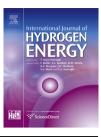


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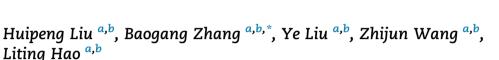
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# Continuous bioelectricity generation with simultaneous sulfide and organics removals in an anaerobic baffled stacking microbial fuel cell



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## ABSTRACT

The malodorous and toxic nature of sulfide governs its removal from wastewaters. An improved anaerobic baffled stacking microbial fuel cell (ABSMFC) was constructed for sulfide removal and energy recovery. With initial sulfide of 60 mg/L, chemical oxygen demand (COD) of 800 mg/L and hydraulic retention time (HRT) of 24 h, sulfide and COD removal efficiencies of 70.0% and 54.6% were achieved, respectively. The maximum voltage reached 2340 mV in series connection while the maximum current increased to 3.29 mA in parallel connection, with 1000  $\Omega$  external resistance. Initial sulfide concentration and HRT affected the performance of ABSMFC. Sulfide was mainly oxidized to elemental sulfur and sulfate. Molecular biology analysis indicated electrochemically activated bacteria, sulfur related bacteria and anaerobic fermentative bacteria interacted together. This study provides promising configurations of MFC to scale up for its actual applications in sulfide wastewater treatment.

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## Introduction

Sulfide is a widespread environmental contaminant resulting from human activities such as chemical production, food processing and petrochemical industry [1]. It is corrosive and toxic to organisms. Sulfide can be treated physically and chemically, while common physical-chemical methods, such as adsorption and chemical oxidation, are not so costeffective [2]. Nowadays, biological oxidation is most widely used as it provides an environmental-friendly alternative for sulfide removal in both liquid and gaseous phases [3,4].

In another aspect, microbial fuel cells (MFCs) are devices that can oxidize organics and recover electricity by using bacteria as catalysts [5–7]. They attract great attention nowadays due to the capability of simultaneous wastewater treatment and bioelectricity generation [8,9]. MFCs have also been employed for effective sulfide removal with bioelectricity recovery [10], while little concern have been showed in MFCs studies for simultaneous organics removal which is often

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presented together with sulfide in sulfide containing wastewater such as anaerobic digestion effluents [11]. Moreover, relatively lower power density of individual MFC restricts its widespread practical applications [12]. MFC units can be stacked together or linked together in series, resulting in an additive increase in total power outputs and treatment capacity [13]. Numerous studies have focused on MFC stacking and configuration improvements based on traditional biological treatment techniques [14–16]. Baffled stacking has been demonstrated to function well for energy harvest from wastewater such as high strength molasses wastewater [17], while behaviors of baffled stacking MFCs on simultaneous sulfide and organics removals with energy recovery have rarely been considered.

In the present study, a promising kind of MFC stacking based on anaerobic baffled reactor, i.e. anaerobic baffled stacking microbial fuel cell (ABSMFC) was developed and further improved and its ability of continuous sulfide and organics removals with bioelectricity generation was evaluated. Operating factors as initial sulfide concentration and hydraulic retention time (HRT) were investigated in terms of sulfide removal and electricity generation. Power outputs by connecting its units in series and parallel were studied, respectively. Products from sulfide oxidation and the involved bacteria were also examined. Results indicate that the improved ABRMFC is effective for sulfide and organics removals with bioelectricity generation, providing a new pathway to successful remediation of sulfide contamination.

## Methods and materials

## ABSMFC construction and solution conditions

The configuration of the proposed ABSMFC consisting of six cell units (U1, U2, U3, U4, U5, U6) in rectangular parallelepipedshape (internal dimensions 60.0 cm  $\times$  10.0 cm  $\times$  14.0 cm, 10 mm wall thickness) was made of plexiglass (Fig. 1), divided by five straight overflow plates (10 mm thickness) with different heights (9 cm, 8.5 cm 8 cm, 7.5 cm, and 7 cm, respectively) for overflow and to intercept the generated nonconducting elemental sulfur from sulfide oxidation. The chamber of each unit was separated into a down-flow part and an up-flow part (width ratio of 1:4) by a perpendicular hanging baffle (45° slant edge at the bottom), with a 10 mm distance to the inner bottom of the reactor. Carbon fiber felt (5 cm  $\times$  5 cm, 3 mm thickness) and 25 g graphite granules (diameter 3–5 mm) served as anode in each unit. The cathode was made of carbon paper (5 cm  $\times$  5 cm) with a surface loading of 0.5 mg Pt/cm<sup>2</sup> on the water-facing side. It was bonded to a current-collecting screen made of copper mesh. Each pair of electrodes in each unit was connected to a data acquisition system (PMD1608LS, MCC Corporation, USA) for voltage measurement, with 1000  $\Omega$ external resistance. The influent was pumped into the reactor by pump and flowed from the water entrance through U1 to U6 in turn and then discharged. Each unit was inoculated with 50 ml anaerobic sludge collected from an anaerobic digester in sewage treatment plants in Shandong province, China. The influent contained sulfide and organics with given concentrations in the form  $C_6H_{12}O_6$  and  $Na_2S \cdot 9H_2O$ , respectively. All experiments were conducted at room temperature ( $22 \pm 2$  °C). Each test was repeated three times and their average results were reported.

### Experimental procedures

After inoculation, each unit was filled with solution containing sulfide of 60 mg/L and chemical oxygen demand (COD) of 800 mg/L under close circuit condition. The solution was refreshed each 72 h for domestication and this lasted about one month. Then the ABSMFC operated in continuous flow mode with influent as mentioned above. This lasted about two months for further domestication. When stabilized, experiments were conducted to evaluate the pollutants removals and the power outputs at 24 h interval. The reduction products were also examined as well. The effects of initial sulfide concentration (30 mg/L, 60 mg/L, 90 mg/L, 120 mg/L, 150 mg/L) with HRT of 24 h and HRT (12 h, 24 h, 36 h, 48 h, 60 h) with initial sulfide concentration of 60 mg/L on the performance of ABSMFC were investigated, respectively. The COD concentration was fixed at 800 mg/L during the whole experiment. After that, microbes on the anode surface were collected and analyzed.

## Analysis and calculations

Sulfide was measured according to the methylene blue method (n = 665 nm). The indication of "sulfide" included all species (H<sub>2</sub>S, HS<sup>-</sup>, and S<sup>2-</sup>). pH was measured using a pH-201 m (Hanna, Italy). COD was monitored in concentrated sulfuric acid basing on digestion with potassium dichromate for 2 h at 150 °C.

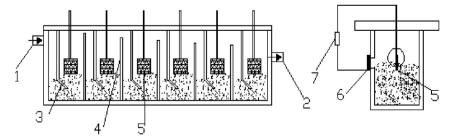


Fig. 1 – Schematic diagrams of the proposed ABRMFC. (1) water inlet; (2) water outlet; (3) baffle with slant edge; (4) straight overflow plate; (5) anode (carbon fiber felt); (6) cathode (carbon paper with Pt catalyst, copper screen); (7) resistance.

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