Treatment of fouling and biofouling by coupling electrocoagulation and nanofiltration processes.

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***Abstract*— The development of the urbanization and the variation of the industrial activities and agricultural, during these last centuries, were accompanied by a general increase in pollution. The latter is regarded as a major problem of humanity nowadays, and represents a threat for the life on our ground; thing which is generally due to discharges in the mediums reception (rivers, lake, river… etc), of rejections causing various reactions suitable for transform the ecological balance of the medium.**

Water treatment of the lake of Reghaia coming from an industrial park Rouiba/Reghaia was examined by coupling processes. The sample is characterized by a COD of 267.6 mg/L, a BOD of 112 mg/L and a turbidity of 132.85 NTU. In the first time we treated the effluent by electrocoagulation by using an aluminum anode and a graphite cathode. The effect of the density of current, of flow, the pH of the solution and the time of electrolysis was examined. The results obtained showed that the continuous process of electrocoagulation is effective with a rate abatement of turbidity of about 97.35 %, DCO of 90.29 %. The process of electrocoagulation is more effective with a current density of 74.6 A.m $^{-2}$, pH of 5, flow=2.10 $^{-3}$ L.s $^{-1}$ and NaCl concentration of 1 g.L^{-1}. Then we applied the nanofiltration which is a technique of refining making it possible to balance the ions in water.

***Keywords:* Wastewater, electrochemical processes, aluminum anode, nanofiltration, fouling and biofouling.**

I. Introduction

The Lake of Reghaia is one of the most significant natural sites in Algeria from where its classification on June 04, 2003 in the convention of Ramsar relating to the wetlands of international importance. Last vestige of old Mitidja, it is currently the single wetland of the biogeographic area of the Inhabitant of Algiers [1].

But because of its proximity of the industrial park, urban and agricultural, the lake is the direct receptacle of various types of urban and industrial wastes. It receives daily approximately 80 000 m^3 of water polluted which a part comes from the station of purification of worn water another part from both wadi (wadi El Biar, Reghaia wadi) [1].For that we applied coupling of an electrochemical and membrane treatment (nanofiltration) to the water of the lake. Initially we applied the process of electrocoagulation to eliminate several types of pollutants which can be responsible of membrane fouling. Then we applied the nanofiltration which is a technique of refining making it possible to balance the ions in water [2, 3].

Electrocoagulation is an electrochemical process of indirect oxidation in which a reagent dissolved exists or is generated starting from the electrolyte or the phase of the electrode with an aim of taking part in the reaction of coagulation. At the time of the process of electrocoagulation, the coagulant is generated in situ by electrolytic oxidation of a suitable solid anode [4]. Electrocoagulation (EC) is a technology which results from the interaction of three fundamental technologies which are: electrochemistry, coagulation and flotation [5].

It is based on the fact that the stability of the colloids, suspensions and emulsions, is influenced by the electric charges [6]. This technology was employed more and more for the industrial water treatment worn of various origins in the last decade [7].Thus, EC. was applied to drinking waters [6], dyes [8], oil [9], arsenic [10], worn water of the Tannery [11], water underground [12].The principal advantages of the process of electrocoagulation highlighted by several authors [13,14,15] are the compactness of the installations, the lower volume of muds and the elimination of the colloidal particles of small sizes.

The nonofiltration is a membrane process of filtration recently developed which is between ultrafiltration and opposite osmosis. This type of processes is relatively simple of operation does not generate any by-product. However, the filling of the membranes is generally regarded as a limiting factor since that tends to decrease the productivity of the membrane systems and to increase the consumption of cleaning products and energy.

ISSN: 2961-6603

II. REACTIONS OF ELECTRODES

The possible reactions which can occur on surfaces of the electrodes are [16, 17]:

A. *At THE ANODE*

Metal will pass from the solid state in an ionic state according to the reaction:

$$Al = Al^{3+} + 3e^-$$

B. *AT THE CATHODE*

The principal reaction indexed in the bibliography leads to the reduction of water

$$H_2O + e^- \rightarrow \frac{1}{2} H_2 + OH^-$$

Ions Al^{3+} and OH^- generated by the electrode react to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al(OH)_4^-$, et $Al_2(OH)_2^{4+}$ and of the polymeric species such as $Al_2(OH)_2^{4+}$, $Al_2(OH)_5^+$, $Al_6(OH)_{15}^{3+}$, $Al_{13}(OH)_{34}^{5+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ who transform finally into $Al(OH)_3$ according to the kinetics of precipitation of the complexes. These species play the role of coagulant and lead to the formation of precipitates, then of flocs easily eliminable. The majority species depends on the pH of the medium [18].

III. MATERIALS AND METHODS

A. *Sampling*

The samples of the studied rejection were taken starting from the station of purification of worn water of Reghaia. They are stored cold (4°C) and are heated at ambient temperature for need for analysis.

B. Experimental device

In our work we chose a pilot with operation continuous with closed loop whose electrochemical engine is provided with two electrodes, the anode is out of aluminum and cathode out of graphite is connected to a generator of current continuous of type DC power supply PS-305D, the two electrodes are cylindrical plates separated between them by an insulator, the diameter of the anode is 0.8 cm and the height is of 16cm (active surface = 40.2 cm^2).They are placed vertically in the electrochemical cell and thus parallel to the direction of passage of the flow of the liquid, in order to minimize the pressure losses in this compartment to support the recirculation of the liquid. The recirculation of the solution is ensured by a peristaltic pump of type ISMATEC.

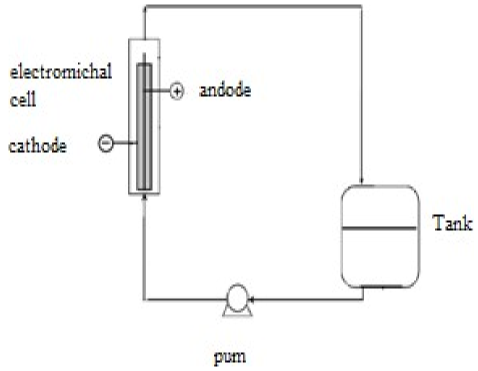

Fig.1. Experimental device of EC

C. *Characteristics of sample*

TABLE I: Characteristics of sample

Parameters	Units	Values
pH		7.8
conductivity	mS/cm	1.56
Turbidity	NTU	133
COD	mgO_2/L	267.6
BOD	mgO_2/L	112.11
Chlorides	mg/L	470.27
NO_3^-	mg/L	2.048
SO_4^{2-}	mg/L	321
SPM	mg/L	262

IV. RESULTS AND DISCUSSION

1) Electrocoagulation

A. *Effect of the density of current*

The density of current is a critical parameter in electrocoagulation, since it is only being able to be controlled directly [19]. When the current density increases, the processing time decreases because of the strong dissolution of the electrodes.

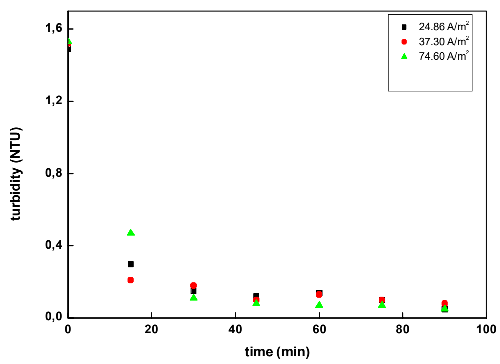

Fig. 2: Variation of turbidity according to times with different density of current (pH =7.50, Q_v= 7.3 10^{-3}L/s).

ISSN: 2961-6603

Figure 2 shows the evolution of turbidity according to times with different density of current. It shows that residual turbidity decreases with the packing of current. This reduction is due to the agglomeration of the suspended matter [21] and to the formation of the complexes of hydroxides which play a significant role in the coagulation of the pollutants which settle or float on the surface. It is noticed that for a density of current to 74.6 A.m^{-2} equalizes and a time T = 90 min the rate of elimination of turbidity is to the maximum 96.73%. This result suggests that electrocoagulation is an effective technique for the elimination of the turbidity of water.

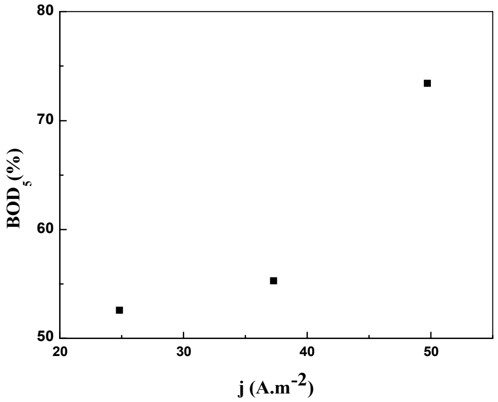

Fig.3 Evolution of rate of abatement of the BOD_5 according to the current density (pH = 7.50; Qv = 7.3 mL/s).

According to figure 3 the output of elimination of BOD increases gradually with the packing of current. Indeed, a density of higher current generates a significant quantity of flocs which contributes to the elimination of the organic matter. Moreover, the rates of generation of bubble increase their size on the other hand decreases with the density of current [22] what is beneficial for the elimination of the pollutant by flotation. In addition, a density of higher current could provide an action of oxidation [16].The effectiveness of elimination of the BOD passed from 55% to 75% when the density of current passes from 24 A.m^{-2} to 49 A.m^{-2}.

B. Effect of flow

In order to study the effect of the residence time on the effectiveness of the treatment, the flow of entry Q was gradually increased in the beaches of 2.10^{-3} L/s with 7.10^{-3} L/s, for j = 75 A.m^{-2}; pHi = 7.5.The results obtained are presented at the lower part:

According to figure 4 the elimination of turbidity is higher with lowest flow, one can conclude that the more time of contacts is larger the effectiveness of the treatment is better. For a flow of 2.10 $^{-3}$ L/s and a time T = 45 min the rate of elimination of turbidity is 99.05% and remains constant until T = 90 min. This is in agreement with the results obtained previously with other researchers [23]. The optimal flow of entry could thus be regarded as 2.10^{-3} L/s.

Figure 5 shows the evolution of the DCO of the effluent during the treatment for a flow of percolation of 2.10-3L/s. We note that the rate of abatement of the DCO increases during the treatment to reach a stage. The rate of elimination is to the maximum (81.25%) with T = 30 min. The reduction in the concentration of the organic matter is due to the reactions redox [24]; the elimination of the pollutants by electrochemical oxidation, adsorption on various species hydroxo and/or aluminium polyhydroxydes [25] which have a great capacity of adsorption of the soluble organic compounds and coagulate solid colloids in the effluent.

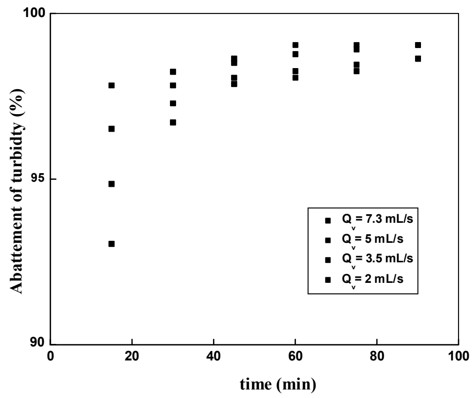

Fig.4 Variation of the abatement of turbidity according to time with different flows (j=74.6 A.m^{-2}, pH = 7.50).

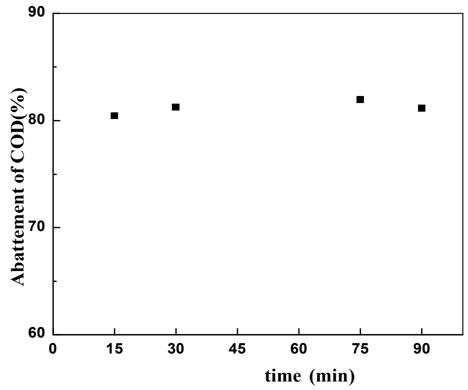

Fig.5 Evolution of COD according to time (j=74.6 A.m^{-2} et Q_v=2.10^{-3} L.s^{-1}, pH = 7.50).

ISSN: 2961-6603

Figure 5 shows the evolution of the COD of the effluent during the treatment for a flow of percolation of 20^{-3}L/s. We note that the rate of abatement of COD increases during the treatment to reach a stage. The rate of elimination is to the maximum (81.5%) with t = 30 min. The reduction in the concentration of the organic matter is due to the reactions redox [24]; the elimination of the pollutants by electrochemical oxidation, adsorption on various species hydroxo and/or aluminum polyhydroxydes [25] which have a great capacity of adsorption of the soluble organic compounds and coagulate solid colloids in the effluent.

C. Effect of pH

To study the effect of the pH, we carried out a series of tests with four values of pH to knowing: 2, 3, 5 and 9.This pH was adjusted with solutions of hydroxide of sodium and hydrochloric acid (NaOH and HCl). The density of current was fixed at 74.6 $A.m^{-2}$ during one hour and half of electrolysis. The results obtained are presented at the lower part:

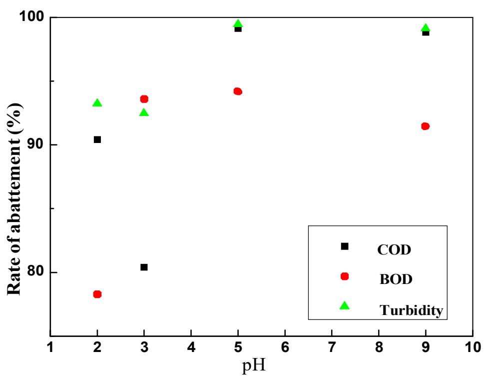

Fig.6 : Evolution of the abatement of the COD, the BOD_5 and turbidity according to different pH (j =74.60 $A.m^{-2}$, $Q_v = 2.10^{-3}$ L/s).

We observe according to figures 7 that for pH =5 the rates of abatement of turbidity, COD and BOD are respectively 99 %, 99.12% and 94.18%. The effectiveness of elimination was low and decreased significantly with the reduction in the pH (pH < 5).The possible explanation of this phenomenon was given starting from the observation of the diagram of solubility of the aluminum hydroxide, which indicates that the degree of hydrolysis of Al^{3+} depends on the pH With a low pH, Al^{3+} is favored in the solution and thus the effect of coagulation was negligible, resulting in a low effectiveness from elimination of pollution. In a range of pH from 5 to 9 the polymerization of $Al3^{+}$ forms of the mononuclear and/or polynuclear complexes of aluminum, for example$Al(OH)_3$, $Al(OH)^{2+}$, $Al_2(OH)^{4+}$, et$Al_{17}(OH)_{32}{}^{7+}$, who are effective agents coagulants for the pollutants.But with high pH of the solution, the dominant species is $Al(OH)_4^-$, which does not want obviously to coagulate the pollutants [26]. Indeed the process of electrocoagulation is more significant with pH 5 with very fast kinetics compared with those obtained for the other pH.

D. Effect of NaCl

In the electrochemical processes which consist of transfer reactions of heterogeneous ions, the conductivity of the solution to be treated is a significant parameter. It affects the output of current, the tension applied, and the electric consumption of power in electrolytic systems, because the current crossing the circuit is a function of conductivity under a certain tension applied.
In this study, the sodium chloride (NaCl), was used as electrolyte to increase the conductivity of the aqueous solution.

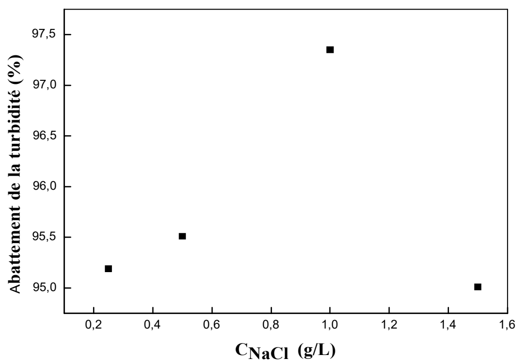

Fig.7: Evolution of the abatement of turbidity according to various NaCl concentrations ($Q_v = 2.10^{-3}$L/s, pH = 5, j = 74,6 $A.m^{-2}$; t = 1h).

The effect of various concentrations of electrolyte on the effectiveness of elimination of turbidity is shown in figure **7**. In general, after 60min of electrolysis, the addition of the electrolyte (NaCl) led to a light increase in the effectiveness of treatment until reaching a maximum (97%). Then, the addition of the NaCl concentration does not have an influence on the effectiveness of treatment.

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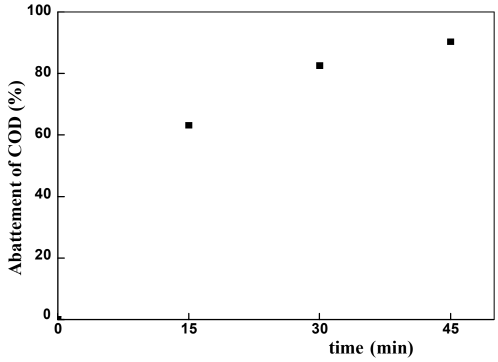

Fig.8 Evolution of the abatement of the COD according to time ($Q_v = 2.10^{-3}$ L.s^{-1} , pH = 5; j = 74,6 A.m^{-2}, C_{NaCl} = 1 g.L^{-1})

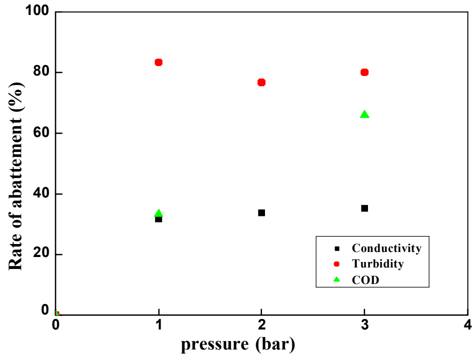

Fig.9 Variation of abatement of the turbidity of conductivity and the DCO according to flow of perméat

The results presented in figure 8 show that the percentage of elimination increased with time. That is due to the oxidation and the reduction of the reactions. For a time of 45 min the rate of abatement is to the maximum 90%. The presence of chlorides in the effluent, Cl_2 and OCl^- can be produced starting from the anodic discharge of the ions chloride [28]. The hypochlorous ions, OCl^-, are strong oxidants, which could oxidize part of the organic molecules present in worn water so that it contributes to the water treatment.

TABLE II
ANALYSES BACTERIOLOGIQUE

Bacteriological Parameters	Units	Before treatment	After treatment
Coliformes totals	ge/mL	>2420	>250<1000
Coliforme fecal	ge/mL	>2420	>250<1000
Germes totals	FCU/mL	>2420	>250<1000

According to table **2** we notice that the process of EC is very effective for the elimination of the bacteria.

2) Waste processing by Coupling of processes EC.-NF

The purpose of the coupling with nanofiltration is to reduce the ions present in the supernatant collected after electrochemical treatment.

According to figure 9 we notice that the rate of abatement increases with the increase in the pressure this is due to the retention of the ions.

V. Conclusion

This research made it possible to show the effectiveness of the electrocoagulation in organic matter the industrial waste processing charged. Indeed, it is well by the in situ formation of agent coagulant (Al^{3+}, Fe^{2+}) coming from anodic dissolution that worn water tested was clarified.

The physicochemical characterization of taken sample showed that water of the Lake Reghaia presents high concentrations of the parameters of study which are the COD, conductivity and turbidity. For that we carried out coupling of an electrochemical and membrane treatment.

The experimental results proved that:

- Residual turbidity decreases with the packing of current. This reduction due to the agglomeration of the suspended matter and the formation of the complexes of hydroxides.
- The output of elimination of DBO increases gradually with the packing of current. Indeed, a density of higher current generates a significant quantity of flocs which contributes to the elimination of the organic matter.
- Highest flow involves less elimination of pollution load.
- With pH=5 the rate of elimination reaches its maximum with this range is made the polymerization of $Al3^{+}$ forms of the mononuclear and/or polynuclear complexes of aluminum.

The experimental results showed that electrocoagulation can reach a percentage of elimination of the turbidity of 97 %, COD of 90%, nitrite of 86 % and nitrates of 11%. Thus the

ISSN: 2961-6603

process of electrocoagulation is more effective with a density of current of 74.6 $A.m^{-2}$; pH of 5, flow of percolation of 2.10^{-3} $L.s^{-1}$.

The process of nanofiltration plays a significant role in traittement of water of the Lake Reghaia, it gives a rate of abatement of about 90%.

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ISSN: 2961-6603