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Azo dyes wastewater treatment and simultaneous electricity generation in a novel process of electrolysis cell combined with microbial fuel cell

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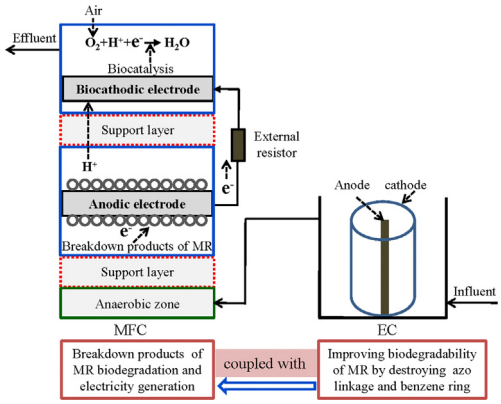
HIGHLIGHTS

- A new process of azo dyes removal coupled with electricity generation was proposed.
- EC had a positive effect on enhancement of biodegradability of dyes wastewater.
- Excellent decolorization and cell voltage output were obtained in the EC-MFC system.

GRAPHICAL ABSTRACT



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ABSTRACT

A new process of electrolysis cell (EC) coupled with microbial fuel cell (MFC) was developed here and its feasibility in methyl red (MR) wastewater treatment and simultaneous electricity generation was assessed. Results indicate that an excellent MR removal and electricity production performance was achieved, where the decolorization and COD removal efficiencies were 100% and 89.3%, respectively and a 0.56 V of cell voltage output was generated. Electrolysis voltage showed a positive influence on decolorization rate (DR) but also cause a rapid decrease in current efficiency (CE). Although a low COD removal rate of 38.5% was found in EC system, biodegradability of MR solution was significantly enhanced, where the averaged DR was 85.6%. Importantly, COD removal rate in EC-MFC integrated process had a 50.8% improvement compared with the single EC system. The results obtained here would be beneficial to provide a prospective alternative for azo dyes wastewater treatment and power production.

1. Introduction

Annually, about 70 million tons of synthetic dyes are produced and mostly used in textile, cosmetics, leather, etc, and approximately 30 ~ 150 thousand tons of dyes included wastewater are discharged into water bodies (Shabbir et al., 2017), probably resulting in severe aquatic environment problems, such as high colority, high COD, biological toxicity, etc. Currently, azo dyes are the most versatile and widely used synthetic dyes in practical applications, accounting for 80% of those (Hou et al., 2017), which are characterized by aromatic compounds with one or more azo groups (–N=N–) (a stable chromophores), such as methyl red (MR). The azo double bound groups are toxic and difficult to biodegrade in nature (Li et al., 2016), probably leading to serious

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environmental problems and threatening public health. Hence, treatment of azo dyes wastewater is necessary before their final discharge into water bodies. Although traditional biological degradation is an environmental friendly and cost-effective technology, it may not be used as a standalone process for treatment azo dyes wastewater with low biodegradability and high biotoxicity.

To date, various physicochemical methods have been developed to treat azo dyes wastewater, mainly including coagulation/flocculation (Akyol et al., 2013), adsorption (Gong et al., 2015), membrane filtration (Sun et al., 2009) and advanced oxidation processes. However, advanced oxidation processes, such as Fenton's reaction (Rahim Pouran et al., 2014), ozonation oxidation (Benincá et al., 2013), catalytic degradation (Bi et al., 2009), etc., can be successful in the decolorization of azo dyes wastewater but are high energy requirement and operational cost and probably result in secondary pollution generated by the breakdown intermediates of azo dyes, commonly being carcinogenic, teratogenic or mutagenic (Hou et al., 2017). Similarly, the inherent drawbacks of other physicochemical techniques are also obvious by the following two points: (a) in coagulation/flocculation processes, a number of chemicals are needed and large quantities of settled sludge can be produced, requiring being further handled and disposed (Shi et al., 2007). (b) secondary waste streams was generated in adsorption and membrane filtration due to reviving adsorbent and backflushing the membranes (Mu et al., 2009). As mentioned above, the physicochemical technology was also not be sufficient as a single azo dyes wastewater process and may be an excellent pretreatment unit. Therefore, an integrated process, in an easy operation, technology economically and environment friendly way, should be proposed to meet the required effluent quality.

Recently, several attempts have been conducted to develop new combination processes of azo dyes wastewater removal (Li et al., 2017; Punzi et al., 2015; Zou et al., 2015). Most of those reported on the combination process about physicochemical coupled with biological degradation. The previous study (Zou et al., 2015) indicated that the process linking Fenton's oxidation with biological aerated filter (BAF) system was effective in MR wastewater treatment. Similarly, Li et al. (2017) demonstrated that complete mineralization of 400 mg/L Orange G was achieved by using a microbial reverse-electrodialysis electrolysis cell-Fenton integrated process. Commonly, the physicochemical units are followed by a biological treatment system in the coupled process, where, firstly, azo dye and its preliminary degradation products (such as aromatic amines) can be degraded to simple molecules (such as hydrocarbons and alcohols) in the physicochemical units and then those were further removed using biodegradation method. However, the conventional biological treatment processes, such as SBR, BAF, A/A/O, etc., were rather complicated and difficult to operate in practice. So, from the standpoints of operation and maintenance, it is more desirable to further simplify the coupling systems.

More recently, microbial fuel cell (MFC) as an innovative and promising technology has attracted much attentions in wastewater treatment and simultaneous energy recovery (Fang et al., 2015, 2016; Wetser et al., 2015), In a MFC, electrons can be generated via oxidizing the organics from wastewater at anode, i.e., electron donors, and then these electrons were transferred through an external circuit to the cathode and consumed by electron acceptors (Fang et al., 2015). Most interestingly, it has been demonstrated that many kinds of breakdown intermediates of azo dyes can be effectively degraded in the MFC systems (Fang et al., 2016). Thus, it should be reasonable to combine MFC with other methods to enhance degradation efficiency of azo dyes in a simpler and more economical way.

Nowadays, some research efforts have been reported regarding combination of MFC with other methods for azo dyes wastewater treatment (Li et al., 2016). Commonly, MFC was used as a pretreatment and followed by other methods. This may present a great challenge due to the fact that both very low biodegradability of azo dyes and its toxicity to microorganisms would greatly decrease MFC sustainability. In fact, physicochemical pretreatment, such as electrolytic cell (EC), is still a quite good method to destroy the azo linkage and benzene ring contained in azo dyes (Mohapatra et al., 2010).

In this content, a novel EC-MFC integrated process was developed in this study for azo dyes wastewater treatment. MR wastewater was used from the Chemical Dye Industrial Factory, Fengyang, China. In the coupled process, EC can enhance biodegradability of MR wastewater and MFC can further degrade the breakdown intermediates of MR and simultaneously generate electric energy. The specific objectives were: (1) to investigate the influence of electrolysis voltages and hydraulic retention time (HRT) on decolorization rate (DR) of MR and current efficiency (CE) in EC system, (2) to enrich microorganisms responsible for MR removal and generation of bioelectricity in MFC system, (3) to assess the feasibility of the EC-MFC integrated process for MR wastewater treatment and bioelectricity generation. Moreover, the shifts in microbial community structure during the MFC startup period were analyzed by using Polymerase Chain Reaction-Denaturing Gradient Gel Electrophoresis (PCR-DGGE). The results obtained from here may provide a good suggestion for non-degradation organic wastewater treatment and simultaneous renewable electric energy generation in practice.

2. Materials and methods

2.1. EC-MFC integrated process setup and operation

A lab-scale EC-MFC integrated process (Fig. 1) proposed here consisted of two systems made from polyvinyl chloride, i.e., an EC and a MFC. For EC system, the total volume was 4.0 L with an internal diameter of 16 cm and a 20 cm in height while the working volume was 3 L. A high-purity graphite rod (2 cm in diameter and 13 cm in length; Zhejiang Leqing Graphite Rod Co., Ltd, China) fixed in the circular groove at the bottom center of the EC was used as the anode. And a stainless steel column (size: 6 cm internal diameter × 12 cm height × 0.15 cm wall thickness; Jiangsu Yixuan Stainless Steel Products Factory, China) placed on the trestle at the bottom center of the EC was adopted as the cathode. An adjustable direct current regulated power supply (PS-305DM; Dongguan Longwei Electronic Technology, Co., Ltd, China) was adopted to provide a power supply for electrolysis of MR. The anode and cathode were connected with titanium wires (1.5 mm in diameter) to the positive and negative output of the PS-305DM respectively, thus constituting an electric current during the electrolysis of MR. The water inlet and outlet were set up at a distance of 1 cm and 15 cm from the bottom of EC, respectively. Besides, an overhead stirrer at 100 rpm was used to maintain well-mixed condition.

The MFC system with a working volume of 3.2 L was mainly comprised of anaerobic zone, anode zone, cathode zone and two support layers. Based on the characteristics of microbial treatment in anaerobic conditions (Henze and Harremoes, 1983), an innovation design of anaerobic zone in MFC can preliminary hydrolysis the intermediate products and eliminate the dissolve oxygen possible existence in effluent from EC system. Along the flow direction (see Fig. 1), the first support layer (16 cm in height) was packed with gravel (4 ~ 5 mm in diameter). The anode zone (8 cm in height) was packed with granular activated carbon (a diameter of 4 ~ 7 mm; Taiyuan Huasheng, Co., Ltd, China) pretreated by soaking in 1 M NaOH and 1 M HCl for 12 h respectively. A thickness of 0.2 mm carbon cloth (Shanghai Miaohan Technology, Co., Ltd,