



## INVESTIGATION OF ELECTROCOAGULATION AND ELECTROOXIDATION METHODS OF REAL TEXTILE WASTEWATER TREATMENT

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### ABSTRACT

In this study, it was investigated the treatability of real wastewater from textile industry by electrochemical treatment methods (electrocoagulation and electrooxidation). Effect of important operating parameters such as, electrode type and combination (Al-Al, Fe-Fe, Al-Fe, Fe-Al, Pt-Fe), pH, reaction time and potential were investigated on removal efficiency of color and chemical oxygen demand (COD). The initial color and COD concentrations of the wastewater were 395 Pt-Co and 1040 mg/L, respectively. At the end of the electrocoagulation experiments, concentrations of color and COD were decreased to 28 Pt-Co and 115 mg/L, respectively. Results showed that at pH 3 and 6 V potential, up to 93% color and 89% COD removal efficiencies were obtained in the reactor consisting of Fe-Fe electrodes. COD and color were removed at the rate of 88% and 92%, respectively in the study done with Al-Al couple at 10 V in natural pH (6.96). COD removal was achieved in the ratio of 93% at 6V as a result of the electrooxidation study with a couple of Pt-Fe electrodes. The study showed that the removal process was promising and it was reached to the discharge limit values for the color and COD with each electrode couple specified in the regulation. When considering the removal efficiencies, electrocoagulation process is the best treatment method is for this study. In terms of energy consumption, the electrooxidation process is more economical to effectively remove COD

**Keywords:** COD, Color, Electrocoagulation, Electrooxidation, Textile wastewater

## 1. INTRODUCTION

Textile wastewater is the wastewater that have high volume and whose composition can show great changes. That it contains non-degradable colorants and toxic compounds with a degree of high probability brings along the potential to create a risk in terms of the receiving waters. Reactive dye is one of the most used dyes in the textile industry. These carcinogenic and mutagenic dyes have harmful effects on the environment. The dyed wastewater that is given to the receiving medium changes the color of the water. It reduces the light permeability and oxygen content in the water environment and affects the photosynthetic activity negatively. Therefore, removing of wastewater which is stemmed from textile industries with appropriate and effective methods is very important [1, 2].

Electrochemical treatment methods have attracted attention as an alternative wastewater treatment method in recent years, and have begun to take traditional processes's place [3]. Electrochemical methods for the treatment of textile wastewater have been a special point of interest. Electrochemical treatment methods have begun to use widely with its many benefits such as high removal efficiency, low cost, minimum level of chemical usage, simple operation parameters. Factors such as treatment time, current density, type of electrode that is used are the most fundamental differences that distinguish electrochemical treatment processes from other processes [4]. Electrochemical treatment processes are classified under three main headings notably, electrocoagulation (EC) and electroflotation (EF), electrooxidation (EO) [5-7]. Electrocoagulation is based on the principle that affecting of pollutants in wastewater from electrical charges. Metal ions come to release by dissolution of anode in process, and removal of contaminant is ensured by coagulation, adsorption and precipitation processes performing by metal hydroxide flocks [8]. The treatment performs with 3

phases in the electrocoagulation process. These are; i) formation of metal hydroxides by reactions occurring at the surface of the electrodes, ii) forming of coagulation by metal hydroxides with contaminants at the liquid phase, and iii) removal of contaminants by adsorption, coagulation sedimentation or flotation mechanisms [9]. The electrochemical reactions occurring in different electrodes during the electrocoagulation process are shown in Table 1.

**Table 1.** Electrochemical reactions occurring in the electrocoagulation process [10]

Anode	Cathode
$2\text{H}_2\text{O} - 4\text{e}^- = 4\text{H}^+ + \text{O}_{2(\text{g})}$	$2\text{H}_2\text{O} + 2\text{e}^- = 2\text{OH}^- + \text{H}_{2(\text{g})}$ (alkali solution)
$4\text{OH}^- - 4\text{e}^- = 2\text{H}_2\text{O} + \text{O}_{2(\text{g})}$	$2\text{H}_3\text{O}^+ + 2\text{e}^- = 2\text{H}_2\text{O} + \text{H}_{2(\text{g})}$ (acidic solution)
	$2\text{H}_2\text{O} + \text{O}_2 + 4\text{e}^- = 4\text{OH}^-$
<i>With Fe electrode</i>	
$\text{Fe}(\text{s}) - 2\text{e}^- = \text{Fe}(\text{aq})^{2+}$	$\text{Fe}(\text{OH})_3 + \text{OH}^- = [\text{Fe}(\text{OH})_4]^-$
$\text{Fe}^{2+} - \text{e}^- = \text{Fe}^{3+}$	$[\text{Fe}(\text{OH})_4]^- + 2\text{OH}^- = [\text{Fe}(\text{OH})_6]^{3-}$ (alkali solution)
$\text{Fe}(\text{aq})^{2+} + 2\text{H}_2\text{O} = \text{Fe}(\text{OH})_2 + 2\text{H}^+$	
$\text{Fe}^{3+} + 3\text{H}_2\text{O} = \text{Fe}(\text{OH})_3 + 3\text{H}^+$	
$\text{Fe}^{2+} + 3\text{OH}^- - \text{e}^- = \text{FeOOH} + \text{H}_2\text{O}$	
<i>With Al electrode</i>	
$\text{Al}(\text{s}) - 3\text{e}^- = \text{Al}(\text{aq})^{3+}$	$\text{Al}(\text{s}) + 4\text{OH}^- = [\text{Al}(\text{OH})_4]^- + 3\text{e}^-$ (alkali solution)
$\text{Al}(\text{aq})^{3+} + 3\text{H}_2\text{O} = \text{Al}(\text{OH})_3 + 3\text{H}^+$	

Iron (III) complexes, formed by dissolution of iron electrodes in water which are frequently used in electrocoagulation, are important. The hydroxyl ions in the water leading to the formation of  $\text{Fe}(\text{OH})_x$  complexes depending on the pH of the medium by joining with the  $\text{Fe}^{3+}$  ions dissolved from the electrodes. This treatment method which is based on the principle of adsorbing of various contaminant parameters in the water caused by the iron complexes or metal hydroxides with high adsorption capacity and removing them from water by precipitation is used for many wastewaters nowadays [11-13].

Aluminium electrode has very low resolution in neutral pH, which means that pH adjustment is not usually required for aluminium electrode. The cost of the treatment is reduced and treatment can be performed with high efficiency in neutral conditions [14-15].

Aluminum ions in the aqueous medium present a complex equilibrium with different monomeric species such as  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}(\text{OH})_3$  and  $\text{Al}(\text{OH})_4^-$  depending on the pH conditions [16].

Very small gas bubbles comes out of the water as a consequence of reactions that occur on the electrodes during the electrocoagulation process. Given that these gas bubbles are also the basis of electroflotation, some contaminants can be removed from the water by electroflotation method during electrocoagulation [17].

The main principle in the electrooxidation (EO) method is to achieve the desired oxidation with the gases ( $\text{O}_2$  and  $\text{H}_2$ ) emerging by means of electrodes using undissolving electrodes (Ti, Ru, Pt, stainless steel etc.) [18]. While many substances can be oxidized with this process, those that are difficult to biodegrade are converted into biodegradable organic compounds or end products such as  $\text{CO}_2$  or  $\text{H}_2\text{O}$ . The electrode that plays an active role is anode in electrooxidation. For that reason, the main parameter which is effective in this process is the catholytic activity of anode. In addition, current, temperature, pH and diffusion rate of organic compounds and other oxidants are also important. The oxidation of organic contaminants in the electrolytic cell realizes in two different ways: i) direct anodic oxidation ii) indirect oxidation. If anode has a potential high enough, chloride ions in the

wastewater can convert to chlorine or secondary reactions such as direct oxidation of organic compounds may occur as well [19-20].

Treatment of textile wastewater is quite important as it can lead to severe contamination of surface and ground waters [21]. The organic dyes are hardly removed by conventional wastewater treatment technologies, such as biological and physico-chemical processes [19]. Recently, the UNESCO's World Water Development Report [22] raised a growing concern to enforce the development of novel technologies that are capable to treat these polluted industrial dye effluents efficiently [20].

Various studies have been reported about electrocoagulation and electrooxidation methods for remediation of textile wastewater [1]. These studies carried out for the treatment of textile wastewater vary within themselves because of pH, electrode type, current density parameters.

Merzouk et al. (2010) investigated the treatment of a textile wastewater by using electrocoagulation and electro-flotation processes in their study. Optimum working conditions in the study were determined as the distance between electrodes in 1 cm, 10 min treatment time, and pH 7.6 and 11.55 mA/cm<sup>2</sup> current density. The 85.5% suspended solids, 76.2% turbidity, 93% color and 79.7% COD removal were achieved under optimum conditions [23]. Khandegar and Saroha (2013) studied on the electrocoagulation of synthetic solution containing Acid Red 131 dye using Al electrodes. The results were indicated only with color removal efficiency (CRE). The maximum CRE was obtained as 98% for a synthetic dye solution at 0.0625 A/ cm<sup>2</sup> and at pH 11 for 120 min [21]. Yüksel et al. (2013) investigated the electrochemical treatment of reactive orange 84 from textile wastewater using stainless steel (SS) and Fe electrodes. At the end of the experiment, SS electrodes were found to be better than Fe electrodes to treat both dye and textile wastewater. The textile wastewater was treated successfully with 89.7% of COD, 91.2% of TOC and 90.3% of turbidity at 110 A/m<sup>2</sup> by using SS electrodes [24]. In a study conducted by Naje et al. (2017) [25], a new rotary bed electrocoagulation reactor was investigated for optimum conditions. Al was used as the electrode and the removal was observed approximately at the rate of 97% in COD and 98% in color in the system where the reaction time was 10 min, pH was 4.57 and the distance between the electrodes was 1 cm.

The selected studies related to electrooxidation in textile wastewater are mentioned below. Aquino et al. (2014) reported that the 100% color and 86% COD removal of raw textile effluent obtained in 180 min using a Ti-Pt/ $\beta$ -PbO<sub>2</sub> at 75 mA/cm<sup>2</sup> [26]. Another study performed by Ling et al. (2016) on electrooxidation of textile wastewater, Ti/PbO<sub>2</sub> was used as anode. After the finishing of the 60 min treatment, the removal of color and COD was found approximately as 100% and 78% respectively at 12 mA/cm<sup>2</sup> [27]. Zou et al. (2017) conducted that the electrooxidation of real textile wastewaters using a boron-doped diamond (BDD) anode. They reported a complete COD removal under the optimum conditions (60 mA/cm<sup>2</sup>, 3 g/L NaCl, pH 2) in 3 h for the real textile wastewaters [28].

When the literature is examined, it is seen that the studies on the textile wastewater treatment concentrate on synthetic or simulated dyes wastewaters. However, compared to the synthetic wastewater, the content of real textile effluents is more complex and usually contain organic and inorganic chemicals such as surfactants, inhibitor compounds, active substances, chloride and sulfate, dyeing substances and total phosphate [20-21,28]. For these reasons, this study focused on remediation of real textile wastewater.

Treatment of textile effluents studies done by using hybrid/composite electrode couple such as Fe/Al and especially Pt-Fe, are inadequate in the literature. While in this study, different and hybrid electrode couples such as Al/Al, Fe/Fe, Fe/Al, Al/Fe and Pt/Fe were used in order that both Fe and Al ions dissolve at the same time during EC and EO processes.

## 2. MATERIAL AND METHODS

### 2.1. Characteristics of Real Textile Wastewater

The treatment efficiency of wastewater obtained from a textile factory producing main woven fabric and located in Seyhan district of Adana province was investigated by using EC and EO processes at different conditions. It is directly collected from the entrance of the reservoir. The textile wastewater was largely composed of dyes and various organic/inorganic substances. The certain content could not be detected due to possible commercial secrets. The characteristics of the textile wastewater are given in Table 2.

**Table 2.** Physico-chemical characteristics of the real textile wastewater

Parameter	Value
pH	6.96
Conductivity (mS/cm)	4.15
Color (Pt-Co)	395
Suspended soil (mg/L)	330
COD (mg/L)	1040

### 2.2. Electrochemical experiments and analysis

Fe and Al electrodes were used for the electrocoagulation process in the study. Iron electrodes have a surface area of 40 cm<sup>2</sup> and Al electrodes have a surface area of 30 cm<sup>2</sup>. Electrocoagulation experiments were carried out by using Al/Al and Fe/Fe (anode-cathode) electrode couples in wastewater at three different potentials (6V, 8V, 10V) during 3 h in definite time intervals. Thus, the effects of potential and time on COD and color removal were determined. The effects of Al/Fe and Fe/Al hybrid electrode couples on the COD and color removal were investigated depending on the potential on condition that keeping Fe as stable at the later stage.

Electrochemical oxidation test was performed by using Pt plate (anode) and Fe plate (cathode) at the natural pH conditions and 3 different potential (6 V, 8 V and 10 V). The distance between the electrodes was determined as 2 cm, and the 2 cm<sup>2</sup> platinum and 40 cm<sup>2</sup> iron electrodes were used in the EO process.

The effect of pH on COD and color removal was investigated after the optimum electrode couple and potential had been determined. Since electrocoagulation had better removal efficiency compared EO, the effect of pH was only investigated for the electrocoagulation process. Experiments of pH were performed by adjusting the initial pH of the wastewater to 3, 5, 7 and 9 by using NaOH and HCl solutions.

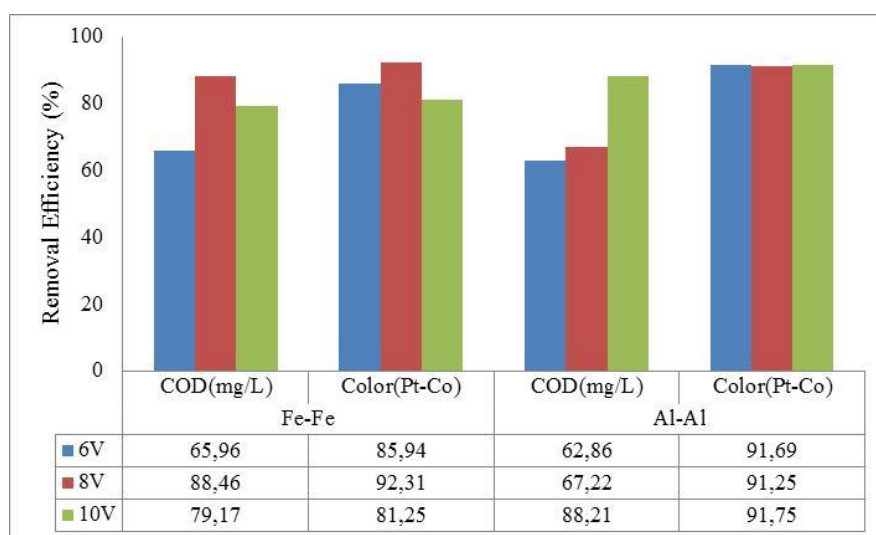
The system was supported with a constant potential during the tests by using PC 2000 DC power supply. COD and color analyzes were made in the study by taking samples from wastewater in volume of 400 ml reactor and after 3 h of treatment period. COD was determined by using HACH LCK 514 thermal reactor and 314 test kits. Color was directly measured with DR 3900 spectrophotometer as Pt/Co.

### 3. RESULTS AND DISCUSSION

#### 3.1. The Effect of Electrode Couple and Cell Potential on Electrocoagulation

Different electrode materials have different electrochemical properties. Selection of the appropriate electrode material is the most important factor affecting the treatment efficiency in the electrocoagulation (EC) process [29]. For this purpose, different electrode types (Al/Al and Fe/Fe) and hybrid electrode couple (Al/Fe and Fe/Al) were used to determine the effect of electrode material on COD and color removal. In the first stage, experiments by using three different potentials (6V, 8V and 10V) were performed to determine the couple and the optimum electrode material. At the end of the experiment, the maximum COD and color removal with the Fe/Fe electrode couple at the 8 V potential (0.88 mA/cm<sup>2</sup> current density) were determined as 88% and 92%, respectively. The maximum color removal with the Al/Al combination which is the other electrode couple were realized as 92% at 10 V (1.05 mA/cm<sup>2</sup>).

The percentages of COD and color removal that were obtained at 3 different potentials with different electrode couples at the end of the experiments are shown in Figure 1.

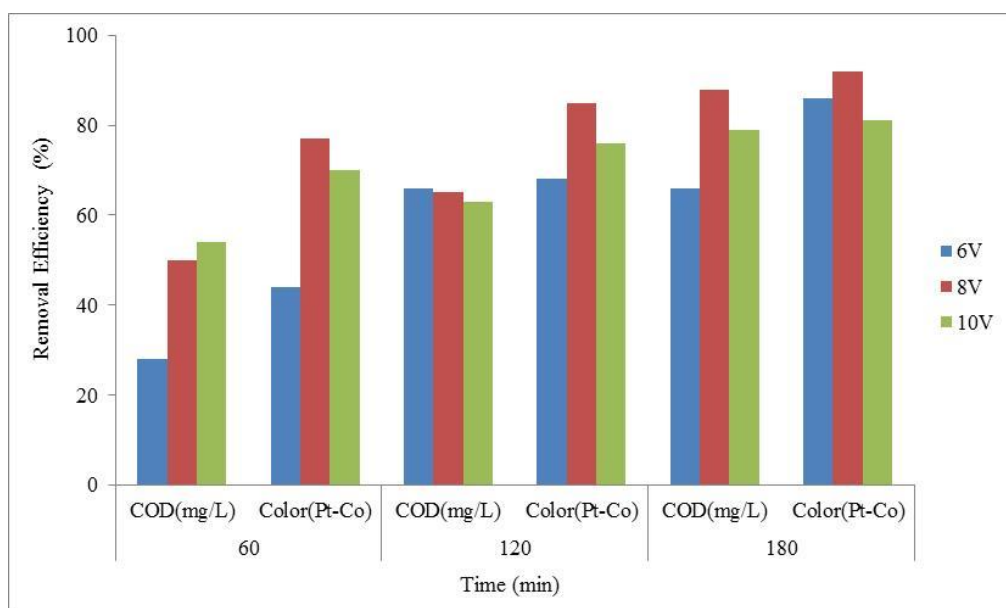


**Figure 1.** Effect of three different potential and different electrode couples on COD and color removal (pH 6.96; current densities are 0.48 mA/cm<sup>2</sup>, 0.88 mA/cm<sup>2</sup> and 1.05 mA/cm<sup>2</sup> for 6V, 8V and 10 V respectively at the Fe/Fe electrode couple. Current densities are 1.5 mA/cm<sup>2</sup>, 1.87 mA/cm<sup>2</sup> and 1.9 mA/cm<sup>2</sup> for 6V, 8V and 10 V respectively at the Al/Al electrode couple.)

When the graph is examined, it is seen that the COD removal was increased by increasing the potential in the Al/Al electrode couples, and the maximum removal was 88% at the end of 3 h. There is a linear relationship between the potential that is applied in electrochemical treatment studies and the removal efficiency. The amount of dissolved metal ions increases by increasing the potential or the current density passing through system, and as a result more hydroxide cationic complexes emerges (Faraday's Law). These complexes adsorb strongly to the contaminants or precipitate with them. Meanwhile, the hydrogen production rate increases and the size of the bubbles which increase the mass transfer and flocculation performance in the EC process decrease. As a result higher efficiency is obtained [10].

The experimental results which were performed with Fe/Fe electrode pair at different potentials as time-dependent are shown in Figure 2. COD and color removal increased as time-dependent. However, the highest COD and color removal (88% and 92%) were obtained at 8V and at the end of 3 h in the Fe/Fe electrode couple. At the end of the experiment done at 8V, while the pH of the solution was 6.96 at the beginning, it was determined as 11.08 at the end of the experiment. When the iron pH-

Eh (pourbaix) diagram is examined, it is understood that the Fe ions were completely in the  $\text{Fe}(\text{OH})_3$  type and precipitated at this pH.



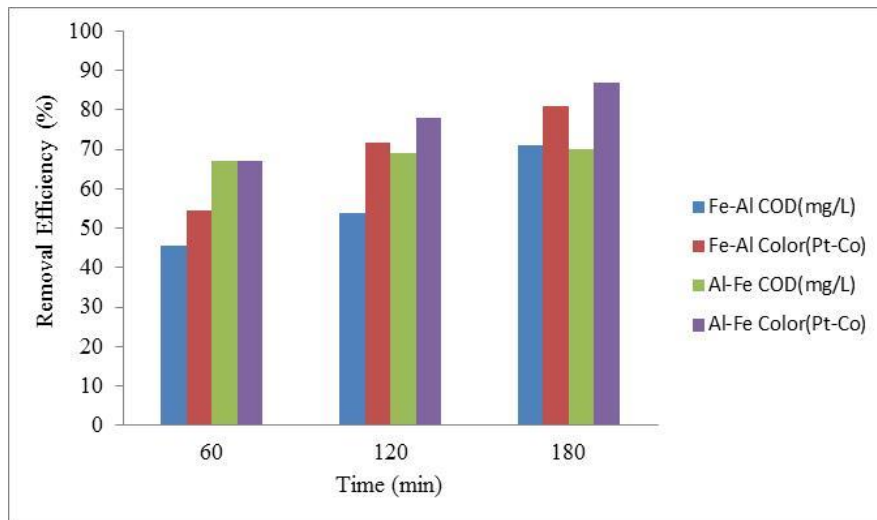
**Figure 2.** Effect of time and potential on COD and color removal when using Fe-Fe electrode couple at pH 6.96 (Current densities are  $0.48 \text{ mA/cm}^2$ ,  $0.88 \text{ mA/cm}^2$  and  $1.05 \text{ mA/cm}^2$  for 6V, 8V and 10 V respectively at the Fe/Fe electrode couple)

Additionally, in the experiments made on Fe/Fe electrode at 8V, while the current passing through the system was initially 35 mA, it decreased to 28 mA at the end of the experiment. This decrease indicates why the removal at 8V is much more. When applying a 6V potential ( $0.48 \text{ mA/cm}^2$ ) to the system, the removal efficiency decreased since the current increased from 19 mA to 23 mA. The current that was measured in the system decreased from 42 mA to 40 mA at the 10 V.

When the results are evaluated, it is seen that the removal efficiencies close to each other was reached with Fe/Fe and Al/Al couples in textile wastewater treatment. It was accepted in the study that the optimum potential was 8V and the best electrode couple was Fe/Fe because of the higher efficiency at lower potential.

The electrode materials which are used the most widely in the EC process are Al and Fe [2, 10]. However, researches on the use of Al and Fe hybrid electrodes for the treatment of textile wastewater are very important [30]. The composite electrode (Fe/Al) was used in the EC process in order to understand the effect of both Fe and Al ions on releasing them simultaneously and effectiveness of the treatment activity during the electrolysis reactions.

Therefore, COD and color removal from textile wastewater with the combination of Al/Fe and Fe/Al hybrid electrodes were investigated after the optimum potential and duration had been determined. There are few studies evaluating composite/hybrid electrode (Fe/Al) activity for the treatment of textile wastewater. It was determined which electrode material is more effective in textile wastewater treatment by hybrid electrode matching. COD and color removal which were realized with hybrid electrode couples at 8V for 3 h at the end of the experiment are shown in Figure 3.



**Figure 3.** Effect of hybrid electrode couples on COD and color removal at the 8V potential for 3 h at pH 6.96

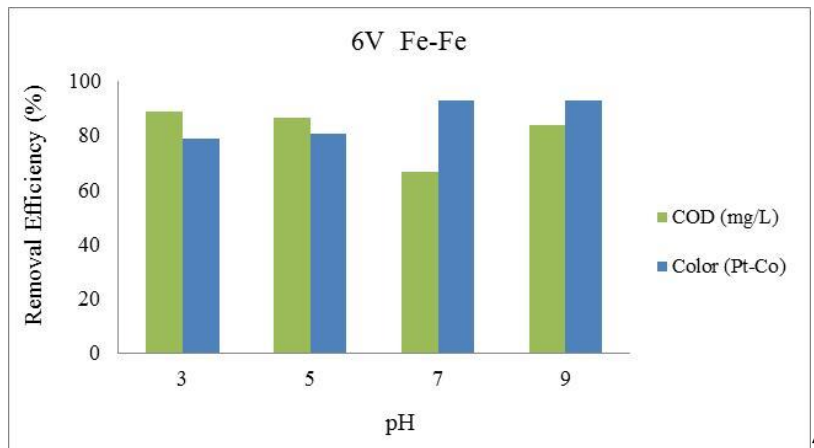
When the Fig. 3 is examined, it is seen that COD removal efficiency at the 8V ( $1.43 \text{ mA/cm}^2$  for Fe/Al) and the end of 3 h was at the level of 71% by using the Fe/Al electrode couple and at the level of 70% by using the Al/Fe electrode couple ( $1.73 \text{ mA/cm}^2$ ). The maximum color removal was achieved in an Al/Fe electrode couple with efficiency at the rate of 87% at 8V. Since these COD removal efficiencies meet the limit values specified in the regulation for discharge of wastewater, the selected 8V potential is proper. The color removal at the rate of 100% and COD removal at the rate of 90% were achieved in the study of Verma (2017) [2] on Fe/Al electrode couple in synthetic textile wastewater that was conducted at 12 V under optimized conditions (pH 8, reaction time 80 min, inter-electrode distance 3 cm). The color removal at the rate of 98% and COD removal at the rate of 87% was obtained in the study of Ghanbari *et al.*, (2014) [30] on textile wastewater by using Fe and Al anode and under optimum operating conditions (pH 7, current 300 mA, distance between two electrodes 3 cm and reaction time 40 min).

### 3.2. The effect of pH on Electrocoagulation of Real Textile Wastewater

The pH in wastewater treatment by electrocoagulation plays a very important role in determining the effectiveness of the treatment. Because it manages the hydrolysed metal forms that are produced in the reactive environment and affects the current mechanisms of the EC [31]. Additionally, pH affects the conductivity and zeta potential of the solution and electrode dissolution [32].

However, since the pH of the treated water changes during the EC process between the pH of the solution and the electrocoagulation activity, it is difficult to establish relationship clearly, so that it is usually referred to the pH of the initial solution [33-34].

The workings with Fe-Fe combination which is optimum electrode couple at 6V potential and having 3, 5, 7 and 9 pH values were done in order to determine the effect of pH on COD and color removal in the study. The aim is to achieve the color and COD removal efficiency obtained at 8 V in the same electrode couple by changing pH. Thus, cost can be reduced by decreasing energy usage. Furthermore, since the pH level of the wastewater is between the values indicated in the discharge criteria after the EC, neutralization will not be needed. The effect of pH on COD and color removal are shown in Figure 4.

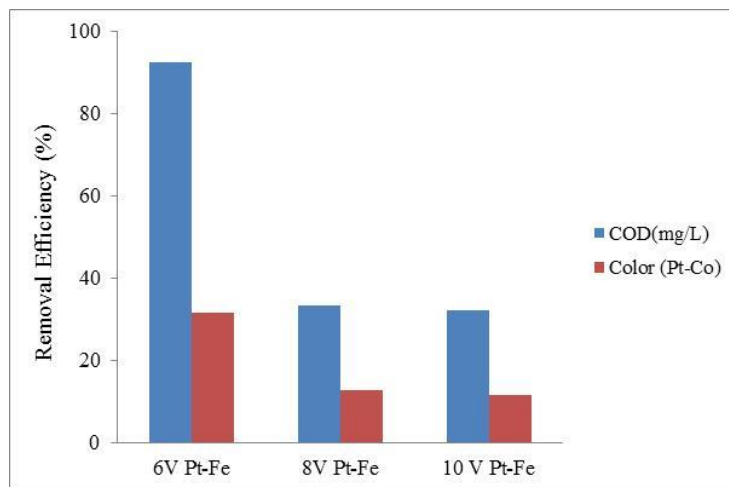


**Figure 4.** Effect of pH on COD and color removal at 6V with Fe/Fe electrode couple

Accordingly, as a result of the EC experiments carried out at the 6V potential with Fe-Fe electrode couple, the COD concentration decreased from 1040 mg/L to 115 mg/L with removal efficiency in the ratio of 89% at pH 3. This value is below the discharge limit value specified in the regulation for COD. The maximum color removal is the same at pH 7 and pH 9, and is at the rate of 93%. The color concentration decreased from 395 Pt-Co to 28 Pt-Co. When the iron Eh-pH diagram is examined, it is seen that it is in the form of  $Fe^{2+}$  and  $Fe(OH)_3$  at pH 3. It is in the form of  $Fe$ ,  $Fe(OH)_2$  and  $Fe(OH)_3$  at pH values higher than 6.5. When compared with the studies in the literature, COD and color removal efficiencies were found to be quite high when studying at 6V in the real textile wastewater only with pH change without chemical addition. Merzouk et al. (2010) achieved 79.7% COD removal and 93% color removal at pH 7.6 in the study [13].

### 3.3. Performance of Electrooxidation Process on Treatment of Real Textile Wastewater

The electrode that plays an active role in the electrooxidation process is anode, and oxidation of the organic contaminants such as  $O_2$  and  $H_2$  gases and dye produced by insoluble electrode is provided as a main principle. The Pt/Fe which was not previously encountered in the literature was used as the electrode couple in the experiments performed in the context of the electrooxidation (EO). The EO experiments were carried out at 3 different potentials and natural pH. The effect of EO experiments on COD and color removal are shown in Figure 5.



**Figure 5.** Effect of electrooxidation experiments on COD and color removal at pH 6.96



As can be seen in the figure, the maximum COD and color removal at 6V (2.5 mA/cm<sup>2</sup>) was realized as 93% and 32%, respectively. As potential increased, color and COD removal from textile wastewater decreased.

Direct anodic oxidation or electrolysis occurs on the anode in the EO process and charge transfer reactions occur between the anode surface and organic contaminants. This mechanism only has the ability to oxidize some organic contaminants at certain potentials. Hence, the fact that the maximum removal in the study realized at 6V and that the removal decreased as the potential increased, are related to this. The more O<sub>2</sub> gas came out along with the increase in potential on the surface of the Pt electrode, and the oxygen evolution reaction (OER) potential became more negative. Thus, the reduction potential of organic contaminant in the wastewater (dye) was exceeded [35-36].

In addition, when direct electrolyse is applied when the potentials below the reaction potential of water oxidation, the electrodes are susceptible to surface poisoning and inhibition of the EO process [20]. Therefore, the removal efficiency may have fallen.

### 3.4. Cost Analysis

Cost analysis was performed to determine whether electrocoagulation and electrooxidation were economical. Consumption of energy during the electrocoagulation and electro-oxidation play an important role which affect the economic feasibility of the process [2].

For the EC process, the most important costs of operation are energy and electrode consumption while in the electrooxidation process, only energy consumption is important because there is no electrode consumption. The energy and electrode consumption was calculated through the following equations suggested by Şengil and Özacar [37].

$$\text{Energy consumption (kWh/kg of removed color, COD)} = \frac{I \times \text{Voltage} \times 1000}{60(Co - Ct) \times v} \quad (1)$$

$$\text{Electrode consumption (kg of electrode/kg of removed color, COD)} = \frac{M \times I \times t}{n \times F \times v \times (Co - Ct)} \quad (2)$$

where I is the applied current (A), t is the electrolysis time (sec), v is wastewater volume (L) and Co is the initial concentration of the color/COD and Ct is the concentration at time t. M is the molar mass of iron/aluminium (g/Mol), n is the number of moles of electron involved in the reaction (Fe =2, Al =3), F is the Faraday constant (F = 96,487 C/Mol). [2, 37]. The energy consumed for COD and color removal for each couple of electrode is shown in Table 3.

**Table 3.** Operating cost of electrocoagulation and electrooxidation at the optimized conditions

Electrode couple	Energy consumption (kWh/kg) for removed COD	Electrode consumption (kWh/kg) for removed COD	Operating cost (kWh/kg) for removed COD
Fe/Fe	12,68	2,75E-05	1,27E+01
Al/Al	28,34	1,59E-05	2,83E+01
Al/Fe	26,18	1,83E-05	2,62E+01
Fe/Al	28,36	6,17E-05	2,84E+01
Pt/Fe	1,29	0	1,29
Electrode couple	Energy consumption (kWh/kg) for removed color	Electrode consumption (kWh/kg) for removed color	Operating cost (kWh/kg) for removed color
Fe/Fe	32,00	0,00	32,00
Al/Al	82,18	0,00	82,18
Al/Fe	60,19	0,00	60,19
Fe/Al	71,16	0,00	71,16
Pt/Fe	92,30	0,00	92,30

Considering the energy related experimental conditions in which the maximum COD removal (93%) was gained (0.05 A, 6 V and 60 min), 1.29 kWh electrical energy is required to remove COD of the studied wastewater by 93% at EO. These value can be accepted to be the economical for EO treatment processes.

The costs were varying between 12.7–28.4 kWh/kg COD<sub>removed</sub>, 32-71 kWh/kg dye<sub>removed</sub> for EC process. It is clearly seen that EC process with Fe/Fe electrodes consumed less operating cost. In both treatment processes, more energy was consumed compared to COD removal for color removal. Similar findings related to operating cost during electrochemical wastewater treatment were also observed by Körbahti and Tanyolaç (2009) [3] and Yüksel et al. (2011) [24].

#### 4. CONCLUSIONS

As a result of the study, it was reached to conclusion that EC and EO processes which are the electrochemical treatment methods can be proposed in the treatment of textile wastewaters containing dye and complex chemicals. In this study, the most important operating parameters were come out as electrode type and combination, pH, reaction time and potential, and these parameters affect the efficiency of electrochemical removal. After EC experiments having been done with different electrode couples at the natural pH of the wastewater, the 88% COD and 92% color removal were achieved at 8 V with the Fe-Fe electrode couple. The maximum COD and color removal at 10 V were realized as 88% and 92%, respectively with the Al-Al combination which is the other electrode couple. The maximum COD removal was 93% at 6V in the EO process in which using Pt/Fe electrode couple was used. As a result, it was determined that the electrode combinations used in the study are suitable in order to produce dischargeable or recyclable quality wastewater from the textile wastewater.

Energy consumptions affecting the operating cost were found to be 1.29 kWh/kg for Pt/Fe electrode couple (EO). At the end of the study, it was seen that electrooxidation was more feasible in terms of cost for COD removal. At the optimised conditions for color removal, minimum operating cost was determined to be 32.0 kWh/kg of dye removed at EC. In view of economic performances, EO which used Pt/Fe electrodes was found superior to EC with Fe/Fe electrodes and it can be recommended for the treatment of the textile wastewater.

#### REFERENCES

- [1] Orts F, Del Río AI, Molina J, Bonastre J, Cases F. Electrochemical treatment of real textile wastewater: Trichromy Procion HEXL®. J Electroanal Chem 2018; 808: 387–394.
- [2] Verma AK. Treatment of textile wastewaters by electrocoagulation employing Fe-Al composite electrode. J Water Process Eng 2017; 20: 168–172.
- [3] Körbahti BK, Tanyolac A. Continuous electrochemical treatment of simulated industrial textile wastewater from industrial components in a tubular reactor. J Hazard Mater 2009; 170: 771–778.
- [4] İlhan F, Kurt U, Apaydın Ö, Arslankaya Ö, Gönüllü MT. Elektrokimyasal Arıtım ve Uygulamaları: Katı Atık Sızıntı Suyu Çalışması. 2007, AB Sürecinde Türkiye’de Katı Atık Yönetimi ve Çevre Sorunları Sempozyumu.
- [5] Vardar B. Treatment of Textile Industries Reactive Dye Baths by Electrochemical Methods. MSc, İstanbul University, İstanbul, Turkey. 2006.
- [6] Welham A. The Theory of Dyeing (and the Secret of Life). J Soc Dyers Colour 2000; 116: 140-143.

- [7] Won S, Han M, Yun Y. Different Binding Mechanisms in Biosorption of Reactive Dyes According to Their Reactivity. *Water Res* 2008; 42: 4847-4855.
- [8] Bayar S, Boncukçuoğlu R, Fil B, Yılmaz A. Elektrokoagülasyon Yöntemi Kullanılarak Direct Red 23 Boyarmaddesinin Gideriminin İncelenmesi. *Iğdır Üniversitesi Fen Bilimleri Enstitüsü Dergisi*, 2012; 2: 21-28.
- [9] Mollah M, Schennach R, Parga J, Cocke D. Electrocoagulation (EC) Science and Applications. *J Hazard Mater* 2001; B84: 29-41.
- [10] Song P, Yang Z, Zeng G, Yang X, Xu H, Wang L, Xu R, Xiong W, Ahmad K. Electrocoagulation treatment of arsenic in wastewaters: A comprehensive review. *Chem Eng J* 2017; 317: 707–725.
- [11] Kobyas M, Demirbas E, Can OT, Bayramoglu, M. Treatment of levafix orange textile dye solution by electrocoagulation. *J Hazard Mater* 2006; 132: 183-188.
- [12] Sahu O, Mazumdar B, Chaudhari PK. Treatment of wastewater by electrocoagulation: a review. *Environ Sci Pollut* 2014; R.21: 2397–2413.
- [13] Merzouk B, Gourich B, Sekki A, Madani K, Vial C, Barkaoui M. Studies on the decolorization of textile dye wastewater by continuous electrocoagulation process. *Chem Eng J* 2009; 149: 207–214.
- [14] Pourbaix M. *Atlas of Electrochemical Equilibria in Aqueous Solution*, second ed. Pergamon Press, 1974 Houston.
- [15] Ozay Y, Ünşar EK, Işık Z, Yılmaz F, Dizge N, Perendeci NA, Mazmanci MA, Yalvac M. Optimization of electrocoagulation process and combination of anaerobic digestion for the treatment of pistachio processing wastewater. *J Clean Product* 2018; 196: 42-50.
- [16] Cotillas S, Llanos J, Cañizares P, Mateo S, Rodrigo MA. Optimization of an integrated electrodisinfection/electrocoagulation process with Al bipolar electrodes for urban wastewater reclamation. *Water Res* 2013; 47: 1741–1750.
- [17] Nidheesh PV, Singh ATS. Arsenic removal by electrocoagulation process: Recent trends and removal mechanism. *Chemosphere*, 2017; 181: 418-432.
- [18] Chen G. *Electrochemical Technologies in Wastewater Treatment*, *Sep Purif Technol* 2004; 38: 11-41.
- [19] Brillas E, Martinez-Huitle CA. Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review. *Appl Catal B*, 2015; 166–167.
- [20] Garcia-Segura S, Ocon JD, Chong MN. Electrochemical oxidation remediation of real wastewater effluents -A review. *Process Saf Environ Prot* 2018; 113: 48–67.
- [21] Khandegar V, Saruha AK. Electrocoagulation for the treatment of textile industry effluent - A review. *J Environ Manage* 2013; 128: 949-963.
- [22] UNESCO, 2012. *The United Nations World Water Development Report 4. Volume 1, Managing Water Report under Uncertainty and Risk*.

- [23] Merzouk B, Madani K, Sekki A. Using Electrocoagulation–Electroflotation Technology To Treat Synthetic Solution and Textile Wastewater, Two Case Studies. *Desalination* 2010; 250: 573-577.
- [24] Yüksel E, Eyvaz M, Gurbulak E. Electrochemical treatment of colour index Reactive Orange 84 and textile wastewater by using stainless steel and iron electrodes. *Environ Progr Sustain Energ* 2013; 32: 60-68.
- [25] Naje AS, Chelliapan S, Zakaria Z, Abbas SA. Electrocoagulation Using A Rotated Anode: A Novel Reactor Design For Textile Wastewater Treatment. *J Environ Manage*, 2016; 176, 34-44.
- [26] Aquino JM, Rocha-Filho RC, Ruotolo LAM, Bocchi N, Biaggio SR. Electrochemical degradation of a real textile wastewater using -PbO<sub>2</sub> and DSA® anodes. *Chem Eng J* 2014; 251: 138–145.
- [27] Ling Y, Hu J, Qian Z, Zhu L, Chen X. Continuous treatment of biologically treated textile effluent using a multi-cell electrochemical reactor. *Chem Eng J* 2016; 286: 571–577.
- [28] Zou J, Peng X, Li M, Xiong Y, Wang B, Dong F. Electrochemical oxidation of COD from real textile wastewaters: Kinetic study and energy consumption. *Chemosphere* 2017; 171: 332-338.
- [29] Linares-Hernández I, Barrera-Díaz C, Roa-Morales G, Bilyeu B, Urena-Nunez F. Influence of the anodic material on electrocoagulation performance. *Chem Eng J* 2009; 148, 97–105.
- [30] Ghanbari F, Moradi M, Eslami A, Emamjomeh MM. Electrocoagulation/Flotation of textile wastewater with simultaneous application of aluminum and iron as anode. *Environ Process* 2014; 1, 447–457.
- [31] Malakootian M, Mansoorian HJ, Moosazadeh M. Performance evaluation of electrocoagulation process using iron-rod electrodes for removing hardness from drinking water. *Desalination* 2010; 255, 67–71.
- [32] Moussa DT, El-Naas MH, Nasser M, Al-Marri MJ. A comprehensive review of electrocoagulation for water treatment: Potentials and challenges, *J Environ Manage* 2017; 186, 24-41.
- [33] Mansoorian HJ, Mahvi AH, Jafari AJ. Removal of lead and zinc from battery industry wastewater using electrocoagulation process: influence of direct and alternating current by using iron and stainless steel rod electrodes. *Sep Purif Technol* 2014; 135, 165-175.
- [34] Gatsios E, Hahladakis JN, Gidaracos E. Optimization of electrocoagulation (EC) process for the purification of a real industrial wastewater from toxic metals. *J Environ Manag* 2015; 154, 117-127.
- [35] Panizza M, Cerisola G. Direct and mediated anodic oxidation of organic pollutants. *Chem Rev* 2009; 109, 6541–6569.
- [36] Garcia-Segura S, Brillas E. Mineralization of the recalcitrant oxalic and oxamic acids by electrochemical advanced oxidation processes using a boron-doped diamond anode. *Water Res* 2011; 45, 2975–2984.
- [37] Şengil İA, Özacar M. The decolorization of CI Reactive Black 5 in a aqueous solution by electrocoagulation using sacrificial iron electrodes. *J Hazard Mater* 2009; 161, 1369–1376.