Comparison of Electrocoagulation Using Iron and Aluminium Electrodes with Chemical Coagulation for removal of Vat Dye Solution

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Abstract

The dyes are widely used in textile industry and causes various environmental problems. There are several methods available for removal of dyes from wastewater but they have been found to exhibit certain limitations. Among these methods, chemical coagulation (CC) and electrocoagulation process (EC) has been successfully used for the treatment of textile wastewater being attractive alternative. The objective of study is to investigate the performance of EC and CC for decolorization of vat dye and to compare between iron and aluminium as electrode and coagulant. The results show EC process had color removal efficiency higher than 90 % for both of Fe and Al electrode. The respective effects of operational parameters (initial dye concentration, reaction time, initial pH and applied voltage) had effect on color removal of EC. And the optimum conditions for Fe and Al electrode are 28 mg/L for initial dye concentration, 30 and 20 min for reaction time, 5 and 7 for initial pH and 30 volt for applied voltage respectively. A comparison with CC using the same amount of Fe and Al as in EC was also carried out. The EC process show more efficient process than CC process to treat vat dye solution. And for color removal of EC did not differ significantly between Al and Fe electrode.The application of EC to treat wastewater from textile household industries had color and COD removal efficiency higher than 80% and 60% respectively. It was concluded that the EC process is a competitive alternative process and presents a high potentiality for the treatment of color from textile wastewater.

Keywords: Electrocoagulation, Chemical Coagulation, Vat Dye, Iron Electrode, Aluminium Electrode

Introduction

The dyes are widely used in textile industry and causes various environmental problems, not only because of its huge quantity of poisonous organic pollutants, but also due to its intense color, high COD, large quantity of suspended solids, broadly fluctuating pH and biotoxicity (Hamad, Bassyouni, El-Ashtoukhy, Amin & El-Latif, 2018; Bassyouni, Hamad, El-Ashtoukhy, Amin, & El-Latif, 2017; Verma, Dash, & Bhunia, 2012). There are several methods available for removal of dyes from wastewater. Many methods have been found to exhibit certain limitations: biological methods suffer by the toxicity of dyestuffs on microbial (Daneshwar, Khataee, & Djafarzadeh, 2006) and other advanced oxidation processes require chemicals which introduce a secondary pollutants (Alshamsi, Albadwawi, Alnuaimi, Rauf, & Ashraf, 2007). Among these methods, conventional coagulation (CC) is the most common and practical method of removal dyes from textile wastewater. However, they present all severe limitations. For instance, CC requires the addition of chemicals that decrease pH and produces large volumes of sludge, requires high treatment times and exhibits a limited effectiveness in the presence of soluble dyes (Kiliç & Hosten, 2010). Electrocoagulation process (EC) has been successfully used for the treatment of textile wastewater being attractive for a simple, reliable, and costly effective method for the treatment of wastewater without need for additional chemicals, and reduces the amount of sludge (Khadim et al., 2016; Fajardo, Martins, Silva, & Martínez-Huitle, Quinta-Ferreira, 2017). However, no general conclusion about the compared efficiency can be made for any type of vat dyes. Vat dye is water insoluble, the structure composed of ketones or least one carbonyl, Bright colors and has the ability to withstand sun and wash well, usually applied to woven knitted fabric fiber and cotton (jiraporn hutakamol, 2011). Therefore, the objective of this study is to investigate the performance of EC for decolorization of insoluble vat dye solution and to propose a rational and systematic comparison between iron and aluminium electrodes and investigated the optimum experimental conditions. The optimum CC conditions (pH and coagulant dose; ferric chloride and PAC dosing) and optimum EC conditions (Fe/Al electrode, initial concentration of vat dye, initial pH of solution, reaction time and applied voltage) were determined and the vat dye removal effectiveness of CC and EC was compared. The results of this study could make reliable comparison of the efficiency for the CC and EC processes to remove vat dye from textile industry scale. The studies on vat dye solution were included to understand the coagulation mechanism in EC and CC. Moreover, the results of study can be widely used in dye removal from textile wastewater and engineering design purpose.

MethodsandMaterials

2.1 Dye solutions

Vat dye was supported by DyStar Co., Ltd, Thailand. The vat dye stock solution was prepared at a concentration of 100 mg/L and the experimental solutions were obtained by diluting the stock solutions to the required initial concentrations. The wavelength of vat dye solution was determined by running a scan of dye solution on a UV-Visible spectrophotometer (GENESYSTM20, USA) and the maximum absorbance wavelengths (λmax) was found at 254 nm.

2.2 Experimental setup

2.2.1 Electrocoagulation Experiments

The EC experimental set was shown in Figure 1 and had two sets for different electrode iron set and aluminium set. They are the most widely used materials for electrode in electrocoagulation process (Verma, 2017). Each set was equipped with four parallel, two anodes and two cathodes with the dimensions of 46 x 55 x 3 mm of iron and aluminium plates. To prepare electrodes by washing with acetone for surface grease removal. The impurities on aluminum and iron electrode surfaces were removed by dipping for 5 min in a solution freshly prepared by mixing 100 cm^3 of HCl solution (35%) and 200 cm^3 of hexamethylenetetramine aqueous solution (2.80%), dried and weighted (Kobya, Can, & Bayramoglu, 2003). The total effective electrode area was 192 cm^2 and the spacing between electrodes was 11 mm and connected to a DC digital power supply. Beaker electrocoagulator with the volume of 1,000 ml was used. The experimental set up was carried out to find the optimum conditions for EC process by varied the initial vat dye concentration from 4 to 28 mg/L, initial pH of solution from 3 to 9 (The pH was adjusted to a desirable value using 0.1 M NaOH and 0.1 M HCl) reaction time from 10 to 35 min and applied voltage from 10 to 30 volt as shown in Table 1. All experimental setups were performed at mixing speed 200 rpm, and with 600 ml of vat dye solution.

2.2.2 Chemical coagulation experiments

In the CC experimental sets were used a Jar test apparatus with four stirring paddles. Two coagulant agents; $FeCl_3$ and PAC were used to compare the removal efficiencies. The operating parameters were kept constant: at ambient temperature, vat dye concentration 28 mg/L for both $FeCl_3$and PAC sets (the volume of vat dye solution 600 mL, rapid mixing of 200 rpm for 1 min and then slow mixing of 40 rpm for 15 min and sedimentation time for 30 min. The concentration of PAC 34.67mg/l as Al (572.50 mg/l as PAC)and $FeCl_3$ 71.78 mg/l as Fe (206.67 mg/l as $FeCl_3$), which equal as using in EC experimental sets. These concentration came from the calculation of the amount of dissolved metal in to the electrocoagulation experiment set by using equation given below (1) (Vepsäläinen, 2012). The polymer concentrations for Fe and PAC set were varied in the range of 1.67-125 mg/L, and initial pH solution was varied in the range of 2-9 to find the optimum conditions of CC for vat dye removal as shown in Table 1.

$$M = \frac{itm_w}{ZF} \qquad (1)$$

The amount of metal cation dissolved during the reactions at the anode can be calculated according to Faraday's law here; I is the current, t is the operation time (s), m_w is molecular weight of the substance (g/mol), F is Faraday's constant (96,485 C/mol), z is the number of electrons involved in the reaction (2 for Fe^{2+} and 3 for Fe^{3+} and Al^{3+}) assuming $Fe(OH)_2(s)$ and $Al(OH)_3(s)$ are supposed to be as main species forms for Fe sets and Al sets respectively, the number of electron moles in dissolution reaction are equal to two and three (Nandi & pastel , 2017) and M is the quantity of metal dissolved (g). The metal dissolved is 34.67 mg/l for Al sets and 71.78 mg/l for Fe set. The reduction of vat dye concentrations was measured and calculated by using equation given below (2).

$$R(\%) = \frac{C_0 - C_t}{C_0} \times 100 \qquad (2)$$

Where C_0 is the initial vat dye concentration before treating with electrocoagulation and coagulation process, and C_t is the vat dye concentration remain after sedimentation for 30 min.

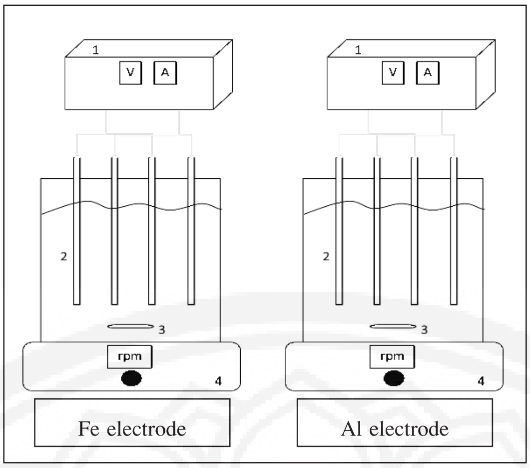

Figure 1 Schematic of experimental set up for Fe and AL electrocoagulation sets (1) DC power supply, (2) electrocoagulation cell, (3) magnetic bar-stirrer, (4) digital magnetic stirrer.

Table 1 The conditions for Chemical coagulation experimental sets.

Factor		**Ferric chloride ($FeCl_3$)**		**Poly aluminum chloride (PAC)**	
Varied	Unit	Polymer dosage (mg/l)	pH	Polymer dosage (mg/l)	pH
Polymer dosage	mg/l	1.67 - 125	5	1.67 - 125	5
pH	-	108.33	3,5,7,9	58.33	3,5,7,9

2.2.3 Comparative performance of vat dye removal efficiency of EC with CC

As the results of optimum conditions finding for CC and EC experiments, the vat dye removal efficiency (%) was calculated using the equation (2) and compared.

2.2.4 Application of EC to treat real wastewater

To apply the EC process to treat real wastewater from textile household industry. The study was carried out to collect raw wastewater from three sampling sites of textile household industries. Three sampling sites of textile household industry were selected; two sampling sites namely, Samko and Sawai which locate in Surin province. Another sampling site namely,Kayabatic which locates in Nakhonratchasima province. Wastewater samples were analyzed for various parameters and analytical methods as shown in Table 2.

Table 2 Parameter and analytical methods for wastewater.

Parameter	**Unit**	**Analytical methods**
pH	-	pH Meter
Color	Pt-Co	2120 C. Spectrophotometric Metod
COD	mg/l	5220 C. Close Reflux , Titrimetric Method

Results and Discusion

3.1 Electrocoagulation Experiments

The EC experimental set up was carried out to find the optimum conditions for EC by varied initial vat dye concentration, reaction time, initial pH solution and applied voltage as shown in Table 1 and investigated their effect on vat dye removal.

3.1.1 The effect of initial vat dye concentration

The effect of the initial vat dye concentration was varied of 4,8,12,16,20,24 and 28 mg/L for both type of electrode sets. Figure 2 presented the vat dye removal efficiency of Fe and Al type electrode sets versus initial vat dye concentration. The results showed the set of Fe electrode had removal efficiencies slightly superior in range of 72.14 - 95.85% than the set of Al electrode in range of 65.91 - 95.67 %. As it can be observed both type of electrode sets are significant increase of removal efficiency with increasing initial vat dye concentration from 8 to 16 mg/ L and after that they are slightly increased of removal efficiency. And the maximum removal efficiency was found at initial vat dye concentration at 28 mg/L for both of Al and Fe electrode sets. One of most significant pathways of vat dye removal by EC is adsorption of dye molecule on metal hydroxide flocs. According to Faraday's law, a constant amount of metal ion is liberated to solution at the same current and time for all initial vat dye concentration. Thus, the same amount of flocs is limited and only a specific amount of flocs is able to adsorb specific amount of vat dye molecule. So the removal efficiency depend on the appropriate of amount form flocs to adsorb all dye molecules (El-Ashtoukhy, Amin, & Abdel-Aziz, 2012; Bassyouni et al., 2017). Therefore, for comparative performance initial vat dye concentration 28 mg/L was used as optimum initial vat dye concentration for further experiment of Fe and Al electrode sets.

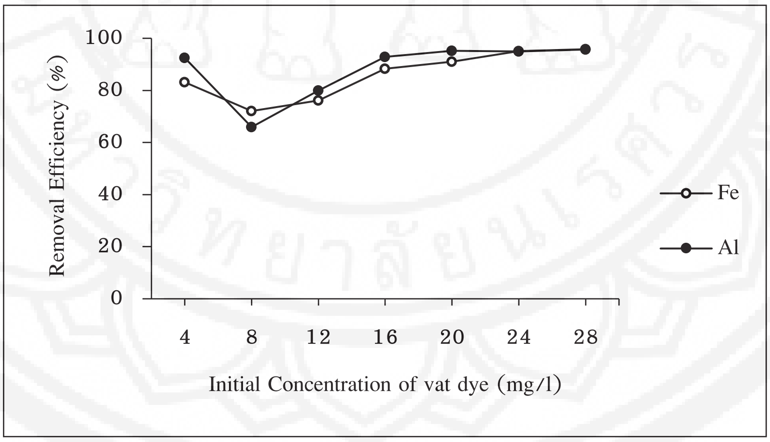

Figure 2 The effect of initial concentration of vat dye on vat dye removal efficiency of electrocoagulation

3.1.2 The effect of reaction time

The results of varies electrolysis reaction time versus at dye removal efficiency was shown in Figure 3 to study their effect on vat dye removal by EC. It can be observed that increasing reaction time over 10 and 15 min for Al and Fe electrode set respectively, the vat dye removal efficiencies significant increased with increasing of reaction time. These results occur due to reaction time relates with the production rate of Fe^{2+} or Fe^{3+} ions from Fe electrodes and Al^{3+} from Al electrode. And the vat dye removal efficiency depends directly on the concentration of hydroxyl and metal ions produced on the electrodes (Daneshvar et al., 2006). The released metal ions and hydroxyl from electrodes formed more hydroxide flocs, which would adsorb more pollutant particles as the time extended (Drouiche et al., 2009). But beyond the optimum electrolysis reaction time resulted in slightly increased in removal efficiency, this would not be applied due to the high energy and electrode consumption (An, Huang, Yao, & Zhao, 2017). This could be related to charge loading and affects the performance of electrocoagulation on colour removal (Verma, 2017) and decreasing extent of cathodic reduction and generation of nascent electrocoagulantflocs (Akyol, 2012). As shown in Figure 3, the maximum vat dye removal efficiency of Al electrode sets was 97.17% occurred at reaction time for 30 min and Fe electrode set was 97.33% occurred at reaction time for 20 min. It can be notice that both of electrode sets had the similar range of vat dye removal efficiencies. These results is corresponds by the previous literatures that found normally the optimal reaction time was 20-30 min (Kobya, Hiz, Senturk, Aydiner, & Demirbas, 2006; Malakootian, Mansoorian, & Moosazadeh, 2010; Xu & Zhu, 2004). Therefore, for further experiments,reaction time for 30 min and 20 min were used as optimum reaction time for Al and Fe electrode sets respectively.

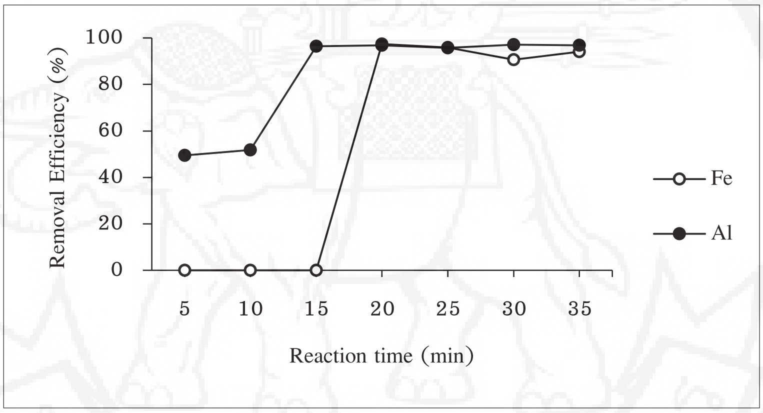

Figure 3 The effect of reaction time on vat dye removal efficiency of electrocoagulation.

3.1.3 The effect of initial pH

In order to investigate the effect of the initial pH of of vat dye solution on removal efficiency batch experiments were performed by adjusting the initial pH in the interval from 3 to 9. In each batch experiment of Fe electrodes, the initial vat dye concentration, reaction time and applied voltage were kept constant at 28 mg/L, 20 min and 30 V respectively. And for each batch experiment of Al electrodes, the initial vat dye concentration, reaction time and applied voltage were kept constant at 28, mg/L, 30 min and 30 V respectively. The effect of the pH on electrocoagulation is illustrated in Figure 4. The results of Al electrode sets showed that when pH of vat dye solutions was between 5 to 9, with maximum removal efficiency at 98.23%. In wastewater treatment using electrocoagulation, pH plays a very important role in determining treatment efficiency (Verma, 2017).And for Al electrode sets, higher efficiencies were recorded in range which is close to the optimal pH for $Al(OH)_3(S)$ solid formation. The flocs of $Al(OH)_3(S)$ have large surface areas, which are useful for a rapid adsorption of soluble organic compounds and trapping of colloidal particles (Bayamoglu, Kobya, Can, & Sozbir, 2004).Previous research of Barrera- Diaz et al.(2006) studied the predominant aluminum species present in the solution at different pH values, where they found that at pH less than 3.5, Al^{3+} is the major specie present, for pH values between 4 and 9.5, $Al(OH)_3(S)$ predominates and when the pH is greater than 10, $Al(OH)_4^-$ forms. These results are quite reasonable since $Al(OH)_3(S)$ traps the colloids/pollutants in a sweep coagulation manner as it precipitates. On the other hand, $Al(OH)_4^-$ is known to be a very soluble weak coagulant and will not be able to destabilize colloids. Several authors reached the same conclusion that the maximum performance of EC using aluminum anodes is around neutral pH (Moussa, El-Naas, Nasser, & Al-Marri, 2017). And the results of Fe electrode sets showed when pH of vat dye solutions was between 3 to 9, dye removal efficiency was optimal with a maximum at 97.33% at pH 7. These results supported the study of Moussa et al. (2017) that concluded the optimum operating pH range of iron electrode for EC is 5-9 and operation at an initial pH of 8-9 is favorable to ensure complete oxidation of ferrous ions which are known to be highly soluble, poor coagulants with no adsorption capacity of pollutants. And at highly alkaline pH, undesired $Fe(OH)_4^-$ forms which is a weak coagulant and deteriorates EC performance (Vepsäläinen, 2012). Therefore, for further experiments, initial pH solution at 5 and 7 were used as optimum pH for Al and Fe electrode sets respectively.

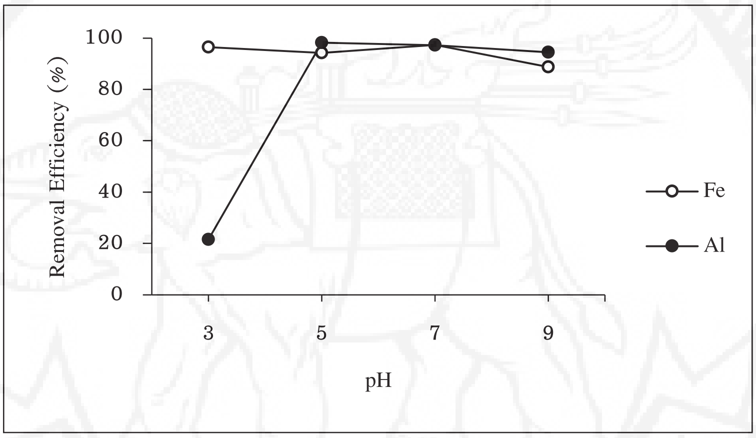

Figure 4 The effect of initial pH on the vat dye removal efficiency of electrocoagulation.

3.1.4 The effect of applied voltage

The effect of current density on vat dye removal efficiency was investigated by varies applied voltage from 10 to 30 V and maintain the constant initial concentration of vat dye 28 mg/l with initial pH of 7 and reaction time for 20 min for Fe electrode sets and for Al electrode sets, the initial concentration of vat dye 28 mg/l with initial pH solution of 5 and reaction time for 30 min. As shown in Figure 5, the efficiency of vat dye removal increase according to increasing of applied voltage. The maximum vat dye removal efficiencies of both Al electrode and Fe electrode sets occur at 30 V with 98.23% and 97.33% respectively. The higher removal efficiency of impurities with increased applied voltage might be related to the higher amount of ions produced on the electrodes that promote destabilization of the pollutant molecules and, finally, the agglomeration of the induced flocs, with increase in the hydrogen evolution (Cañizares, Martínez, Rodrigo, & Sáez, 2009). It is well known that current density determines the coagulant production rate, regulates bubble production, and therefore affects the growth of flocs and therefore current density is one of the most important parameter for controlling the reaction rate in the electrocoagulation (Kobya, Demirbas, Sik, & Oncel, 2015). Therefore, for further experiments, applied voltage at 30 V was used as optimum applied voltage for both of Al and Fe electrode sets.

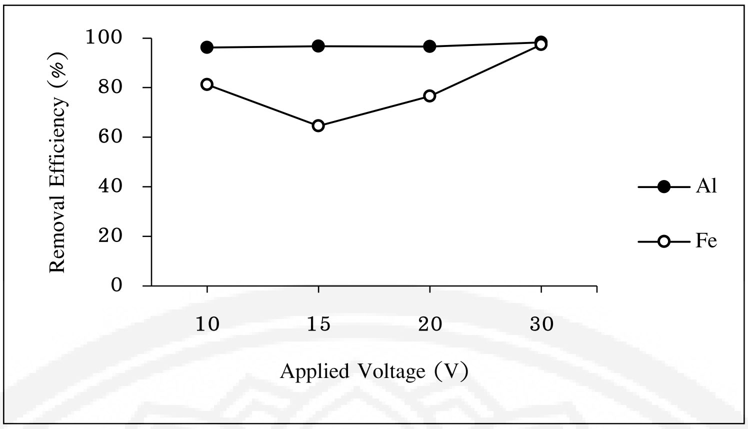

Figure 5 The effect of applied voltage on the vat dye removal efficiency in electrocoagulation

3.1.5 Conclusion of optimum condition for EC process

The results of optimum conditions for EC as summarized in Table 3 were carried out to measure the performance of EC process. They were found the optimum condition for Al and Fe electrode have same initial concentration of 28 mg/L and applied voltage 30V whereas, reaction time 30 and 20 min and initial pH were 5 and 7 respectively.

Table 3 The conclusion of optimum conditions for EC process.

Electrode	initial concentration of vat dye (mg/l)	reaction time (min)	pH	applied voltage (volt)	%Vat dye removal
Al	28	30	5	30	98.23
Fe	28	20	7	30	97.33

3.2 Chemical coagulation experiments

The optimum condition of Chemical coagulation

The chemical coagulation (CC) studies were carried out by using of two kind of coagulant; PAC and $FeCl_3$. The PAC concentration of 34.67 mg/l as Al (572.50 mg/l as PAC) and $FeCl_3$ concentration of 71.78 mg/l as Fe (206.67 mg/l as $FeCl_3$), which calculated amount of dissolved metal the equal as using in EC experimental sets were used.The CC studies were carried out using the jar test method to determine the optimum pH range, and polymer dosage for PAC and $FeCl_3$ sets. And the optimum initial vat dye solution of 28 mg/L that was obtained from the results of EC experiments, was used. Overall optimum parameters with results were given in Table 4 for CC process.They were found the optimum condition for PAC sets, at polymer dosage of 58.33 mg/L and pH 2with the removal efficiencies of 9.88% and the optimum condition for $FeCl_3$ sets, at polymer dosage of 108.33 mg/L and pH 5 with the removal efficiencies of 27.66% as shown in Figure 6 and 7. The results showed the CC process had poor removal efficiency in vat dye solution.

Table 4 The conclusion of optimum conditions for chemical coagulation studies.

Set	coagulant dosage (mg/l)	Polymer (mg/l)	pH	Removal (%)
$FeCl_3$	206.67	108.33	5	27.66%
PAC	572.50	58.33	2	9.88%

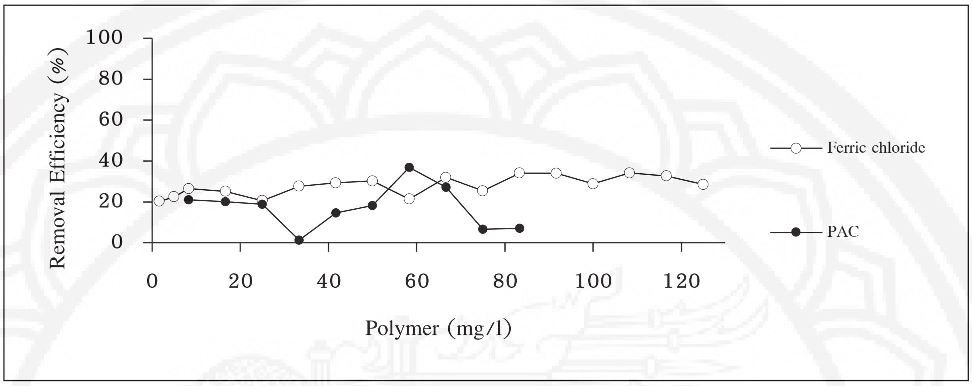

Figure 6 The results of CC study to determine the optimum polymer dosage.

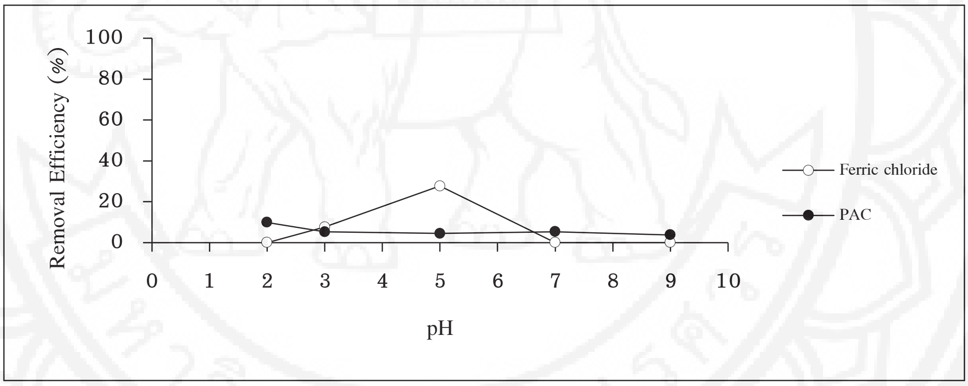

Figure 7 The results of CC study to determine the optimum pH .

3.3 Comparative performance of EC and CC

The results of optimum conditions for EC and CC studies were carried out to treat vat dye solution and measuring removal efficiency of each process for comparison of performance. The conclusions of vat dye removal efficiencies for EC and CC processes are presented in Figure 8. The results showed the vat dye removal efficiencies of EC were in range of 97.33-98.23% respectively and were significant higher than removal efficiencies of CC in range of 9.88–27.66%. And the comparison of vat dye removal efficiencies for Al and Fe of EC processes showed similar removal efficiency and for using Fe as coagulant had much effective than using Al. These result are similar to previous researches that concluded aluminum or PAC had not effectives for the vat dye because vat dye has too small particle for coagulant (Hai, Yamamoto, & Fukushi, 2007). As a conclusion, the superiority of EC much over CC is clearly established for vat dye removal.

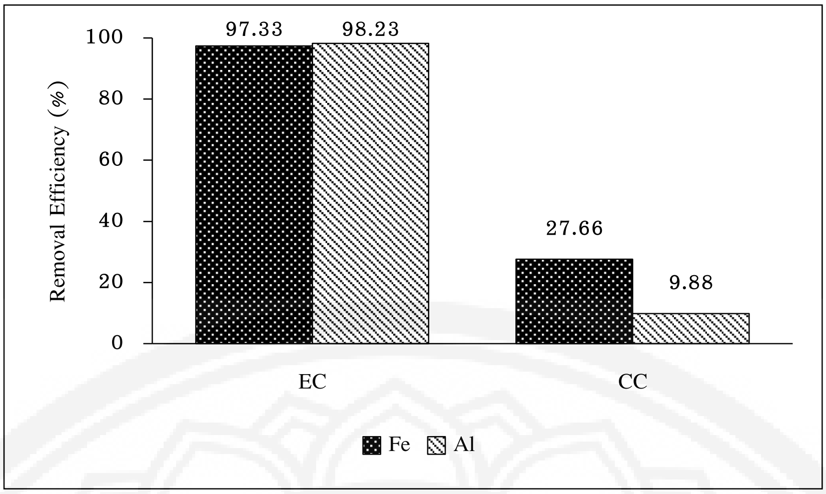

Figure 8 The conclusion of overall color removal efficiencies for EC and CC processes with different type of electrode and coagulant

3.4 Application of EC to treat wastewater from textile household industies

The results of optimum conditions as shown in Table 3 for EC were applied to treat raw wastewater from three sampling sites of textile household industries for measuring the performance. The results of COD and color removal efficiencies of EC were presented in Table 5 and Figure 9. They showed EC had COD and color removal efficiencies were 60.86 % and 82.63% respectively for using Al electrode, and were 65.85% and 87.24% respectively for Fe electrode. The results showed EC process had color removal efficiencies slightly lower than found in batch experiments of dye solution. And the results showed the EC process using Fe and Al electrode were in the similar range of COD and color removal efficiencies. These results proved that EC process cloud be used as an effective wastewater treatment for color removal from textile wastewater. However, the COD of the effluent had not meet the standard, thus the author recommend to use EC process as pretreatment of biological treatment. The EC process will be benefit for color removal that is toxic to micro-organism.

Table 5 The removal efficiencies of EC process to treat wastewater from textile household industies

Parameter	Unit	Influent (min - max)	Al electrode set		Fe electrode set	
			Effluent (min - max)	Removal efficiency (%)	Effluent (min - max)	Removal efficiency (%)
pH	-	5.13-10.25	7.91-8.06	-	8.13-9.54	-
COD	(mg/l)	1,726-2,560	395-1,536	60.86±20.3	436-1,280	65.85±14.53
Color	(Pt-Co)	511-9,256	74 - 1,278	82.63±5.65	26-497	87.24±6.68
	ADMI	426-7758	64.24-1,074	82.63±5.65	23.99 - 418.89	87.24±6.68

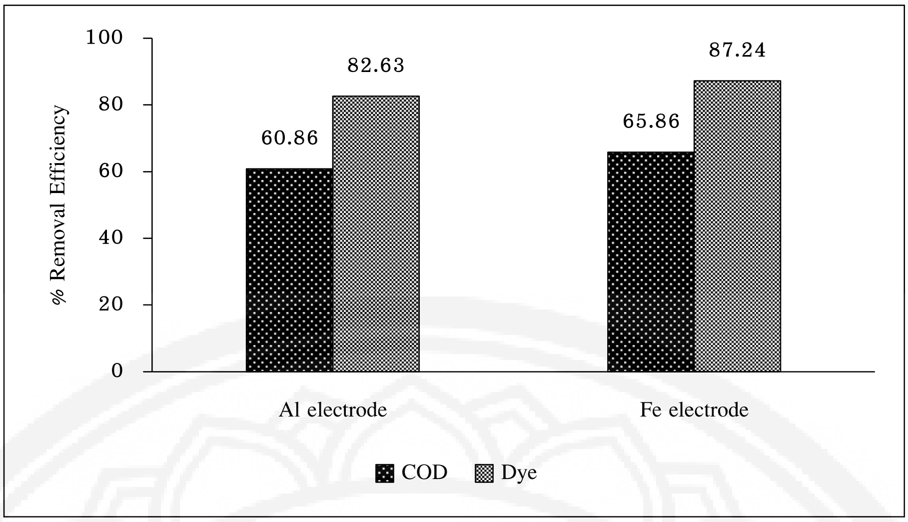

Figure 9 The COD and color removal efficiencies of EC process to treat wastewater from textile household industry

Conclusions

The studies on comparison of EC and CC in vat dye removal led to the following conclusion; the EC process is more efficient process than CC process to treat vat dye solution. The initial concentration, initial pH, reaction time and applied voltage come out to be major operating variables which regulate the vat dye removal efficiency of EC. The application of EC to treat wastewater from textile household industries had color and COD removal efficiency higher than 80% and 60 % respectively.It was concluded that the EC process is a competitive alternative process and presents a high potentiality for the treatment of color from textile wastewater. And the EC has advantages over CC such as high efficiency and easiness of process control. The further works should address a better understanding of EC mechanisms in order to improve scale up methodology.

Acknowledgments

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