



## Removal turbidity and separation of heavy metals using electrocoagulation–electroflotation technique A case study

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### ABSTRACT

The electrocoagulation (EC) process was developed to overcome the drawbacks of conventional wastewater treatment technologies. This process is very effective in removing organic pollutants including dyestuff wastewater and allows for the reduction of sludge generation. The purposes of this study were to investigate the effects of the operating parameters, such as pH, initial concentration ( $C_0$ ), duration of treatment ( $t$ ), current density ( $j$ ), interelectrode distance ( $d$ ) and conductivity ( $\kappa$ ) on a synthetic wastewater in the batch electrocoagulation–electroflotation (EF) process. The optimal operating conditions were determined and applied to a textile wastewater and separation of some heavy metals. Initially a batch-type EC–EF reactor was operated at various current densities (11.55, 18.6, 35.94, 56.64, 74.07 and 91.5 mA/cm<sup>2</sup>) and various interelectrode distance (1, 2 and 3 cm). For solutions with 300 mg/L of silica gel, high turbidity removal (89.54%) was obtained without any coagulants when the current density was 11.55 mA/cm<sup>2</sup>, initial pH was 7.6, conductivity was 2.1 mS/cm, duration of treatment was 10 min and interelectrode distance was 1 cm. The application of the optimal operating parameters on a textile wastewater showed a high removal efficiency for various items: suspended solid (SS) 86.5%, turbidity 81.56%, biological oxygen demand (BOD<sub>5</sub>) 83%, chemical oxygen demand (COD) 68%, and color over 92.5%. During the EC process under these conditions, we have studied the separation of some heavy metal ions such as iron (Fe), nickel (Ni), copper (Cu), zinc (Zn), lead (Pb) and cadmium (Cd) with different initial concentrations in the range of 50–600 mg/L and initial pH between 7.5 and 7.8. This allowed us to show that the kinetics of electrocoagulation–electroflotation is very quick (<15 min), and the removal rate reaches 95%.

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### 1. Introduction

The large quantity of aqueous waste generated by metal finishing and textile industries has become a significant environmental problem. Dye bath effluents, in particular, are not only aesthetic pollutants by nature of their color, but may also interfere with light penetration in the receiving bodies of water, thereby disturbing biological processes. Furthermore, dye effluent may contain chemicals, which are toxic, carcinogenic, mutagenic, or teratogenic in various microbiologic, fish species [1]. The characteristics of wastewater from textile dyeing are high or low pH, high temperature and a high concentration of coloring material. The method of treatment of dyeing wastewater are biological treatment, chemical coagu-

lation, activated carbon adsorption, ultrafiltration, ozonation and electrocoagulation (EC) [2,3]. The costs of adsorption, ultrafiltration and ozonation exceed that of chemical coagulation. When chemical coagulation is used to treat dyeing wastewater, the pollution may be caused by chemical substance added at a high concentration. Excessive coagulant material can be avoided by EC. The coagulant in this case is generated by dissolution of a sacrificial anode [2,4]. EC uses an electrical current to produce several metal ions in solution. In fact, the EC systems can be effective in removing suspended solids, dissolve metals, tannin and dyes. The contaminants present in wastewater are maintained in solution by electrical charges. When metal ions are neutralized with ions of opposite electric charge, provided by an EC system, they become unstable and precipitate in a form that is usually very stable [5]. The characteristics of EC are simple equipment and easy operation, brief reactive retention period, decreased or negligible equipment for adding chemicals and decreased amount of sludge [2]. The elec-

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### Nomenclature

$A_{740}$	absorbance measured at 740 nm
AAS	atomic absorption spectrometer
BOD <sub>5</sub>	biological oxygen demand (mg O <sub>2</sub> /L)
C	concentration (mg/L)
COD	chemical oxygen demand (mg O <sub>2</sub> /L)
$d$	interelectrode distance (cm)
EC	electrocoagulation
EF	electroflotation
$I$	current intensity (A)
$j$	current density (mA/cm <sup>2</sup> )
$M$	anode material
$n$	number of electrons
$S$	electrode surface area (cm <sup>2</sup> )
SS	suspended solids (mg/L)
$t$	time (min)
TR	turbidity removal efficiency (%)
$U$	measured potential (V)

### Greek letters

$\phi_{50}$	mean diameter
$\kappa$	wastewater conductivity (mS/cm)
$\lambda_{\max}$	maximum wavelength of the absorption spectrum (nm)

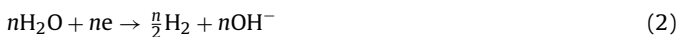
trochemical process ideal to take advantage of the combined effect due to the production of polyvalent cations from the oxidation of corrodible anodes (such as Fe and Al). The gas bubbles carry the pollutant to the top of the solution where it can be more easily concentrated, collected and removed. The metallic ions can react with the OH<sup>-</sup> ions produced at the cathode during the evolution of H<sub>2</sub> gas, to yield insoluble hydroxides that will sorb pollutants out of the solution. It also contributes to coagulation by neutralizing any negatively charged colloidal particles which have been reported to be more compact than sludge obtained by chemical methods.

The electrocoagulation process is as follows:

At the anode,



At the cathode,



where  $M$  = anode material and  $n$  = number of electrons involved in the oxidation/reduction reaction.

Metal (soluble metal such as Al, Fe) ions are generated at the anode and react with the hydroxide ions formed at the cathode, and the metal hydroxides are produced as below. These insoluble metal hydroxides react with the suspended and/or colloid solids and precipitate.



Recently, investigations have been focused on the treatment of wastewaters using electrocoagulation. The electrocoagulation has successfully been used for the treatment of wastewaters such as electroplating wastewater [6], laundry wastewater [7], latex particles [8], restaurant wastewater [9] and slaughterhouse wastewater [10]. Meanwhile, EC process has been widely used in the removal of arsenic [11], phosphate [12], sulfide, sulfate and sulfite [13], boron [14], fluoride [15], nitrate [16] and chromate [17]. Treatments of wastewaters containing textile dyes have been studied by electrocoagulation method. The results of these studies show that COD,

color, turbidity and dissolved solids at varying operating conditions are considerably removed [18,19].

In addition, it is clear that a technically efficient process must also be economically feasible with regard to its initial capital and operating costs, and practically applicable to the environmental problems. The economic aspect of the electrocoagulation process is not investigated well by the researches except a few of them [18]. Electrical energy consumption is a very important economical parameter in EC process like all other electrolytic processes.

In this study, experiments were conducted to examine the effects of the operating parameters, such as pH, initial concentration ( $C_0$ ), retention time ( $t$ ), current density ( $j$ ), interelectrode distance ( $d$ ) and conductivity ( $\kappa$ ) on simulated model of wastewater in the batch electrocoagulation process. The optimal operating conditions were determined and applied to a textile wastewater and separation of some heavy metal ions.

## 2. Experimental

### 2.1. Materials and methods

The experimental equipment schematically is shown in Fig. 1. The electrocoagulation–electroflotation (EF) unit consisted of an 1.5 L electrochemical reactor with two aluminum electrodes of rectangular shape (27 mm × 17 mm × 1 mm), which corresponds to  $S=4.59\text{ cm}^2$  electrode surface area, installed horizontally in the middle of the reactor. The electrodes were connected to a DC power supply (Didalab, France) providing 0–30 V (0–8 A) with galvanostatic operational options for controlling the current density. All the runs were performed at room temperature. In each run, 1000 ml of the synthetic wastewater was decanted into the electrolytic cell. All samples were taken from the effluent port located 05 cm above the bottom of the reactor and measured over time after sedimentation. Neither centrifuging nor filtration was performed. All experiments were repeated three times, and the experimental error was around 2%. The synthetic wastewater was prepared using 1000 mL of tap water and 300 mg/L of silica gel (fluka) sieved to particle dimensions of 43–500  $\mu\text{m}$ . The wastewater used in this research was taken from the effluent of the textile wastewater. Simulated solutions of heavy metal ions were prepared to further evaluate the efficacy of the EC–EF process. The procedure has been described later.

### 2.2. Chemical analysis

The measurements of COD (chemical oxygen demand), BOD<sub>5</sub> (biological oxygen demand), SS (suspended solids) and color followed the procedure of a standard method [20]. The conductivity of solutions was measured using a CD810 conductimeter (Radiome-

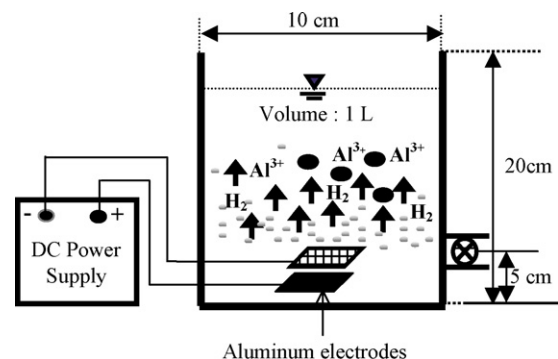


Fig. 1. Batch electrocoagulation–electroflotation unit.

ter Analytical, France). For the analysis of particle distribution of the simulated wastewater, a Retsch apparatus was utilized. Turbidity was determined by UV–vis spectrophotometer (Shimadzu UVPC1601, Japan) and pH was measured by using EC30 pH meter. AAS (Atomic Absorption Spectrometer, Shimadzu, Japan) was used to determine heavy metals. Silica gel concentration was estimated from its absorbance characteristics in the UV–vis rang (200–800 nm) at maximum wavelength  $A_{740}$  ( $\lambda_{max} = 740$  nm). The calculation of turbidity removal efficiency (TR%) after electrocoagulation treatment was performed using this formula:

$$TR(\%) = \frac{C_0 - C}{C_0} \cdot 100 \quad (4)$$

where  $C_0$  and  $C$  are concentrations of silica gel before and after electrocoagulation in mg/L.

### 3. Results and discussions

When current passes through an electro-chemical reactor, it must overcome the equilibrium potential difference, anode overpotential, cathode overpotential and ohmic potential drop of the solution [21]. When aluminum is used as an electrode material, there are three major reactions in the electrochemical reactor as follows [22]:

(a) The oxidation reaction at the anode,



(b) The reduction reaction at the cathode,



The hydrolysis reaction,



#### 3.1. Effect of current density and interelectrode distance on the turbidity removal

The current density is expected to exhibit a strong effect on EC [23–33], especially on the kinetics of turbidity removal: higher the current, shorter the treatment. This is ascribed to the fact that at high current density, the extent of anodic dissolution of aluminum increases, resulting in a greater amount of precipitate for the removal of pollutants. Moreover, bubble generation rate increases and the bubble size decreases with increasing current density. These effects are both beneficial for high pollutant removal by  $H_2$  flotation [27]. To investigate the effect of current density and interelectrode distance on the efficiency of turbidity removal, electrocoagulation process was carried out using various current densities and distance between the electrodes. An initial pH (7.6) value was used in this study. The current density and interelectrode distance ranged from 11.55 to 91.5 mA/cm<sup>2</sup>, 1 to 3 cm, respectively, in the experiments, and the results are shown in Figs. 2–4. When the current density was increased from 11.55 to 91.5 mA/cm<sup>2</sup> and interelectrode distance was increased from 1 to 3 cm, it was observed that the turbidity removal efficiency decrease and do not change after 10 min retention time of the wastewater in the EC unit. For this reason, the optimal current density, distance between electrodes and retention time for treatment of the simulated wastewater are considered to be 11.55 mA/cm<sup>2</sup>, 1 cm and 10 min, respectively. With these experimental conditions, the turbidity removal efficiency reached up to 89%.

From Figs. 2–4, it can be seen that the turbidity has a very particular variation: it increases at the beginning of treatment under the

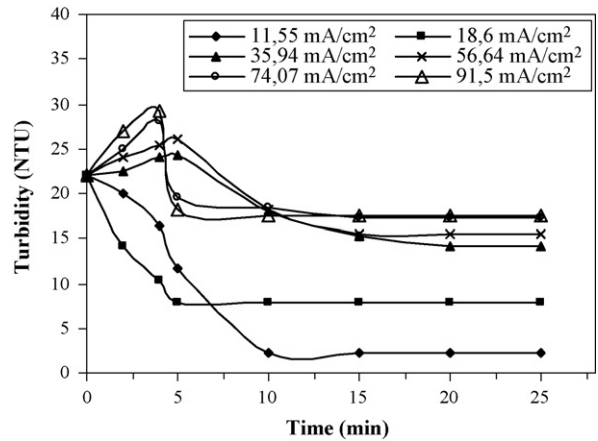


Fig. 2. Effect of current density  $j$  on the turbidity removal:  $C_0 = 300$  mg/L, interelectrode distance  $d = 1$  cm, initial pH 7.6, conductivity  $\kappa = 2.1$  mS/cm.

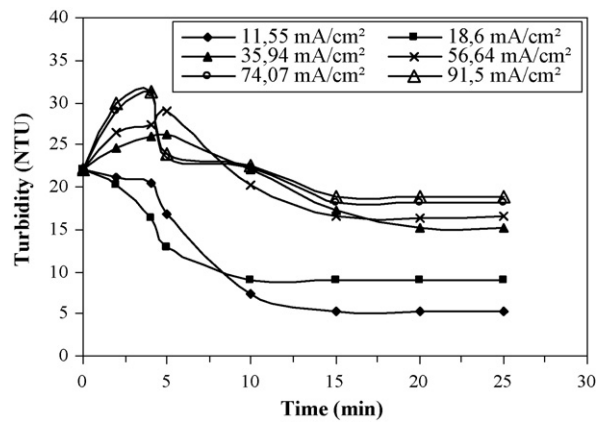


Fig. 3. Effect of current density  $j$  on the turbidity removal:  $C_0 = 300$  mg/L, interelectrode distance  $d = 2$  cm, initial pH 7.6, conductivity  $\kappa = 2.1$  mS/cm.

effect of the accumulation of the aluminum hydroxide in the polymer shape to reach very strong values. A decrease starts to reach, in all the studied cases, a very low turbidity. The processing time necessary depends on the operating conditions. The negatives removal efficiencies of the turbidity at low times and high current densities can be explained: during the flow of the electrical current, additional convection occurs. According to EDELIN [34,35], dissolved aluminum is polymerized in the shape of aluminum hydroxide and thus increases turbidity.

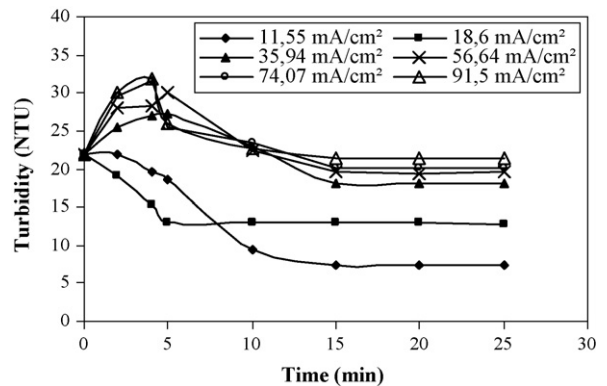


Fig. 4. Effect of current density  $j$  on the turbidity removal:  $C_0 = 300$  mg/L, interelectrode distance  $d = 3$  cm, initial pH 7.6, conductivity  $\kappa = 2.1$  mS/cm.

The decrease of turbidity removal with the increase of current density is probably due to the adsorption of the oxygen bubbles produced by the anode, which is horizontally arranged under the cathode; although the latter is perforated, oxygen is adsorbed on the lower face of cathode and remains blocked on this one. This technical problem induces a reduction in the turbidity removal.

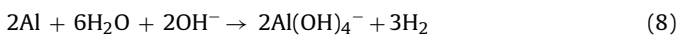
Thus, the following investigation of removal efficiency for different items was accomplished only at the experimentally selected optimum parameters: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$  and retention time  $t = 10 \text{ min}$ .

### 3.2. Effect of initial pH on the efficiency of turbidity removal

It has been established that the influence pH is an important parameter influencing the performance of the electrochemical process [3,9]. To examine its effect, the sample was adjusted to a desired pH for each experiment by using sodium hydroxide or sulfuric acid. Fig. 5 demonstrates the removal efficiencies of turbidity as a function of the influent pH. The maximum removals of turbidity were observed at pH around 8. However, the pH changed during batch EC, as already mentioned in the above-mentioned papers. Its evolution depended on the initial pH. EC process exhibits some buffering capacity because of the balance between the production and the consumption of  $\text{OH}^-$  [32], which prevents high change in pH (Fig. 6). The buffering pH seems just above 7: when the initial pH is above this value, pH decreases during EC; otherwise, the opposite behavior is observed.

The effect of pH can be explained as follows.

At low pH, such as 2–3, cationic monomeric species  $\text{Al}^{3+}$  and  $\text{Al}(\text{OH})_2^+$  predominate. When pH is between 4 and 9, the  $\text{Al}^{3+}$  and  $\text{OH}^-$  ions generated by the electrodes react to form various monomeric species such as  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}(\text{OH})_2^{2+}$ , and polymeric species such as  $\text{Al}_6(\text{OH})_{15}^{3+}$ ,  $\text{Al}_7(\text{OH})_{17}^{4+}$ ,  $\text{Al}_{13}(\text{OH})_{34}^{5+}$  that finally transform into insoluble amorphous  $\text{Al}(\text{OH})_{3(s)}$  through complex polymerization/precipitation kinetics [25]. When pH is higher than 10, the monomeric  $\text{Al}(\text{OH})_4^-$  anion concentration increases at the expense of  $\text{Al}(\text{OH})_{3(s)}$ . In addition, the cathode may be chemically attacked by  $\text{OH}^-$  ions generated together with  $\text{H}_2$  at high pH values [36]:



Two main mechanisms are generally considered: *precipitation* for pH lower than 4 and *adsorption* for higher pH. Adsorption may proceed on  $\text{Al}(\text{OH})_3$  or on the monomeric  $\text{Al}(\text{OH})_4^-$  anion depending on the pollutant chemical structure. The formation of  $\text{Al}(\text{OH})_{3(s)}$

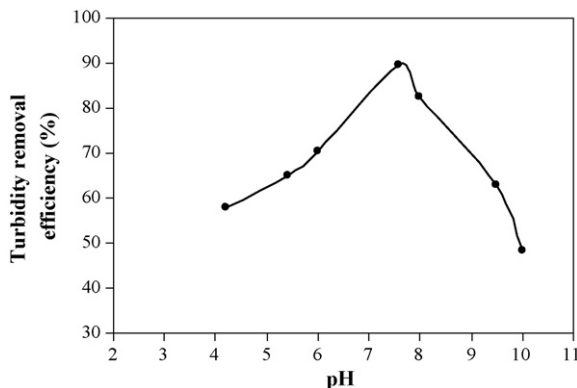


Fig. 5. Effect of initial pH on the turbidity removal efficiency:  $C_0 = 300 \text{ mg/L}$ , current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , retention time  $t = 10 \text{ min}$ , conductivity  $\kappa = 2.1 \text{ mS/cm}$ .

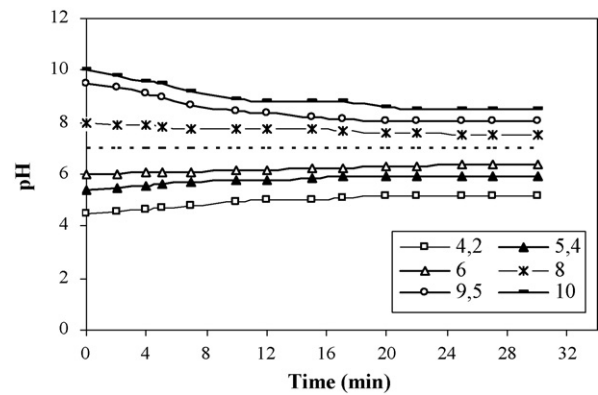


Fig. 6. Evolution of pH values during EC for different values of initial pH:  $C_0 = 300 \text{ mg/L}$ , current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , conductivity  $\kappa = 2.1 \text{ mS/cm}$ .

is therefore optimal in the 4–9 pH range, which corresponds to the optimum pH values investigated in this work.

However, pH affects also bubble size [37]. Typical bubble sizes in electroflotation always fall in the range of 20–70  $\mu\text{m}$  [38], far smaller than those observed in conventional air-assisted flotation, which provides both sufficient surface area for gas–liquid–solid interfaces and mixing efficiency to favour the aggregation of tiny destabilized particles. Hydrogen bubbles, which obey usually to a lognormal size distribution, are known to be the smallest about neutral pH [39].

As a conclusion, pH may be adjusted in the optimum range in order to achieve a compromise between best coagulation and best flotation. The optimum range may however vary as a function of electrode material and silica gel structure. In the following sections, initial pH will be fixed at about 7.6 to maximize turbidity removal efficiency.

### 3.3. Effect of initial concentration on the efficiency of turbidity removal

The variations of rate of turbidity removal for six different initial concentrations are shown in Fig. 7. Using aluminum as sacrificial anode, the removal efficiency of turbidity was effectively reduced to <54%, when the initial concentration of silica gel increased from 100 to 800 mg/L. As seen, with increasing initial concentration, removal efficiency of turbidity decreases linearly. The reason for this is the lack of sludge for adsorption of excess silica gel in high concentra-

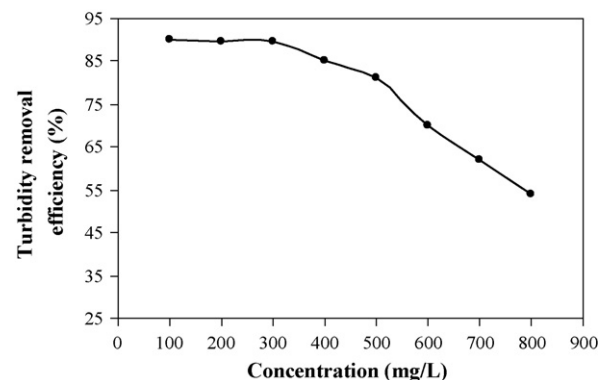


Fig. 7. Effect of initial concentration on the turbidity removal efficiency: current density  $j = 11.55 \text{ mA/cm}^2$ , initial pH 7.6, interelectrode distance  $d = 1 \text{ cm}$ , retention time  $t = 10 \text{ min}$ , conductivity  $\kappa = 2.1 \text{ mS/cm}$ .

tions. It is necessary to mention that the total amount of sludge is constant for all concentrations.

### 3.4. Effect of conductivity

The increase of the conductivity by the addition of sodium chloride is known to reduce the cell voltage  $U$  at constant current density due to the decrease of the ohmic resistance of wastewater [40,25,27]. Energy consumption, which is proportional to  $UI$ , will therefore decrease. Chloride ions could significantly reduce the adverse effects of other anions, such as  $\text{HCO}_3^-$  and  $\text{SO}_4^{2-}$ , for instance by avoiding the precipitation of calcium carbonate in hard water that could form an insulating layer on the surface of the electrodes and increase the ohmic resistance of the electrochemical cell [40,41]. Chloride anions can also be oxidized and give active chlorine forms, such as hypochlorite anions, that can oxidize dyes.

The conductivity of the simulated wastewater was adjusted to the desired levels by adding an appropriate amount of 1N NaCl solution. The experimental conditions were: initial pH of 7.6; current density  $j = 11.55 \text{ mA/cm}^2$ ; and EC retention time of 10 min. The effect of conductivity on performance of the EC process is shown in Fig. 8. In Fig. 8, turbidity removal efficiency remains almost unchanged between the conductivity range of 2100 and 3000  $\mu\text{S/cm}$ .

### 3.5. Case study: textile wastewater

The textile wastewater used in this research was taken from the effluent of a textile factory in Algeria (M'sila) producing approximately 15000  $\text{m}^3$  of wastewater per day. The initial characteristics of textile wastewater are shown in Table 1.

Under the optimal operating conditions: current density  $j = 11.55 \text{ mA/cm}^2$ , retention time  $t = 10 \text{ min}$  and interelectrode distance  $d = 1 \text{ cm}$ , the final characteristics of the wastewater used are shown in Fig. 9.

It showed that most of the SS that generated turbidity in textile wastewater were removed by 86.5%, forming a floating scum layer. Moreover, the removal efficiencies of COD and  $\text{BOD}_5$  showed values of 68% and 83%, respectively. This remarkable reduction of COD and  $\text{BOD}_5$  can be explained by the fact that EC-EF has some effects on non-biodegradable organics in the textile wastewater. The great reduction of the color (>92.5%) shows that the EC-EF is a technique of choice for the clarification of wastewater.

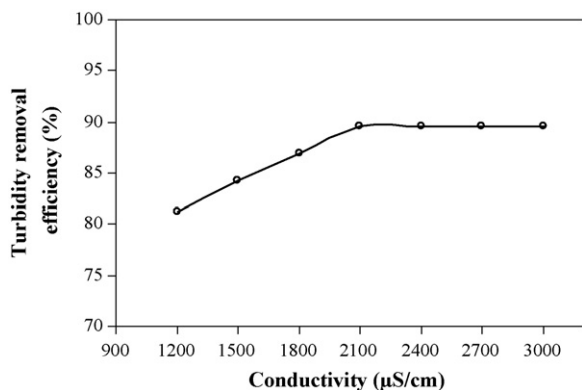


Fig. 8. Effect of conductivity on the turbidity removal efficiency:  $C_0 = 300 \text{ mg/L}$ , current density  $j = 11.55 \text{ mA/cm}^2$ , initial pH 7.6, interelectrode distance  $d = 1 \text{ cm}$ , retention time  $t = 20 \text{ min}$ .

Table 1

Characteristics of textile wastewater used (TINDAL)

Characteristics	Value
pH	8.7
Biological oxygen demand $\text{BOD}_5$ ( $\text{mg O}_2/\text{L}$ )	708
Chemical oxygen demand COD ( $\text{mg O}_2/\text{L}$ )	2000
Suspended solids SS ( $\text{mg/L}$ )	722
Turbidity (NTU)	217
Color (Co-Pt)	>200
Conductivity $\kappa$ ( $\mu\text{S/cm}$ )	4571

### 3.6. Separation of heavy metals

The possibility of removal of heavy metal ions such as copper (Cu), nickel (Ni), zinc (Zn), cadmium (Cd), lead (Pb) and iron (Fe) from wastewater by electrocoagulation was investigated.

#### 3.6.1. In the presence of sulfates

Synthetic solutions of  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ,  $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ ,  $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$ ,  $\text{PbSO}_4$  and  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  with different initial concentrations in the range of 50–600  $\text{mg/L}$  and initial pH between 7.5 and 7.8 were used for conducting the experiments. All solutions were prepared using distilled water with the following characteristics: initial concentration  $C_0 = 50$  to 600  $\text{mg/L}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ . The different experiments were carried out at  $20^\circ\text{C}$  using interelectrode distance  $d = 1 \text{ cm}$  and a current density  $j = 11.55 \text{ mA/cm}^2$ .

The experimental results are shown in Figs. 10–15.

According to the results obtained, we can note that the EC-EF process functions for all studied metal concentrations. Comparing the results obtained in this work with similar ones [42–44], it can be observed that the EC-EF process has a high average removal efficiency. As the concentration of metal solution was increased from 50 to 600  $\text{mg/L}$ , the removal efficiency decreased from 99% to 70%. The removal extent was higher for lower metal concentrations. From 50 to 400  $\text{mg/L}$ , it can be seen that the rate of removal of all metals is high (90–99%) and the kinetics of electrocoagulation is fast and does not exceed 5 min. Except for iron and copper, a time of 15 min is required to reach more than 90%. For instance, after 15 min of operation, 98.61% of the 50  $\text{mg/L}$  iron concentration was eliminated against 70.24% for the 600  $\text{mg/L}$  iron concentration.

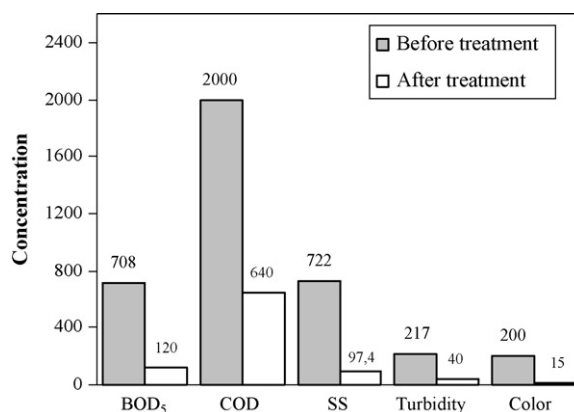
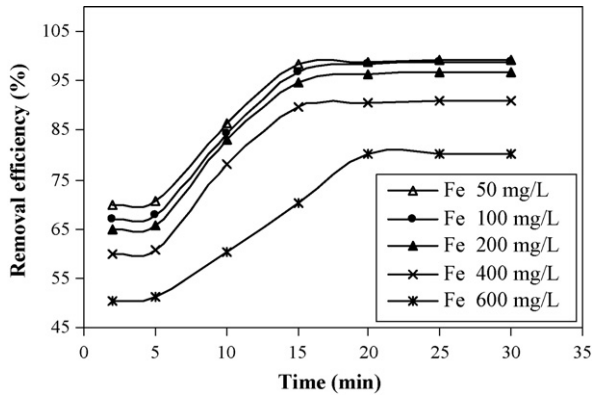
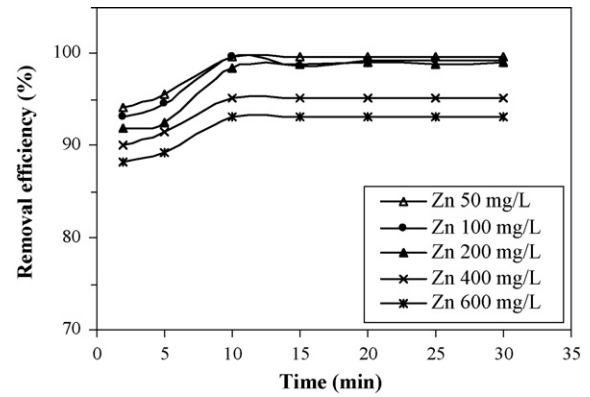


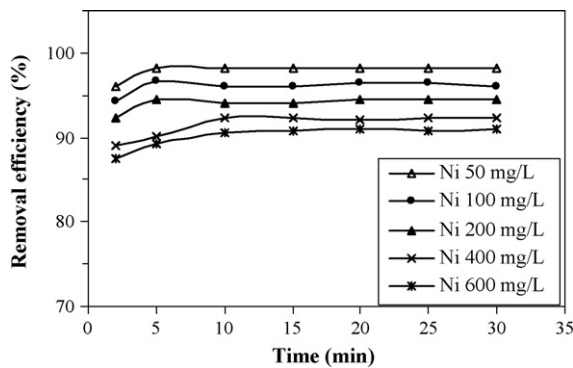
Fig. 9. Removal efficiency for various items: current density  $j = 11.55 \text{ mA/cm}^2$ , retention time  $t = 10 \text{ min}$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 8.7, conductivity  $\kappa = 4.571 \text{ mS/cm}$ .



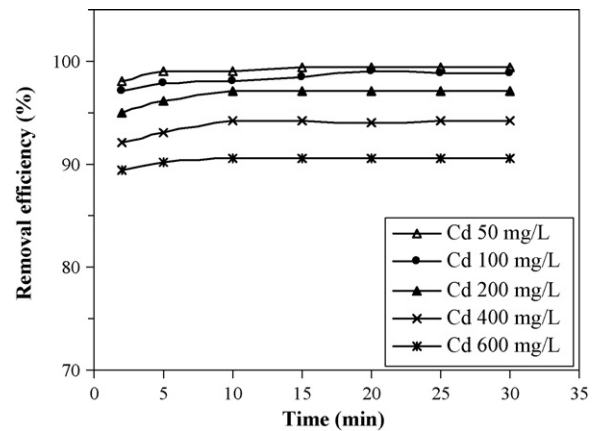
**Fig. 10.** Removal efficiency as a function of iron concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .



**Fig. 13.** Removal efficiency as a function of zinc concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .



**Fig. 11.** Removal efficiency as a function of nickel concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .



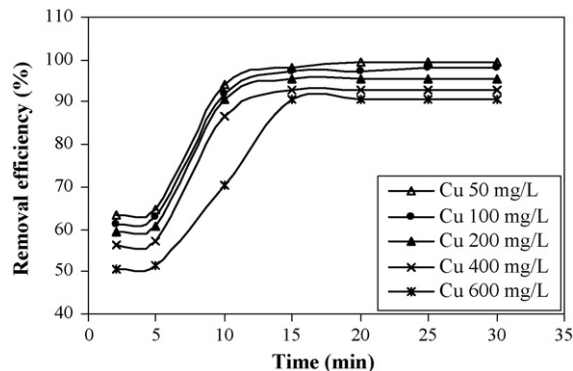
**Fig. 14.** Removal efficiency as a function of cadmium concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .

### 3.6.2. Kinetics of electrocoagulation of polymetallic solutions

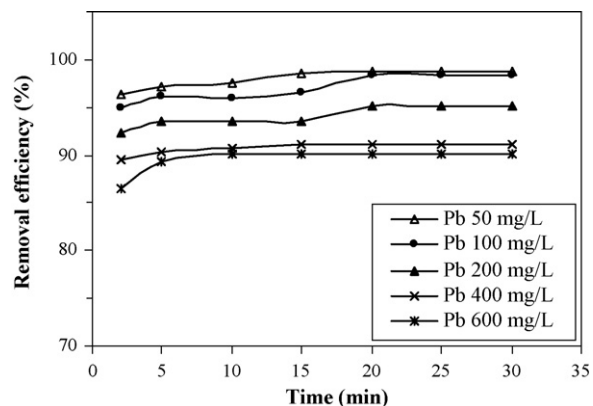
In order to see the effectiveness of EC–EF in the case of solution containing several metals, a mixture of  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ,  $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ ,  $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$  and  $\text{PbSO}_4$  in a same solution was studied. The operating conditions are the same ones with the first case. The experimental results are represented in Fig. 16.

Fig. 16 shows that the removal efficiencies of the polymetallic solutions containing sulfate is faster than the monometallic solutions (Figs. 10–15). Only one type of hydroxide particles found

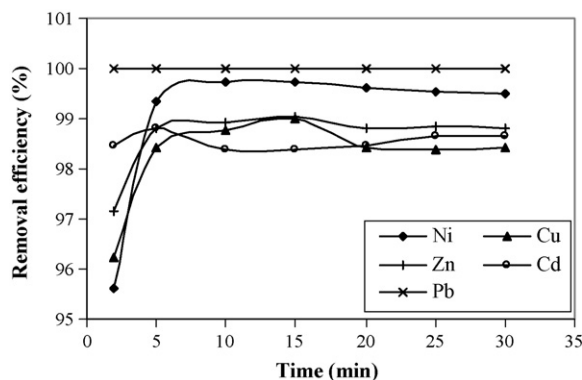
in monometallic solutions which is involved by gases, when in polymetallic solutions there were so many, and their numbers is equal to various metals found in the solution. We know that during the EC–EF functioning, its speed depends on the ascension effect between aggregates; so, the ascension effect activation is probably caused by various aggregates numbers; these results the increase



**Fig. 12.** Removal efficiency as a function of copper concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .



**Fig. 15.** Removal efficiency as a function of lead concentration and time in the presence of sulfates: current density  $j = 11.55 \text{ mA/cm}^2$ , interelectrode distance  $d = 1 \text{ cm}$ , initial pH 7.8, conductivity  $\kappa = 2.4 \text{ mS/cm}$ .



**Fig. 16.** Removal efficiency of various metals in a polymetallic solution:  $C_0 = 100$  mg/L, Current density  $j = 11.55$  mA/cm<sup>2</sup>, interelectrode distance  $d = 1$  cm, initial pH 7.8, conductivity  $\kappa = 2.4$  mS/cm.

in the efficiency for the polymetallic solutions compared to the monometallic ones.

It should be noted that the removal efficiencies were around 98.5% for all metals. For PbSO<sub>4</sub>, we did not record any remaining traces.

### 3.6.3. In the presence of acetates

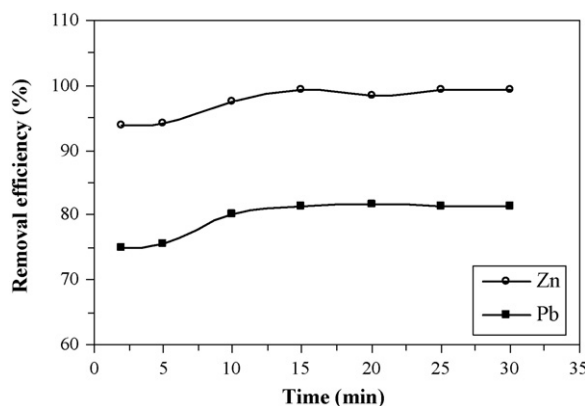
A model solution of Zn(CH<sub>3</sub>COO)<sub>2</sub> and Pb(CH<sub>3</sub>COO)<sub>2</sub> was used for conducting the experiments. All solutions were prepared using distilled water with the following characteristics:  $C_0 = 100$  mg/L, initial pH 7.5, conductivity  $\kappa = 2.4$  mS/cm.

The experimental results are represented in Fig. 17.

Fig. 17 shows that the kinetics of EF process is fast. We also note that the effectiveness of treatment of Zn(CH<sub>3</sub>COO)<sub>2</sub> is 97.52% and Pb(CH<sub>3</sub>COO)<sub>2</sub> is 80.06% after 10 min.

The removal efficiencies for zinc are almost the same ones in presence of sulfates and acetates (Figs. 15 and 17), but the difference was observed for lead.

For lead, in presence of acetates and with the basic pH (7.6), lead acetate can be formed in the solution, not very soluble to this pH, and more probably can be adsorbed on certain points of electrodes surfaces. This can cause a blocking of the surface. There will be no more electrochemical reactions (water electrolysis), which make the process inefficient comparatively to the solution containing sulfates.



**Fig. 17.** Removal efficiency of various metals in the presence of acetates:  $C_0 = 100$  mg/L, current density  $j = 11.55$  mA/cm<sup>2</sup>, interelectrode distance  $d = 1$  cm, initial pH 7.5, conductivity  $\kappa = 2.4$  mS/cm.

## 4. Conclusions

The results obtained in this study suggest an opportunity for the application of electrocoagulation–electroflotation technology. It can be concluded that the treatment of wastewater by EC–EF is effective based on the following:

- The application of EC–EF to a synthetic wastewater, consisted of drinking water and silica gel ( $\phi_{50} < 25$   $\mu$ m) showed a removal turbidity of 89.54% at a current density  $j = 11.55$  mA/cm<sup>2</sup>, retention time  $t = 10$  min, interelectrode distance  $d = 1$  cm, initial pH 7.6, conductivity  $\kappa = 2.1$  mS/cm.
- Based on the characteristics of particles in simulated wastewater ( $\phi_{50} < 25$   $\mu$ m), EC–EF is applicable to the treatment of industrial wastewater.
- Under the optimal operating conditions (current density  $j = 11.55$  mA/cm<sup>2</sup>, interelectrode distance  $d = 1$  cm, retention time  $t = 10$  min), removal efficiencies for various items of the textile wastewater used were as follows: BOD<sub>5</sub> (83%), COD (68%), turbidity (81.56%), SS (86.5%) and color (>92.5%).
- For concentrations ranging from 50 to 600 mg/L and initial pH of 7.8, the process of EC–EF functions for all studied metals and the removal efficiency decreased from 99% to 70%. From 50 to 400 mg/L, it can be seen that the rate of removal of all metals is high (90–99%) and the kinetics of electrocoagulation is fast and does not exceed 5 min. Except for iron and copper, a time of 15 min is required to reach more than 90%.
- The efficiency of treatment of the polymetallic solutions containing sulfates is faster than the monometallic solutions.
- Anions of sulfates and acetates contained in wastewater influence neither the kinetics nor the effectiveness of removal of different metal ions.

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## References

- [1] A. Willcock, M. Brewster, W. Tinchler, Using electrochemical technology to treat textile wastewater, three case studies, *Am. Dye Stuff Rep.* (1992) 15–22.
- [2] J.S. Do, M.L. Chen, Decolorization of dye-containing solutions by electrocoagulation, *J. Appl. Electrochem.* 24 (1994) 785–790.
- [3] S.H. Lin, M.L. Chen, Treatment of textile wastewater by electrochemical methods for reuse, *Water Res.* 31 (1997) 868–876.
- [4] J. Thampi, R. Paul, Azo dye effluent treatment: an environmental challenge—(Part-I), *Colourage* (1997) 47–49.
- [5] J.F. Blais, S.G. Dufresne, State of the art of technologies for metal removal from industrial effluents, *Rev. Sci. Eau* 12 (4) (2000) 687–711.
- [6] N. Adhoum, L. Monser, N. Bellakhal, J. Belgaied, Treatment of electroplating wastewater containing Cu<sup>2+</sup>, Zn<sup>2+</sup> and Cr(VI) by electrocoagulation, *J. Hazard. Mater. B* 112 (2004) 207–213.
- [7] J. Ge, J. Qu, P. Lei, H. Liu, Newbipolar electrocoagulation–electroflotation process for the treatment of laundry wastewater, *Sep. Purif. Technol.* 36 (2004) 33–39.
- [8] E. Vorobiev, O. Larue, C. Vu, B. Durand, Electrocoagulation and coagulation by iron of latex particles in aqueous suspensions, *Sep. Purif. Technol.* 31 (2003) 177–192.
- [9] X. Chen, G. Chen, P.L. Yue, Separation of pollutants from restaurant wastewater by electrocoagulation, *Sep. Purif. Technol.* 19 (2000) 65–76.
- [10] M. Kobya, E. Senturk, M. Bayramoglu, Treatment of poultry slaughterhouse wastewaters by electrocoagulation, *J. Hazard. Mater. B* 133 (2006) 172–176.
- [11] P.R. Kumar, S. Chaudhari, K.C. Khilar, S.P. Mahajan, Removal of arsenic from water by electrocoagulation, *Chemosphere* 55 (2004) 1245–1252.
- [12] N. Bektaş, H. Akbulut, H. Inan, A. Dimoglo, Removal of phosphate from aqueous solutions by electrocoagulation, *J. Hazard. Mater. B* 106 (2004) 101–105.
- [13] M. Murugananthan, G.B. Raju, S. Prabhakar, Removal of sulfide, sulphate and sulfite ions by electrocoagulation, *J. Hazard. Mater. B* 109 (2004) 37–44.
- [14] A.E. Yilmaz, R. Boncukcuoglu, M.M. Kocakerim, B. Keskinler, The investigation of parameters affecting boron removal by electrocoagulation method, *J. Hazard. Mater. B* 125 (2005) 160–165.

- [15] A.S. Kopalal, U.B. Ogutveren, Removal of nitrate from water by electroreduction and electrocoagulation, *J. Hazard. Mater. B* 89 (2002) 83–94.
- [16] P. Gao, X. Chen, F. Shen, G. Chen, Removal of chromium(VI) from wastewater by combined electrocoagulation–electroflotation without a filter, *Sep. Purif. Technol.* 43 (2004) 117–123.
- [17] M.N. Pons, A. Alinsafi, M. Khemis, J.P. Leclerc, A. Yaacoubi, A. Benhammou, A. Nejmeddine, Electrocoagulation of reactive textile dyes and textile wastewater, *Chem. Eng. Process.* 44 (2005) 461–470.
- [18] M. Kobya, O.T. Can, M. Bayramoglu, M. Sozbir, Operating cost analysis of electrocoagulation of textile dye wastewater, *Sep. Purif. Technol.* 37 (2004) 117–125.
- [19] N. Daneshvar, H. Ashassi-Sorkhabi, M.B. Kasiri, Decolorization of dye solution containing Acid Red 14 by electrocoagulation with a comparative investigation of different electrode connections, *J. Hazard. Mater. B* 112 (2004) 55–62.
- [20] A.E. Greenberg, L.S. Clesceri, A.D. Eaton, *Standard Methods for the Examination of Water and Wastewater*, 18th ed., APHA, 1992.
- [21] K. Scott, *Electrochemical Processes for Clean Technology*, The Royal Society of Chemistry, Cambridge, 1995, pp. 12–62.
- [22] G. Chen, X. Chen, P.L. Yue, Electrocoagulation and electroflotation of restaurant wastewater, *J. Environ. Eng.* (2000) 858–863.
- [23] M. Bayramoglu, M. Eyvaz, M. Kobya, Treatment of the textile wastewater by electrocoagulation: economical evaluation, *Chem. Eng. J.* 128 (2007) 155–161.
- [24] M. Kobya, O.T. Can, M. Bayramoglu, Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes, *J. Hazard. Mater.* 100 (2003) 163–178.
- [25] M. Bayramoglu, M. Kobya, O.T. Can, M. Sozbir, Operating cost analysis of electrocoagulation of textile dye wastewater, *Sep. Purif. Technol.* 37 (2004) 117–125.
- [26] O.T. Can, M. Kobya, E. Demirbas, M. Bayramoglu, Treatment of the textile wastewater by combined electrocoagulation, *Chemosphere* 62 (2006) 181–187.
- [27] M. Kobya, E. Demirbas, O.T. Can, M. Bayramoglu, Treatment of levafix orange textile dye solution by electrocoagulation, *J. Hazard. Mater.* 132 (2006) 183–188.
- [28] M.Y.A. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC)-science and applications, *J. Hazard. Mater.* 84 (2001) 29–41.
- [29] M.Y.A. Mollah, P. Morkovsky, J.A.G. Gomes, M. Kesmez, J.R. Parga, D.L. Cocke, Fundamentals, present and future perspectives of electrocoagulation, *J. Hazard. Mater.* 114 (2004) 199–210.
- [30] P.K. Holt, G.W. Barton, C.A. Mitchell, The future for electrocoagulation as a localised water treatment technology, *Chemosphere* 59 (2005) 355–367.
- [31] K. Rajeshwar, J.G. Ibanez, G. Swain, *Electrochemistry and the environment*, *J. Appl. Electrochem.* 24 (1994) 1077–1091.
- [32] G. Chen, *Electrochemical technologies in wastewater treatment*, *Sep. Purif. Technol.* 38 (2004) 11–41.
- [33] M.Y.A. Mollah, S.R. Pathak, P.K. Patil, M. Vayuvegula, T.S. Agrawal, J.A.G. Gomes, M. Kesmez, D.L. Cocke, Treatment of orange II azo-dye by electrocoagulation (EC) technique in a continuous flow cell using sacrificial iron electrodes, *J. Hazard. Mater.* 109 (2004) 165–171.
- [34] M. Khemis, J.P. Leclerc, G. Tanguy, G. Valentin, F. Lapique, Treatment of industrial liquidwastes by electrocoagulation: experimental investigations and an overall interpretation model, *Chem. Eng. Sci.* 61 (2006) 3602–3609.
- [35] F. Edeline, *l'épuration physico-chimique des eaux, théorie et technologie*, Cebedoc Editeur Lavoisier Tec. et Doc, 1996.
- [36] A. Alinsafi, M. Khemis, M.N. Pons, J.P. Leclerc, A. Yaacoubi, A. Benhammou, A. Nejmeddine, Electrocoagulation of reactive textile dyes and textile wastewater, *Chem. Eng. Process.* 44 (2005) 461–470.
- [37] A. Livingston, S.F. Zhang, Hydrodynamic behaviour of three-phase (gas–liquid–solid) airlift reactors, *Chem. Eng. Sci.* 48 (1993) 1641–1654.
- [38] C. Freitas, M. Fialovia, J. Zahradnik, A. Teixeira, Hydrodynamic model for three-phase internal- and external-loop airlift reactor, *Chem. Eng. Sci.* 54 (1999) 5253–5258.
- [39] C. Vial, E. Camarasa, S. Poncin, G. Wild, N. Midoux, J. Bouillard, Study of hydrodynamic behaviour in bubble columns and external loop airlift reactors through analysis of pressure fluctuations, *Chem. Eng. Sci.* 55 (2000) 2957–2973.
- [40] N. Daneshvar, A. Oladegaragoze, N. Djafarzadeh, Decolorization of basic dye solutions by electrocoagulation: an investigation of the effect of operational parameters, *J. Hazard. Mater.* 129 (2006) 116–122.
- [41] X. Chen, G. Chen, P.L. Yue, Investigation on the electrolysis voltage of electrocoagulation, *Chem. Eng. Sci.* 57 (2002) 2449–2455.
- [42] C. Escobar, C.S. Salazar, M.I. Toral, Optimisation of the electrocoagulation process for the removal of copper, lead and cadmium in natural waters and simulated wastewater, *J. Environ. Manag.* 81 (2005) 384–391.
- [43] R.G. Casqueira, M.L. Torem, Flotation applied to the removal of heavy metals, *CETEM/MCT Environ. Technol.* 28 (2003) 1–74.
- [44] A. Khelifa, S. Moulay, A.W. Naceur, Treatment of metal finishing effluent by the electroflotation technique, *Desalination* 181 (2005) 27–33.