Science of the Total Environment 634 (2018) 616–627

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Enhanced performance and microbial community analysis of bioelectrochemical system integrated with bio-contact oxidation reactor for treatment of wastewater containing azo dye

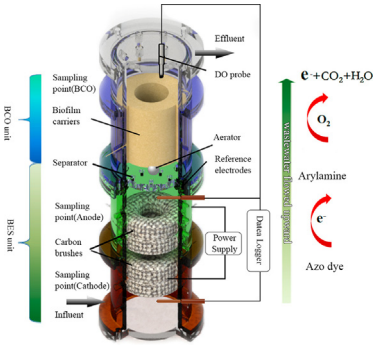
Youzhao Wang [a,1], Yuan Pan [a,*,1], Tong Zhu [a,*], Aijie Wang [b], Yalun Lu [a], Liting Lv [a], Kuo Zhang [a], Zijun Li [a]

[a] School of Mechanical Engineering and Automation, Northeastern University, Shenyang 110004, China
[b] Key Laboratory of Environmental Biotechnology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

HIGHLIGHTS

- The effects of HRT, applied voltage and DO on the performance of BES-BCO without and with BCO were investigated.
- The optimised condition was an applied voltage of 0.59 V, HRT of 12 h, and DO concentration of 0.96 mg/L at the bioanode.
- Under the optimization conditions, the DE, COD removal efficiency, and SEC values were 94.62 ± 0.63%, 89.12 ± 0. 32%, and 687.57 ± 3.86 J/g.
- Several aerobic aniline-degrading bacteria and anode-respiration bacteria were found to dominate the community of the anode biofilm.
- The removal of azo dye degradation by-products was correlated with the o-bioanode and the BCO bacterial community structure.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 January 2018
Received in revised form 27 March 2018
Accepted 28 March 2018
Available online xxxx

Editor: Dr F Frederic Coulon

Keywords:
Bioelectrochemical systems
Bio-contact oxidation
Azo dye removal
Response surface methodology
Microbial community structure

ABSTRACT

Feasibility and superiority of the bioelectrochemical system integrated with biocontact oxidation (BES-BCO) for degradation and/or mineralization of azo dyes have been confirmed. In this study, the effects of hydraulic retention time (HRT), applied voltage, and dissolved oxygen (DO) concentration at the bioanode on the performance of BES-BCO and traditional BES were investigated. Using the response surface methodology, the optimum values of HRT, applied voltage, and DO concentration at the bioanode of BES-BCO were investigated to obtain the maximum decolouration and COD removal efficiency and minimum specific energy consumption (SEC). The microbial community structure in BES-BCO was studied for analyzing the change following the introduction of oxygen. The optimised solution was an applied voltage of 0.59 V, HRT of 12 h, and DO concentration of 0.96 mg/L at the bioanode. Under such conditions, the DE, COD removal efficiency, and SEC values were 94.62 ± 0.63%, 89.12 ± 0. 32%, and 687.57 ± 3.86 J/g, respectively. In addition, after changing from BES to BES-BCO, the bacterial community structure of the bioanode underwent significant changes. Several aerobic aniline-degrading bacteria and anode-respiration bacteria (ARB) were found to dominate the community of the anode biofilm. The results showed that the removal of azo dye degradation by-products was closely correlated with the o-bioanode and the BCO bacterial community structure.

* Corresponding authors.
E-mail addresses: panyuanneu@gmail.com, (Y. Pan), tongzhu@mail.neu.edu.cn. (T. Zhu).
[1] These authors contributed equally to this work and should be considered co-first authors.

https://doi.org/10.1016/j.scitotenv.2018.03.346
0048-9697/

1. Introduction

Dyes are used widely in textile, pharmaceutical, cosmetic, food, and other industries. The deleterious aspects of dye effluents are their high chroma, high toxicity, and high chemical oxygen demand (COD). Azo dyes with the azo bond (—N═N—) are considered to be the largest and most versatile class of organic dyes and are used widely in the textile industry. Because of the complex aromatic structures of azo dyes, they are difficult to degrade (Selvam et al., 2003).

Conventional technologies, including physical-chemical technologies and electrochemical technology, have been employed successfully to remove azo dyes from wastewater (You et al., 2016; Robinson et al., 2001). However, these methods have significant drawbacks, including high capital costs and high operating costs. Conventional anaerobic biological treatment can remove chromaticity colour (Van der Zee et al., 2001), but further aerobic treatment is required (Pandey et al., 2007) because the aromatic amines formed from decolourisation of azo dyes are toxic and difficult to degrade under anaerobic conditions. Moreover, it has a slower decolouration rate. The bioelectrochemical system (BES) is a new treatment technology that can accelerate the decolouration process. BES integrates advantages from both conventional electrochemical and biological processes (Mu et al., 2009; Wang et al., 2013). In recent years, researchers have gained great interest by developing BES technology, like Microbial fuel cell (MFC), for azo dye wastewater decolourisation (Khan et al., 2014; Khan et al., 2015). However, BES has disadvantages similar to those of conventional anaerobic processes as it requires further aerobic treatment (Sultana et al., 2015).

For years, the bioanode of BES has been operated under the anaerobic condition because oxygen is considered to be a competitive electron acceptor. However, a positive role of oxygen in the biodegradation of recalcitrant organics on the anode as well as on electricity generation was reported recently (Cheng et al., 2015). Small amounts of oxygen helps biotransformation or conversion of recalcitrant organics, like aromatic amines, to non-ring organic products by micro-aerophilic bacteria. Meanwhile, the energy extraction or recovery from aromatic amine removal would be facilitated by introducing oxygen to bioanode (o-bioanode) in BES. The concept of o-bioanode has provided new insights into degrading azo dyes and their toxic degradation products.

In a previous study, the feasibility and superiority of the up-flow membrane-less bioelectrochemical system integrated with bio-contact oxidation (BES-BCO) for the degradation and/or mineralization of azo dyes was confirmed (Pan et al., 2017). Several key factors may significantly influence the degradation process and energy consumption of the system, including hydraulic retention time (HRT), applied voltage, and dissolved oxygen(DO). However, little is known about the effects of these key factors on the performance of BES-BCO and the influence on microbial community structures after the introduction of oxygen into the bioanode.

Response surface methodology (RSM) is a statistical method based on the multivariate non-linear model that is useful for studying interactions of various parameters affecting the process (Mundra et al., 2007; Chou et al., 2010). It has been used widely for the investigation and optimisation of the factors affecting the environmental pollution (Halim et al., 2009; Shanmugam et al., 2017; Wu et al., 2017). For the microbial electrolysis cell, the applied voltage can provide external electrons for azo dye reduction and accelerate the azo dye removal rate (Mu et al., 2009). HRT determines the reaction time between pollutants and bacteria. The DO concentration facilitates the reduction of azo dye degradation products and bio-contact oxidation biofilm growth. Both of these significantly influence degradation performance and energy consumption; thus, RSM can be used to evaluate and optimise the novel BES-BCO integrated system for best removal efficiency and energy consumption.

In this study, the effects of HRT, applied voltage and DO on the performance of BES-BCO without and with BCO were investigated. RSM was performed to obtain the optimal values of HRT, applied voltage and dissolved oxygen, for maximising the removal efficiency and minimising the energy consumption in the BES-BCO system. To gain insight into how the introduction of BCO into BES influences microbial communities, the 16S rRNA gene-based high-throughput Majorbio Bio-pharm sequencing platform was employed to analyse biofilms on the contact-oxidation carrier and bioelectrodes.

2. Materials and methods

2.1. Reactor configuration

In a previous study, a laboratory-scale integrated BES-BCO bioreactor, made of polymethyl methacrylate, was developed (Pan et al., 2017). The schematic diagram of BES-BCO reactor is given in Fig.1. The BES-BCO system comprised two units, a BES unit as the lower part and a BCO unit as the upper part. The BES unit was membrane-less system and included bioanode and biocathode. A plate with even distribution holes was installed between BES unit and BCO unit to reduce the negative oxygen impact on BES unit and ensure even distribution of up-flow fluid.

The total empty reactor volume was 5.95 L.

An aerator was placed at the bottom of the BCO unit to supply the oxygen, and a dissolved oxygen (DO) probe was installed at the BES anode to monitor the concentration of DO at the anode in real time. Two saturated calomel reference electrodes (SCE) were installed between the anode and the cathode to measure the potential. The anode and cathode were connected by a direct-current power supply. A precise resistor (10 Ω) was connected into the circuit. The bio-electrodes and the reference electrodes were connected to a data acquisition system, and the voltage of the resistor and the potentials of the anode and cathode were recorded every 15 min using the data acquisition system.

2.2. Inoculation and operation

The original inoculum source was anaerobic sludge collected from the anaerobic tank of a wastewater treatment plant in Shenyang, China. Biofilm carriers of BCO was inoculated with aerobic sludge collected from the wastewater treatment plant. The composition of the synthetic azo dye wastewater was glucose; acid orange 7 (AO7); $NaH_2PO_4 \cdot 2H_2O$, 2.77 g/L; $Na_2HPO_4 \cdot 12H_2O$, 11.55 g/L; NH_4Cl, 0.31 g/L; KCl, 0.13 g/L; trace element solution (1 mL); and vitamin solution (Donlon et al., 1997). The reaction solution was purged with nitrogen gas for 15 min to remove possible dissolved oxygen in the solution.

To compare the performance of BES-BCO without and with BCO under different conditions, the experimental period can be divided into six stages. In Stages I and III, the reactor was operated as BES unit. The BCO unit was not installed into the system. In Stage I and II, the system was investigated the influence of different power supplied and HRT under anaerobic condition. After Stage II, graphite fibres were cut from the anodes and cathodes for DNA extraction. The effect of oxygen to the traditional BES was investigated at Stage III. The aerator was installed at 10.0 cm above the BES unit at Stage III. The intermittent aeration was applied. According to changing the aerator situation and the aeration time, the concentration of DO was controlled within limits. From stage IV to VI, the BCO unit was installed system. The reactor was operated as a BES-BCO system. After Stage V, graphite fibres of o-bioanode and BCO biofilm carriers were cut for DNA extraction. Table 1 showed operation conditions at different stages.

The Box–Behnken design response surface methodology (BBD-RSM) was used to optimise different parameters. The optimal experiment was further expanded using Design Expert (Version 8.0.6, Stat-Ease Inc. USA). Table 2 showed the Box–Behnken design for three independent variables. Finally, the morphological characteristics and bacterial community structure of the biofilm on electrodes and BCO carriers were further analysed.