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Electrocoagulation of a raw water of Ghrib Dam (Algeria) in batch using aluminium and iron electrodes

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ABSTRACT

Laboratory experiments were carried out to investigate electrocoagulation (EC) of Ghrib Dam's water (Algeria) in batch using aluminium and iron electrodes without addition of chemicals, without pH modification, and without agitation. The optimal conditions are an applied voltage of direct current of 8 V and a current density 74 A m^{-2} (0.35 A) during 45 min for both electrodes. The microbial pollution is completely removed by essentially electrical field whereas conductivity and turbidity are reduced at 27 and 85% respectively for Fe electrodes and 22 and 97% for Al electrodes by metallic cations. These results prove that water treatment by EC using Al electrodes (flotation) for low turbid water (7 NTU) is more convenient than Fe electrodes (sedimentation) for both turbidity and organic matter removal. A new parameter to be taken in consideration for EC reactor design the ratio r active volume on reactor volume which is full of water is introduced where the *active volume* is the active surface multiplied by the distance between the electrodes.

Keywords: Surface water; Drinking water; Electrocoagulation; Iron; Aluminium

1. Introduction

Suspended particles are present in almost all surface waters. These fine turbidity particles often cause taste, odour, and colour problems. The physicochemical interactions (destabilisation and aggregation) that enable them to separate from an aqueous medium are the objective of coagulation. Usually chemicals (coagulants, such as $Al_2(SO_4)_3$ and $FeCl_3$) are added to a flash mix and given a few minutes of agitation to disperse. Afterward the solution is gently agitated between half an hour and an hour for floc formation (flocculation) [1,2].

Electrocoagulation (EC) is the process of destabilising suspended and emulsified contaminants in aqueous medium by introducing an electrical current into

the medium. As the current passes through the media, ion dissolution from reactive electrodes (iron or aluminium) can destabilise colloidal particles. As this occurs, the contaminants form hydrophobic entities precipitate, and can be easily removed by flotation or sedimentation [2]. EC involves the generation of coagulants in situ by dissolving electrically either aluminium or iron ions from respectively aluminium or iron electrodes. The metal ions generation takes place at the anode; hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water [3].

The chemical reactions taking place at the anode are given as follows [3].

For aluminium anode:

 $Al_{(s)} - 3e^- \rightarrow Al^{3+}_{(aq)}$ (1) at alkaline conditions

$$Al_{(aq)}^{3+} + 3OH_{(aq)}^{-} \rightarrow Al(OH)_{3(s)}$$

$$\tag{2}$$

at acidic conditions

$$Al_{(aq)}^{3+} + 3H_2O_{(l)} \rightarrow Al(OH)_{3(s)} + 3H_{(aq)}^{+}$$
 (3)

For iron anode:

 $Fe_{(s)} - 2e^{-} \rightarrow Fe^{2+}_{(aq)} \tag{4}$

at alkaline conditions

$$\operatorname{Fe}_{(aq)}^{2+} + \operatorname{3OH}_{(aq)}^{-} \to \operatorname{Fe}(\operatorname{OH})_{2(s)}$$
(5)

at acidic conditions

$$4Fe^{2_{(aq)}} + O_{2(g)} + 2H_2O_{(l)} \to 4Fe^{3_{(aq)}} + 4OH^{-_{(aq)}}$$
(6)

In addition, there is oxygen evolution reaction

$$2H_{2}O_{(l)} - 4e^{-} \rightarrow O_{2(g)} + 4H^{+}_{(aq)}$$
⁽⁷⁾

The reaction at the cathode is

$$2H_2O_{(1)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(ag)}$$
 (8)

The nascent Al³⁺ or Fe²⁺ ions are very efficient coagulants for particulates flocculating. The hydrolyzed aluminium ions can form large networks of Al–O–Al–OH that can chemically adsorb pollutants such as F⁻ [3]. Aluminium is usually used for water treatment and iron for wastewater treatment. The advantages of EC include high particulate removal efficiency, compact treatment facility, relatively low cost and possibility of complete automation [3].

EC being proved efficient for different waters and solutions [4–7] is a good alternative because it can remove the disadvantages of the chemical coagulation [8]. Water treatment feasibility by EC in batch, at laboratory scale, of Ghrib Dam's water (Ain Defla, Algeria) is studied. For this object, EC tests have been realised on raw water samples.

2. Experimental details

2.1. Experimental procedure

EC tests have been realised using an equipment which is composed of two ordinary steel electrodes (iron: 99.8% and carbon: 0.2%) and two others in commercialised aluminium. The electrodes are of the same dimensions and plunged in a device with 0.5 Las volume. For each electrode, the immerged (active) surface is 47.25 cm² (4.5 cm × 10.5 cm) and the distance between them is fixed at 4 cm. The electrodes are connected to direct current power supply (Enyl1 Elektrolyser) with 15 V as maximal voltage and 10 A as maximal intensity. Applied voltage *U* (V) and current intensity *I* (A) are measured by voltammeter and ammeter connected in parallel and in series respectively.

In order to show only EC efficiency, no agitation was applied. The electrophoretic motion and light turbulence

which is created by $H_{2(g)}$ bubbles ascension towards the surface are preferred alone. Electrochemical migration, which is imposed by electrical field between the anode and the cathode, constitutes the principal diffusion mode of the charged species in water. Moreover, no chemicals were added to raw water neither for adjusting pH nor for accelerating the process. On the other hand, raw water samples after EC are submitted to decantation during 30 min before aspiration and Buchner filtration (funnel with glass-frit (0.45 µm) and vacuum-pump) for their analyses.

Before EC tests and in order to avoid any interference, Fe electrodes are prepared as follows: (1) skim with solution composed of: NaOH: 25 g, Na₂CO₃: 25 g, K_2CO_3 : 25 g, distilled water: q.s.p. 1000 mL, (2) rinse with distilled water and polish using abrasive paper, (3) clean in sulphuric acid solution (H₂SO₄ at 20%) at 40°C during 5 min, (4) rinse with distilled water; and Al electrodes: (1) rinse with distilled water, (2) clean in sodium hydroxide solution (10%), (3) rinse with distilled water. All used chemicals are of analytical grade.

2.2. Analytical techniques

The frequently performed analyses are turbidity in NTU (Turb 550 (wtw) turbidimeter), conductivity in mS cm⁻¹ at 25°C (EC215 Hanna Instruments conductimeter), and pH (Inolab pH level 1 pH-meter).

Water which is used in EC tests as raw water to treat comes from Ghrib Dam (Ain Defla) supplying Algiers, Medea, and Berrouaghia (Algeria). Raw water samples were taken on April 25th, 2005. During this spring period, even though the climate was relatively rainy, water was practically limpid (turbidity 6.5 NTU). The bold and underlined values (Table 1, second column) exceed accepted standards which are close to WHO standards. Surface water, which supplies Ghrib Dam, comes from a saline area (conductivity 2.7 mS cm⁻¹ at 25°C and total hardness 825 mg L⁻¹ as CaCO₃) and goes through calcareous and gypseous soils [9]. In fact, for a water to treat by EC, a high conductivity is necessary [10,11] for process acceleration. The KMnO4 oxidability indicates the presence of organic matter (born from the degradation of vegetable fragments and animal rubbish). Total coliforms, faecal coliforms, and Streptococcus indicate natural microbial pollution. This raw water is consequently very hard and organically and microbiologically polluted.

3. Results and discussion

3.1. EC mechanism

In order to explain EC effects, several mechanisms have been cited [6,11–15]. To get an appropriate

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Table 1

Physicochemical and bacteriological parameters of Ghrib Dam's raw water (second column, April 25th, 2005) and treated water by EC with Fe and Al electrodes (third and fourth columns respectively) at optimal conditions (8 V, 45 min) and their accepted standards (close to WHO standards, numbers between brackets indicate removal percentage).

Parameters	Raw water	After Fe EC	After Al EC	Accepted standards
Turbidity (NTU)	6.50	1.02 (84%)	0.51 (92%)	<5
pH	7.92	9.5	6.8	6.5-8.5
Temperature (°C)	9.2	20	20	<25
Conductivity (mS cm ⁻¹) at 25°C	2.7	2.12 (21%)	2.34 (13%)	3.1
Total coliforms	<u>Uncountable</u>	0	0	<10/100 mL
Faecal coliforms	28	0	0	0/100 mL
Streptococcus	<u>3</u>	0	0	0/100 mL
Sulphates (mg L ⁻¹)	371	120	176	<400
Ammonium (mg L ⁻¹)	0.0	2.25	0.0	< 0.5
Phosphates (mg L ⁻¹)	0.0	0.0	0.0	<0.5
Nitrates (mg L ⁻¹)	0.01	0.01	0.01	<50
Iron (mg L^{-1})	0.01	0.15	0.01	< 0.3
KMnO ₄ oxidability (mg L ⁻¹)	4.6	3.8	3.0	<5
Dry residue (mg L ⁻¹)	<u>2007</u>	1357	1481	<2000
Total hardness (mg L^{-1} as CaCO ₂)	<u>825</u>	350	500	<500
Calcium (mg L ⁻¹)	<u>224</u>	140	164	<200
Magnesium (mg L ⁻¹)	<u>198</u>	84	132	<150
Nitrites (mg L ⁻¹)	0.01	0.01	0.01	< 0.1
TAC (mg L^{-1} as CaCO ₃)	149	51.5	15.5	<500
Chlorides (mg L ⁻¹)	440	355	361	<500

mechanism, a preliminary EC test of raw water has been accomplished. This test consists to raise the applied voltage U gradually from 0 to 15 V on a raw water sample and to note the observed phenomena which are presenting in EC reactor:

3.1.1. Iron electrodes

When *U* is at 6 V, clouds of green flocs come from the anode surface and sediment at the recipient bottom under the anode. The observed green colour indicates ferrous Fe²⁺ and/or bihypoferrite HFeO₂⁻ ions presence [16]. Near the cathode, H_{2(g)} bubbles are intensely formed especially close to its lateral extremities and a froth of redbrown flocs appears on the solution surface (photo (a), Fig. 1). The observed red-brown colour indicates ferric hydroxide Fe(OH)_{3(s)} and/or hematite Fe₂O₃ presence [16]. For Kovacheva–Ninova [15], yellow-orange-redbrown colour indicates Fe(OH)_{3(s)} presence.

When *U* is at 9 V, the before carried flocs towards the surface by $H_{2(g)}$ bubbles start to come down again in the solution near the anode. This is likely affected to their alkaline nature [15–17].

When *U* is at 10 V, green flocs deposit near the anode at the side of the recipient wall.

When U is at 11 V, green flocs appear at the bottom of the recipient with red-brown flocs between the cathode and the recipient. At the surface, two froth layers are



Fig. 1. Photos showing Ghrib Dam's water after EC tests using Fe (a) and Al (b) electrodes during sedimentation. Fe flocs are more dense (sedimentation > flotation) than Al flocs (sedimentation < flotation).

observed: the first with red-brown colour is in touch with air, and the second with green colour is under the surface.

Finally when *U* is at 12 V, solution becomes limpid with a continuous formation of $H_{2(g)}$ bubbles.

Consequently, the following mechanism can be proposed:

Anode:

$$2Fe_{(s)} - 4e^{-} \rightarrow 2Fe^{2+}_{(aq)}$$
 (E° = +0.447 V) (9)

$$\begin{aligned} & Fe^{2_{+}}_{(aq)} - e^{-} \to Fe^{3_{+}}_{(aq)} & (E^{\circ} = -0.771 \text{ V}) & (10) \\ & Fe_{(s)} - 3e^{-} \to Fe^{3_{+}}_{(aq)} & (E^{\circ} = +0.037 \text{ V}) & (11) \end{aligned}$$

Solution:

$$\operatorname{Fe}_{(aq)}^{2+} + 2OH_{(aq)}^{-} \to \operatorname{Fe}(OH)_{2(s)}$$
(12)

$$2Fe^{3+}_{(aq)} + 6OH^{-}_{(aq)} \rightarrow 2Fe(OH)_{3(s)}$$
(13)

Cathode:

$$8H_2O_{(l)} + 8e^- \rightarrow 4H_{2(g)} + 8OH^-_{(aq)}$$
 (E° = -0.828 V) (14)

Total:

$$3Fe_{(s)} + 8H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + 2Fe(OH)_{3(s)} + 4H_{2(g)}$$
 (15)

This mechanism takes in consideration the apparition of green flocs of $\text{Fe}(\text{OH})_{2(s)}$ and then red-brown flocs of $\text{Fe}(\text{OH})_{3(s)}$ in the solution with hydrogen production at the cathode.

3.1.2. Aluminium electrodes

When *U* is at 4 V, small hydrogen bubbles appear at the cathode.

When U is at 7 V, the solution becomes slightly cloudy (white), bubbles speed and their sizes increase, and small oxygen bubbles appear at the anode.

When *U* is at 12 V, white-grey froth layer is observed at the solution surface (photo (b), Fig. 1).

Consequently, the following mechanism can be proposed:

Anode:

$$Al_{(s)} - 3e^{-} \rightarrow Al^{3+}_{(aq)}$$
 (E° = + 1.66 V) (16)

$$(\frac{1}{2})H_2O_{(1)} - e^- \rightarrow H^+_{(aq)} + (1/4)O_{2(g)} \quad (E^\circ = -1.23 \text{ V})$$
(17)

Solution:

$$Al_{(aq)}^{3+} + 3OH_{(aq)}^{-} \rightarrow Al(OH)_{3(s)}$$
 (18)

Cathode:

$$4H_2O_{(1)} + 4e^- \rightarrow 4OH_{(ac)} + 2H_{2(c)}$$
 (E° = -0.828 V) (19)

Total:

$$Al_{(s)} + (7/2)H_2O_{(l)} \rightarrow (1/4)O_{2(g)} + Al(OH)_{3(s)} + 2H_{2(g)}$$
 (20)

This mechanism takes in consideration the apparition of white-grey flocs of $Al(OH)_{3(s)}$ in solution with hydrogen and oxygen production at the cathode and the anode respectively.

3.2. Determination of optimal conditions

To set in focus optimal conditions for EC, two important parameters influencing directly EC efficiency are studied in this work: time t_{EC} (min) and applied voltage U (V). On the other hand, turbidity T (NTU) and conductivity C (mS cm⁻¹ at 25°C) removal percentages (Rt and Rc respectively) are selected to compare rapidly between the different tests in terms of efficiency.

The obtained results for Fe and Al electrodes are shown in Table 2 ((a) and (b) respectively). For the applied voltage, EC efficiency is most important at 8 V and less important at 4 and 12 V. For the treatment period, EC efficiency is most important for 45 min and less important for 30 and 15 min. Consequently optimal conditions are 8 V and 45 min.

3.3. *Physicochemical and bacteriological results for optimal conditions*

After determining optimal conditions for Ghrib Dam's water EC process (after EC, water samples are submitted to decantation during 30 min before aspiration and Buchner filtration), physicochemical and bacteriological analyses were done for the optimal conditions (Table 1, third and fourth columns). Microbial pollution was completely eliminated; similar results have been achieved by several authors [13,18-21]. However, KMnO₄ oxidability remains near the accepted standards (Table 1). This can be explained by the liberation of great a quantity of cell constituents which are released after the swelling and rupture of the cells [18–20]. For a better removal of these dissolved matters by charge neutralisation and adsorption, more intimate contact between water and electrodes must be designed by horizontal or vertical electrodes disposition, water passage direction between the electrodes and mechanical agitation.

3.4. Comparison between iron and aluminium EC

Fig. 1 shows the photos of Ghrib Dam's water after EC tests using iron (a) and aluminium (b) electrodes during sedimentation. Iron flocs are more dense (sedimentation > flotation) than aluminium flocs (sedimentation < flotation). This can be explained by the fact that iron is denser than aluminium. On the other hand, turbidity is better reduced with aluminium (92%) than iron (84%) electrodes (Table 1) indicating that Al flotation is better than Fe sedimentation at least for this relatively low turbid water (initial turbidity 6.50 NTU).

Final pH for Al electrodes is lower than for Fe electrodes (6.8 and 9.5 respectively, Table 1). This can be explained by the cathode reactions ((14) and (19)) which produce respectively 4OH⁻ and 8OH⁻.

Table 2 Initial and final conditions at three applied voltages U (4, 8 and 12 V) for three fixed times t_{EC} (15, 30 and 45 min) for Fe and Al electrodes.

U (V)	Stage	рН	T (NTU)	C (mS.cm ⁻¹)	I (A)	Anode mass (g)	Cathode mass (g)
			Fe ele	ectrodes, $t_{\rm EC} = 15$ mi	in		
4	Initial	7.92	6.50	2.7	0.12	271.48	273.28
	Final	7 57	1.60 (75%)	2.35 (12%)	0.15	271.45	273.28
8	Initial	792	6.50	2.00 (1270)	0.32	271.36	273.26
0	Einal	7.40	1.20 (81%)	2.7	0.02	271.30	273.20
10	Fillal Indition	7.40	1.20 (0170)	2.20 (10 %)	0.45	271.23	273.20
12	Initial	7.92	6.50	Z./	0.45	2/1.23	2/3.25
	Final	7.38	2.50 (61%)	1.98 (26%)	0.60	271.12	273.25
			Fe ele	ectrodes, $t_{\rm EC} = 30$ mi	in		
4	Initial	7.92	6.50	2.7	0.12	271.48	273.28
	Final	7.57	1.60 (75%)	2.35 (12%)	0.15	271.45	273.28
8	Initial	7.92	6.50	2.7	0.32	271.36	273.26
	Final	7.40	1.20 (81%)	2.20 (18%)	0.43	271.23	273.26
12	Initial	792	6.50	27	0.45	271.23	273 25
12	Final	7.38	2.50 (61%)	1.98 (26%)	0.60	271.12	273.25
			Fe ele	ectrodes, $t_{\rm EC} = 45$ mi	in		
4	Initial	7.92	6.50	2.7	0.12	260.64	264.20
	Final	7.65	1.60 (75%)	2.10 (22%)	0.15	260.60	264.20
8	Initial	702	6 50	27	0.32	260.50	264.04
0	Final	7.92	0.07 (95%)	1.07(070/)	0.25.2	60.20	264.04
10		7.41	0.97 (0376)	1.97 (27 /0)	0.33 2	00.29	204.04
12	Initial	7.92	6.50	2./	0.45	260.21	264.01
	Final	7.40	1.25 (80%)	1.85 (31%)	0.50	259.84	264.01
			Al ele	ectrodes, $t_{\rm EC} = 15$ mi	in		
4	Initial	7.92	6.50	2.7	0.13	43.82	44.20
	Final	8.01	0.48 (92%)	2.30 (14%)	0.13	43.80	44.20
8	Initial	792	6 50	27	0.30	43.77	44 19
0	Einal	7.70	0.00	2.7 2.25 (16%)	0.30	12.77	44.10
10		7.70	0.17 (97 70)	2.25 (10 %)	0.30	43.74	44.19
12	Initial	7.92	6.50	Z./	0.50	43.51	44.10
	Final	7.81	0.19 (97%)	2.24 (17%)	0.51	43.45	44.10
			Al ele	ectrodes, $t_{\rm EC} = 30$ mi	in		
4	Initial	7.92	6.50	2.7	0.13	43.26	44.04
	Final	8.01	0.25 (96%)	2.25 (16%)	0.12	43.23	44.04
8	Initial	7.92	6.50	2.7	0.30	43.16	44.01
0	Final	6.86	0.11 (98%)	2 20 (18%)	0.31	43.10	44.01
10	Initial	7.02	6 50	2.20 (1070)	0.51	42.00	44.00
12		7.92	0.30	2.7	0.50	43.09	44.00
	Final	/./0	0.54 (91%)	2.14 (20%)	0.51	43.02	44.00
			Al ele	ectrodes, $t_{\rm EC} = 45$ mi	in		
4	Initial	7.92	6.50	2.7	0.13	42.96	43.97
	Final	7.73	0.36 (94%)	2.21 (18%)	0.13	42.92	43.97
8	Initial	7.92	6.50	2.7	0.30	42.90	43.94
	Final	7.47	0.18 (97%)	2.10 (22%)	0.31	42.83	43.94
12	Initial	792	6.50	27	0.50	42.73	43.93
	Final	7.56	0.00	1.94(28%)	0.52	42.64	43.03
	rmai	7.50	0.00 (90%)	1.94 (2070)	0.52	72.07	-1J.JJ

Because this work was carried out by using a low raw water turbidity, the performances in terms of pollutant removal (turbidity and others elements) are difficult to analyse. But in terms of water quality (accepted standards in Table 1), the only weak point of Fe electrodes appears to be the pH value (too high) then other criteria (Ca²⁺, Mg²⁺, SO₄²⁻, total hardness) which are better than with Al electrodes. Moreover, the Al³⁺ that remains in solution when working at pH lower than 7 (in its ionised form) may be toxic for people with kidney problem. According to the price and the problem of sustainable development linked to aluminium industry, iron electrodes could appear in fact as the best solution.

Al and Fe electrodes are at least in accord concerning microbial pollution which is completely eliminated. This is essentially the consequence of the electrical field which is improved with metallic cations [21]. However, ammonium for Al does not exist but for Fe is 2.25 mg L⁻¹ (which is higher than accepted standard 0.5 mg L⁻¹, Table 1) indicating that NH_4^+ is better adsorbed on Al(OH)_{3(s)} than on Fe(OH)_{2(s)}/Fe(OH)_{3(s)} [22] after the swelling and rupture of the cells as mentioned above.

Natural water, highly contaminated with coliforms, was electrochemically treated in a stirred batch system with the use of two Ti electrodes by Patermarakis and Fountoukidis in 1990 [23]. The process was found to be effective and the percentage of the initial concentration of bacteria which were destroyed was found to be proportional to both treatment time and the square of current density; consequently the time needed for complete disinfection was inversely proportional to the square of current density. The percentage above was found to be independent of the initial concentration of germs at least for the range of concentrations employed [23].

In order to reduce the risks of Legionnaires' disease, caused by the bacterium *Legionella pneumophila*, disinfection of tap water systems contaminated with this bacterium is a necessity. The study by Delaedt *et al.* [24] investigates if electrochemical disinfection is able to eliminate such contamination. Hereto, water spiked with bacteria (104 CFU *Escherichia coli* or *L. pneumophila* mL⁻¹) was passed through an electrolysis cell. The spiked tap water was completely disinfected during passage through the electrolysis cell [24].

3.5. The concept of the "ratio r active volume on reactor volume"

In other context, optimising EC of Ghrib Dam's water has conducted us to propose the concept of the *ratio r active volume on reactor volume*. Indeed, in the same procedure as previously, laboratory experiments were carried out to investigate EC of Ghrib Dam's water in

batch using iron electrodes which are also of the same dimensions and plunged in a device with 4 L as volume (eight times greater than the first one which was 0.5 L). For each electrode, the immerged (active) surface is 56.64 cm² (4.8 cm \times 11.8 cm) and the distance between them is variable from 2 to 10 cm (it was maintained constant at 4 cm in the first experiments).

3.5.1. Optimal voltage determination

The optimal voltage for which the best turbidity and conductivity reductions are obtained is determined for a treatment time fixed at 60 min. Three voltages 5, 10, and 15 V have been selected according to the maximal capacity (15 V) of the power supply. The initial and final current intensity *I* values, corresponding to each applied voltage *U*, for every test are read (Table 4). The noted observations are as follows.

For U = 5 V, 10 min after treatment start, green flocs come out from the anode with intense H_{2(g)} bubbles emanation from the cathode. At the end of treatment, the solution becomes less limpid.

Table 3

Physicochemical and bacteriological parameters of Ghrib Dam's raw water (April 17th, 2004), which was used in the introduction of the r ratio concept, and their accepted standards (close to WHO standards).

Parameters	Raw water	Accepted standards
Turbidity (NTU)	5.3	<5
pH	8	6.5 to 8.5
Temperature (°C)	17.9	/
Conductivity	2.7	3.10
(mS cm ⁻¹) at 25°C		
Total coliforms	Uncountable	<10/100mL
Faecal coliforms	21	0/100mL
Streptococcus	84	0/100mL
Sulphates (mg L ⁻¹)	200	<400
Ammonium (mg L ⁻¹)	0.0	< 0.5
Phosphates (mg L ⁻¹)	0.0	< 0.5
Nitrates (mg L ⁻¹)	0.0	<50
Iron (mg L ⁻¹)	0.0	< 0.3
KMnO ₄ Oxidability	7.9	<3
(mg L ⁻¹)		
$COD (mg O_2 L^{-1})$	9.6	3
Dry residue (mg L ⁻¹)	1482	<2000
Total hardness	675	<500
(mg L ⁻¹ as CaCO ₂)		
Calcium (mg L ⁻¹)	194	<200
Magnesium (mg L ⁻¹)	162	<150
Nitrites (mg L ⁻¹)	0.0	< 0.1
TAC (mg L^{-1} as CaCO ₂)	110.5	<500
Chlorides (mg L ⁻¹)	440	<500

For U = 10 V, at the end of treatment, clouds of green flocs appear on the anode surface and yellow broth appears on the solution surface.

For U = 15 V, 5 min after treatment start, a more important H_{2(g)} bubbles emanation is observed at the cathode, a red-brown colour appear in the solution, and a start of yellow broth formation is observed on the solution surface. After 10 min, red-brown flocs aggregate at the recipient bottom under the anode.

As can be expected, according to Table 4, the found results for U = 15 V (I = 0.68 A) comply better with turbidity and conductivity accepted standards. For Holt *et al.* [25], the current density (*i*), which is the delivered current at the electrode divided by electrode active area, is a used parameter for EC design. The current densities ranking from 10 to 2000 A m⁻² have been reported from 16 literature sources. The majority of these sources report a current density in the range 10–150 A m⁻² [25]. For this voltage U = 15 V, *i* being 120 A m⁻² (I = 0.68 A, $S = 56.64 \times 10^{-4}$ m²) is in the range of the majority of bibliographic sources (10–150 A m⁻²).

The S/v ratio, surface area on water volume, was expected as being a parameter of significant development. It has been reported that when the S/v ratio increase, the optimal current density decrease. The reported values of S/v ratio are all of the same order: 18.8 to 42.5 m² m⁻³ [25]. For this present case, the S/v ratio is 1.62 m² m⁻³ ($S = 56.64 \times 10^{-4}$ m² and $v = 3.5 \times 10^{-3}$ m³). This ratio is very lower than the first reported value (18.8 m² m⁻³). It will be necessary to increase the active surface to meet the literature interval.

3.5.2. Optimal time determination

The time $t_{\rm FC}$ for which EC process is better is determined. Thus, the voltage U being fixed at 15 V, four values of the time $t_{\rm FC}$ are selected: 15, 30, 45, and 60 min (for which, the test was previously accomplished). The initial and final conditions are presented in Table 5. The noted observations during these tests are always the same as for $t_{\rm FC} = 60$ min and U = 15 V test. Finally, optimal time is tEC = 45 min since turbidity and conductivity are reduced at 69.81 and 29.62% respectively and their final values comply with accepted standards. These results are confirmed by other researchers [26,27] whose Holt et al. [27] who talk about three stages for EC in batch: inert stage (0-15 min), active stage (15-35 min), and stable stage (more than 35 min). For this found optimum at this step, *i* is 102 A m⁻² (I = 0.58 A, $S = 6.64 \times 10^{-4} \text{ m}^2$) which is in the bibliographic majority range (10–150 A m⁻²).

Table 4 Initial and final conditions at three different voltages of applied current for EC of Ghrib Dam's water using Fe electrodes (*U* is the voltage and *I* is the current intensity, numbers between brackets indicate removal percentage).

<i>U</i> (V)	Stage	pН	Turbidity (NTU)	Conductivity (mS cm ⁻¹) at 25°C	I (A)
5	Initial	8	5.3	2.7	0.21
	Final	8.93	2.7 (49.05)	2 (25.92)	0.21
10	Initial	8	5.3	2.7	0.43
	Final	8.44	2 (62.26)	2 (25.92)	0.43
<u>15</u>	Initial	8	5.3	2.7	0.68
	Final	<u>9.46</u>	<u>2 (62.26)</u>	<u>1.9 (29.62)</u>	0.68

Table 5

Initial and final conditions of EC of Ghrib Dam's raw water at U = 15 V for four times t_{EC} 15, 30, 45, and 60 min (I is the current intensity, numbers between brackets indicate removal percentage).

Time (min)	Stage	рН	Turbidity (NTU)	Conductivity (mS cm ⁻¹) at 25°C	I (A)
15	Initial	8	5.3	2.7	0.70
	Final	8.25	1.9 (64.15)	2.1 (22.22)	0.71
30	Initial	8	5.3	2.7	0.63
	Final	8.75	1.8 (66.03)	2 (25.92)	0.64
45	Initial	8	5.3	2.7	0.58
	Final	9.59	1.6 (69.81)	<u>1.9 (29.62)</u>	0.58
60	Initial	8	5.3	2.7	0.68
	Final	9.46	2 (62.26)	1.9 (29.62)	0.68

Distance (cm)	Stage	pН	Turbidity (NTU)	Conductivity (mS cm ⁻¹) at 25°C	I (A)
2	Initial	8	5.3	2.7	1.05
	Final	10.74	2.3 (56.6)	1.8 (33.33)	1.05
4	Initial	8	5.3	2.7	0.58
	Final	9.59	1.6 (69.81)	1.9 (29.62)	0.58
10	Initial	8	5.3	2.7	0.35
	Final	<u>8.5</u>	<u>1.7 (67.92)</u>	<u>1.9 (29.62)</u>	0.35

Initial and final conditions for optimisation of distance between the two electrodes for EC of Ghrib Dam's raw water (U = 15 V and tEC = 45 min, numbers between brackets indicate removal percentage).

3.5.3. Optimal distance between the two electrodes determination

After determination of these parameters U = 15 V and tEC = 45 min, the optimal distance separating the electrodes is determined. For this the distance between the two electrodes is varied: 2, 4 and 10 cm. The S/v ratio remains unchanged since it did not depend on separation between two electrodes. The initial and final conditions of electrocoagulated waters are presented in Table 6. The anode mass decreased as follows: 0.32% for 2 cm, 0.18% for 4 and 10 cm.

The noted observations during these tests are the same as for previously accomplished tests. One can expect that more the distance between electrodes is lower more the process efficiency is higher. But according to the found results for different distances between the two electrodes, the optimal distance is 10 cm (Table 6). Indeed, better turbidity (68%) and conductivity (30%) reductions are obtained with final results in the accepted standards.

For these conditions (Table 6), when the distance between the two electrodes increase (2, 4 and 10 cm), current intensity decrease (1.05, 0.58 and 0.35 A respectively). The current density *i* follows hence the same evolution: 185, 102 and 62 A m⁻² respectively ($S = 56.64 \times 10^{-4}$ m²). These values are in the majority bibliographic range (10–150 A m⁻²) with the exception of the first value (185 A m⁻²) which is correspondent to the distance 2 cm.

According to these results, it is useful to introduce a new parameter to take simultaneously in consideration with inter electrode distance during EC design. Indeed, it is about *ratio r active volume on reactor volume which is*



Fig. 2. Illustration of the evaluation of *ratio r active volume on EC reactor's volume full with water to treat* using two electrodes with same dimensions.

full of water. The *active volume* is the active surface multiplied by the distance between the electrodes. Fig. 2 illustrates the ratio r calculus where *L*, *l*, and *d* represent the electrode immerged height, the electrode width, and the distance separating the electrodes, respectively:

$$r = \frac{active \ volume}{reactor \ volume \ full \ of \ water} = \frac{d \times 56.64 \times cm \times cm \times cm}{3.5 \times 1000 \times cm \times cm \times cm} = 0.0162 \times d$$
(21)

For d = 2 cm, r = 0.0324; for d = 4 cm, r = 0.0648; and for d = 10 cm, r = 0.162. The ratio r = 0.162 (d = 10 cm) is then optimal for these tests.

The concept of the *ratio r active volume on reactor volume which is full of water* joins the concept of *feed zone* and *reaction zone* (maybe considered as active volume) which is explained by Buso *et al.* [28].

3.6. From camp and stein's coagulation paper to vik et al.'s EC paper

From 1943 [29] to 1984 [22], the concept of chemical coagulation has been developed to electrochemical coagulation (EC). Indeed, a renewed interest in EC has been spurred by the search for reliable, cost-effective water treatment processes. By comparison, conventional chemical dosing typically adds a salt of the coagulant, with settling providing the primary pollutant removal path [30–33].

4. Conclusion

The laboratory tests show that EC process is highly efficient for surface water treatment. The *in situ* formed metal cations (Fe²⁺/Fe³⁺ or Al³⁺) neutralise colloidal particles and microorganisms and conduct to hydroxides (Fe(OH)_{2(s)}/Fe(OH)_{3(s)} or Al(OH)_{3(s)}) which adsorb organic matter. Since good removal of colloids and total removal of microorganisms are reached, water treatment by EC can be presented as a potential process for the next years. To enhance the EC process, a more direct contact between water contaminants and the anode and the cathode must be designed especially in continue

Table 6

mode (zigzag passage) where the *ratio r active volume on reactor volume which is full of water* would be increased. Finally, the pH evolution with time during EC experiments would give more information on EC process.

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