PERSPECTIVES OF ELECTROCOAGULATION IN WATER DISINFECTION

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Abstract: Today effective electrochemical disinfection system seems to be an alternative to conventional chlorination, ozone or UV processes. This paper reports on a series of experiments evaluating the disinfection efficiency of electrocoagulation cell using Al electrodes. Solutions contaminated by *E. coli* and surface waters were used as test media. The process significantly outperformed chemical coagulation reducing the amount of aluminum used. In all cases, the treated solutions were effectively disinfected and almost total removal of coliforms and algae were rapidly performed. A relationship for the disinfection rate of *E. coli* cells has been proposed. This equation takes into account the electrophoretic migration of the cells and the subsequent release of discharged cells into the bulk.

1 INTRODUCTION

Nowadays water quality and available quantity are being challenged by increased pollution from various sources such as industry and agriculture. Innovative, cheap and efficient methods of purifying water as well as to clean wastewaters from industrial effluents are needed. Electrochemical techniques such as electroflotation, electrodecantation or electrocoagulation offer the possibility to be easily installed and they require few chemicals. According to Rajeshwar et al. (Rajeshwar 1994) benefits from using electrochemical techniques include: environmental compatibility, versatility, energy efficiency, safety, selectivity and cost effectiveness. In addition to these, the systems employ only electrons to facilitate water treatment. Of the known techniques, there is a considerable interest in using electrocoagulation. The process is well known for the treatment of suspended particles (Domini 1994, Abuzaid 2002), phosphates (Idermez 2006a, Idermez 2006b) and oil-in-water emulsions (Canizares 2008 , Asselin 2008, Un 2006). Another approach of the electrocoagulation is water disinfection where this technique can reduce notably the amount of chlorine used. It has been reported that electrochemical and

magnetic fields can destroy a wide variety of microorganisms from viruses (Zhu 2005) to bacteria (Dao 2004, Li 2004).

In this study, *Escherichia coli* cells were used to show the disinfection effect of electrocoagulation using aluminum electrodes. The technique was then used to treat total coliforms and algae.

2 THEORETICAL

Electrocoagulation is a process involving many chemical and physical phenomena that use soluble ("sacrificial") anode to supply ions into the treated water. In the electrocoagulation process the coagulation ions are produced *in situ*. The process involves three successive stages: coagulant formation by electrolytic oxidation of the soluble anode; contaminants destabilization and emulsion breaking; aggregation of the destabilized phases leading to floc formation.

In the case of an electrocoagulation, the reactor contains one cathode and one anode (Figure. 1). When a potential is applied with a current generator, the anode material undergoes oxidation, while water

reduction occurs at the cathode. The reactions can be presented as follows:

At the anode: $\qquad M_s = M_{aq}^{n+} + ne^-$

$$2H_2O = 4H^+ + O_2 + 4e^-$$

At the cathode: $\quad 2H_2O + 2e^- = H_2 + 2OH^-$
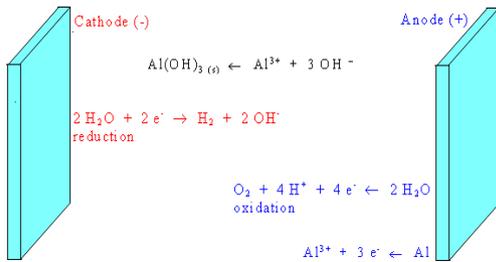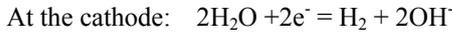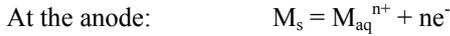
Figure 1: Schematic diagram of an electrocoagulation cell.
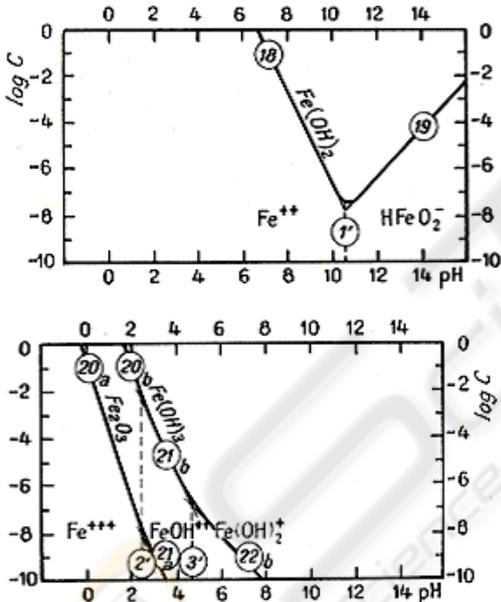
Figure 2: Influence of pH on the solubility of iron hydroxides.

Generally iron or aluminum electrodes are proposed both allowing good process performance. These metal ions generated in the solution react in solution and afford corresponding hydroxide and/or polyhydroxide compounds. These compounds have strong affinity for suspended particles, droplets and eventually bacteria to cause coagulation. In fact, the choice of the sacrificial electrode is a very important parameter which depends on the operating conditions, the goal being to use all the metal hydroxides dispersed in the solution. The solubility

of these hydroxides depends on the pH of the treated solution as it can be seen in Figure 2 and Figure 3.
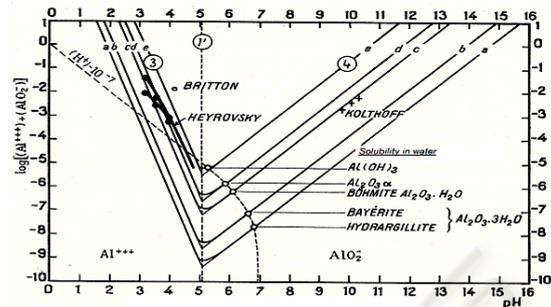
Figure 3: Influence of pH on the solubility of aluminum hydroxides.

It is clear that aluminum hydroxides have a lower solubility than ferrous hydroxide, and therefore a better efficiency in wastewater treatment. This is why aluminum electrodes have been used in this study.

When aluminum electrodes are used, the generated Al_{aq}^{3+} ions undergo spontaneous reactions to generate:

Mononuclear complexes :
$Al(OH)_2^+, Al(OH)_4^-, Al(OH)^{2+}$
Polynuclear complexes : $Al_2(OH)_2^{4+}, Al_2(OH)_5^+, Al_6(OH)_{15}^{3+}, Al_{13}(OH)_{34}^{5+}$
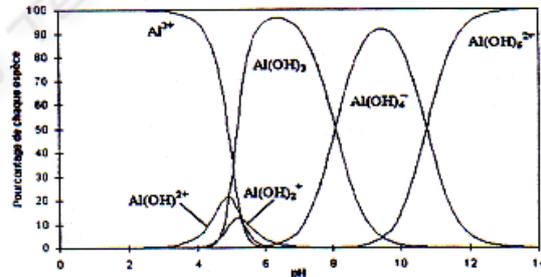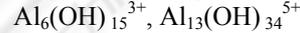
Figure 4: Al hydroxides and polyhydroxides species as function of pH.

For pH between 5 and 8, the predominant specie is $Al(OH)_3$ (almost 95% when pH = 6.5) with small amounts of $Al(OH)_2^+$ and $Al(OH)^{2+}$. These compounds show strong affinity for counter ions to cause coagulation. The gases evolved at the electrodes may impinge on and cause flotation of the coagulated material. It can be assumed that the process may be summarized as follows: charge neutralization resulting from the production of counter ions during the electrochemical dissolution of the sacrificial anode, van der Waals interactions followed by coagulation and floc formation.

In the present study we are presenting results obtained with artificial and real aqueous solutions for *E. coli* and indigenous flora to illustrate the possibilities offered by an electrochemical disinfection process using aluminium anode.

3 EXPERIMENTAL

3.1 Artificial Solutions containing E. Coli

These solutions were prepared by diluting an *E. coli* culture in a PCA medium. The *E. coli* was grown on a shaker at 37°C for 24 hours and then 100 µl were used to inoculate the medium.

Surface waters were obtained from local river and pool.

E. coli, total coliforms and algae were counted in accordance with standard methods.

3.2 Pilot Plant
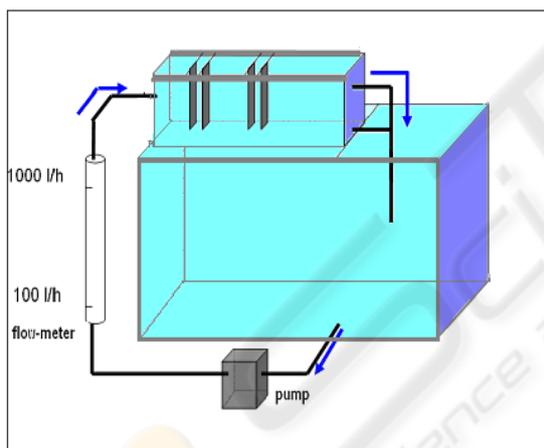
Figure 5: Schematic diagram of the bench-scale reactor with monopolar electrodes.

The experiments were performed with two types of reactor. The first one was composed of two electrodes which were plunged in a beaker (V = 1 L). Different distances between the electrodes have been tested (1, 2 and 4 cm). The electrodes ware connected to a direct current power supply able to furnish a controlled tension or current up to 30 V or 10 A, respectively. Applied tension, current, temperature, conductivity and pH were measured. After each run, the electrodes were dried and weighted.

The obtained results have been used to build the pilot plant shown in Figure 5. It consists of an electrocoagulation reactor (V = 2 L) having two pairs of monopolar aluminum electrodes and a decantation chamber with V = 10 L. The flow in the pilot is controlled with a centrifugal pump (Prolabo, France). All tests were performed at a constant flow rate of 100 $L.h^{-1}$(flow velocity of 1.85 x 10^{-3} $m.s^{-1}$).

4 RESULTS AND DISCUSSION

4.1 Comparison between Coagulation and Electrocoagulation

In order to illustrate the interest of the technique, a comparison between the chemical coagulation and the electrocoagulation is presented in Table 1. The chemical coagulation was performed using the same amount of aluminum as in the electrocoagulation. It can be seen that the electrocoagulation improves considerably thee bacteria removal. Using the same amount of aluminium a 3-log (99.9%) reduction can be achieved in this case, while only 1-log bacteria reduction was reached with the chemical coagulation in the same Al dosage range.

Table 1: Comparison between chemical and electro-coagulation (Coa = coagulation).

	Coa 1	Coa 2	Coa 3	Electo coagulation
T0 (CFU/mL)	5.75E+04	2.38E+04	8.30E+04	3.00E+05
Tf (CFU/mL)	9.75E+03	1.12E+04	1.40E+04	5.40E+02
E (%)	83	53	83	99.9

The fact that the electrocoagulation significantly outperformed chemical coagulation deserves explanation. According to Oss (Oss 1994) bacterial adhesion to surfaces results from the Lifshitz-Van der Waals interaction free energy and the Lewis acid-base interaction free energy. Bacteria either donate or accept electrons to the surface of the substrate (in this case the gas bubbles). Adhering bacteria may decrease electrostatic repulsion

Table 2: Reduction of the cellular concentration of *E.coli* as function of the gap between the electrodes.

Distance	1 cm			2 cm			4 cm	
Run N°	1	2	3	1	2	3	1	2
T_0(CFU/mL)	6.60E+04	1.20E+04	3.95E+04	1.60E+04	5.35E+04	2.48E+04	2.50E+04	5.00E+04
T_f(CFU/mL)	3.50E+02	9.30E+02	6.35E+02	7.70E+01	2.85E+03	4.45E+02	8.30E+01	1.70E+01
E (%)	99.5	92	98	99.6	95	98	99	99
N/N_0	0.0053	0.0775	0.0161	0.0048	0.0518	0.0179	0.0033	0.0003

allowing floc formation. The charge transfer, however, takes place over a range shorter than 0.5 nm, so close contact is needed. This contact is realized easily in electrocoagulation process where the negatively charged bacteria could electrophoretically move resulting in higher bacteria concentrations near the positively charged anode. At the same time, the coagulation (chemical or electrochemical) creates a sludge blanket that entraps and can bridge colloidal particles as well as bacteria still remaining in the aqueous solution.

At the same time, aluminum cation is generated continuously over the extended area of the anode in electrocoagulation compared with the point addition in chemical coagulation. So aluminum is more effectively used. Thus, locally higher coagulant and bacteria concentrations might have been one reason of the better performance of electrocoagulation compared to chemical coagulation.

4.2 Influence of the Gap between the Electrodes

The tests were performed at an initial pH = 7.2 at 15.6°C. The initial conductivity of the medium was 0.4 mS/cm. The applied tension was 30 V and the current 0.22 A. The electrocoagulation time was estimated according to the well-known sterilization chart assuming a first order reaction for the cell elimination and an imposed efficiency of disinfection of N/N_0 = 0.001. The experiment duration was 30 min. Three distances between the electrodes have been tested: 1, 2 and 4 cm. The obtained results are presented in Table 2.

As it can be seen, the best results have been obtained with the distance of 4 cm where a 3-log reduction was always obtained so this distance was used in the experiments with real effluents. These results show that electrical phenomena occurring in the solution, and particularly the ion electromigration, are involved in the disinfection process, may be thanks to an electroporation mechanism.

4.3 Coagulation Rate in a Batch Electrocoagulation

The electrocoagulation of *E. coli* cells in a batch continuously flowing system consisting of two cells in series has been studied using the pilot plant shown in Figure 5. The results given in Figure 6 show the reduction in cell concentration in time as well as the coagulation rate. The coagulation rate can be described by a first order rate equation (1):

$$dC/dt = k_1 e^{-k_2 t} \tag{1}$$

where C is the concentration at time t, $k_2 = 4kT/3\mu$ (k is the Boltzman's constant and μ the viscosity of the aqueous medium), k_1 depending on the electrophoretic velocity of the cells and the geometric parameter of the electrode size. This equation takes into account the electrophoretic migration of the cells and the subsequent release of discharged cells into the bulk. According to Matteson et al. (Matteson 1995), the constant terms in this equation derive from the basic electrophoretic and coagulation phenomena, which are depending on the zeta potential of the cells, the applied voltage, the initial concentration and the electrode geometry.
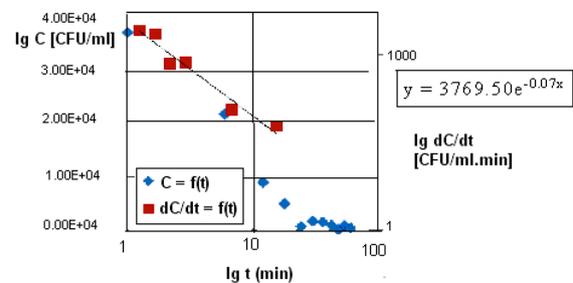
Figure 6: Cell reduction and coagulation rate for *E. coli* cells.

4.4 Electrocoagulation of Surface Waters

In order to confirm the electrocoagulation performances, two surface waters were studied. A river water (conductivity *0.4 mS/cm, pH = 7.2)* containing indigenous coliforms and a pool water (conductivity *0.55 mS/cm, pH = 6.8*) containing algae were used. The former was disinfected at *I = 0.35 A*, the latter at *I = 0.22 A*. In both cases the applied tension was 30 V. In both cases after 10 min a froth layer appears at the surface increasing in time with flocs floating on the surface. The obtained results after 30 min of electrocoagulation are given in Table 3. They show a total coliform removal and a good disinfection efficiency for the algae population.

Table 3: Coliform and algae reduction in surface waters.

	River water			Pool water	
T_0 (CFU/mL)	1.12 E+02	1.12 E+02	1.12 E+02	4.15 E+04	6.05 E+05
T_f (CFU/mL)	0	0	0	2.50 E+02	2.20 E+02
E (%)	99.99	99.99	99.99	99.4	99.6

It can be assumed that the applied current originate potential difference from an extremity to the other of the cellular membrane on account of its electrical resistance. This potential difference modifies consequently the trans-membrane potential producing destruction of the cellular membrane (Li 2004). The cell membrane is constituted essentially by layers of phospholipids and proteins can be included allowing ionic change with the cell environment. Proteins are easily destroyed by direct effect of an electric field while the phospholipidic membrane is not easily oxidable. So, some cells can be reactivated in a favorable medium. This is probably the case of the algae present in the solution.

5 CONCLUSIONS

The effects of disinfection of *E. coli* cultures, coliforms and algae using aluminum electrocoagulation were studied. The process significantly outperformed chemical coagulation reducing the amount of aluminum used. In many cases, *E. coli* cells in the treated model solutions were effectively disinfected and almost total removal of coliforms and algae were rapidly reached. A relationship for the coagulation rate of *E. coli* cells has been proposed. This equation takes into account the electrophoretic migration of the cells and the subsequent release of discharged cells into the bulk.

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