



Applicability Assessment of Electrocoagulation in Real Dyeing Wastewater Treatment

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ABSTRACT

In this study, the applicability of electrocoagulation using iron electrodes in real dyeing wastewater treatment was assessed based on pollutants removal efficiency, sludge generation, energy consumption and operation cost in practice. The effects of current density, pH, conductivity, and reaction time on treatment performance were evaluated. The operation cost of electrocoagulation was calculated including the energy cost, the iron plate cost, generated sludge treatment cost, and added substances cost. The results indicated that the colour, COD_{Cr} and TSS removal efficiencies were high and quite stable with short reaction time (reached $92.07 \pm 1.21\%$, $65.7 \pm 1.47\%$, and $89.8 \pm 1.2\%$, respectively, with only 15 min). Average sludge generation, specific energy consumption, and operation cost were determined respectively as $0.645 \pm 0.0543 \text{ kg/m}^3$, 1.182 kWh/m^3 and 0.517 USD/m^3 . Coagulation-flocculation using FeSO_4 was performed as a control experiment as well. Compared to coagulation-flocculation, electrocoagulation has the same removal efficiency but has less generated sludge (only 50%) and little to no added chemicals. Therefore, the operating cost was quite less than the others, with only 0.517 USD/m^3 instead of 1.99 USD/m^3 (equal to 1/3.5).

INTRODUCTION

The real dyeing wastewater has usually extremely variable characteristics depending on the kind of dyes used and production capacity according to customer orders. It is highly coloured and viscous caused by residual dye and suspended solids. It also contains a lot of sodium and chloride ions due to high sodium chloride consumption in processing units (Hussain et al. 2004). In general, developing countries and namely, in Vietnam, the conventional and popular treatment method applied in primary treatment to remove colour and TSS (Total suspended solid) in dyeing wastewater is coagulation-flocculation-sedimentation. However, the amount of generated hazardous sludge and high chemical consumption are the biggest problems of this method.

Electrocoagulation (EC) is well known as the method that can remove colour, COD (chemical oxygen demand), TSS in the textile wastewater by electrolysis reaction using iron or aluminium or stainless steel electrodes. The pollutants can be removed by flocculation, adsorption of contaminants on flocs, sedimentation and floatation in the EC process. Iron electrodes out-performed others (Wang et al. 2016). Easy operation, none used chemicals, low sludge settling time, and less sludge formation are the main

advantages of electrocoagulation technology (Khorram & Fallah 2018, Chaturvedi & Satish 2013).

In recent years, most results of the previous studies on electrocoagulation using iron or aluminium electrodes to remove various dye types in textile wastewater showed that the dye and colour removal efficiencies were too high, from 83% to 100%. Each dye type in synthetic wastewater had a different optimal operating condition when treated by the electrocoagulation method. The operating factors significantly affected the treatment performance of dyeing wastewater such as current density, pH, conductivity, and reaction time (Aleboyeh et al. 2008, Arslan-Alaton et al. 2009, Charroenlarp & Choyphan 2009, Daneshvar et al. 2003, 2007, Kashefialasl et al. 2006, Khandegar & Saroha 2013, Khorram & Fallah 2018, Ahmed et al. 2018, Wang et al. 2016, Korbahti et al. 2011, Huynh et al. 2016, Parsa et al. 2011, Yang & McGarrahan 2005, Yuksel et al. 2013). Although the previous studies illustrated that electrocoagulation was able to remove dye and colour with high performance, most of these researches were performed with synthetic wastewater containing a specific dye. Therefore, it is difficult to apply these results to the general practical treatment of real wastewater containing various pollutant's concentration with different kinds of dye. In this study, the

electrocoagulation method using iron electrodes to treat the real dyeing wastewater from textile manufactory was investigated. The effect of pH, current density, reaction time, and conductivity was evaluated to find out the optimal operating condition. The treatment efficiency, sludge generation, energy consumption, and operation cost of the EC process were determined. In addition, the coagulation-flocculation method using FeSO_4 was performed as a control experiment. A comparison of the electrocoagulation method and conventional method based on generated sludge, specific energy consumption, and operation cost were evaluated to provide a scientific basis for selecting and replacing technology in the primary treatment of dyeing wastewater.

MATERIALS AND METHODS

Materials

The real dyeing wastewater was taken every day from the output of the equalization tank of the wastewater treatment system at Shoelace dyeing manufactory, Ho Chi Minh City, Vietnam, during October 1st, 2018 to November 16th, 2018 (Toan Hung Co., Ltd., Vietnam). The characteristics of real dyeing wastewater are presented in Table 1. $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was used as the coagulant in the control experiment. pH and conductivity were controlled by $\text{NaOH}/\text{H}_2\text{SO}_4$ and NaCl , respectively. $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{NaOH}/\text{H}_2\text{SO}_4$ and NaCl were procured from Xilong Chemical Co. Ltd., China.

Experimental Set Up and Analysis Methods

The experimental bench scale was set up as shown in Fig. 1. In this study, the temperature of wastewater was maintained at the same temperature as the coagulation-flocculation tank in the wastewater treatment system, in the range of 37°C - 40°C by the electrical heater before supplying into the reaction tank. 8 litres of the wastewater was supplied into a polyacrylic reaction tank (200mm × 200mm × 250mm) containing a pair of iron plate electrodes. The dimensions of the 5mm thick plate electrodes are 150mm × 200mm. The gap between the electrodes is 2.5cm. The electrical current was directly supplied from a laboratory DC power supply (DC Regulated power supply - QJ3010E, China).

Table 1: Characteristics of the real dyeing wastewater.

No.	Parameters	Unit	Range of value
1	pH	-	6.58 - 7.21
2	Colour	Pt - Co	500 - 2,513
3	Conductivity	mS/cm	0.36 - 0.78
4	CODcr	mg/L	430 - 1,620
5	TSS	mg/L	478

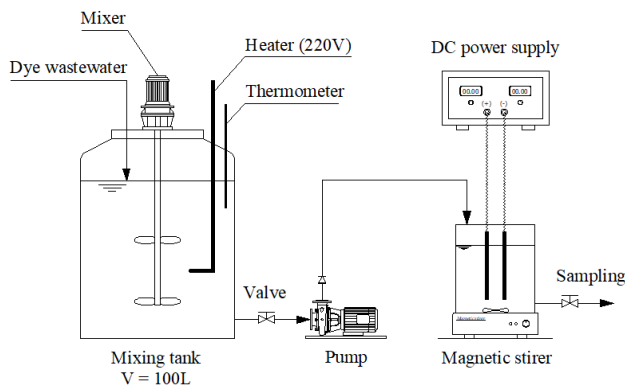


Fig. 1: Schematic diagrams of the electrocoagulation system.

500 mL sample was taken and removed the deposited sludge by settling for 60 min. CODcr, colour, and TSS were determined following the Standard Methods for the Examination of Water and Wastewater (APHA 1995). The mass of generated sludge was evaluated by the determination method of TSS in the treated wastewater. The pH value, temperature, and conductivity were observed by a pH meter (Hana, Germany), thermometer (Okaya Handy Thermo/T200, Japan), conductivity meter (Hana Model 130, Germany), respectively.

Each experiment was repeatedly carried out 3 times and experimental results were analyzed by ANOVA with $\alpha = 0.05$. Each experiment time was performed with the same homogeneous wastewater sample.

Calculation Methods

Removal efficiency, current density, energy consumption, and quantity of dissolved iron were calculated as previous studies of Huynh et al. (2016) and Chaturvedi & Satish (2013) as follow:

The removal efficiency was calculated as

$$E_{ff} = \frac{C_0 - C_t}{C_0} \times 100 \quad \dots(1)$$

Where, E_{ff} is the removal efficiency, %. C_0 and C_t are pollutants concentration at initial and t (min) reaction time, respectively.

The current density was calculated through the equation:

$$CD = \frac{I}{S} \quad \dots(2)$$

Where, CD is the current density, A/m^2 .

I is the current (A) and S is total surface area of the electrodes (m^2).

Electrical energy consumption by EC process:

$$E_{EC} = \frac{E_{cell} \times I \times t}{3.6 \times V} \quad \dots(3)$$

Where, E_{EC} is energy consumption for EC, Wh/m³. E_{cell} is the cell potential (V). I is the total current (A). t is the electrolysis time (s). And V is the working volume of the reactor (L)

The dissolved iron quantity in the EC process was calculated by equation .

$$W = \frac{i \times t \times M}{n \times F} \quad \dots(4)$$

Where, W : the amount of dissolution of iron electrode (g); i : current (A); t : reaction time (s); M : Molecular weight of iron; n : number of electrons in oxidation/reduction reaction, $n_{Fe} = 2$; F : Faraday's constant, 96485 C/mol.

RESULTS AND DISCUSSION

The Effect of Current Density

Current density is the most important factor that affects treatment efficiency and energy consumption in the EC process. In this study, the current density was changed in the range of 12.5 A/m² - 33.3 A/m². pH and electrical conductivity were controlled in the range of 6.7 - 7 and 2.5 mS/cm, respectively. The reaction time was 20 min. The results indicated that current density significantly affected the colour removal (P-value: 0.000749). Colour removal efficiency increased rapidly from 10.78% to 90.8% when current density rose from 12.5 A/m² to 20.8 A/m² and approximate 94% with a current density of 25 A/m². When current density was continuously increased, colour removal performance was slightly increased. It reached nearly 95% when the current density rose to 33.3A/m² (Fig. 2a). The higher current density led to consuming more energy. Therefore, the suitable current density value was chosen based on colour treatment efficiency and energy consumption. In this case, the current density of 25 A/m² was chosen.

The Effect of pH

pH is the key parameter in the electrocoagulation process (Chafi et al. 2011). It possibly directly affects the removal efficiency and chemical consumption to adjust pH before and after treatment. In this study, the effect of pH on colour removal efficiency was determined with experimental conditions such as pH range from 3 to 9, the current density of 25 A/m², the electrical conductivity of 2.5mS/cm, and reaction time of 20 min. The ANOVA analysis results showed that the effect of pH on colour treatment was significant with P-value equal to 2.2×10^{-7} . The observed data is depicted in Fig. 2b. The colour removal efficiency rapidly increased when pH

increased from 3 to 7 and reached 94.07% at pH 7. Then, it slightly increased to 95.59% when pH rose from 7 to 9. It was caused by an acidic medium, OH⁻ ions generated from the cathode reacted with H⁺ ions led to lack of OH⁻ for iron hydroxide formation. When increasing pH, the presence of OH⁻ ions in solution was risen to enhance the formation and precipitation processes of iron ions. In addition, Fe²⁺ was generated at anode and oxidized to Fe³⁺ by Cl₂, HOCl, and ClO⁻ in acidic, neutral and alkaline mediums, respectively (Chafi et al. 2011). HOCl is more effective than others. Precipitation of Fe(OH)₂ starts at pH = 8 and Fe(OH)₃ starts at pH = 3.5 (Magdalena & Aneta 2011).

With this actual dyeing wastewater, pH of influent wastewater nearly equalled to 7, making the optimal pH as 7 for treatment by electrocoagulation process using iron electrodes because no or little chemicals were used to adjust pH value allowing the colour removal efficiency nearly reached the optimal value.

The Effect of Electrical Conductivity

Electrical conductivity was varied at 1.5 mS/cm; 2.0 mS/cm, 2.5 mS/cm and 3.0 mS/cm. Current density, pH and reaction time were fixed at 25 A/m², 7 and 20 min, respectively. Although colour removal efficiency in these experiments slightly increased from 92.3% to 93.7% when electrical conductivity rose from 1.5 mS/cm to 2.5 mS, ANOVA analysis results indicated that electrical conductivity didn't affect significantly the colour removal, P-value of 0.119. It was the same with a previous study (Huynh et al. 2016). With the same current density input, the voltage was inversely proportional to electrical conductivity and followed an equation: $y = -2.23x + 16.878$ ($R^2 = 0.9967$). So, the electrical conductivity directly affected energy consumption. Increasing electrical conductivity led to reducing energy consumption in the EC process (Huynh et al. 2016). In this study, electrical conductivity increased from 1.5 mS/cm to 3.0 mS/cm, energy consumption decreased 2.14 times.

The Effect of Reaction Time

Reaction time was surveyed from 5 to 35 min with the current density of 25 A/m², pH 7 and EC of 2.5 mS/cm. The obtained results presented in Fig. 2c showed that colour removal performance increased markedly in the first 15 min reaction time and archived at nearly 91%. Extending reaction time, treatment efficiency slowly and trivially increased, reached upto 94.5% within 30 min. It is easy to explain because the amount of dissolved iron ion generated in the first 15 min reaction time was nearly enough for the coagulation and flocculation process. When reaction time rose to 35 min, colour removal slightly reduced to 93% caused by residual iron ions in solution.

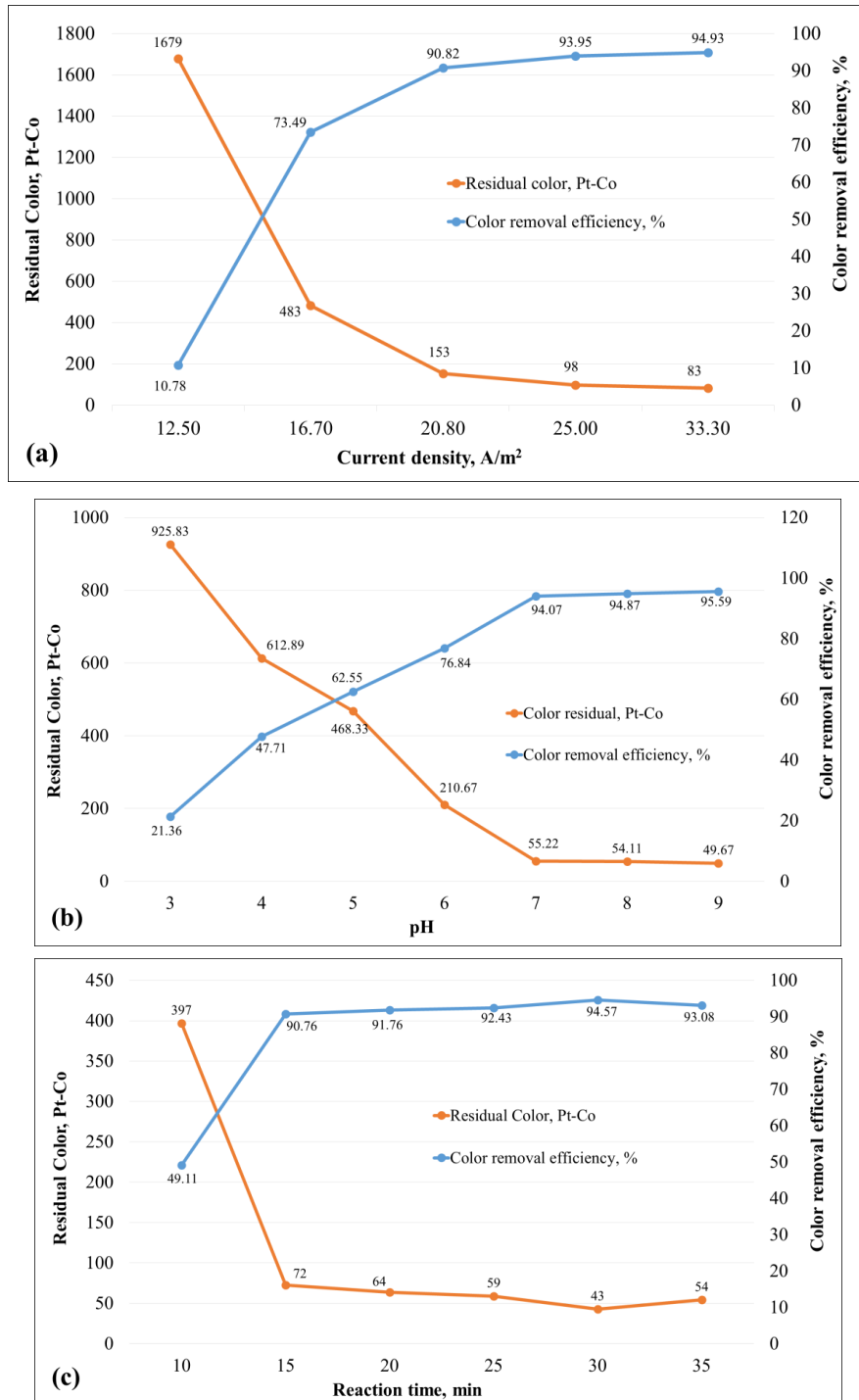


Fig. 2: The average colour removal versus various current density (a), pH value (b) and reaction time (c).

The Treatment Efficiency by Electrocoagulation

In order to determine the applicability of this method into wastewater treatment with the real condition at the factory,

the testing was performed with the real wastewater taken in different 25 days. The suitable operating parameters were obtained as current density of 25 A/m², non-adjusted pH, electrical conductivity of 2.5 mS/cm and 15 min reaction

time. As depicted in Fig. 3, the results showed that the treatment performance was very high and quite stable even though the characteristics of wastewater in input flow much fluctuated. Namely, during the observed period, the pH, colour, COD_{Cr}, TSS, and conductivity were in range of 6.58 - 7.21, 725 Pt-Co - 2,513 Pt-Co, 415 mg/L - 1,692 mg/L, 320 mg/L - 478 mg/L and 0.33 mS/cm - 0.78 mS/cm, respectively. It may be explained as: in the EC process, iron hydroxo complexes formed at the anode has high adsorption properties, forming strong aggregates with pollutants. In addition, the flocculation and adsorption capacity was increased when the pollutant

concentration increased due to the higher contacting opportunities between pollutants and coagulants (Kabdasli et al. 2014). It may buffer the fluctuation in inlet flow. The average treatment efficiency was $92.04 \pm 1.21\%$, $65.71 \pm 1.47\%$ and $89.8 \pm 1.2\%$ corresponding to colour, COD_{Cr}, and TSS removal respectively (Table 2). Compared to the discharge standard in Vietnam (QCVN 13:2015/BTNMT, column B), the residual colour and TSS in effluent were lower but residual COD_{Cr} was still too much higher than the discharged standard. Therefore, COD_{Cr} in the effluent of electrocoagulation reaction was continuously treated by the next stage.

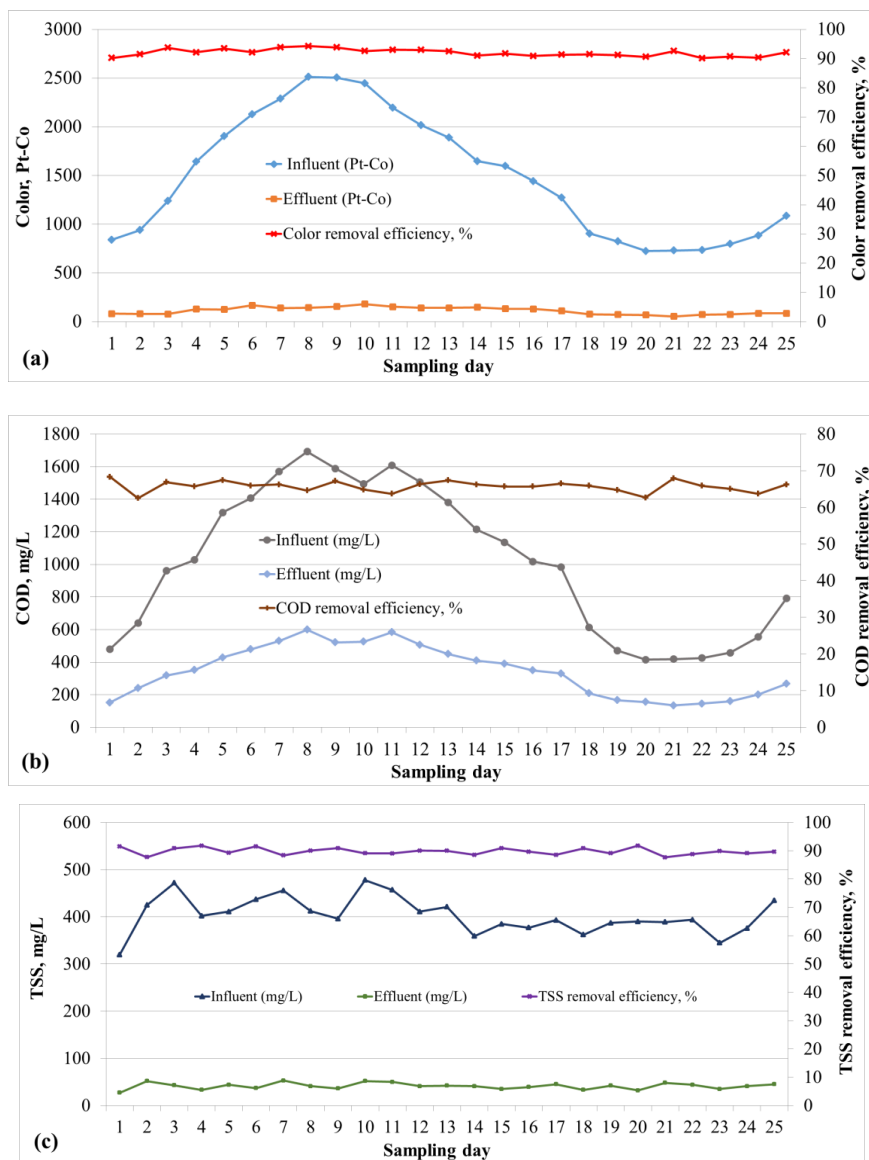


Fig. 3: Variation of concentration and removal efficiencies of Colour, COD_{Cr}, TSS.

The average mass of generated sludge including flotation and sedimentation process was 0.645 ± 0.054 g/L (Table 2). Specific energy consumption was evaluated by equation (3), approximately 1.182 kWh/m³ (Table 2). Iron consumption was determined based on the dissolved iron quantity (Eq. 4) and the total volume of treated wastewater, equal 0.0975 kg/m³.

Comparisons of Electrocoagulation and Coagulation-Flocculation

The optimal value of pH and coagulant dose of FeSO₄·7H₂O in the coagulation-flocculation process using the same raw wastewater used in the electrocoagulation process were determined by Jar-test experiment. The treatment performance, sludge generation, and chemical use were calculated in coagulation-flocculation as well. With the optimal condition, COD_{Cr}, TSS and decolourization efficiency reached $56.8 \pm 2.2\%$, $82.4 \pm 2.9\%$, and $93 \pm 0.6\%$, respectively. Sludge generation was 1.5 ± 0.0419 kg/m³.

The comparison of the electrocoagulation and coagulation-flocculation based on treatment efficiency, sludge generation, energy consumption, chemical use, and operation cost were summarized and presented in Table 2. Colour removal efficiency was the same in both methods but COD_{Cr} and TSS treatment performances in the electrocoagulation were higher. Sludge generation, chemical use in electrocoagulation were quite less than coagulation-flocculation. This is because, in electrocoagulation, TSS was mainly removed by flotation mechanism so the small flocs formed was immediately floated to the surface by many fine bubbles (H₂). It did not need as much iron to form big flocs to settle to the bottom as in coagulation-flocculation. Moreover, the coagulant in electrocoagulation was directly generated in situ from the anode, did not contain impurity materials. OH⁻ participated in the flocculation reaction that was formed from H₂O. OH⁻ generation had been consumed that buffered the pH of the solution during electrocoagulation led to the final pH of effluent changed only slightly. Therefore, there was no need to add chemical to provide OH⁻ and adjust pH after treating it as the traditional chemical coagulation.

Calculation of the operating cost included the energy cost, iron plate cost, generated sludge treatment cost, and added substances cost. The results showed that the operation cost of electrocoagulation was far much lower than coagulation-flocculation as well, with only 0.517 USD/m³ instead of 1.99 USD/m³ as in conventional coagulation, approximately equal to 1/3.5. The main reasons were because of less generated sludge and no added chemical in the electrocoagulation process (with only small amount of NaCl).

Table 2: Comparison of electrocoagulation using iron electrodes and coagulation-flocculation using FeSO₄.

Criteria	Electrocoagulation	Coagulation-Flocculation
pH	7.0	11.0
Colour removal efficiency (%)	92 ± 1.21	93 ± 0.61
COD _{Cr} removal efficiency (%)	65.7 ± 1.47	56.8 ± 2.2
TSS removal efficiency (%)	89.8 ± 1.2	82.4 ± 2.9
Sludge generation (kg/m ³)	0.645 ± 0.0543	1.5 ± 0.0419
Specific energy consumption (kwh/m ³)	1.182	0.135
NaOH 98% (kg/m ³)	0	0.8 ± 0.02
H ₂ SO ₄ 98% (kg/m ³)	0	0.472 ± 0.011
FeSO ₄ (kg/m ³)	0	0.75
NaCl (kg/m ³)	1.17 ± 0.33	0
Iron plate consumption (kg/m ³)	0.0975	0
Operation cost* (USD/m ³)	0.517	1.99

*The operation cost in both methods was calculated based on the price in Vietnam (Pham Phu Gia Chemical Co. Ltd.).

CONCLUSIONS

This study found the optimal operating parameters for real dyeing wastewater treatment by electrocoagulation using iron electrodes as follows: current density of 25 A/m², non-adjusted pH, electrical conductivity of 2.5 mS/cm and 15 min reaction time. Electrocoagulation using iron electrodes may be applied in actual dyeing wastewater treatment of shoelace manufactories because it was able to remove colour, COD_{Cr}, and TSS in the actual dyeing wastewater with high performance, achieved $93 \pm 0.6\%$, $56.8 \pm 2.2\%$, and $82.4 \pm 2.9\%$, respectively. Furthermore, it adapted to the various characteristics of influent very well, released less generated sludge, and consumed low operating cost.

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