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## Accelerated decolorization of azo dye Congo red in a combined bioanode-biocathode bioelectrochemical system with modified electrodes deployment



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#### HIGHLIGHTS

- CR decolorization was accelerated with combined bioanode-biocathode BES.
- The biocathode played an important role in accelerating CR decolorization.
- Three electrodes deployments were compared based on startup, DE and EIS analysis.
- The surrounding mode was the best, followed by vertical and horizontal ones.
- The possible mechanism for the combined bioanode-biocathode was discussed.

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#### ABSTRACT

In this study, BES with bioanode and biocathode was applied to decolorize an azo dye Congo red (CR). Results showed that the Congo red decolorization efficiency (CR-DE) within 23 h in a combined bioanode–biocathode single chamber BES was  $98.3\pm1.3\%$ , significantly higher than that of mixed solution in a dual chamber BES ( $67.2\pm3.5\%$ ) (P<0.005). Various electrodes deployments (horizontal, vertical and surrounding) in the combined bioanode–biocathode BES were further compared based on the decolorization performance and electrochemical characterization. Results indicated that CR-DE within 11 h improved from  $87.4\pm1.3\%$  to  $97.5\pm2.3\%$ , meanwhile the internal resistance decreased from 236.6 to  $42.2~\Omega$  as modifying the horizontal deployment to be a surrounding deployment. It proved that the combination of bioanode and biocathode with suitable electrodes deployment could accelerate the decolorization of azo dye Congo red, which would be great potential for the application of bioelectrochemical technology in azo dye wastewater treatment.

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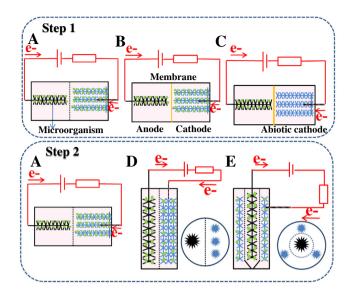
#### 1. Introduction

Bioelectrochemical technology has recently drawn increasing attention in azo dyes treatment (Fernando et al., 2012; Hou et al., 2012; Luo et al., 2011; Mu et al., 2009; Solanki et al., 2013; Wang et al., 2013a,b). There are two modes for azo dyes decolorization in BES. One is cathode decolorization in a dual chamber BES: Substrate is oxidized by bacteria at anode to produce protons and electrons, which are transferred to the cathode via membrane and external circuit, respectively. At the cathode, the azo bonds of dye are broken using proton and electron, resulting in the formation of colorless products (Mu et al., 2009; Solanki et al., 2013). In this mode, azo dye decolorization is mainly attributed to the electrochemical reduction for the absence of microorganism (Mu

et al., 2009). Few papers focused on the azo dye decolorization with dual microbial or enzymatic cathode BES (Cui et al., 2011; Savizi et al., 2012). The other is anode decolorization in an aircathode single chamber BES: Azo dye is readily transformed by anode microorganisms through reductive cleavage of azo bond to form aromatic amines in the presence of co-substrate. Due to the fact that electrons produced from substrate were transferred to the cathode for oxygen reduction rather than azo dye reduction for the higher oxygen redox potential (Cao et al., 2010; Hou et al., 2011a,b). The co-metabolism-dependent bioreduction is mainly responsible for the decolorization while less electrochemical reduction does (Cao et al., 2010).

The biocathode is proving to be a promising feature for development of BES. It has been reported that the reduction reactions at cathode especially biocathode can enhance wastewater treatment, such as pentachlorophenol, hexavalent chromium, nitrobenzene and chlorinated nitroaromatic antibiotic chloramphenicol etc.

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**Fig. 1.** Bioelectrochemical reactor configurations and operation. (A) Single chamber BES with combined bioanode-biocathode (horizontal electrodes deployment); (B) dual chamber BES with bioanode and biocathode; (C) dual chamber BES with bioanode and abiotic cathode; (D) single chamber BES with combined bioanode-biocathode (vertical electrodes deployment); (E) single chamber BES with combined bioanode-biocathode (surrounding electrodes deployment).

**Table 1**The main characteristics of the activated sludge.

Parameter	Value
pH TSS (total suspended solids)	6.73 2.26 g L <sup>-1</sup>
VSS (volatile suspended solids) TCOD (total chemical oxygen demand) SCOD (soluble chemical oxygen demand)	$1.40\mathrm{g}\mathrm{L}^{-1}$ $1.84\mathrm{g}\mathrm{L}^{-1}$ $42.70\mathrm{mg}\mathrm{L}^{-1}$
Solute protein (as COD) Solute carbohydrate (as COD)	12.88 mg $L^{-1}$ 1.47 mg $L^{-1}$

(Aulenta et al., 2011; Huang et al., 2011, 2013; Liang et al., 2013; Wang et al., 2011; Xafenias et al., 2013), gaining more and more interest as it accelerates reduction reactions and circumvents the utilization of artificially added mediators, however, less literature refers to azo dyes decolorization with biocathode BES (Cui et al., 2011; Wang et al., 2013a). Thus, it is necessary to investigate the decolorization performance of azo dyes using biocathode BES. Furthermore, there are some studies on combination of anode and cathode in BES for wastewater or groundwater treatment (Li et al., 2010; Lohner et al., 2011; Sun et al., 2011a; Wen et al., 2010). Considering the bioanode decolorization at present, air-

cathode single chamber MFC has been used extensively (Cao et al., 2010; Hou et al., 2011b, 2012; Xu et al., 2013). The air-cathode was chosen for minimizing the effect from dye and cathode (Cao et al., 2010) but was limited by the unavailable cathode reduction role in the decolorization process. This will make no effective use of the whole BES reactor in wastewater treatment. If the feasibility of biocathode decolorization is further verified, the development of a combined bioanode–biocathode system will become a promising technology for enhanced azo dye decolorization.

Present BES reactor configurations for decolorization vary from rectangular to tubular and to sleeve-type (Cui et al., 2011, 2012; Kong et al., 2013). It has been demonstrated that a suitable BES structure will be a necessary consideration for large-scale MFC design, including electrode surface area and electrode spacing (Liu et al., 2008; Zhang et al., 2013b). However, it is inevitable to consider the key design factors when the number of electrodes increases to a certain amount in the scale-up process (Zhu et al., 2013). Previous work in the laboratory has demonstrated the feasibility of reducing internal resistance and enhancing decolorization performance in a modified sleeve-type dual chamber BES for azo dye acid orange decolorization (Kong et al., 2013). If modifying the electrodes deployment of single chamber BES to be sleevetype, namely the cathode electrodes surrounding the anode electrode, it may also facilitate electron transfer and increase the effective area of anode and cathode, further improving azo dye decolorization performance in the combination of bioanode and biocathode BES. Although different reactor structures have been used during the last years, there is no systematic information regarding the influence of electrodes deployment on decolorization and electrochemical characterization in the single chamber BES.

Based on the above consideration, this study aims to develop a single chamber BES with a combination of bioanode and biocathode, and make full use of bioanode and biocathode in azo dye decolorization. Further focus on the comparison of biocathode and abiotic cathode was also studied to confirm the important role of microbial in azo dye decolorization at biocathode. Three different electrodes deployments (horizontal, vertical and surrounding) were compared based on the azo dye decolorization performance and electrochemical catalytic characterization to determine the optimum electrodes deployment in single chamber BES.

#### 2. Methods

#### 2.1. Configurations of bioelectrochemical system

The single chamber reactor was constructed by a cylindrical Plexiglas tube (ID 8 cm  $\times$  H 10 cm) without membrane (Fig. 1A, D and E). The dual chamber reactor with an anode chamber and a cathode chamber separated by a cation exchange membrane

Experimental setup.

No.	Purpose	Reactor	Chamber	Electrolyte	Time (h)
1	Performance in combined bioanode-biocathode BES and separated bioanode and biocathode BES	Combined bioanode and biocathode (Fig. 1A) Separated bioanode and biocathode (Fig. 1B)	Single Dual	CR + glucose Anolyte: CR + glucose Catholyte: CR + glucose	23 23
2	Microbial role at biocathode	Biocathode (Fig. 1B)	Dual	Anolyte: CR + glucose Catholyte: CR + glucose	23
		Biocathode (Fig. 1B)	Dual	Anolyte: CR + glucose Catholyte: CR	23
		Abiotic cathode (Fig. 1C)	Dual	Anolyte: CR + glucose Catholyte: CR	23
3	Effect of electrodes deployments	Horizontal (Fig. 1A) Vertical (Fig. 1D) Surrounding (Fig. 1E)	Single Single Single	CR + glucose CR + glucose CR + glucose	23 23 23

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