



Treatment of lixivate from Jebel Chakir-Tunis by electrocoagulation

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Received 26 October 2009; accepted 13 May 2010

ABSTRACT

The removal of organic pollutants from lixivate was experimentally studied using direct current electrocoagulation (EC) with aluminum electrodes. The effects of operating parameters such as current density, initial pH and electrolysis time were further studied in order to optimize conditions for the treatment of lixivate. Two different electrode connections (monopolar and bipolar) were examined for choosing the best alternative in order to intensify the performance of the process. It was observed that high removal capacity was better for monopolar connection than for bipolar connection. The subsequence of EC tests performed with Al electrodes showed that optimal operating conditions are an initial pH of 6; a current density 15 A cm^{-2} and EC time of 90 min. Treatment reduces chemical oxygen demand (COD) to 94%. The color removal efficiency reached 96%. Cost estimation was adopted and presented as well. Total operating costs for monopolar connection was evaluated at about $43.315 \text{ US\$ m}^{-3}$ for the optimal operating conditions. These findings might be useful in order to treat lixivate contaminated groundwater, rivers and grounds.

Keywords: Lixivate; Electrocoagulation; Electrode of aluminum; Chemical oxygen demand; Decolorization

1. Introduction

Lixivate is a solution of complex chemical composition particularly harmful to the natural environment and also to the threatening for human health [1,2].

It can mix in surface waters as in subterranean waters and then the soluble organic matter, the constituents inorganic as the heavy metals can be a source of pollutants. The biggest risk bound to the production of lixivate is the contagion of the groundwater. It

would consequently pollute the well waters to be consumed and thus deprive populations of a vital element for its survival.

Tunisia produces about $90,000 \text{ m}^3/\text{year}$ of lixivate [3]. It has high concentrations of organic compounds. Electrochemical treatment techniques attract a great deal of attention because of their versatility and environmental compatibility. They yield very effective removal of organic pollution.

Strong color of the wastewater effluents is particularly troublesome because of its negative visual impact. Conventionally, biological, chemical, and

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physical methods have been employed for the treatment of lixiviate. There are many processes to treat the contaminants of drinking water and industrial effluents, such as adsorption, precipitation, chemical degradation, photodegradation, biodegradation, chemical coagulation and electrocoagulation (EC).

The important advanced oxidation processes to remove pollutants from wastewater are Fenton systems ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$), ($\text{H}_2\text{O}_2/\text{Fe}^{3+}$) [4,5]; ozonation ($\text{O}_3/\text{H}_2\text{O}_2$) [6]; (O_3/UV) [7], homogeneous ultraviolet irradiation ($\text{H}_2\text{O}_2/\text{UV}$) [8], electro-Fenton and photoelectro-Fenton [9–12]. Tizaoui et al. [13] explored the treatment of lixiviate of Jebel Chakir by means of oxidizing action of $\text{H}_2\text{O}_2/\text{O}_3$. They obtained high reduction of COD and intensity color.

Bellakhal et al. [14] have treated olive oil mill wastewater by Electro-Fenton Process and have obtained good removal efficiencies. The adsorption process is one of the effective methods used to remove dyes from aqueous solutions [12]. Activated carbon is the most widely used as an adsorbent for organic matter removal [15] but it is too expensive.

Ziyang et al. [16] have applied the technique of ultra-filtration for the treatment of lixiviate, they have showed that this method permits to reduce the COD to 75%. Although these methods present disadvantage of being expensive because of their operating cost and/or the relatively high cost for the chemical reagent used.

EC process provides a simple, reliable and cost-effective method for the treatment of wastewater without any need of additional chemicals.

The main purpose of this work was to investigate the removal of color and cloudiness from lixiviate of dumping-ground of Jebel Chakir (south-west of Tunis, in Tunisia) by aluminum EC.

EC is based on the in situ formation of the coagulant as the sacrificial anode corrodes due to an applied current. In an electrochemical cell, aluminum cations are produced at the anode. Similarly to a conventional coagulation process these coagulant reagents destabilize the colloidal pollutants breaking the emulsion. The gases evolved at the electrodes cause flotation of the coagulated materials. The EC process is intrinsically associated with electroflotation (EF) since bubbles of hydrogen and oxygen are produced at the cathode and anode, respectively.

In this paper, the most effective operational parameters, two different electrode connections, initial pH, current densities and time of treatment on Jebel Chakir lixiviate were described in terms of reduction of COD and decolorization efficiency. Lixiviate was characterized before and after treatment. Evolution of % COD and of color intensity, has been studied and discussed.

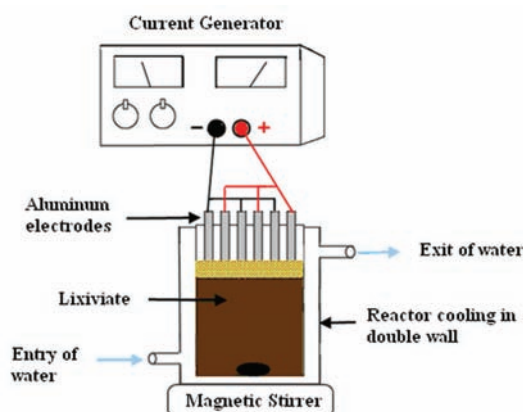


Fig. 1. Configuration of Electrolytic Cell.

2. Material and methods

2.1. Wastewater samples

Lixiviate samples were collected in plastic containers and cooled to 4°C, then transported to laboratory for analysis and electrochemical treatment. The initial pH of wastewater was previously adjusted to the electrochemical process and it was maintained by adding the required amount of H_2SO_4 [10^{-2} M].

2.2. Experimental device

The electrochemical reactor consists of a cylindrical vessel in the cooler system and two shafts supporting series of aluminum plates (60 mm × 40 mm × 0.5 mm of 99% purity) (Fig. 1). Three plates were connected as cathodes and three others as anodes in a monopolar mode. The volume of treated liquid each time was 300 cm³. A small hole was drilled into the cover serving as sampling port. The stirrer was used in the electrochemical cell in order to maintain an unchanged composition and to avoid the association of the flocks in the solution.

2.3. Methods of characterization

All other chemicals were of analytical grade and supplied by Fluka. All experiments were carried out at ambient temperature. The D.C. source was used to power supply (Metrix, model AX 322) the system with galvanostatic operational options for controlling the current density. Electrodes were washed with dilute HCl between the experiments.

A digital calibrated pH-meter (Metrohm, 744) was used to measure the pH of the lixiviate wastewater samples.

The effect of electrochemical treatment was determined by analysis of the chemical oxygen demand (COD) and color intensity of lixiviate at different time

Table 1
Physico-chemical characteristic of lixiviate of Jbel Chakir.

Parameters	Values
pH	8.8
Conductivity (mS.cm ⁻¹)	39.7
TSS (mg.L ⁻¹)	270
COD (mg _{O2} .L ⁻¹)	8200
D.O (390 nm)	8.8
Chloride (g.L ⁻¹)	26.98
Sulphate (g.L ⁻¹)	2.8
Cd (μg.L ⁻¹)	< 10
Cu (mg.L ⁻¹)	0.56
Fe (mg.L ⁻¹)	10.7
Pb (mg.L ⁻¹)	1.8

intervals. Analysis of COD was determined by the procedure described in the standard method [17]. The total suspended solids (TSS), was obtained by centrifugation then drying at 105°C [18]. The classic potentiometric method was used to determine chlorides.

The absorption spectrometry allowed the analysis of many inorganic materials (Fe, Cd, Cu, SO₄²⁻, etc.). The color intensity was determined by measuring the samples absorbance at 390 nm (UV-vis spectrophotometer, Beckman, DU 530). The samples were scanned in quartz cells with 1 cm optical path.

Samples of lixiviate were chemically analyzed before treatment. Some characteristics are presented in Table 1.

The table shows that lixiviate is a basic solution (pH = 8.8). It also indicates that the COD and color intensity have elevated concentration.

3. Results and discussion

3.1. Electrocoagulation

EC is an alternative approach which was widely used for the decolorization of different organic dyes [19,20] and wastewater from pasta and cookies [21]. This process includes the generation of coagulants inside by dissolving electrically either aluminum or iron ions in aluminum or iron electrodes, respectively. The metal ions generation takes place at the anode and the hydrogen gas is released from the cathode. The hydrogen gas would also help the flocculated particles float out of water. When a potential is applied from an external power source, the anode material undergoes oxidation while the cathode will be subjected to reduction of water [22]. EC of wastewater using aluminum electrodes takes place according to the following reactions [23,24]:

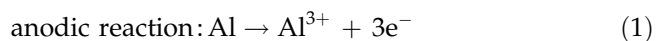
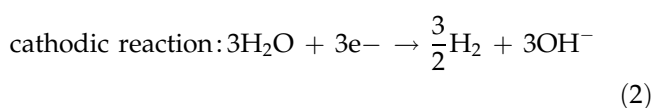
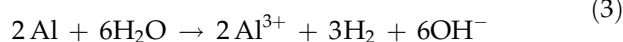


Table 2
Results of repeatability tests.

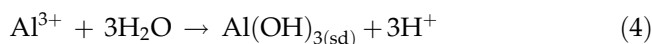
	Removal COD %	Absorbance %
Test 1	44.9	66
Test 2	46.4	66.2
Test 3	45.4	66.1
Test 4	46.8	66.5
Test 5	44	66.3
X	45.5	66.4
S _r	1.1	0.36
RSD(%)	2.4	0.55



overall reaction:



The generated Al³⁺ and OH⁻ ions by reactions (1) and (2) are immediately going to undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. These hydroxides/polyhydroxides/polyhydroxymetallic, finally transform into Al(OH)_{3(sd)} according to complex precipitation kinetics [25,26].



Al(OH)_{3(sd)}, which are formed can be soluble, insoluble or colloidal [27]. They have strong affinity with dispersed/dissolved organic compounds as well as the counterions to cause coagulation/adsorption [28,29]. Finally, the flocks formed are removed easily from aqueous medium by sedimentation or H₂ flotation [26,29,30].

Bubbles of hydrogen produced at the cathode may cause flotation of the coagulated materials. Then EC process is intrinsically associated with EF [11].

The performance of EC process in terms of reducing COD, decolonization efficiency, was studied.

3.2. Repeatability tests

In order to estimate the relative standard deviation [11–31] during the treatment of lixiviate of Jebel Chakir by EC, a series of five experiments was carried out with aluminum electrodes under the same experimental conditions ($J = 12 \text{ mA cm}^{-2}$, $t = 20 \text{ min}$, initial pH = 8). From the results presented in Table 2, we can conclude that EC makes it possible to carry out an electrochemical treatment of the lixiviate with a good repeatability

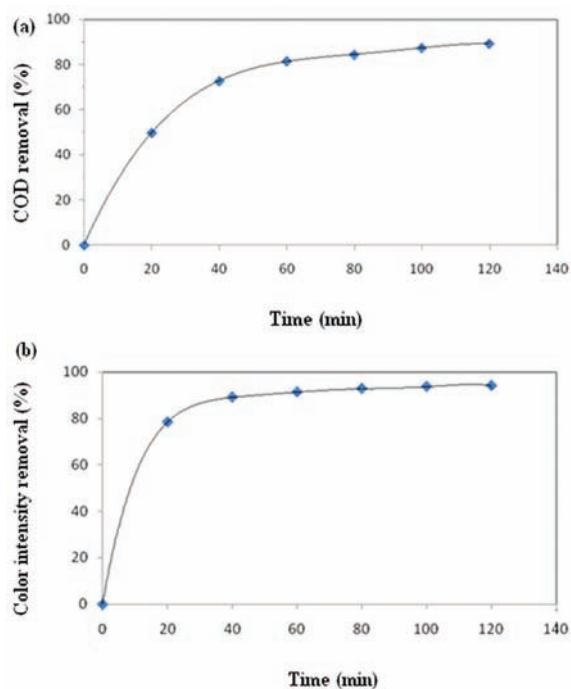


Fig. 2. Effect of the electrolysis time with aluminum electrode on the removal efficiency of COD (a) and color intensity (b) (initial pH = 8, $J = 16 \text{ mA}\cdot\text{cm}^{-2}$, $V = 300 \text{ cm}^3$).

(R.S.D. < 3%). Where R.S.D. is the reactive standard deviation expressed by the following equation:

$$\text{R.S.D.} = \left(\frac{\text{Sr}(x)}{\bar{x}} \right) \times 100, \quad (5)$$

where $\text{Sr}(x)$ is the standard of repeatability

$$\text{Sr}(x) = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n-1}}, \quad (6)$$

\bar{x} designates the arithmetic mean of variable X (Eq. (7)) and (n) the number of measurements carried out.

$$\bar{x} = \frac{\sum_{i=1}^n x_i}{n}. \quad (7)$$

3.3. COD and intensity color of EC process

In this section the efficiency of EC process on the reduction of COD and decolorization of lixiviate solution was evaluated by aluminum electrode. Fig. 2 shows the electrolysis time versus the color and COD removal efficiency at different times. It appears from

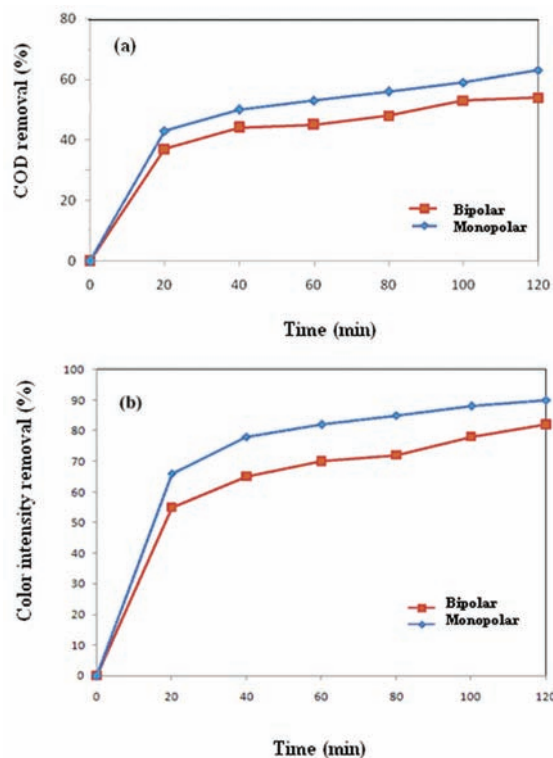


Fig. 3. Effect of mode on electrode connection on the removal efficiency of COD (a) and color intensity (b), as a function of time ($J = 12 \text{ mA}\cdot\text{cm}^{-2}$, initial pH = 8, $V = 300 \text{ cm}^3$).

the results that the 49.8% of COD and 70% of the color intensity were rapidly reduced just after 20 min. Consequently, EC can be an efficient method for organic pollutants removal from industrial wastewater.

3.4. Effect of electrode connection

In order to estimate the connection efficiency for EC treatment, two modes of connections were studied for choosing the best alternative so as to intensify the performance of the process. The performance of these processes in terms of reduction of COD and decolorization efficiency was compared. Results obtained for COD and color intensity removal for these two modes of connection using the same current density same current density ($J = 12 \text{ mA cm}^{-2}$) are shown in Fig. 3.

As observed by other investigators [32] for the monopolar connection, the applied voltage is the same in the electrolytic cell but for bipolar mode the resistances induce a high voltage in the electrodes.

3.5. Effect of initial pH

The pH of solution has important influence on the EC process. In this order, effect of pH on COD and color removal efficiency of lixiviate was investigated.

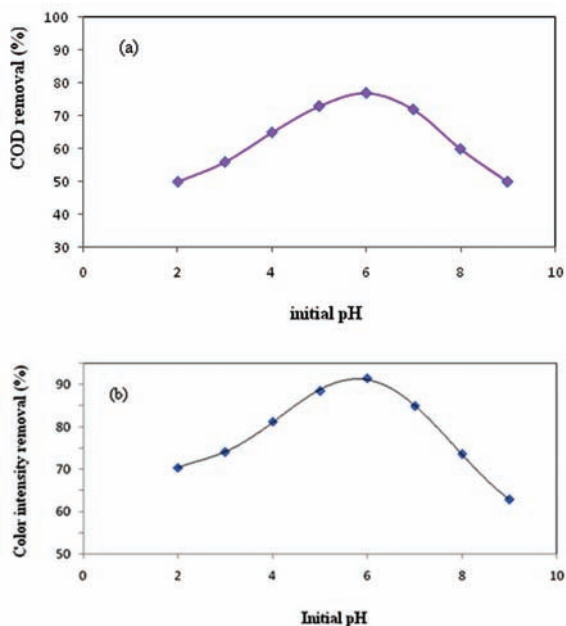


Fig. 4. The comparative removal COD (a) and decolorization (b) of solutions as function of initial pH. ($J = 12 \text{ mA}\cdot\text{cm}^{-2}$, $t = 60 \text{ min}$, $V = 300 \text{ cm}^3$).

Fig. 4 shows the comparative COD and decolorization of 300 cm^3 solutions at different pH.

The decrease of removal efficiency at a pH less than 5 and higher than 7 was observed by many investigators and was attributed to an amphoteric behavior of $\text{Al}(\text{OH})_3$ which leads to soluble Al^{3+} cations, when the initial pH is low and monomeric anions $\text{Al}(\text{OH})_4^-$, when the initial pH is high [33]. These soluble species are useless for water treatment. When the initial pH was kept in the range between 5 and 7, all aluminum cations produced at the anode formed polymeric species such as $\text{Al}_6\text{O}_{15}(\text{OH})_{24}^{3+}$, $\text{Al}_7\text{O}_{17}(\text{OH})_2^{4+}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ [34,35] and precipitated $\text{Al}(\text{OH})_3$ leading to a more effective treatment. As seen, the highest efficiencies have been obtained at a level pH near to 6 (Fig. 4). High removal capacity of COD (>70%) and color intensity (>90%) were observed.

As observed by other investigator [36], adsorption on $\text{Al}(\text{OH})_3$ flocks, followed by coagulation to form particles, mainly at $\text{pH} > 6.5$. The large surface areas of freshly formed amorphous $\text{Al}(\text{OH})_3$ flocks can adsorb soluble organic compounds and/or trap colloidal particles, which are separated from the aqueous solution.

A final pH increase occurs when the influent pH is low [23,24,37], it is due to hydrogen evolution at cathodes [23]. However, Chen et al. [37] explained this increase by the release of CO_2 from wastewater owing to H_2 bubble disturbance. Indeed, at low pH, CO_2 is over saturated in wastewater and can release

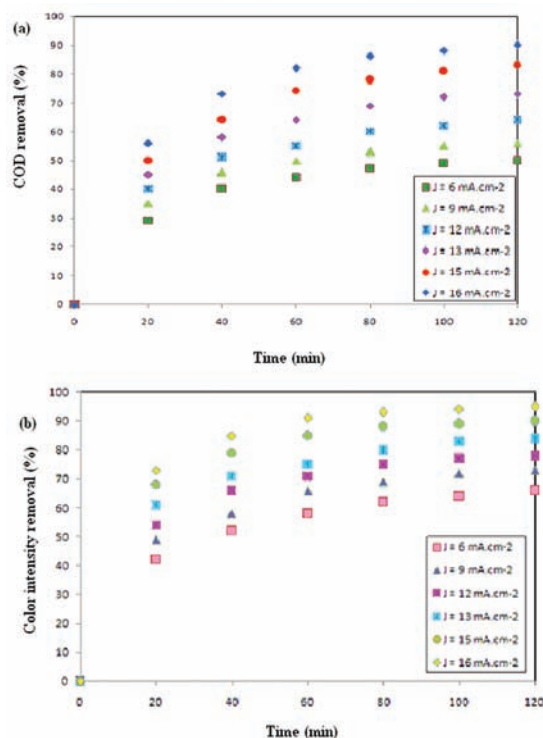


Fig. 5. Effect of current density on (a) the COD removal and (b) color intensity removal (initial pH = 8, $V = 300 \text{ cm}^3$).

during H_2 evolution, causing pH increase. In alkaline medium ($\text{pH} > 8$), the pH final does not varied very much and a slight drop was recorded [24,37]. The lixivate is an alkaline effluent (initial pH 8.8) and the optimum initial pH required to achieve high removal yield was adjusted to 5–7, the final pH after EC treatment reached 7.6–8.2.

3.6. Effect of current density

Fig. 5 shows the electrolysis time vs. the COD and the color removal efficiency at different CD. The current density varied from 6 to $16 \text{ mA}\cdot\text{cm}^{-2}$. As it can be seen, when current density and/or electrolysis time raises, the COD and the color removal efficiency are improved. The highest current ($16 \text{ mA}\cdot\text{cm}^{-2}$) produced the quickest treatment with 81% and 91% COD and color reduction respectively occurring after only 60 min.

It appeared that at lower current densities, less aluminum was released from the anode and hence the color removal efficiency was low. When the current density significantly increased, amount of flocks generated which in turn trapped the organic matter, led to the improvement of the COD and the color removal efficiency.

On the other hand, it was demonstrated that bubbles density increases and their size decreases with

increasing current density [35,38] resulting in a greater upward flux and a faster removal of pollutants and sludge flotation.

The results indicated that under the optimal conditions, about 81% of COD and 93% of residual color could be effectively removed from the lixiviate effluent with further contribution of the EC technology used as a post-treatment unit.

3.7. Effect of charge loading

From the results presented in Table 3, we can conclude that the current density has not significant effect on the COD removal in the range from 6 to 15.6 mA cm⁻² at fixed loading ($Q = It$) of 3120°C. The results show that the charge loading affects the treatment efficiency and not current density.

Since it is necessary to optimize the treatment efficiency with the lowest cost, we have to reach a compromise between the current density and electrolysis time. Three comparative tests carried out at $J = 6, 15$ and 14 mA cm^{-2} and at $\text{pH} = 6$, show that the optimum value of current density, allowing fast removal (90 min) of pollutants with two electrodes consumption, was found to be 15 mA cm^{-2} .

3.8. Evaluation of total operating costs

It is interesting to evaluate the total operating cost for treatment of lixiviate solution by electro dialysis in the optimal conditions. It depends on the cost of material particularly electrodes and the energetic cost and of manpower and the maintenance.

The cost depends on the energetic consumption (C_{energy}) as well as electrodes one ($C_{\text{electrodes}}$) [39]:

$$C_{\text{energy}} (\text{kWh/m}^3) = \frac{U \times I \times t}{V} \times 10^{-3} \quad (8)$$

and

$$C_{\text{electrodes}} (\text{kg/m}^3) = \frac{I \times t \times M}{n \times F \times V} \times 10^{-3}. \quad (9)$$

With U is the electric voltage between electrodes (V), I is the current intensity (A), t is the time (h), V is volume of lixiviate in the reactor (m³), M is the molar mass of aluminum (g mol⁻¹), n is number of electrons transferred in the reaction (3 for aluminum) and F is the Faraday's constant (96 486 C mol⁻¹).

The cost of treatment was evaluated by relation [39]:

$$\text{Cost} (\text{US}\$ \text{m}^{-3}) = a C_{\text{energy}} + b C_{\text{electrodes}}. \quad (10)$$

With a is the electricity cost (US\$ /kWh) and b is the cost of 1 kg of aluminum (US\$ /kg). As we know the

Table 3

Effect of current density on the COD removal at a fixed loading charge $Q = 3120^\circ\text{C}$.

J (mA.cm ⁻²)	t (S)	Removal COD (%)	Color removal (%)
6	3120	47.3	76.9
9	2080	47.8	78.2
12	1560	47.3	76.9
13.1	1418	48.29	77.3
14.9	1248	48.78	77.8
6	3120	47.3	76.9

cost of kWh and 1 kg of aluminum are about 0.093 US\$ and 1.950 US\$, respectively. Total operating costs for monopolar connection was estimated at 43.315 US\$ m⁻³ for the optimal operating conditions.

4. Conclusion

The EC process, with aluminum electrodes, presented in this study was investigated for the reductions of the concentration of organic pollutants in lixiviate solution of Jebel Chakir-Tunis.

The influence of variables such as electrolysis time, current density, mode of the connection and initial pH on the run oval of COD and color intensity have been studied and explained as well. Monopolar connection has been preferred for this EC than the bipolar connection. It was found that for treatment of lixiviate solution, COD removal and decolorization efficiency were of 94% and of 96%, respectively, when the pH was about 6, time of electrolysis was 90 min and current density was approximately 15 mA cm^{-2} .

For the optimal operating, conditions operating cost was evaluated to 43.315 US\$ m⁻³.

Finally, EC was found to be a very effective technology treatment for the reduction of organic pollutants in lixiviate solution in Jebel Chakir-Tunis.

Acknowledgements

The authors thank Prof. M.Dachraoui (L.C.A.E. of F.S. T-Tunis) for his financial support of this research.

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