

Suspended Solids and Organic Matter Elimination from Superficial Raw Waters by Electrocoagulation Process in an Airlift Reactor Internal Loop

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Abstract-

In this work, an airlift reactor with internal loop using electrocoagulation technology for superficial raw water treatment has been studied. The objective was the evaluation of the effect of applied voltage on turbidity reduction. For this purpose, synthetic solutions with known turbidities were prepared and tested. The airlift reactor has two parallel compartments, separated by a partition: an upward flow compartment, called riser, in which aluminum electrodes were immersed, and another down flow compartment called downcomer. Different potentials generated by an electric generator were applied on electrodes (0 to 12 Volts). During electrocoagulation, small gas bubbles generated on electrodes (hydrogen at the cathode and oxygen at the anode) cause recirculation synthetic solutions between the riser and the downcomer.

Evaluation of removal performance of the turbidity in the reactor internal airlift loop with electrocoagulation led to conclusive results. The yield increases with time, it is best when the applied voltage increases. At 12 V, it reached 96%.

Keywords -Electrocoagulation, Airlift reactor internal loop, turbidity, superficial raw water.

I. INTRODUCTION

Superficial raw waters destined to drinking water production contains usually many inconvenient elements (suspended solids, bacteria, viruses, organic matter, etc.), which could causes some limits for its human consumption. So, superficial raw water should be treated to meet drinking water standards.

Organic matter in superficial raw water presents the risk of formation of chlorinated products [1], which can be produced by the pre-chlorination or post-chlorination, mainly used in a lot of water treatment plants in Morocco. These chlorinated products are also the source of bad organoleptic quality of drinking water and can be highly toxic and even carcinogenic to humans[2], [3]. The organic matter removal could be done by a pre-oxidation

process using different agents: chlorine, chlorine dioxide, ozone, potassium permanganate.

The presence of suspended matter induces a number of undesirable effects, such as the fixation of the microorganisms or the adsorption of organic matter, which may induce bacterial reminiscence or chlorinated products, after release of organic matter in the case where the suspended matter of superficial raw water is not sufficiently treated.

Removal of the suspended solid matters is made by coagulation. It requires the use of metal cations coagulants. The electrocoagulation process is derived from classical coagulation and is widely used in the treatment of drinking water [4]-[7].

II. ELECTROCOAGULATION TECHNIQUE

Electrocoagulation consists on two metallic electrodes immersed on water with a continuous voltage, which allows the release of metal cations as coagulant. Electrode metals are cylindrical and made of iron and aluminum [8], [9].

The experimental setup of the electrodes may be in serial mode [10], or in parallel mode [11].

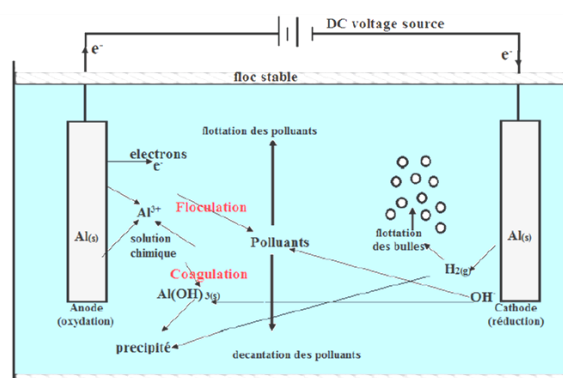
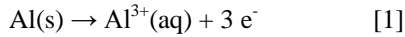


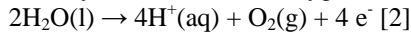
Fig. 1 Shows Electrocoagulation Setup.

The main reactions which have been encountered at the aluminum electrodes are:
At the anode:

The electrolytic dissociation of the aluminium anode products Al^{3+} :

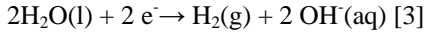


At high current densities, this reaction may be accompanied by the formation of oxygen:

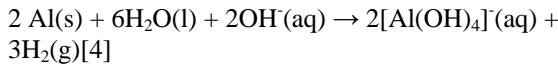


At the cathode:

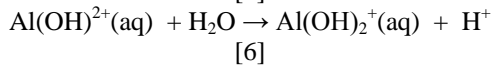
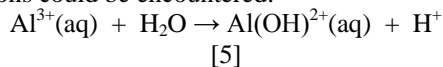
water reduction is produced according to the following reaction:



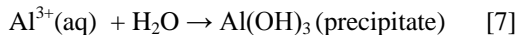
Hydrogen bubbles are generated on the surface of the cathode. Several authors have shown that these bubbles are causing transport of coagulated flocs to the free surface (flotation processes)[12] – [15]. When the pH of the solution is high, the cathode could be attacked by the OH^- ions to form aluminates and hydrogen according to the following reaction:



In acidic medium, hydrolysis reactions of Al^{3+} were made, with formation of monomers ($Al(OH)^{2+}$, $Al(OH)_2^+$) or polymers ($Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{3+}$), and protons, H^+ . For examples, in the case of monomers, the following reactions could be encountered:



The polymerization degree increases with increasing pH. These hydrolyzed cations neutralize, with their positive charges, the negative potential of the double layer surrounding the colloidal particles (ζ Potential). When the $pH \geq 4$, the Al^{3+} cations give precipitation $Al(OH)_3$ according to the following reaction:



The contact between the coagulant and the elements to remove in water is a key element for the success of electrocoagulation. It can be ensured in various ways, by mechanical agitation using magnetic stirrer [16], or mobile agitation blade type [17], [18]. Recently, some authors [19]-[22], have used the flow of airlift loop to ensure the transport and increase contact between the coagulant and the elements to remove. A recent study on the application of electrocoagulation in an airlift reactor internal loop was done for treatment of colored wastewater [23].

The gas bubbles produced at the electrodes allow the separation of the coagulated flocs by the electroflotation process. The flocs reaching the surface of the water form a foam containing 9 to 12% of solid, easy to separate from water. Also, the coupling between electrocoagulation and electroflotation can be envisaged for the treatment of water,

especially when electrocoagulation is performed with high current density.

III. AIRLIFT TECHNIQUE

The airlift reactor contains two compartments where there is a movement of gas bubbles causing a circulation of the liquid. The area where this movement is created is called riser, this movement is upward. It is separated from another zone, called the down comer, by a vertical partition. In this area, the movement of gas bubbles from the liquid is down (e.g. Fig. 2). Fig. 3 illustrates the airlift systems for internal and external loops.

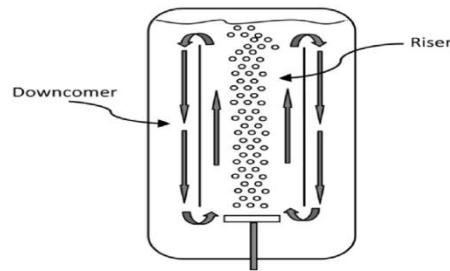


Fig.1 Schematic Diagram of the Airlift Internal Loop

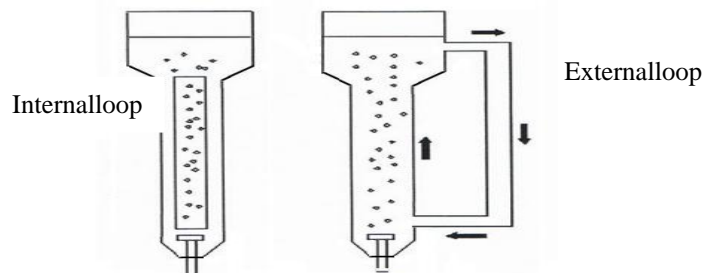


Fig.2 Both Types of airlift

IV. MATERIALS AND METHODS

A. Experimental Setup

The removal of suspended solids and organic matter was carried out in a laboratory pilot type airlift reactor internal loop containing two electrodes for the electrocoagulation. A simplified scheme of the pilot is shown in Fig. 4.

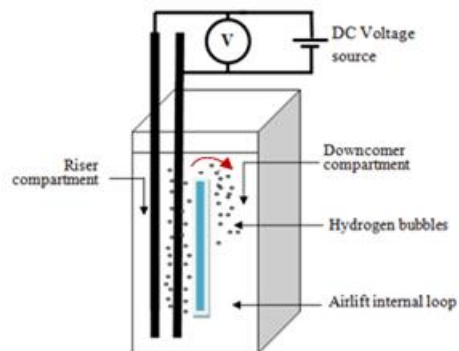


Fig.3 Schematic Description of the Installation

Electrocoagulation was performed according to the diagram of Fig.4. Two aluminum parallel and plate electrodes were made with a thickness of 3mm, width of 24mm and having an inter-electrode distance of 18 mm. Electric generator model G823 feeds the reactor by a direct current and at variable potential (0 to 12 volts). The electrodes are immersed in a synthetic solution of 1200 ml and 40 cm of height under water surface.

B. Synthetic Solution

In a container, a water solution with clay is prepared, and then exposed to an agitation at 100 tour/ min for 30 min. After decanting for 30 min, the supernatant is recovered to prepare a synthetic solution with a given turbidity.

C. Experimental Protocol

The protocol of experimental tests was as follows:

- airlift reactor cleaning using distilled water;
- electrodes cleaning with HCl (4%);
- electrodes fixing in the airlift reactor at the riser compartment;
- the airlift reactor is then filled with the solution to be treated;
- the electrocoagulation test is activated.

With the electric generator, one fixes the voltage applied to the electrodes. Electrocoagulation is followed using the physicochemical parameters, which are measured in samples taken using a pipe at the bottom of the reactor.

Monitoring parameters and measurement apparatus used are presented in the following table:

Table I : Physicochemical Parameters Monitored and Measuring Devices

Turbidity	Palintest photometer 7000
pH	HACH HQ40d pHmeter
Conductivity	Conductimeter type Orion model 125

The oxydability measured with $KMnO_4$ method is determined by assaying samples filtered in a filter of porosity $0.45\mu m$.

V. RESULTS AND DISCUSSION

A. Visual Observations

The visual monitoring of this reactor has identified some phenomena illustrated in Fig. 5.

These phenomena are as follows:

- 1) Because of electrolysis of water, micro bubbles were appearing at the two electrodes surface: hydrogen (H_2) at the cathode and oxygen (O_2) at the anode. The amount of hydrogen bubbles was higher than that of the oxygen;
- 2) Formation of a wake of a solid matter (clay) near the anode: this is probably due to attraction of

the clay by Al^{3+} dissolved in solution near the anode;

- 3) Formation of a floating layer, which thickness increases over time;
- 4) In the downcomer compartment, there is slow recirculation of water compared to the riser compartment;
- 5) The speed of the agitation between the two compartments (riser and downcomer) increases by increasing the intensity of current with the applied voltage;
- 6) After some time of electrocoagulation, the wake at the neighborhood of the anode was disappeared. This is due to the homogenization of solutions by recirculation;
- 7) After a certain time depending on the applied voltage, there is clarification of the solution.

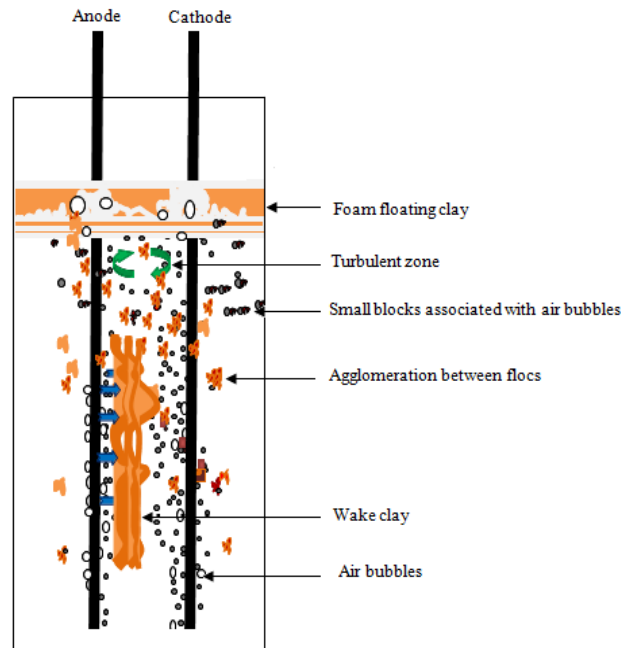


Fig.4 Electrocoagulation Phenomenon in the Riser Compartment

B. Effect of Applied Voltage on Synthetic Solution

At different tensions of 3, 6, 9, and 12 V, results of the electrocoagulation tests in a reactor airlift internal loop were as follows:

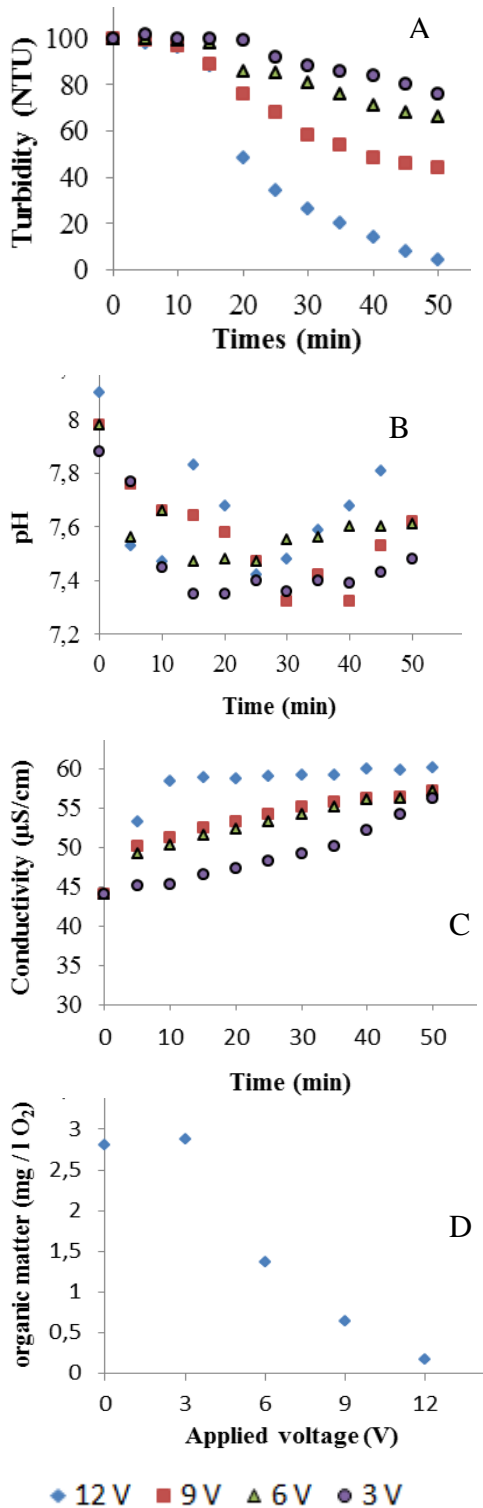


Fig.5 Physicochemical Parameters Evolution Versus Time for Different Applied Voltages

Fig.6-A shows that during electrocoagulation test, abatement rates remain low until almost 10 minutes (20 minutes for the test of 3 V). Al^{3+} cations released at the anode were hydrolyzed (e.g. reactions 5 and 6), which results in the decrease in pH (e.g. Fig. 6-B). These cations are not capable to reduce the double layer of the colloidal particles and then ensure electrostatic coagulation of

the suspended matters. Then, there is a clear reduction in turbidity as a function of the applied voltage between 15 and 30 min. That may be associated to increasing the concentration of Al^{3+} cations released at to the anode (increase the ionic strength of the solution), then, the $\text{Al}(\text{OH})_3$ were formed (e.g. equation 7) at minimum pH (between 7.3 and 7.45).

Turbidity abatement rates increases with time and depending on the applied voltage. After 50 minutes, they reach 24% for 3 V, 34% for 6 V, 56% for 9 V and 96% for 12 V. This reduction is more improved by the recirculation of oxygen and hydrogen bubbles between the riser and the downcomer allowing a good mix between the clay and the flocs of $\text{Al}(\text{OH})_3$ and contributes to the improve of treatment performance [18].

The removal of suspended matters made by adsorption on $\text{Al}(\text{OH})_3$ [24], or on other solid structures that can be also be formed, particularly by reaction between Al^{3+} and clay (as it was observed visually near to the anode in the airlift reactor), establishing some interactions particularly by metal bridges.

The X-ray diffraction analysis (XRD) diagrams (e.g. Fig. 7) show crystallographic structures of raw clay, wet clay without electrocoagulation and wet clay after electrocoagulation. The examination of these diagrams shows the presence of a number of structures such as quartz, kaolinite, montmorillonite, dolomite, illite, ... etc.

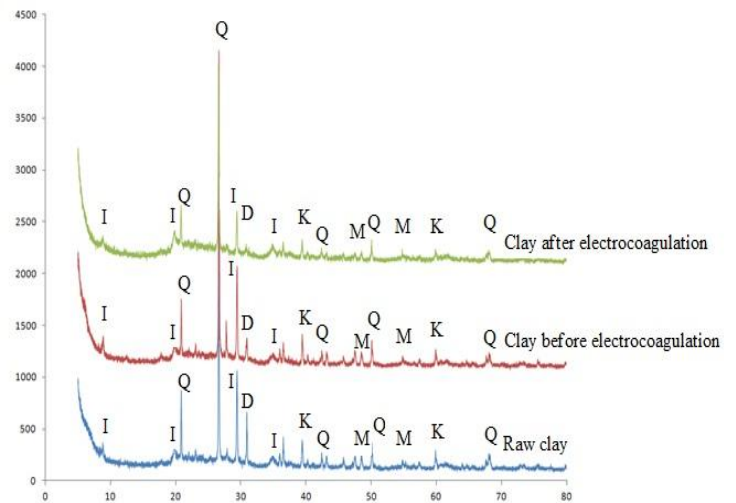


Fig.6 DRX Diagrams of Clay in the Raw State, in Water and After Electrocoagulation (I: Illite, K: Kaolinite, Q: Quartz, D: Dolomite, M: Montmorillonite)

Concerning the conductivity (e.g. Fig.6-C), it increases despite its low initial value (lack of electrolytes promoting electrocoagulation). This increase is slow for the essay of 3V and more or less fast for the other essays, depending on the applied voltage. It increases over time because of the dissolution of Al^{3+} and OH^- ions in the solution. It

quickly reached a level for the voltage of 12 V, which remains constant because of precipitation of $\text{Al}(\text{OH})_3$, when Al^{3+} and OH^- are formed in large quantities at this potential (e.g. equation 7).

Fig. 6-D shows that the increasing elimination of the organic matter was function of the applied voltage. This can be explained by the nascent oxygen liberated at the cathode. It is very reactive with the organic matter. This result was found by other authors [25]-[28], which showed that the reduction of organic matter, expressed in terms of COD increases with the current density.

Furthermore, increasing the voltage applied across two electrodes promotes the formation of $\text{Al}(\text{OH})_3$, allowing more adsorption of organic matter. Zidane et al. [29], using hydroxides prepared based on aluminum and iron electrocoagulation, made adsorption tests; they showed that these hydroxides fix the dissolved organic matter with a removal rate varies between 51.9 and 68.8%. The highest rate is observed among hydroxides containing aluminum.

VI. CONCLUSIONS

The use of electrocoagulation, combined with the airlift internal loop process seems to be an effective solution for the removal of suspended solids and organic matters. Hydrogen and oxygen micro bubbles generated at electrodes ensure good mixing of the solution during electrocoagulation, and also increase the residence time of the oxygen in the reactor.

This work has highlighted the following conclusions:

- At the start of electrocoagulation, suspended matter (clay) forms a wake in front of the anode releasing Al^{3+} cations, probably due to electrostatic attraction;
- Recirculation of oxygen and hydrogen bubbles between the riser and the downcomer provides a good mix between the clay and the flocks of $\text{Al}(\text{OH})_3$ and contributes to improve the performance of the process;
- During electrocoagulation, the suspended matter floats by electroflotation;
- The abatement rate reaches, after 50 min, 24% for 3V, 34% for 6 V, 56% for 9 V and 96% for 12V. The removal of suspended solids is mainly done by adsorption on $\text{Al}(\text{OH})_3$ or on other solid structures that can also be formed, especially by means of reaction between Al^{3+} and clay through metal bridges, or by means of exchange promoting the release of cations that allow the coagulation of suspended matter. This can be corroborated by XRD diagrams of the clay before and after coagulation, where disappearance of some rays or decrease their intensities demonstrate the interaction between $\text{Al}(\text{OH})_3$ and clay;
- Despite of the low conductivity of the solution, a slow increase is observed mainly at 3V test, but more or less fast for the other tests, depending on the applied voltage. The availability of Al^{3+} and OH^- ions in solution by electrocoagulation over time is a factor promoting the electrocoagulation performance;
- The increased removal of organic matter depended on the applied voltage. The oxygen released at the anode was in a nascent state, it was very reactive against organic matter.

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