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HIGHLIGHTS

• A new type of bio-cathode MFC-based biosensor was developed for COD monitoring.

• The performance of bio-cathode MFC-based biosensor was affected by HRT, MLSS and COD.

• The range of COD concentrations detected by biosensor was increased.

• The MFC-TMBR system produced electricity for biosensing while organic was degraded.

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ABSTRACT

A bio-cathode microbial fuel cell (MFC) with tubular membrane was integrated to construct a microbial fuel cell-tubular membrane bioreactor (MFC–TMBR) system, in which the bio-cathode MFC was developed as a biosensor for COD real-time monitoring in TMBR and the performance was analyzed in terms of its current variation caused by operation parameters. With a constant anode potential, the effect of HRT demonstrated that higher rate of mass transport increased the response of the system. The system was further explored an inverse relationship between TMP and current peak by using EPS concentration under the different MLSS concentration. The sensor output had a linear relationship with COD up to 1000 mg/L (regression coefficient, $R^2 = 0.97$) and MLSS (regression coefficient, $R^2 = 0.94$). The simple and compact bio-cathode MFC biosensor for TMBR using MFC–TMBR integrated system showed promising potential for direct and economical COD online monitoring, and provided an opportunity to widen the application of MFC-based biosensor.

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1. Introduction

Microbial fuel cell (MFC) is a bio-electrochemical device which use microorganisms as catalyst for converting chemical energy into electrical energy (Logan, 2008). MFC consists of an anode exposed to an electron donor, and a cathode exposed to a terminal electron accepted. The two chambers are typically separated by a permeable membrane. As current can be monitored easily on line, MFC can act as an inexpensive online biosensor which can be operated steady (Chang et al., 2004, 2005; Kim et al., 2003a,b) with naturally-occurring bacteria and constructed with low-cost materials. In the general scheme of a biosensor, the biological recognition element responds to the target chemicals and the transducer converts the biological response to a detectable signal, which can be measured electrochemically, mechanically, acoustically, or optically (Su et al., 2011). Such an online MFC-based biosensor measured with an electrical current as the signal, which does not need a mediation to read the signal or convert it to an electrical signal. Various types of MFC-based biosensors have been reported (Di Lorenzo et al., 2009; Stein et al., 2010; Feng et al., 2013), showing that it has a major advantage over many other types of biosensors (Hyun et al., 1993; Chee et al., 2005).

A number of researchers have used biosensors to monitor water quality (e.g. Lee et al., 2005, 2007; Kim et al., 2007;





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Di Lorenzo et al., 2009). Online MFC-based biosensors are widely used to monitor water quality of water flows like food wastewater, drinking water or processing water, or used to be integrated with artificial neural networks (ANNs) to identify specific chemicals present in water samples. The relationship between water quality and MFC signals depends on the physical parameters of the MFC, operating conditions (e.g. pH, temperature, organic loading rate and electrical load), as well as the ecology of the biofilm. There are some direct evidences that MFC-based biosensor can be used to monitor water quality in real time. For example, Di Lorenzo et al. found out a linear relationship between chemical oxygen demand (COD) concentration and MFC current output (Di Lorenzo et al., 2009), demonstrating the applicability of this system to real-time COD monitoring. Feng et al. demonstrated that the peak area generated by the electrical signals correlated well with the chemical oxygen demand (COD) present in water samples (Feng et al., 2011). Kumlanghan et al. developed a biosensor based on a single-chamber microbial fuel cell, and the current study showed the development of a MFC sensor system for fast estimation of easily biodegradable organic matter in which glucose was used as the only preliminary substrate (Kumlanghan et al., 2007). Zhang and Angelidaki developed a submersible microbial fuel cell (SBMFC) as a biosensor for real-time monitoring of dissolved oxygen (DO) in environment waters (Zhang and Angelidaki, 2012). Tront et al. explored a MFC sensor to in suit monitoring of substrate concentration, showing that current generation was mirrored by acetate concentration, and a correlation $(R^2 = 0.92)$ was developed (Tront et al., 2008). Similar results have been retrieved for other MFC-based biosensor researches (Chang et al., 2004, 2005; Shen et al., 2012; Modin and Wilen, 2012). However, most previous studies of MFC-based biosensor have not discussed the practical application in detail.

Di Lorenzo et al. suggested that the MFC-based biosensor could be installed between the clarifier and the aeration tank to determine the quality of the effluent in terms of COD content after the primary clarifier in a wastewater treatment plant (Di Lorenzo et al., 2009). In this study, compared with this design, a MFC-based biosensor for membrane bioreactor (MBR) was developed, in which the aeration tank of a MBR was directly used as cathode chamber. Using bio-cathode MFC combined with tubular membrane, a combined system of microbial fuel cell and tubular membrane bioreactor (MFC-TMBR) for biosensor and wastewater treatment appeared to be more attractive in terms of costs and the practical application (Wang et al., 2013). The electrical signals (e.g. electrical current) generated by the bio-cathode MFC were thus a direct linear measure for metabolism of the electrochemically active microorganisms under the condition of the stability of anode potential. The aim of the series of tests in this study is to investigate the interrelationship between MFC electricity generation and tubular membrane bioreactor (TMBR) operation parameters under different operation conditions. The bioelectricity performance of MFC-TMBR system was evaluated in terms of its chemical oxygen demand (COD) range, loading rate, mixed liquor suspended solids (MLSS) concentration, extracellular polymeric substances (EPS) concentration and transmembrane pressure (TMP). A microbial fuel cell-based biosensor for monitoring COD in tubular membrane bioreactor was also investigated, which could be potentially useful for a wider COD concentration range. In this paper, we demonstrate the new type of COD sensor for on-line monitoring TMBR operation using a bio-cathode MFC-based biosensor. The development of MFC-TMBR system will provide an advantageous sensor design for COD monitoring and expand the application of the MFC technology.

2. Methods

2.1. Integrated MFC–TMBR system

The MFC-TMBR system was constructed with a bio-cathode MFC and single tubular membrane, as shown in Fig. 1a. The ultrafiltration membrane (PES: MWCO = 30 k; Shanghai SINAP Membrane Tech Co., Ltd., China) separated the anode and the biocathode. The two chambers (2.5 L, $10 \text{ cm} \times 10 \text{ cm} \times 25 \text{ cm}$) were filled with 2.2 L liquid. The anode and cathode electrode each was a piece of carbon felt (126 cm², Beijing Honghaitian Tech Co., China) without any pretreatment. The cathode chamber was directly used as the aeration tank. The anode and cathode were connected through a 1000 Ω resistor. The cell voltage and electrode potential were recorded automatically every 5 min using a data acquisition system (USB-FS1208, Measurement Computing™, America). For the TMBR section, the single tubular membrane (10 cm, polyvinglidene fluoride with an average pore size of 0.1 µm, Tianjin Motimo Membrane Tech Co., China), as shown in Fig. 1b, with a filtration area of 37.68 cm² was placed out of the aeration tank. Aeration unit was placed at the bottom of the chamber and the aeration flow rate was 78 L/h. Peristaltic pumps (BT100-2], Baoding Langer pump company, China) rotating at different revolutions per minute with different flux were used to conduct the control of constant flow for drawing out effluents, and the hydraulic retention time (HRT) had a corresponding change. The TMP was measured by pressure sensors (Danfoss, MBS 3000, Denmark) for monitoring membrane fouling. Tubular membrane was washed with sodium hypochlorite (0.5%) for 1 h and back-flushed with tap water for 3 h when TMP reached -50 kpa.

2.2. Operating conditions

4.4 g/L KH₂PO₄, 3.4 g/L K₂HPO₄, 0.5 g/L NaCl, 0.1 g/L NH₄Cl, 0.05 g/L MgCl₂, and 0.05 g/L CaCl₂ were dissolved in tap water to synthesize simulative influent wastewater for the whole experiment (Shan et al., 2011). The COD concentration of the artificial water in the cathode chamber was controlled by adding an appropriate amount of glucose, and the COD concentration of the artificial water in the anode chamber was 1000 mg/L consistently. The aerobic active sludge was taken from Jizhuangzi Wastewater Treatment Plant (Tianjin, China). A 1000 ml bacterial suspension collected from laboratory bioreactors was injected as inoculum into the anodic zone. Copper wire was used to connect the cathode and anode electrode, respectively. After reaching a stable status for about 200 days, the MFC finished executing and the coupled system began to work. To achieve the constant anode potential, the MFC-TMBR system was operated at a room temperature of 25 ± 3 °C, a pH of 7 and a substrate of glucose (1 g/L in anode chamber).

During the tests, the MLSS concentration of the aeration tank was varied from 3400 mg/L to 12240 mg/L. Due to the cultivation of the sludge, the MLSS concentration was increased step by step. The method of maintaining the MLSS concentration at each stage test was sludge wasting, which meant that about 100 ml mixed sludge liquor was discharged from aeration tank every day.

2.3. Electrochemical and chemical measurements

The polarization curves of MFC were obtained by varying the circuit external resistance from 10 to 10,000 Ω when the cell voltage of the MFC was relatively stable. The working potential of the electrodes was measured against a reference electrode (type of

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