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Efficiency of a Pretreatment by Electrocoagulation with Aluminum Electrodes in a Nanofiltration Treatment of Polluted Water

Mourad Taleb Ahmed*[a], Toufik Chaabane[a], Rachida Maachi[a], André Darchen[b]

[a] *Environmental Department / FGM_GP / USTHB / Algiers 16111, Algeria*
[b] *UMR CNRS 6226 Sciences Chimiques de Rennes, ENSCR, CS 50837, avenue du Général Leclerc, 35708 Rennes Cedex 7, France*

Abstract

The decrease of quality and availability of waters induces the research of hyphenated technologies for improving purification processes. The paper is devoted to the water treatment of a lack which accumulates a diversified pollution arising from a lot of industries. The investigated lack water contained heavy metals, fecal bacteria and high chemical pollution as measured by its chemical oxygen demand (COD). An electrocoagulation was applied as water pretreatment of a nanofiltration process. The electrocoagulation was carried out in a batch experiment with two parallel aluminum plates of 15 cm^2. Water analysis showed a decrease of the COD from 60 to 5 mg L^{-1}, depending upon current density, current charge and solution pH. Thanks to electrocoagulation efficiency, the most probable number (MPN) of bacteria decreased from 2500 to 2 MPN/100 mL. This disinfection allowed the performing of a continuous coupling treatment of electrocoagulation and nanofiltration by using a cylindrical aluminum anode and an organic Nanomax50 membrane respectively.

Keywords: Pollution**,** Heavy metals; Bacteria; Lake Water; Hyphenated processes.

1. Introduction

The decrease of quality and availability of waters induces the research and development of new materials and processes in membrane filtration [1-3]. Membrane technologies are frequently used in water and wastewater treatments for purification of drinking water [5-6] or for water reuses [7]. These technologies are able to clarify or purify waters by eliminating a lot of undesirable particles. According to the nature of the membrane and applied pressure, these techniques give the advantage of carrying out a separation without addition of chemicals into the solution, and so they allow a control of the ionic composition of the solution.

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* Corresponding author. Tel/Fax.: 213 2 21247919
E-mail address: mdtaleb_fr@yahoo.fr.

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The major disadvantage of these technologies lies in the filling of the membrane pores leading to a reduction of flow rate [7]. This performance decrease is the result of the development of fouling or biofouling on the membrane surfaces [8-11]. So, the goal of obtaining an efficient treatment of highly polluted waters implicates the use of hybrid or hyphenated processes where the first process acts as a pretreatment. The reduction of fouling and biofouling is the main objective in order to improve the efficiency of membrane separations. The decrease of biofouling involves the disinfection of water before its treatment in a membrane process. Generally, this disinfection is performed with oxidant chemicals. The reduction of fouling may be carried out by using chemical or electrogenerated coagulants [12]. Electrocoagulation is an electrolytic process which works with soluble anodes, generally aluminum or iron electrodes. In electrocoagulation [13-21] the anode dissolution affords ions and hydroxides which are well known as coagulant involved in a flocculation process. The stability of the formed hydroxides depends primarily on the pH of the solution. Electrocoagulation is well known in wastewater treatments [18-19]. One main advantage of electrocoagulation is that this treatment does not use chemicals which remain in the treated water. Recent papers have showed that electrocoagulation can also remove living species from water [20-22]. The coupling of electrocoagulation with membrane separation has already been performed [23-26]. In this study, treatment of landfill leachate by a novel electrocoagulation – nanofiltration (EC/NF) hybrid system is investigated. The electrocoagulation leads to the decrease of the total organic compounds (TOC) and turbidity of 67% and 80%, respectively. In comparison, the chemical coagulation with the best dosage of $Al_2(SO_4)_3$ allows an abatement of TOC and turbidity of 10% and 65%, respectively. Therefore, this better performance of the electrocoagulation process may be explained by a higher coagulant concentration and by the formation of polymeric aluminum species which are known to be more efficient for small organic compounds. Coupling of the processes with aluminum instead of iron in electrocoagulation pretreatment avoids the filling of the membranes in the surface water treatment [23]. In addition, the electrocoagulation was shown more effective in front of coagulation for pretreatment of the solution containing bacteria by using soluble iron electrodes and this before treatment by microfiltration [24]. For the treatment of brackish water by osmosis inverse process, the electrocoagulation is used in order to avoid clogging in the membranes, the coupling gives good result even with concentration raised out of treated silica [25]. For treatment of a textile effluent [26], the hybrid process (electrocoagulation/nanofiltration) was used to improve permeate quality of biologically treated textile wastewater. In this paper we show a new example of the coupling of an electrocoagulation process with a nanofiltration in order to treat natural water arising from a highly polluted lake. In this coupling we investigated the treatment of both biological and chemical pollutions in batch and continuous experiments.

2. Experimental

2.1. Chemicals

All reagents were of analytical reagent grade and were used as supplied. Reagent solutions were prepared in distilled water and were stored under dark conditions at 4 °C.

2.2. Characterization of the samples

Initially, we carried out the characterization of the initial sample. This water sample is taken at the exit of a purification station which ensures a primary treatment. The analysis results are gathered in table 1.

Table1. Physicochemical parameters on the level of the STEP (Eng Dept 2009).

Parameters	Raw waters	Treated waters
MES (mg/L)	383	9
COD » »	553.5	70
BOD » »	263.5	7.83
Conductivity (mS/cm)		1,3
Turbidity (NTU)		3,5
Total Coliformes (NPP/100mL)		>2420
Fecal Enterocoques (NPP/100mL)		>2420

2.3. Tests in electrocoagulation batch mode

The electrocoagulation batch mode consists of a cell out of cylindrical glass of form acting as decanter. The tests are carried out between two aluminum electrodes using a galvanometer. The treated volume of the solution being of 300 cm^3 and in which the electrodes of an active surface equal to 15 cm^2 are immersed. The solution is subjected to an agitation along the experiment.

For the batch mode, we fixed the initial pH and the electrolysis time. Following this operation, a decantation during 24 hours was estimated for a separation. In order to have reproducible surfaces a cleaning with the hydrochloric acid and water is necessary. A measurement of parameters (pH, conductivity, turbidity, COD, J) is taken for the supernatant obtained after electrocoagulation (table2).

Table 2. Parameters in batch mode after Electrocoagulation.

pH initial	**Current density (mA/cm^2)**	**pH After EC**	**Conductivity (mS/cm)**	**Turbidity (NTU)**	**COD (mg/L)**
8.1	6.66	8.25	1.41	2.97	8.64
	13.33	8.40	1.22	1.28	4.80
	12.5	8.46	1.19	31.9	-
6	4.16	6.98	1.64	16.6	-
	8.33	7.47	1.53	30.7	-
	12.5	7.52	1.52	0.2	9.46
5	4.16	6.71	170	18.8	6.72
	8.33	6.28	1.63	19.5	31.68
	19.16	7.16	1.53	1.51	4.69
4	8.33	6.6	1.58	1.23	48.96
	16.66	6.52	1.51	81.3	11.1
	25	7.19	1.48	8.74	25.92

2.4. Tests of electrocoagulation in continuous mode

Into an engine piston, we introduced cylindrical aluminum anode with 3.5 cm in internal diameter, 16.5 cm of height (H) and with a surface activates S = 362.67 cm^2. Cathode used is out of graphite. The assembly is with external anode with a radial configuration compared to the ascending percolation of the solution. A calming section is added at the entry of the engine to ensure a flow piston. The electrodes are connected to a galvanometer (Fig. 1).

1. DC power supply
2. Cathode Carbone electrode
3. Anodic cylindrical of aluminum electrode
4. Anodic electrical connection
5. Volumetric pump
6. Tubular reactor
7. Feed tank
8. Tank of decantation
9. Flow meter

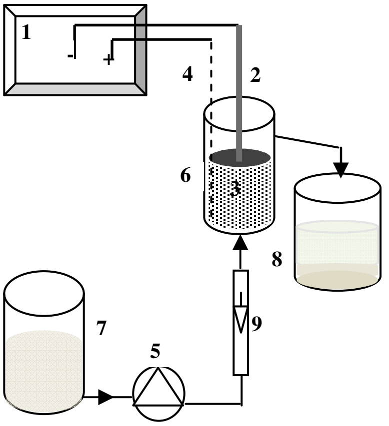


Fig. 1. Electrocoagulation pilot description

For this part of work we used a sample, taken in the same intake point that the first whose characteristics are summarized in table 3.

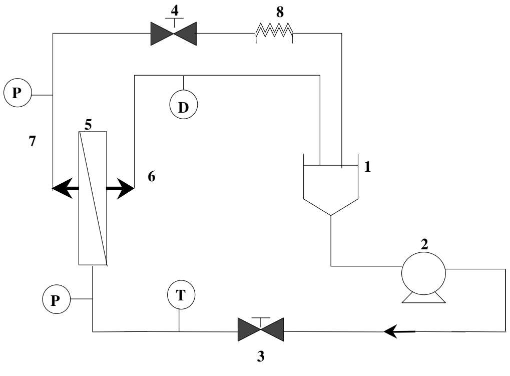
Table3. Results in continuous mode of Electrocoagulation

Sample	**pH**	**Conductivity** (mS/cm)	**Turbidity** (NTU)	**COD** (mg/L)	**m_{Al}^{the}** (g)
Sample 1					
Before EC	7.8	2.3	3.84	70	**0.80**
After EC	8.3	2.14	2.96	34.56	
Study Conditions	Volumetric flow =2.72. 10^{-3} m^3/ h ; Current density = 1.93 mA/cm^2; Time of electrocoagulation = 3.42 h.				
Sample 2					
Before EC	6.80	2.4	3.84	70	**0.53**
After EC	7.90	2.3	1.67	23.04	
Study Conditions	Volumetric flow = 3.24. 10^{-3} m^3/ h ; Current density = 1.93 mA/cm^2; Time of electrocoagulation = 2.25h				
Sample 3					
Before EC	4.5	2.43	3.84	70	
After EC	6.5	2.33	0.57	34.56	**0.47**
Study Conditions	Volumetric flow = 3.33. 10^{-3} m^3/ h ; Current density = 1.93 mA/cm^2; Time of electrocoagulation t = 2h				

Three experiments are undertaken in continuous mode. Three starting parameters are fixed which are: pH of the solution, current density and circulation flow.

2.5. Coupling of the nanofiltration and electrocoagulation

The installation of nanofiltration thus realized consists of a stainless cell, in which the spiral polyamide polysulfone membrane is maintained. The active surface of the membrane is of $0.37m^2$. The effluent is introduced into the feedback (Fig.2). In addition, the permeability of membrane being given starting from the law's Darcy and is found equal to $L_P = 10^{-6}$ m/s bars. = 10^{-11} m/s Pas.



(1) Feed tank; (2) Volumetric valve
(3) - (4) Regulator pressure valves
(5) Membrane
(6) Permeate flow; (7) Retentat flow
(8) Temperature control; (D) Flow meter
(P) Pressure control; (T) Thermometer

Fig. 2. Nanofiltration pilot description

The nanofiltration is applied to the supernatant recovered after electrocoagulation. It is applied to a pressure of 4bars after having fixed the pH of circulation respectively at 8.1; 7.9; 6.9. The test sample selection (retentate, permeate and circulation) is carried out after 20minute operation (Table 4).

Table 4. Nanofiltration test of Electrocoagulation sample

Parameters		Sample 1	Sample 2	**Sample 3**
pH	Circulation	8.13	7.86	**6.88**
	Permeat	8.35	8.34	**7.27**
	Retentate	8.38	8.35	**7.43**
Conductivity (mS/cm)	Circulation	1.73	1.81	**2.01**
	Permeat	1.45	1.43	**1.63**
	Retentate	1.70	1.79	**2.02**
Turbidity (NTU)	Circulation	0.98	0.54	**0.68**
	Permeat	0.07	0.03	**0.03**
	Retentate	0.13	0.13	**0.09**
COD (mg/L)	Circulation	44.16	38.4	**16.32**
	Permeat	26.88	13.44	**15.36**
	Retentate	46.08	34.56	**17.28**
Volumetric flow .10^3 (m^3/s)	Retentate	233	257	**300**
	Permeat	1.55	2.5	**2.72**
Ionic retention		16%	21%	**19%**

3. Results and discussion

3.1. Mode batch

Studied industrial water comes from an industrial park of which the parameters of pollution are marked by organic and mineral compounds [1-3]. In addition an analysis compared to the total coliform, fecal coliform, and enterocoq bacteria shows that their concentrations are largely higher than the standard water so that can be rejected into nature. The industrial activity which generates studied pollution results in major part of a processing industry of metals, units of textiles and units agro-alimentary.

The results make it possible to note a state of degradation advanced as well water of the Reghaia Lake [3]. Indeed, all the parameters present significant values with the majority of the concentrations exceeding sometimes largely the acceptable limits.

The applicability of electrocoagulation in this part is based on fixing of certain parameters to knowing: treated volume, the electrolysis time and variation of other parameters such as the initial pH and the current density.

In the theoretical part, we evoked the role of the pH of the solution on the elimination of the organic and mineral matter. We considered it significant to study the influence of the initial pH in this process of treatment. The results obtained are presented in Fig. 3-5.

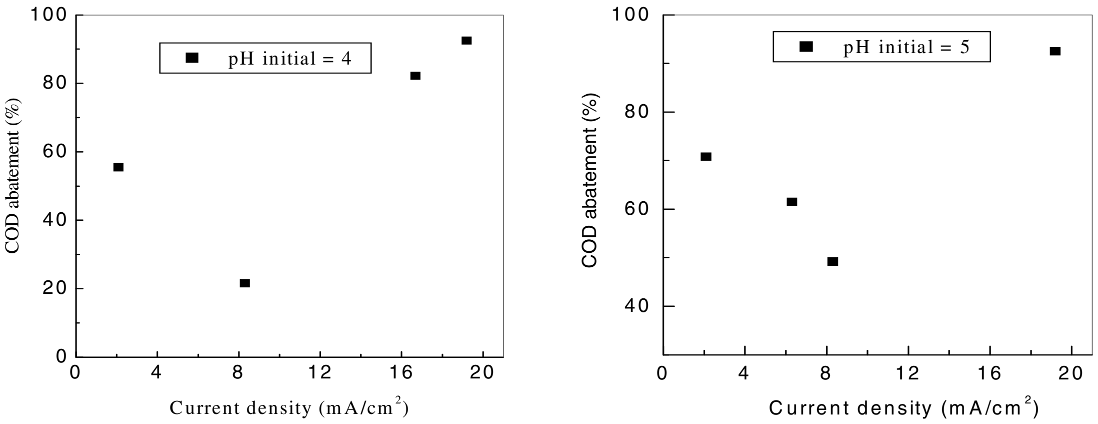


Fig. 3-4. Influence initial pH on the percentage of abatement of the COD for different density of current. (Batch mode)

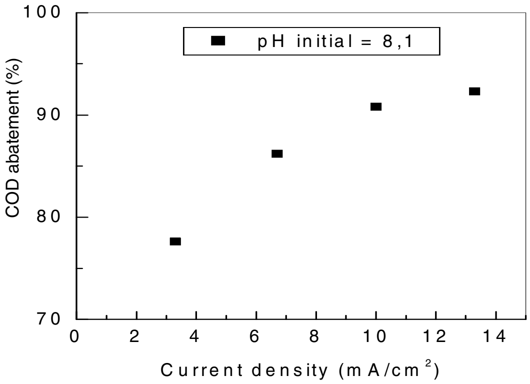


Fig. 5. Influence initial pH on the percentage of abatement of the COD for different density of current. (Batch mode)

A significant difference between the pH before and after EC is noted for the pH acid zones. While a light variation is noticed for the zone of the pH basic. In order to meet the standards of rejection for parameter pH estimated in the zone (6.5 - 8.5), an initial pH ranging between (6 - 8) and a current density lain between (4.16 - 6.66 mA/cm^2) are the favorable conditions to have a pH which meets the standard. The pH with him alone is insufficient for saying that water meets the standards of rejection. This water must be subjected to other conditions to know: Conductivity and COD…

For the zone of the basic and acid pH one notices a progressive reduction in conductivity and this same one by increasing the density of current. While for pH bordering neutrality (slightly acid), an abrupt reduction in conductivity is noticed with the strong densities of current.

The Algerian standard of rejection concerning conductivity is set for values which should not exceed 3 mS/cm. A pH ranging between (6 and 8) and a density of current which varies between (3.33-6.25) mA/cm^2 are operating conditions which meet the standard of conductivity.

The results obtained for parameter COD in batch mode EC are related to the following histograms:

It is noticed that the rate of abatement of the COD increases gradually according to the density of current, this remark is valid for the basic pH. While a fluctuating variation of the rate of abatement of the DCO according to the density of current is observed in the zone of the acid pH.

A COD strictly lower than 40 mg/L is fixed by the Algerian standard, this value can be reached for an initial pH of electrocoagulation ranging between (5-8) and for a density of current ranging between (2.08-3.33) mA/cm^2.

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3.2. Continuous mode

The results in continuous mode are given in Fig.6. One notices an increase in the pH after EC., it comes probably due to ions (OH^-) from the reduction of water to cathode. But a reduction in conductivity is observed after each EC., in the same way for the COD.

A rather significant reduction in the COD is observed for an initial pH of 6,8.

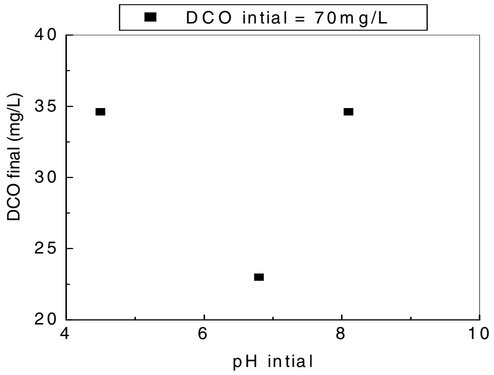

Fig. 6. Influence initial pH on the COD continuous mode of Electrocoagulation

The second test of continuous electrocoagulation mode characterized by (pH = 6,8 and current density equal at 93 mA/cm^2), seems to satisfy the standards of rejection to knowing:

- pH ranging between 6.5 and 8.5. Conductivity lower than 3mS/cm; COD lower than 40mg/L.

One notices an increase in the pH after EC., it comes probably due to ions (OH$^-$) from the reduction of water to cathode. But a reduction in conductivity is observed after each electrocoagulation, in the same way for the COD. A rather significant reduction in the COD is observed for an initial pH of 6,8.

The second test of continuous electrocoagulation mode characterized by (pH = 6,8 and current density at 1,93 mA/cm^2), seems to satisfy the standards of rejection to knowing:

- pH ranging between 6.5 and 8.5; conductivity lower than 3mS/cm; COD lower than 40 mg/L.

4. Treatment by the nanofiltration processes

4.1. pH influence pH on conductivity

To follow demineralization, we had the measurement of the conductivity of circulation, permeat and retentat**.** The reduction in the conductivity of the permeat after the membrane treatment is due to the elimination of excess of ions, ionized salts multivalent (calcium, magnesium, aluminum, sulfates), and of not ionized organic compounds of molar mass higher approximately than 250 g/mol. Monovalent salts ionized and organic compounds not ionized of mass molar lower approximately than 200 - 250 g/mol are not retained by this type of membrane, which leads us to a higher conductivity of retentat.

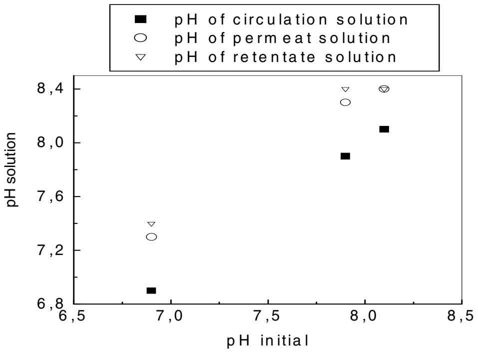

Fig. 7. Evolution of the pH of the solution in nanofiltration according to the initial pH.

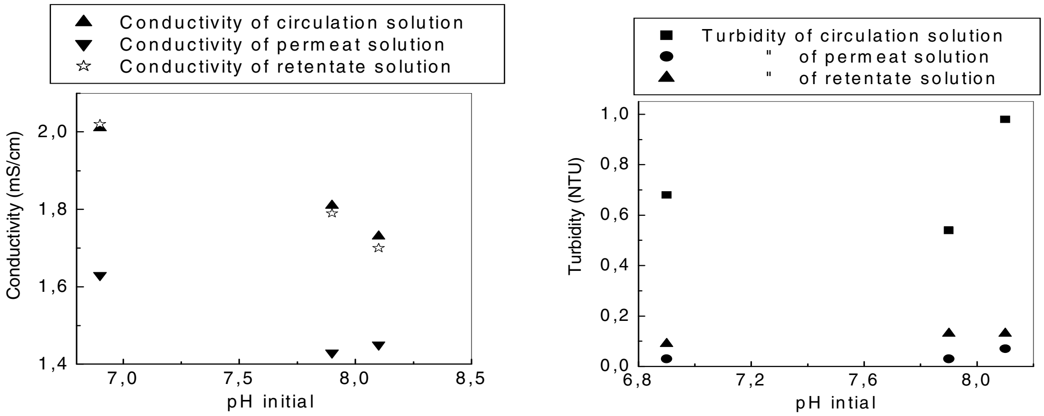

Fig. 8-9. Evolution of the conductivity and turbidity of the solution in nanofiltration according to the initial pH..

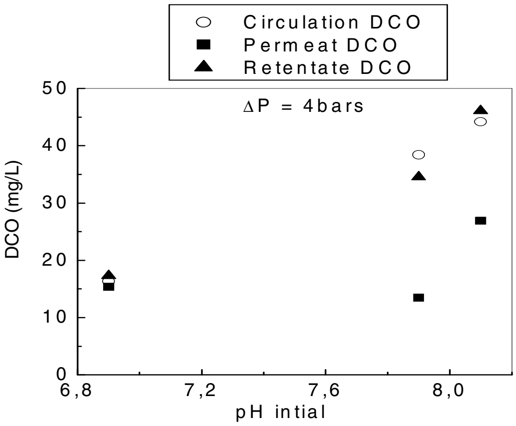

Fig. 10. Evolution of the DCO of the solution in nanofiltration according to the initial pH

The treatment by membranes Indeed plays a great role in the water treatment by increasing the percentage of abatement of the COD according to the results obtained; more than 56 % of the carbonaceous pollution expressed in COD was obtained.

4.2. Influence pH on the rate of retention

We noted that the nanofiltration carried out with pH = 7.9 give the rate of retention more raised, which reached 19.8% (Figure11).

The interpretation of these results requires the knowledge of the behavior of the active layer of the membrane and the chemical physic properties of the aqueous solutions with the change of the pH of the medium.

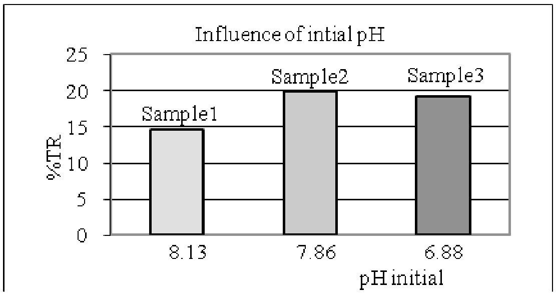

Fig. 11. Ionic retention according to the initial pH of circulation

5. Conclusion

The environmental protection for a long time became a need. Among the priorities, the safeguard of the water resources appears. All the countries of the world are concerned with the lack and the pollution water. Our work aims at the water treatment of surface and to achieve our goal, we based ourselves on the analysis and the water treatment of the Lake Reghaia which is a fish pond of natural treasure. The results of analysis (Table5) obtained which exceed the standards enabled us to note a state of advanced decomposition of water of the lake by noticing high concentrations of the parameters of study of surface water which are the COD, conductivity as we found a biomass bacterial very significant with all the harmful consequences on public health bordering, causing damage with the environment.

Table5. Parameters pollution before and after coupling of process Electrocoagulation / Nanofiltration

Parameters	Raw water	Final Treated Water
pH	7.82	7.27
Conductivity (mS/cm)	2.3	1.63
Turbidity (NTU)	3.8	0.03
COD (mg/L)	70	15.36
Fecal Enterocoques (NPP/100ml)	>2419.6	2
Total Coliformes (NPP/100mL)	>2419.6	2

This degradation reveals that the lake is confronted with a problem of pollution and harmful effects primarily related to the urban and industrial water effluents worn without treatment. That carried out us to adapt a coupling of electrochemical and membrane treatment applied to water of the lake. Initially we applied the process of electrocoagulation to eliminate several kinds of pollutants. Then, we carried out the treatment by a membrane technique of treatment, nanofiltration, which is a process of refining making it possible to balance the ions in water [27-29].

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