



# Increasing the bio-electrochemical system performance in azo dye wastewater treatment: Reduced electrode spacing for improved hydrodynamics

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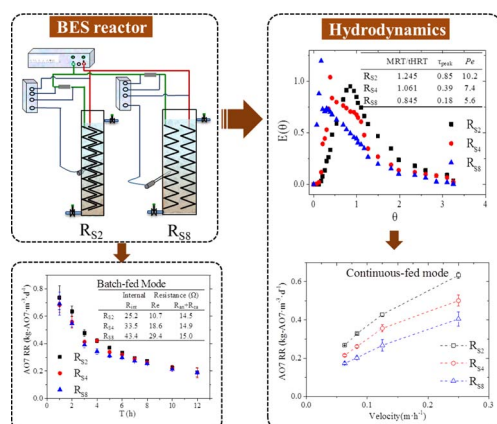
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## GRAPHICAL ABSTRACT



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## ABSTRACT

The electrodes spacing would exert a pronounced effect on bio-electrochemical systems (BESs) performance, especially for the scaling-up of reactors and practical applications. In this study, we traced the effect of electrode spacing on wastewater treatment performances from the aspects of hydrodynamics and electrochemical characteristics. Three series of folded stainless steel mesh (f-SSM) electrodes with electrode spacing of 2, 4 and 8 mm were designed for azo dye (acid orange 7 (AO7)) wastewater treatment. Results showed that BES with electrode spacing of 2 mm (R<sub>S2</sub>) obtained the highest efficiencies of AO7 decolorization (90.9 ± 0.4%) and COD removal (36.8 ± 3.8%) at HRT of 8 h, which was 30.7% and 15.2% higher than that in BES with electrode spacing of 8 mm (R<sub>S8</sub>), respectively. Moreover, the relationship between pollutants removal, internal resistance and hydrodynamics of BESs with different electrode spacing supported the hydrodynamics was significantly influence the pollutants removal performance.

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## 1. Introduction

As the most important industry in national economy in China, textile industry produced large quantities of azo dyes containing wastewater, which are toxic and highly persistent to biodegradation, raising attention for environmental concerns, as biological toxicity, “three-induced” (carcinogenicity, teratogenicity, mutagenicity) and undesirable aesthetic effects etc. (Pandey et al., 2007). Azo dyes are generally persistent under aerobic conditions but may be reduced under anaerobic conditions, and anaerobic decolorization is a cost-efficient process; however, it is usually with slow rate and requires extra organic co-substrate to provide electrons for the reduction (dos Santos et al., 2005). The co-substrate addition typically far exceeds the stoichiometric requirements, leading to additional costs and unwanted methane production (van der Zee and Villaverde, 2005). Thus, treatment process of azo dyes containing wastewater is a challenging issue that requires improvement. As all kinds of azo dye, acid orange 7 (AO7) is the most widely used dye in the textile industries, thus it is a significant component of wastewaters produced by these manufacturers that requires removal (Mu, 2009).

Recently, BESs have been explored extensively for its innovative features and environmental benefits for recalcitrant contaminants degradation in wastewater (Logan et al., 2006a,b; Rozendal et al., 2008), especially for azo dyes (Cui et al., 2016a; Mu, 2009; Sun et al., 2011), nitro-aromatics (Liang et al., 2014; Wang et al., 2011), and halogenated aromatics (Liang et al., 2013) reduction. However, most bio-electrochemical technologies developed until now have been restricted to lab scale as lacking of appropriate strategies to scaling-up. To overcome this challenge, numerous of researchers have focused on the BES scaling-up from many aspects, such as selection of suitable anode materials (Baudler et al., 2015; Guo et al., 2015; Kumar et al., 2013; Zhou et al., 2011), improving catalyst activity in cathode (Heidrich et al., 2013; Zhang et al., 2014), as well as optimization of reactor architectures (Escapa et al., 2016), to name a few. Among these problems, electrode configuration and electrode spacing were of crucial importance and had attracted a great deal of attentions (Wei et al., 2011).

Different electrode spacing and configuration has significant impacts on internal resistance and thereby resulted in different power generation, COD removal and coulombic efficiency (CE) (Ghangrekar and Shinde, 2007; Zhang et al., 2009). It is reported the maximum power output of air-cathode microbial fuel cells (MFCs) was increased 68%, which corresponded to a decrease of internal resistance from 161 to 77  $\Omega$  when the electrode spacing was reduced from 4 to 2 cm (Liu et al., 2005). The maximum power of MFC with continuous mode increased from 826 mW/m<sup>2</sup> to 1540 mW/m<sup>2</sup> as 75% overall decrease in internal resistance when the electrode spacing was decreased from 3 to 1 cm (Cheng et al., 2006). Besides, several studies demonstrated that the variation of electrode configurations has a strong relationship with hydrodynamics and takes an important role on mass transfer, convection and flow path of BESs. It demonstrated that MFC with full brush anodes spaced evenly between two cathodes (S2C) has slightly higher COD removal (6.4–24.4%) and lower coulombic efficiency than the MFC with trimmed brush anodes near a single cathode (N1C). These results were attributed to the different hydrodynamics resulted by N1C configuration and S2C configuration, for example, at theoretical HRT of 8.8 h, the measured HRT, dispersion number and Peclet number of the MFCs with N1C was 10.7 h, 0.020 and 49, respectively, which were 7.4 h, 0.028 and 36 for MFCs with S2C configuration, respectively (Kim et al., 2015). Therefore, the importance of internal resistance and hydrodynamics to performance becomes more important when design electrode spacing for BES in full-scale application. However, there was no studies consider both internal resistance and hydrodynamics impact on BES performance when varying the electrode spacing.

In this study, an innovative folded stainless steel mesh (*f*-SSM) electrode was developed (Wang et al., 2017) and built-in BES with different spacing (2, 4 and 8 mm) to treat azo dye wastewater. The

performances of BESs from the aspects of hydrodynamics and internal resistance simultaneously investigated and carried out the research from three-folded. Firstly, three pairs of *f*-SSMs were deployed in three BESs with different electrode spacing. They were operated in batch-fed mode to investigate the relationship of electrode spacing and internal resistance of BESs and its effect on azo dye removal. Secondly, BESs were operated in continuous flow mode with four theoretical hydraulic retention times (HRTs) of 8, 6, 4, and 2 h. In addition, the hydrodynamic characteristics of BESs with different electrode spacing under different HRT conditions were investigated by tracer test and residence time distribution (RTD) curve. Finally, the relationship between hydrodynamics, internal resistance and pollutants removal performance were explored to reveal the significant effect of electrode spacing on BES performance.

## 2. Materials and methods

### 2.1. BES construction

Single-chamber BESs with up-flow channel were used in the present study. The electrode was made of SAE 304 stainless steel mesh with wire diameter of 0.18 mm and a filter rating of 20 mm (Kaian Ltd., China). The electrode was prepared as following: (1) cutting the single layer stainless steel mesh (SSM) into 7 cm × 144 cm pieces (one side surface area of 0.1008 m<sup>2</sup>); (2) folding the single layer SSM pieces according to Zheng and colleagues' method (Zheng et al., 2015) to maintain folding angle with 40°; (3) the folded SSM electrode was thoroughly treated in 1 M H<sub>2</sub>SO<sub>4</sub> for 24 h to remove surface oxides and obtain a rough surface; (4) cleaning SSM by deionized water and drying in air. Titanium wire ( $\phi$ 1 mm) was used as the current collector, copper wire was used to connect the electrodes to an external resistor ( $R_{ex}$  = 10  $\Omega$ ).

The spacing of anode and cathode in the plexiglas reactor was varied in order to systematically investigate the effect of electrode spacing on BES performance. The electrode spacing, *D*, defined as the vertical distance between anode and cathode, was set as 2, 4 and 8 mm (Fig. 1). The BES with electrode spacing of 2, 4 and 8 mm, was denoted as  $R_{S2}$ ,  $R_{S4}$  and  $R_{S8}$ , respectively. Correspondingly, the effective reactor volumes were 1.55, 1.85 and 2.25 L. The specific surface areas of the cathode electrode per volume of reactor were 65, 55, and 45 m<sup>2</sup>SSM m<sup>-3</sup>, respectively. The *f*-SSM electrodes were all placed close to the wall of reactor to make sure all of wastewater penetrated the electrode. An Ag/AgCl reference electrode (+197 mV vs standard hydrogen electrode) (Shanghai Precision Scientific Instruments Co., Ltd., China) was installed between the anode and the cathode to enable potential measurement. Electric power (0.5 V) was provided to each pair of electrode with a power supply (PS-3005D, Zhaoxin Co., Ltd., China) during the whole experiments. The voltage across external resistor, cathode potential and anode potential were recorded using the data acquisition system (Keithley 2700, Keithley Co., Ltd., USA) every 10 min.

### 2.2. Operating conditions

#### 2.2.1. Inoculation and start-up

Three BESs ( $R_{S2}$ ,  $R_{S4}$  and  $R_{S8}$ ) were all inoculated with 10 mL of concentrated anaerobic sludge from a laboratory-scale biocatalyzed electrolysis system that had been operated for about 200 days, with mixed liquor suspended solid (MLSS) concentration of 7 g L<sup>-1</sup>. During the start-up period, the reactors were continuously fed domestic wastewater containing 100 mg L<sup>-1</sup> AO7, the theoretical HRT was fixed at 8 h by a peristaltic pump (Longer Co., China). Current output was observed in all reactors after 2 days and finally reached plateau after a rising duration.

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