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Evaluation of anaerobic sludge volume for improving azo dye decolorization in a hybrid anaerobic reactor with built-in bioelectrochemical system

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HIGHLIGHTS

- A hybrid anaerobic reactor with built-in BES was efficient for azo dye removal.
- The appropriate volume ratio between anaerobic sludge and cathode was 1:1.
- BES module presented higher electron utilization efficiency.

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ABSTRACT

A hybrid anaerobic reactor with built-in bioelectrochemical system (BES) has been verified for efficiently treating mixed azo dye wastewater, yet still facing many challenges, such as uncertain reactor construction and insufficient electron donors. In this study, an up-flow hybrid anaerobic reactor with built-in BES was developed for acid orange 7 (AO7) containing wastewater treatment. Cathode and real domestic wastewater both served as electron donor for driving azo dye decolorization. The decolorization efficiency (DE) of AO7 (200 mg/L) in the hybrid reactor was 80.34 ± 2.11% with volume ratio between anaerobic sludge and cathode ($VR_{slu:cat}$) of 0.5:1 and hydraulic retention time (HRT) of 6 h, which was 15.79% higher than that in BES without sludge zone. DE was improved to 86.02 ± 1.49% with $VR_{slu:cat}$ increased to 1:1. Further increase in the $VR_{slu:cat}$ to 1.5:1 and 2:1, chemical oxygen demand (COD) removal efficiency was continuously improved to 28.78 ± 1.96 and 32.19 ± 0.62%, but there was no obvious DE elevation (slightly increased to 87.62 ± 2.50 and 90.13 ± 3.10%). BES presented efficient electron utilization, the electron usage ratios (EURs) in which fluctuated between 11.02 and 13.06 mol e^-/mol AO7. It was less than half of that in sludge zone of 24.73–32.06 mol e^-/mol AO7. The present work optimized the volume ratio between anaerobic sludge and cathode that would be meaningful for the practical application of this hybrid system.

1. Introduction

Industrial parks drive China's rapid development in the past decades. There are hundreds of national grade and thousands of middle and small industrial parks all over the country (Geng and Doberstein, 2008). It is enforced to pretreat the wastewaters produced in these industries in situ before discharging to the centralized wastewater treatment plants or merging into municipal wastewater treatment plants. The industrial wastewaters are usually mixed with domestic wastewater and treated together, for instance, Shaoxing wastewater treatment plant, the largest plant for treating mixed printing and dyeing wastewater in Asia, has a capability of 900,000 m^3/d (approximately 8% municipal sewage and 92% industrial wastewater) (Chen et al., 2011). Centralized wastewater treatment plant in Shijiazhuang pharmaceutical industrial park is responsible for mixed wastewater treatment that

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contains 98% of pharmaceutical wastewater and 2% municipal wastewater (Lei et al., 2010).

The mixed wastewater is usually characterized as fickle composition, biorefractory, and insufficient available electron donor source (COD<1000 mg/L). To remedy these mixed refractory wastewaters to meet the stringent discharge standard about COD and chroma, anaerobic methods are extensively employed for recalcitrant compounds biotransformation (Cui et al., 2016c). However, anaerobic bioprocesses require plentiful electron donors (far exceeds stoichiometric requirements) for microbial metabolism and reduction of oxidized groups (Mu et al., 2009). The commonly used strategy to solve this problem is to add co-substrate (such as lactate, glucose and ethanol) as extra electron donors, but it undoubtedly increase the operation cost and carbon footprints (Rasool et al., 2015).

The emergence of bioelectrochemical systems (BESs) provides a new way to make up for the deficiency of anaerobic process. Anode respiration bacteria degrade organic matters to release electrons to the anode, which are then transfer to the cathode. The cathode works as efficient electron donor for oxidized pollutants reduction and resource recovery, including electricity generation (Cheng et al., 2015), biogas (H_2 or CH_4) production (Wang et al., 2014), heavy metals recovery (Huang et al., 2014), nitro reduction (Wang et al., 2011), azo bond cleavage (Cui et al., 2012, 2016a) and dehalogenation (Liang et al., 2013), etc. One of the attractive characters of BES is lower organic electron donor requirement. Mu et al. reported that BES presented two orders of magnitude lower organic matter usage ratio than traditional anaerobic methods (Mu et al., 2009). Recently, one feasible strategy to improve the performance of present processes is assembling BES into traditional anaerobic units to build hybrid systems, in view of the fluctuating and practical conditions, as well as the challenges in scaling-up (Cui et al., 2016b). Several studies have reported the concept of hybrid anaerobic system. Cui et al. developed a hybrid anaerobic baffled reactor (ABR) with built-in BES to decolorize azo dye, which improved about 8% decolorization efficiency (DE) compared to the sole ABR (Cui et al., 2014); Zhao et al. enhanced biogas production from a hybrid up-flow anaerobic sludge blanket (UASB) and BES reactor (Zhao et al., 2014); Shen et al. reported that the *p*-nitrophenol removal efficiency was significantly enhanced by a BES-UASB hybrid reactor comparing to the control UASB (Shen et al., 2014); Cui et al. investigated the position of electrodes in a BES-UASB hybrid reactor and concluded that the installing BES in liquid phase was superior to that in sludge phase (Cui et al., 2016b). However, there are lots of significant issues, for instance, the volume matching between anaerobic sludge and BES, development of economical and sustainable electrode materials, optimization of the hydraulic flow pattern, that should be identified to improve this newly designed system and target its application.

In this study, an UASB with built-in BES was used for synthetic low strength wastewater (real domestic wastewater containing azo dye) treatment. The volume matching between anaerobic sludge and BES was optimized to enhance azo dye decolorization, the contributions of anaerobic sludge and cathode to azo dye decolorization were particularly analyzed, electrons utilization efficiencies of anaerobic sludge and cathode zone for azo dye decolorization were further evaluated.

2. Material and methods

2.1. Reactor configuration

A dual-chamber up-flow anaerobic reactor was manufactured with plexiglass as shown in Fig. 1. Anode chamber and cathode chamber respectively had an empty volume 2 L with the

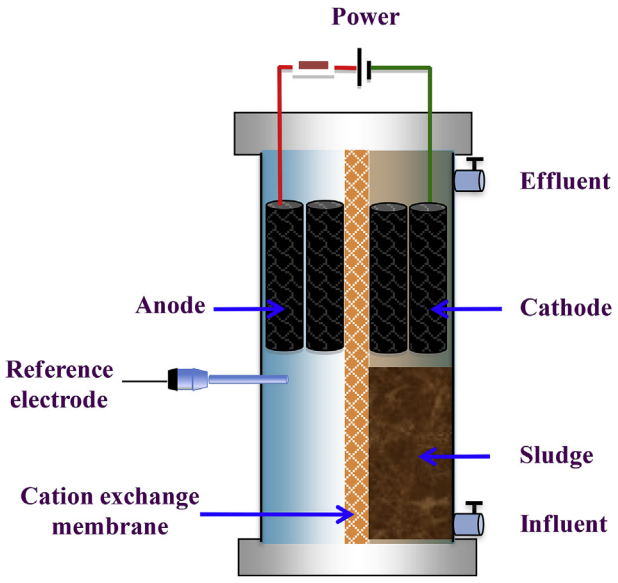

Fig. 1. Schematic diagram of the dual-chamber up-flow hybrid anaerobic reactor with built-in bioelectrochemical system.

dimensions of L 10 cm × W 5 cm × H 40 cm. Cation exchange membrane (Ultrex CMI-7000, Membranes International, U.S.) was used for separation of anode and cathode. Both anode and cathode were consisted of two bundles of graphite fiber brushes with 8 cm height, given a total volume was 0.0004 m^3. Each bundle of graphite fiber brushes was constructed of 8 small carbon brushes (2.5 cm in diameter and 4 cm in length, graphite fiber produced by TOHO TENAX, Co., Ltd., Japan). All carbon brushes were pretreated according to the literature (Feng et al., 2010): cleaned by soaking in the acetone overnight, acid treated by soaking in a solution of ammonium peroxydisulfate and concentrated sulfuric acid (100 mL/L) for 15 min, heat-treated in a muffle furnace at 450 °C for 30 min. Titanium wire (Baoji LiXing Titanium Group Co., Ltd., China) that used as current collector was stretched out of the reactor and connected to external circuit. The reactor was operated with external resistance of 10 Ω and applied voltage of 0.5 V using a DC power supply (FDPS-180, Fudan Tianxin Scientific and Educational Instruments Co., Ltd, Shanghai, China). A saturated calomel electrode (SCE) (+247 mV vs. standard hydrogen electrode) was used as reference electrode and placed in the anode chamber in view of the relative stable condition in anode chamber than cathode one. In this work, all the potentials were reported against SCE. Anode, cathode and reference electrode were connected to a data acquisition system (Keithley 2700, Keithley Co. Ltd., U.S.) for collecting potentials and currents every 10 min.

2.2. Inoculation and operation

The anode was inoculated with effluent from a long-term operating BES and mixed with active sludge from Taiping municipal wastewater treatment plant (Harbin, China) for fast enrichment of anodic biofilm. In order to acquire stable and continuous electrons supply, anode was installed in an individual chamber and provided ideal condition. The composition of anodic medium was NaAc (2000 mg/L), phosphate buffer solution (50 mmol/L, pH ~7.2), KCl (130 mg/L), NH_4Cl (310 mg/L), distilled water, trace element solution (10 mL/L) and vitamin solution (10 mL/L) (Wolin et al., 1963). Real domestic wastewater with AO7 (50–200 mg/L) addition was used as cathodic medium. The main characteristics of the domestic wastewater were as follow: COD of 120 ± 20 mg/L, total