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Enhanced decolorization of azo dye in a small pilot-scale anaerobic baffled reactor coupled with biocatalyzed electrolysis system (ABR–BES): A design suitable for scaling-up

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HIGHLIGHTS

- A pilot-scale refractory wastewater treatment process (ABR–BES) was developed.
- ABR–BES showed superior decolorization performance.
- VFAs could be efficiently utilized for azo dye decolorization in ABR–BES.
- The mechanism of azo dye transformation in ABR–BES was proposed.
- Power supply and HRT were the key impact factors of ABR–BES.

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ABSTRACT

A four-compartment anaerobic baffled reactor (ABR) incorporated with membrane-less biocatalyzed electrolysis system (BES) was tested for the treatment of azo dye (alizarin yellow R, AYR) wastewater (AYR, 200 mg L^{-1}; glucose, 1000 mg L^{-1}). The ABR–BES was operated without and with external power supply to examine AYR reduction process and reductive intermediates with different external voltages (0.3, 0.5 and 0.7 V) and hydraulic retention times (HRT: 8, 6 and 4 h). The decolorization efficiency in the ABR–BES (8 h HRT, 0.5 V) was higher than that in ABR–BES without electrolysis, i.e. 95.1 ± 1.5% versus 86.9 ± 6.3%. Incorporation of BES with ABR accelerated the consumption of VFAs (mainly acetate) and attenuated biogas (methane) production. Higher power supply (0.7 V) enhanced AYR decolorization efficiency (96.4 ± 1.8%), VFAs removal, and current density (24.1 A m^{-3} TCV). Shorter HRT increased volumetric AYR decolorization rates, but decreased AYR decolorization efficiency.

1. Introduction

Biocatalyzed electrolysis systems (BESs) can generate electrical power from organic waste and wastewater, while the power density produced from BESs is too small to be used as renewable power source. Recently, more researches have been focused on wastewater treatment, bioremediation of contaminants and stabilization of recalcitrant compounds using BESs to improve cost-effectiveness and sustainability (Mu et al., 2009; Rabaey et al., 2010). Laboratory scale BESs have been successfully tested for the removals of pyridine (Zhang et al., 2009), quinolone (Zhang et al., 2010), indole (Luo et al., 2010), antibiotics ceftriaxone sodium and penicillin (Wen et al., 2011a,b) and 1,2-dichloroethane (Pham et al., 2009) present in wastewater. The main goal of these literatures was to oxidize reduced forms of compounds. In comparison, BESs can be applied to reduce oxidized forms of contaminants on the cathode to less toxic or more stable products. For example, a number of reduction reactions have been achieved using BESs for reducing nitrobenzene (Wang et al., 2011, 2012), antibiotics chloramphenicol (Liang et al., 2013), copper (II) (Tao et al., 2011),

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halogenated compounds (Lohner et al., 2011; Strycharz et al., 2010) and azo dyes (Mu et al., 2009; Cui et al., 2012, 2014). Recently, our research group developed a membrane-less, up-flow biocatalyzed electrolysis reactor (UBER) that successfully reduced azo dye up to 97.5 ± 1.0% (Cui et al., 2012, 2014). However, previous studies used acetate as the electron donor for exoelectrogens to generate circuited electrons that were utilized for reducing azo dyes on the cathode in UBER. Since azo dye wastewater contains organics more complex than acetate (Şen and Demirer, 2003), biofilm density of exoelectrogens on the anode was readily diluted by fermenting bacteria in BESs fed with fermentable, complex substrate, and consequently current density could drop seriously (Lee et al., 2008). Then, azo dye reduction on the cathode would be negligible in UBER directly receiving the substrate. UBER would achieve high current density and azo dye reduction efficiency on the cathode if exoelectrogens utilized simple fatty acids and hydrogen gas fermented from complex substrate using dark fermentation or methanogenesis prior to UBER as pre-treatment (Freguia et al., 2008; Hamelers et al., 2010). Such pre-treatment also mitigate the fluctuation of azo dye wastewater quality that often occurred in industrial wastewater.

Anaerobic baffled reactor (ABR) can work efficiently as pre-treatment to UBER, due to its several merits. ABR steadily treats recalcitrant wastewater (Barber and Stuckey, 1999). The unique structure of ABR provides separate phases of acidogenesis and methanogenesis in a single reactor, and this phase separation also enhances protection of microorganisms against toxic substances or fluctuation of environmental parameters (e.g., pH, temperature, etc.). Therefore, ABR can improve treatment flexibility to recalcitrant wastewater that is typically fluctuated (Bachmann et al., 1985; Grover et al., 1999). The configuration and operation of ABR are relatively simple over other anaerobic processes, such as the lack of liquid–gas separation devices, packing materials, and mechanical mixing (Metcalf, 2003). Despite of these advantages, ABR would not meet strict effluent standards for recalcitrant wastewater effluent, such as azo dye wastewater, especially at short hydraulic retention time (HRT) (Van der Zee and Villaverde, 2005). The volatile fatty acids (VFAs) produced from fermentation of organics in ABR, especially acetate, are normally poor electron donors for decolorization of azo dyes under anaerobic conditions (Dos Santos et al., 2005, 2007). ABR reduction rate became slow at low concentration of azo dye (less than 40 mg L^{-1}) (Bell et al., 2000; Seshadri et al., 1994), and its reduction at low concentration was readily competed with other reduction reactions (Dos Santos et al., 2006). For these reasons, it is challenging to decrease azo dye concentration below textile wastewater discharge standards solely with ABR and post-treatment is needed to further reduce azo dye. Biological aerobic processes would be an option as post-treatment. Intensive aeration with long HRT could remove azo dye enough to meet the standards, but such oxygen supply substantially increases operating and maintenance costs, which significantly dilutes the merits of ABR – lack of oxygen. Taking the advantages of UBER and ABR into account, the combination of the two processes can complement each other. UBER needs a pre-treatment step to generate VFAs as substrate to exoelectrogens, and ABR meets this requirement. ABR cannot reduce azo dye below the discharge standards but UBER can decrease azo dye concentration below the standards without aeration.

In this study, we developed a new system integrating UBER with ABR by installing UBER module into each compartment of ABR (called, ABR–BES) and tested this novel process at a small pilot-scale for the treatment of azo dye (alizarin yellow R: AYR) wastewater. ABR–BES has several advantages for scale-up as compared to existing bio-systems. First of all, ABR–BES is membrane-free system. The lack of membrane can decrease costs for investment and maintenance significantly over other BESs having membrane. Besides, ABR–BES employs low-cost, granular graphite as electrodes without chemical catalysts, which not only increases the contact surface but also reduces capital costs. Moreover, the design of ABR–BES can be readily retrofitted to existing anaerobic wastewater treatment systems, simply with installation of electrodes and baffles in the systems. Therefore, the successful outcomes of this study can facilitate the transfer of this technology into field. This study has three objectives. Firstly, we tracked AYR reduction process, its intermediates, VFAs, and biogases production throughout the compartments of the ABR–BES with and without power supply at a constant HRT of 8 h. Secondly, we tested different applied voltages for AYR reduction at the fixed HRT. Finally, we varied HRT to determine the maximum influent loading rate of AYR at a fixed applied voltage of 0.7 V.

2. Methods

2.1. Anaerobic baffled reactor coupled with biocatalyzed electrolysis system (ABR–BES)

The schematic diagram of ABR–BES is presented in Fig. 1. ABR–BES was made of PVC (polyvinyl chloride) with external dimensions L 80 cm × W 15 cm × H 55 cm. The reactor was divided into four compartments (#1, #2, #3 and #4; the volume of each chamber: 10 L) by three straight over-flow plates. The four compartments design of ABR in this study was to improve methane capture by stimulating separate metabolism between fermentation and methanogenesis (Barber and Stuckey, 1999). The over-flow plates have different water levels (42 cm, 40 cm and 38 cm in height, respectively), which helped influent flowing from one compartment to the next compartment. Each compartment was separated into an up-flow part and a down-flow part (width ratio of 4:1) by a vertical baffle (having a 45° slant edge at the bottom) with a 1 cm distance from the bottom of the reactor. This configuration allows ABR–BES to have anaerobic granular sludge bed (~3 L) on bottom and a bioelectrolysis zone in upper portion (2 L), in individual compartment. As illustrated in Fig. 1, the influent first flowed via anaerobic granular sludge bed at the bottom of compartments where anaerobic biological decolorization reactions would mainly occur. Biocatalyzed electrolysis reactions for decolorizing AYR were simulated in the following. Four samples ports (SP 1#, SP 2#, SP 3# and SP 4# with inner diagram 1 cm) were vertically placed in the ABR–BES for monitoring chemical variation.

The cathode and anode insets in ABR–BES were both made of granular graphite (ID from 3.0 to 5.0 mm, Harbin Northern Electrical Carbon Co., Ltd., China). The graphite was tightly filled in a cuboid basket made up of titanium (L 13 cm × W 10 cm × H 7 cm) as a current collector. The hole of the basket was rhombic with side of 3 mm. A titanium rod (30 cm) was welded on the basket for electricity conductance and external circuit connection. The anode insets were placed over the cathode in the four compartments. Four Ag/AgCl electrodes (+195 V vs standard hydrogen electrode) (Shanghai Precision Scientific Instruments Co., Ltd., China) were installed between the anode and the cathode insets in each compartment to measure electrode potentials. The distances between the reference electrodes and the bottom of the four compartments are 35 cm, 34 cm, 33 cm and 32 cm. Electric power was provided to the ABR–BES with a power supply (IT6921, Itech Co., Ltd., USA). The anode and cathode insets, and the reference electrode were connected to a data acquisition system (Keithley 2700, Keithley Co., Ltd., USA) to record electrode potentials and current every 10 min.

Medium was fed into the ABR–BES with a pump (Longer YZII25, Longer pump Co., China) at different flow rates of 87.5–175 mL min^{-1}, corresponding to hydraulic retention time (HRT) of 8–4 h.