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Electrocoagulation Process: A Mechanistic Review at the Dawn of its Modeling

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Abstract

During the two last decades, electrocoagulation method (EC) was the focus of many industrial applications and remains a fascinating domain of research. Most published researches concern uses in treating potable water and wastewaters to increase both the removal of dissolved and undissolved contamination. Significant achievements have been realized comprising participations to fundamental comprehension, electrode metals, working parameters, device conception, and economic aspects determinations. Despite the fact that there are several benefits mentioned through the specialized references, the EC large-scale use is not until now viewed as a recognized wastewater technique due to the absence of viable models used in designing device. The present review discusses the mechanisms involved in the EC process and opens a broad perspective on its modeling. The scientific community is near to suggest empirical/theoretical models to present the EC technology as a viable green process. However, more great efforts remain to be accomplished. Technological software developers such as COMSOL™ Multiphysics are invited to insert the EC process in their electrochemistry module to better commercialize this intensified technique and encourage its massive use through the world.

Keywords

COMSOL™; Electrocoagulation; Modeling; Potable water

Abbreviations

CCD: Central Composite Design

CFD: Computational Fluid Dynamic

EC: Electrocoagulation

FD: Factorial Design

RSM: Response Surface Methodology

Introduction

Preserving water resources becomes one of the humankind greatest challenges [1,2]. It has to treat many problems such as population growth, deforestation, rapid urbanization, industrialization and warming global climate change [3-5]. Now, the availability of potable water is restricted and not assured [6]; water contamination may significantly affect aquatic ecosystems and the accessibility to healthy freshwater [7]. Consequently, there is a necessity to promote performant techniques and methods for depolluting and controlling used waters [8], to preserving widely quality and increase quantity whereas assuring environmental safeguard [9,10]. At the same significance, more performant potable water processes treatment is needed to monitor hazards developed by ecological pollution [11-13]; as an illustration, existence of nitrate or fluoride ions at elevated levels [5,14].

The electrocoagulation (EC) technique may be employed for treating potable water and depolluting wastewater [15]. EC includes producing cationic metallic species inside the reactor through accelerated corrosion of consumable metallic anode induced via electric power exercised over the plaques [16]. Following the water acidity, the metallic cations formed electrochemically are instantaneously subjected to aquatic hydrolysis, producing different metallic forms comprising hydroxide flocs (eliminating contaminants through adsorption/settling). Usually, aluminum and iron metals are employed as electrodes due to several benefits: they are abundantly available at low cost, their relative low poisoning effect, and their elevated (+2/3 and +3, respectively) valence that conducts to a performant elimination of contaminant [17-19]. Moreover, concurrent cathodic reaction lets contaminant elimination either upon attachment on cathode or through fixation on H2(g) bubbles liberated at the negative pole. In a general manner, the anode and the cathode materials are selected from the identical metal, even if electrochemical dissolution should happen exclusively at the positive pole.

Usually, electrochemical coagulation is performed in two configurations: a batch or uninterrupted mode [20]. There is a vast number of cases where EC was successfully applied [21,22]. Relatively, EC is an ancient technique, at the same age as electricity [22]. Employing this electrochemistry-based method in treating potable water factories was mentioned during the 19th century in UK, and treating wastewater factories [23] in the United States in the dawn of the 20th [24]. In the late 30s, this electrical process has been substituted by conventional coagulation [25,26] and by biological techniques for eliminating colloids and dissolved organic matters in wastewater, respectively [27]. This was due to the more elevated functioning price; indeed, the cost of electricity at this time was insupportable. Naturally, the trend has radically varied, and the benefits of EC have been shown once again from the 80s. Several advantages have been mentioned in the written works [28] with the particular disadvantages of EC comparatively with competitive techniques, as listed in (Table 1). However, EC is known for additional problems than those mentioned in Table 1. As an example, the requirement for sludge treatment, even if conventional coagulation [29] and activated sludge method have to resolve the identical problem, remains to be resolved. The constitution of EC sludge is similar to that formed employing conventional coagulation [30,31] when either $Al_2(SO_4)_3.18H_2O$ or $FeCl_3$ are injected; this significates that sludge handling must be identical. On the contrary, a particular problem of EC is that, until recently, there are approximately no detailed surveys of EC modeling for treating water [5].

Consequently, this work aims to abridge, debate, and examine breakthroughs on modeling methods realized for simulating the scaling-up the EC process. First, different mechanisms for contamination reduction are presented. After that, the fundamental process variables and device conception characteristics will be reviewed intending to initiating the specifics of modeling features. Subsequently, the principal techniques and design approaches will be assessed and associated with the techno-economic study of the EC method. Ultimately, several views for subsequent investigation and progress will be proposed.

Hypothetical Context of EC Technique

EC merges distinct mechanisms that may be electrochemical (metal dissolution and water reduction, pollutant electrooxidation or electro-reduction, etc.), chemical (acid/base equilibria with varying pH, hydroxide precipitation, a redox reaction in the solution, etc.) and physical (physical adsorption, coagulation, flotation, etc.) [34,35]. All the mentioned stages may happen successively or in parallel (Figure 1). Figure 1 focuses on the intricacy and the interaction among the routes of the electrochemical method. These routes are described in the following section [5].

EC reactions

During the electrochemical coagulation technique, metal species are formed inside the recipient through dissolving electrochemically the metallic plaque, frequently in Fe or Al [5].

The involved reactions may be outlined:

- At the anode, the metal is oxidized into cations:

$$M \rightarrow M^{Z+} + Ze^- \qquad (1)$$

In Equation (1), Z is the number of electrons carried during the anodic dissolution phenomenon per mole of metal. In the situation of an elevated anode potential, there is a great probability

Benefits	Disadvantages
General technique	Requires repair
Treats potable water and used water	Electrode passivation over time [5]
Integrates oxidation, coagulation, and precipitation (conducting to minimal capital costs [28])	The necessity of high-conductivity water
Decreased requirement for chemicals (substituted with metal electrodes and electric power) [32]	Lack of systematic reactor design [24]
Reduced operating expense	
The decreased hazard of ancillary contamination	
Minimal sludge formation	
Without moving parts [5]	
Reduced power needs	
Solar energy may be employed [33]	

Table 1: Benefits and disadvantages of the EC process [5,28].

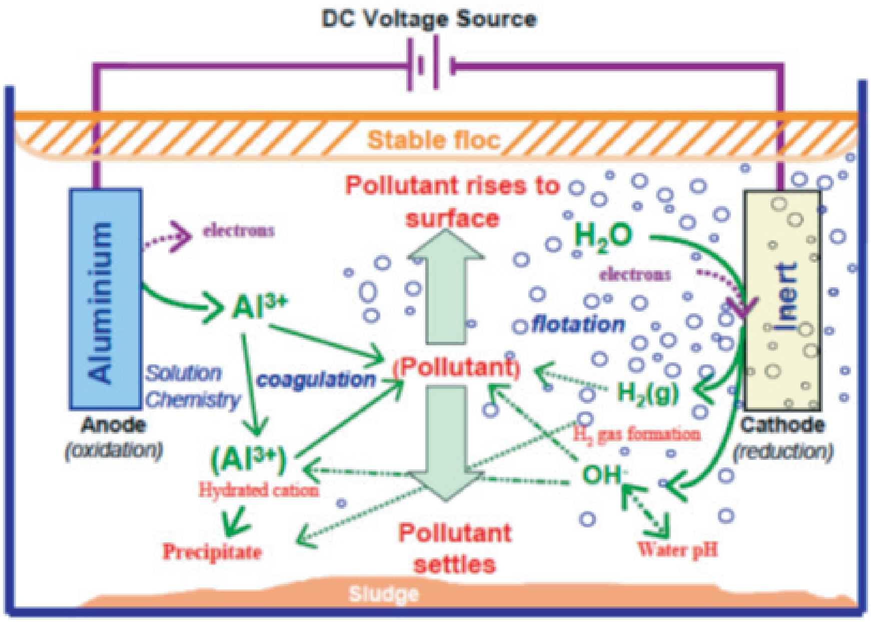

Figure 1: Mutual actions happening inside the EC device [36].

that auxiliary reactions may happen [37,38]. Moreover, oxidation of water may conduct to hydronium cation and oxygen gas; in addition, if the chloride anions are present, they may be oxidized into Cl2. The last-mentioned being a powerful oxidant can take part in oxidizing the soluble organic matters or can conduct to generating ClOH that as well functions as an oxidizer [5,39].

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$ ($E° = 1.23$ V/ENH) (2)

$2Cl^- \rightarrow Cl_2 + 2e^-$ ($E° = 1.36$ V/ENH) (3)

$Cl_2 + H^2O \rightarrow ClOH + Cl^- + H^+$ (4)

- At the cathode: water is reduced into hydrogen gas and hydroxyl anions:

$3H_2O + 3e^- \rightarrow (3/2)H_2 + 3OH-$ ($E° = 0.00$ V/ENH) (5)

The quantity of metal solubilized through oxidization at the anode may be computed employing Faraday's law (Equation 6). Consequently, the amount of metal m depends on the residence period t and the electric current I [5]:

$$m = \phi \frac{It}{ZF} M \qquad (6)$$

In Equation (6), M is the atomic weight of the electrode metal, and F is Faraday's constant. Nevertheless, Faraday's law (ϕ=1) is correct if all the electrons in the reactor take part exclusively in the metal-liberation phenomenon at the anode. In the case of parallel reactions happening, a corrective term, designated *current efficiency or faradic yield* (ϕ), is employed to take into consideration the difference among the theoretical and experimental solubilization of the anode [40,41]. Generally, this term is smaller than 1 [42]; however, ϕ may be bigger than one if the chemical and the electrochemical oxidation routes of the metal happen concurrently. This final case is recurrent with Al [43,44]. The metal cations liberated inside the solution go through diverse equilibrium reactions that correspond to the acid/base, complexation, precipitation and redox reactions. The contribution of these elimination routes is a function of pollutant species (Table 2) [5].

Dissolved contaminants	Removal mechanism	References
Sulfide anions	Precipitation	[45]
Calcium cations	Co-precipitation	[46]
Phosphate anions	Precipitation, adsorption, complexation	[47,48]
Organic compounds	Complexation, co-precipitation	[49,50]
Fluoride anions	Complexation, precipitation	[41,51]
Arsenate anions		[52]

Table 2: Equilibrium reactions of metal hydroxides with dissolved contaminants in the solution [5].

Nevertheless, concerning the positive ions liberated from the positive pole, the frequent event is the generation of $Fe(OH)_{3(s)}$/$Al(OH)_{3(s)}$ that show low solubility and easily settle. Consequently, dissolved contaminants in water can as well be attached by physical or chemical means onto the flocs. Possible contamination removal routes implicate [5]:

- The completely physical entrapment of soluble matters happens through $Fe(OH)_{3(s)}/Al(OH)_{3(s)}$, adsorption, and complexation. As an example, phosphates are eliminated upon complexing and/or by precipitating with $Fe(OH)_{3(s)}/Al(OH)_{3(s)}$ and/or by attachment on the latter. At the same time, the soluble organic matter reduction may be explained by the co-precipitation and/or to complexation and/or to the electrostatic attraction on the surface of $Fe(OH)_{3(s)}/Al(OH)_{3(s)}$. Concerning the complexation, the contaminant may play the role of a ligand (L) to fix a hydrous iron moiety or $Al(OH)_{3(s)}$ (Equations (7) and (8)):

$$L\text{-}H_{(aq)} + (OH)OFe_{(s)} \rightarrow L\text{-}OFe_{(s)} + H_2O \qquad (7)$$

$$L\text{-}H_{(aq)} + (OH)(OH)_2Al_{(s)} \rightarrow L\text{-}(OH)_2Al_{(s)} + H_2O \qquad (8)$$

- Electro-oxidating anodically or electro-reducing cathodically electro-active ions or molecules, like reducing Cr (VI) into Cr (III) may be followed by: (1) Cr (III) hydroxide settling [53], (2) reducing anionic nitrates into nitrite, (3) ammonia and nitrogen gas [54], and (4) oxidizing instead reducing if arsenic As (III) is oxidized into arsenic As(V) [55]. It was mentioned that heavy metals could as well go through electro-reducing cathodically throughout the electrochemical coagulation operation [56,57].

- Adsorbing directly contaminants on the electrodes surfaces: particularly for F- because of the electro-condensation where F- are fixed anodically upon the electric effects [58,59].

These pathways for dissolved contaminants are shown in (Figure 2). Practically, there is frequently a dominant mechanism for each pollutant depending on the type of this contaminant [5].

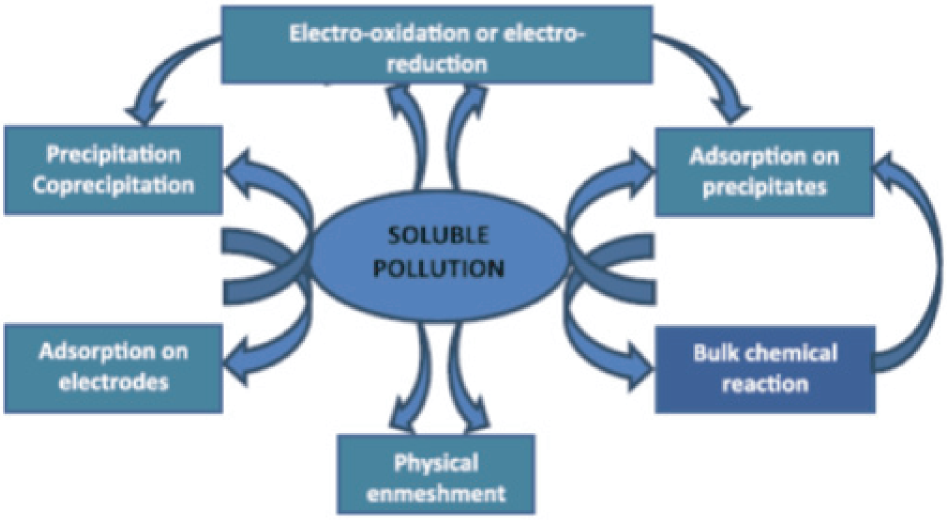
Electro-oxidation or electro-reduction
Precipitation Coprecipitation
Adsorption on precipitates
SOLUBLE POLLUTION
Adsorption on electrodes
Bulk chemical reaction
Physical enmeshment

Figure 2: Principal mechanisms of dissolved contamination removal throughout EC [5].

Frequent cases are listed in Table 2, and more information may be noted in survey articles on EC [22,54].

Simultaneously, colloids and emulsions are as well neutralized through injecting coagulant without interruption from electro-dissolution like the conventional coagulation [60]. The penchant of colloids to destabilize or to persist distinctly and scattered is a consequence of the net inter-particles force that is

Mechanism	Description
Compression of the double layer	The compression of the double layer of a colloid is formed by mutual actions with the soluble ionic species produced from the electrochemical dissolution of the sacrificial anodes. This compression impacts the electrical potential disparity among the particle surface and the bulk solution and conducts to decreasing the repulsive forces among particles [49].
Charge neutralization	Charge neutralization is realized through adsorption of ionic metal species/hydronium cations/hydroxyl anions or by the precipitation of charged hydroxide precipitates onto the surface of charged colloids existent in water/wastewater [64]. This charge neutralization may be illustrated through a variation of the zeta potential throughout the isoelectric point.
Sweep coagulation	Colloids entrapment (enmeshment mechanism or sweep coagulation): Hydroxide precipitates entrap the colloids existing in water/wastewater. This destabilization mechanism is a function of metal hydroxide precipitates that may be generated [64]. This mechanism dominates around pH 7 [65].

Table 3: Destabilization mechanisms of colloidal particles [5].

envisaged upon the addition of opposing forces among the attractive van der Waals and the repulsive forces of the electrical double-layer as described by the DLVO [61,62] theory as a first approximation [63]. Neutralizing routes are listed in Table 3 [5]. During the EC operation, these neutralization routes (Figure 3) can happen together or one by one following the characteristics of the water/wastewater to remedy, the contaminants to be eliminated, the working situations (particularly electric current), and the type of the metal. Opposite to dissolved contaminants, the controlling pathway is, consequently, more laborious to describe since it is frequently more related to working situations than on the type of the colloids. Then, the neutralization stage is pursued by flocculation that is composed of the assemblage of the destabilized colloids and the precipitates [66,67]. Flocculation performance is a function of the efficiency of the destabilization step and the collision velocity of colloids [5,49].

Since reducing dissolved matters implicates attachment and entrapment, the routes of dissolved and solid matters are in themselves related. The aggregated pollutants, constituting EC sludge, may thus be eliminated physically from water, in the device or employing a downstream process (Figure 4). In the recepient, the two principal means implicate electroflotation (EF) and decantation [68,69]. Moreover, decantation and EF may as well be employed as downstream processes; however, in this situation, additional downstream techniques are accessible for sludge elimination comprising filtration, centrifugation, hydrocycloning, and flotation [5].

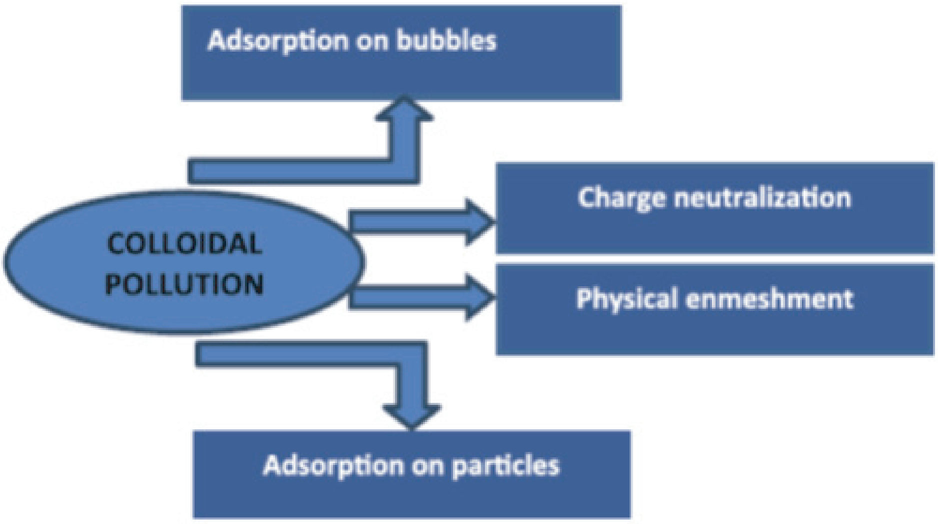

Figure 3: Principal mechanisms of insoluble colloidal contamination reduction employing EC [5].

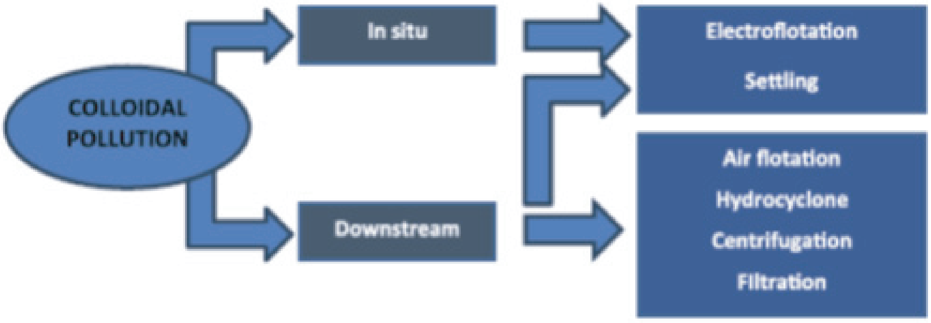

Figure 4: Principal mechanisms of EC sludge elimination [5].

Case of Al electrodes

For Al electrodes, only just the half-oxidation reaction among Al^{3+}/Al pursues Equation (1), because $Z = 3$. More than the chemical equations presented above, additional monomeric species are produced from the instantaneous hydrolysis of Al^{3+} cations following the acid/base reactions (Equations (9)-(12)) and Al^{3+} amount [5,38].

$$Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+ \quad (9)$$

$$Al(OH)^2+ + H_2O \rightarrow Al(OH)_2^+ + H+ \quad (10)$$

$$Al(OH)^2+ + H_2O \rightarrow Al(OH)_3 + H+ \quad (11)$$

$$Al(OH)^3 + H_2O \rightarrow Al(OH)_4^- + H+ \quad (12)$$

Reaction	**pK**
$Al^{3+} + H_2O = Al(OH)^{2+} + H^+$	4.997
$Al^{3+} + 2H_2O = Al(OH)_2^+ + 2H^+$	10.094
$Al^{3+} + 3H_2O = Al(OH)_{3(aq)} + 3H^+$	16.791
$Al^{3+} + 3H_2O = Al(OH)_{3(s)} + 3H^+$ (amorphous)	8.578
$Al^{3+} + 2H_2O = AlO(OH)_{(s)} + 3H^+$ (boehmite)	10.800
$Al^{3+} + 4H_2O = Al(OH)_4^- + 4H^+$	22.688
Reaction	***E*° (V)**
$Al^{3+} + 3e^- = Al_{(s)}$	-0.41

Table 4: Equilibrium constant (pK) and standard reduction potential aluminum species (E° (V)) of Al and Fe species [5].

Aluminum speciation and partition may be drawn from E-pH Pourbaix diagrams if the implied reactions are under thermodynamic control. This may be concluded from the equilibrium constants for acid/base reactions and standard reduction potentials listed in Table 4 [5].

Practically, dissolved Al^{3+} cations dominate if pH is less high than 4; dissolved aluminate anions take over if pH is more significant than 10, during the time that the insoluble $Al(OH)_{3(s)}$ form takes control apart from that [5].

The production of polymeric species has as well been mentioned: $Al(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$ and $Al_{13}O_4(OH)_{24}^{7+}$ [5]. The Lewis acidity of aluminum equilibrates the generation of OH^- anions at the cathode, which produces a buffered impact and conducts to a final pH among 7 and 8 which powerfully changes from conventional coagulation employing Al salts [49]. Therefore, monomeric and polymeric species provoke lately the production of the amorphous $Al(OH)_{3(s)}$ "sweep flocs" which possess important surface areas useful for fast adsorbing dissolved organic matters and trapping colloids [70-73]:

$$nAl(OH)_3 \rightarrow Al^n(OH)_{3n} \tag{13}$$

Auxiliary reactions can happen on the electrodes because of a simple chemical attack of aluminum under acid or alkaline conditions, respectively [8,37]:

$$2Al + 6H^+ \rightarrow 2Al^{3+} + 3H_2 \tag{14}$$

$$2Al + 6H_2O + 2OH- \rightarrow 2Al(OH)_4^- + 3H_2 \tag{15}$$

The result is that the quantity of soluble Al liberated through the EC process overpasses the expected level predicted using Faraday's law [5]. Consequently, the faradic yield φ is more important than 100% and may attain 200% [38].

A problematic issue of EC technique is the passivation of the cathode since it elevates both the cell voltage and the energy consumption. The passivation phenomenon may be surmounted through optimizing the current reversal frequency [74] or NaCl injection to accelerate pitting corrosion upon a chemical reaction between Cl^- fixed on the aluminum oxide film with Al3+ species in the oxide lattice [5]. The velocity of chemical corrosion of dissolved Al electrodes is a function of two mechanisms [75]:

- The generation and construction of a passive aluminum-oxide film;

- The following partial demolition of this film by pitting.

The pitting corrosion is a function of the initial pH, the type and the amount of supporting electrolyte [76] and the current density [5]. The favorable action of anions on passive aluminum-oxide film in the decreasing classification [77] is Cl^-, Br^-, I^-, F^-, ClO_4^-, OH^-, SO_4^{2-}. The pitting corrosion may significantly influence the global dissolution of sacrificial Al anode. The pitting potential E_{pit} (V) diminishes with the logarithm of chloride concentration (ppm), as illustrated through Equation (16) [38]:

$$E_{pit}=1.75-0.72\times Ln[Cl^-] \tag{16}$$

The pathway of chemical dissolution involved through pitting corrosion in the existence of chloride ions may be described [78]:

$$2Al + 6HCl \rightarrow 2AlCl_3 + 3H_2 \tag{17}$$

$$AlCl_3 + 3H_2O \rightarrow Al(OH)_3 + 3HCl \tag{18}$$

Case of Fe electrodes

The electrochemical reactions happening at Fe electrodes or in aqueous medium through Fe-EC are more complicated than with Al, because anode oxidation may conduct either to ferrous or ferric cations [5,79,80]:

$$Fe \rightarrow Fe^{2+} + 2e^- (19)$$

$$Fe \rightarrow Fe^{3+} + 3e^- (20)$$

Fe^{2+} and Fe^{3+} may be hydrolyzed in the recipient. Moreover, they may generate several monomeric and polymeric species whose proportions are function of the Fe^{3+} level and the pH degree: $Fe(OH)^{2+}$, $Fe(OH)_2^+$, $Fe(OH)_2^{4+}$, $Fe(OH)_4^-$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)^{2+}$, $Fe(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_4^{2+}$ and $Fe(OH)_3$. This difficulty is as well illustrated in Table 5 that sums up the acid/base and equilibrium constants and standard reduction potentials of monomeric species [5].

Reaction	**pK**
$Fe^{2+} + H_2O = Fe(OH)^+ + H^+$	9.397
$Fe^{2+} + 2H_2O = Fe(OH)_2 + 2H^+$	20.494
$Fe^{2+} + 2H_2O = Fe(OH)_{2(s)} + 2H^+$	13.564
$Fe^{2+} + 2H_2O = Fe(OH)_3^- + 2H^+$	28.991
$Fe^{3+} + H_2O = Fe(OH)^{2+} + H^+$	2.187
$Fe^{3+} + 2H_2O = Fe(OH)^+ + 2H^+$	4.594
$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+$	12.56
$Fe^{2+} + 2Fe^{3+} + 8H_2O = Fe_3(OH)_{8(s)} + 8H^+$	20.222
$Fe^{3+} + 2H_2O = \alpha FeO(OH)_{(s)} + 3H^+$	0.491
$Fe^{3+} + 2H_2O = \gamma FeO(OH)_{(s)} + 3H^+$	1.371
$Fe^{3+} + 4H_2O = Fe(OH)_4^- + 4H^+$	21.588
$2Fe^{3+} + 2H_2O = Fe_2(OH)_2^{4+} + 2H^+$	13.771
$2Fe^{3+} + 4H_2O = Fe_3(OH)_4^{5+} + 4H^+$	6.228
Reaction	**$E°$ (V)**
$Fe^{2+} + 2e^- = Fe_{(s)}$	-0.41
$Fe^{3+} + 3e^- = Fe_{(s)}$	-0.04
$Fe^{3+} + 2e^- = Fe^{2+}$	+0.77
$FeO_4^{2-} + 3e^- + 8H^+ = Fe^{3+} + 4H_2O$	+2.20

Table 5: Equilibrium constant and standard reduction potential of iron species [5].

In spite of certain confusion in large part of the literature about the implied stages of Fe-EC, contemporary references largely hypothesis that the anode oxidation liberates Fe^{2+} since it has been established that the dissolution proportion of Fe^{3+} may be neglected [81-83]. The oxidation of Fe^{2+} to Fe^{3+} is a function of pH and the dissolved oxygen amount [84]. When pH is acid, ferrous cations oxidize very slowly in the presence of dissolved O_2 (Equation (21)); however, if pH is neutral or alkaline, ferrous cations are instantaneously converted into $Fe(OH)_2$ (Equation (22)) which is immediately oxidized by dissolved O_2 to $Fe(OH)_3$ (Eq. (23)) [5]:

$$Fe^{2+} + O_2 + 2H_2O \rightarrow Fe^{3+} + 4OH^- \tag{21}$$

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \tag{22}$$

$$4Fe(OH)_2 + O_2 + 2H_2O \rightarrow 4Fe(OH)_3 \tag{23}$$

As a result, several references mentioned that the Fe dissolution obeys to the Faraday's law with a faradic yield comprised in the range of 80 and 100% [42,43,75]. However, different authors affirm that there is a gap between the quantity of Fe theoretically solubilized evaluated using Faraday's law and the quantity of detected solubilized Fe founded on $Z = 2$ [5]. If pH is acid, ϕ is bigger than 100%; at the same time, if pH alkaline, the opposite is correct. At smaller pH degrees, acceptable interpretations are identical to those mentioned for aluminum: chemical corrosion and pitting corrosion at both electrodes in the existence of certain ionic species, like Cl^- [5,75]. At bigger pH degrees, the dissolution efficiency diminishes under the Faraday's law levels since additional reactions happen next to the anode, comprising oxygen formation (Equation (2)). If pH is alkaline, Fe oxidation conducts to Fe(III) production following Equation (20) in the form of oxide or hydroxide species. This oxidation reduces the Fe amount generated since Fe(III) production needs 3 electrons instead of 2 for Fe(II) and consequently, a more important current level for attaining the equal Fe amount is necessitated [5,81]. The several monomeric and polymeric compounds ultimately convert into amorphous $Fe(OH)_3$ flocss that are performant in quick adsorption of dissolved organic matters and enmeshing of colloids [85,86].

Moreover, there are two additional dissimilarities with aluminum electrodes: (1) the buffering impact mentioned for iron is less significant than for aluminum. Ultimate pH frequently attained is ~9-10 with iron electrodes even though pH is acid [84,87]. (2) Fe^{2+} is greatly dissolvable and thus not able of a performant colloidal particles destabilization upon $Fe(OH)_3$, consequently inducing low EC efficiency [42]. As a result, a performant action of Fe-EC necessitates one or more of the next optimization methods for the Fe^{3+} formation [5,84]:

- Aerating the water to augment the solubilized O_2 amount and Fe^{2+} oxidation;
- Augmenting the pH level to 7.5 or bigger to encourage the Fe^{2+} oxidation velocity;

Adding oxidant like Cl_2 that may be formed upon oxidization of the Cl^- existent in the water on the Fe anode [5]. Thus, ferrous oxidation happens in the bulk solution, following:

$$Cl_2 + H_2O \rightarrow ClOH + Cl^- + H^+ \qquad (24)$$

$$2Fe^{2+} + 2HOCl \rightarrow 2Fe^{3+} + 2OH^- + Cl_2 \qquad (25)$$

Considering the electric current consumed through Cl^- oxidation, this method is not performant unless the water/wastewater to purify holds more than 600 mg Cl^-/L.

- Elevating the operation period to attain total ferrous oxidation.

Fe shows as well two extra benefits than Al: Fe is not poisonous; therefore, it may be employed for drinking water. The second one is the smaller cost of Fe, about 0.5-0.8 US$/kg, while Al cost is comprised between 1.5 and 3 US$/kg [5].

Fundamental Factors Affecting the EC Efficiency

The factors influencing EC performance are linked to (1) the working situations like current or voltage and residence period, to (2) water characteristics like pH, alkalinity and conductivity and (3) configuration of the EC device and its electrodes (electrode surface area, electrode inter-distance) [5].

Impact of the electric current

The electric current *I* is the most important parameter of EC. Practically, EC depends on the current density i that is described as the ratio of electric current over electrode surface area S. The continuity equation implies current conservation amongst the anode and the cathode; in fact, the current density may vary amongst the electrodes [5]:

$$I = i_A \times S_A = i_C \times S_C \qquad (26)$$

Current density dictates the coagulant injection from the anode and the hydrogen gas (H2) production from the cathode following the Faraday's law [5]. The bubble intensity influences the device hydrodynamics, which in turn affects mass transfer among contaminants, coagulant and gas micro-bubbles, and ultimately imposes collision significance of destabilized colloids and dissolved matters that conducts to flocs production [88]. Moreover, current density influences metal species during pH variation through EC operation in the form of a function of water alkalinity. As a result, the current seems to form a dynamic physical/chemical medium that dictates the coagulation/flocculation stages [89] and encourages the electromigration of ions and charged colloidal particles [18].

Cell voltage is linked in the form of equilibrium potential, anode and cathode overpotential as described through Eq. (35). Thus, electric energy consumption may be formulated in the form of contact period t employing [5]:

$$P = \int_0^t U \times I \times dt \qquad (27)$$

Since the electric energy needed for the EC method is related to the electric current and potential as described in Equation (27), EC may be performed either upon the (1) galvanostatic or (2) potentiostatic mode. For the first one, EC technique is realized through monitoring and/or modifying the current imposed over electrodes; at the same time, for the second one, it is the imposed cell voltage that is monitored and/or changed in the form of quantity of coagulant needed to be liberated in the EC device [5]. The second mode is not frequently employed for EC [90] and usually used for different electrochemical techniques like electro-oxidation and electro-reduction where sacrificial anodes are not employed [91,92].

Nevertheless, too high current amounts cam badly influences the EC performance. As an example, additional reactions can happen mainly, and overdosing may reverse the charge of the colloidal particles and disperse them another time conducting consequently to a diminution of the coagulant performance and a decrease of the anode lifetime [5].

The levels of the current density may largely change following the characteristics and the concentration of contaminants to be eliminated from water; as an example, from 0.01 A/m^2 to 880 A/m^2. Optimum current density must be evaluated following additional working indicators. To employ the EC reactor during a large time without stopping for maintenance, the current density is proposed to be among 20 and 25 A/m^2 [77]. Moreover, high current augments voltage and ohmic drop among anode and cathode. Ohmic drop or IR drop is a consequence of the ohmic resistance of the electrolyte R, which may be described as follows [5]:

$$R = \frac{d}{S} \times \frac{1}{K} \qquad (28)$$

here *d* designates the inter-electrode gap, and k the water electric conductivity. If *i* augments, and *U* has tendency to the *IR* drop term, RI derived from Equation (28), which significates that electric power changes as RI^2. Therefore, power input may be diminished upon reducing the gap among the plaques and augmenting the electrode surface area and the water conductivity [66]. Employing current reversal (switching anode and cathode electrically) is helpful to decrease maintenance price; however, its impact on contamination elimination is not ascertained until this moment.

Impact of the water pH

Besides the electric current, pH is an additional fundamental parameter touching the EC efficiency, specifically coagulating stages because it dictates the hydrolyzed metal species produced in reactive solution and affects the active steps of EC [82]. As shown in Section 2, investigating the Al and Fe speciation as a result from hydrolysis of their corresponding cations dictated upon thermodynamic equilibrium is crucial to explain the manner by which pH participates in imposing the stages of the electrochemical coagulation. There is no doubt that adsorption and coagulation are importantly depending on pH. The superficial charge of the Al or Fe flocs may be interpreted upon the adsorption of the charged soluble monomeric species on their respective hydroxide flocs [81]. The behavior among the pH-dependent coagulant species and their near contaminants may be determined from electrostatic interactions. Researchers such as Jiménez et al. [81] mentioned a detailed description taking into account these stages, i.e., double-layer compression, neutralization and sweep flocculation, for Al and Fe-EC intending to optimize the removal of several contaminants depending on the predominating hydrolyzed metal species. The significance of pH on EC efficiency may be at the same degree understood throughout the thermodynamics linked to the electrochemistry as described upon the Nernst equation. This relationship permits us showing the E-pH diagram of a useful electrode material that, once superimposed on E-pH diagram of water, conducts to a diagram well described as Pourbaix diagram. The Pourbaix diagram illustrates the areas of thermodynamically stable metal species in the aqueous medium, i.e., immunity, passivation, and corrosion, which lets to predict the corresponding electrode stability and its dissolution behavior in water throughout describing the stable aqueous species in a specific region of electrochemical potential and pH [5].

Researchers [51] examined the Al and Fe speciation intending to determine the predominance diagrams of corresponding hydroxides and to evaluate a fraction of undissolved hydroxides as a function of pH taking into account only monomeric species. For Al electrode, it has been established that the quantity of undissolved $Al(OH)_3$ augments importantly with elevating pH from 4.5 to 7 to the detriment of aluminum hydroxide ions and the reverse is correct for a pH from 7 to 10; at the same time, amorphous metal hydroxide is not detected above the latter pH value. For the Fe electrode, the amount of undissolved iron hydroxide significantly augments with pH varying from 4 to 7 [93]. At pH 7, iron hydroxide ions are not present in the predominance diagram. These predominance diagrams were realized from the theoretical computation founded on equilibrium constants and pH for a concentration of 10^{-2} M for both electrode metals [5].

It should be mentioned that solution pH after EC operation would augment for acidic pH; however, it may diminish for alkaline pH, which is attributed to the buffering impact of EC [94]. The augmentation of pH in acidic water is explained by hydrogen production at the cathode; while the diminution of pH is linked firstly to the apparition of hydroxide precipitates that liberate H^+ protons near the anode and the additional reactions like water oxidation and chlorine generation and its hydrolysis [95]. This fact focuses on the buffering impact of EC that enters in action in addition to that of water alkalinity [5]. This impact is significantly elevated with Al electrodes due to the generation of aluminate anions at elevated pH [39].

It was mentioned that the bicarbonate alkalinity ameliorates at a small degree the contaminants elimination performance [96]; moreover, it aids to reduce the hardness throughout precipitation of $CaCO_3$ due to the hydroxyl anions formed upon water reduction near the cathode [5,46].

Impact of cell geometry and electrodes conception

The EC apparatus is mostly constituted of electrodes and enclosure. The electrodes are placed in a ring that is a non-conductive recipient in which the treatment of water happens [5].

Electrodes arrangement: The electrode system primarily influences EC method through electrodes arrangement and inter-electrode gap. Electrodes arrangement may either be easily constituted of an anode and a cathode or be formed of several anodes and cathodes complicatedly arranged in EC tank. The complicated electrodes settlement may be categorized in monopolar and bipolar electrodes (Figure 5) [5]. Table 6 summarizes a description of the different configurations of the electrodes.

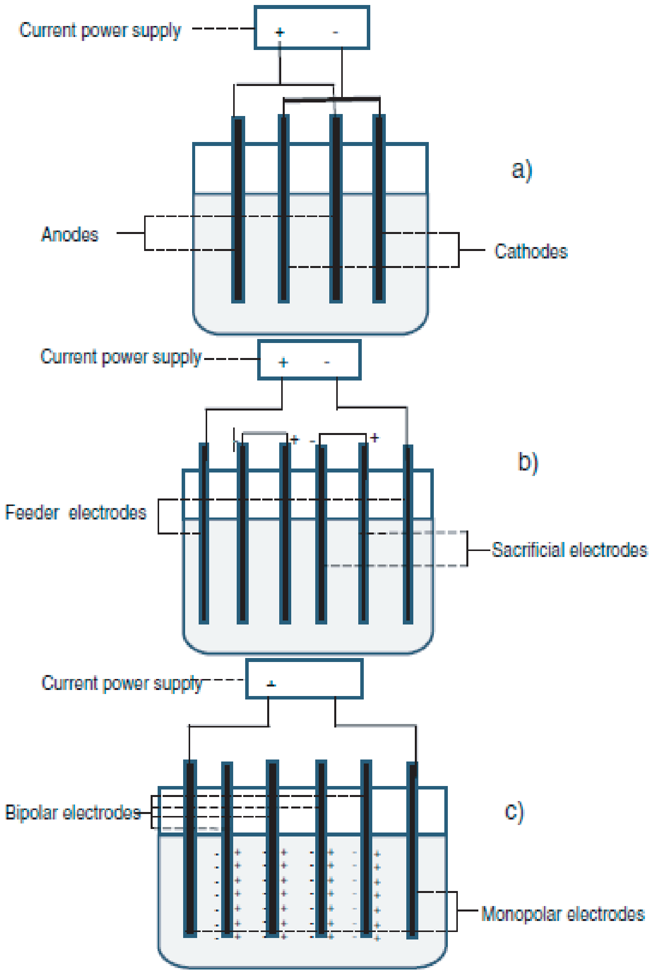

Figure 5: Monopolar electrodes: (a) in parallel connections, (b) in series connections; (c) bipolar electrodes in series connections [5].

In a general manner, monopolar electrodes necessitate a low voltage and a more significant current contrary to the bipolar electrodes that work under high tension and a smaller current [5]. It is not easy to affirm which electrodes settlement is more significant than the other taking into account solely EC performance considering that it has been established that at the same level BP-S may show an elevated EC performance [97,98]. Monopolar electrodes can be viewed as significant because in several situations this electrodes arrangement provides an elevated contaminant reduction with a lower energy consumption [99,100], taking into account that bipolar electrode frequently consumes high energy [101-103]. The previous configuration is not difficult to manipulate and requires less maintenance cost throughout function; therefore, the effect of maintenance cost on global operation cost should at the same level be taken into account to

Electrodes configuration	Description
Monopolar electrodes in parallel connection (MP-P)	MP-P are illustrated in Figure. 6a. The electrode settlement is composed of cathodes and anodes arranged alternatively at the same anodic or cathodic potential, respectively. Each pair of cathode/anode corresponds to a small electrolytic cell in which the voltage is the same. Thus, the reactor is composed of electrolytic cells in parallel. Therefore, the current of each electrolytic cell is additive.
Monopolar electrodes in series connections (MP-S)	MP-S are defined in Figure. 6b. Each pair of internal sacrificial electrodes is internally linked with each other and has no interconnections with the two outer electrodes. The electric current traveling through all the electrodes is the same, whereas the global voltage is the sum of tension in each electrolytic cell.
Bipolar electrodes in series connections (BP-S)	BP-S comprise two outer electrodes attached to the electric power supply and the sacrificial electrodes fixed among the two external electrodes (Figure. 6c). Outer electrodes are monopolar, and the inner ones are bipolar. The bipolar electrodes are not interconnected, and each of their sides acts simultaneously as an anode and a cathode. This fact implies that the opposite sides of each bipolar electrode are oppositely charged, the anodic dissolution happens on the positive side; at the same time, the negative side is prone to cathodic reactions [28].

Table 6: Various electrodes configurations used in EC process [5].

select a convenient electrode configuration [37].

More than the typical rectangular electrodes, there are additional geometrical forms like circular and cylindrical. Electrodes may be placed either vertically or horizontally in EC cell [5]. Even if being scarcely employed, horizontal electrodes in EC batch reactor may possess a more significant mixing [104] performance [105].

Inter-electrode gap: It is well-known that if the IR-drop augments, the difference among electrodes increases. This fact means that energy consumption diminishes with reducing the distance among the electrodes (Equations (27) and (28)) [5]. As the separation among the electrodes becomes lower, more electrochemically produced gas bubbles bring about turbulent hydrodynamics. This fact conducts to a high-mass transfer as well as to a high reaction rate amongst the coagulant species and contaminants [106]. Moreover, inter-electrode distance describes the contact period between the anode and the cathode for a continuous system and the period of operation for a batch reactor for attaining a wanted EC performance. For a complicated electrode placement, inter-electrode separation dictates as well the number of electrodes to arrange in EC cell, if its volume is fixed [107]. On the other hand, EC reactor conception is crucial because it influences the global efficiency of the EC method in the matter of its impact on the working variables, i.e., flow regime, flocs production, reduction percentage, and flotation/settling features [5,74].

An in-depth discussion of the EC reactor conception may be found in the excellent review of Hakizimana et al. [5].

Influence of water conductivity

The current density contribution is a function of conductivity and ionic strength of the solution. The current density effect augments if the electrolytic conductivity is elevated thanks to the reduction of the ohmic resistance of water. Moreover, the electric conductivity contributes to reducing the residence period needed to attain the desired reduction percentage [108]. As a result, energy consumption (*UI*) is decreased. Usually, NaCl is employed to augment the electrolytic conductivity. In addition, Cl- contributes to the decrease of the negative impacts of different anions to reduce the precipitation of calcium carbonate in hard water that may constitute an insulating film on the electrodes surface [94]. If the current density is elevated, Cl- may as well be oxidized to active chlorine forms, like hypochlorite anions, that may oxidize organic matters [37] and ferrous ions [5] or participate in killing microorganisms in water [109-112]. To providing an efficient function of EC in treating water, it is suggested that 20% of the anion's existent must be Cl- [77].

Nevertheless, there are the limits dictated on conductivity augmentation in drinking water treatment [113]. Indeed, conductivity augmentation throughout the treatment of potable water using EC is significantly restricted in following the standard norms that describe the maximum chloride level in industrial effluents at 250 mg/L [5].

Influence of temperature

Temperature is an important parameter affecting EC process. Usually, EC process is realized at ambient temperature. During EC operation, the water temperature may increase due to the

Joule effect. This phenomenon is more pronounced if the water conductivity is elevated especially in the case of brackish water or seawater treatment [49,109]. In this case, some precautions should be taken such as reducing the applied voltage and decreasing the residence time to avoid overheating of the electric components and circuit [80].

EC Modeling

At the best of our knowledge, the consulted references through this review proved the absence of an algorithmic and direct method of EC reactor conception. The significant enhancements in EC device efficiency and function require better design, materials, and optimization. These need a fundamental comprehension of the respective contributions of several phenomena implied in EC, comprising electrochemical mechanisms, coagulation, flotation, and settling [66]. EC modeling is importantly useful to enhance the conception and decrease equipment and operating costs simultaneously. It may easily ensure viable and correct solutions to EC issues, and therefore let us evaluate EC setup efficiency under a considerable interval of working situations [5].

Hakizimana et al. [5] discussed the EC models previously mentioned and examined in the specific references, and that may assist in reaching more comprehensions in EC devices conception. In a general manner, there are two principal categories of EC modeling: statistical modeling and modeling funded on knowledge. Mathematical modeling usually is destined to searching optimum working parameters in which EC performance will be enhanced. Since EC is a complicated method, modeling founded on knowledge embraces specific models that adopt EC as one method and some others that are employed to define a given physical or chemical process happening throughout the technique. Moreover, a particular focus is given to the Computational Fluid Dynamics (CFD) modeling; for which computational

Modeling	**Description**
Statistical modeling	Taking into account several physical/chemical processes implicated in EC, the contaminants elimination performance complicatedly is the function of the separated and additional impacts of the key technique parameters (factors). Until now, in the largest part of the investigations performed on water/wastewater treatment using EC, optimization has been reached through modifying a single parameter at the same time maintaining all remaining parameters set at a particular series of situations. This usual fashion for optimization of EC process needs several practice tests and conducts to a weak optimization, like underestimation or overestimation of the impact of the method factors on EC efficiencies because of neglecting the mutual contributions inside those parameters [114]. Response surface methodology (RSM) has been employed as a method in many studies to show the impacts of main process factors and their mutual contributions. RSM is used in multiple forms such as the full or partial factorial design (FD) [115,116], central composite design (CCD) [117-121], D-optimal design (DOP) [114], and Box-Behnken design (BBD) [122-125], etc.
Modeling funded on knowledge	*a) Phenomenological models* In several studies, EC kinetics has been investigated to model and simulate the EC process. Several researchers such as Mameri et al. [78] have tried to model EC kinetics of the removal of fluoride anions. Kinetics of defluoridation was shown to obey an exponential law with time so that it turned out to be first-order for fluoride concentration. Moreover, the elimination of contaminants like nitrates [54,126] and heavy metals by EC obeyed to an n-order kinetic model [127]. *b) Modeling detailed mechanisms* *1. Electrochemical phenomena* EC technique is mainly founded on electrochemistry since the electrochemical events are the initiating heart of the whole method. Electrochemistry is viewed as a complicated knowledge since it concerns at the same time charge transport, electrochemical kinetics, comprehension of electrodes interface and thermodynamics. *2. Adsorption* To better assess the stages implicated in EC and for modeling aim, pure adsorption isotherm and adsorption kinetics models have mainly been employed. Because the quantity of coagulant formed may be evaluated for a specific period by applying Faraday's law, contamination reduction may be seen from the adsorption phenomenon point of view [128]. The most common adsorption isotherms employed for EC modeling founded on the hypothesis of thermodynamic monitoring are mainly the Langmuir and Freundlich isotherms and the Langmuir–Freundlich model that uses the two previous ones. The Langmuir isotherm supposes a monolayer deposition of the adsorbate on a homogenous adsorbent surface, while the Freundlich isotherm defines reversible adsorption that may be followed by multilayer formation [129].

	3. Flocculation modeling Flocculation is a complicated method during which two or more colliding destabilized colloids adhere together and constitute a floc. Stages happening throughout flocculation may be summarized into two steps: transport conducting to the collision and fixation. The first one is realized through (a) Brownian motion of the colloids (perikinetic flocculation), (b) fluid motion (orthokinetic flocculation) and (c) differential settling velocities attributed to gravity. Fixation is a consequence of the interparticle forces [130]. *4. Flotation and settling* The formed flocs during EC may be eliminated upon various physical manners, as illustrated in Figure 4. EF appears thanks to buoyancy forces and settling thanks to gravity. EF is a function of current density, hydrogen micro-bubbles size (20-50 µm) and particle collection performance by the micro-bubbles [24]. Even if the current density imposes a convenient contaminant reduction mechanism, particularly in batch systems, it is not easy to entirely escape to either settling or EF in favor of the remaining stages, whatever the current density that may be employed. *5. Complexation* Complexation model is a modern phenomenological model illustrating adsorption equilibrium that is imposed by complexation of suspended matter upon Fe or Al hydroxides for chemical oxygen demand removal.
Modeling utilizing CFD	Recently, like in several research domains, CFD simulations have reached a significant role to play in the comprehension and enhancement of the EC reactors design for the next future. Many authors have focused on different features of flows inside electrochemical cells [131-134]. CFD seems to be useful and exciting in comprehending the hydrodynamic and contact period distribution to describe the flow pattern and may be successfully used to predict the principal local characteristics of the method, such as velocity profiles, the reaction rate distribution at the electrodes and the cell voltage. CFD has been employed to examine the distribution of potential and current density because the latter shows the distribution of attack on electrode surface throughout its dissolution and may be helpful in the EC reactor conception for energy efficiency [131].

Table 7: EC process statistical modeling and modeling funded on knowledge [5].

techniques have been used to investigate the fluid flow and current density inside EC devices and predict complicated inherent processes, particularly if technical restrictions limit an experimental method.

Table 7 summarizes the main features of the statistical modeling and modeling funded on knowledge of the EC process [5].

Conclusions

From this review, the main points drawn are listed as below:

1. Similar experiments, as jar test experiments used in conventional coagulation to determine the optimal coagulant doses, should be conceived using Zeta-meters to control the EC optimal residence time better and metal amount liberated. An empirical method would be suggested to facilitate a direct approach to calculate the EC optimal parameters following the (1) water main characteristics such as electric conductivity, pH, pollutants concentrations and the (2) EC device features like metal type, reactor geometry and batch/continuous mode.

2. The scientific community specialized in the EC process is near to suggest empirical/theoretical models to present the EC technology as a viable green process. However, more great efforts remain to be accomplished.

3. Technological software developers such as COMSOL™ Multiphysics are invited to insert the EC process in their electrochemistry module in their future versions to better commercialize this intensified technique and encourage its massive use through the world.

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