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Ozone assisted electrocoagulation in a rectangular internal-loop airlift reactor: Application to decolorization of acid dye

Jamshid Behin*, Negin Farhadian, Mojtaba Ahmadi, Mehdi Parvizi

Department of Chemical Engineering, Faculty of Engineering, Razi University, Kermanshah, Iran

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ABSTRACT

Decolorization of synthetic wastewater containing Acid Brown 214, a typical triazo acid dye, was carried out by ozone assisted electrocoagulation as an advanced oxidation process. Ozonation and electrocoagulation were performed simultaneously in the riser and downcomer sections of a lab-scale rectangular airlift reactor, respectively, where ozonated gas had no influence on dielectric constant. A good mixing was achieved with the liquid circulation induced by gaseous ozone, without the need of mechanical mixing. Taguchi experimental design method with orthogonal arrays L27 and the "larger is better response" category were applied to determine optimum conditions for dye removal. The operating variables considered were the initial pH, initial dye concentration, salt concentration, current density, superficial gas velocity and decolorization period with three levels. Under optimum conditions, the synergistic effect of the combined process reaching complete decolorization in 30 min with energy consumption 7.4 kWh/kg dye removed. Results indicated that combination process in innovative configuration of single airlift reactor is cost effective as compared with that obtained individually in separate conventional reactors.

1. Introduction

Synthetic organic dyes are one of the major pollutants and water contaminants in effluents of textile, leather, paper, plastic, printing, food, and mineral processing industries [1–3]. Aromatic dyes, especially azo dyes, cause severe ecological problems and are classified as environmentally hazardous materials due to their toxicity and slow degradation [4–6]. Treatment of industrial effluents containing carcinogenic and mutagenic azo dyes is necessary prior to their final discharge to the environment and meets the stringent environmental regulations [7–9].

Conventional methods for removing organic dyes from wastewater consist of biological, physical, chemical/electrochemical treatments and their combinations, but these methods are not as efficient as advanced oxidation processes (AOPs) [10–13]. The AOPs generally use a combination of oxidation agents (such as hydrogen peroxide, oxygen or ozone), irradiation (such as ultraviolet or ultrasound), catalysts (such as metal ions, metal oxides like titanium oxide) and adsorbents (such as activated carbon) [14–24]. AOPs primarily involve the generation of a very powerful and non-selective oxidizing agent, the hydroxyl radical, $OH^{\bullet}$, which is readily available to react instantly with any organic matters present in water, like the dyes [25–28].

Among AOPs, the integration of ozonation (O) and electrocoagulation (EC) treatment has an excellent potential of decolorization and resulted in increased removal efficiency due to the following reasons: (1) no need for the external addition of flocculating agents since they are electrochemically generated inside the treatment vessel; (2) minimal pH changes during the treatment process not requiring pH adjustment; (3) a significantly lower amount of sludge since only Fe or Al ions were released in the solution; (4) flocs formed by the electrocoagulation can be easily separated by the filtration; (5) the final effluent is clear, colorless and odorless [29,30].

The airlift reactor (ALR) has been regarded as a promising type of gas-liquid reactor due to the good mixing with low shear stress and energy consumption as well as the advantages of high gas-liquid mass transfer [31]. There are few reports make reference to the use of external ALR for dye removal by EC alone [32–36]. Further, Few researchers have investigated the ozone assisted electrocoagulation (OEC) process for the treatment of synthetic effluent and wastewater [37–43]. They used conventional reactors such as stirred tank or bubble column reactors to perform the OEC process. Based on our knowledge, the conventional decolorization reactors used in literature have some disadvantages which are presented

* Corresponding author. Department of Chemical Engineering, Faculty of Engineering, Baghe Abrisham, Kermanshah, Iran. Fax: +98 831 8250167
E-mail address: Behin@razi.ac.ir (J. Behin).

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Table 1
Limitation of various decolorization methodologies (O: ozonation, EC: electrocogulation, OEC: ozone assisted electrocogulation).

Process	Reactor	Limitations
O	Bubble column	- Required large amount of energy due to high reaction time
EC	Conventional cell	- Flocs move downward by settling or upward by flotation - The recovery of the floc obtained by the EC needs a long time or an additional secondary treatment (like filtration or sedimentation) - Needs to mechanical mixer
	External-loop ALR	- Liquid recirculation induces only by H_2 gas microbubbles - Mixing conditions is acceptable
OEC	Two batch reactors Stirred or bubble column	- Needs to a relatively high treatment period - Ohmic resistance between electrodes due to presence of ozone gas bubbles

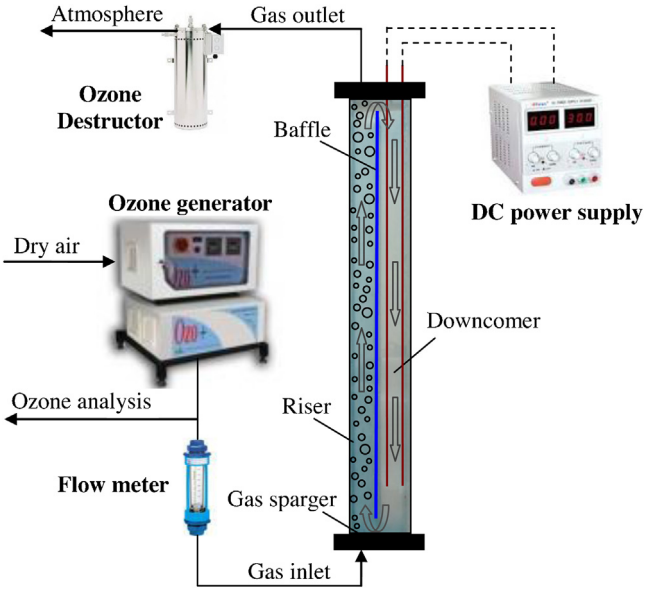

Fig. 1. Schematic of experimental OEC set-up.

in Table 1. However, the reactor with more efficient mass transfer was rarely used.

The objective of this study was to investigate the feasibility of using internal ALR for removal of acid brown 214 using hybrid technique OEC system. Moreover, in order to determine the influence of the operating variables such as initial pH, initial dye concentration, current density, salt (Na_2SO_4) concentration and superficial gas velocity on the decolorization efficiency, the statistical evaluation of experimental results was conducted using Taguchi method as an efficient experimental design. The experiments were carried out according to L27 orthogonal arrays experimental design with two replicate. Statistical calculations were done using MINITAB software.

2. Material and methods

Ozone was produced from dried air using an ozone generator module 20 G/H (Arda, France). Its flow rate was measured by a rotameter before entering the reactor and its concentration was determined using an iodimetric method [44]. The gas flows through an antenna sparger which consists of 4 holes each with 0.25 mm diameter. The glass reactor has a volume of 1.5 L and consists of a square cross sectional area of 10 cm × 3 cm and height of 50 cm (Fig. 1). The riser and the downcomer separated by straight rectangular baffle. The bottom clearance was 4 cm and the top clearance was 6 cm. The riser of ALR serves as ozonation cell. A pairs of flat iron plates with a total anodic surface area of 30 cm^2 was used as electrode and placed in downcomer as an electrocogulation cell. The distance between two electrodes was 1 cm. The electrodes were connected to a digital DC power supply equipped with current-and voltage-reading meters to supply the current options. Ozone in the gas phase leaving the reactor was removed in a thermal gas ozone destructor.

The dye used in this study was acid brown 214 (AB214) obtained from Boyakh Saz Company (Iran) and its chemical structure and characteristics are shown in Table 2.

A stock solution of AB214 was prepared by dissolving an accurate quantity of dye in deionized water (3 g/l). The initial liquid pH was adjusted using minute addition of 0.1 M H_2SO_4 or NaOH solutions. All the chemicals used were analytical grade and all experiments were conducted at room temperature (20 ± 2 °C) and atmospheric pressure until the steady state condition was achieved. Before each experimental run, the iron electrodes were immersed in dilute HCl solution for 5 min to remove oxide and were rinsed with deionized water. Then, they were polished by sandpaper to further remove oxide. The current intensity was then set to the desired value and the voltage was automatically regulated to match the current value. To monitor degradation, 10 ml volume samples were collected at regular time intervals and centrifuged (2000 rpm, 7 min) for sequent analysis.

2.1. Decolorization criterion

The optical absorption of the AB214 dye in water with 100 mg/L concentration shows a maximum absorbance (λ_{max}) at 485 nm. Dye concentration was measured at the wavelength corresponding to maximum absorbance, using spectrophotometer (Unico, UV-2100, USA). The removal yield of AB214 was determined using following equation:

$$Y = \frac{A_o - A_t}{A_o} \quad (1)$$

where,

Y: Decolorization yield (%)

A_o: The light absorbance of dye before decolorization (−)

A_t: The light absorbance of dye afterdecolorization (−)

Specific energy consumption (E_{dye}) per kg of dye removed was estimated as follow [45]:

$$E_{dye} = \frac{U \cdot I \cdot t}{V \cdot C_o \cdot Y}$$

where,

C_o: Initial dye concentration (kg/m^3)

I: Current intensity (A)

Y: Decolorization yield (%)

U: Measured cell potential (V)

V: Liquid volume (m^3)

t: Electrolysis time (h)

E_{dye}: Specific energy consumption (Wh/kg dye removed)