Electrocoagulation of *Escherichia coli* Culture: Effects of Temperature and Cell Concentration

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Abstract

Laboratory tests were performed to examine electrocoagulation (EC) as electrochemical disinfection of synthetic wastewater infected by non-pathogenic *Escherichia coli* species in batch culture and two surface waters employing ordinary steel, stainless steel and aluminum electrodes. Aluminum electrodes were observed most performant in killing *E. coli* cells relatively with stainless steel and ordinary steel electrodes. About thirty minutes are requested for EC to attain total *E. coli* cell elimination. Identical performance toward algae and coliform removal in two kinds of surface waters was noticed. EC technology relies on charge neutralization of pathogens via electrical field application and Al^{3+} or $Fe^{2+}/^{3+}$ followed by their flotation thanks to hydrogen and/or oxygen bubbles or flocculation/decantation thanks to $Al(OH)_{3(s)}$ or $Fe(OH)_{2(s)}/Fe(OH)_{3(s)}$ flocs.

Subject Areas

Chemical Engineering & Technology

Keywords

Electrocoagulation (EC), Electrochemical Disinfection (ED), Electric Field (EF), Mechanism, Pathogens, Water Treatment

1. Introduction

During the last decades, there is no doubt that water and wastewater treatment industry has known a marked advance [1] [2]. However, water contamination

has likewise greatly augmented due to the uncontrolled industrial expansions [3] [4] [5]. As a result, the at-hand water resources became contaminated with a large range of contaminants generating various health issues [6] [7]. Some of such contaminants, the pathogenic and non-pathogenic microorganisms are categorized as the most elevated danger than the remaining contaminants because of the towering cases of illness and death that they could provoke [8] [9] [10] [11]. For example, it has been mentioned that the pathogens trigger diverse waterborne diseases, like diarrhea and gastrointestinal, which successively lead to around 2,000,000 deaths/year [8] [12]. Thus, several disinfection techniques, like chlorination [13] [14] [15] [16], ozonation, and irradiation with ultraviolet, have been employed to kill pathogenic and non-pathogenic microorganisms from water [17]-[24]. As an illustration, the chlorination process has been largely employed in the course of the 1970s as an efficacious and low-cost disinfection technique. During this chemical method, the strong oxidizing capacity of chlorine destructs the fundamental enzymes of microbes, which conducts to killing such biological contaminants [25] [26]. The primary drawback of the chlorination process remains the formation of very poisonous disinfection by-products. For example, the reaction between chlorine and natural organic matter (such as humic substances [27]-[32]) forms trihalomethanes, which are famous as carcinogenic chemicals [25] [26]. Membrane processes, such as microfiltration and ultrafiltration techniques, are additionally efficient barriers to eliminating microbes; nevertheless, their implementation is considerably restricted via the fouling issues and the elevated operational cost [1]. Ozonation is a different method that has been utilized as a disinfection technology; indeed, it has been noted that the ozone is a strong oxidant that can demobilize the microbes via breaking down their cell membrane [25] [33]. Even if the ozonation method is very efficacious and it does not form trihalomethanes like chlorination, it remains costly as contrasted to various conventional techniques [34]; further, it has been established that it may provoke the production of N-nitrosodimethylamine [33].

Lately, disinfection engineering has known outstanding progress through merging several techniques or via employing novel composite materials. For instance, researchers [35] suggested a fresh disinfection process that employs a tubular coaxial-electrode copper ionization cell to disinfect drinking water. The acquired findings depicted that this technique eliminated 6-log of *Escherichia coli* during 2 min of application at a running voltage of 1.5 V. Scientists [36] employed an advanced electrochemical cell that was furnished with boron-doped diamond electrodes to disinfect seawater. The results of this investigation proved that this advanced electrochemical cell reduced 4.8-Log of natural marine heterotrophic bacteria at an energy consumption of 0.264 kWh/m^3. The same researchers [36] implemented nanotechnology [37] to present a disinfection technology that comprises an anodic multiwall carbon nanotube filter to kill viruses and *E. coli* in water. They discovered that such technique diminished the number of viruses and *E. coli*, during 30 s at a voltage of 3 V, to below the detection limit [1].

Electrocoagulation (EC) process has lately received a big deal of focus as an effi-

cient technology to eliminate microbes from wastewater and water thanks to its simplicity, selectivity, and comparatively low operating cost [38]-[44]. Further, the EC technique does not require chemicals injections to elevate the treatment performance (except for the case where the ionic strength is weak, so a supporting electrolyte should be added to increase the solution electric conductivity [45]), and it may be with ease automated and combined with additional treatment setups [46] [47] [48]. In addition, EC technology hugely decreases the volume of the formed solid waste (sludge) that needs elevated treatment cost [49] [50]; which successively greatly reduces the working price of the EC technique [51] [52]. These merits of the EC process place it an extremely encouraging choice to the classical treatment techniques [53] [54] [55]. On the other hand, the absence of reactor design (simple horizontal or vertical arrangement of square or rectangular plate electrodes inside a container) and the care of the EC to the chemical composition of the liquid being handled constitute the major obstacles of the EC technology [1] [51] [56] [57] [58].

The literature presents numerous explications for the routes of killing microorganisms via electrochemical technologies, which could be listed in **Table 1** [19] [59] [60] [61] and shown in **Figure 1**.

Table 1. Principal actions proposed explaining the deadliness of the electrochemical disinfection (ED) [19].

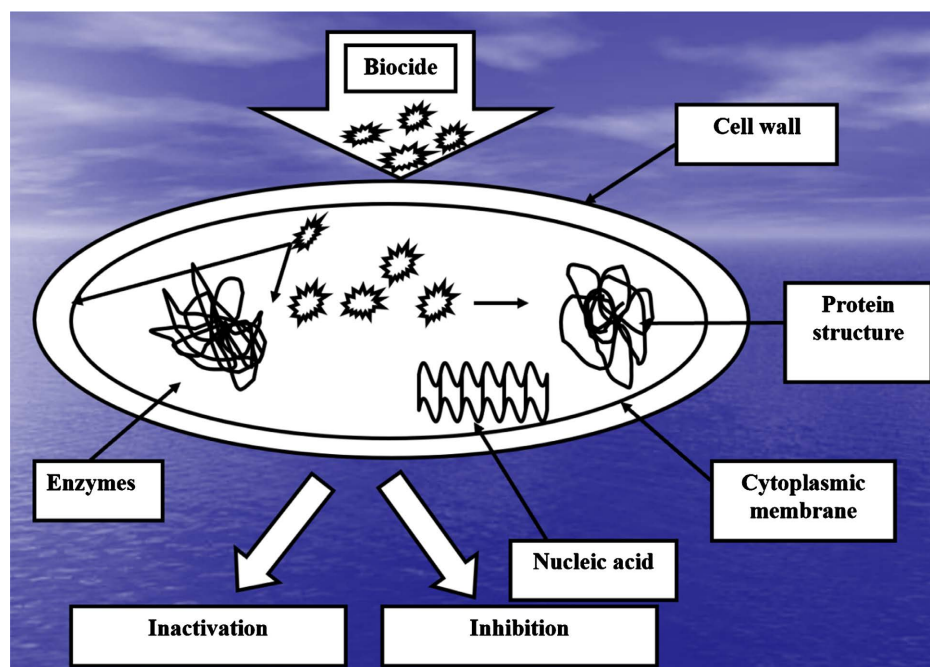| Electrochemical Disinfection (ED) Tools	
Oxidants	**Electric Field (EF)**
Oxidative stress and cell loss of life.	1) Irreversible permeabilization of cell membranes.
	2) Electrochemical oxidation of vital cellular constituents.
	3) Electrosorption of negatively charged *E. coli* cells to the anode surface + direct electron transfer reaction.

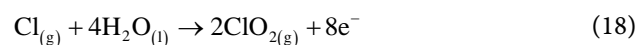**Figure 1.** Targeted sites of biocides in microbial cells.

In the EC process, in addition to the aforesaid routes, the microbes may be demobilized thanks to the direct adsorption on the surface of the anode pursued by electron transfer, and physical elimination through floating pathogens with formed hydrogen gas and/or precipitating with the produced flocs [34] [59] [60]. Table 2 presents the detailed EC reactions in the case of Fe [62] and Al electrodes.

Table 2. Electrocoagulation (EC) mechanisms using Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [63] [64] [65] [66].

Fe Mechanisms	Medium	Reaction	
Mechanism # 1 (pH 2)	Anode	$2\text{Fe}_{(s)} - 4\text{e}^- \rightarrow 2\text{Fe}^{2+}_{(aq)}$ $\left(E^\circ = +0.447\text{ V}\right)$	(1)
		$2\text{H}_2\text{O}_{(l)} - 4\text{e}^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)}$ $\left(E^\circ = -1.229\text{ V}\right)$	(2)
	Solution	$2\text{Fe}^{2+}_{(aq)} + 4\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{2(s)}$	(3)
	Cathode	$8\text{H}^+_{(aq)} + 8\text{e}^- \rightarrow 4\text{H}_{2(g)}$ $\left(E^\circ = 0.000\text{ V}\right)$	(4)
	Total	$2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow \text{O}_{2(g)} + 4\text{H}_{2(g)} + 2\text{Fe}(\text{OH})_{2(s)}$	(5)
Mechanism # 2 (pH 7)	Anode	$2\text{Fe}_{(s)} - 4\text{e}^- \rightarrow 2\text{Fe}^{2+}_{(aq)}$ $\left(E^\circ = +0.447\text{ V}\right)$	(1)
		$\text{Fe}^{2+}_{(aq)} - \text{e}^- \rightarrow \text{Fe}^{3+}_{(aq)}$ $\left(E^\circ = -0.771\text{ V}\right)$	(6)
		$\text{Fe}_{(s)} - 3\text{e}^- \rightarrow \text{Fe}^{3+}_{(aq)}$ $\left(E^\circ = +0.037\text{ V}\right)$	(7)
	Solution	$2\text{Fe}^{2+}_{(aq)} + 4\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{2(s)}$	(3)
		$2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)}$	(8)
	Cathode	$8\text{H}_2\text{O}_{(l)} + 8\text{e}^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ $\left(E^\circ = -0.828\text{ V}\right)$	(9)
	Total	$3\text{Fe}_{(s)} + 8\text{H}_2\text{O}_{(l)} \rightarrow \text{Fe}(\text{OH})_{2(s)} + 2\text{Fe}(\text{OH})_{3(s)} + 4\text{H}_{2(g)}$	(10)
Mechanism # 3 (pH 12)	Anode	$\text{Fe}_{(s)} - 3\text{e}^- \rightarrow \text{Fe}^{3+}_{(aq)}$ $\left(E^\circ = +0.037\text{ V}\right)$	(7)
	Solution	$2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)}$	(8)
	Cathode	$8\text{H}_2\text{O}_{(l)} + 8\text{e}^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ $\left(E^\circ = -0.828\text{ V}\right)$	(9)
	Total	$2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)} + 3\text{H}_{2(g)}$	(11)
Al Mechanism (pH 7)	Anode	$\text{Al}_{(s)} - 3\text{e}^- \rightarrow \text{Al}^{3+}_{(aq)}$ $\left(E^\circ = +1.660\text{ V}\right)$	(12)
		$2\text{H}_2\text{O}_{(l)} - 4\text{e}^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)}$ $\left(E^\circ = -1.229\text{ V}\right)$	(2)
	Solution	$\text{Al}^{3+}_{(aq)} + 3\text{OH}^-_{(aq)} \rightarrow \text{Al}(\text{OH})_{3(s)}$	(13)
		$\text{Al}(\text{OH})^-_{4(aq)} \rightarrow \text{OH}^-_{(aq)} + \text{Al}(\text{OH})_{3(s)}$	(14)
	Cathode	$8\text{H}_2\text{O}_{(l)} + 8\text{e}^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ $\left(E^\circ = -0.828\text{ V}\right)$	(9)
	Total	$\text{Al}_{(s)} + 5\text{H}_2\text{O} \rightarrow \text{O}_{2(g)} + (7/2)\text{H}_{2(g)} + \text{Al}(\text{OH})_{3(s)}$	(15)

More importantly, so powerful oxidizing agents, like HOCl, OCl⁻, $ClO_{2(g)}$ and $Cl_{2(g)}$, are formed throughout the EC technology following *Reactions* (16), (17) and (18) [59] [67]:

$$2Cl^-_{(aq)} \rightarrow Cl_{2(g)} + 2e^- \tag{16}$$

$$Cl_{2(g)} + 2OH^-_{(aq)} \rightarrow H_2O_{(l)} + OCl^-_{(aq)} + Cl^-_{(aq)} \tag{17}$$

$$Cl_{(g)} + 4H_2O_{(l)} \rightarrow 2ClO_{2(g)} + 8e^- \tag{18}$$

Such chemicals may harm the membrane of the cell that leads to killing microbes.

In the previous work [34], *E. coli* cells were used as clear application example of ED by using stainless steel (SS), ordinary steel (OS), and aluminum (Al) electrodes. Two surface waters (from Ghrib and Keddara dam's waters, north of Algeria) were also taken to test EC efficiency for coliforms and algae removal using Al electrodes.

In the present study, the effect of temperature on *E. coli* removal is investigated besides the influence of cell concentration.

2. Experimental Procedures

2.1. Artificial Wastewater Containing *E. coli* Preparation

For the experimental investigation, synthetic wastewater infected by *E. coli* culture was employed [34]. The test wastewater was prepared by diluting a pure culture of *E. coli* in distilled water (**Figure 2**). The *E. coli* culture was cultivated via inoculating seed of *E. coli* into a 500 mL flask filled with 250 mL of the growth medium (seed of *E. coli* and growth medium were gratefully supplied by Hygiene Laboratory of Blida Hospital). The culture was grown on a shaker in a water bath for 24 h at 37°C to reach its stationary growth phase. By addition of 250 mL of distilled water to this culture, a 500 mL fresh culture was then used to make the model water for the *E. coli* EC experiment. Surface waters were obtained from the Ghrib dam, which is known for its hardness (800 mg/L as CaCO₃),

Figure 2. Culture medium inoculated with *E. coli*.

and the Keddara dam famous for its high algae content [34].

2.2. EC Experiments

EC tests were conducted using equipment that was composed of two electrodes, which have the same dimensions and plunged in a beaker (V = 0.5 L and Ø = 8 cm) (Figure 3). For each electrode, the immerged (active) surface was 49.5 cm² (4.5 cm × 11 cm) and the distance between them was fixed at 5 cm. Electrodes were connected to direct current power supply (Stell Trafo) with 30 V as maximal tension and 10 A as maximal intensity. Applied tension U (V) and current intensity I (A) were measured by a voltammeter and ammeter connected in parallel and in series, respectively. Before the EC test and in order to avoid any interference, electrodes were prepared as follows: 1) rinse with distilled water, 2) clean in sodium hydroxide solution (10%), 3) rinse with distilled water. They were then dried with absorptive paper and weighted. After introduction of solution to treat in the reactor, pH was adjusted at its selected initial value using H_2SO_4 or NaOH (2N) solutions. Electrodes were plunged in the reactor and fixed before their connection to power supply. Conductivity and pH measures were immediately realized. Finally, electrodes were dried and weighted again. All used chemicals were of analytical grade [34].

Samples (5 mL) are aseptically pipetted during EC at every 5 min from the solution and filtered for analysis. *E. coli* cells were enumerated following the visible spectrophotometry method at 620 nm in accordance with the standard methods [68]. Total coliforms and algae are counted in accordance with the standard methods [69].

3. Results and Discussion

First, several experiments were carried out using OS electrodes to optimize EC parameters of artificial wastewater contaminated by *E. coli* culture such as time
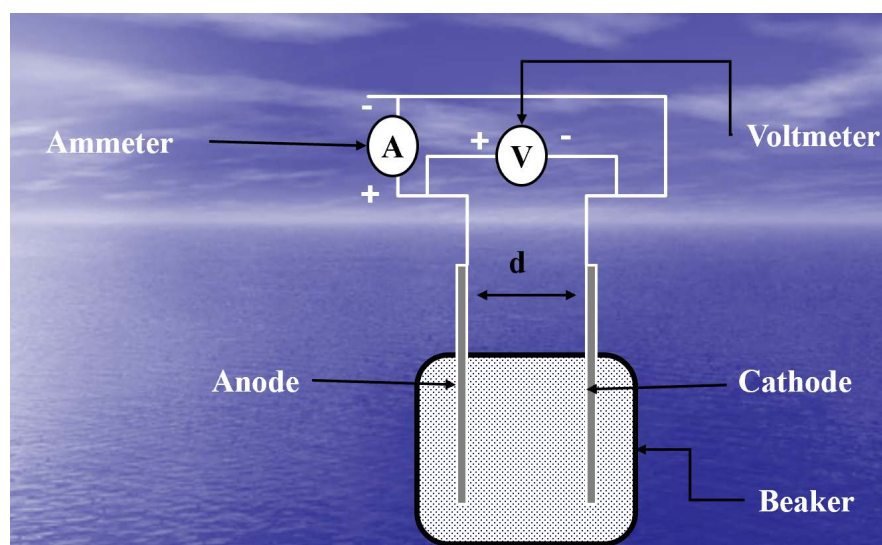

Figure 3. Experimental electrochemical disinfection (ED) device.

(t_{EC}, in min), current intensity (I, in A), pH, electrodes nature (*i.e.*, using SS and Al electrodes), temperature and initial cellular concentration. Finally, these optimal parameters were also applied for Ghrib and Keddara waters.

3.1. EC of *E. coli* Culture

3.1.1. Effect of EC Time (t_{EC}) and Current Intensity (I)

Several tests were first performed to understand how EC efficiency varies with time. EC time was fixed at 60 min, and samples were taken at every 5 min (Figure 4) during this period to control its ED efficiency at $I = 0.5$ A ($U = 7.5$ V). These observations are noted:

For $t_{EC} = 1$ min, medium emission of $H_{2(g)}$ bubbles from the cathode and white froth formation at the surface of solution (Figure 5). For $t_{EC} = 10$ min, solution color becomes yellow green. For $t_{EC} = 15$ min, there is a formation of a green cloud in the solution which migrates to the space between the anode and beaker. For $t_{EC} = 20$ min, the solution becomes limpid. For $t_{EC} = 40$ min, there is formation
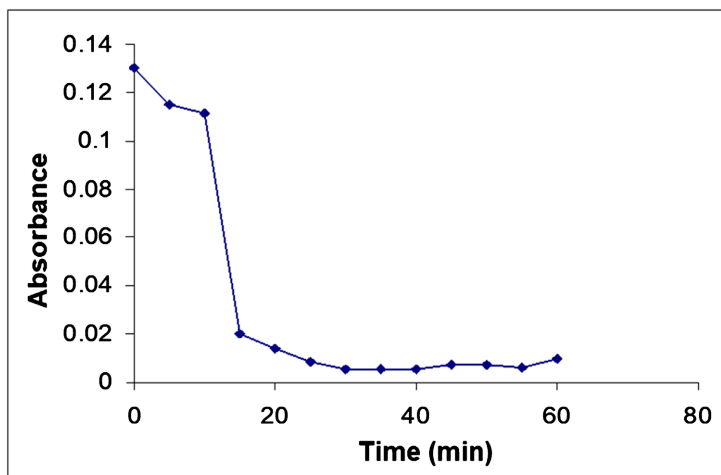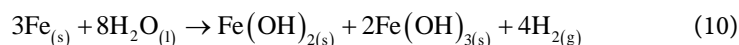

Figure 4. Reduction of cellular concentration of *E. coli* as a function of time using ordinary steel (OS) electrodes during electrocoagulation (EC) application [34].

Figure 5. White froth formation at the surface of solution during EC application.

of a small deposit on the anode. *Reaction* (10) (see Table 2) is proposed:

Reaction (10) (neutral pH):

$$3Fe_{(s)} + 8H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + 2Fe(OH)_{3(s)} + 4H_{2(g)} \tag{10}$$

Reaction (10) (see Table 2) takes into consideration the apparition in solution of green colloids ($Fe(OH)_{2(s)}$) and then yellow colloids ($Fe(OH)_{3(s)}$) with hydrogen ($H_{2(g)}$) production. It is evident that colloid formation is affected to the anode dissolution, which is reduced at 0.26%. Initial number of *E. coli* cells decreased (89%) rapidly during the first 20 min and then decreased slowly before reaching a relatively constant rate (96%) when time exceeds 35 min. Consequently, optimal EC time is around 35 min, which was also found by Holt *et al.* [70] for clay suspension removal by EC.

To understand the influence of current intensity on EC performance, three values other than the first one ($I = 0.5$ A, $U = 7.5$ V) are studied: $I = 0.1$ ($U = 2$ V); 0.25 ($U = 3.7$ V); and 1 A ($U = 12$ V). These observations are noted:

- *For I* = 0.1 *A*:

When $t_{EC} = 1$ min, slight emission of $H_{2(g)}$ bubbles at the cathode. When $t_{EC} = 5$ min, formation of a small quantity of froth at the solution surface. When $t_{EC} = 10$ min, green colloids in the solution appear.

- *For I* = 0.25 *A*:

When $t_{EC} = 1$ min, emission of $H_{2(g)}$ bubbles at the cathode and formation of more important froth than for $I = 0.1$ A. When $t_{EC} = 10$ min, solution becomes green.

- *For I* = 1 *A*:

When $t_{EC} = 1$ min, intense emission of $H_{2(g)}$ from the cathode and froth formation at the surface. When $t_{EC} = 10$ min, a green color appears. When $t_{EC} = 12$ min, formation of clouds (blue green to black particles) and appearance of green deposit in the bottom of the beaker near the anode. When $t_{EC} = 15$ min, solution starts to be limpid and deposit volume increases.

The obtained results are shown in Figure 6. When current intensity *I* increased from 0.1 to 1 A, reduction of *E. coli* cells is enhanced by 80% - 98%. In other words, more applied current is increased more its disinfectant effects become more efficient [71] [72]. Applied current originate potential difference
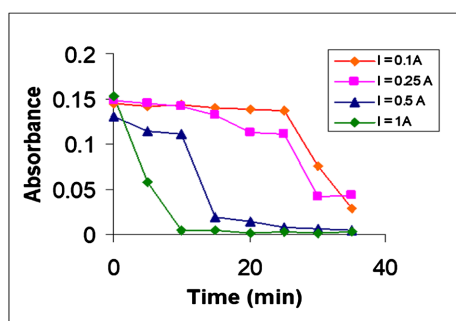

Figure 6. Reduction of cellular concentration of *E. coli* as a function of current density using ordinary steel (OS) electrodes during $t_{EC} = 35$ min [34].

from an extremity to the other extremity of the cellular membrane on account of its electrical resistance [73] [74] [75]. This potential difference modifies consequently the transmembrane potential producing destruction of the cellular membrane [76] [77] [78]. On the other hand, ions motion is restricted in a limited area [79] [80] [81]. These ions could not consequently pass through the membrane simply and vital physiological functions are not available for cells. For biomacromolecules such as enzymes, their conformations are crossed or transformed (Figure 1). As a consequence, vital physiological functions of cells are destroyed [76] [82] [83].

3.1.2. Effect of pH

It is well known that pH plays an important role in EC processes [84] [85] [86]. Three pH values were selected to illustrate its influence ($I = 1$ A): acid ($U = 12.6$ V), neutral ($U = 12$ V) and alkaline pH ($U = 11$ V). These observations are noted:

For pH = 2:

When $t_{EC} = 5$ min, intense emission of $H_{2(g)}$ bubbles at the cathode and significant formation of $O_{2(g)}$ bubbles at the anode with white froth at the solution surface. When $t_{EC} = 15$ min, formation of green flocs. When $t_{EC} = 20$ min, formation of deposit near the anode with some green suspensions in the bottom of the beaker. When $t_{EC} = 30$ min, solution becomes transparent.

For pH = 7.1:

The same observations as cited above for $I = 1$ A (Section 3.1.1).

For pH = 9.5:

When $t_{EC} = 1$ min, emission of $H_{2(g)}$ bubbles from cathode and froth formation at the surface. When $t_{EC} = 2$ min, formation of white deposit in the bottom of the beaker, its volume increases with time in comparison with acid pH, and a red-brown color appears. When $t_{EC} = 10$ min, the solution starts to be clear.

Based on these observations, *Reactions* (5) and (11) (see Table 2) for acid and alkaline pH, respectively, could be proposed:

Reaction (5) (*acid pH*):

$$2Fe_{(s)} + 6H_2O_{(l)} \rightarrow O_{2(g)} + 4H_{2(g)} + 2Fe(OH)_{2(s)} \qquad (5)$$
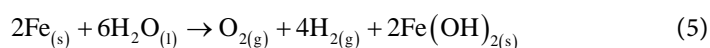
Reaction (5) (Table 2) accounts for observed production of oxygen at the anode and hydrogen at the cathode and the appearance of a green color ($Fe(OH)_{2(s)}$) in solution [87].

Reaction (11) (*alkaline pH*):

During the first minutes after electrodes are introduced into the recipient, solution becomes yellow-red-brown with flocs appearing because of ferric ion spontaneous discharge. Ferric ions in intense presence of OH^- give birth to ferric hydroxide following *Reaction* (11) (Table 2):

$$2Fe_{(s)} + 6H_2O_{(l)} \rightarrow 2Fe(OH)_{3(s)} + 3H_{2(g)} \qquad (11)$$
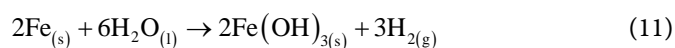
Reaction (11) reflects red-brown flocs ($Fe(OH)_{3(s)}$) appearing in solution and hydrogen production at the cathode.

These three *Reactions* ((10), (5) and (11)) were reported by several authors [88]. The reduction of *E. coli* cells is shown in **Figure 7**. For t_{EC} = 20 min, EC efficiency is less important when pH is acid (29%) and becomes more significant when pH is neutral (99%) and alkaline (100%). For t_{EC} = 35 min, EC efficiency is nearly the same for every pH (100% for acid and alkaline pH, 98% for neutral pH). In fact, at alkaline condition (*Reaction* (11)), produced $Fe^{3+}_{(aq)}$ neutralizes directly and efficiently *E. coli* cells charge. On the other hand, $Fe(OH)_{3(s)}$ acts by *sweep flocculation* or enmeshment [89]: $Fe(OH)_{3(s)}$ being dense settles out and moves the *E. coli* cells, which could be adsorbed on these ferric hydroxides. At neutral condition (*Reaction* (10)), $Fe^{3+}_{(aq)}$ is produced directly and indirectly. The indirect production seems to decrease process efficiency. In solution, $Fe(OH)_{2(s)}$ and $Fe(OH)_{3(s)}$ settle out on the one hand and float to the solution surface in presence of $H_{2(g)}$ bubbles on the other hand. Sedimentation and flotation, being countercurrent, seem to be in concurrence. At the acid condition (*Reaction* (5)), $Fe^{2+}_{(aq)}$ is produced simultaneously with $O_{2(g)}$. Even if $O_{2(g)}$ is well known as an excellent oxidant, it does not compensate for the absence of $Fe^{3+}_{(aq)}$. In solution, $Fe(OH)_{2(s)}$ is less efficient in *sweep flocculation* than $Fe(OH)_{3(s)}$. For Zhu *et al.* [89], iron EC was also found very efficient in virus removal.

3.1.3. Effect of Nature of Electrodes

The nature of electrodes plays an important role in EC process, so two other electrodes than OS were used: SS and Al (I = 1 A). OS (U = 12 V) and Al (U = 11.8 V) give to the solution $Fe^{2+}_{(aq)}$ and $Fe^{3+}_{(aq)}$ (neutral pH) and $Al^{3+}_{(aq)}$, respectively; however, SS (U = 10.7 V) does not give iron ions to the solution. These observations are noted:

1) *Stainless steel* (*SS*) *electrodes*

For t_{EC} = 1 min, emission of $H_{2(g)}$ bubbles from cathode and froth formation at the surface solution [90] [91]. For t_{EC} = 10 min, formation of colloids that migrate to the anode [92]. For t_{EC} = 15 min, formation of a green deposit at the bottom of the beaker and at the surface solution near anode. For t_{EC} = 17 min, solution becomes clear yellow, until the end of experiment when there was no more change.
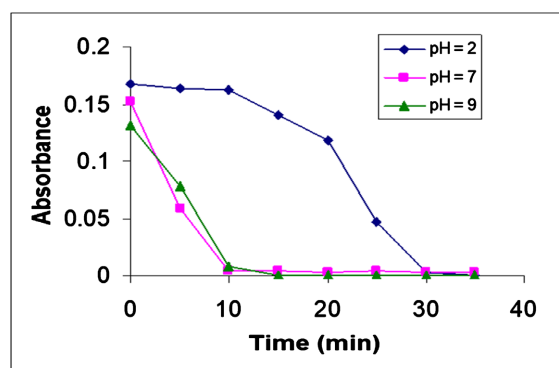

Figure 7. Reduction of cellular concentration of *E. coli* as a function of pH using ordinary steel (OS) electrodes during t_{EC} = 35 min (I = 1 A) [34].

2) *Aluminum electrodes*

For t_{EC} = 1 min, intense emission of $H_{2(g)}$ bubbles at cathode and $O_{2(g)}$ bubbles at anode and formation of white froth at the solution surface. For t_{EC} = 5 min, appearance of white particles in solution that migrate to the anode and constitute a white deposit. For t_{EC} = 10 min, solution becomes clear yellow and froth formation increases with time. The first samples (t_{EC} = 1, 5 and 10 min) give after filtration limpid solutions in comparison with OS electrodes. For t_{EC} = 11 min, white deposit volume increases at the surface and near the anode. For t_{EC} = 25 min, the solution becomes more limpid. Based on these observations for Al electrodes, *Reaction* (15) (Table 2) can be proposed:

$$Al_{(s)} + 5H_2O \rightarrow O_{2(g)} + (7/2)H_{2(g)} + Al(OH)_{3(s)} \tag{15}$$
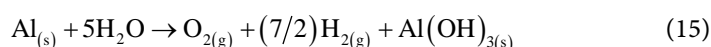
Reduction of absorbance at 620 nm as a function of electrode nature is shown in Figure 8. For the first 10 min (Figure 8), SS (55.45%) is less efficient than OS (97.18%), which is less than Al (98.16%). In fact, ED takes place with direct and indirect effects. Direct effect is produced by the EF application (as for SS electrodes) [93] [94]. On the other hand, the indirect effect results from microorganisms contact with oxidants that are generated by water electrolysis and anode dissolution (as for OS and Al electrodes) [76] [82] [95].

Vital centers of bacterial cells are protected by a membrane that is constituted essentially by a biomolecular layer of phospholipids with hydrophobic and hydrophilic parties. Protein inclusions inside the membrane authorize ionic change with the cell environment. A phospholipidic membrane is not easily oxidable whereas proteins are easily destroyed by direct effect of an electrical field. Cells cannot then change more ions but can be reactivated in a favorable medium. Its total destruction requires an oxidant capable of passing through the membrane and reaching vital centers [76]. Direct effect of electrical field can then reduce the number of microbes in water even if is not sufficient to remove this number in order that the water becomes drinkable [82]. Al electrodes form $Al^{3+}_{(aq)}$ whereas OS electrodes generate $Fe^{2+}_{(aq)}$ (which can be $Fe^{3+}_{(aq)}$ at neutral pH). Consequently, valence (+3) is an advantage for Al. On the other hand, Al flocs are more easily floatable than iron ones. Consequently, Al electrodes, being more efficient than OS and SS electrodes, are used in the next experiments.
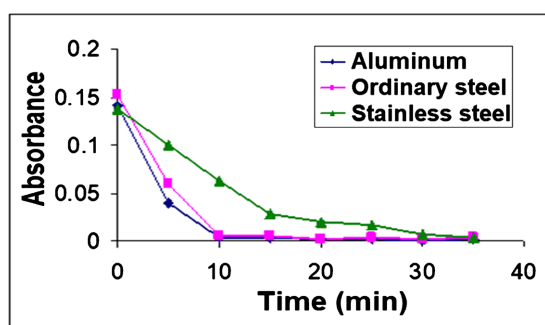

Figure 8. Reduction of cellular concentration of *E. coli* as a function of electrodes nature during EC (I = 1 A).

3.1.4. Effect of Temperature

Here we examine the effect of the initial temperature of the solution containing *E. coli* on the EC efficiency. The temperature is adjusted to 12˚C, 25˚C, 50˚C and 65˚C either by cooling the solution (*i.e.*, for 12˚C) or by heating it to reach the desired temperature (50˚C and 65˚C) that is kept constant for the duration of the treatment.

The same observations cited above (for $I = 1$ A), except that we can add that at the high temperature (*i.e.*, 50˚C and 65˚C), the volume of the white deposit increases and the synthetic wastewater solution becomes clearer in comparison with $T = 12$˚C.

The results obtained are represented graphically in Figure 9. For a treatment time not exceeding 30 min, we note that the reduction in absorbance at 620 nm is greater for the highest temperature (65˚C).

Living cells can be killed in the first place under the effect of heating when the temperature was raised to 50˚C and 65˚C. In fact, for the test at 50˚C, the absorbance of the solution went from 0.5063 to 0.1338 (*i.e.*, a reduction in absorbance of around 73.57%) after heating before EC. For the test at 65˚C, the absorbance of the solution went from 0.3046 to 0.1312 (56.93%) after heating before EC. The cooling effect was also noticed. In fact, cooling the solution to 12˚C reduced the absorbance, which went from 0.2046 to 0.1385 (32.31%) after cooling before EC.

Indeed, the temperature of the medium in which living cells are suspended has a significant influence in determining the properties of membrane fluidity. At low temperature, the phospholipids are tightly packed in a rigid gel structure, while at high temperatures they are less ordered and the membrane has a 'liquid crystalline' structure. The phase transition from gel to liquid crystal is temperature dependent and therefore can affect the physical stability of the cell membrane. A rise in temperature is known to increase the rate of lateral diffusion of lipids by at least two orders of magnitude as lipids change phase from gel to liquid crystal. As seen in Figure 9, a greater reduction in survivability of *E. coli* was observed when the temperature of the liquid medium was higher ($T = 65$˚C) at a comparable magnitude of treatment time than at lower temperatures. Based on this, it is proposed that the temperature-related phase transition of phospholipid
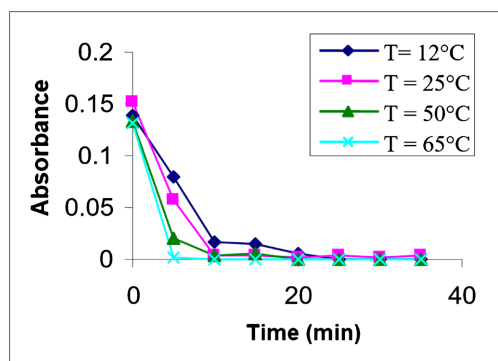

Figure 9. Variation in the cell concentration of *E. coli* (expressed by absorbance) as a function of EC time at different temperatures.

molecules from gel phase to liquid crystal and the associated reduction in bilayer thickness may make the cell more susceptible to EF effects at a relatively high temperature.

3.1.5. Effect of Cell Concentration

In this experiment, we investigated the *E. coli* concentration effect on the efficacy of EC ($I = 1$ A, $T = 25°$C). We took three different strengths: low, medium, and high. The low concentration had the equivalence of an absorbance of 0.1525; the medium and high concentration had an absorbance of 0.3224 and 0.7759, respectively. During these experiments, we observed for:

- A low cell concentration: (See previous paragraph for $I = 1$ A).
- Average cell concentration: The volume of the foam increases at $t_{EC} = 10$ min; the release of hydrogen decreases at $t_{EC} = 25$ min.
- A high cell concentration: The color of the solution (synthetic *E. coli* wastewater) remains light yellow.

The change in absorbance at 620 nm as a function of the EC time, for these three concentration values, is shown in Figure 10.

When the EF and the temperature were constant, it was observed that an increase in the elimination rate of *E. coli* was proportional with the decrease in the initial bacterial cell concentration. In other words, cells can be killed more easily when their microbial density is low compared to when it is high. This can be explained by the fact that the bacterium of *E. coli* has an ability to withstand the EF at high density populations compared to low density populations. Therefore, the lethal effect of ED targets microorganisms separately and not in masse.

3.2. EC of Two Surface Waters Using Aluminum Electrodes

In order to confirm EC efficiency, two surface waters were used. First, Ghrib raw water (2700 µS/cm at 25°C as conductivity and 660 mg $CaCO_3$/L as total hardness) EC was realized using Al electrodes at $I = 0.8$ A ($U = 17.8$ V) for $t_{EC} = 35$ min. Total coliforms are controlled by colonies counting in specific culture medium (Figure 11). These observations are noted: For $t_{EC} = 1$ min, emission of $H_{2(g)}$ bubbles from the cathode and $O_{2(g)}$ bubbles from the anode with little froth
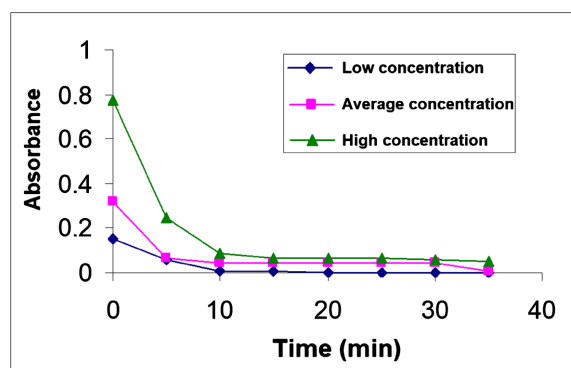

Figure 10. Variation in the cell concentration of *E. coli* (expressed by absorbance) as a function of EC time at different microbial concentrations.

at the surface. For t_{EC} = 10 min, froth layer increases with white flocs floating to the surface. An important efficiency (99.73%) of EC as ED for t_{EC} = 35 min was reached.

Finally, Keddara raw water (considered as soft water), which is known for its algae content, is disinfected at I = 0.25 A (U = 18.5 V) for t_{EC} = 35 min. These observations are noted:

For t_{EC} = 1 min, emission of $H_{2(g)}$ bubbles from the cathode and $O_{2(g)}$ bubbles from the anode. For t_{EC} = 5 min, algae suspension rises to the solution surface and at the beaker bottom with white froth formation (**Figure 12(a)**). For t_{EC} = 10 min,

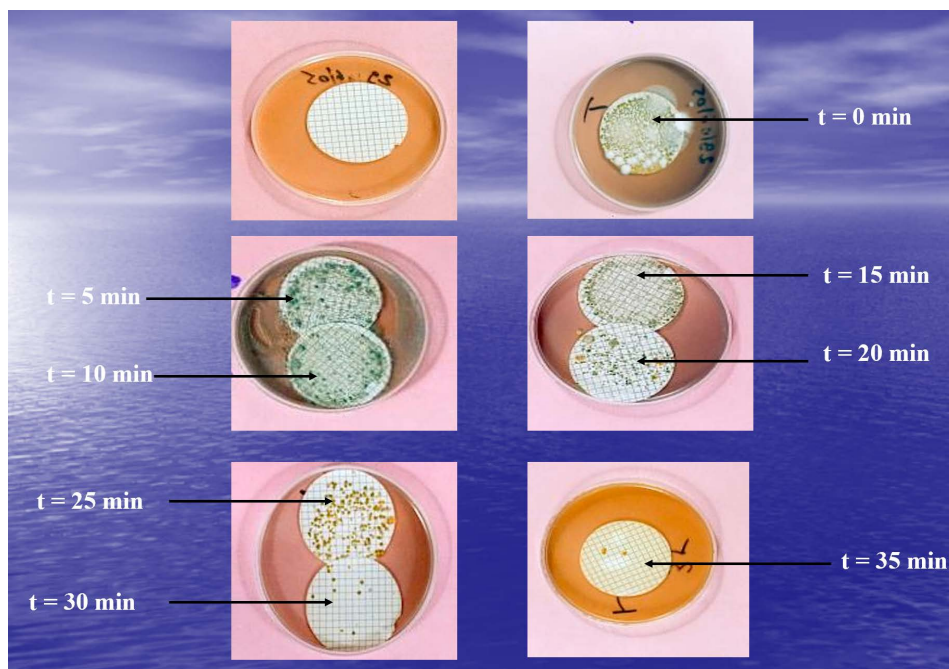**Figure 11.** Total coliforms removal as a function of t_{EC} for Ghrib raw water.
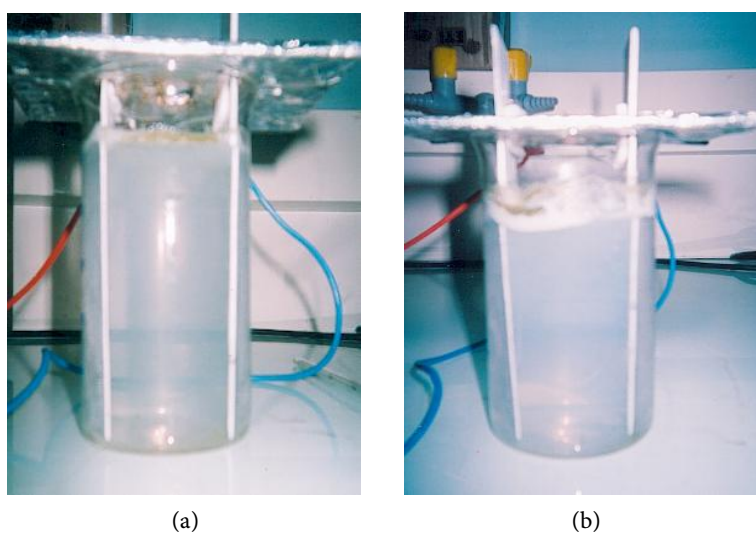

(a)   (b)

Figure 12. Electrocoagulation (EC) of Keddara raw water rich in algae (Al electrodes). (a) t_{EC} = 5 min, (b) t_{EC} = 10 min.
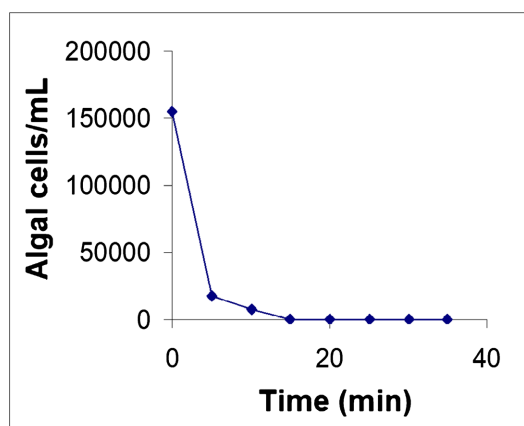
Figure 13. Number of algae cells/mL removal as a function of EC time.

solution becomes more transparent (**Figure 12(b)**). Total removal of algae is reached by EC for t_{EC} = 30 min. The reduction in the number of algae cells as a function of the EC time is shown in **Figure 13**.

Similar results have been obtained by several authors, proving that ED is effective in eliminating algae and pathogens [93] [96].

4. Conclusions

Laboratory tests were performed to examine electrocoagulation (EC) as electrochemical disinfection (ED) of synthetic wastewater infected by no pathogenic *Escherichia coli* species in batch culture and two surface waters employing ordinary steel (OS), stainless steel (SS) and aluminum (Al) electrodes. Further, two surface waters (from Ghrib and Keddara dam's waters, north of Algeria) were also taken to test EC efficiency for coliforms and algae removal using Al electrodes. The effect of temperature on *E. coli* removal is investigated besides the influence of cell concentration. The main points drawn from this work may be listed below:

1) The impacts of disinfection by the EC setup designed for *E. coli* culture and two surface waters were studied. The survivability of *E. coli* decreased with current intensity and treatment time. *E. coli* cells were efficiently demobilized, and total elimination of coliforms and algae were obtained in 30 min. Al electrodes were slightly more effective than OS and SS electrodes. Applying EC in algal toxins elimination would be more helpful. It remains important to check the optimum operating parameters of a continuing process and to perform a detailed comparative study of energy consumption by the treatment system and the conventional methods before constructing an industrial application system in the future.

2) A greater reduction in the survivability of *E. coli* was observed when the temperature of the liquid medium was higher (T = 65˚C) at a comparable magnitude of treatment time than at lower temperatures. Based on this, it is proposed that the temperature-related phase transition of phospholipid molecules from gel phase to the liquid crystal and the associated reduction in bilayer

thickness may make the cell more susceptible to electric field (EF) effects at a relatively high temperature.

3) When the EF and the temperature were constant, it was observed that an increase in the elimination rate of *E. coli* was proportional to the decrease in the initial bacterial cell concentration. In other words, cells can be killed more easily when their microbial density is low compared to when it is high. This can be explained by the fact that the bacterium of *E. coli* has an ability to withstand the EF at high-density populations compared with low-density populations. Therefore, the lethal effect of ED targets microorganisms separately and not in masse.

Acknowledgements

Conflicts of Interest

The authors declare no conflicts of interest.

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Abbreviation

EC	Electrocoagulation
ED	Electrochemical disinfection
EF	Electric field
I	Current intensity (A)
OS	Ordinary steel
SS	Stainless steel
t_{EC}	Electrocoagulation (EC) time (min)
U	Applied tension (V)