

Treatment of Surface Treatment Effluents by Electrocoagulation Process Using Aluminium Electrodes

Soukaina Namoussi^{1*}, Chaimaa Merbouh¹, Mohamed Kabriti^{1,2},
Abdelmottalib Nahli¹, Ayoub Naamane¹, Mohamed Chlaida¹, Nadia Iounes¹

¹ Laboratory of Ecology and Environment (LEE), Faculty of Sciences Ben M'Scik, University Hassan II, BP 7955-Sidi Othmane, Casablanca, Morocco

² National Laboratory for Studies and Pollution Monitoring (LNESP), Rabat, Morocco

* Corresponding author's e-mail: soukaina.env@gmail.com

ABSTRACT

The surface treatment industry generates effluents with a high load of highly toxic chemicals which must be treated under increasingly stringent regulation. The aim of this study was to treat the effluents of surface treatment unit of an aeronautical industry by the electrocoagulation process using aluminium electrodes. This process is used to study the performance to remove colloidal load, significant amount of oxidizable material and high levels of various metal elements (Cr, Fe, Zn, Cu and Al) from these effluents, under optimum conditions of pH 7, 8.6A of current intensity and 60 min of application. The electrocoagulation process was found to be effective in reducing turbidity (97.12%), COD (97.5%), SS (97.84%) and conductivity (96.82%), hexavalent chromium (99.99%), Zn (96.82%), Cu (94.3%), Iron (99.9%), Al (91.96%). The treated effluent conformed to the Moroccan standards of surface treatment discharge.

Keywords: Electrocoagulation, aluminium electrodes, surface treatment, effluents, pollutants

INTRODUCTION

The industrial sector is considered to be one of the most polluting in terms of discharges into aquatic environments, because it uses large amounts of water in manufacturing processes as well as chemicals through various commercial formulations, which could damage the local sanitation network as well as the equipment of the urban wastewater treatment plants to which these industries are connected. There is also a risk of contamination of water resources, both surface and ground (Bouderka et al., 2016) or even marine resources (Ayah et al., 2015; Siba et al., 2018) and coastal areas (Izougarhane et al., 2016). Among industrial sectors, the surface treatment sector (ST), because they use many chemicals, in particular metals and organic substances, known to be toxic to humans and the environment (Elise Euvrard., 2017). The major problem with this type of effluent lies in the fact

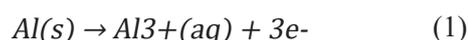
that they are not always treated by the majority of industries and can affect the local sanitation network or even the equipment of centralized wastewater treatment plants. Indeed, the results obtained by the characterization show that this effluent contains hexavalent chromium, well known for its toxicity (Brignon et al., 2015). The effluent has other characteristics, such as a high concentration of COD, suspended solids (SS) and heavy metals (Al, Zn, Cu, Fe, etc.). Because of their environmental toxicity (Rabia et al., 2019), these industrial effluents must be treated in accordance with the limit values set by national standards for discharge into the sewer or into the environment (AVLRBG.,2010: loi n° 10–95.,1995). The objective of this study was to apply a treatment of the effluents generated by this activity by electrocoagulation (EC). This electrochemical technique is commonly used in the treatment of water and industrial effluents (Mickova et al., 2015), the treatment of surface

water (Bejjany et al., 2017), the treatment of sulphated salinity and the associated toxicity of mining effluents (FOUDHAILI, 2019), the elimination nickel in wastewater (Jerroumi et al., 2019), treatment of textile effluents (Yibor et al., 2021), treatment of peat water (Rusdianasari et al., 2019), purification of river water (Suryaningsih et al., 2021). Electrocoagulation technology is a simple, and more economical than conventional chemical treatment (Crini et al., 2019) contains less water and is more stable than those obtained by using the classical chemical coagulation method (Tlaiaa et al., 2020), because it is able to simultaneously eliminate a very wide variety of pollutants. These include, inter alia, colloidal particles, suspended particles, turbidity, heavy metals (El-Naas et al., 2013). This process is characterized by simple equipment and easy operation (Copra et al., 2012). The factors influencing the performance of the EC process depend on various parameters, such as the electrode material type, current intensity, inter-electrode distance, pH and treatment time (Alam et al., 2021).

Electrocoagulation mechanisms

The electrocoagulation process is based on the principle of soluble anodes in an electrocoagulation reactor which is composed of an electrochemical cell, in which cathodic metal electrodes immersed in the solution to be treated and connected externally to a direct current supply and sacrificial metal anodes are used to treat wastewater (Fayad, 2018).

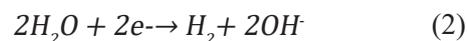
For aluminum oxidation in an electrolytic system, aluminum hydroxide, $Al(OH)_n$, is produced, where $n = 3$. At the anode: electro-dissolution of the anode leads to the release of the soluble Al^{3+} cations in the clay suspension according to the mechanism of the reaction (equation 1) (Bejjany et al., 2017).



The Al^{3+} ions thus produced undergo spontaneous hydrolysis reactions leading to the formation of several monomeric species as $Al(OH)_3$, and optionally its polymers. They are coagulants, the pH of which ranges between 4 and 9 (Hakizimana, 2017).

Other reactions, called secondary reactions, may take place at the anode, particularly the

generation of oxygen when the anodic potential is high. At the cathode: The water reduction reaction occurs, which results in the generation of hydrogen bubbles on its surface (equation 2):



Freshly formed amorphous $Al(OH)_3$ hydroxides have large surfaces which are beneficial for rapid adsorption different soluble pollutants and trapping of colloidal particles (Bharath et al., 2018). In this work, the electrocoagulation technique was used to decontaminate raw effluent of the surface treatment process of aeronautical parts.

MATERIALS AND METHODS

During this study, the experiments were performed in the laboratory using a rectangular electrocoagulation cell with a capacity of 1.5 L and a size of 15 cm long, 10 cm wide and 12 cm high (Figure 2). The electrodes are made of aluminium with a width of 8 cm, a length of 12.5 cm and a thickness of 1 mm, after several studies, it turned out that the treatment efficiency changes with the change of the distance between the electrodes. The work of Copra et al. (2012) showed that the treatment is effective with an inter-electrode distance of 2.5 cm while Balark et al. (2019) found that the distance of 2.5 cm is the most efficient. In turn, the work of Dindas et al. (2020) showed that the inter-electrode distance of 2 cm is the most efficient. This is why the electrodes were separated in parallel with a distance of 2 cm one from the other. The electrodes are connected to a digital DC power supply, capable of supplying an adjustable current intensity in the range of 0–10 A (Long Wei Instruments). The samples treated by electrocoagulation were collected after 15 minutes of decantation and filtered through the filtration equipment to remove undissolved flocs.

In order to achieve the objectives, set in terms of effective and complete treatment of pollutants, the influence of the operating parameters of this electrocoagulation system (amperage, electrocoagulation time and pH) and the effluent quality status were monitored through the measurements of physicochemical parameters (COD, SS, conductivity, turbidity, heavy metals (Cr^{6+} , Zn, Al, Fe, Cu)). During this

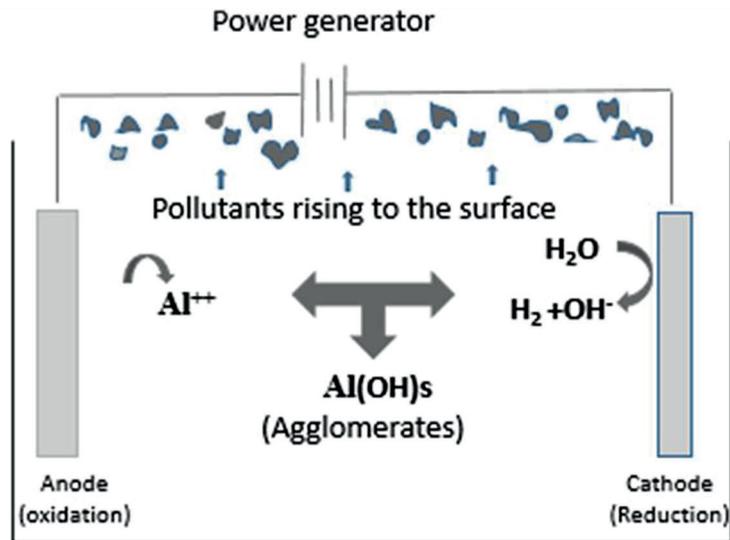


Fig. 1. Principle of electrocoagulation

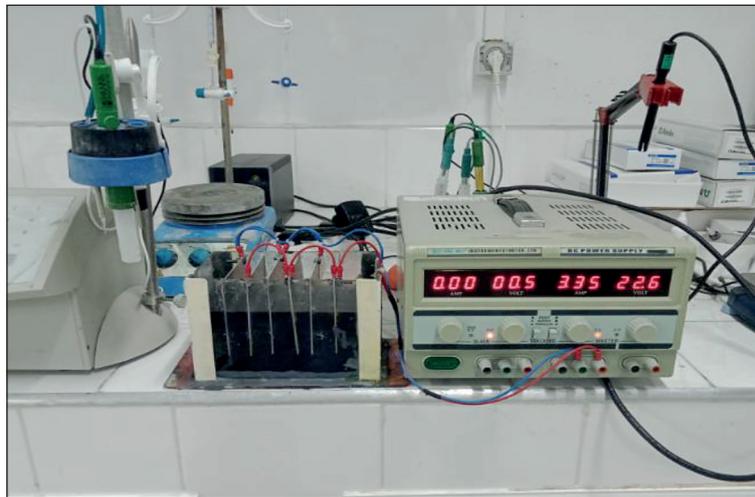


Fig. 2. Experimental system of electrocoagulation

study, the samples were taken from the four production lines in full operation and during the various production cycles. This sampling is carried out in a place where there is sufficient turbulence / agitation to ensure the homogeneity of the sample, an average of the 3 values obtained following the measurements and analysis before each experimental test was carried out. Table 1 presents the monitoring and measurement methods used in this study.

These electrocoagulation treatment tests were conducted under variable conditions of pH (4, 7 and 10), current intensity (2.8 A, 5.7 A and 8.6 A) and for an overall period of 60 min with different treatment controls at 20 min, 30 min, 40 min and 60 min.

RESULTS AND DISCUSSION

Physicochemical characteristics of the raw effluent

The results of characteristics of the investigated wastewater (conductivity, turbidity, pH, COD, SS, Cr, Zn, Al, Fe and Cu) are shown in Table 2.

The analysis of the effluent composition revealed a colloidal pollution (turbidity=98 NTU), a significant oxidizable charge (COD= 587 mg/l), suspended solids (663 mg/l), a mixture of different metallic (Zn, Al, Fe, Cu) and a high chromium concentration. These effluents do not comply with the Moroccan standard for surface treatment discharges (AVLRBG., 2010).

Table 1. Parameters, materials and method

Parameter	Material	Device reference or analytical method
Temperature (°C)	Thermometer	TH CA 004
pH	pH meter	pH 3310 SET 2
Chemical Oxygen Demand "COD" (mg·L ⁻¹)	Thermoreactor with kits type DCO10119	Methode DIN ISO 15705
Turbidity (NTU)	Turbidity meter	EUTECH TN-100
Suspended Substances "SS" (mg·L ⁻¹)	Filtration on a Wattman GFC filter	AFNOR, 1999-NF EN 872
Conductivity (µs/Cm)	Conductivity meter	HI86303
Heavy metals (Zn, Al, Fe, Cu) (mg·L ⁻¹)	Spectrophotometry with standard kits	WTW Spectroflex 6100
Cr ⁶⁺	Automatic titrator	HI900 series titration systems

Table 2. Characteristics of the raw effluent

Parameters	Values	Moroccan limit value of dumping of the surface treatment industry (AVLRBG, 2010)
pH	3	6–6,8
Temperature (°C)	25	≤30
Conductivity (µs/Cm)	4560	2700
COD (mg/l)	587	500
Turbidity (NTU)	98	30
Suspended Substances «SS (mg/l)	663	50
Cr ⁶⁺ (mg/l)	10020	0,1
Zn (mg/l)	4.91	10
Al (mg/l)	1.8	10
Fe (mg/l)	4.83	20
Cu (mg/l)	1.47	4

Effects of physicochemical parameters on the electrocoagulation process

Effect of pH

It has been established in previous studies that pH has a considerable effect on the efficiency of the electrocoagulation process (Sharma et al., 2021). Moreover, as observed by other investigators (Igwegbe et al., 2021) it was found that the pH of the medium changed during the process depending on the type of electrode material used and the initial pH. The performance of electrocoagulation process is known and performs an important role in the removal efficiency of pollutants (Madi et al., 2019; Tchamangoa, 2018). In this first experiment, the pH adjustments were made using 1N NaOH or HCl solutions under an initial current intensity of 5.7 A. The choice of current intensity referred to previous work, in particular that of Bazrafshan et al. (2015) who obtained a good treatment efficiency by electrocoagulation with a current intensity of 5 A or those of Parsa et al. (2011) at 6.329 A.

The removal of chromium, COD, suspended solids, conductivity, turbidity and heavy metals

with aluminium electrode using different pH is illustrated in Figures 3 and 4.

According to the results obtained, the elimination rates of the targeted pollutants gradually increase with the time of treatment and prove to be higher at pH 7 than for the other pH. These rates reach, after 60 minutes of treatment, 91.12% for the turbidity, 76.82% for the COD, 76.82% for conductivity, 77.54% for the SS, 86.82% for chromium and 90% for copper.

These elimination rates recorded during the conducted experiment agree with those reported by Secula et al. (2012) and Gomathi et al. (2011) who, under the same pH conditions, obtained a good percentage of elimination of sulphates (97%) and SS matter (80%). This result would be linked to an influence of the pH on the state of solubility of the products formed and on other species in solution. Indeed, it has been proven that this parameter has an effect on the dissolution of the electrodes (Al Anbari et al. (2012); Zaroual et al. (2006) at pH 6.8. This result agrees with the previously published works (Fayad, 2018; Tezcan, 2015) and suggests that electrocoagulation can act as a pH buffer.

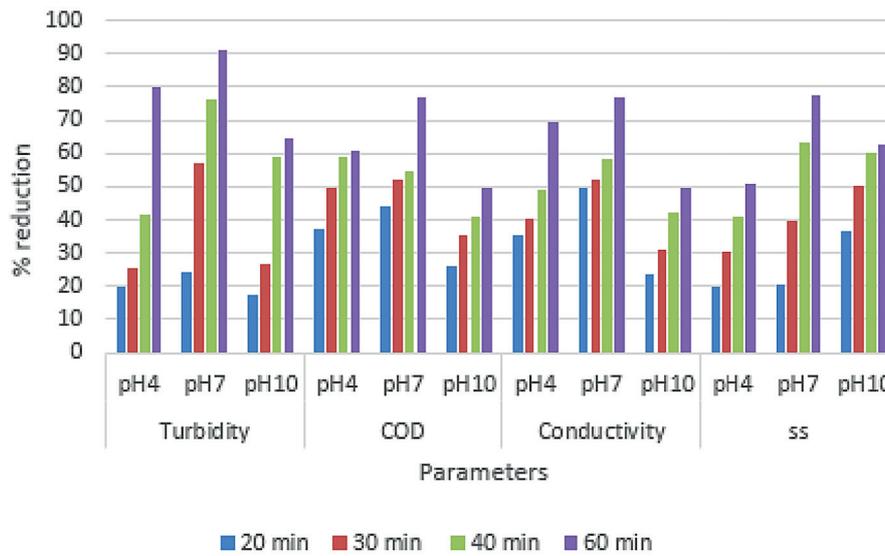


Fig. 3. Effect of pH on the reduction rate of Turbidity, COD, Conductivity and SS of effluents treated at 5.7 A

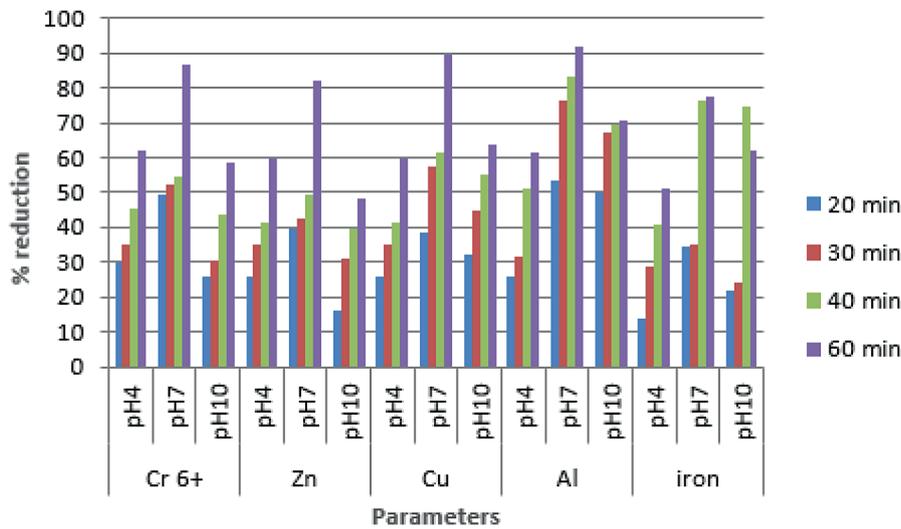


Fig. 4. Effect of pH on the reduction rate of heavy metals of the effluents treated at 5.7 A

Effect of current intensity

In order to determine the optimum current intensity to the removal percentage of different pollution parameters: COD, conductivity, turbidity, suspended substances and heavy metals (Cr, Zn, Al, Fe, Cu) in the studied effluent, the experiments were carried out at pH 7 and at different intensities 2.8A, 5.7A and 8.6 A. The results obtained (Figures 5 and 6) show that the rate of abatement of pollutants in treated effluents increases along with intensity and treatment duration. These rates reach, after 60 minutes and at a current intensity of 8.6 A, up to 97.12% for turbidity, 97.5% for COD, 97.84% for SS and 96.82% for conductivity. Similarly, high reduction rates are recorded for

metal pollutants, particularly chromium and iron, reaching up to 99.9% after 60 minutes of processing and under a current intensity of 8.6 A.

The results obtained from the conducted experiments, in particular with regard to metallic contaminants, are consistent with those obtained by several authors. Chen et al. (2004) and Adhoun et al., (2004) reported that the effectiveness of treatment, by electrocoagulation, is mainly affected by current intensity. They showed that certain metal-based dyes are eliminated at 98.5% with an increase in the current voltage. At the same time, metal ions decreased in the treated effluent with removal efficiency of 99%, 99% and 83% for Cu^{2+} , Zn^{2+} and Cr^{6+} , respectively. Finally, the results obtained by Parsa et al. (2011) go in

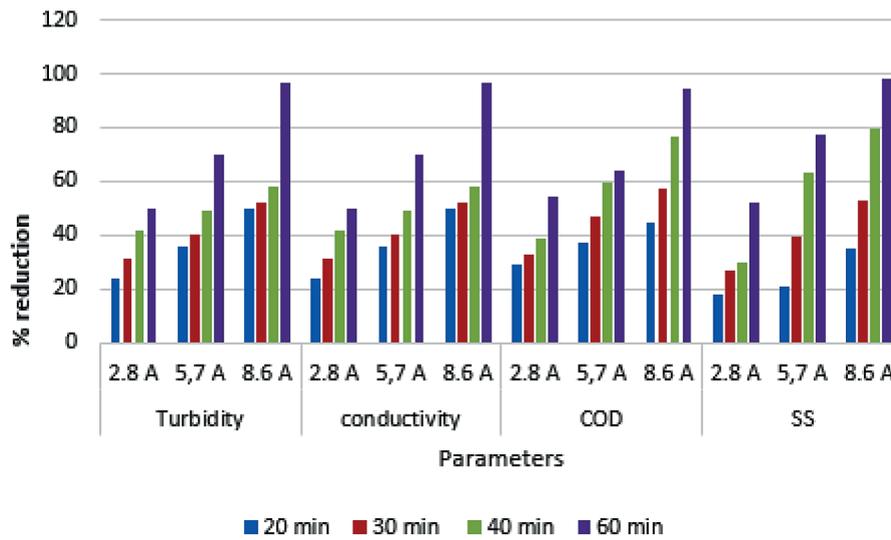


Fig. 5. Effect of current intensity on the reduction rate of Turbidity, DCO, Conductivity and SS of effluents treated at pH 7

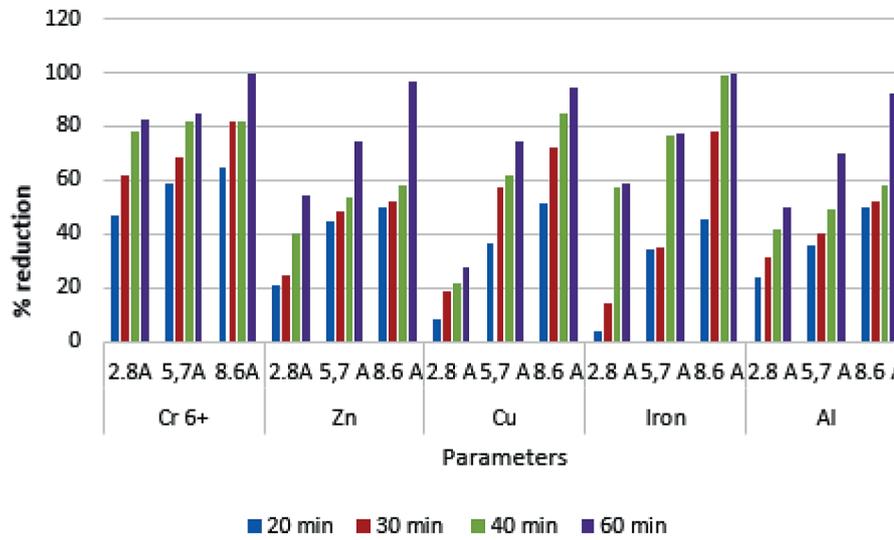


Fig. 6. Effect of current intensity on the reduction rate of heavy metals of effluents treated at pH 7

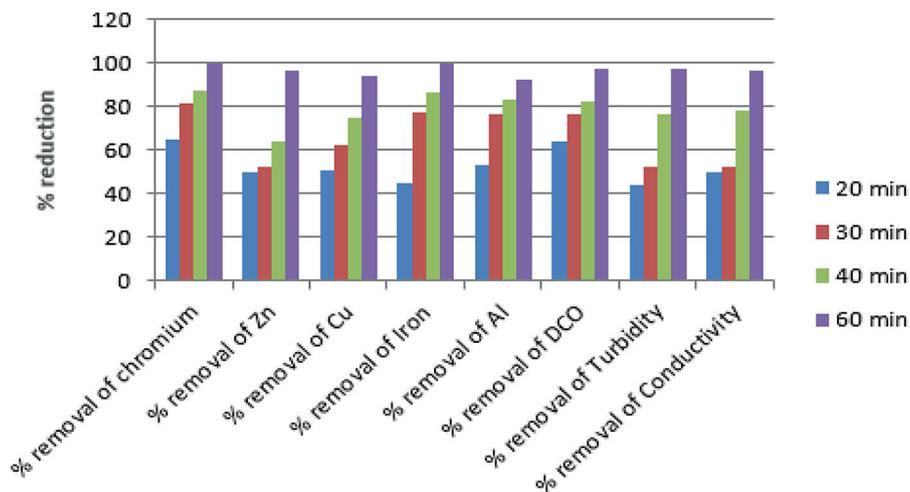


Fig. 7. Effect of processing time under optimal pH and current intensity conditions (I=8.6 A and pH 7)

Table 3. Physicochemical results of the treated wastewater under optimal electrocoagulation conditions and their reduction.

Parameters	Raw wastewater values	Waste water treated by electrocoagulation	Abatement (%)	Moroccan limit value of dumping of the surface treatment industry (AVLRBG, 2010)
Conductivity ($\mu\text{s}/\text{cm}$)	4560	145.08	96.82	2700
SS (mg/l)	663	18.96	97.15	50
Turbidity (NTU)	98	2.82	97.12	30
COD (mg/l)	587	17.31	97.05	500
Cr ⁶⁺ (mg/l)	10020	0.01	99.9	0.2
Zn (mg/l)	4.91	0.15	96.82	10
Al (mg/l)	1.8	0.144	91.96	10
Fe (mg/l)	4.83	0.004	99.9	20
Cu (mg/l)	1.47	0.083	94.3	4

the same direction with a removal efficiency of up to 91% for Cr⁶⁺ under optimal conditions of 6.329 A and a treatment time of 18 min.

Effect of processing time under optimal pH and current intensity conditions

The effectiveness of the removal of pollutants during electrocoagulation (EC) also depends on the duration of the treatment. Indeed, previous studies showed that the pollutant removal efficiency increases along with the EC time (Castañeda-Díaz et al., 2018; Hakizimana et al., 2017). In this third series of experiments, the optimum conditions of pH 7 and current intensity of 8.6A were ensured, whereas the treatment time varied between 20 mn, 30 mn, 40 mn and 60 mn.

The results obtained (Figure 7) show a significant increase in the abatement rate of the different pollutants when there is an extension of EC treatment time. For example, after 60 minutes of application of this technique, the elimination rates reached quite high values for global pollutants (97.5% for COD, 97.84% for SS, 97.12% for turbidity, 96.82% for conductivity) and metallic contaminants (up to 99.99% for Cr and Fe, 96.82% for Zn, 94.3% for Cu and 91.96% for Al). An increase in the operating time from 20 to 60 min in the treatment of the studied industrial wastewater by electrocoagulation has led to increase in the removal efficiencies of various pollutants: heavy metals, COD, turbidity, conductivity and suspended solids. These results are in agreement with the previous research, Tezcan and Eren (2015) achieved the highest removal percentages for Cd, Ni and Cu with 99.78%, 99.98%, 98.90% respectively, with a current intensity of 30 mA/cm², a pH of

7 and 90 minutes of electrocoagulation operation. Bazrafshan et al. (2015) determined that the percentage of chromium removal increased from 91% to 96%, as long as the treatment was between 20 and 60 minutes at pH 4 with a current intensity of 5A.

The results of the physicochemical analysis of the water treated under optimal conditions show that the values of all the parameters exhibit significant reduction rates of up to 99.9% (Table 3). At the same time, all the levels recorded for the other parameters remain well below the discharge limit values specific to the surface treatment industries (AVLRBG, 2010). Furthermore, adjusting the pH to a value of 7, allowed conforming this parameter to these same limit values and optimizing the EC conditions.

CONCLUSIONS

The analysis of raw effluents from the surface treatment chain of an aeronautical industry revealed a significant overall load (COD, SS and Turbidity) and in metals (Zn, Al, Fe, Cu and Cr) exceeding the limit values stipulated by the Moroccan standard specific to surface treatment discharges.

In this regard, an experimental process of electrocoagulation treatment was applied to its wastewater under different pH, current intensity and treatment time conditions. The results showed that the best rates of reduction of chromium (99.9%), Zn (96.82%), Cu (94.3%), iron (99.9%), Al (91.96%), DCO (97.5%), SS (97.84%) turbidity (97.12%) and conductivity (98%) are obtained under the optimum conditions of pH 7, a current intensity of 8.6 A and a reaction time of

60 minutes. At the same time, electrocoagulation allowed the recovery of treated water that largely met the limit values recommended by the Moroccan standards for surface-industry discharges. However, this treatment unfortunately generates sludge which must be studied, treated and highly valued for a zero discharge objective.

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