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Enhancing simultaneous response and amplification of biosensor in microbial fuel cell-based upflow anaerobic sludge bed reactor supplemented with zero-valent iron

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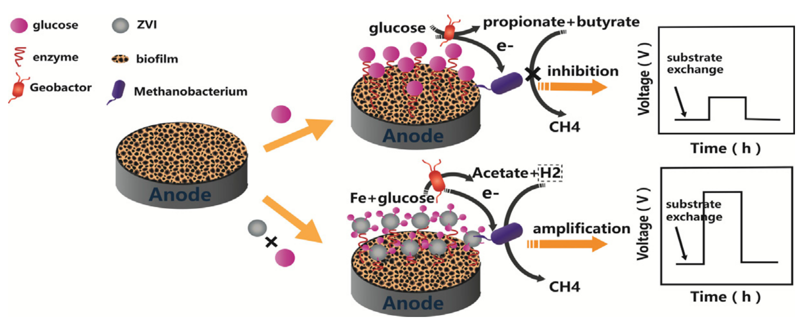
HIGHLIGHTS

- A new manner of ZVI based MFC-biosensor was developed for the properties of UASB-MFC biosensor accumulating.
- The electrical signal response and amplified properties were affected by electron transport and pH of system.
- ZVI enhanced the direct interspecies electron transfer through intensified anaerobic digestion.
- The effects of volatile fatty acid component proportion on the exoelectrogen and biosensing activities were revealed.

GRAPHICAL ABSTRACT



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ABSTRACT

The development of a convenient and sensitive sensor such as a microbial fuel cell (MFC) to monitor the operation of upflow anaerobic sludge blanket (UASB) is indispensable. However, the biosensor's properties were affected due to excessive acidification and suffocation of the electron transport. In this study, zero-valent iron (ZVI) was applied to restrain excessive acidification and improve the sensing performance. According to the results, the response rate of electrical signal accumulated with the addition of ZVI compared to the control reactor. As well as the electrical signal amplified and the subsidence rate maximum reached 0.059 V/h with 30 mg/L ZVI added that 883% higher than the control one during the stage (COD concentration 500 mg/L–1000 mg/L). With the electrochemical analysis, the internal resistance of ZVI-UASB-MFC decreased and redox activity promoted effectively with ZVI added. During the overloading phase, the fractional content of butyric acid changed from 53% to 31%, while that of acetic acid rose from 18% to 39% after 30 mg/L ZVI addition. These results indicated that adding ZVI to the digestion could retard excessive acidification by promoting butyric acid conversion and accumulating direct interspecies electron transfer simultaneous for enhancing the biosensor's performance. According to the Fe^{2+} and Fe^{3+} of effluent were 2.25 mg/L and 0.39 mg/L with 50 mg/L ZVI addition, moderate amount of ZVI was effective for system and safety to the environment. It might helpfully provide a promising way to enhance biosensing.

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1. Introduction

Currently, microbial fuel cell (MFC) devices that use bacteria as catalysts to oxidize organic or inorganic matter and generate current, are attracting increasing attention because they directly convert chemical energy into electricity in more than one step [1–3]. Due to the limitation of the theoretical electrogenesis voltage of the MFC and resultant possible losses, the maximum open circuit voltage of MFC is typically less than 0.8 V [4]. Even in the latest stacking MFCs in series can improve the voltage output [5]. However, the advantage gained by increasing the number of series-stacked MFCs may be offset by the concomitant electrical energy loss [6]. The MFCs inferior ability to generate power seriously restricts their application. Thus, utilized MFC as a sensor for pollutant detecting and in situ process monitoring is the potential application. Compared to other existing sensors, the MFC biosensor worked as a small bioreactor with characteristics of high sensitivity and long stability. For these reasons the MFC biosensor has received more attention and been used for Biochemical Oxygen Demand (BOD) [7], Dissolved Oxygen (DO) [8], toxicity [9], and Volatile Fatty Acid (VFA) sensing [10], etc.

Up-flow anaerobic sludge blanket (UASB) is an efficient biological approach for treating organic wastewater. However, the lack of reliable dynamic information and robust online sensors is one of the system's major problems. To achieve a more efficient and stable operation, the UASB-MFC biosensor has been investigated for real-time monitoring of the UASB process through the electrical signal converted by MFC in a recent study, specifically in an easy operating format [11]. Biodegradable organic matter directly converted to electricity via MFC, and MFC itself is an integration of signal generator and transducer, which reduces the costs associated with external transducers. A major disadvantage of the biosensor is its reliance on biofilm for mediator-less electron transportation. Once acidification appears with microorganisms' inactivation, this causes the electron transmission rate slow down and biosensor become insensitive. To improve the properties of the UASB-MFC biosensor, new methods that vastly improve the electron transport rate from the biofilm covering an anode to the cathode and inhibiting acidification are much needed.

Zero-valent iron (ZVI) is an economic, efficient, and environment friendly reductive material that has been used widely in wastewater treatment and hazardous substance stabilization [12]. In anaerobic digestion, ZVI can provide electrons due to its low oxidation-reduction potential (ORP) ($E_0 = -440$ mV) and serve as an acid buffer, which are crucial abilities that help maintain a stable and favorable state for methanogenic archaea, denitrifying bacteria, and sulfate reducing bacteria [13]. The most recognized mechanism of ZVI bio-availability is linked to the corrosion-induced H_2, which can be utilized to reduce CO_2 to methane by hydrogenotrophic methanogens [14]. The ZVI corrosion process in water releases Fe^{2+} irons, which contains a series of oxydrolysis and finally generates many types of hydrous ferric oxides. Since then, researchers have focused on utilizing ZVI (including nanoscale zero-valent iron (nZVI)) to treat pollution in the last decades which including dyes [15,16], nitrate [17,18], phenol [19,20], heavy metals [21,22], arsenic [23,24], nitroaromatic compounds [25,26], chlorinated organic compounds [27,28], and aliphatic compounds [29,30].

The combination of ZVI and anaerobic reactor has been investigated to facilitate the degradation of pollutants and been successfully applied to treat dairy wastewater [31]. One study confirmed that ZVI could significantly promote the diversity of microbial strains responsible for high efficiency in sludge [32]. Kaan Yetilmezsoy et al. [33] investigated the Fe-electrocoagulation technique for decolorization and chemical oxygen demand reduction of anaerobically pretreated poultry manure wastewater, about 90% COD and 92% of residual color be effectively removed. Cheng et al. [34] studied the pretreatment of concentrated wastewater from triazine manufactured by coagulation, electrolysis and internal microelectrolysis using iron chips. Wu et al. [12] observed that the moderate addition of ZVI can promote COD removal and methane production of the anaerobic system treated using swine wastewater. Liu et al. [35] reported that using a mathematical model, ZVI simultaneously enhances methane production and sulfate reduction in anaerobic granular sludge reactor.

Although many studies have documented that ZVI enhanced anaerobic digestion of various wastewaters, their effect on the MFC based biosensor has not been investigated. Considering its reductive properties, the main interest in applying ZVI in an anaerobic digestion-bioelectrochemical field using wastewater or sludge as substrate was to create an enhanced anaerobic environment. It may help improve the performance of an anaerobic reactor. In this context, a marriage of the UASB-MFC and ZVI technologies would potentially provide an efficient way of utilizing the iron released by ZVI which could transfer electrons to an electrode for improving biosensor properties. The objective of this research was to investigate the acceleration effects of ZVI on the biosensor's signal response at different organic loading rates (OLR) and explore the mechanism of electron transport process variation when ZVI is added. The evolution of VFA in acidified reactors as the response to ZVI addition was investigated to explore the process of the positive inhibition effect of ZVI.

2. Material and methods

2.1. Experimental setup

One kind of laboratory-scale microbial fuel cell-based up-flow anaerobic sludge blanket was set up (Fig. S1). The UASB-MFC reactor comprised a plexiglass cylinder consisting of an anode and cathode in which the UASB reactor worked as an anode chamber with a volume of 2.56 L (internal diameter of 6 cm and height of 50 cm). The structure of the MFC was a single chamber without a proton exchange membrane (PEM) as described in a previous study [11]. The electrode was submerged in the UASB reactor and the anode electrode was made of carbon felt, with an apparent surface area of 50 cm^2 and thickness of 5 mm (Beijing Fengxiang Co., Ltd., China). The air-cathode electrode was made of carbon cloth, with an apparent surface area of 50 cm^2 and thickness of 1.5 mm (HCP330 N, Shanghai Hesen Co., Ltd., China). The anode electrode was previously washed in 1 M HCl and 1 M NaOH to remove possible metal and organic contamination. The cathode electrode via the pretreatment according to previous research by Liu et al. [36]. The UASB-MFC was operated by feeding the wastewater through the bottom of the UASB using a peristaltic pump (BT100-2J, Baoding Longer Precision Pump Co., Ltd., China). Electrodes were connected to the data logging equipment via copper wires and titanium sheet with stainless steel crocodile clips under 0–9999 Ω controlled by a resistance box. Ag/AgCl electrode (3 M KCl, AgCl saturated, 0.210 V versus standard hydrogen electrode, 25 °C, Shanghai Leici Co., Ltd., China) was set in the flange of the reactor as a reference electrode for measuring electrode potential, and its distance from anode and cathode was 2 cm and 2 cm, respectively. A flow meter (LMF-1, Changchun Lvqingqi Co., Ltd., China) was connected to the gas outlet of the UASB. The system's operation temperature was thermostatically controlled at 26 ± 2 °C.

Five reactors were used in the experiments to study the changes in electrochemical characteristics caused by ZVI in microbial fuel cell-based up-flow anaerobic sludge blanket systems. One reactor