaked for 10 min and then put in a muffle furnace (SX-GO5163, Tianjin Zhonghuan Co., Ltd., China) calcination for 30 min at a temperature of 330 °C. A mixture of carbon powder (Shanghai Hesens Co., Ltd., China) and 40% PTFE solution were then applied to one side of the carbon cloth, air dried at room temperature for 2 h, followed by heating at 330 °C for 30 min. Additional brushing 60% PTFE solution onto the coating side, followed again by dyeing at room temperature and heating at 330 °C for 30 min as Cheng described (Cheng et al., 2006). Pt catalyst (HPT010, Shanghai Hesens Co., Ltd., China) was then applied to the other side of the carbon cloth, and this has been previously described using Nafion (D520, Shanghai Hesens Co., Ltd., China) as a binder (Cheng et al., 2006).

2.2. Operating conditions

The anaerobic active sludge was taken from Jizhuangzi Wastewater Treatment Plant (Tianjin, China). It was then injected as an inoculum into the anode chamber after domestication that last for 30 days. To control the anode chamber in an anaerobic state, the nitrogen stripping method was used to eliminate all dissolved

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