

# Removal of color from real dyeing wastewater by Electro-Fenton technology using a three-dimensional graphite cathode

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## Abstract

This work investigates the removal of color from wastewater that contains low dyestuff concentrations by the Electro-Fenton process. The color was removed by in situ electrogenerated hydrogen peroxides at a three-dimensional graphite cathode with added ferrous sulfates. Experimental runs were conducted to evaluate the effect of the operating parameters, such as the oxygen contact mode, the oxygen sparging rate, the applied current density, the concentration of ferrous ions, the solution temperature and the pH among others, on the removal of color. The removal efficiency of the color in the cathodic chamber reached 70.6% under specified operation conditions in 150 min. The removal efficiency was controlled by the mass transfer when the oxygen-sparging rate was less than 0.3 dm<sup>3</sup>/min for the reactor configuration herein. The optimal applied current density was 68 A/m<sup>2</sup> when the energy consumption was considered. The highest removal efficiency was obtained by adding 20 mM Fe(II) to the solution. The optimal solution pH was 3 in this work. The temperature negatively affected color removal.

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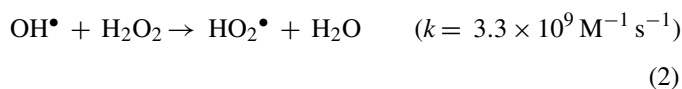
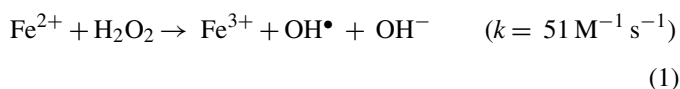
**Keywords:** Electro-Fenton technology; Color removal; Dyeing wastewater; Graphite cathode; Hydrogen peroxide; Three-dimensional electrode

## 1. Introduction

The textile dyeing and finishing industry is a major industrial water consumer. The satisfactory treatment of the effluent from the dyeing industry is difficult because of the effluent's strong color and high COD. Although several traditional approaches are known for the decomposition of the dye in wastewater, some limitations still exist. Interest in the use of electrochemical methods to treat wastewater has been increasing in recent years [1–10]. The most efficient electrochemical process proceeds by indirect anodic oxidation of the treated chemicals. The traditional indirect oxidation deals with the use of chloride ions as redox mediators [11,12]. Although it is very effective in treating some organics and inorganics, its main shortcomings is that some toxic substances are likely to be formed. Hydrogen peroxide is a green chemical and can be usually cathodically generated from dissolved oxygen in alkaline media. Therefore, several researchers used the in situ electrogenerated hydrogen peroxide to destroy

the organics, even though its oxidizing power is not very strong [13–16].

Fenton's reagent is an acidic mixture of hydrogen peroxide and Fe(II). The use of Fenton's reagent to destroy partially or completely the organic compounds is promising. The main reaction mechanism of the formation of hydroxyl free radicals in Fenton's reagent is very complex and may be described by a series of chain reactions [17,18]:



Despite the high oxidative efficiency of Fenton's reagent, its application is limited by the storage and shipment of con-

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centrated  $\text{H}_2\text{O}_{2(\text{aq})}$  and the generation of Fe(III) sludge. In situ electrochemical production of  $\text{H}_2\text{O}_2$  in acidic media and regeneration of Fe(II) by the simultaneous reduction of oxygen and Fe(III) on a cathode can solve this problem. In recent years, several researchers have electrochemically produced considerable amounts of hydrogen peroxide in mildly acidic media. Therefore, the formation of Fenton's reagent becomes feasible. The technology has been called Electro-Fenton technology [19–26]. The efficiency of oxygen reduction at the cathode is crucial in the Electro-Fenton process [27]. Oxygen can be reduced at the cathode in two ways, yielding two products. The electrochemical reduction of oxygen in the two- or the four-electron process depends strongly on the cathode materials. The carbon-based electrode and the gas-diffusion electrode have been reported to have good current efficiencies in the production of hydrogen peroxide [12–16].

In addition to the electrode materials, an increase in the electrode surface is important in the mass production of hydrogen peroxide. In this aspect, a three-dimensional electrode is attractive in industrial applications over a two-dimensional electrode, for its large electrode surface and higher mass transfer. However, little work using a three-dimensional graphite cathode to produce hydrogen peroxide in acidic media was done. Furthermore, most of the previous studies focused on the treatment of alkaline lab-made wastewater of a simple composition. The data of using the Electro-Fenton technology to treat the real wastewater is still limited [20].

This work investigates the removal of the color from the real dyeing wastewater by the Electro-Fenton technology. The hydrogen peroxide was generated in situ at the new flow-by three-dimensional graphite cathode. The Fe(II) was added externally. The electrochemical cell was of the divided type to ensure that the color removal was by the oxidants generated in the cathodic chamber. The constant-current mode was adopted to evaluate the efficiency of color removal. We are concerned with the efficiency of the three-dimensional graphite cathode and the influences of several operating parameters on the color removal from the real dyeing wastewater in this study.

## 2. Experimental

Fig. 1 schematically depicts the experimental setup of this work. The electrochemical generation of hydrogen peroxide and the subsequent removal of the color of the wastewater were performed in a divided flow-by cell. The cell was made of 0.2 cm thick acrylic material, 15 cm high, and was 5 cm long and 5 cm wide. It was divided into an anodic and cathodic chamber with a separator made of a plastic plate, 0.1 cm thick. A cation-permeable membrane Nafion<sup>®</sup> 417 was employed to prevent mixing between the anolyte and catholyte. The volume of the catholyte reservoir exceeded that of the reaction zone of the electrochemical flow cell, so the behavior of the whole system was approximated as that of a well-mixed batch reactor. The cathodic chamber was packed randomly with 50 graphite Rasching rings (Wei-Chang Carbon) with a total surface area of 220 cm<sup>2</sup> for use as the three-dimensional cathode. The anode used in this work was a Pt/Ti plate (8 cm × 2 cm × 1 cm). All

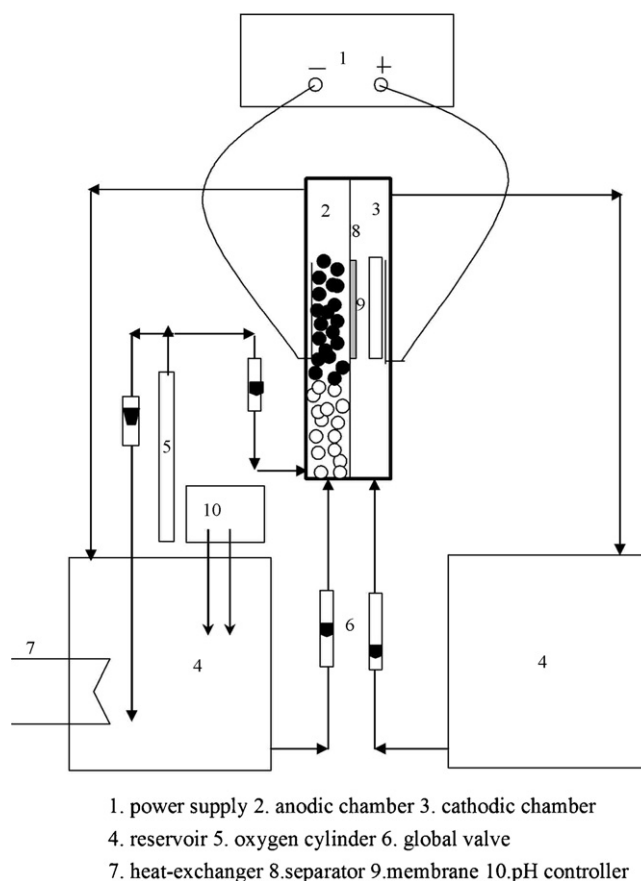


Fig. 1. The schematic diagram of the experimental setup.

of the chemicals used were of reagent grade. Two current feeders, made of titanium plates, were placed in the anodic and the cathodic chamber. Small glass beads (0.3 cm in diameter) were packed at the bottom of the cathodic chamber to increase the uniformity of the flow velocity distribution.

The graphite cathodes and the Pt/Ti anode were pickled in the alkaline solution (1 M NaOH) and acidic solution (1 M  $\text{H}_2\text{SO}_4$ ) alternatively for cleaning, and were then rinsed with deionized water before the electrolysis was conducted. The solution flowed in a recirculating loop. A catholyte volume of 1.5 dm<sup>3</sup> was introduced into the bottom of the cathodic chamber using a pump; flowed out of the top, and returned to the reservoir. The flow conditions of the anolyte were the same as those of the catholyte. The flow rate of each run was controlled. The oxygen was introduced into the bottom of the cathodic chamber to constitute a three-phase graphite cathode. The temperature of the solution was controlled using a heat exchanger.

The mixed wastewater samples were taken from a textile plant located at Tainan Hsien in Taiwan and then stored in a refrigerator 4 °C. Most of the dyes used in the mill are direct dyes. The exact composition of real wastewater has seldom been examined. The mixed wastewater was first filtered using the cellulose acetate filter papers with 0.45 μm pore diameter. Some of the original characteristics of the wastewater were analyzed. The measured wastewater characteristics were as follows: ADMI color value = 1094, pH = 6.2, COD = 2942 mg/dm<sup>3</sup>,  $[\text{Cl}^-] = 238 \text{ mg/dm}^3$ . Various amounts of ferrous sulfate were

added into the reservoir to provide ferrous ions in electrolysis. The direct current was from a DC power supply (Istec, Model GPR-M/H-D Series). The electrolytic duration was 150 min in all experimental runs. The color value was obtained by the ADMI Tristimulus Filter method [28]. It was carried out using a spectrophotometer (Perkin-Elmer, Lambo 40). The pH of the solution was measured and controlled by adding 0.1 M HCl<sub>(aq)</sub> and 0.1 M NaOH<sub>(aq)</sub> during the experiment using a pH controller (Istec, Model 310P). At regular time intervals, 2 ml samples of solution were extracted from the reservoir for measurement.

### 3. Results and discussion

The production rate of hydrogen peroxide plays an important part on the treatment efficiency when using Electro-Fenton technology. In the cathodic process, several possible reactions may occur at the cathode, as presented in Fig. 2. The oxygen must be first transported to the aqueous phase. The dissolved oxygen is transferred to the cathode surface, and finally adsorbed and reduced electrochemically to hydrogen peroxides in acidic media.



Three possible side reactions may occur simultaneously at the cathode.

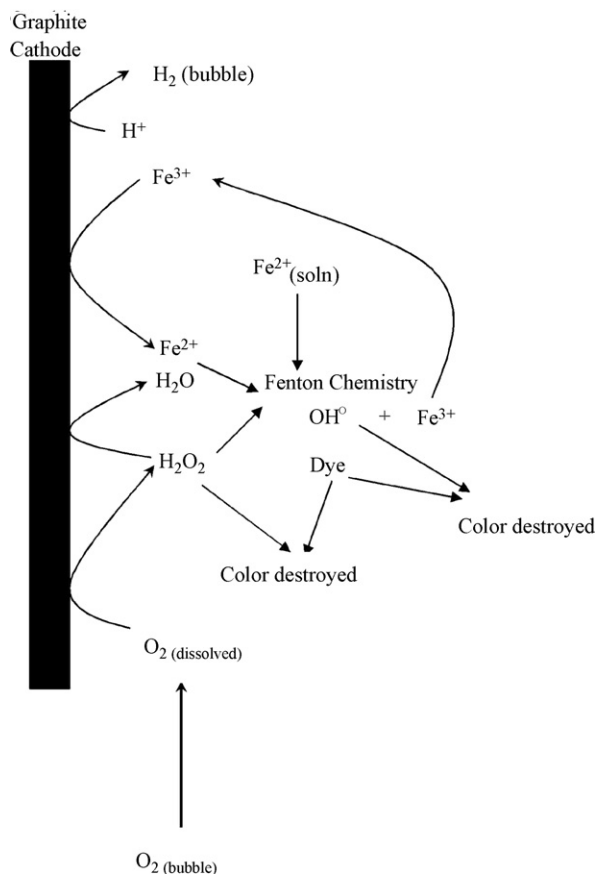
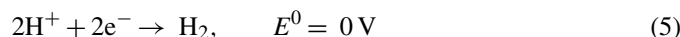
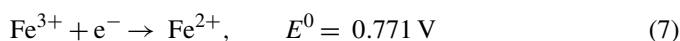
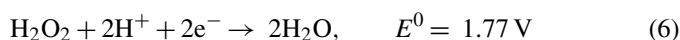


Fig. 2. The possible reactions at the three-phase graphite cathode and in the bulk solution.



The generated hydrogen peroxide reacts with the ferrous ions, produced by the reduction of ferric ions, as described in Eq. (7), or those present originally in the bulk solution, producing the hydroxyl radicals, according to Eq. (1). The hydroxyl radicals are strong oxidants for the oxidation of the pollutants in wastewater.

#### 3.1. Effect of oxygen contact mode

Fig. 3 plots the results of color removal using different oxygen contact modes. In the case of gas–liquid–solid three-phase contact mode (case 1), the solution that surrounds graphite electrodes was saturated with the oxygen gas bubbles sparged at the bottom of the electrochemical reactor. For comparison, the liquid–solid two-phase contact mode (case 2) was investigated. The above mode (case 2) was implemented by sparging the oxygen into the reservoir, such that no oxygen gas bubbles reached the cathode surface. The results show that the three-phase mode operation was more efficient. The removal efficiency was increased with increasing time in both cases. After 150 min of electrolysis, the removal efficiencies in the three-phase contact mode and the two-phase contact mode were 70.6% and 52.5%, respectively. The color removal efficiency in the three-phase contact mode was around 1.33 times that in the two-phase contact mode. The difference between the color removals was probably attributable to the higher dissolved oxygen concentration around the graphite cathode and enhancement of mass transfer by the flow of oxygen bubbles in the three-phase contact mode. Fig. 3 also shows the result obtained without a continuous

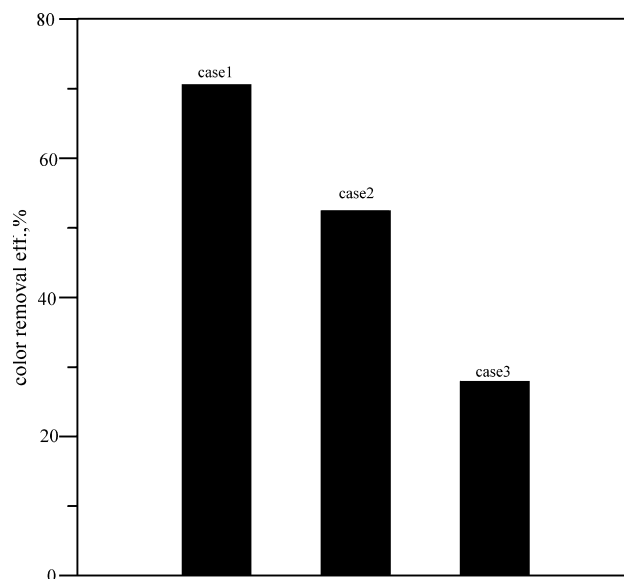


Fig. 3. Effect of the oxygen contact mode on the color removal; initial pH=3; oxygen sparging rate: 0.3 dm<sup>3</sup>/min; applied current density: 68 A/m<sup>2</sup>; [Fe<sup>2+</sup>]=15 mM; 25 °C; case 1: three-phase contact mode; case 2: two-phase contact mode with a continuous oxygen supply; case 3: two-phase contact mode without a continuous oxygen supply.

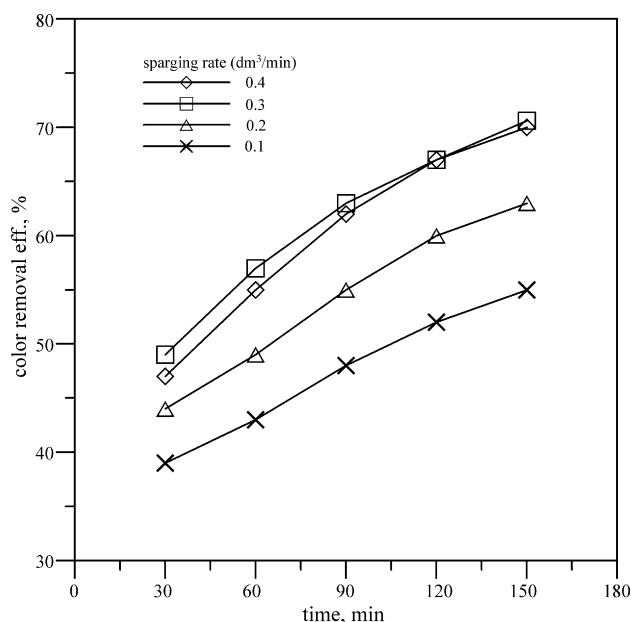


Fig. 4. Effect of the oxygen sparging rate on the color removal; initial pH = 3; applied current density: 68 A/m<sup>2</sup>; [Fe<sup>2+</sup>] = 15 mM; 25 °C.

supply of oxygen gas during the electrolysis (case 3). After 150 minutes of electrolysis, the removal efficiency was about 28%.

### 3.2. Effect of oxygen sparging rate

Fig. 4 displays the experimental results for the oxygen sparging rates ranging from 0.1 to 0.4 dm<sup>3</sup>/min. The color removal efficiencies were about 55.3%, 63.2%, 70.0%, and 70.6% for sparging rates of 0.1, 0.2, 0.3 and 0.4 dm<sup>3</sup>/min, respectively, at a current density of 68 A/m<sup>2</sup>. The removal efficiency increased with the oxygen sparging rate. The result is consistent with the report from Do and Chao [16] using a CSTER. Although the oxygen sparging rate increased from 0.3 to 0.4 dm<sup>3</sup>/min, the removal efficiency remained almost constant at a current density of 68 A/m<sup>2</sup>. The results indicate that color removal began to be controlled by the kinetics of the production of hydrogen peroxides when the oxygen sparging rate exceeded 0.3 dm<sup>3</sup>/min. Clearly, an oxygen sparging rate of 0.3 dm<sup>3</sup>/min at a current density of 68 A/m<sup>2</sup> was adequate to maximize the removal rate in this work.

### 3.3. Effect of current density

Fig. 5 shows the effect of current density on the color removal, which was significant. Increasing the current density increased the production rate of hydrogen peroxide on the cathode. Therefore, the color removal efficiency increased with the current density. In this work, the removal efficiencies were 56.1%, 58.2%, 63.5%, 69.2%, 70.0% and 70.8% for current densities of 11, 23, 34, 57, 68 and 80 A/m<sup>2</sup>, respectively. The results further indicated that the effect of the apparent current density on the removal efficiency was insignificant when the apparent current density exceeded 68 A/m<sup>2</sup>, indicating that the production rate of hydrogen peroxide was controlled by mass transfer when

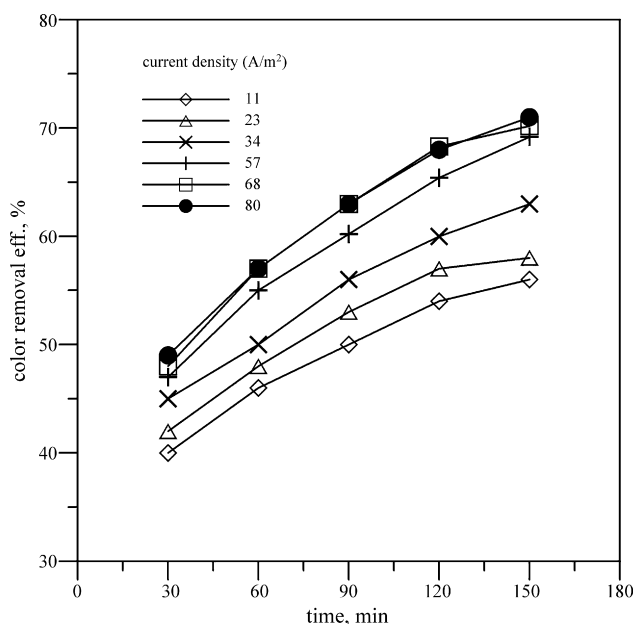


Fig. 5. Effect of the applied current density on the color removal; initial pH = 3; oxygen sparging rate: 0.3 dm<sup>3</sup>/min; [Fe<sup>2+</sup>] = 15 mM; 25 °C.

the current density exceeded 68 A/m<sup>2</sup> at a gas sparging rate of 0.3 dm<sup>3</sup>/min. Since the consumption of electric energy increased with the applied current, the best applied current density in this system was 68 A/m<sup>2</sup> when the accepted removal efficiency and the energy consumption were considered simultaneously. In this case, the removal efficiency was 70% and the energy consumption was 20 kWh/m<sup>3</sup> after 150 min.

### 3.4. Effect of initial Fe(II) concentration

Fig. 6 plots the effect of the concentration of the externally added ferrous ions on the color removal efficiency. The color removal efficiency was poor (9%) when no ferrous ions were added to the solution. This result was expected because the hydrogen peroxide is not a very strong oxidant. The oxidizing power of hydrogen peroxide was not enough to destroy large molecules, such as dyestuffs. A series of experiments was performed to investigate the amount of Fe(II) added for color removal at the start of the electrolysis. The added amounts of ferrous ions were related to the amounts of hydroxyl free radicals produced, according to Eq. (1). In Fig. 6, the color removal efficiency was markedly increased from 9% to 46% by externally adding a small amount of ferrous ions (5 mM). The removal efficiency of color should increase with the amount of added ferrous ions. However, the color removal efficiency did not increase significantly as the dosage of ferrous ions increased from 15 and 20 mM. In comparison with the results obtained from the treatment of synthetic wastewaters by the Electro-Fenton technology, a very high Fe<sup>2+</sup> content as 15 mM was added to obtain the highest color removal efficiency. This can be ascribed to the formation of complexes of Fe(II) and Fe(III) with acids, alcohols, etc. present in the initial effluent or formed during degradation. Furthermore, the concentration of ferrous ions negatively affected on the color removal at [Fe<sup>2+</sup>] > 20 mM. A possible

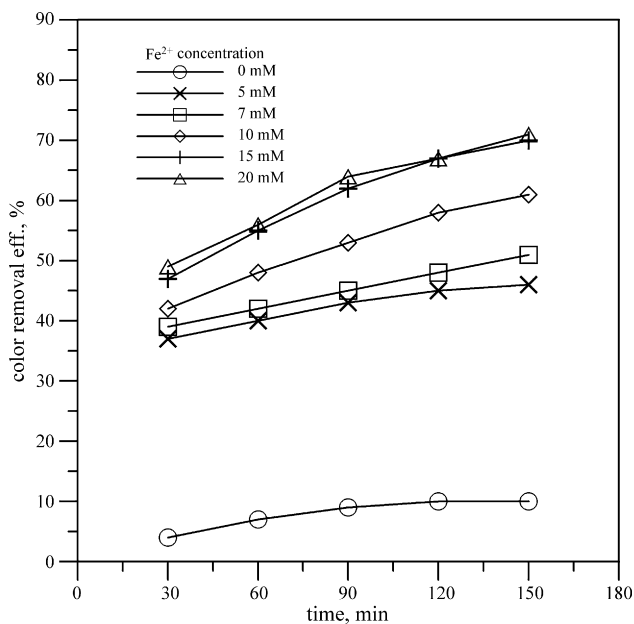


Fig. 6. Effect of the added Fe(II) concentration on the color removal ratio; initial pH = 3; oxygen sparging rate:  $0.3 \text{ dm}^3/\text{min}$ ; applied current density:  $68 \text{ A/m}^2$ ;  $25^\circ\text{C}$ .

explanation is as follows. When the concentrations of the ferrous ions and hydroxyl free radicals are high, the ferrous ions can react with the hydroxyl free radicals according to Eq. (3). The rate constant in Eq. (3) has a value of several orders of magnitude higher than that in Eq. (1). Hence, the excess ferrous ions consumed the hydroxyl free radicals with a high oxidative potential. Additionally, the active sites on the electrode surface are presumably occupied by the ferric ions probably generated by Eqs. (3) and (7). The number of effective sites on the cathode surface for hydrogen peroxide production was thus reduced. The experiments also demonstrated that some colloidal particles were formed as the electrolysis time increased when the concentration of ferrous ions exceeded  $40 \text{ mM}$ .

### 3.5. Effect of temperature

Fig. 7 displays the effect of temperature on the removal of color. The temperatures used herein were  $25^\circ\text{C}$ ,  $35^\circ\text{C}$  and  $45^\circ\text{C}$ . The result shows that the temperature negatively affected the color removal efficiency. The color removal efficiencies decreased from  $70.6\%$  to  $39.1\%$  as the temperature increased from  $25^\circ\text{C}$  to  $45^\circ\text{C}$ . Since the color was removed by an indirect process, the removal rate was essentially limited by the rate of production of hydrogen peroxide. The negative effect of temperature on the production of hydrogen peroxide can be explained by the lower concentration of dissolved oxygen and the self-decomposition of hydrogen peroxide. The concentration of hydrogen peroxide decreased as the temperature was increased because of the decrease in the concentration of dissolved oxygen. Hence, increasing the temperature lowered the color removal efficiency. Additionally, the rate of self-decomposition of the hydrogen peroxide to water and oxygen increased with the temperature. A  $10^\circ\text{C}$  rise approximately dou-

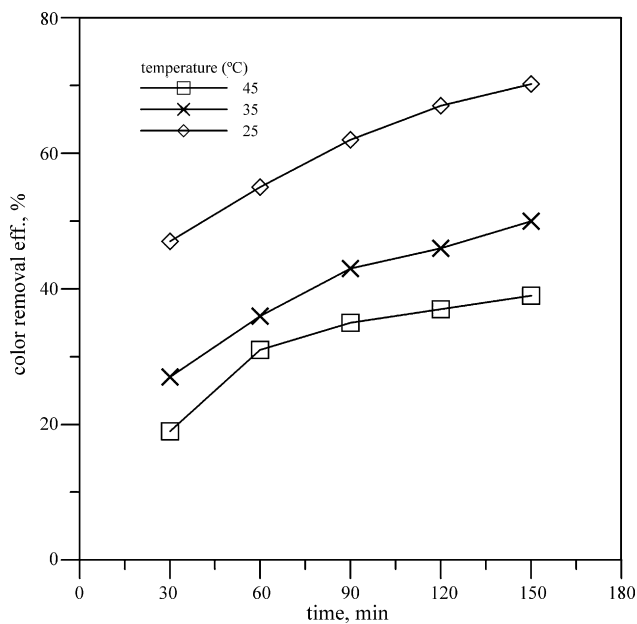


Fig. 7. Effect of the temperature on the color removal; initial pH = 3; oxygen sparging rate:  $0.3 \text{ dm}^3/\text{min}$ ; applied current density:  $68 \text{ A/m}^2$ ;  $[\text{Fe}^{+2}] = 15 \text{ mM}$ .

bles the decomposition rate. In this respect, a lower temperature favored the production and accumulation of hydrogen peroxide, thereby increasing the rate of removal of color.

### 3.6. Effect of solution pH

A low pH is favorable for the production of hydrogen peroxide because the conversion of dissolved oxygen to hydrogen peroxide consumes protons in acidic solution, according to Eq. (4). However, a low pH also promotes hydrogen evolution, according to Eq. (5), reducing the number of active sites for

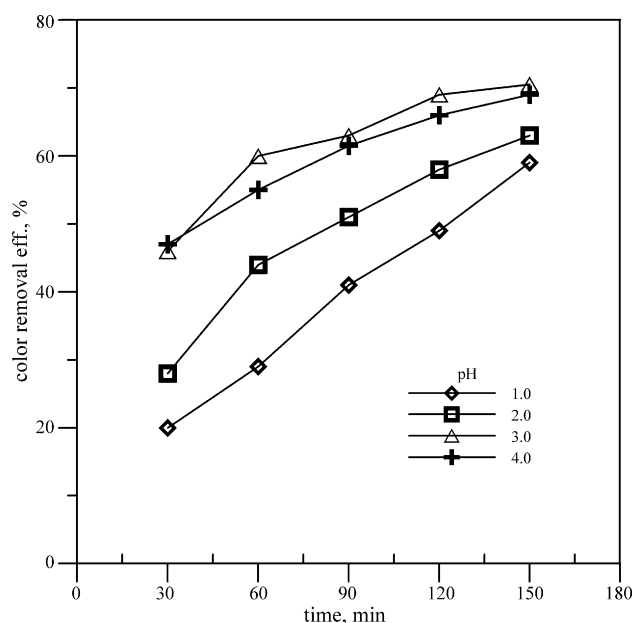


Fig. 8. Effect of the solution pH on the color removal; oxygen sparging rate:  $0.3 \text{ dm}^3/\text{min}$ ; applied current density:  $68 \text{ A/m}^2$ ;  $[\text{Fe}^{+2}] = 15 \text{ mM}$ ;  $25^\circ\text{C}$ .

generating hydrogen peroxide. Therefore, an optimal solution pH might be expected in this work. The effect of the solution pH on color removal is shown in Fig. 8. In a mildly acidic solution the removal efficiency observably increased with solution pH. Experimental results indicated that the removal efficiency was optimal at pH 3 in this work.

#### 4. Conclusion

This study has demonstrated that the removal of color in real dyeing wastewater by the electrogenerated hydrogen peroxide with the Fe(II) ions is an efficient way. The best oxygen contact mode for removing the color was the three-phase contact mode, resulting in the optimal transference of the dissolved oxygen to the electrode surface. The removal efficiency was controlled by mass transfer when the oxygen-sparging rate was less than 0.3 dm<sup>3</sup>/min for the reactor configuration adopted herein. The optimal applied current density in this work was 68 A/m<sup>2</sup> when the energy consumption was considered. Adding a few ferrous ions greatly enhance the efficiency of removal of the color. Adding extra ferrous ions traps the hydroxyl free radicals and reduces the efficiency of color removal, which was increased by increasing the pH to the optimal value of 3 and reducing the temperature of the solution.

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