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Research article

Multiple response optimization for high efficiency energy saving treatment of rhodamine B wastewater in a three-dimensional electrochemical reactor

Jing Ji [a], Yang Liu [b], Xue-yuan Yang [a], Juan Xu [a,*], Xiu-yan Li [a,**]

[a] *Shanghai Key Lab for Urban Ecological Processes and Eco-Restoration, School of Ecological and Environmental Sciences, East China Normal University, Shanghai, China*

[b] *School of Environment and Energy Engineering, Anhui Jianzhu University, Hefei, China*

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ABSTRACT

The removal of high-concentration rhodamine B (RhB) wastewater was investigated in a three-dimensional electrochemical reactor (3DER) packed with granular activated carbon (GAC) particle electrodes. Response surface methodology (RSM) coupled with grey relational analysis (GRA) was used to evaluate the effects of voltage, initial pH, aeration rate and NaCl dosage on RhB removal and energy consumption of the 3DER. The optimal conditions were determined as voltage 7.25 V, pH 5.99, aeration rate 151.13 mL/min, and NaCl concentration 0.11 mol/L. After 30 min electrolysis, COD removal rate could arrive at 60.13% with an extremely low energy consumption of 6.22 kWh/kg COD. The voltage and NaCl were demonstrated to be the most significant factors affecting the COD removal and energy consumption of 3DER. The intermediates generated during the treatment process were identified and the possible degradation pathway of RhB was proposed. It is worth noting that 3DER also showed an excellent performance in total nitrogen (TN) removal under the optimal condition. The activated chlorine generated from chloride had great contributions to eliminate carbon and nitrogen of RhB wastewater. The treatment effluent had a good biodegradability, which was suitable for subsequent biological treatment.

1. Introduction

The negative effect of dyeing wastewater on the ecological environment has received wide concern due to their inherent high toxicity and low biodegradability (Robinson et al., 2001). Various physical, chemical methods such as adsorption, membrane separation, coagulation, flocculation have been adopted to deal with the dyeing wastewater. Biological methods such as white-rot fungi are also capable of decomposing dyes by enzymatic oxidation (Zhang et al., 2016). However, satisfactory effluents could not be achieved as these methods are either expensive, ineffective or inducing secondary pollution (Natarajan et al., 2011). To get a better treatment performance, the new technology known as three-dimensional electrochemical reactor (3DER) was proposed recently. 3DER has been applied in the decomposition of refractory contaminants such as dyeing wastewater for its high efficiency, environmental compatibility and easy operation (Wu et al., 2008).

The 3DER is established based on 2D electrochemical process by packing the particles such as activated carbon, ceramic and γ-Al_2O_3 between the anode and cathode as the third electrode (Zhang et al., 2013). Applying an appropriate external voltage, these particles will be polarized to form amounts of charged microelectrodes, significantly increasing the specific surface area of the electrodes and improving the electrolytic efficiency. Particularly, the 3DER with activated carbon particle electrodes shows a good performance in dyeing wastewater treatment (Ya et al., 2003).

As a complex system, the operation of 3DER are influenced by many parameters (voltage, solution pH, aeration rate etc.), that were optimized by using 'one-at-a-time' strategy in previous studies (Fu et al., 2010). However, this approach is time consuming. Sometimes, contradictory results were obtained as the interactive effects between the influential factors could not been addressed for the optimization of a single factor. For example, it was observed that extensive aeration played a positive role in the electrochemical oxidation of formic acid wastewater (Xiong et al., 2003). While

* Corresponding author.
** Corresponding author.
E-mail addresses: jxu@des.ecnu.edu.cn (J. Xu), xyli@des.ecnu.edu.cn (X.-y. Li).

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other studies indicated that the gas bubbles formation lowered the treatment efficiencies due to the shifts of the concentration profiles (Saleh, 2008). Moreover, for a 3DER, the energy consumption is quite high without optimization of the operation parameters. The energy consumption for dyeing wastewater and petroleum refinery wastewater are reported as high as 865 kWh/kg TOC and 53.65 kWh/kg COD (Liu et al., 2011; Wei et al., 2010), limiting the practical application in wastewater treatment. Thus, the optimal operation parameters of 3DER is needed to enhance the removal efficiency and reduce the energy consumption simultaneously.

The central composite design (CCD) combined with response surface methodology (RSM) may provide a superior alternative for investigation of such a complex system (Caliman et al., 2007). By application this method, it is possible to evaluate the treatment performances comprehensively with a limited number of designed experiments. The interactions between the influential factors of 3DER could be also considered. Furthermore, it is essential to identify the most significant influential parameters of the 3DER affecting contaminants removal process. Grey relational analysis (GRA) is a part of grey system theory. It can be used for searching primary relationships among the influential factors and determining important factors significantly affecting the defined objectives (Zeng et al., 2007). This method has been adopted well to estimate the operation parameters of the wastewater treatment plant relating to the effluents quality (Xu et al., 2011).

In this work, a 3DER packed with granular activated carbon (GAC) particle electrodes was applied for the treatment of simulated dyeing wastewater with high concentration. Rhodamine B (RhB) was selected as a typical dye due to its stable chemical structure (Fig. S1) existing in several industrial wastewater such as textile, leather, jute and food industries. Four operation parameters including voltage, initial pH, aeration rate and NaCl dosage were considered, and COD removal rate and energy consumption were set as the optimal objectives. The compromise optimal conditions for the two responses were obtained using the desirability function approach. And the GRA was used to quantitatively evaluate the significance of these operation parameters. Additionally, the degradation pathway of RhB in 3DER were proposed by intermediates analysis and the effluents were further characterized. The main objective is to optimize the operation conditions of the 3DER, achieving a satisfactory performance in contaminants removal with a reasonable energy consumption. The results would be beneficial to the practical application of 3DER in the wastewater treatment.

2. Materials and methods

2.1. Chemicals and experimental setup

GAC particles were supplied by Jiacheng environmental protection science and technology Co., Ltd., (Zhejiang Province, China). Chemicals of reagent grade were from Sinopharm Chemical Reagent Co., China.

The 3DER was a rectangular tank made of polymethyl methacrylate (PMMA) with an effective volume of 600 mL (60 mm × 100 mm × 100 mm). The main electrodes including a Ti/RuO_2-IrO_2 anode (100 mm × 100 mm × 1 mm) and a titanium plate cathode with the same dimension (Shuerde industrial machinery Co., China) were positioned vertically and parallel to each other with an inner gap of 6 cm. The GAC were packed between the anode and cathode with the filling amount of 125 g. Air was purged into the reactor through a microporous aeration stone at the bottom of the tank and the aeration rate was adjusted by a flowmeter. The voltage was supplied using a digital DC power (CE0036030S, RainWorm, China). In each run, 250 mL RhB solution with the concentration of 1000 mg/L was fed into the 3DER for electrolysis under the designed conditions. NaCl was added into the RhB solution as electrolyte to enhance the electrochemical process. The initial pH of the RhB simulated wastewater was adjusted by H_2SO_4 and NaOH. After electrolysis for 30 min, the effluent was withdrawn from the reactor for further analysis.

2.2. Analytical methods

The COD and BOD of the liquid samples were analyzed on DR3900 and BODTrakII (Hach, USA) respectively. Ammonium nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N) and nitrite nitrogen (NO_2^--N) were determined with selective electrodes (Hach, USA). Total nitrogen (TN) was measured by alkaline potassium persulfate digestion method. All the samples were measured in triplicate. Organic nitrogen (Organic-N) in the solution was calculated as following equation:

$$\text{Organic-N} = \text{TN - } NH_4^+\text{-N - } NO_3^-\text{-N - } NO_2^-\text{-N} \quad (1)$$

The intermediates of RhB decomposition were analyzed by a GC-MS system (Agilent 7890A-5975C, USA). The pretreatment process was described in previous work (Li et al., 2017) as: a 100 mL liquid sample was extracted using 10 mL CH_2Cl_2 three times at pH 2.0, then dehydrated with anhydrous sodium sulfate and dried under nitrogen. The residue was dissolved in 1.0 mL of CH_2Cl_2 for further analysis. For GC system, helium gas was used as carrier in splitless mode, and a DP-5MS capillary column (30 m × 250 μm × 0.25 μm) was adopted. The temperature program followed an initial temperature of 50 °C holding for 5 min, and then ramped to 60 °C at 5 °C/min, ramped to 290 °C at 10 °C/min and finally held at 290 °C for 15 min. For the MS system, electron impact (EI) mode at 70 eV was selected with electron multiplier voltage of 1500 V. The intermediates were analyzed based on the NIST11 mass spectral library database (Diao et al., 2017).

2.3. Energy consumption

Energy consumption dominants the operation cost and determines the feasibility of scaling up to a full scale for the 3DER. In order to evaluate the economic benefits of the 3DER technology, electrical energy consumption was calculated using the following equation:

$$EC = \frac{UIt \times 1000}{(COD_0 - COD_t) \times V} \quad (2)$$

where U is voltage (V), I is the current (A) and t is the treatment time (h), V is the volume of wastewater treated (L), COD_0 and COD_t are the initial and final concentrations of the wastewater calculated as COD (mg/L).

2.4. Experimental design with RSM

Four operation parameters including voltage (X_1), initial pH (X_2), aeration rate (X_3) and NaCl concentration (X_4) were selected as the independent input variables and further optimized by CCD combined with RSM. For statistical calculations, the variables were coded according to the following equation: