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Intensification of phosphate removal using electrocoagulation treatment by continuous pH adjustment and optimal electrode connection mode

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Abstract. This paper discusses two possible ways of intensifying the electrocoagulation treatment of water containing phosphate. The first one consists of controlling the chemical composition of the water by continuous adjustment of the pH. The second aspect focuses on electrode connection modes and four different modes were tested mono-polar in series, in parallels and independents and finally bi-polar. Experiments were conducted in a small batch reactor containing water with 100 mg P/L using electrodes of aluminum. Several electrical connections were used for the water treatment under favorable operating conditions determined by a study and presented in a previous paper. The influence of pH adjustment was conducted using different adjustment step times (no adjustment, every 30 minutes, 10 minutes and 5 minutes). The kinetics of the treatment was found to be accelerated by continuous adjustment of the pH. The choice of the electrode connection modes was found to strongly affect the efficiency of the treatment, the Faradic yield and the consumption of energy. The mono-polar in parallels connection mode was found to be the most economic whereas bi-polar in parallels was found to favor a higher kinetic rate of treatment.

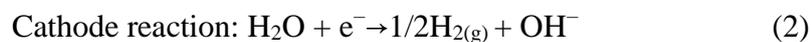
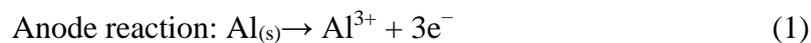
Keywords: Aluminum, electrode connection modes, pH adjustment, electrocoagulation, phosphate, intensification

1. Introduction

Electrocoagulation is an effective primary treatment for highly polluted industrial wastewater. It has been used successfully for the treatment of various industrial effluents including those from food industries [1,2], tannery wastewater [3], water containing metals or heavy metals [4, 5, 6], wastewater containing soluble oil from mechanical workshops [7, 8], polymerization manufacturing processes, and textile industries [9-12]. Over the ten last years, many publications have been

devoted to the optimization of operating parameters for the treatment of given industrial wastewaters. Most of these authors used response surface methodology or direct analysis of experimental results obtained for different operating conditions. In all cases, the operating parameters were fixed throughout the treatment. The results of these studies were often satisfactory with a high abatement of organic and colored pollution usually characterized by turbidity, Chemical Oxygen Demand (COD), Total Organic Carbon (TOC), dyes and absorbance but the researchers also successfully removed heavy metals and more generally undesirable anions and cations. This paper focuses on the specific problem of phosphate removal in water. A complete review of existing treatments including the advantages and disadvantages of each method has been published by Yeoman *et al.* [13]. They are relatively few papers (less than 10) [14-20] dealing specifically with the use of an electrocoagulation process to remove phosphate from water. The main conclusions were detailed in a recent article exploring a detailed experimental analysis of the influence of operating parameters on phosphate removal efficiency [20]. Electrocoagulation presents some advantages insofar as it is a very efficient treatment process and easy to implement. Since its efficiency is mainly affected by current intensity and the time of treatment for a given pH, it can be easily controlled online even during flow-rate or pollutant concentration variations. Moreover, the cost of the treatment is reasonable. Coagulation using chemical coagulants is the most essential process for the conventional treatment of drinking water but remains costly, difficult to control and often necessitates high consumption of chemicals. The quantity of the coagulant produced at the electrode during electrocoagulation is lower due to the easy adjustment while the cost of maintenance is lower and the produced sludge more compact [21].

The most widely used electrode materials in the EC process are aluminium and iron because of these metals' trivalent form. In the case of aluminium, the main reactions are:



Al^{3+} and OH^{-} ions generated by electrode reactions (1) and (2) react to form various monomeric species which finally transform into $\text{Al}(\text{OH})_{3(s)}$ according to complex precipitation kinetics.



The efficiency of the treatment depends on the pH. In the case of water containing phosphate, one part of the phosphate is eliminated by the formation of insoluble phosphate precipitation (see equation (4)) while another is removed by coagulation due to the formation of hydroxides of aluminium (see equation (3)). The results depend on the particular speciation of metals. When electrodes are used in aluminium, optimum coagulation and electrocoagulation are obtained around pH=7 and therefore our experiments were conducted at pH=7.

In most cases, the publications deal with the effect of operating parameters on the treatment efficiency of a given wastewater. The objective was usually to determine the best operating conditions to obtain maximum abatement with minimum energy consumption. The initial pH, electrodes gap, mean residence time (for continuous reactor) or treatment time (for batch reactor), nature of electrodes, initial concentration of pollutant, conductivity are the main parameters studied. In most cases, the anodes and cathodes were made of aluminum or iron. Katalet *et al.* [22] studied the

influence of different combinations of aluminum and iron electrodes for the treatment of paper mill wastewater and showed that the best configuration thereof varied depending on the pollutant involved. However there are very few papers focused on electrical connection systems and the online adjustment of operating parameters to intensify the treatment. The objective of this paper is to focus on these two rarely investigated aspects with a view to improving the efficiency of the treatment. The first is the online control of pH which govern the speciation of the hydroxides formed by anode dissolution while the second is the electrical connection of electrodes aimed at optimizing energy consumption.

2. Materials and methods

2.1 Experimental set-up

Fig. 1 shows the experimental set-up used in the present work and already described in detail [21]. The experiments were carried out in a jacketed batch reactor 10 cm diameter and 23 cm high. The liquid was stirred at 150 rpm with a magnetic stirrer (Fisher Bioblock). The stirring speed was kept low enough to avoid the flocsshearing. The temperature of the reactor was measured during the experiment and kept constant (30°C) by the water circulating in the jacket and heated by a thermostatic bath.

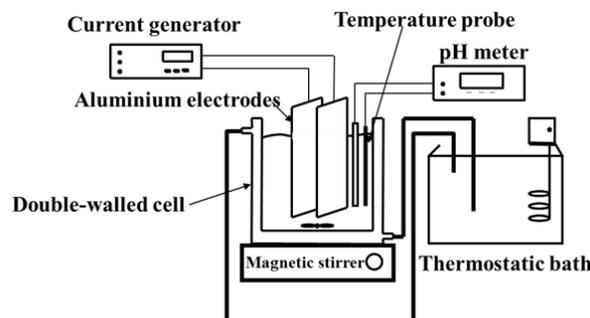
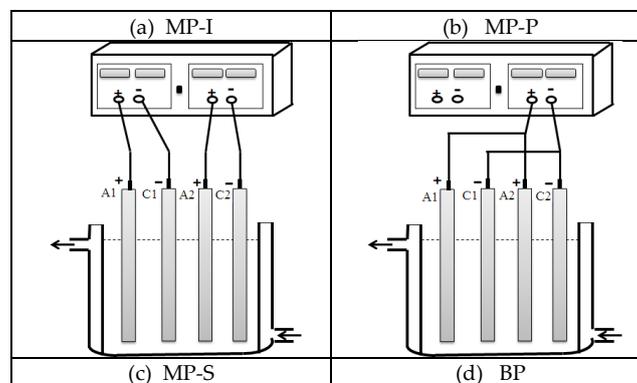


Fig. 1. Experimental set-up, the configuration with two electrodes is represented.



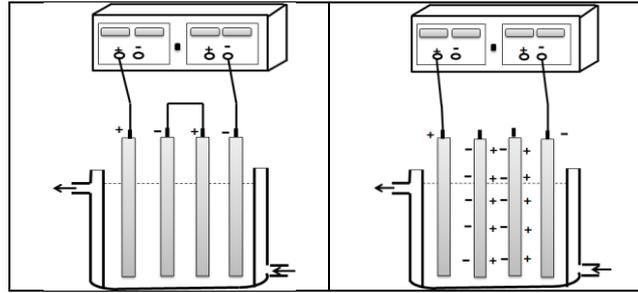


Fig. 2. Four electrodes connection mode configurations

The electrodes are vertically immersed in the solution for an active surface of 50 cm^2 ($5 \times 10 \text{ cm}^2$). The distance between anode and cathode was set at 5 mm as previously suggested [20]. After each experiment and prior to the next experiment, the electrodes were pumiced and degreased with 400 grit super fine sandpaper and rinsed with 0.1 M HCl and clean water to avoid any effects due to the history of the electrodes. The electrodes were connected to a direct current power supply (ALR 3002 M) delivering current up to 2.5 A with a voltage ranging from 0 to 30 V. Electrochemical coagulation experiments were carried out under galvanostatic conditions. The time $t=0$ of the run corresponds to switching on the current supply.

Several electrical connecting configurations with 4 electrodes were tested as illustrated in Fig. 2. Three monopolar configurations were used - Independent (MP-I), Parallels (MP-P) and in Series (MP-S) and a Bipolar configuration. These are shown in Figures 2a, 2b, 2c and 2d respectively. The results of the experiments using only two electrodes have been already published [21] and are used on occasions in the comparison presented below.

2.2 Solutions and chemical analysis

The phosphate synthetic solution 100 mg P/L was prepared by dissolving 0.439 g of potassium dihydrogénophosphate KH_2PO_4 (Prolabo) per liter of distilled water. The initial pH was adjusted with $2 \text{ mol.L}^{-1}\text{NaOH}$ (Chemi-pharma) or with $3 \text{ mol.L}^{-1}\text{HCl}$ (Reagent grade ACS, ISO Chemicals Developing and Manufacturing (CDM)) solutions. The conductivity of the solution was fixed at 1 mS.cm^{-1} by the addition of 0.38 g.L^{-1} of sodium chloride (Chemi-pharma).

A UV spectrophotometer (HACH DR/4000U) at 430 nm was used for phosphate analyses in accordance with the standard vanomolybdophosphoric acid calorimetric method [23] after separating the particles through the $0.45 \mu\text{m}$ mixed cellulose ester syringe filters.

The conductivity was measured continuously using a consort D292 conductimeter. pH and temperature measurements were performed continuously using a pH meter (HANNA instruments, pH 213).

A plasma emission spectrometry ICP / AES (HORIBA) was used for residual aluminum analysis.

3. Results and discussion

3.1 Influence of time treatment

Before analyzing the influences of the pH adjustment online and electrical connections, it is necessary to first discuss again certain explanations of the influence of treatment time even though this has been detailed in a previous paper [20]. In fact, although many authors present the influence of time of treatment and current intensity separately, the production of hydroxide is linked to the charge loading. This means that efficiency can be improved by increasing the current intensity for a fixed treatment duration or by increasing the time of treatment for a given current intensity. At fixed current (here 0.5 A), the time of electrocoagulation determines the amount of Al^{3+} ions released from the electrode. At the beginning, the elimination of phosphate increased with time and progressively the efficiency rate decreased until it reached the maximum percentage of abatement. The time taken to reach this value represents the kinetics of the process and, depending on the pollutants, it is possible to obtain a fast kinetic rate with a limited percentage of abatement or a low kinetic rate until the pollutant is eliminated completely. Our main dual objective was to improve firstly the kinetics and secondly energy consumption to obtain the maximum percentage of phosphate elimination.

3.2 Influence of continuous adjustment of pH

It has often been reported that an optimal initial pH is essential for efficient electrocoagulation treatment [24]. There are numerous experiments in literature on the subject which analyze the effect of the initial pH on the final abatement of the pollution. It has been shown in a previous paper [21] that an acid pH favors the kinetic of abatement because of the predominant form of $\text{AlPO}_4(\text{s})$ except for the very low value of $\text{pH}=2$. When the pH is higher, both phosphate precipitation and adsorption on aluminum hydroxides compete. The kinetics of adsorption is slower than the kinetics of precipitation. When the pH increases, the formation of $\text{AlPO}_4(\text{s})$, and $\text{Al}(\text{OH})_3$ decrease and abatement is limited. Lower pH values are thus preferable both from efficiency and maintenance standpoints. It is thus interesting to adjust the initial pH to obtain the higher abatement of the target pollutant. Usually the initial pH is fixed and its value changes progressively with the time evolution of the reactions. Xu *et al.* [25] demonstrated the relation between pH and Chromium concentration during the electrocoagulation process and proposed a real time control strategy. Our idea was to continuously adjust the pH to keep the reactive volume in optimum chemical conditions to favor the desired speciation. In this section, we maintained the pH at the optimal value of 7 to intensify the treatment rate since the preponderant form of aluminum is $\text{Al}(\text{OH})_3$. Three step times were tested to adjust the pH - at 5, 10 and 30 minutes as illustrated in Fig.3. Fig. 4 shows that the regular adjustment of pH to the desired value helps improve the kinetics of phosphate removal abatement. The results obtained with an adjustment every 5 minutes and every 10 minutes was found to be very close which means that the online control does not need to be overly strict. Moreover, the residual concentration of aluminum was found to be lower with the adjustment of pH (0.28 mg.L^{-1} with a regular adjustment of pH every 10 minutes and 0.46 mg.L^{-1} without pH adjustment).

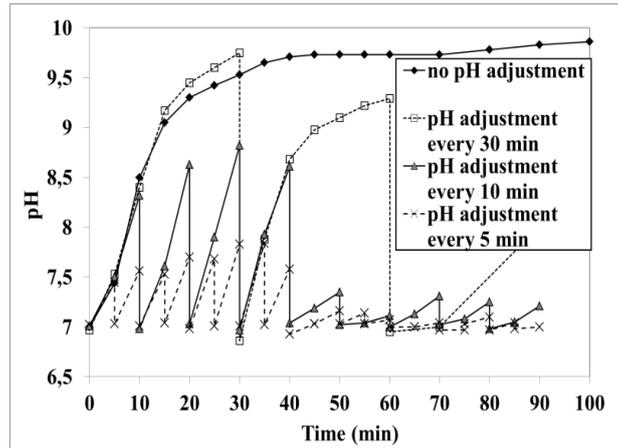


Fig. 3. pH versus time for different types of continuous pH adjustment experiments ($[PO_4^{3-}]_0 = 100$ mg P.L⁻¹, $[NaCl] = 6.5$ mM, $pH_i = 7$, $T = 30^\circ C$, $i = 10$ mA.cm⁻²).

The total removal of phosphate was obtained after 140 minutes without pH adjustment and after only 40 minutes with adjustment of pH every 10 min. Moreover, as table 1 shows, the quantity of aluminum dissolved to fully remove the phosphate with a pH adjustment every 10 minutes is 3.5 times less than without pH adjustment.

The pH can easily be continuously adjusted at the industrial scale and will result in a lower consumption of energy and sludge production for the maximum of efficiency.

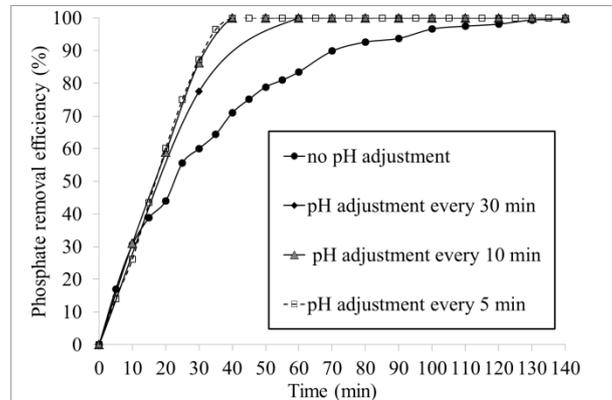


Fig. 4. Phosphate removal efficiency versus time for several pH adjustment periods ($[PO_4^{3-}]_0 = 100$ mg P.L⁻¹, $[NaCl] = 6.5$ mM, $pH_i = 7$, $T = 30^\circ C$, $i = 10$ mA.cm⁻²).



Fig. 5. Visual aspect of the water after treatment (5a left side) without pH adjustment (5b right side) and with continuous pH adjustment every 30 minutes ($i = 10 \text{ mA} \cdot \text{cm}^{-2}$, $[\text{PO}_4^{3-}]_i = 100 \text{ mg P} \cdot \text{L}^{-1}$, $[\text{NaCl}] = 6.5 \text{ mM}$, $\text{pH}_i = 7$, $T = 30 \text{ }^\circ\text{C}$)

The continuous adjustment of pH also affected the separation process. Fig. 5 shows the effluent after treatment without (Figure 4a left side) and with adjustment of pH every 30 minutes (Fig. 4b right side). Without pH adjustment, the volume of sludge was found to be very large and relatively uncompact. This was due to the gelatinous structure of some hydroxides at basic pH.

3.3 Influence of electrical connections

The influence of the nature of the electrodes has been studied several times although there are relatively few studies regarding the efficiency of different types of electrodes to treat the same effluent. As an illustration, Llanos *et al.* [26] compared the efficiency of iron and aluminum bipolar electrodes on the disinfection process using electrocoagulation. The relative efficiency of aluminum or iron depends on the pollutant target. Recently, several research studies have been run using different metals for the anode and cathode.

	Time (min)	Mass of dissolved anode	Mass of dissolved cathode	$[\text{Al}]_{\text{dissolved}}$ ($\text{g} \cdot \text{L}^{-1}$)
10-minute period adjustment	40	0.128	0.036	0.164
No adjustment	140	0.461	0.121	0.582

Table 1. Quantity of dissolved aluminum electrodes and aluminum concentration in the solution for optimal time of treatment under 10-minute period adjustment and with no pH adjustment

However, the influence of the connecting mode has been rarely evaluated. Solaket *et al.* [27] evaluated the monopolar-parallel and monopolar-serial connections using both aluminum and iron electrodes during the treatment of marble processing wastewater but they focused much more on the electrode material. Recently, Vasquez *et al.* [28], studied the impact of primary potential and current density distribution on electrocoagulation. They showed that the non-distribution of current density impacts coagulant formation and thus the efficiency of electrocoagulation treatment. In this paper, we tested four different electrical connected modes using the same material to evaluate how

this can affect the efficiency of electrocoagulation treatment. Figure 6 shows the phosphate removal efficiency versus time for the different connection systems. The bipolar mode is much more efficient compared with monopolar connections systems. This complies with the results obtained by Ghosh *et al.*[29] for the treatment of water containing fluorides.

The classical system featuring 2 electrodes which is regularly chosen in most studies gives a poor level of efficiency when compared with four-electrode systems.

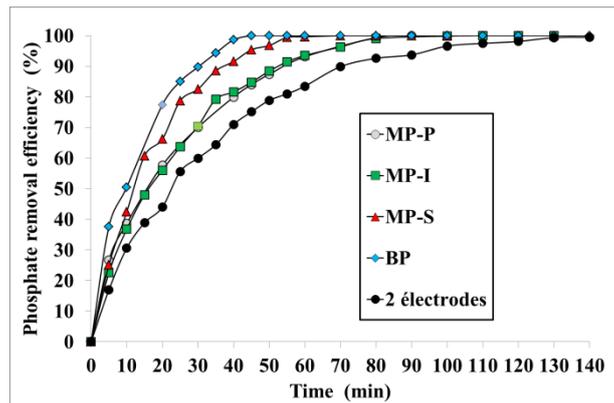


Fig. 6. Phosphate removal efficiency versus time for the five different connection systems ($[PO_4^{3-}]_0 = 100 \text{ mg P.L}^{-1}$, $[NaCl] = 6.5 \text{ mM}$, $pH_i = 7$, $T = 30^\circ\text{C}$, $i = 10 \text{ mA.cm}^{-2}$).

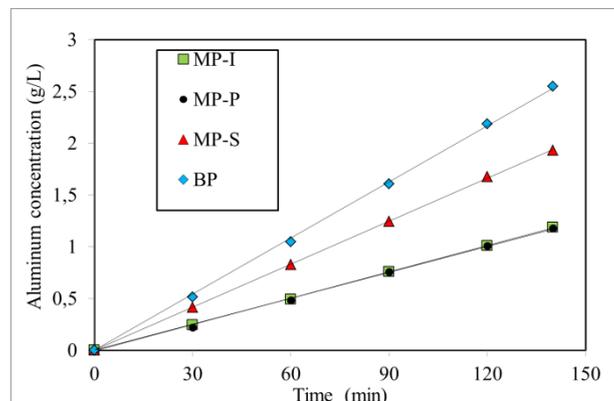


Fig. 7. Aluminum concentration versus time for the four electrode connection systems ($[PO_4^{3-}]_0 = 100 \text{ mg P.L}^{-1}$, $[NaCl] = 6.5 \text{ mM}$, $pH_i = 7$, $T = 30^\circ\text{C}$, $i = 10 \text{ mA.cm}^{-2}$).

Fig. 7 represents the aluminum concentration versus time for the four different connection systems. The quantity of dissolved aluminum varies greatly from one connection to another. These differences may be explained by the varied current distributions and the possible higher formation of hydroxides by over-dissolution of anode for MP-S and BP connections. The measured concentration corresponds to a Faradic yield of 1.15 for MP-P and MP-I, around 2 for MP-S and 3 for B-P. Moreover the sludge was found to be much more compact for the B-P connection mode. Nevertheless, the energy consumption would need to be estimated for the different connecting modes since the measured cell voltage between the electrodes may be different.

3.4 Estimation of energy consumption and operation cost

Electrical energy consumption was calculated in terms of Kwh per m³ of treated effluent using the equation given below:

$$C_{energy} (Kwh. m^{-3}) = \frac{U.I.t_{EC}}{V} \quad (4)$$

where U is cell voltage (V), I is current (A), t_{EC} is the time of electrocoagulation treatment (s) and V is the volume (m³) of effluent to be treated. Fig. 8 represents the time evolution of energy consumption per unit of treated volume for the different connection systems. While the BP mode is better from the kinetic standpoint, energy consumption is lower using the MP-P connection. In fact the tension voltage between the electrodes is lower as illustrated Fig. 9. This result is consistent with the findings of Kobya et al. [30] obtained in the case of the treatment of potable water containing a low concentration of Arsenic.

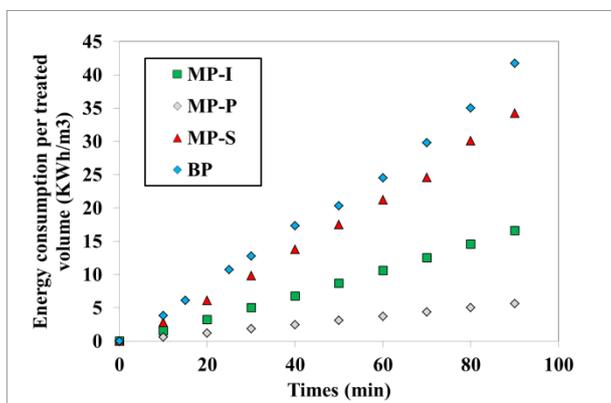


Fig. 8. Energy consumption versus time for the 4 electrodes different connection systems ($[PO_4^{3-}]_0 = 100 \text{ mg P.L}^{-1}$, $[NaCl] = 6.5 \text{ mM}$, $pH_i = 7$, $T = 30^\circ\text{C}$, $i = 10 \text{ mA.cm}^{-2}$).

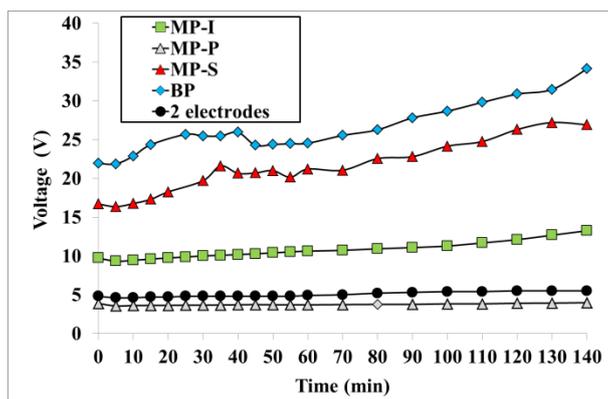


Fig. 9. Voltage versus time for the 5 electrodes different connection systems ($[PO_4^{3-}]_0 = 100 \text{ mg P.L}^{-1}$, $[NaCl] = 6.5 \text{ mM}$, $pH_i = 7$, $T = 30^\circ\text{C}$, $i = 10 \text{ mA.cm}^{-2}$).

These results show that more attention is required when setting up electrical connection systems. This is particularly the case for continuous industrial reactors in which numerous electrodes are used. It is clear that most of the energy consumption figures published in the literature and based on simple cathode-anode system are overestimated. The benefit of the design of an electrode

connection remains to be confirmed and validated in the case of complex industrial wastewater since the few papers dealing with this aspect focus on the low concentration of fluoride [29], Arsenic [30] and phosphate (this paper).

The real operating cost includes electrodes, electrical energy cost, as well as labour, maintenance, sludge dewatering and disposal and fixed costs [28]. Electrodes and electrical energy costs are specific to the electrocoagulation methods and as they are the major cost items, a preliminary economic investigation can be done using the following relation [28]:

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrodes}} \quad (5)$$

Where C_{energy} is the energy consumption calculated by equation (4) and $C_{\text{electrode}}$ is the electrode consumption to treat 1 m³ of effluent calculated in first approximation by equation (6):

$$C_{\text{electrodes}} = \frac{I.M.t_{EC}}{n.F.V} \quad (6)$$

where M is the molecular mass of aluminum (26.98 g.mol⁻¹), n is the number of electrons transferred (n=3), F is Faraday's constant (96487 C.mol⁻¹). When available, it is better to estimate the real consumption of aluminum since the Faradic yield is often higher than 100%.

The equation (5) has two parameters which are strongly variable from one week to the next. The first parameter "a" is the cost of electricity and second parameter "b" is the cost of aluminum. These also depend on the country involved's metal and electricity production and costs. A reasonable average estimation of operating cost for the French market can be made using a = 0.07 euros per Kw.h⁻¹ and b=0.2 euros per Kg. The best operating conditions to treat the water were obtained with a current intensity of 0.5 A for 40 minutes of treatment. In these conditions, the average voltage during the treatment was 6.37 V. The consumption of aluminum obtained by relation (6) is 0.11 Kg.m⁻³, whereas the experimental result was 0.164 Kg.m⁻³ (see table 1). This is why theoretical estimation often underestimates the real consumption of aluminum. The final operating cost to treat one cubic meter of water is around 0.45 euros.m⁻³ with 40% for electrical costs and 60% for the electrodes.

4. Conclusions

The treatment of industrial wastewater and water by electrocoagulation is very efficient for a wide range of pollutants. Many experimental studies in the literature have aimed to determine the best fixed operating parameters to treat a given wastewater (pH, temperature, conductivity, current intensity, pollutant types and concentration level, nature of electrodes...). The aim of this paper was to optimize the treatment of water containing phosphate using two options which have been the subject of very little study - continuous pH adjustment and the selection of the optimal electrical connection mode.

Four connection modes were studied and notable important differences were observed both from efficiency and energy consumption standpoints. The bi-polar connection had the best results in terms of phosphate removal rate but, from the energy cost point of view, mono-polar in parallel was more efficient. The continuous adjustment of the pH can greatly improve efficiency compared to a fixed value. Treatment time under the tested operating conditions was reduced from 140 to 40 minutes by pH adjustment to the optimum value of pH=7 every 10 min. From the industrial point of view, online adjustment of pH value is credible and easy to implement and treatment cost is relatively low. Moreover, the sludge is more compact and the residual level of aluminum in the

treated water is lower. These first results would need to be confirmed for other types of wastewater but are still promising for intensification of the treatment. To achieve the same goal of finding new ways to improve treatment, different types of alternating currents should be tested to evaluate how these can also improve treatment performance levels.

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