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Electrocoagulation as a Pioneering Separation Technology—Electric Field Role

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Abstract

Electrocoagulation (EC) is a very efficient process in dealing with effluent streams and separating complicated contaminants prior to the discharge of the treated water. Attention to such a technique augmented thanks to its large set of utilizations, zero—or minimal—chemical dosing demands, low waste formation, and low price. EC appears as an efficacious option to traditional water treatment techniques for the separation of a large collection of contaminants. This work examines the theories of the EC method and its application for the separation of contaminants from wastewater streams. Such a technique depends on the integration of electrochemical and coagulation methods. Basic parameters that touch the effectiveness comprise the electrode material (Fe or Al), current density, the electrical charge per unit volume, and solution pH. Electrode fouling could constitute a hard running dare even if it could be reduced by the alternating current operation. Next studies have to follow the routes of the EC technique for numerous kinds of pollutants at a set of working parameters, in particular for continuous mode, and the expansion of convenient models that could be utilized for scale-up and techno-economic evaluation of EC is required. Running as a destabilization agent and aiding to separate contaminants from the wastewater, the electric field should attract more attention to highlight its key contribution.

Subject Areas

Chemical Engineering & Technology

Keywords

Electrocoagulation (EC), Wastewater Treatment, Separation Technology, Iron, Aluminum, Electric Field

1. Introduction

Global investment in techniques and consumer demand for energy, goods, and natural resources conducted to the growing diffusion of contaminants in nature [1] [2] [3]. Numerous ecological programs focused on controlling and decreasing pollution [4] [5] [6]. Following their poisoning, contaminants have been classified with those that are highly threatening to nature known as "Black List" contaminants [1] [7] [8]. Black List contaminants are so dangerous, persistent, or bioaccumulative in nature, so procedures should be followed to remove their pollution [9] [10] [11]. The list of Black List pollutants persists to increase and comprises primarily persistent organic pollutants (POPs) (like organohalogen, organophosphorus), and toxic metals and their organometallic compounds [12] [13] [14]. Treatment of effluents carrying POPs remains hard since they provoke hurdles to conventional biological treatment plants which are resilient to biological digestion, bioaccumulate, and persist in nature [15] [16] [17].

The waste management hierarchy has to prefer the decrease in usage of POPs or institute regulations to prohibit their employment [18] [19] [20]. The following stage in the management hierarchy should also be adopted (that is to say, elimination, minimization, reuse, recycling, recovering, and, if possible, safe quarantine or disposal) [21] [22] [23]. Consequently, to guarantee the implementation of sustainability, it remains vital to decrease waste without periling human health, employing practical technologies that induce no damage to nature [24] [25] [26]. As a rule, environmental techniques employed different properties of physical [27] [28] [29] [30], biological [31] [32] [33], photolytic [34], chemical [35] [36] [37], and physicochemical methods [38] [39] [40] for pollution remediation [41] [42] [43]. Nevertheless, regulatory limits dare the techniques to stay cost-effective and reach an elevated degree of pollution reduction while dealing with complicated systems, comprising the occurrence of mixed contaminants, watery degrees of pollutants (e.g., micropollutants), and the exigency to recuperate contaminants (like metal ions) in a reusable form [44] [45] [46]. Among treatment processes, electrolytic techniques, regarded as physiochemical ones, could deal with such dares, allow ecologically benign treatment, and frequently give an easy solution to pollution troubles linked to industrial effluents [12] [47] [48]. Lately, some global guidance on the usage of electrochemical techniques for efficacious ecological treatment, monitoring, energy conversion, and pollutant separation has been furnished [12] [49] [50]. Such a guidance underlined the significance of external control of electron exchange as the major ecologically friendly reagent in the electrochemical treatment techniques [1] [51] [52]. This work defines the usage of electricity to treat water by means of accepted electrocoagulation (EC) technique and the merit of employing the method for performant water recuperation and pollutant separation.

The phenomenon of selective separation has a crucial contribution to the chemical and biochemical industries [53] [54] [55]. Indeed, the phenomenon of separation participates in about 40% to 70% of the total capital and operating

expenses [56] [57] [58]. Nevertheless, taking into account the techniques of ecological treatment and water purification, where the pollutants are often existing at very small levels, the dare is to eliminate pollutant species in a cost-effective and environmentally sustainable procedure [1] [59] [60]. The troubles related to contaminant separation are usually because of a small level in effluent (related to the low mass transfer of the dilute pollutants), the steric effects of complexing species, and the occurrence of a mixture of different end-of-pipe products that stop the phenomenon of continuous recuperation and separation [12] [61] [62].

Employing EC for ecological treatment involves the elimination and separation of pollutants via catching them within a precisely selected coagulant to finally facilitate water reuse and recycling [63] [64] [65]. Even if the technology has been accepted, a selective capturing of a pollutant for elimination and separation furnishes a novel procedure to allow a performant treatment technique [1] [66] [67].

This work examines the EC background. A special focus is accorded to EC as a manner for separating contaminants. An apercu is dedicated to the chemical theories and electrochemical reactions. Features affecting the EC process and its mean fields of usage are discussed.

2. Electrocoagulation (EC) Background

The theory of EC has long been mentioned [68] [69] [70]. Nevertheless, the practical employment of the setup for ecological treatment and separation has only lately acquired attention [1] [71] [72]. The earliest account to refer to the use of the EC process to separate water from sewage emerged in 1889 in England [73] [74] [75]. A 1909 patent filed in the US was the first to mention the probable capture of contaminants from wastewater by the manner of an electrochemical cell employing sacrificial aluminum (Al) and iron (Fe) electrodes [76] [77] [78]. Nevertheless, the first full-scale utilization of EC for water treatment was noted in 1946 [61] [79] [80]. In such configuration, the electrogenerated flocs were discovered to be efficacious in decoloring water [81] [82] [83]. An identical device, employed in England in 1956, injected Fe electrolytically into river water to separate turbidity and color [84] [85] [86].

At a certain level, the EC technique was restricted to some usages; nevertheless, it has attracted universal attention during the last three decades [1] [87] [88]. Even if the technique is more than 100 years old, there was little interest in academic literature until around the 1980s [89] [90] [91]. The resumed concern in EC for separating pollutants from wastewater has happened worldwide thanks to its wide applicability, low sludge formation, scalability, running at ambient temperature and pressure, simple design and ease of control, and low capital and operating costs [92] [93] [94]. Further, the capability to change the applied current to improve the treatment performance facilitates process automation and control [95] [96] [97].

EC is a method that employs the theories of chemical coagulation and elec-

trochemistry to treat and separate pollutants from wastewater [1] [61] [65]. Conventional coagulation implicates the injection of chemical products to react, aggregate, and separate pollutants via the generation of large flocs that could be physically separated from the mixture [38] [53] [98]. The word traditional coagulation is utilized inconsistently with the word flocculation to explain the phenomenon related to colloidal separation from effluents [38] [99] [100]; nevertheless, the two words differ [101] [102] [103]. Coagulation implies the aggregation of colloids because of the reduction of electrostatic repulsive forces between the colloids in the suspension [104] [105] [106]. Flocculation includes the production of chemical bonds between the particles to enmesh them in relatively large masses called floc networks [107] [108] [109]. In EC, nonetheless, sacrificial metal anodes are used to dose the contaminated water with a coagulating agent, like Al or Fe ions [30] [63] [110]. Therefore, coagulation, flocculation, and electrochemical processes take place together (**Figure 1**) [1] [111] [112].

In EC technology, applying an adequate voltage to the sacrificial anodes that are composed of the coagulant species (*i.e.*, Fe or Al) will permit their oxidation at the electrode/solution interface (**Figure 1**). Further, the dissolution of the coagulant species into the electrolyte lets them interact with each other and with the suspended matters or dissolved pollutants that can be more separated from the suspension by sweep coagulation [38] [107] [113]. Therefore, the EC technique inserts coagulant *in situ* rather than via external injection [43] [61] [67]. Employing such a tool, a specific separation of pollutants could be possible with careful control of the method, and a suitable selection of dissolved coagulant [1] [114] [115]. Such phenomena take place in both sides of the cell (*i.e.*, anode and cathode); thus, the coagulation, flocculation, and gas evolution will happen together and participate in the global treatment technology [52] [58] [68]. As an illustration, in addition to the enhanced coagulation due to the reduced electrostatic repulsion between aggregating particles, in the EC process, the electrical

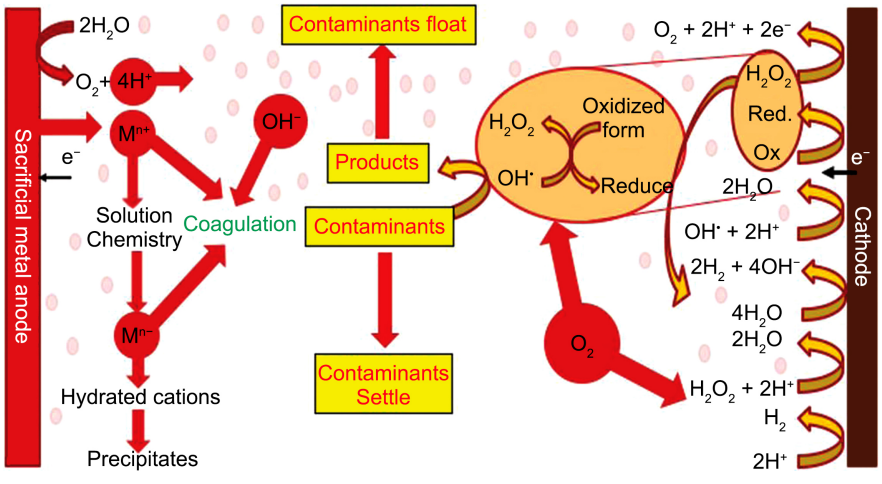

Figure 1. A schematic illustration of the major reactions happening during the EC method. In some conditions, redox reactions may occur directly on the electrode surface or in the bulk electrolyte [1].

field ameliorates this technique via augmenting the migration of ions through the electrophoresis impact and the charge redistribution on the pollutants [47] [58] [63]. The electrolysis of H_2O at the cathode produces hydroxyl ions (OH^-) that combine with the dissolved metal coagulants (Al^{3+} or $Fe^{2+/3+}$) from the anode to generate the corresponding hydroxide (or oxide) precipitate at a convenient pH, eliminating contaminants via sweep coagulation [38] [47] [55]. Further, the evolution of gases due to water electrolysis (usually O_2 and H_2) takes place at both electrodes at sufficient overpotentials that could raise floatation of some fractions of the coagulated contaminants to the surface (**Figure 1**) [58] [72]. The agglomerated species possess the potential to adsorb other species [58]. Such adsorption phenomenon could be improved by the applied potential between the electrodes that let separation and removal phenomena to happen along with the majority of the suspended matters [61]. Like in any electrochemical setup, additional responses happen jointly with the coagulation reactions, comprising cathodic reduction of reducible particles, anodic oxidation, and mass transfer of ions in solution, which adds some complexity to the technology [13] [52] [93]. It is evident that EC is a complex synergistic method with numerous reactions and pathways happening together to eliminate contaminants [58] [63] [68].

3. Electrocoagulation (EC) for Separating Contaminants

Since the early application of EC to deal with wastewater in the eighteenth century, the technique has been adopted as a separation method to separate water from sludge [1] [61] [63]. Founded on ecological and physicochemical concepts, the EC method could be viewed as one of the most efficient techniques for contaminant separation [64]. From an ecological point of view, an EC remediation of effluent is frequently applied to eliminate a mixture of contaminants from water at a level that it could be safely discharged. Taking into account the theories of chemical separation, EC implies a phase-transformation process (**Figure 1**), where the phase of dissolved contaminants is modified during elimination (from the liquid- to solid-phase) [58] [63] [68]. Identical to several chemical separation techniques, extracting contaminants by agglomeration inside the coagulated flocs (metal hydroxide) happens, in many situations, without changing the chemical or biological properties of the original contaminants [69]. In such a situation, the separated contaminants are aggregated inside the coagulants with no big modification of their chemical structure [30]. Agglomerating species renders the recuperation of such chemicals easy by extraction from the recuperated solids. Such a separation phenomenon is greatly significant when treating high-value chemicals for separation or recuperation. Oxidation or reduction reactions could happen at the anode or cathode surfaces, conducting to chemical alterations in some organic pollutants and could render recuperation of the original species more demanding or inappropriate [74].

In the absence of oxidation or reduction of the pollutant, EC could be adopted as a physicochemical separation technique [58] [116]. Numerous in-

vestigations have established the EC technique for the concurrent separation of several contaminants from a set of diverse wastewater streams [68]. Illustrations of waste water that have been treated via EC for contaminant separation are well-documented [1]. Pollutants that have been separated comprise emulsified oil, nanoparticles, antimicrobial chemicals (Bronopol), microalgae, heavy metals, organometallic mixtures, nuclear fission products, and heavy metals leachates [1]. As a rule, EC applies the theory of phase separation to eliminate contaminants and treat water [116].

Extracting contaminants by coagulation is founded on the physicochemical features of both coagulate and coagulant. The physicochemical conduct is affected by both chemical interactions between the coagulant and coagulate (like coprecipitation, solid-solution interface, and complexation) and electrostatic interactions (that is to say surface charge and colloidal destabilization). In EC, the coagulant arrives directly from the selection of the anode material and should have a high affinity for coagulating the target pollutants. Dissolved silica has a very high affinity to generate very stable coordination with Al cations. The phenomenon is largely employed in the coagulation technique to separate silica as aluminosilicates [117]. Researchers [1] proved a selective separation of Si from a petroleum process effluent via a chemical coagulation technique employing alum as a coagulant. Using the equivalent method of EC employing Al electrodes has also been illustrated to selectively retain silica from treated water from the oil and gas industry [1].

4. Chemical Theories and Electrochemical Reactions

Theoretically, during the electrolysis of sacrificial metal anodes, the solubilized metal ions at the anode have a tendency to hydrate, particularly those with a charge of +3 or more tend to donate hydronium (H_3O^+) cation from the surrounding hydration species (Brönsted acids) [1]. Equations (1)-(3) show the acidic reaction for Fe^{3+} cation and Equations (4)-(6) for Al^{3+} cations along with their equilibrium constants in aqueous media [118]. Some of such species show amphiprotic conduct, as they are acids when they appear on the left side of the equilibria and bases on the right side [1].

$$Fe\left(H_2O\right)_{6(aq)}^{3+} + H_2O \leftrightarrow Fe\left(H_2O\right)_5\left(OH\right)_{(aq)}^{2+} + H_3O^+ pK = 2.187 \quad (1)$$

$$Fe\left(H_2O\right)_{6(aq)}^{3+} + 2H_2O \leftrightarrow Fe\left(H_2O\right)_4\left(OH\right)_{2(aq)}^{+} + 2H_3O^+ pK = 4.594 \quad (2)$$

$$Fe\left(H_2O\right)_{6(aq)}^{3+} + 3H_2O \leftrightarrow Fe\left(H_2O\right)_3\left(OH\right)_{3(aq)} + 3H_3O^+ pK = 12.56 \quad (3)$$

$$Al\left(H_2O\right)_{6(aq)}^{3+} + H_2O \leftrightarrow Al\left(H_2O\right)_5\left(OH\right)_{(aq)}^{2+} + H_3O^+ pK = 4.997 \quad (4)$$

$$Al\left(H_2O\right)_{6(aq)}^{3+} + 2H_2O \leftrightarrow Al\left(H_2O\right)_4\left(OH\right)_{2(aq)}^{+} + 2H_3O^+ pK = 10.094 \quad (5)$$

$$Al\left(H_2O\right)_{6(aq)}^{3+} + 3H_2O \leftrightarrow Al\left(H_2O\right)_3\left(OH\right)_{3(aq)} + 3H_3O^+ pK = 16.791 \quad (6)$$

In coupling with the metal ions, the occurrence of OH^- could work as a bridging group to join two or more metal hydroxides jointly (**Figure 2(a)** and **Figure 2(b)** for Fe and Al, respectively), allowing more dimerization or polymerization (Equations (8) and (9)). The produced species of hydroxyl bridges are most probable to carry positive charges that can donate another hydrogen ion to provide OH^- anions and bond more with additional metals to produce polymeric hydrolytic species (gelatinous hydroxide) [1]. Polymerizing gelatinous hydroxide can further carry suspended solids or dissolved matters with it as it grows and settles (sweep coagulation) [7] [38] [53]. The capability and the specificity of the pollutants' aggregation could change following the physicochemical properties of the produced gelatinous hydroxides, surrounding media, and the type of the suspended solids or solubilized pollutants in the solution [46] [61] [98]. Key properties of the generated gelatinous hydroxides comprise electrical charge, porosity, and types of bonding produced either in the hydroxide or with the pollutants.

$$2\left(\mathrm{Fe}\left(\mathrm{H_2O}\right)_5\left(\mathrm{OH}\right)\right)^{2+} \rightarrow \left(\mathrm{Fe_2}\left(\mathrm{H_2O}\right)_8\left(\mathrm{OH}\right)_2\right)^{4+} + 2\mathrm{H_2O} \quad (7)$$

$$2\left(\mathrm{Al}\left(\mathrm{H_2O}\right)_5\left(\mathrm{OH}\right)\right)^{2+} \rightarrow \left(\mathrm{Al_2}\left(\mathrm{H_2O}\right)_8\left(\mathrm{OH}\right)_2\right)^{4+} + 2\mathrm{H_2O} \quad (8)$$

In the coagulation chemistry, the conduct of hydrated metal ions to work as the acid can greatly touch the separation and treatment methods of different metal elements from effluents [1] [119]. The acidity of solubilized metal ions augments with the charge and decreases with the radius of the ionized species [120]. Hydrated metal species, other than Fe and Al, can likewise tend to donate protons, produce bridging compounds, and finally, work as a coagulant when polymerization happens [121] [122] [123]. Metal species that have been mentioned as coagulants comprise Be(II), Bi(III), Ce(IV), Co(III), Cu(II), Ga(III), Mo(V), Pb(II), Sc(II), Sn(IV), and U(VI) [1]. Even if the favored anode materials in commercial EC devices remain Fe and Al and if one or a mixture of the other metal species mentioned above are available in the effluent, they will coagulate and separate out of the solution. Such a phenomenon has been exploited in the EC technique, where either Al or Fe anodes have been utilized to separate and treat heavy metals from effluents [124]. Employing Al and Fe (or in some situations Zn [125]) in EC for industrial implementations is affected to their low

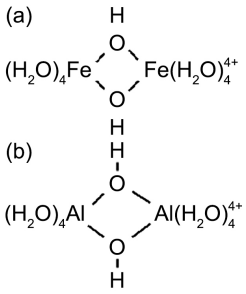

Figure 2. (a) Structure of the ferric hydroxide dimer. (b) Structure of the aluminum hydroxide dimer [1].

price and availability, large efficient pH span (4 - 11) for insoluble hydroxides generation, and low environmental footprint (especially for Fe [5] [75]).

5. Features Influencing Electrocoagulation (EC) Technique

Variables that touch the coagulation method could identically affect the EC phenomena [126]. In EC where electrochemical reactions are utilized to inject coagulants, the metal complexation is a function of the pH circumstances and the kind of the sacrificial anode employed [1] [63] [64]. The electric field can run as a destabilization agent, aiding to separate contaminants from the wastewater [47]. During electrolysis phenomena, the formation of hydronium and hydroxide ions work to enhance a pH difference between the oppositely polarized electrodes inside the reactor. Such a change in pH inside the electrochemical setup is a function of the applied current, voltage, supporting electrolyte, and time of treatment [57]. Consequently, pH change greatly affects the level of separation and treatment. In this context, numerous practical factors could touch the efficiency of the EC device. Nevertheless, the main elements could be classified into four interconnected groups that are related to the electrode setup (material, design, and electrode spacing), running circumstances (current density, operating period, over potential), the device design, the characteristics of the effluent treated (pH, conductivity, temperature, turbidity).

Such elements are interrelated and their contributions to the EC technology are examined elsewhere [1].

6. Employing Electrocoagulation (EC) Fields

In order to make possible efficacious and economic water treatment, EC could be utilized either on its own or as a stage in a treatment train especially for small- or medium-sized implementations where a wide water treatment plant is not economically practicable. Illustrations of pollutants that have been remedied by EC setups along with the parameters used and their performance may be found elsewhere [1]. EC has been proved to be efficient for a large set of implementations like removal of toxic arsenic [127] or fluoride [95] from groundwater, remediation of dairy effluent [128], treatment of wastewater from a set of sources comprising slaughterhouses, textile dyehouses, pharmaceutical processes, oilfields, municipal effluents, paper mills, olive mill processes, metal finishing processes, and disinfecting effluents [47] [61]. The technology stays efficient in eliminating colloidal particles from surface water [64], reducing turbidity in algae [58] [72], microorganisms [63] [67] [92], and micropollutants [1]. Whereas EC remains performant in eliminating a so large variety of pollutants, its efficiency in dealing with organics changes largely [30] [74]. The main route for eliminating organic matters in EC is by adsorption on the coagulant and coprecipitates [65]. Elimination achievement is complicated by the organic interactions with both coagulants and different aqueous species [54] [69].

EC treatment techniques usually involve a pretreatment reservoir that is uti-

lized to monitor and adjust the pH, followed by an electrolysis chamber that may implicate a plurality of electrodes connected to a power supply, followed by reservoirs for flocculation and/or chemical dosing, and finally solid-liquid separation (e.g., by sedimentation, induced flotation [58] [72], or filtration [129] [130] [131]) [61]. Designing the device should consider numerous features, comprising the fluid flow and distribution between the electrodes, floatation, and precipitation, in addition to the running factors (like electrode materials, electrode arrangement, electrode connections, current density, etc.) [61]. Electrical power is applied to the electrodes utilizing either an applied voltage or constant current, frequently with alternating polarity, with the same material employed for both electrodes [61]. A constant current is mostly utilized to attain the wanted coagulant injection and so treatment efficacy [61]. The reactor has to be conceived to deal with the gas formation (usually O_2 and H_2) in the device. In most cases, the H_2 bubbles will be liberated in the solid-liquid separation process [1].

7. Conclusions

EC separation techniques supply credible and green technology for eliminating a large variety of pollutants from water. The technology is more than 100 years old and it is only during the past two decades that its application has pulled important attention from academia and industry. Several features of the process remain to be well known because of the complexity of the interactions between electrochemistry, chemical coagulation, flow, mixing, and transport phenomena. This work focused on the key chemical and electrochemical theories ruling the method and the fundamental parameters that affect the separation efficacy [1] [132]. The main points drawn from this work are listed below:

1) The EC method takes advantage of the concepts of traditional coagulation while furnishing important benefits over a chemical dosing procedure. In EC, the coagulants are injected via integrating electrochemical dissolution of metal at the anode and formation of hydroxide at the cathode. The first dissimilarity between chemical coagulation and EC is simply the way of introduction of the coagulant; nevertheless, EC averts the injection of the counter ion in a coagulant salt and does not need the introduction of alkali to balance the pH change related to hydrolysis of the coagulant. In EC, the pH of the water to be treated must be in the neutral or alkaline interval; however, very little pH change happens during the treatment application [1].

2) The chemical phenomena happening during EC are a function of the electrode material and the composition of the solution [1]. The geometry, electrochemical variables (e.g., current, voltage, and resistance), type of current flows (direct or alternative), and the pH are crucial elements that dictate the separation efficacy. Convenient selection of the electrode material, the design of electrochemical reactors, and systematic control of the electrochemical variables could conduct to an efficient setup. Even if the EC separation method remains an interesting technique, there are numerous dares as the generation of a fouling

layer on the electrode surface that could conduct to running difficulties, as well as the restricted effectiveness for eliminating persistent organic pollutants. Regardless of such dares, for several industrial implementations, the efficacy of the system is proved and the technique of EC furnishes a relatively compact and robust manner for handling end-of-pipes industrial effluent (e.g., textile, leather tanning, pulp and paper, olive mill, metal-bearing industrial effluent, arsenic, and fluoride-containing effluents).

3) Despite the progress of the technology, more comprehension of the basic phenomena happening during EC is requested to guarantee effective design and utilization for novel implementations. Even if EC has been employed for a wide set of usages and pollutants, only a few investigations have tried to interpret the complicated physicochemical phenomena taking place during EC application. Most investigations concentrated on the effectiveness of EC for particular contaminants or implementations, rather than assessing the technology at a mechanistic degree, considering the complicated responses and physical processes happening. An additional dare is the scale-up of EC to satisfy industrial demands. Most investigations realized batch treatment; however, industrial usage is mostly founded on a continuous mode. Scale-up of a chemical method is a function of the usage of established models to define process efficacy as a function of running variables; however, very few such models have been suggested for EC technology. Modeling and scale-up researches remain demanded to permit the easier utilization of EC for industrial implementations without complicated and costly tests. Next studies have to follow the routes of the EC technique for numerous kinds of pollutants at a set of working parameters, in particular for continuous mode, and the expansion of convenient models that could be utilized for scale-up and techno-economic evaluation of EC is required [1]. Running as a destabilization agent and aiding to separate contaminants from the wastewater, the electric field should attract more attention to highlight its fundamental contribution.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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