COMPARISON OF ELECTROCOAGULATION AND CHEMICAL COAGULATION IN THE TREATMENT OF ARTISANAL TANNERY EFFLUENTS

S. Tchamango[1],*, O. Kamdoum[2], D. Donfack[3], D. Babale[4] and E. Ngameni[5]

[1] UNIVERSITE DE NGAOUNDERE, FACULTE DES SCIENCES, DEPARTEMENT DE CHIMIE, P.O. BOX 454, NGAOUNDERE, CAMEROON

[2, 3, 4, 5] UNIVERSITE YAOUNDE I, FACULTE DES SCIENCES, DEPARTEMENT DE CHIMIE INORGANIQUE, LABORATOIRE DE CHIMIE ANALYTIQUE, P.O. BOX 812, YAOUNDE, CAMEROON

E-mail addresses: [1] *tchamangos@yahoo.fr,* [2] *kamdoumolivier@yahoo.fr;* [3] *plastik1983@yahoo.fr,* [4] *innadoudou@yahoo.com,* [5] *engameni@yahoo.fr*

ABSTRACT

In this study, the treatment of the effluents of an artisanal tannery by electrocoagulation with iron electrodes was carried out. During electrolytic treatment, a current intensity of 0.4 A was used, and the electrolysis time was varied from 0 to 40 minutes. The performance of electrocoagulation was compared with that of chemical coagulation with ferric sulfate. The results obtained showed that highest removals after electrocoagulation were 71.15, 98.26, and 86.59% for the COD, colour and turbidity respectively. Chemical coagulation reduced the COD up to 83.17%, while colour and turbidity removals were 99.81 and 98.62% respectively. Although chemical coagulation leads to highest percentage removals after treatment, electrocoagulation results in near neutral pH values and also decrease conductivity weakly. By contrast, chemical coagulation increases the acidity of the effluent and its conductivity. Hence with close to neutral pH and weak conductivity values obtained for effluents treated by electrocoagulation, it is inferred that such treated effluents can be recycled.

Keywords: Electrocoagulation; Chemical coagulation; Iron electrodes; Tannery effluents; Removal efficiency

1. INTRODUCTION

Leather, derivative product of the activities of the slaughterhouses, can be transformed in very varied products, used for example in the leatherwork. The city of Maroua, situated in far north region of Cameroon, is recognized for the big activity resulting in industrial and artisanal tanning of animal's skins that serve in manufacture of leather. Both processes of tanning are practiced in this city since a very long time and in spite of the modernization, those involved in artisanal tanning use rudimentary means as against the industrial ones that use chromium salts. In fact, artisanal tanning is achieved with the acacia fruits which contain the tannin which stops the putrefaction of animal skins and also prevent its shrinkage [1]. The environmental consequences of the activities of tanneries in the city of Maroua pose danger because the leather wastes from some artisanal factories and effluents from industrial factories which could contain toxic chemicals are disposed on land or in nearby rivers. People and animals that drink and bathe in these rivers expose themselves to serious

dangers. This threat does not only come from the chemicals used during tanning, but particularly of the bacterial infections that can occur in the rudimentary facilities of artisanal tanneries. For example in the sixties, cases of contamination by the bacillus of coal of the industrial residuary waters were observed in Germany [2].

The dangers induced by the effluents of artisanal tanneries do not come principally from the chemicals as in the case of industrial tanning (chromium salts), but in fact of the organic load resulting from the animals skins used for the tanning process. It is therefore important to sensitize the neighbouring populations of these rivers on their use and to propose the solutions aiming to suppress the source the pollution then, without forbidding the activity of the tannery however that presents an economic interest.

Chemical coagulation uses aluminium and iron salts to eliminate the pollutants. However this technique produces a huge quantity of mud and can be source of secondary pollution because of the chemical reagents

that intervene in the treatment. The adsorption and the membrane filtration are also used with success, but the major inconvenience of these techniques resides in the high treatment cost [3-7].

To palliate these defects, it is necessary to find efficient and less costly techniques that are environmentally friendly. Biological techniques such as biosorption or bioaccumulation offer potential alternatives in the treatment of tannery effluents. In spite of their efficiency, the loss of the biomass at the time of the treatment inhibits these processes. Besides in the case of the plant tanning, it has also been demonstrated that the tannins are recalcitrant to the biologic treatments [8, 9]. Recent works on the treatment of the tannery effluents enhanced the efficiency of the electro-chemical techniques on the destruction of recalcitrant and non-biodegradable organic matters [8]. Electrocoagulation is among these promising processes used in the treatment of aqueous effluents.

Electrocoagulation is the electrolytic version of the chemical coagulation. During this electrolytic process of treatment of the aqueous wastes, the passage of the current in a reactor containing metallic electrodes (iron, aluminum or their alloys) permits to free metallic ions (Fe^{2+}, Fe^{3+} or Al^{3+}) by oxidization of the anode. These metal ions combine then with hydroxyl ions released at the cathode, leading to the formation of metal hydroxides which favour flocs constitution [10-14]. Although the mechanism of electrocoagulation is similar to that of chemical coagulation, the cationic species responsible of the neutralization of the colloids charge, and the flocs coagulated differ in each of these processes: during electrocoagulation, the coagulants (iron or aluminum cations) are gotten in situ by dissolution of the anode, what is not the case of the chemical coagulation where these cations are produced by the dissolution of iron or aluminum salts, which is also accompanied by the liberation counter-ions [15,16].

During electrocoagulation, the amount of metal electro generated (g) is proportional to the quantity of electricity (It) according to the relation: $m = \frac{It}{zF} M$.

In this relation, I the intensity of current imposed, t the electrolysis time, z the number of electrons transferred during anodic oxidation of the metal (z = 2 for iron), F the Faraday's constant (F = 96.486 C. mol^{-1}) and M the molecular weight (g.mol^{-1}) [11, 13, 17].

Comparatively to chemical coagulation, electrocoagulation process provides a simple, reliable and cost-effective method for the treatment of wastewater without any need for additional chemicals, and thus the secondary pollution. It also reduces the amount of sludge, which needs to be disposed [18].

In this work, electrocoagulation and chemical coagulation were applied in the treatment of the effluents of an artisanal tannery. The investigations were focused in one hand on the effect of the physico-chemical parameters (pH, conductivity) and on the other hand on the elimination of some parameters of pollution (DCO, color, turbidity).

2. MATERIALS AND METHODS
2.1 Experimental
All experiments were carried out in a 500 mL beaker at room temperature. For electrocoagulation process, a pair of iron electrodes of size 15 cm×1.8 cm×0.2 cm, immersed to a 5 cm depth was used in the experiments. These electrodes were separated by an insulator made of Teflon. A Stuart magnetic stirrer was used in order to get a good homogenization of flocks and wastewater. The current was provided by an ERMES M10-SP-303, 3A-30V DC power supply. The pH and conductivity measurements were achieved with a Multi340i/SET of Weight Watchers International (WTW). These devices are connected to electrodes comprising a temperature sensor. The Chemical Oxygen Demand (COD) and color were analyzed with a HACH DR 2500 Odyssey spectrophotometer, while turbidity was measured using a Lovibond turbidimeter.

2.2 Reagents and Solutions
Solutions were prepared at room temperature with deionized water. All reagents were of analytical grade and were used without further purification. Before electrolytic treatment, the pH was adjusted by adding 10^{-1} M H_2SO_4 (98%, from Riedel de Haën), or NaOH (from Prolabo) at the same concentration. The coagulant used for chemical coagulation was ferrous sulphate $Fe_2(SO_4)_3$, nH_2O (22% of Fe(III), Prolabo). Support electrolyte was not used to increase the conductivity before the electrolytic treatment because the effluent used contained initially sufficiently sodium chloride (kitchen salt), which interfere with the tanning process. Before each treatment, electrodes were cleaned with H_2SO_4 1M and rinsed several times with deionised water.

2.3 Description of the Tanning Process

The artisanal tannery where effluents were sampled was situated in full air. All operations of tanning took place in watertight pits. Not far away of this place, is a seasonal river in which the content of these pits was emptied after tanning. The process of tanning is described by the following stages:

(i) Salting: the skins of sheeps, goats or cows were soaked in the pits where kitchen salt is poured. This operation permitted the preservation of these skins;

(ii) After salting, these skins were exposed to the sun;

(iii) in order to have flexible skins, one eliminated the salt that they contained while soaking them in the watertight pits filled of hot water;

(iv) The fourth stage consisted in depilating these skins. To achieve this, the skins were soaked in other pits containing anyone of the following products: the lime, sodium carbonate, wood ash, the mixture sodium carbonate and lime or the mixture ash and lime. This operation lasted 48 hours and had for goal the elimination of hairs (keratin) and the fat matter in skins;

(v) Deliming with birds excrements: this operation lowered the pH and to eliminate the last hairs;

(vi) Tanning: after the stage of deliming, these skins were soaked in pits containing a hot solution of acacia fruits previously crushed. The action of the tannin contained in these fruits returned these flexible skins and unputrescible;

(vii) These skins were dried and smeared of fat matters. They were soaked then again in pits containing acacia fruits crushed to which one added some stains;

(viii) The drying was the last stage of the artisanal tanning process. After this stage, skins presented a good aspect.

At the end of tanning, the pits were emptied and the content poured in the seasonal river situated close to the tannery. The excrements of birds and animals hairs used at the time of tanning were recycled to manufacture some bricks.
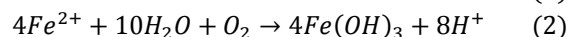
2.4 Description of the Treatment Procedure

For electrocoagulation experiments, 500 mL of the tannery effluent were first fed in a beaker acting as reactor. Then, iron electrodes connected to a DC power supply were introduced in this solution and the homogenization made by a magnetic stirrer. When a current was passed, the treatment occurred and flocks formation was observed after few minutes. These flocs

were greenish first, and then turned yellow and turbid. The green and yellow colours resulted from Fe^{2+} and Fe^{3+} ions formation. Fe^{2+} is the common ion formed *in situ* of electrolysis of iron anode, and can be oxidized easily into Fe^{3+} by dissolved oxygen in water [11]. After40 minutes of electrolysis, the effluent treated is left at rest for about 10 minutes. Then, the physicochemical (pH and conductivity) and the pollution parameters (turbidity, colour and COD) were measured to evaluate the level of the treatment achieved.

Two mechanisms for the production of hydroxides have been proposed to describe the electrocoagulation process when iron is used as the electrode material [16, 19]:
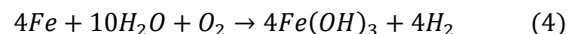
Mechanism1
Anode:
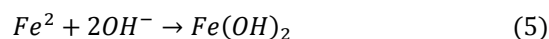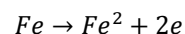$$4Fe \rightarrow 4Fe^{2+} + 8e \qquad (1)$$
$$4Fe^{2+} + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 8H^+ \qquad (2)$$
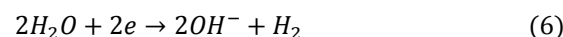
Cathode:
$$8H^+ + 8e \rightarrow 4H_2 \qquad (3)$$
Overall reaction
$$4Fe + 10H_2O + O_2 \rightarrow 4Fe(OH)_3 + 4H_2 \qquad (4)$$
Mechanism 2
Anode:
$$Fe \rightarrow Fe^2 + 2e$$
$$Fe^2 + 2OH^- \rightarrow Fe(OH)_2 \qquad (5)$$
Cathode:
$$2H_2O + 2e \rightarrow 2OH^- + H_2 \qquad (6)$$
Overall reaction:
$$Fe + 2H_2O \rightarrow Fe(OH)_2 + H_2 \qquad (7)$$
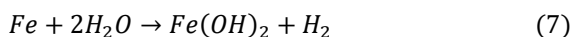
Ferrous or ferric hydroxide formed is in the solution as a gelatinous suspension, and can remove pollutants from aqueous effluents either by complexation or by electrostatic attraction followed by coagulation. Ferric ions electrogenerated may form monomeric ions, ferric hydroxo complexes with hydroxide ions and polymeric species, namely, $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5OH^{2+}$, $Fe(H_2O)_4(OH)^{2+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_4^{2+}$ and $Fe(OH)_4^-$, depending on the pH of the aqueous medium[18, 20].The complexes (i.e. hydrolysis products) can polymerize at pH 3.5–7.0 [16, 18 and 20].Chemical coagulation was done in order to make a comparative study towards electrocoagulation process. For these experiments, ferric sulphate was used as coagulant and added to the tannery effluent in the beaker, under magnetic stirring for about 30 minutes. The amount of the coagulant weighed corresponded to that of Fe^{2+} theoretically

electrogenerated (according to the Faraday's law) during various electrocoagulation times chosen. Calculations were made assuming that during electrolyses, all the Fe(II) species oxidized completely in those of Fe(III).

3. RESULTS AND DISCUSSIONS

### 3.1 Characteristics of the Tannery Effluent Before Treatment

The effluents were withdrawn in the pits, because in the dry season (period of withdrawal), the rivers where they are usually disposed are dry. Consequently, these effluents had a milky aspect and contained in fact more polluting load compared to the case where they would be disposed into a river. They were thus diluted with tap water before to pre-treatment. The characteristics of this water are presented in Table 1.

Table1: Characteristics of the tannery effluent before the treatment.

Parameters	pH	χ (mS. cm^{-1})	Turbidity (FNU)	Colour (Pt-Co)	COD (mg. L^{-1})
Values	8.32	1.46	138	5192	2005

From Table 1, it can be seen that:
- COD and colour values are high. Indeed, according to environmental standards' set by the Ministry in charge of Environment and the Nature Protection of Cameroon,effluents must have COD value lower than 200mg.L^{-1} and should be colourless before it is disposed on land or water[21];
- The value of pH is alkaline, and can this could inhibit the electrolytic treatment since it is known that neutral and acidic pH enhance pollutants elimination during electrocoagulation [19, 22].
- This effluent is turbid, (turbidity more than 50 FNU) [23]
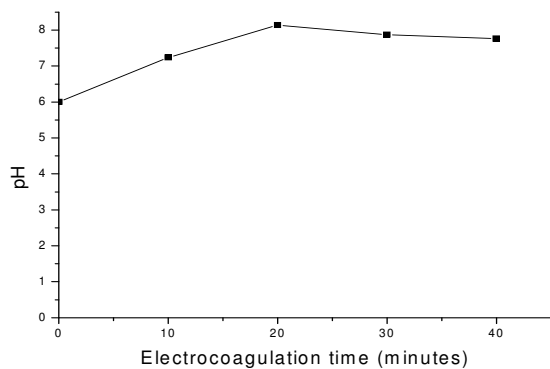
3.2 Electrocoagulation of the Tannery Effluent

Because the initial pH of the effluent was alkaline, we adjusted this value to 6 before all the experiments (electrocoagulation or chemical coagulation). At this pH value, metallic hydroxides that trap colloidal particles are formed [12].

On Fig. 1, is depicted the variation of pH during electrocoagulation of the tannery effluent. As seen on this figure, one notes initially an increase of this parameter with the amount of iron electrogenerated up to 8.14. Beyond this value, the pH decrease to 7.76 at the end of the treatment. The pH increase is due to the water reduction at the cathode that releases continuously OH$^-$ ions. A drop observed then could be explained by acid-base reaction

$$Fe^{2+} + H_2O \leftrightarrow Fe(OH)^+ + H^+ \qquad (8)$$
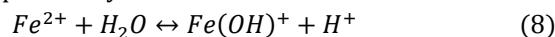
(pKa Fe^{2+}/FeOH$^+$ = 7.9) [24].

The H$^+$ formation in this reaction justify the pH reduction noted. By this phenomenon, one can conclude that electrocoagulation has a buffering capacity and acts as pH neutralization [11].

From Fig. 2, it can be observed that the conductivity varies slightly in the range 1.72-1.85 mS. Cm^{-1}. The increase of this parameter at the beginning of treatment is the consequence of hydroxyl ions released at the cathode. The fall which follows could be attributed to Fe(OH)$_2$ and Fe(OH)$_3$ formation, not conducting and amorphous.

Fig. 3 reports removal efficiencies of colour, COD and turbidity during electrocoagulation. By analyzing this figure, we note that colour and COD evolve in similar way: removal efficiencies of these parameters increase quickly during the first ten minutes of electrolysis. At this stage, 95.69 and 66.56 % of colour and COD respectively are removed.

Figure 1: Variation of the pH during electrocoagulation of the tannery effluent. I= 0.4 A and t = 40 minutes.
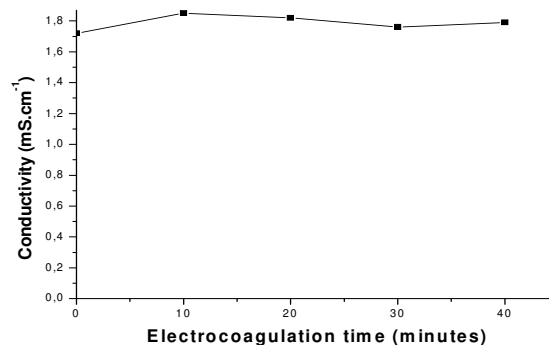
Figure 2: Variation of the Conductivity Variation During Electrocoagulation of the Tannery Effluent. I= 0.4 A and t = 40 minutes.
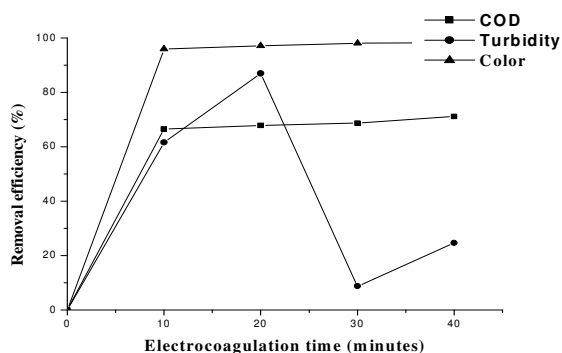
Figure 3: variation of the Removal Efficiencies of the Parameters of Pollution Against Electrocoagulation Time.
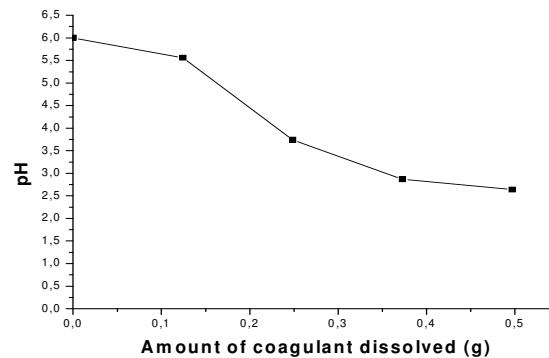

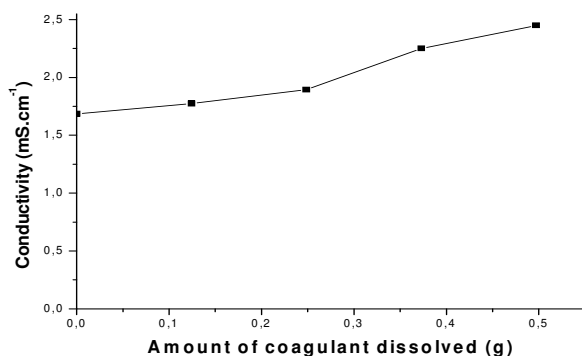Figure 4: Variation of the pH against the quantity of the coagulant added.

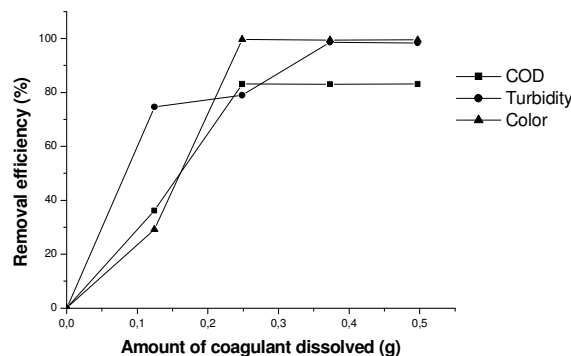Figure 5: Variation of the conductivity against the quantity of the coagulant added.

Figure 6: Variation of the removal efficiencies against the quantity of the coagulant added.

The relative weak removal of COD could be due to soluble organic compounds present in the tannery effluent and which are not eliminated by electrocoagulation [11, 13, and 20].

The variation of turbidity is different from that of the other parameters of pollution studied. Indeed, after the beginning of electrolysis a great increase of this parameter is observed, and after ten minutes of electrocoagulation, one obtains a removal efficiency of 86.96%.A fall is observed then, which brings back this removal to 8.7%.This drop is due to post-flocculation effect, when the rust is formed in the treated effluent after Fe(II) to Fe(III) oxidation in the presence of oxygen, as described above [11].

3.3 Chemical Coagulation of the Tannery Effluent
During chemical coagulation experiments of the tannery effluent, the same parameters (physico-chemical and of pollution) were also studied and the initial characteristics of the effluent remained unchanged.

The variation of pH upon the amount of ferric sulphate is illustrated on Fig. 4. As observed on this figure, the pH of the effluent during chemical coagulation

decreases when the amount of the coagulant added is increased. In fact, the hydrolysis of Fe^{3+} releases protons during the formation of $Fe(OH)_3$ according to the equations below:

$$Fe_2(SO_4)_3 \rightarrow 2Fe^{3+} + 3SO_4^{2-} \tag{9}$$

$$F_e{}^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \tag{10}$$
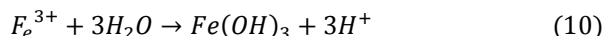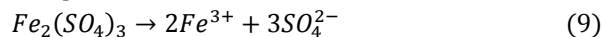
As depicted on Fig. 5, the conductivity on the other hand increases when the coagulant is added to the effluent during chemical coagulation. The conductivity increase is due to two main causes as explained by equations 9 and 10 namely: the protons produced by the hydrolysis of Fe $^{3+}$ and the sulphate ions formed by the dissolution of the coagulant which did not take part in the coagulation of the effluent.

As in the case of the electrocogulation process, we indexed on Fig. 6 the results inherent to the removals of the parameters of pollution. By examining this figure, we note that colour and COD evolve in similar way: an increase of the removal efficiency occurs first when the amount of the coagulant added in the effluent goes up from 0 to 0.25 g. Then, removals vary very slightly until the end of the treatment. But for the

turbidity, 0.37 g of the coagulant is added to reach a plateau. Contrary to the case of electrocoagulation, a drop of this parameter was not observed because the coagulant used is a ferric salt: as a consequence Fe(II) to Fe(III) oxidation that colours the solution does not occurred. At the end of the treatment, removals of 83.14, 98.40 and 99.52% are obtained for COD, turbidity and colour respectively.

By comparing the efficacy of both processes after treatment, following conclusions can be established:

- electrocoagulation presents after treatment of the effluent of the artisanal tannery a water whose pH is slightly alkaline and a conductivity relatively weak comparatively to chemical coagulation. Water resulting from the treatment by chemical coagulation is thus corrosive, more acidic and consequently harmful for the environment;

- electrocoagulation requires less reagent than chemical coagulation. In fact during electrolysis, the really amount of iron dissolved after 20 minutes (by weighing iron anodes) is 0.2703 g whereas the quantity of salt corresponding having been used for chemical coagulation is 0.4971 g;

- whatever the parameter of pollution studied, chemical coagulation presents highest removals. Besides, the amounts of ferric salt dissolved corresponded to that of iron theoretically electrogenerated after 5, 10, 15 and 20 minutes respectively. This situation is explained by the assumption emitted, assuming that during electrocoagulation all Fe(II) is transformed into Fe(III). But actually, $Fe(OH)_2$ was also formed when electrocoagulation takes place, and knowing that it is less effective than $Fe(OH)_3$ for trapping colloidal particles [22]; it is obvious that chemical coagulation with ferric salt which produces only Fe^{3+} ions has best results.

As illustrated on Fig. 7 which presents highest removals obtained by both processes (electrocoagulation and chemical coagulation) the elimination of the parameters of pollution increases in the order COD < turbidity< colour.
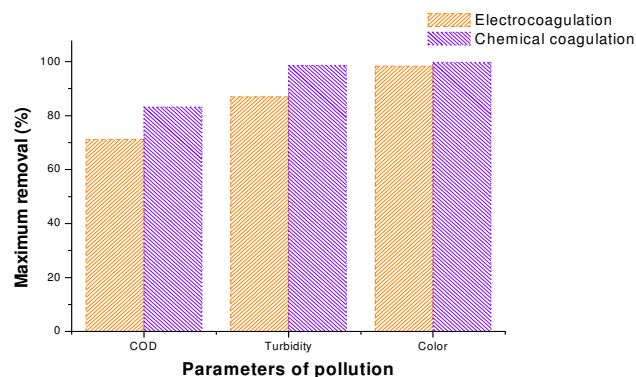

Figure 7: Comparison of the efficacy of electrocoagulation and chemical coagulation.

4. CONCLUSION

In this work, electrocoagulation and chemical coagulation were applied successfully for the treatment of an artisanal tannery, which by its great quantity of organic matter is harmful for the environment. The results revealed that more than 98% of colour was eliminated by both processes. The oxidation of Fe^{2+} to Fe^{3+} during electrocoagulation increase the turbidity of the effluent, and this situation was not observed when chemical coagulation occurred. Water obtained after treatment by electrocoagulation is less conducting and has a pH not far from neutral value and can be recycled. Besides, iron anode consumption during electrolytic treatment is weak compared to that of the ferric salt which acted as coagulant. In spite of its highest removals, chemical coagulation generates after treatment acidic and very conducting water. Thus, in the perspective of preserving the environment this process is not recommended.

5. ACKNOWLEDGMENT

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