Performance of an Electrocoagulation Process in Treating Direct Dye: Batch and Continuous Upflow Processes

C. Phalakornkule, S. Polgumhang, and W. Tongdaung

Abstract—This study presents an investigation of electrochemical variables and an application of the optimal parameters in operating a continuous upflow electrocoagulation reactor in removing dye. Direct red 23, which is azo-based, was used as a representative of direct dyes. First, a batch mode was employed to optimize the design parameters: electrode type, electrode distance, current density and electrocoagulation time. The optimal parameters were found to be iron anode, distance between electrodes of 8 mm and current density of 30 A·m^{-2} with contact time of 5 min. The performance of the continuous upflow reactor with these parameters was satisfactory, with >95% color removal and energy consumption in the order of 0.6-0.7 kWh·m^{-3}.

Keywords—Decolorization, Direct Dye, Electrocoagulation, Textile Wastewater, Upflow Reactor.

I. INTRODUCTION

ELECTROCOAGULATION is an alternative technology for wastewater treatment and for recovery of valuable chemicals from wastewater. The main advantages of electrocoagulation over other conventional techniques, such as chemical coagulation and adsorption, are "*in situ*" delivery of reactive agents, no generation of secondary pollution, and compact equipment. Many studies have reported the potential of electrocoagulation in the treatment of a variety of wastewater. The method has been used for removing suspended solids [1]-[3], removing dyes [4]-[10], removing heavy metals [11],[12], breaking oil emulsions in water [13],[14], removing complex organics [15],[16], and removing bacteria, viruses and cysts [17].

The textile industry consumes considerable amounts of water during dyeing and finishing operations. Dye-containing effluent can be toxic to the environment since dyes are stable compounds that are not easily biodegradable and they are liable to be carcinogenic [18]. Especially, azo dyes are known to be highly toxic [19]. Dyes are normally very large aromatic molecules consisting of many linked rings. Four common classes of dye, classified according to the fibers to which they can be applied, and their chemical nature, are: 1. acid dyes; 2. reactive dyes; 3. disperse dyes and 4. direct dyes [8]. The dyes are also different in terms of their resistance to destruction or to removal by physical and chemical methods. One of the main influences on the performance of dye removal by electrocoagulation is the solubility of the dyes.

Conventional methods for textile wastewater treatment are the combination of physical, chemical and biological methods. However, physical methods such as adsorption and precipitation are relatively time-consuming and costly, while most chemical methods such as additions of aluminum sulfate and chlorine can generate secondary pollutants. Some other advanced technologies such as photo oxidation by UV/H$_2$O$_2$ or UV/TiO$_2$ also generate secondary pollutants [20]. Biological treatment of textile wastewater has low efficiency because of the toxicity of dye molecules to active microorganisms.

In this study, we have investigated the operational parameters that are suitable for treating a direct dye by electrocoagulation. Direct red 23 (DR 23), which is azo-based, was used. This dye is one of the dyes used in a number of factories in Thailand and the treatment of wastewater containing this dye is therefore important in reducing environmental pollution. The effect of environmental factors, i.e. pH and temperature, on the process efficiency was investigated. A set of optimal operational parameters including electrode type, electrode distance, current density and electrocoagulation time for a batch mode were then used to design a continuous-flow electrocoagulation reactor. Technical and energy consumption assessments of the electrocoagulation process were made.

II. MATERIALS AND METHOD

A. Synthetic and Textile Wastewater

Direct red 23 (Disodium 3-[(4-acetamidophenyl)azo]-4-hydroxy-7-[[[[5-hydroxy-6-(phenylazo)-7-sulphonato-2 naphthyl]amino]carbonyl]amino]naphthalene-2-sulphonate)

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was used for preparing synthetic dye wastewater. Its chemical structure is shown in Fig. 1. The synthetic wastewater was prepared by dissolving 0.10 kg·m^{-3} of the direct dye into tap water. The pH was adjusted to 9.6±0.1 with NaOH and the conductivity was adjusted to 875±25 μS·cm^{-1} with NaCl. The values of pH and conductivity were chosen to simulate the properties of wastewater from a textile industry in Thailand. The initial concentration of 0.10 kg·m^{-3} was selected because it provided a Carbon Oxygen Demand (COD) value in the same order of magnitude as the effluent from the textile factory.

The textile effluent was obtained from the textile industry in Bangkok, Thailand, which produced approximately 1000 m^3 of wastewater per day. The effluent was a mixture of wastewater from dyeing and finishing operations and other washing and rinsing activities.

B. Batch-Mode Electrocoagulation

The batch system consisted of a DC power supply, a power control and measurement units, an electrochemical reactor and a magnetic heater and stirrer (Fig. 2). The electrochemical reactor was a 2-L beaker with a set of five pairs of electrodes made of either iron or aluminum with a total area of 1047 cm^2. Each electrode had a circular shape with a diameter of 12.2 cm. The electrodes were connected vertically with an adjustable gap of 5-12 mm between electrodes. The electrodes were connected to the DC power supply in monopolar mode and the electrochemical reactor was operated in batch and galvanostatic mode.

Before and at the end of each run, electrodes were washed thoroughly with water, dipped in HCl solution (5% vol/vol) for at least 15 min and rinsed again with tap water. Typical runs were conducted at 301.15 K. To investigate the effect of temperature, the temperature was raised to 333.15 K. During the runs, the reactor unit was stirred at 150 rev·min^{-1} by a magnetic stirrer (IKA, C-MAG HS, Germany). The experiments were carried out in a batch mode with a liquid sample of 1.8 L. Four different current densities of 10, 20, 30, 40 A·m^{-2} were applied. For each current density, electrocoagulation time of 2, 5, 10, and 15 min were used. After the specified electrocoagulation time, the system was allowed to stand at room temperature for 20 min and a supernatant was pipetted from the middle of the supernatant portion for the analysis of water properties.

C. Continuous-Mode Electrocoagulation

The electrochemical reactor was a 10-L acrylic column with a set of twenty-five pairs of electrodes made of iron with a total area of 5235 cm^2. The unit consisted of 1. a wastewater storage tank; 2. a pump with an operating flowrate between 1-5 L·h^{-1}; 3. a wastewater inlet located at the bottom of the electrochemical reactor; 4. the electrochemical reactor with a set of twenty-five pairs of iron electrodes; 5. a treated effluent outlet located at the top of the reactor; and 6. a 120-L sedimentation tank (Fig. 3a). Each electrode had the same configuration as those employed in the batch experiments. The electrodes were connected vertically with a gap distance of 8 mm (Fig. 3b).

D. Analytical Method

The pH and conductivity were measured by a pH meter (Schott, Lab 850, Germany) and a conductivity meter (Milwaukee, EC59, Italy), respectively. The dye concentration was indicated by an absorbance value measured by a UV-Vis spectrophotometer (Spectronic Genesys 20, USA) at the wavelength of 500 nm. The color removal efficiency was determined according to (1):

$$\text{Color removal efficiency (\%)} = 100 \times \left(\frac{A_o - A}{A_o} \right) \qquad (1)$$

where A_o and A are the light absorbance of dye before and after electrocoagulation, respectively.

The amount of iron released into solution by electrolytic oxidation of the anode was estimated from the Faraday's law:

$$w = \left(\frac{ItM_{Fe}}{ZF} \right) \qquad (2)$$

where w is the iron dissolved (g), I is the current (A), t is the electrocoagulation time (s), M_{Fe} is the molecular weight of Fe (55.85 g·mol^{-1}), Z is the number of electrons involved in the redox reaction, and F is the Faraday's constant (96500 C per mole of electrons).
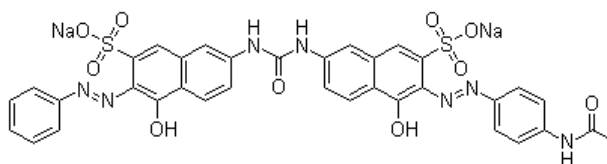

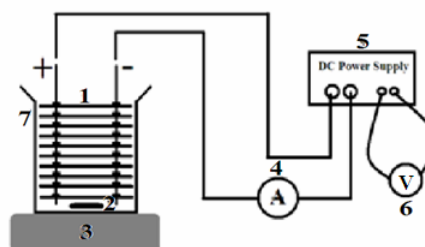Fig. 1 The chemical structure of direct dye 23

Fig. 2 Set-up of the electrochemical system 1: Electrodes 2: Magnetic Bar Stirrer 3: Magnetic Stirrer Controller 4: Ammeter 5: D.C. Power Supply 6: Voltmeter 7: 2-L Beaker
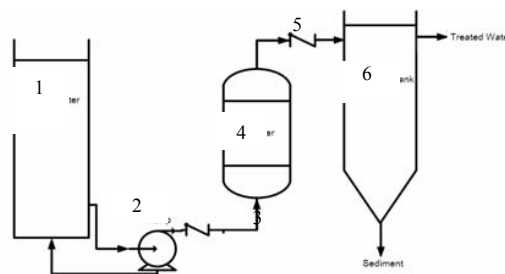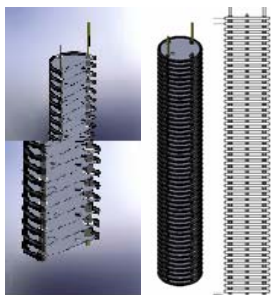

Fig. 3 (a) The electrochemical continuous system
1: Wastewater Storage Tank 2: Pump 3: Wastewater Inlet 4: Set of Electrodes 5: Treated Effluent Outlet 6: Sedimentation Tank

(b) Twenty-five pairs of electrodes sealed with rubbers connected vertically and located in an acrylic column

III. RESULTS AND DISCUSSION

A. Treatment of Direct Dye in a Batch Mode

Fig. 4 shows the removal efficiency of the direct dye DR 23 as a function of current density and electrocoagulation time. The distance between electrodes was 12 mm. Sharp increases of removal efficiencies can be observed in the first 2 min for all of the investigated current densities, especially by using Fe-anode. Except with the current density of 40 $A \cdot m^{-2}$, electrocoagulation with Al-anode required the electrocoagulation time for at least 5 min to achieve the removal efficiency greater than 80%. For the electrocoagulation time longer than 5 min, the efficiency of color removal by Fe-anode was comparable with that by Al-anode. A removal efficiency of 80% has previously been reported to be the minimum acceptable for an electrocoagulation process [19]. For a current density of 10 $A \cdot m^{-2}$, the removal efficiency was below 80% for all electrocoagulation times under investigation (data not shown in the figure).
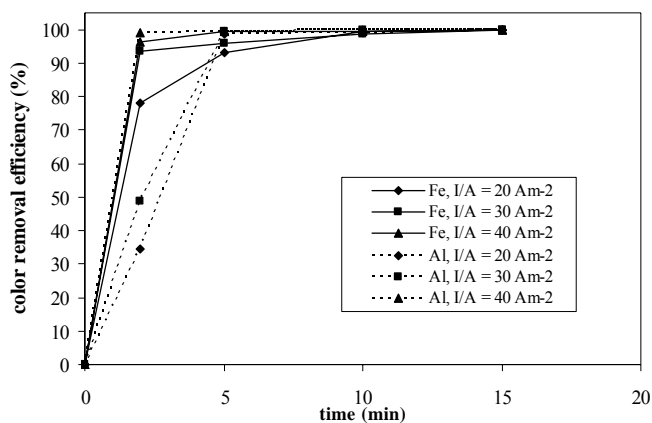

Fig. 4 The removal efficiency of the direct dye DR 23 as a function of current density and electrocoagulation time (distance between electrodes: 12 mm, temperature: 301.15 K, pH: 9.5-9.7, conductivity: 850-870 $\mu S \cdot cm^{-1}$)

In a further selection process of electrode type, energy consumption was investigated. Fig. 5 compares the energy consumption of each electrode type with a current density of 30 $A \cdot m^{-2}$. The iron anode required lower energy ranging between 0.6-2.5 $kWh \cdot m^{-3}$. Therefore, based on both technical and energy consumption assessment, Fe-anode was ranked above Al-anode in treating the direct dye DR 23.

The distance between electrodes is another important design parameter. In general, a closer distance between electrodes is desirable, because electrical resistance increases with an electrode gap distance. Fig. 6 shows the removal efficiency of the direct dye DR 23 and energy consumption as a function of distance between electrodes and electrocoagulation time with the current density of 30 $A \cdot m^{-2}$. Unexpectedly, the electrical energy consumption of the electrode distance of 8 mm was lower than that of 5 mm. This was in contrast with a previous study [21] which has reported that the power consumption for electrocoagulation increased proportionally when the electrode gap was increased from 1 to 3 cm. The explanation was that at the electrode distance of 5 mm, the gap between anode and cathode was too closed and solid and fluid transfer was obstructed. The accumulated solid particles and bubbles between the anodes and cathodes caused a consequent higher electrical resistance.
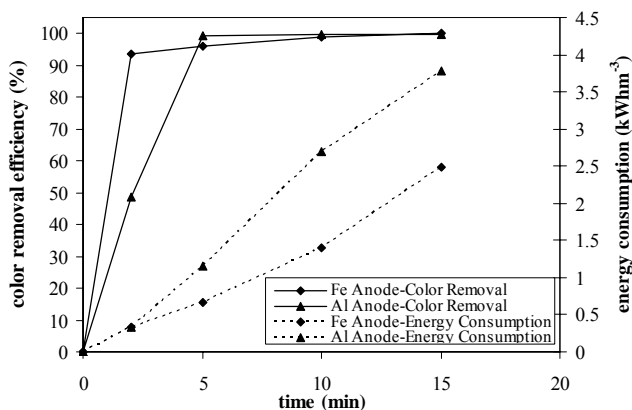

Fig. 5 The removal efficiency of the direct dye DR 23 and energy consumption as a function of anode type and electrocoagulation time (current density: 30 $A \cdot m^{-2}$, distance between electrodes: 12 mm, temperature: 301.15 K, pH: 9.5-9.7, conductivity: 850-870 $\mu S \cdot cm^{-1}$)
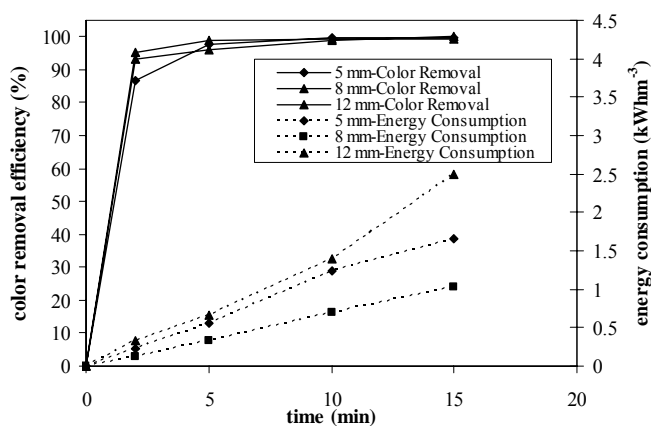

Fig. 6 The removal efficiency of the direct dye DR 23 and energy consumption as a function of distance between electrodes and electrocoagulation time (current density: 30 $A \cdot m^{-2}$, anode type: iron, temperature: 301.15 K, pH: 9.5-9.7, conductivity: 850-870 $\mu S \cdot cm^{-1}$)

Based on the removal efficiency and energy consumption, an effective set of design parameters for treating the direct dye DR 23 was as follows: iron anode, distance between electrodes of 8 mm and current density of 30 A·m^{-2} and contact time of at least 2 min. It was of an interest whether these parameters were still effective when the two environmental factors, i.e. pH and temperature, fluctuated. Even though the typical pH of textile effluent was high, ranging from 8.5 to 10, a fluctuation in pH was possible. Discharge temperature of the textile wastewater is actually around 333 K, making treatment at the temperature possible without extra cost. Fig. 7 and 8 show the effect of pH and temperature on the removal efficiency of the direct dye by electrocoagulation, respectively. The effect of pH was not
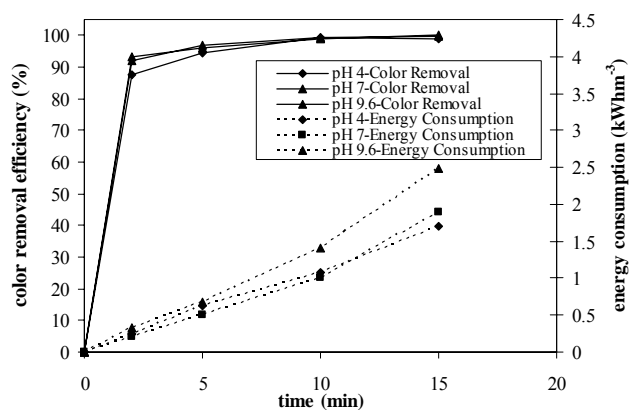

Fig. 7 The effect of pH on the removal efficiency of the direct dye DR 23 by electrocoagulation and energy consumption (current density: 30 A·m^{-2}, anode type: iron, distance between electrodes: 8 mm, temperature: 301.15 K, conductivity: 850-870 µS·cm^{-1})
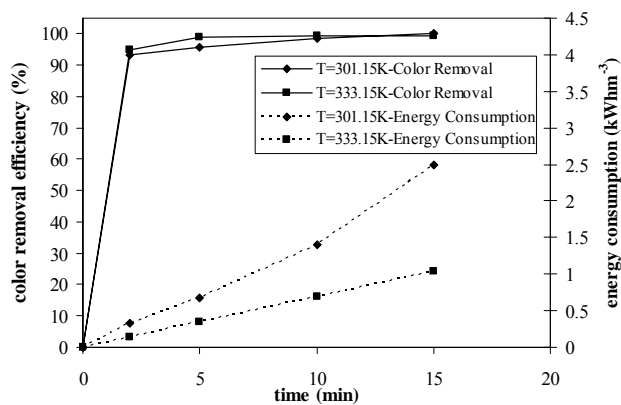

Fig. 8 The effect of temperature on the removal efficiency of the direct dye DR 23 by electrocoagulation and energy consumption (current density: 30 A·m^{-2}, anode type: iron, distance between electrodes: 8 mm, pH: 9.5-9.7, conductivity: 850-870 µS·cm^{-1})

significant, and the removal efficiency at different temperatures of 301.15 and 333.15 K was also not significantly different. Even though pH can influence the form of Fe-species [22], the effect of form of Fe-species was not

revealed in case of the direct dye. It should also be noted that the operation of electrocoagulation process at higher temperature significantly reduced electrical energy consumption. The reason is believed to be that fluid conductivity increases with temperature.

B. Performance of the Continuous Upflow Electrochemical Reactor

A set of design parameters that was effective for treating the direct dye in a batch-mode was used in the construction and operation of the continuous upflow electrochemical reactor. The following combinations of current density and contact time were investigated: 30 A·m^{-2} with electrocoagulation time of 5 min (CD30T5), 40 A·m^{-2} for 5 min (CD40T5), 30 A·m^{-2} for 10 min (CD30T10) and 40 A·m^{-2} for 10 min (CD40T10). The pump flowrate was adjusted to control the fluid residence time to the required electrocoagulation time. Fig. 9 shows the removal efficiency and energy consumption as a function of current density and electrocoagulation time. The performance of the continuous upflow electrochemical reactor agreed fairly well with the performance of the batch-mode both in terms of color removal efficiency and energy consumption. Fig. 10 shows the amount of iron released as a function of current density and electrocoagulation time by the continuous upflow electrocoagulator. For one operation in this study (either 30 or 40 A·m^{-2} for 5-10 min), the weight loss of the anode was between 1-4 g. The performance was rather independent of the amount of iron released. Therefore, the set of parameters that yielded the lowest loss of anode materials, i.e. 30 A·m^{-2} for 5 min, should be chosen. In this case, this set of parameters also required the lowest energy consumption.
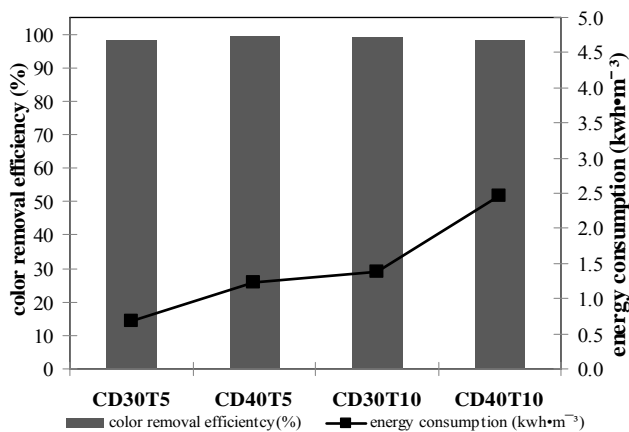

Fig. 9 The removal efficiency of the direct dye DR 23 and energy consumption as a function of current density and electrocoagulation time by the continuous upflow electrocoagulator (temperature: 301.15K, pH: 9.5-9.7, conductivity: 850-870 µS·cm^{-1})
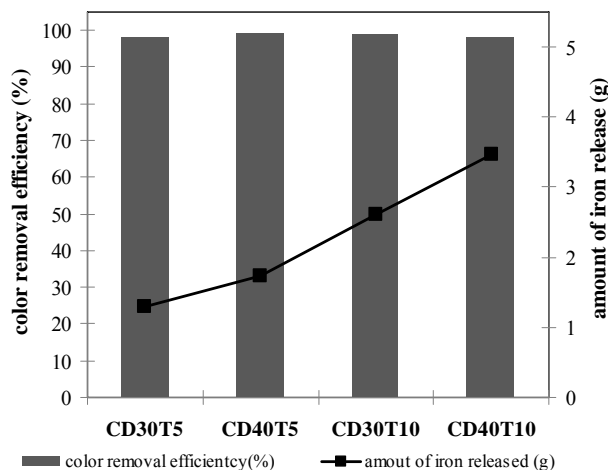
Fig. 10 The removal efficiency of the direct dye DR 23 and amount of iron released as a function of current density and electrocoagulation time by the continuous upflow electrocoagulator (temperature: 301.15 K, pH: 9.5-9.7, conductivity: 850-870 μS·cm^{-1})

C. Treatment of Textile Wastewater

This set of parameters was further employed to treat the textile wastewater. The initial COD of the textile wastewater ranged between 278-736 mg·dm^{-3}. The color removal efficiency as shown in Fig. 11 was not always satisfactory, i.e. from six samplings, four were satisfactory (> 80%), but two failed (< 80%). Removal of color from the textile wastewater was more difficult than the synthetic wastewater. This may be because the real textile wastewater contained other dye molecules that could not be effectively removed by the electrocoagulation process operated with the design parameters. Direct dyes have been generally reported to be more easily removed by the electrocoagulation method than reactive dyes which are water-soluble [21]. Another possible explanation was that the real textile wastewater may contain some chemicals which can intervene the electrocoagulation mechanism. Further studies are necessary in order to increase the reliability of the technique.
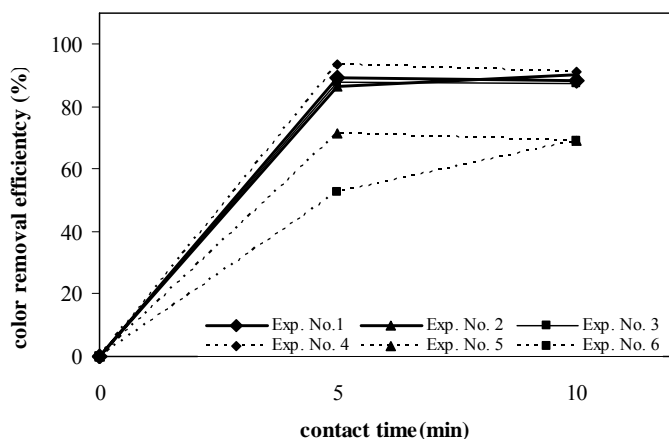

Fig. 11 The removal efficiency of the textile wastewater as a function of electrocoagulation time with the current density of 30 A·m^{-2}
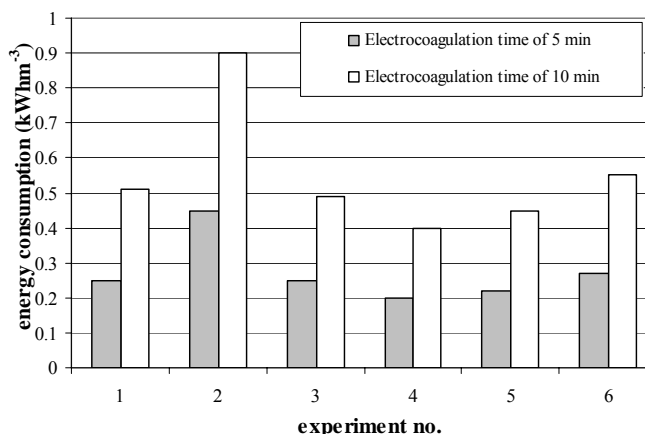

Fig. 12 Energy consumption of the electrocoagulation system in treating the textile wastewater as a function of electrocoagulation time with the current density of 30 A·m^{-2}

Fig. 12 shows the energy consumption of the electrocoagulation system in treating the textile wastewater. The electric cost ranged from 0.2 to 0.9 kWh·m^{-3}. Considering the cost of electricity at 3 baht·kWh^{-1} (1 USD = 35.5baht, [23]), the cost of water treatment by the electrocoagulation unit was lower than 3 baht·m^{-3}. Fresh water was available for an industry at 12 baht· m^{-3} in a normal situation. When there were incidents of drought, the price of fresh water rose up to 100 baht·m^{-3} [24]. Therefore, besides sharing the responsibility for protecting the environment, an industry might gain benefit from the water treatment and reuse in its processes.

D. Comparison of System Performance with Other Studies

TABLE I
PERFORMANCE COMPARISON WITH OTHER STUDIES

Ref.	Wastewater Characteristics	Process Description	Color Removal
[19]	Red disperse dye solution of 2-naphthoic acid and 2-naphtol Initial Concentration 25-200 mg·L^{-1} pH 6-9 Conductivity 2400 μS·cm^{-1}	Continuous Anode type aluminum Current density 312.5 A·m^{-2} Electrode distance 1 cm Contact time 14 min	> 80%
[21]	Disperse blue 106 Initial Concentration 500 mg·L^{-1} NaCl Concentration 1000 mg·L^{-1}	Continuous Anode type aluminum Current density 45 A·m^{-2} Electrode distance 1 cm Contact time 13 min	> 98%
	Reactive yellow 84 Initial Concentration 500 mg·L^{-1} NaCl Concentration 1000 mg·L^{-1}	Continuous Anode type aluminum Current density 45 A·m^{-2} Electrode distance 1 cm Contact time 13 min	> 85%
	Reactive blue 49 Initial Concentration 500 mg·L^{-1} NaCl Concentration 1000 mg·L^{-1}	Continuous Anode type aluminum Current density 45 A·m^{-2} Electrode distance 1 cm Contact time 13 min	< 80%

	Disperse yellow 54 Initial Concentration 500 mg·L^{-1} NaCl Concentration 1000 mg·L^{-1}	Continuous Anode type aluminum Current density 45 A·m^{-2} Electrode distance 1 cm Contact time 13 min	> 95%
	Dyeing wastewater from a dyeing industrial complex located in Banwol, Korea	Continuous Anode type aluminum Current density 45 A·m^{-2} Electrode distance 1 cm Contact time 13 min	< 50%
This study	Direct red, azo-based Initial Concentration 100 mg·L^{-1} pH 9.6±1 Conductivity 860±10 μS·cm^{-1}	Batch and continuous Anode type iron Current density 30 A·m^{-2} Electrode distance 0.8 cm Contact time 5 min	>90%

Table I compares the performance of electrocoagulation of this study with other two studies. In treating direct dyes, all studies were successful, i.e. >80% color removal. However, Kim et al. (2002) [21] reported the failure in treating reactive dye (Reactive blue 49) and real textile wastewater with the same set of operating parameters. It was suggested that the performance of dye removal by electrocoagulation depended upon the solubility of the dyes, the final floc formation and its settling quality. Since dyeing activities of a factory could vary according to its customer's requirements, resulting in different levels of dye types in wastewater, optimization of operating parameters for treating textile wastewater remains a challenging task.

IV. CONCLUSION

This work was among relatively few studies linking a batch mode to a continuous mode of electrocoagulation. The removal efficiency showed that iron was in competition with aluminum in treating the direct dye DR 23. Based on energy expense, iron was more superior as it required lower energy than aluminum. In contrast to a previous study [21], the power consumption for electrocoagulation did not increase in proportion with the electrode gap. The optimal electrode gap distance was 8 mm. With the initial dye concentration of 0.10 kg·m^{-3}, the energy cost in achieving >95% color removal with a current density of 30 A·m^{-2} in a batch mode was 0.67 kWh·m^{-3}. The application of the set of optimal parameters was implemented successfully to an up scaled continuous upflow electrocoagulator. The performance of the continuous upflow electrochemical reactor agreed fairly well with the performance of the batch-mode process. With the initial dye concentration of 0.1 kg·m^{-3}, the energy cost in achieving >95% color removal with a current density of 30 A·m^{-2} in the continuous mode was 0.69 kWh·m^{-3}.

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