A continuous-flow sparged packed-bed electrocoagulator for dye decolorization

Chantaraporn Phalakornkule [a,b,*], Tanapat Luanwuthi [a], Paschanok Neragae [a], Elvin J Moore [c]

[a] Department of Chemical Engineering, Faculty of Engineering, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand
[b] The Research Center for Renewable Energy and Product, Science and Technology Research Institute, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand
[c] Department of Mathematics, Faculty of Applied Science, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand

ARTICLE INFO

ABSTRACT

A sparged packed-bed electrocoagulator was developed for decolorization of dye wastewater. The anode of the electrocoagulator was a perforated polyvinyl chloride (PVC) pipe packed with iron nuts. At the center of each anode, a gas sparging tube was installed, and the hydrogen generated from the oxidation reduction reactions was recycled back to the reactor as the sparging gas. When running with a DC power supply, the operation of the packed-bed electrocoagulator with gas sparging can both enhance the decolorization efficiency and reduce the resistance and the energy consumption of the operation. When running with a photovoltaic (PV) panel, the wastewater feed flowrate was adjusted in response to the varying PV output current. The gas sparging was found to help enhance mass transfer rates of the electroactive ions to the bulk solution, and to make the decolorization efficiency independent of the varying feed flowrates.

© 2016 Taiwan Institute of Chemical Engineers. Published by Elsevier B.V. All rights reserved.

1. Introduction

Wastewater treatment in the textile industry appears to be well developed. Aerobic treatment of dye-containing wastewater is widely used in the textile industry. Even though aerobic treatment can typically remove most of the biochemical oxygen demand (BOD) and chemical oxygen demand (COD) in dye-containing wastewater, the biological treatment neither destroys nor removes dye molecules and their color from the wastewater [1]. Considerable research effort has been aimed at improving the existing treatment processes, *e.g.* to increase the efficiency of color removal. Low voltage direct current electrocoagulation is one of the techniques that has been extensively studied [2–4]. The electrocoagulation technique typically removes 98–100% of color in dye-containing wastewater [5–23]. Surface complexation and electrostatic attraction between dye molecules and the electroactive monomeric ions, metal hydroxides and polymeric hydroxyl complexes, are the underlying mechanism for color removal in the electrocoagulation technique [24]. The generation of metal ions from electrochemical oxidation of

metal electrode and the spontaneous conversion of the metal ions to the electroactive species do not produce secondary pollutants such as sulfate or chloride ions in the water, making the technique more environmentally friendly than more traditional coagulation processes [25].

As attraction and complexation between dye molecules and the electroactive species are the underlying mechanism, the rate of mass transfer of metal ions from the electrode surface to the bulk solution is an important factor. Mass transfer at the electrode surface is typically diffusion controlled [26]. In batch experiments, mechanical stirring is a convenient method for enhancing the transfer of metal ions from the electrode surface to the bulk solution. However, mechanical stirring cannot be easily implemented in most designs of continuous-flow electrocoagulators [24]. For continuous-flow electrocoagulators, it is necessary to develop mechanisms of mass-transfer enhancement that are suitable for particular electrode configurations, *e.g.*, for electrodes such as steel wool cathode [27], wrapped iron wire netting [28] and packed-bed anode [29] that have complex shapes. Mechanisms of mass-transfer enhancement are also important in electrocoagulators operated in a continuous flow mode and with time-varying flow velocities. The latter case may occur in the operation of an electrocoagulator powered by solar PV panels because the flowrates are adjusted to the fluctuating solar energy incident on the PV solar panels [30,31].

* Corresponding author at: Department of Chemical Engineering, Faculty of Engineering, King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand. Tel.: +66 8 9 135 3253; fax: +66 2 587 0024.

E-mail address: cpk@kmutnb.ac.th, cphalak21@yahoo.com (C. Phalakornkule).

In this study, a continuous-flow packed-bed electrocoagulator was developed. In order to reduce the increased concentration overpotential associated with the packed-bed anode, a gas sparging tube was installed at the center of each packed-bed anode, and the hydrogen product from the underlying redox reactions was recycled back to the reactor for gas sparging. The color and COD removal efficiencies of a continuous-flow sparged packed-bed electrocoagulator with a DC power supply were investigated for treatment of a synthetic wastewater containing Reactive Blue 21. The color and COD removal efficiencies of a continuous-flow sparged packed-bed electrocoagulator with solar power and time-varying flow velocities were also investigated.

2. Materials and method

2.1. Synthetic wastewater

A blue reactive dye (Reactive Blue 21) supplied by a textile industry in Thailand was used to prepare synthetic wastewater by dissolving 500 mg/L of dye into tap water, and then adjusting the pH with NaOH to 8.3±0.1 and the conductivity with NaCl to 2 mS/cm. Reactive Blue 21 belongs to the phthalocyanine class, and its chemical structure is given in the supplementary material. An initial concentration of 500 mg/L was selected because it provided a COD value of the same order of magnitude as that of actual wastewater from a textile factory [32]. The values of pH and conductivity were selected to represent the most extreme pH and conductivity conditions for electrocoagulation of the real textile wastewater, namely high pH and low conductivity.

2.2. Reactor configurations

As shown in Fig. 1, the continuous-flow packed-bed electrocoagulator consisted of three packed-bed anodes and four tubular cathodes. Each anode was a perforated PVC pipe packed with iron nuts. The PVC pipe had an internal diameter of 28 mm, a length of 970 mm and a thickness of 3 mm. The packed-bed anode had an apparent density of 3376 kg/m^3 and a void fraction of 0.57. The holes on the perforated PVC pipe were 5 mm in diameter (Fig. 1a). A gas sparging tube was installed at the center of each anode. Each gas sparging tube had an internal diameter of 4.0 mm and a length of 1000 mm and had four 2 mm-holes along its height (Fig. 1b). Each cathode was a stainless steel tube with an internal diameter of 16.7 mm, a length of 1000 mm and a thickness of 1.2 mm. The three packed-bed anodes and four cathodes were assembled as an electrode set with the configuration shown in Fig. 1c. The electrode set was then placed inside a 7.85-L acrylic column with an internal diameter of 100 mm and a height of 1000 mm.

The continuous-flow electrochemical reactor was designed to work by an upflow process. The dye-containing wastewater was fed in at the bottom of the reactor, flowed upward through the electrode set and drained out at the outlet located at the top of the reactor. Fig. 2 shows the components of the system which consist of a feed pump, the electrochemical reactor, a power supply, a gas–liquid–solid separation tank and a gas line-valve system. The power supply can be either a DC power supply or a 200 W-PV array (Fig. 2).

2.3. Experimental procedures

2.3.1. Operation with a DC power supply
The DC power supply was first set up to provide a voltage of 7.4 V. The feed flowrate was controlled between 0.1–1 L/min to provide hydraulic retention times (HRT) for the treatment of the wastewater in the electrocoagulator of 5.7, 6.3, 7.1, 8.1, 9.5, 11.4,

14.2, 19.0, 28.5 and 57.0 min. For each specified HRT time, the system was run until a steady-state performance was achieved. Current and voltage across the circuit were monitored throughout the course of the experiments. The effluent was collected at the reactor outlet and was allowed to stand at room temperature for 20 min. The supernatant was then collected for the analysis of water properties.

2.3.2. Operation with a 200 W-PV array
The electrochemical reactor was directly powered by a 200 W-PV array without a storage battery system, a charge regulator or controller. Current and voltage across the circuit and the temperature at the surface on the PV array were monitored throughout the course of the experiment. The feed flowrate was controlled in response to fluctuations of the current supplied by the PV array so that the ratio of current to flowrate (J_V^*) remained in an appropriate range. The J_V^* is defined as follows:

$$J_V^* = \frac{I}{Q} \left(\frac{A}{\frac{m^3}{s}} \right) \text{ or } \left(\frac{C}{m^3} \right) \qquad (1)$$

That is, J_V^* represents the amount of electrical charge passed per unit volume of the feed. As in the operation with the DC power supply, the effluent was collected at the reactor outlet and was allowed to stand at room temperature for 20 min before the supernatant was collected for the analysis of water properties.

2.3.3. Pattern of gas sparging and the employed frequencies
In order to recycle the hydrogen product back to the electrochemical reactor for its use as the sparging gas, the pressure difference between that in the gas line and that in the electrochemical reactor was set at 1.2 bar (absolute) through the manipulation of valves. The gas sparging cycle consisted of four steps: (1) normal operation with continuous feed and effluent flow; (2) build up of pressure in the gas line to 1.2 bar; (3) recycling and sparging hydrogen and (4) release of built up back pressure in the system.

Three frequencies of gas sparging were employed: (1) continuous operation of the gas sparging cycle; (2) a cycle of gas sparging followed by release of built-up pressure and (3) a cycle of gas sparging followed by release of built-up pressure for twice the sparge time.

2.4. Estimation of experimental and theoretical amounts of hydrogen product

The amount of gas product from the experiments was estimated from the ideal gas law: $PV_{\text{gas space}} = nRT$. P is the pressure built up in the system recorded by a pressure gauge, $V_{\text{gas space}}$ is the system volume occupied by gas, T is the system temperature, R is the ideal gas constant, and n is the number of moles. The molar amount of hydrogen was calculated from the product of the number of moles and the product gas composition on a mole fraction basis.

The theoretical amount of hydrogen generated was estimated from the stoichiometry of the oxidation and reduction reactions at anodes and cathodes. When 1 mole of Fe dissolves, 2 moles of electrons are produced by oxidation at the anode, and 1 mole of hydrogen is produced by water electrolysis at the cathode. The relationship between the supplied current and the theoretical amount of hydrogen generated is:

$$n_{H_2} = \frac{I \cdot t}{F} \cdot H \qquad (2)$$

where n_{H2} is the amount of hydrogen generated (mole), I is the supplied current (A), t is the electrocoagulation time (s), F is Faraday's constant (96,500 C per mole of electrons), H is the number of hydrogen molecules generated per electron involved in the redox reactions. H is equal to $\frac{1}{2}$ [33].