



The effects of alternating current electrocoagulation on dye removal from aqueous solutions

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ABSTRACT

The main goal of this study is to investigate the effects of alternating current (AC) on dye removal from aqueous solutions by electrocoagulation (EC). An EC system with parallel-connected aluminium electrodes was operated in batch mode. Two different aqueous dye solutions were used: one was obtained from Dianix Yellow CC (DY) and the other was obtained using Procion Yellow (PY). The experiments employing direct current (DC) were carried out using a DC power supply. The AC experiments were conducted using rectangular wave, which is produced with an adjustable time relay connected to the output of DC power supply. This current is called alternating pulse current (APC) in order to refer AC system in this study. Total organic carbon (TOC) and dye removal efficiencies were measured to assess treatment efficiency. Operating cost was calculated for both power supply systems and alternating pulse current was found superior to direct current for the treatment of reactive and disperse dyes used in this study.

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1. Introduction

Dyeing effluents produced in large amounts from textile industry have high pH and temperature, detergents, oil, suspended and dissolved solids, toxic and non-biodegradable matter, strong color and alkalinity as well as high COD and TOC values [1–3]. These effluents can exhibit serious environmental problems and public health concerns if improperly disposed. These highly colored components, when discharged with wastewater into the water bodies, stop the reoxygenation capacity of the receiving water and cut-off sunlight, thereby upset biological activity in aquatic life [4].

The main problem that environmental engineers have to deal with is the elimination of the wastewater's color, which is due to the remaining dyes. However, color removal has been a great challenge over the last decades, and up to now there is no single and economically attractive treatment that can effectively decolorize dyes and new technologies for wastewater decolorization are especially needed [5–8].

Various physiochemical treatment methods, such as chemical coagulation and adsorption, biological methods, such as anaerobic reduction and aerobic oxidation, advanced oxidation processes, such as ozonation and UV/H₂O₂ have been employed so far in order to effectively purify dyeing effluents; decolorization being among the main targets to achieve. Chemical coagulation/flocculation techniques, usually combined with activated-sludge treatment,

have been among the most common processing methods mainly due to their application simplicity. However, the above methods require high amounts of chemicals and also generate large amounts of sludge leading to the elevation of the total treatment cost [9–11]. Another physico-chemical method, adsorption is quite effective for decolorization, but safe and inexpensive disposal of the settled sludge is becoming increasingly difficult [12–15]. Advanced oxidation processes (ozonation, UV/H₂O₂) are based on the generation of hydroxyl radicals, which are highly reactive oxidants. They are environment-friendly techniques since they produce no solid wastes. However, they are not cost-effective due to the high consumption of both energy and raw material (e.g. hydrogen peroxide) [16–18]. Directly biological treatment of dye wastewater is considered unsatisfactory because of the low efficiency and low reaction rate of the treatment. Consequently, dye wastewater is usually treated using physical and chemical methods before biodegradation [19]. Anaerobic processes have been shown to be more suitable for azo bond cleavage via bio-reductive processes [20]. Unfortunately, anaerobic processes often are not feasible for dyeing wastewater, owing to the extremely high hydraulic volume typical of textile dye wastewater [21]. So, there is a need to find alternative methods that are effective in removing dyes from large volumes of effluents and are low in cost [22].

In recent years, electrocoagulation of synthetic or real wastewater containing textile dyestuffs has been investigated by some researchers [23–26] and good removal efficiencies of COD, color, turbidity and dissolved solids at varying operating conditions have been revealed. EC process provides a simple, reliable and cost-effective method for the treatment of wastewater without and need

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for additional chemicals, and thus the secondary pollution. It also reduces the amount of sludge, which needs to be disposed. EC technique uses a direct current source between metal electrodes immersed in polluted water [27].

In this method, soluble metal electrodes form metal hydroxides when subjected to a suitable current. The metal hydroxides act as coagulants and lead to the removal of various contaminants [26].

Usually, direct current is used in EC systems. In this case, an impermeable oxide layer may form on the cathode material as well as corrosion formation on the anode material due to oxidation. This prevents the effective current transfer between the anode and cathode, so the performance of EC reactor declines. These disadvantages of direct current have been diminished by the addition of parallel plate sacrificial electrodes in the EC unit configuration. However, many have preferred the use of alternating current in EC unit [28].

It is believed that the cyclic energization between the anode–cathode in AC system delays the cathode passivation and anode deterioration that are experienced in DCE system, and thus, ensure reasonable electrode life [29].

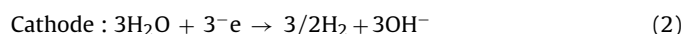
Although many researches on treatment of synthetic or real wastewaters with EC are available, very few researches have been carried out on the economical applicability of EC [27,30,31]. Therefore, in this study, the effects of power supply type on dye removal from aqueous solutions by electrocoagulation (EC) were investigated both technically and economically.

2. A brief description of EC process

EC consists of in situ generating M^{3+} ions in electrolyte solution electrochemically using sacrificial anode materials, according to



where M is usually iron or aluminium. Hydrogen gas is released at the cathode which is made of sacrificial material as the anode or inert one, such as stainless steel.



M^{3+} and OH^- ions generated by electrode reactions (1) and (2) react to form various monomeric species, which finally transform into $M(\text{OH})_{3(s)}$ according to complex precipitation kinetics.



3. Materials and methods

3.1. Dye solutions

Disperse dye, Dianix Yellow CC, and reactive dye, Procion Yellow H-E6G, were commercially obtained from Dystar. Demineralized water was used to prepare the 1000 ppm of dye solutions in all runs. The properties of dye were given in Table 1.

3.2. EC reactor

The schematic figure of electrochemical reactor is shown in Fig. 1. The thermostated unit was made of Plexiglas with the dimensions of 130 mm × 130 mm × 125 mm. The unit consists of two pair of electrodes made of plate aluminium with effective area of 137 cm² and the distance between the electrodes is 20 mm. Electrodes were connected to a digital DC power supply (Maksimel LPS 991, 0–600 VA) in monopolar-parallel mode.

3.3. Description of alternating (pulse) current electrocoagulation

An adjustable time relay plugged into the DC power supply was employed to obtain alternating pulse current. It represents alter-

Table 1
Characteristics of dye solutions.

Parameter	Value	
C.I. number	Disperse Yellow 241	Reactive Yellow 135
Molecular formula	C ₁₅ H ₁₀ I ₂ N ₄ O ₂	C ₄₆ H ₃₆ C ₁₆ N ₁₂ O ₂₀ S ₆
CAS number	83249-52-9	77907-38-1
Water solubility at 293 K (g l ⁻¹)	Diffusible	45
Melting point (°C)	>200	>300
Combustion temperature (°C)	>500	–
Bulk density (kg m ⁻³)	580–630	400–800
pH	6.0–6.5 (100 g/l water)	8.0–9.0 (10 g/l water)
Acute oral toxicity LD ₅₀ (mg kg ⁻¹)	>5000	>5000
Biological degradation (%)	>70	<10
Fish toxicity LC ₅₀ (mg l ⁻¹)	>100	>100
Bacterial toxicity IC ₅₀ (mg l ⁻¹)	>100	>100
Chemical oxygen demand (CSB) (mg g ⁻¹)	1.250	1.100
λ _{max} (nm)	419	421

nating current in our study. According to EC unit with time relay system, turn on and turn off modes switch to positive pole to negative pole or reversion to it. For example, when the time relay is turned on in an EC reactor including two electrodes namely 1 (anode) and 2 (cathode), electrode 1 is then converted to cathode while electrode 2 is becoming anode. When the time relay is turned off, only DC system is in circuit, electrode 1 becomes anode this time. Current wave in real alternating current is shown in Fig. 2a; alternating pulse current's wave used in this study is shown in Fig. 2b, as well. In Fig. 2b, T is the period (turn on time + turn off time).

3.4. Experimental procedure

All experiments were done at constant temperature of 20 °C. In each run, 1500 ml of dye solution was placed into the reactor. The conductivity, pH and current density were adjusted to the desired value before the process. After each run, electrode surfaces were removed by dipping for 1 min in a solution prepared by mixing 100 cm³ HCl solution (36.5%) and 200 cm³ of hexamethylenetetramine aqueous solution (2.80%) and washed thoroughly with demineralized water to remove any solid residues on the surfaces, dried and re-weighed.

Total operation cost includes direct cost items, such as electricity, electrodes and chemicals, as well as indirect cost parameters, such as labor, maintenance and depreciation of the major equipments including power supply and EC tank. Economic data used for the evaluation of the total operating costs dye given for the second quarter 2008, Turkey, in Table 2.

3.5. Analytical techniques

To measure dye concentration an UV spectrophotometer (HACH Co., model DR5000) was employed. The absorbance peak decreasing was directly proportional to the reduction of the dye concentration. The characteristic peak of pure DY was observed at

Table 2
Economic factors used in the operation cost calculations.

Item with warranty period	Cost
Power supply and installing (\$) (5 years)	9000
EC tank and installing (\$) (10 years)	500
Maintenance and depreciation (\$ m ⁻³)	0.005
Electricity (\$ (kWh) ⁻¹)	0.17
Labor costs (\$ m ⁻³)	0.1
Aluminium electrode (\$ kg ⁻¹)	2.95
Chemicals (acid, salt, etc.) (\$ m ⁻³)	0.04

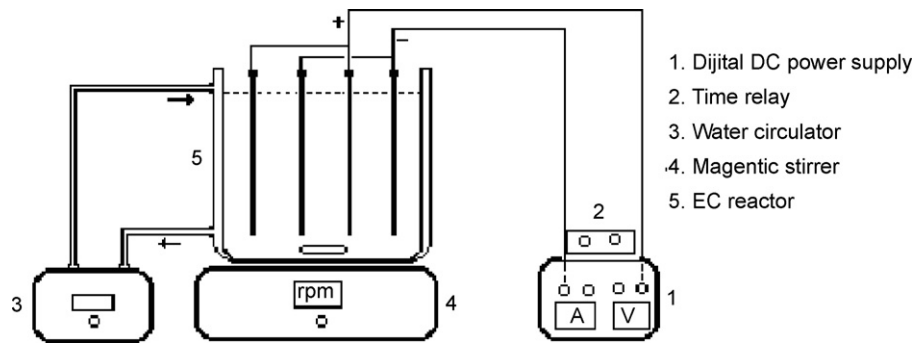


Fig. 1. Experimental setup.

419 nm and of RY was 421 nm. Total organic carbon was analyzed using a TOC analyzer (HACH IL 550 TOC-TN model). The initial pH was adjusted to a desired value using NaOH or H₂SO₄.

The dye removal efficiency R_{dye} is calculated as follows:

$$R_{\text{dye}} = \frac{C_{\text{dye}} - C_{\text{dye}}}{C_{\text{dye}}} \times 100 \quad (4)$$

Likewise, TOC removal efficiency R_{TOC} is

$$R_{\text{TOC}} = \frac{C_{\text{TOC}} - C_{\text{TOC}}}{C_{\text{TOC}}} \times 100 \quad (5)$$

4. Results and discussion

4.1. Determining the optimum experiment parameters to comparison DC & APC systems

The EC process is highly dependent on the initial pH of the solution [26]. Therefore, five pH values (3.5, 4.5, 5.5, 6.5 and 7.5) were selected to investigate the optimal pH at which maximum removal efficiencies and minimum electrode and energy consumptions were observed. The effects of initial pH of dye solution on EC have been investigated at constant current density of 35 A m⁻², 15 min of operating time and 1000 mg l⁻¹ initial dye concentration. TOC removal and decolorization efficiencies are featured in Fig. 3a. Both TOC and dye removal efficiencies showed similar trends. TOC and dye removal efficiencies are pH dependent parameters as aluminium hydroxide flocs which destabilize and aggregate the suspended particles occur at optimum pH. The highest TOC and dye removal efficiencies were obtained at pH 4.5 for both dyes. Electrode consumption in respect of initial pH was given in Fig. 3b. At pH 4.5, the least electrode consumption was obtained depends on the

highest removal efficiencies at this pH. Likewise, the lowest energy consumptions were obtained (Fig. 3c). It is shown from the figures that pH 4.5 is suitable for the treatment of dyes in this study.

Current density is an important factor influencing the treatment performance of EC process [32]. All the experiments were applied under pH 4.5, 15 min of operating time for DY; 20 min for RY and at 1000 mg l⁻¹ dye concentration to examine current density effects. Fig. 3d depicts the current density effects on dye and TOC removal. Both dye and TOC removal efficiencies increased as the current density increased up from 35 to 105 A m⁻², then, it decreased with increasing of current density from 105 to 150 A m⁻². Increment in current density raised the formation of hydroxide flocs and increased the outputs of coagulation and so of removal. But after a certain value of current density, cathode passivation occurred and dissolution of anode material reduced and also floc formation and removal efficiencies diminished. Electrode consumption in terms of current density could be seen in Fig. 3e. While slight changes occurred in electrode consumption, respectable variation was obtained in energy consumption at given current densities and similar trends could be observed therein. Increasing of current density causes an increase in energy consumption. While this increment was very explicit for TOC removal, these consumptions were at close values in dye removal (Fig. 3f). Depending on dye and TOC removal efficiencies, 105 A m⁻² was chosen as the appropriate current density value.

Finally, some experiments were performed to specify optimum anode–cathode polarization period. Four various periods, such as 3 min (180 Hz⁻¹), 5 min (300 Hz⁻¹), 10 min (600 Hz⁻¹), 15 min (900 Hz⁻¹) were tried in APC system. Results are depicted in Fig. 4. Similar removal efficiencies were laid out for both DY and RY dyes. A little decrement occurred only at 180 Hz⁻¹. That is to say, these polarization periods could said to not affect the removal efficien-

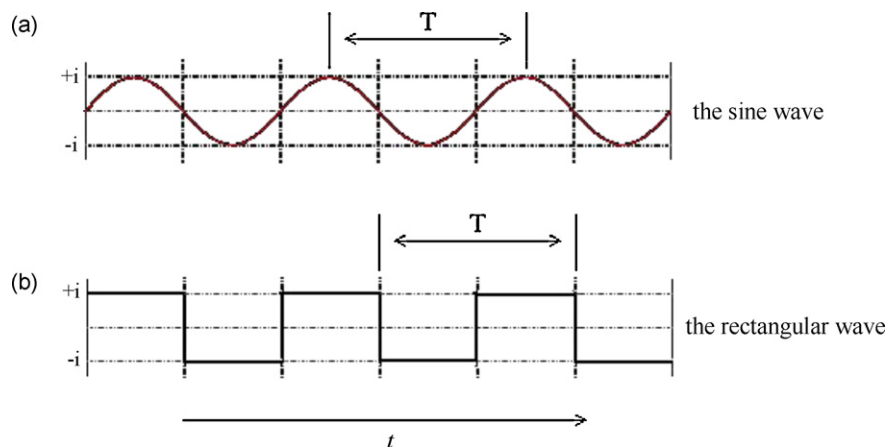


Fig. 2. (a) Current wave of alternating current and (b) Current wave of alternating pulse current.

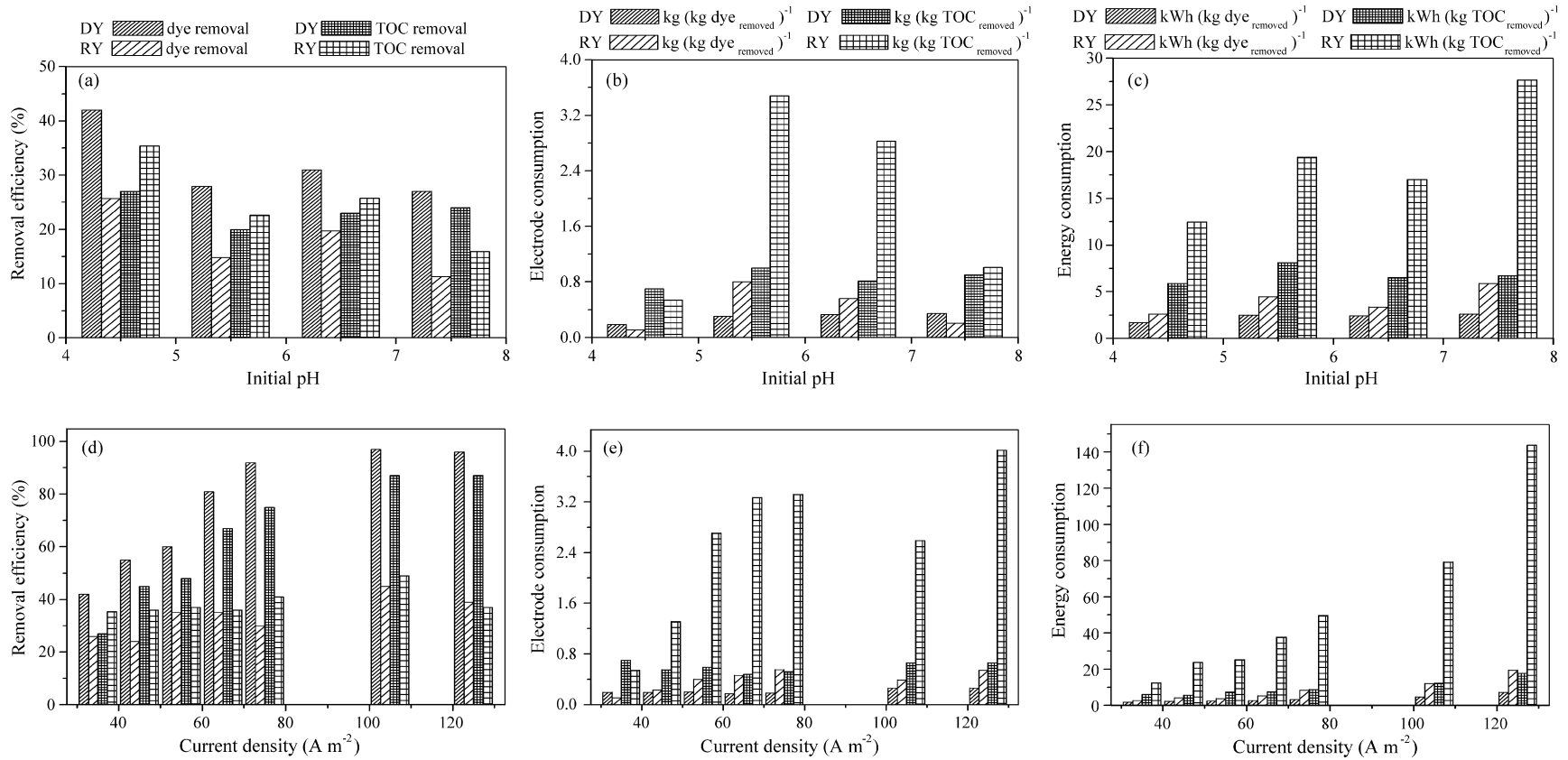


Fig. 3. Effects of initial pH: (a) on dye and TOC removal, (b) on electrode consumption, and (c) on energy consumption. Effects of current density: (d) on dye and TOC removal, (e) on electrode consumption and (f) on energy consumption.

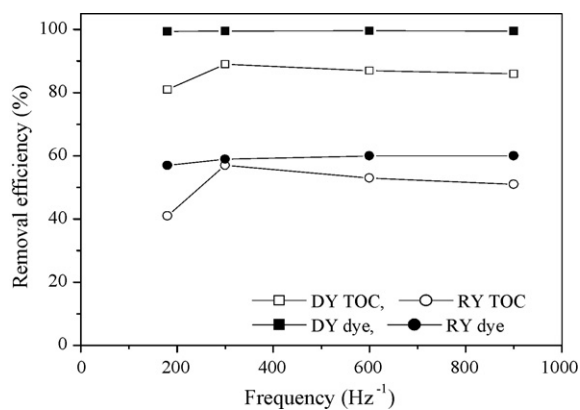


Fig. 4. Effects of frequency on dye and TOC removal.

cies. In conclusion, 300Hz^{-1} was selected as APC frequency in following experiments.

Dye removal efficiencies are presented in Fig. 5. DY removal efficiency is generally higher than that of RY in both power supply systems. Maximum RY removal with DC was obtained as 59% at 20 min of operation time and it dropped to 25% at 100 min. Seeing that the other parameters except operation time, e.g. pH and current density, were optimized before, the decrease of removal efficiency after reaching its maximum value at any specific time may be due to the cathode passivation. When DY removal is taken into account, a major difference is stand out between DC and APC at 5 min of operating time. This period of time in question is inadequate for reactions which are required for the formation of metal hydroxides. So, dye removal efficiency just reached to 11%. As for AC power supply, 69.7% removal efficiency was obtained. At this operation time, positive and negative electrodes changed their charge inversely for once with time relay by 300Hz^{-1} . The anode cathode polarization may have promoted the metal hydroxide formation reactions for DY removal. Thus a major difference came out between two power systems for DY removal. After 10 min, at which 89% and 90.3% dye removal percentages were attained for DC and APC, respectively, the given percentage of removal efficiencies stays nearly constant. For APC system, approximately 99.5–99.9% removal efficiencies were obtained at all operation times consequent to 10 min. However, it decreases right after 35 min, and then it sharply drops after 70 min and decreases to 42% when 100 min of operation time is applied for DC system. It can be said that, increasing the operation time increases the accumulation of dye residue causing passivation

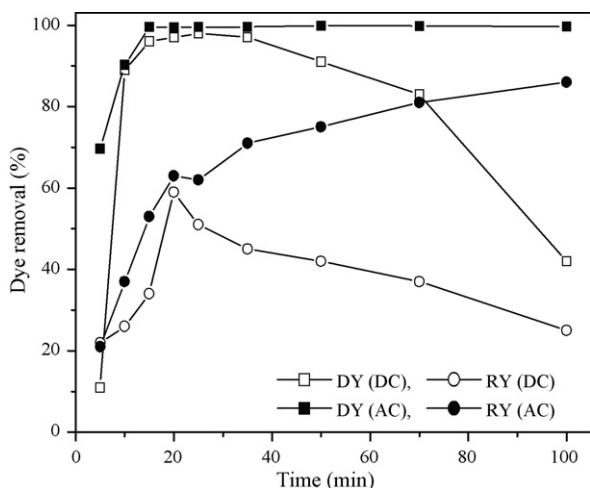


Fig. 5. Effects of power supply type on dye removal in view of operation time.

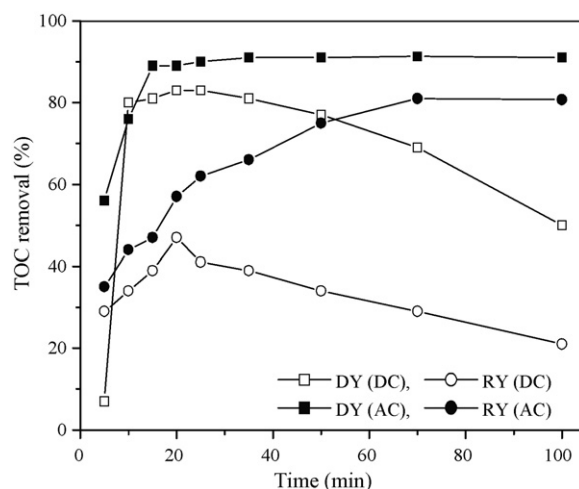


Fig. 6. Effects of power supply type on TOC removal in view of operation time.

on cathode material. Thus, anodic dissolution diminishes and then metal hydroxide formation diminishes too. As a result, dye removal efficiency decreases.

TOC removal from aqueous solutions in terms of power supply type delineated in Fig. 6. It is shown that TOC removal variations appear to be parallel with dye removal efficiencies presented in Fig. 5. Maximum TOC removal (47%) was reached for RY at 20 min of operation time while the DC power supply is being used, then it decreases to 21% with increasing of time to 100 min. However, TOC removal continuously increases with the increase of electrolysis time in APC regime. It reaches to 81% of removal efficiency at 70 and 100 min of electrolysis time. This increment with APC system may be due to no passivation which may occur in DC regime. Additionally, APC is superior to DC in TOC removal from aqueous solutions prepared with RY for all operation times. TOC removal from DY solution is similar to dye removal at 5 min of operation time for DC system. In APC regime, TOC removal varies between 56% and 91% from 5 to 100 min. However, it changes from 7% to 83% up to 20 min at which was attained maximum removal efficiency value, removal percentage decreases slightly when the electrolysis time is changed from 25 to 70 min, and then it sharply decreases to 50% at 100 min.

The EC with different power supplies' operating costs for both dyes are given by means of dye and TOC removal in Figs. 7 and 8,

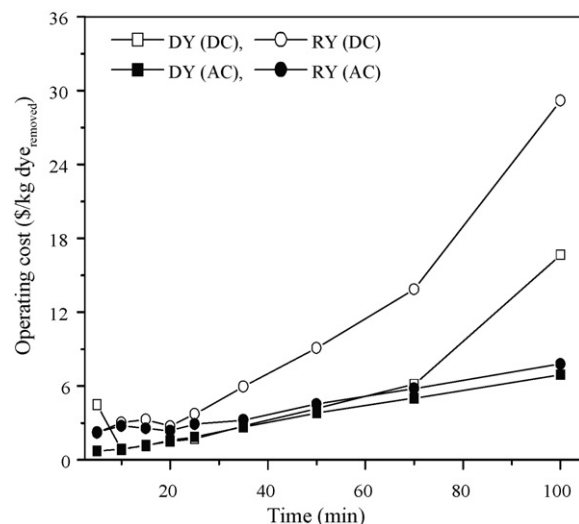


Fig. 7. Effects of power supply type on dye removal cost in view of operation time.

Table 3
Optimum experimental conditions determined before power supply-time experiments.

Dye	DY			RY		
	pH	Current density ($A\ m^{-2}$)	Operating time (min)	pH	Current density ($A\ m^{-2}$)	Operating time (min)
Optimum experimental conditions	4.5	105	15	4.5	105	20
Dye removal (%)	42	97	99	25.7	45	62
TOC removal (%)	27	87	87	35.4	49	53
Electrode consumption ($kg\ (kg\ dye)^{-1}$)	0.19	0.26	0.26	0.11	0.39	0.65
Electrode consumption ($kg\ (kg\ TOC)^{-1}$)	0.7	0.66	0.66	0.54	2.59	5.41
Energy consumption ($kWh\ (kg\ dye)^{-1}$)	1.7	4.74	4.74	2.58	14.99	9.65
Energy consumption ($kWh\ (kg\ TOC)^{-1}$)	5.9	12.16	12.16	12.47	79.22	80.38

Table 4
Technical and economic comparison of DC and APC.

Dye	Procion Yellow		Dianix Yellow	
	DC	APC	DC	APC
Process ^a	DC	APC	DC	APC
Initial pH	4.5	4.5	4.5	4.5
Final pH	10.43	9.85	9.72	9.63
Current density ($A\ m^{-2}$)	105	105	105	105
Electrolysis time ^b (min)	20	20	15	15
TOC removal (%)	49 (29 at 70 min)	57 (81 at 70 min)	81	89
Dye removal (%)	59 (25 at 100 min)	63 (86 at 100 min)	96	99.9
Operating cost ($\$/kg\ TOC_{removed}$) ^c	3.43 (17.7 at 70 min)	2.61 (5.8 at 70 min)	1.43	1.3
Operating cost ($\$/kg\ dye_{removed}$) ^c	1.59 (29.2 at 100 min)	1.51 (7.8 at 100 min)	3.27	2.57

^a All data in table are given according to technical and economic optimum values obtained in DC experiments. Results of APC experiments are also stated in these conditions. However, since TOC and dye removal efficiencies still increase with increasing time in APC system, the operation time exhibiting maximum removal percentages and related operation costs are also given in brackets. On the other hand, the performances of DC system at operation times in question are shown in the table.

^b Two different optimum electrolysis times were attained separately for DY and PY in DC experiments.

^c Economic factors used in operating cost calculations were considered for second quarter of 2008 in Turkey.

respectively. Operation costs of dye and TOC removal from RY dye are higher than those of DY dye in DC system while nearly same operation costs come out for both RY and DY dyes when APC system is employed. Overall results are given in Table 3.

The following results may be drawn when technical and economical results shown in Table 4 are considered:

- The TOC removal performance of APC system is 10% higher than that of DC system and the dye removal is nearly the same at 15 min of operation time when DY dye is considered. Operating cost per kg TOC removed by DC system is 1.1 times as high as the operating cost of APC system while operation cost per kg dye removed in DC regime is 1.3 times as high as the operating cost of APC regime. In short operation times, especially at 5 min, APC system is faster and more economic than DC system.

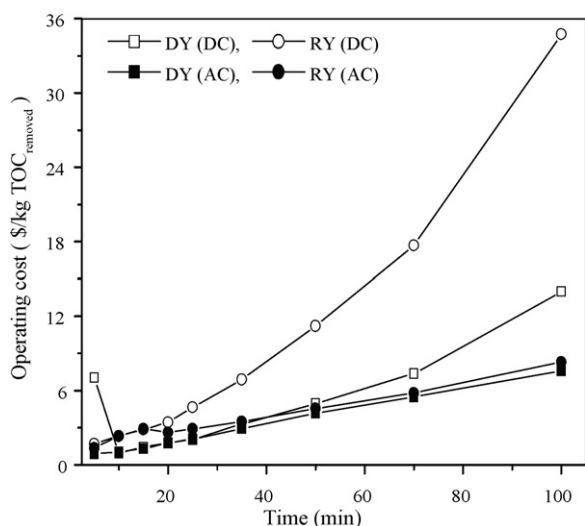


Fig. 8. Effects of power supply type on TOC removal cost in view of operation time.

- When RY dye is taken into account, more removal efficiencies can be reached by increasing of operation time with APC system while it can be just reached to 59% and 49% for dye and TOC removal, respectively at 20 min of operation time with DC system. Removal performances decrease with increasing of electrolysis time. Both power systems exhibit nearly same operating costs in terms of kg dye removed, but operating cost of DC system is 30% higher than that of APC system.

5. Conclusions

In this study, electrocoagulation of two textile dyes from aqueous solutions by different power supplies was carried out. Aluminium electrodes were used as sacrificial electrodes in parallel connection modes. Direct current (DC) was obtained from a DC power supply operated at galvanostatic mode while alternating pulse current (APC) was obtained by a time relay integrated with DC power supply. Dye and TOC removal efficiencies were considered when DC and APC were compared technically. Furthermore, various cost items were used to calculate the total operation cost of both DC and APC systems by means of TOC and dye removal.

In the light of experimental results, following conclusions may be exposed:

- Higher removal efficiencies of TOC and dye can be acquired in shorter operation times by using APC system (e.g. 5 min of operation time). Thus, it can be said that anode–cathode polarization reduces the reaction time which is necessary for metal hydroxides removing TOC and dye from aqueous solutions.
- For RY dye, TOC and dye removal percentages increase until a certain operation time, and then they decrease so long as DC system goes on working. It may be due to the cathode passivation arisen from accumulation of contaminants on the cathode material. Therefore, electrode surfaces are needed to be cleaned and then put into use again. However, removal efficiencies increase in APC system after optimum operation time belongs to DC sys-

tem as well. Thus, APC system can be used to overcome these drawbacks came out from DC system.

- Fill-and-draw periods of reactor could be easily increased for batch EC processes by using time relay to eliminate cathode passivation.

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