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Techno-economical evaluation of electrocoagulation for the textile wastewater using different electrode connections

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Abstract

The bench scale of an electrocoagulation (EC) unit requires a detailed study discerning the effects of continuous variables such as pH, current density and operating time, and type variables such as electrode material and connection mode. This paper presents the results of the treatment of a textile wastewater by EC process. Two electrode materials, aluminum and iron, were connected in three modes namely, monopolar-parallel (MP-P), monopolar-serial (MP-S), and bipolar-serial (BP-S). COD and turbidity removals were selected as performance criteria. For a high COD removal, acidic medium is preferable for both electrode materials. For a high turbidity removal, acidic medium is preferable for aluminum, and neutral medium for iron. High current density is favorable for both removals in the case of iron. In the aluminum case, the current density exhibits a pronounced effect on COD removal, depending strongly on the connection mode, but it has a negligible effect on the turbidity removal. MP-P with iron or MP-S with aluminum electrode are suitable configurations in regard with the overall process performance.

Moreover, process economy is as important as removal efficiencies during the process evaluation task. Various direct and indirect cost items including electrical, sacrificial electrodes, labor, sludge handling, maintenance and depreciation costs have been considered in the calculation of the total cost. The results show that MP-P mode is the most cost-effective for both electrode types. Both electrodes show similar results in reducing COD and turbidity, but iron is preferred as a low cost material. Finally, a comparative study showed that EC was faster and more economic; consumed less material and produced less sludge, and pH of the medium was more stabilized than chemical coagulation (CC) for similar COD and turbidity removal levels. For CC, $FeCl_3$ was the preferable salt in view of its techno-economic performance. On the other hand, iron was the preferred electrode material in EC with MP-P system in experimental conditions such as, 30 A m^{-2} of current density and 15 min of time, the treatment cost was \$ 0.245 m^{-3}. Consequently, the operating cost of CC was 3.2 times as high as the operating cost of EC.

Keywords: Textile wastewater; Electrocoagulation; Electrode connection; Electrode material; Economic analysis; Operating cost

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1. Introduction

In recent years, electrochemical treatment methods such as electro-oxidation and electrocoagulation (EC) have attracted great attention as an eco-friendly and cost-effective process [1–5]. EC involves the in situ generation of coagulants by electrolytic oxidation of an appropriate sacrificial anode (e.g., iron and aluminum) upon application of a direct current. The metal ions generated hydrolyze in the electrocoagulator to produce metal hydroxide ions and neutral $M(OH)_3$. The low solubility of the neutral $M(OH)_3$, mainly at pH values in the range of 6.0–7.0, promotes the generation of sweep flocs inside the treated waste and the removal of the pollutants by their enmeshment into these flocs. EC process removes pollutants principally by coagulation, adsorption, precipitation and flotation [6,7]. EC has been successfully used decades to treat the wastewaters of olive mill [8,9], phosphate [10,11], surfactant [12], food process [13], semiconductor [14], chemical mechanical polishing [15,16], restaurant [17], metal plating [18], tannery [19], chromium(VI) [20], potato chips manufacturing [21], dairy [22], poultry slaughterhouse [23,24], pulp and paper mill [25].

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Textile wastewaters are one of the most polluted wastewaters due to their characteristics such as high COD concentration, strong color, high pH and temperature and low biodegradability [26]. Since diversity of textile products increases, different dyestuffs with highly varying chemical characteristics are used in this sector, which complicates further the treatment of textile wastewaters [26]. Several conventional methods have been applied for this purpose such as adsorption, biological treatment, oxidation, coagulation, and flocculation [26]. Although these methods have been widely applied, they have some disadvantages. For example, adsorbents are usually difficult to regenerate [26]. Chemical coagulation causes extra pollution due to the undesired reactions in treated water and produces large amounts of sludge [20]. Biological methods are not suitable for most textile wastewaters due to the harmful effects of some commercial dyes on the organisms used in the process. Furthermore, these conventional methods are also usually expensive and treatment efficiency is inadequate because of the large variability of the composition of textile wastewaters [27]. Various studies, including those recently conducted in our laboratory have shown the high performance of EC for the treatment of textile dye or wastewater with good removals of COD, turbidity and dissolved solids [28–38].

Variables which are commonly explored at laboratory-scale studies include wastewater characteristics such as pH and conductivity, and process variables such as current density, operating time and electrode material type. Meanwhile, an important design variable not fully investigated in these researches is the electrode connection mode which needs to be considered also for a complete technical–economic evaluation before the process design. Thus, the aim of this study is to accomplish a complete process analysis by comparing the performances of various electrode connection modes and sacrificial electrode material types as function of wastewater pH, current density and operating time. Two performance criteria are considered namely, COD and turbidity removals. Further goal of this study is to select process configurations with highest removals, to compare them with conventional chemical coagulation, and to gather data for an economic evaluation which will also be presented in this study.

Table 1
Characteristics of textile wastewater used

Characteristics	Value
Chemical oxygen demand (COD) ($g\,m^{-3}$)	2031
Total suspended solids (TSS) ($g\,m^{-3}$)	102
Conductivity ($\mu S\,m^{-1}$)	2310
Turbidity (NTU)	671
pH	8.88

2. Experimental

2.1. Materials

The wastewater was obtained from a tank containing a mixture of exhaust dyeing solutions at a textile factory in Turkey (Gebze) producing approximately 1000 m^3 of wastewater per day. The composition of the wastewater is shown in Table 1. The wastewater was first filtered using a screen filter to remove large suspended solids before it was used for the subsequent studies.

2.2. Apparatus and instruments

The experimental setup is shown in Fig. 1. The thermostated electrocoagulator was made of Plexiglas with the dimensions of 120 mm × 110 mm × 110 mm. There are four electrodes used in each configuration. Both aluminum or iron cathodes and anodes were made from plates with dimensions of 45 mm × 53 mm × 3 mm. The total effective electrode area was 143 cm^2 and the spacing between electrodes was 20 mm. The electrodes were connected to a digital dc power supply (Topward 6306D; 30 V, 6 A) operated at galvanostatic mode. The following electrode connection modes have been considered.

2.2.1. Monopolar electrodes in parallel connections (MP-P)

As shown in Fig. 1(a); anodes and cathodes are in parallel connection, the current is divided between all the electrodes

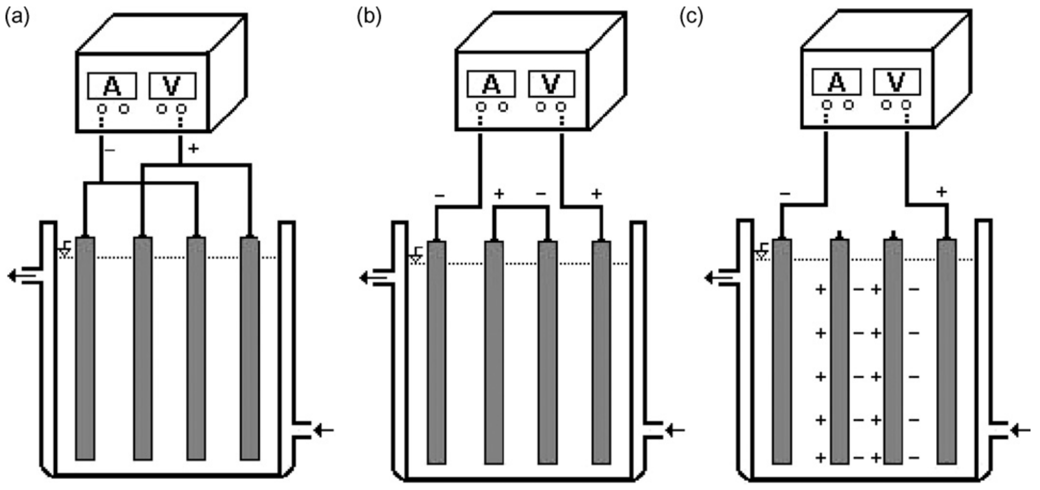

Fig. 1. (a) MP-P system, (b) MP-S system, and (c) BP-S system.

in relation to the resistance of the individual cells. Hence, a lower potential difference is required in parallel connection, when compared with serial connections.

2.2.2. *Monopolar electrodes in serial connections (MP-S)*

As shown in Fig. 1(b); each pair of sacrificial electrodes is internally connected with each other, because the cell voltages sum up, a higher potential difference is required for a given current.

2.2.3. *Bipolar electrodes in serial connections (BP-S)*

As shown in Fig. 1(c); there is no electrical connection between inner electrodes, only the outer electrodes are connected to the power supply. Outer electrodes are monopolar and inner ones are bipolar. This connection mode has simple setup with and has less maintenance cost during operation.

2.3. *Procedure*

All the runs were performed at constant temperature of 20 °C. A constant magnetic stirring at 250 rpm was applied to enhance mass and heat transfer. In each run, 750 cm^3 of the wastewater solutions was placed into the electrolytic cell. The current density was adjusted to a desired value and the coagulation was started. At the end of electrocoagulation, the solution was filtered and then was analyzed. The solid residue is air-dried until constant weight. Before each run, electrodes were washed with acetone to remove surface grease, and the impurities on the aluminum or iron electrode surfaces were removed by dipping for 5 min in a solution freshly prepared by mixing 100 cm^3 HCl solution (35%) and 200 cm^3 of hexamethylenetetramine aqueous solution (2.80%). At the end of the run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, dried and re-weighted.

2.4. *Analysis*

Chemical oxygen demand (COD), total suspended solids (TSS) and turbidity were carried out according to the Standard Methods for Examination of Water and Wastewater [39]. The turbidity (NTU) and COD of samples were analyzed using a Shimadzu Model UV-160 double beam spectrophotometer. The pH was measured using AZ 8601 model pH meter, and the conductivity was determined with Lutron CD-4303 model conductivity meter.

2.5. *Economic analysis*

Total operation cost has been calculated for a plant with a capacity of 1000 m^3 wastewater per day. It includes direct cost items such as electricity, material (electrodes and chemical reagents), sludge transportation and disposal costs, as well as indirect cost items such as labor, maintenance and depreciation of the major equipments including rectifier and electrocoagulator. Economic data used for the evaluation of the total operating cost are given for the third quarter 2005, Turkey, in Table 2.

Table 2
Economical data used in calculating of the operating cost

Item	Cost ($)
Rectifier installing cost	10,000
EC tank installing cost	500
Maintenance ($\$\,m^{-3}$)	0.003
Electricity ($\$\,kWh^{-1}$)	0.06
Labor costs ($\$\,m^{-3}$)	0.06
Sludge transportation and disposal ($\$\,kg^{-1}$)	0.01
Materials and chemical costs	
Fe electrode ($\$\,kg^{-1}$)	0.3
Al electrode ($\$\,kg^{-1}$)	1.8
Chemicals ($\$\,m^{-3}$)	0.025
$FeCl_3{\cdot}6H_2O$ ($\$\,kg^{-1}$)	0.34
$Fe_2(SO_4)_3{\cdot}7H_2O$ ($\$\,kg^{-1}$)	0.4
$AlCl_3{\cdot}6H_2O$ ($\$\,kg^{-1}$)	0.8
$Al_2(SO_4)_3{\cdot}18H_2O$ ($\$\,kg^{-1}$)	0.4

3. Results and discussion

The effects of wastewater pH, current density and operating time are presented separately for two sacrificial electrode materials, Fe and Al, and three electrode connection modes, MP-P, MP-S and BP-S.

3.1. *Effect of initial pH*

The effect of initial pH has been explored at constant current density of 30 $A\,m^{-2}$ and operating time of 15 min. Generally, similar trends are observed, as seen in Fig. 2. In the case of iron electrode, the electrode connection mode is more effective on the pH change. EC exhibits some buffering capacity observed especially at alkaline conditions and more clearly in the aluminum case.

The COD removal efficiency is strongly pH-dependent as presented in Fig. 3. For iron electrode, in acidic medium, connection modes have very different COD removal performances and BP-S mode have the highest COD removal efficiency near 70% at pH 5, while in neutral–alkaline mediums, all modes perform in

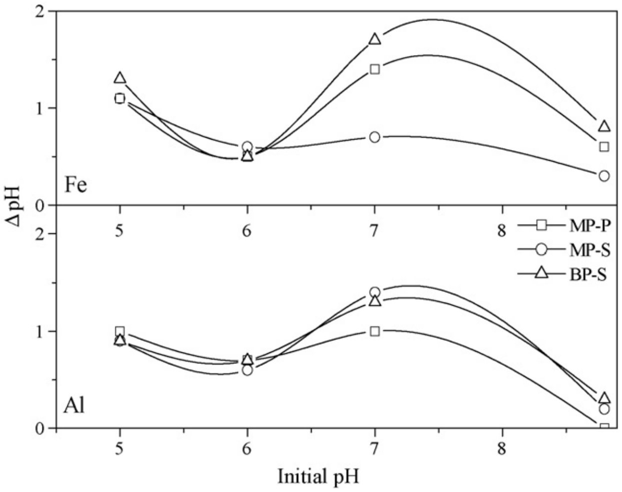

Fig. 2. pH change ($pH_{final} - pH_{initial}$) after electrocoagulation.

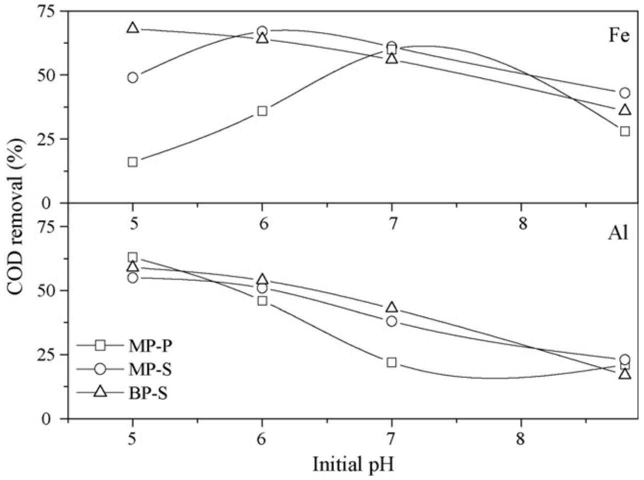

Fig. 3. Effect of initial pH on COD removal.

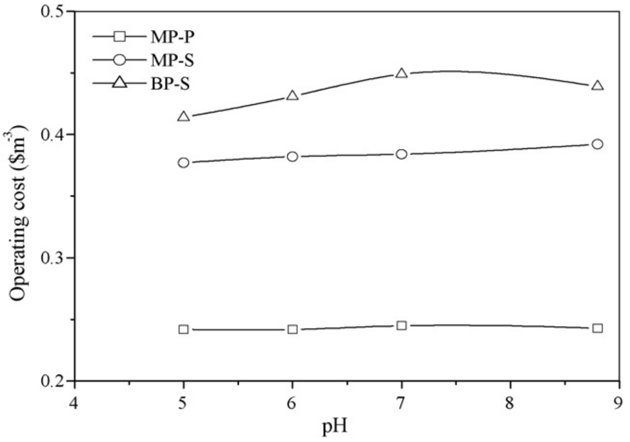

Fig. 5. Effect of initial pH on operating cost for iron electrodes.

a decreasing trend, MP-S mode performing better than the others. In the case of aluminum, no great differences are observed between connection modes; in acidic medium (pH 5), MP-S have the maximum performance (63%), while the removals lie near 20% at pH 10.

Turbidity removal-pH dependency is presented in Fig. 4; as a general trend, turbidity removals drop dramatically with increasing pH. In the iron case, the best performance, as high as 90%, is obtained at neutral pH (6–7) with MP-S mode. At low pH, process causes extra turbidity in wastewater due to colloidal sized hydroxide flocs which need filtering under pressure using denser filter paper; this filtering technique revealed to be efficient at pH 6 (shown as black points in Fig. 4) but inefficient at pH 5. On the other hand, aluminum electrode exhibits different trend than iron electrode; acidic medium at pH 5 favors high turbidity, greater than 90% with BP-S mode.

In conclusion, acidic pH 5 is preferable for COD removal for both electrode materials, which is also beneficial for the turbidity removal in aluminum case; thus, pH 5 is optimum for aluminum electrode. Meanwhile, despite high COD removal, the poor turbidity removal or poor filterability of the flocs at pH 5, dictate pH 7 to be more suitable for the iron electrode.

For a given connection mode, the electrode consumption is weakly dependent on the initial pH. The highest consumption is obtained with BP-S; near 0.27 kg m^{-3} for iron electrode and between 0.18 and 0.23 kg m^{-3} for aluminum electrode material. MP-P system shows the lowest electrode consumption for both electrode materials; 0.12 kg m^{-3} for aluminum electrode, and 0.16 kg m^{-3} iron electrodes.

When energy consumptions are compared, weak dependence on pH is observed for all of the systems; MP-S and BP-S systems exhibit high consumptions as the consequence of the serial connection requiring higher potential. The lowest consumption values are approximately, 0.63 kWh m^{-3} for iron electrode, and 0.70 kWh m^{-3} for aluminum electrode, with MP-P mode.

On the other hand, sludge amounts vary between 0.65–1.0 kg m^{-3} in the case of iron, and 0.9–1.3 kg m^{-3} in the case of aluminum. In general, more sludge is produced in BP-S mode and less sludge with MP-P mode. Meanwhile, the differences between various connection modes are not very different.

As seen in Figs. 5 and 6, for both electrode materials, MP-P system is economically more feasible owing to low electrode material and electrical energy consumptions, and low amount

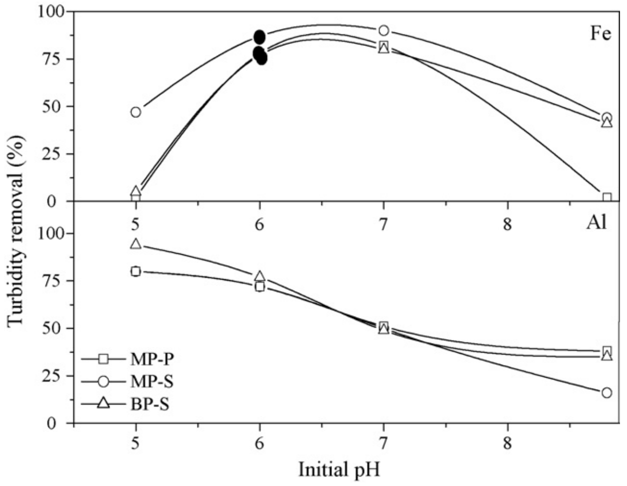

Fig. 4. Effect of initial pH on turbidity removal.

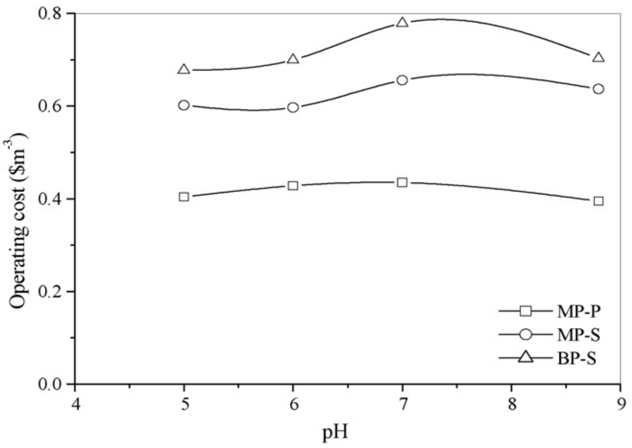

Fig. 6. Effect of initial pH on operating cost for aluminum electrodes.

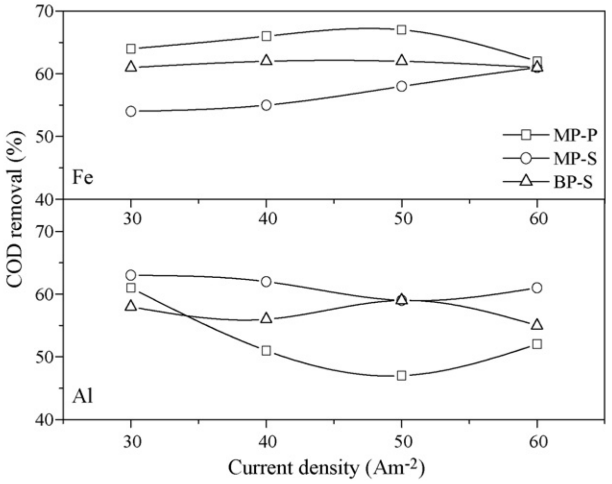

Fig. 7. Effect of current density on COD removal.

of sludge produced; \$ 0.18 and 0.34 m^{-3}, in the case of iron and aluminum electrodes, respectively. BP-S system has the highest cost values which reaches \$ 0.73 m^{-3} with aluminum, and \$ 0.34 m^{-3} with iron electrode.

3.2. Current density

Fig. 7 depicts the effect of current density on COD removal efficiency with operating time 15 min, at constant pH 7 for iron electrode, and pH 5 for aluminum electrode. In the case of iron, the current density is not very effective on the performances of MP-P and MP-S modes especially; COD removal reach the maximum 67% with MP-P mode for a current density of 50 A m^{-2}. For aluminum electrode, the effect of the current density is more pronounced on COD removal, especially for MP-P mode. Lower current density is preferable; maximum removal (63%) is attained at 30 A m^{-2} with MP-S mode.

Turbidity removal is plotted against current density in Fig. 8. In the iron case, turbidity removal increase with increasing current density, MP-P system exhibit slightly lower performance than two others which performs nearly equally; a current density of 50 A m^{-2} ensures 92% turbidity removal in the case of MP-S and BP-S modes, while 89% for MP-P mode. In the aluminum case, current density has almost no effect on the turbidity removal lying between 82 and 85%.

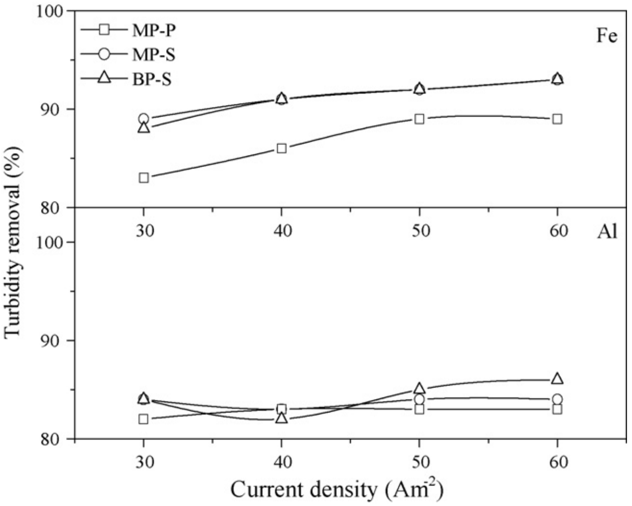

Fig. 8. Effect of current density on turbidity removal.

When both removal efficiencies are taken into account, MP-P mode using iron electrode and operating at 50 A m^{-2} ensure 67% COD and 89% turbidity removals, while at 30 A m^{-2}, 65% COD and 83% turbidity removals are obtained. As low current density is beneficial for a low operating cost, further analysis is needed to choose the optimum current density. On the other hands, with aluminum electrode, MP-S mode operating at 30 A m^{-2} is preferable, which ensures 63% COD and 84% turbidity removals.

Electrode consumption values increase with increasing current density. At 60 A m^{-2} current density and for BP-S system, it reaches the highest value of 0.58 kg m^{-3} in the iron case, and 0.32 kg m^{-3} for aluminum electrode, while the lowest consumption is obtained with MP-P mode; 0.27 kg m^{-3} in iron case and 0.11 kg m^{-3} in the aluminum case. It is clear that electrode consumptions are higher with iron electrode material, but on a weight basis. By considering atomic weights of aluminum (27) and iron (56.5), the consumption on molar basis are not very different; with MP-P mode, for example, these are calculated as 4.78 and 4.07 mole g m^{-3} for iron and aluminum electrodes, respectively.

All of the systems exhibit similar trends with respect to energy consumption as function of current density; it increases with increasing current density. Similar to pH effects on energy consumption, BP-S system consumes the highest energy, while MP-P system is the most economical with 30 A m^{-2} of current density. Its consumption values are 0.68 and 0.72 kWh m^{-3} for iron and aluminum electrodes, respectively.

Sludge amount increases with increasing current density for all connection modes and electrode material types. BP-S and MP-S systems show similar values. MP-P system produces the lowest amount of sludge as 0.81 and 0.9 kg m^{-3} for iron and aluminum electrodes, respectively, at a current density of 30 A m^{-2}. When these latter values are revaluated on a molar basis by considering molar electrode consumptions, it is seen that aluminum hydroxide flocs bound more water, chemically or physically, than iron hydroxide flocs.

The effect of the current density on the operating cost of EC process is presented in Figs. 9 and 10. For both electrode materials, operating cost increases more rapidly for MP-S and BP-S modes when compared with MP-P mode. BP-S system exhibits highest cost values as \$ 1.02 and 1.48 m^{-3} for iron and aluminum electrodes, respectively. MP-P mode using iron as the electrode material is the most economical one with \$ 0.2 m^{-3} operating cost, at a current density of 30 A m^{-2}.

3.3. Operating time

To investigate the effect of operating time; the current density is selected as 30 A m^{-2}, pH is hold at 5 for aluminum electrode, and at pH 7 for iron electrode. The results are given in

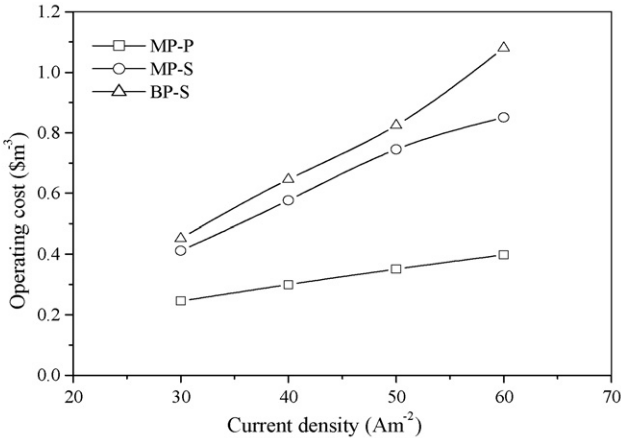

Fig. 9. Effect of current density on operating cost for iron electrodes.

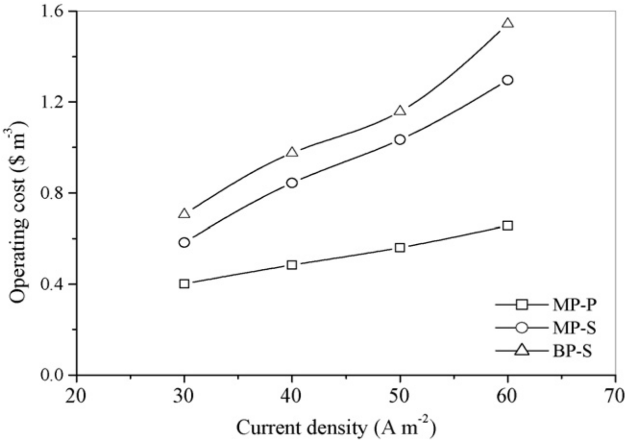

Fig. 10. Effect of current density on operating cost for aluminum electrodes.

Figs. 11 and 12, for COD and turbidity removals, respectively. In the iron case, COD removal is between 64 and 54%. It is clear that for both COD and turbidity removal, an operation time of 5 min is insufficient, after which the removal rates increase rapidly and reach steady values above 15 min for both removals.

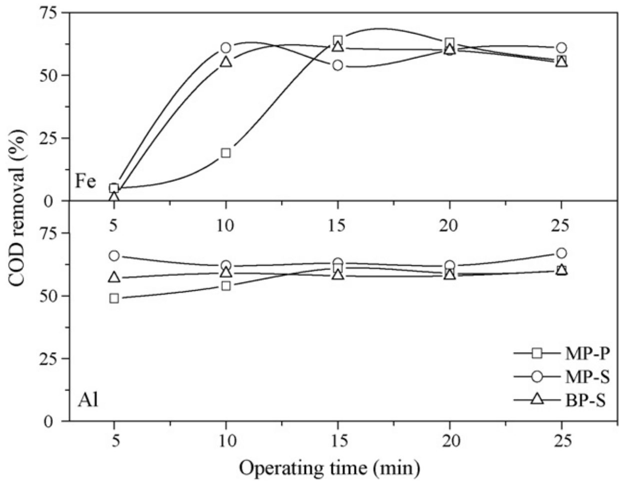

Fig. 11. Effect of operating time on COD removal.

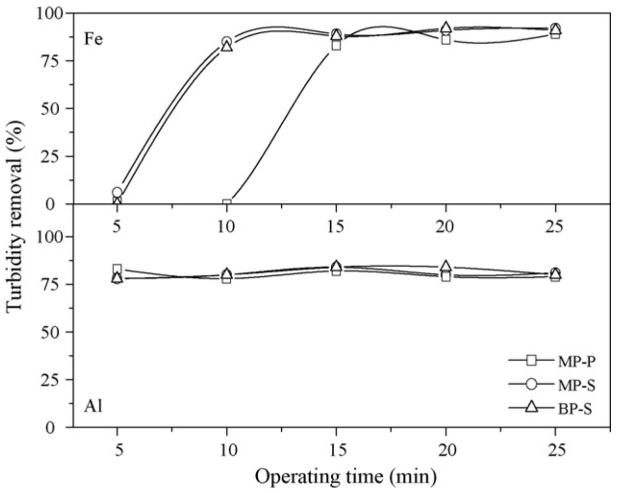

Fig. 12. Effect of operating time on turbidity removal.

In the case of aluminum, nearly steady removal efficiencies are reached within 5 min for all three systems among which MP-S system exhibits the highest performance; 65% COD removal and 78% turbidity removal.

Electrode consumption values are higher for iron electrode material. Higher electrode material has been sacrificed in BP-S system for both electrode materials. Energy consumption for MP-S and BP-S modes have similar slopes, almost 4–6.5 times greater than that of the MP-P mode. Thus, longer operating times may be used with MP-P mode, to obtain higher removal performances using less electric energy. Effect of the operating time on sludge formation is not very profound as in the case of energy consumption. Generally, more amount of sludge is formed in the aluminum case. BP-S mode produces higher amount of sludge and MP-P mode lower ones.

For a complete technical analysis, it is worth to compare EC with conventional chemical coagulation, in regard with removal efficiencies and various important aspects. For this purpose, jar-tests were performed at laboratory scale in order to determine the adequate coagulant dosage. After choosing the best amount, same experiments have been performed to determine optimum pH value for each coagulant. Experimental conditions, removal efficiencies and some other pertinent data of EC and CC process variations are shown in Table 3. At first sight, it is clearly seen that EC is faster, consumes less material and produces less sludge, and pH of the medium is more stabilized than chemical coagulation for similar COD and turbidity removal levels. The process using aluminum electrodes connected in MP-S mode seems to be the best choice.

MP-P mode is the most cost-effective for both electrode materials. Both materials show similar results in reducing COD and turbidity, but iron is preferred as a low cost one. pH 7 and 5 is suitable for iron and aluminum electrodes, respectively. Meanwhile, 30 A m^{-2} of current density and 15 min of operating time are sufficient for both materials. For a concluding economic analysis, it is worth to compare technical–economical performances of EC and chemical coagulation (CC).

Operating cost values with regard to operating time is shown in Figs. 13 and 14. As expected, operating cost increases with

Table 3
Technical comparison between electrocoagulation and chemical coagulation

Process electrode/coagulant	Electrocoagulation (EC)[a]		Chemical coagulation (CC)			
	Fe electrode	Al electrode	$FeCl_3 \cdot 6H_2O$	$Fe_2(SO_4)_3 \cdot 7H_2O$	$AlCl_3 \cdot 6H_2O$	$Al_2(SO_4)_3 \cdot 18H_2O$
Sacrificial electrode or coagulant consumption						
$kg\,m^{-3}$ wastewater	0.163	0.107	1.500	1.500	1.000	1.500
$kg\,kg\,COD^{-1}$ removed	0.126	0.095	1.761	1.586	0.828	1.896
$Me(OH)_3$ produced						
$kg\,m^{-3}$ wastewater	0.309	0.168	0.595	0.615	0.325	0.355
Operating time[b] (min)	15	10	25	25	25	25
Initial pH	7.0	5.0	7.0	7.0	6.0	6.0
Final pH	7.9	5.7	2.9	3.1	4.1	4.1
COD removal (%)	65	63	71	68	68	59
Turbidity removal (%)	83	80	87	63	89	90
Operating cost ($\$\,m^{-3}$)	0.25	0.40	0.67	0.75	0.96	0.75

[a] Connection mode: Fe electrode: MP-P, Al electrode: MP-S, current density for both electrode: $30\,A\,m^{-2}$.
[b] Operating time of CC includes 5 min of rapid mixing at 250 rpm and 20 min of slow mixing at 50 rpm.

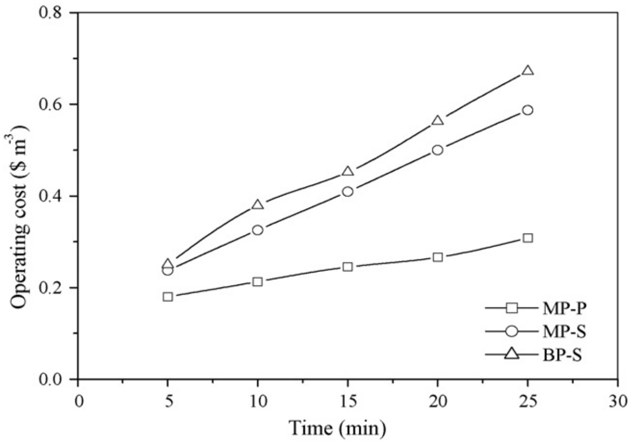

Fig. 13. Effect of operating time on operating cost for iron electrodes.

increasing operating time. All connection modes with iron electrode exhibit low cost values than aluminum counterparts. For an operating time of 25 min, with BP-S system using aluminum electrode, the operating cost reaches high values as \$ 1.1 m^{-3}, while with MP-P mode with iron electrode, the operating cost is 5.5 times lower. For CC, $FeCl_3$ is the preferable salt in view of its techno-economic performance. On the other hand, iron is the preferred electrode material in EC.

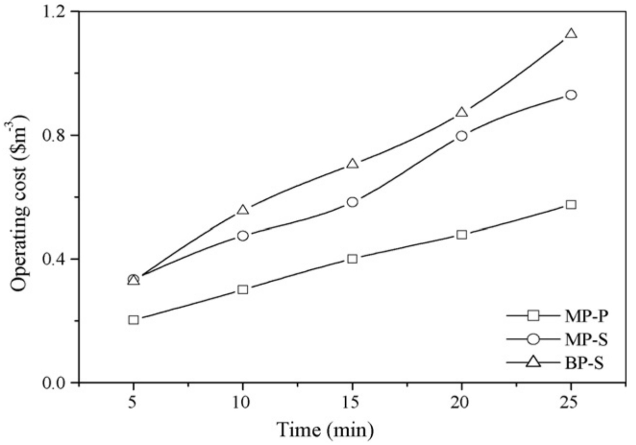

Fig. 14. Effect of operating time on operating cost for aluminum electrodes.

4. Conclusions

The following conclusions may be drawn from the experimental results:

1. Acidic medium is preferable for a high COD removal for both electrode materials; iron electrode performs clearly better with BP-S mode, while the performance of aluminum is not strongly dependent on connection mode. For a high turbidity removal, the optimum pH depends on the electrode material; aluminum electrode connected in BP-S mode performs better in acidic medium, while the poor filterability of the flocs dictates pH 7 to be more suitable for the iron electrode connected in MP-S mode.
2. High current density is generally favorable for high COD and turbidity removals in the case of iron; at low current density, MP-P mode performs better, while at high current densities, the three modes perform equally well. In the aluminum case, the effect is more pronounced on COD removal and it depends strongly on the connection mode, but it has nearly a negligible effect on the turbidity removal which also unaffected by the connection mode.
3. In the case of aluminum, steady removal efficiencies are reached within 5 min for all type of the connections, while for iron electrode, serial connection systems, BP-S and MP-S reach steady values in 10 min, while MP-P needs longer operating time.
4. The COD removal performance of CC is 10% higher than EC, the turbidity removal is nearly the same, but in 60% longer retention time.
5. With the same initial pH, the final pH is 7.9 in EC, but 2.9 in CC. The final acidic and chloride bearing medium is an important drawback of CC, causing severe corrosion problems which may necessitate high-cost building materials.

From this point, $Fe_2(SO_4)_3{\cdot}7H_2O$ may be used despite of its higher operating cost.

6. High coagulant consumption in CC means high chloride concentration in the effluent.
7. Finally, and more importantly, the operating cost of CC is 3.2 times as high as the operating cost of EC.

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