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Review Article

Disinfection via Electrocoagulation Process: Implied Mechanisms and Future Tendencies

Djamel Ghernaout[1,2]*

[1]*Chemical Engineering Department, College of Engineering, University of Ha'il, Ha'il, Saudi Arabia*

[2]*Chemical Engineering Department, Faculty of Engineering, University of Blida, Blida, Algeria*

***Corresponding Author**: Djamel Ghernaout, Chemical Engineering Department, College of Engineering, University of Ha'il, Ha'il, Saudi Arabia and Chemical Engineering Department, Faculty of Engineering, University of Blida, Blida, Algeria.

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Abstract

Electrocoagulation (EC) is an electrochemical method of applying an electric current on wastewater and producing and injecting metallic coagulant *in situ* and eliminating colloids and metals, as well as other dissolved solids from wastewater. EC technology has been successfully used in eliminating pollutants, pesticides, and radionuclides. This method as well kills pathogenic microbes. This review focuses on implied mechanisms of killing microorganisms' during EC application. The electrical current contribution in EC efficiency through the destruction of microbes via building pores in the cytoplasmic membrane, which augments the cell permeability, is well established. Physical elimination and chemical deactivation mechanisms are described for microbes' removal procedure through the EC technique: (i) enmeshment of microbial pollutants upon EC flocs, (ii) sweep flocculation is preferentially for the destabilization of negatively charged microorganisms, and (iii) deactivation of bacteria membranes via electrochemically formed reactive oxygen species or direct interaction of the electric field.

Keywords: *Electrocoagulation (EC); Disinfection; Electric Field; Electrodes; Electrochemistry; Microorganisms*

Abbreviations

EC: Electrocoagulation; Fe-EC: Electrocoagulation Using Iron Electrodes; Al-EC: Electrocoagulation Using Aluminum Electrodes; MF: Microfiltration; NOM: Natural Organic Matter; OM: Organic matter

Introduction

Electrocoagulation (EC) process is considered as the most promising technology to eliminate simultaneously organic matter (OM) and microorganisms from wastewater [1-4]. EC is performant and green phase-separation technique founded on the liberation of $Fe^{2+/3+}$ (or Al^{3+}) ions from sacrificial Fe (or Al) anodes [5-8]. Table 1 presents EC mechanisms employing Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [9].

Iron hydroxides ($Fe(OH)_{2(s)}$ and $Fe(OH)_{3(s)}$) are comparatively harmless and constitute flocs that let contaminant elimination, forming a precipitable sludge [10,11]. The flocs enmesh colloids via surface complexation or electrostatic attraction and throughout sweep flocculation [12,13]. Moreover, the H_2 gas bubbles formed at the cathode induce the flotation of various contaminants and, as a result, the separation operation is promoted [14,15]. Like traditional electrochemical technique, EC needs easy equipment and is simple to run [16,17]. The periodic replacement of the sacrificial anodes, their passivation, and the electricity cost have been reported as the main drawbacks of this technology [1,18,19].

Many researchers employed EC for killing pathogens in urban wastewater treatment facilities, showing total deactivation (> 99.99%) of *Escherichia coli* [10,20-22], total coliforms [20,23-25] or *Staphylococcus aureus* [26] and microalgae [20,27-29]. EC has as well been

Fe mechanisms	
Mechanism #1 (pH 2)	Anode: $2Fe_{(s)} - 4e^- \rightarrow 2Fe^{2+}_{(aq)}$ (E° = +0.447 V) (1) $2H_2O_{(l)} - 4e^- \rightarrow O_{2(g)} + 4H^+_{(aq)}$ (E° = -1.229 V) (2) Solution: $2Fe^{2+}_{(aq)} + 4OH^-_{(aq)} \rightarrow 2Fe(OH)_{2(s)}$ (3) Cathode: $8H^+_{(aq)} + 8e^- \rightarrow 4H_{2(g)}$ (E° = 0.000 V) (4) Total: $2Fe_{(s)} + 6H_2O_{(l)} \rightarrow O_{2(g)} + 4H_{2(g)} + 2Fe(OH)_{2(s)}$ (5)
Mechanism #2 (pH 7)	Anode: $2Fe_{(s)} - 4e^- \rightarrow 2Fe^{2+}_{(aq)}$ (E° = +0.447 V) (1) $Fe^{2+}_{(aq)} - e^- \rightarrow Fe^{3+}_{(aq)}$ (E° = -0.771 V) (6) $Fe_{(s)} - 3e^- \rightarrow Fe^{3+}_{(aq)}$ (E° = +0.037 V) (7) Solution: $Fe^{2+}_{(aq)} + 2OH^-_{(aq)} \rightarrow Fe(OH)_{2(s)}$ (3) $2Fe^{3+}_{(aq)} + 6OH^-_{(aq)} \rightarrow 2Fe(OH)_{3(s)}$ (8) Cathode: $8H_2O_{(l)} + 8e^- \rightarrow 4H_{2(g)} + 8OH^-_{(aq)}$ (E° = -0.828 V) (9) Total: $3Fe_{(s)} + 8H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + 2Fe(OH)_{3(s)} + 4H_{2(g)}$ (10)
Mechanism #3 (pH 12)	Anode: $2Fe_{(s)} - 6e^- \rightarrow 2Fe^{3+}_{(aq)}$ (E° = +0.037 V) (7) Solution: $2Fe^{3+}_{(aq)} + 6OH^-_{(aq)} \rightarrow 2Fe(OH)_{3(s)}$ (8) Cathode: $6H_2O_{(l)} + 6e^- \rightarrow 3H_{2(g)} + 6OH^-_{(aq)}$ (E° = -0.828 V) (9) Total: $2Fe_{(s)} + 6H_2O_{(l)} \rightarrow 2Fe(OH)_{3(s)} + 3H_{2(g)}$ (11)
Al mechanism	
Mechanism (pH 7)	Anode: $Al_{(s)} - 3e^- \rightarrow Al^{3+}_{(aq)}$ (E° = +1.66 V) (12) $2H_2O_{(l)} - 4e^- \rightarrow O_{2(g)} + 4H^+_{(aq)}$ (E° = -1.229 V) (2) Solution: $Al^{3+}_{(aq)} + 3OH^-_{(aq)} \rightarrow Al(OH)_{3(s)}$ (12) $Al(OH)^-_{4\,(aq)} \rightarrow OH^-_{(aq)} + Al(OH)_{3(s)}$ (13) Cathode: $4H_2O_{(l)} + 4e^- \rightarrow 2H_{2(g)} + 4OH^-_{(aq)}$ (E° = -0.828 V) (9) $Al_{(s)} + 4OH^-_{(aq)} - 3e^- \rightarrow Al(OH)^-_{4\,(aq)}$ (14) Total: $2Al_{(s)} + 8H_2O_{(l)} \rightarrow 5H_{2(g)} + 2Al(OH)_{3(s)} + O_{2(g)}$ (15)

Table 1: *EC mechanisms using Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [9].*

used to decrease the organic load of dye and textile wastewater [30-36], urban wastewater [37], olive oil mill wastewater [38] and cheese whey or dairy wastewater [39,40].

This review focuses on implied mechanisms of killing microorganisms' during EC application.

Behavior of microorganisms in EC setups

Killing pathogens electrochemically has been largely established employing carbon, mixed metal oxides, boron doped diamond, and other dimensionally stable anodes widely employing model waters not containing natural organic matter (NOM) [9,20,41-43]. The main benefit of purely electrochemical deactivation is that *in situ* killing agent production diminishes the hazard linked to transport and on-site storage of big quantities of greatly poisonous chemical products [44,45]. In such setups, deactivation happens via oxidants formation (like chlorine [46]) and free radical intermediates [41,42,47]. On the other hand, EC is in fact usually realized employing sacrificial (not dimensionally stable) anodes and real-world surface waters hold NOM [48]. It is proved that intact microfiltration (MF) membranes alone have been found to eliminate > 99.9999% (6-logs) of bacteria and protozoa easily contributed to size exclusion but they let nearly unhindered virus passage [49]. Moreover, EC alone is greatly performant for bacteria and algae control [20,50,51]. Consequently, Chellam and Sari [48] focused viruses, which are smaller than MF membrane pores and discussed information collected from experiments concerning the integration of Al-EC/MF (or Fe-EC/MF) for potable water treatment. It is established that Al-EC importantly decreases MF fouling via causing the generation of a cake composed of particles bigger than in the raw water [48]. Nevertheless, Al flocs have the capacity to compact or compress and relatively worsen MF fouling at more elevated pressures. Moreover, novel findings illustrated crucial enhancements in microfiltered water quality throughout EC pre-treatment. Indeed, $Al(OH)_{3(s)}$ flocs sorb NOM and disinfection by-products precursors [52,53], which are thus fixed on the MF membrane surface. Viruses are found efficiently sweep coagulated via EC and eliminated later by MF. A thick cake layer of $Al(OH)_{3(s)}$ flocs further enhances virus elimination upon functioning as a dynamic membrane [48]. Chellam and Sari [48] concluded that EC/MF setups are encouraging choices for small-scale decentralized facilities since they really give many barriers against pollutants and decrease membrane fouling while needing limited operator attention [54,55].

Even if the idea of using electrical energy to dissolve metal coagulants and produce oxidants *in situ* is not recent, EC has not until now reached a solid foothold in water/wastewater treatment industry [48]. Nowadays, more development has been performed to establish diverse efficiency aspects comprising pH variations among electrodes, precipitated solid phases, electrode passivation, and additional parameters touching EC efficiency. Nevertheless, much more research has to be performed because a global comprehension of the type and composition of precipitated phases founded on water chemistry and electrolysis parameters remains elusive. In addition, first economic evaluations announce that EC is competitive to traditional coagulation [56] particularly for smaller installations [57]. Chellam and Sari 2016 [48] concluded that EC may be applicable for a large interval of water chemistries and removes the necessity to inject base to fix pH throughout coagulation (conducting to decreased chemical handling and enhanced method simplicity). Enhanced conceptions comprising multiple electrode configurations have also been suggested [58,59]. All these factors show the augmenting focus on electrochemical and MF technologies particularly for small, portable systems for localized treatment [55].

EC improved with peroxide

During the last three decades, employing hydroxyl radicals in aqueous medium has been proposed to enhance the oxidation of toxic contaminants existing in the effluent to be handled [60,61]. The hydroxyl radical is a very powerful oxidant. In 1993, researchers [62] assessed the benefits and inconvenient of many advanced oxidation processes. They found that the techniques founded on hydrogen peroxide (H_2O_2) to induce the production of hydroxyl radical are the most interesting methods, because such technologies do not implicate the injection of highly poisonous chemical agents, their handling is comparatively easy and they are not too expensive. In addition, such approaches present more performant mass transport characteristics than those implying different hydroxyl radical promoting species like ozone [62]. Indeed, the hydroxyl radical has an oxidation potential of 2.8 V, just below fluorine, 3.0; however, ozone has 2.1 and hydrogen peroxide only has 1.8 [63].

EC for disinfecting seawater

EC method may be employed as a substitutional pretreatment with a view to evaluating its pertinence to exchange the traditional pretreatments (like chemical coagulation, chlorination and scale inhibitors) applied to diminish membrane fouling before seawater desalination via reverse osmosis process [64]. Hakizimana., *et al*. [65] performed EC tests in a batch cell using Al electrodes driven in the galvanostatic mode. With a view to controlling the EC performance in eliminating OM [66,67] from seawater, they measured the absorbance

(UV_{254} nm) and dissolved organic carbon. Similarly, cultivable heterotrophic bacteria were counted to monitor the killing microorganisms potential of the electrochemical technology and total hardness was followed. Eliminating OM [68-70] from seawater via EC was enhanced with higher current density and lower pH. Indeed, EC eliminated around 70% dissolved organic matter, while absorbance decrease was about 90% with total elimination of bacteria at elevated current density. On the contrary, decreasing total hardness was low, around 10%; consequently, EC could not be utilized as a softening approach. Practical findings proved the elevated ability of EC as a pretreatment technique to mitigate potential organic fouling and biofouling of reverse osmosis membrane thanks to its capacity to eliminate efficaciously dissolved OM and bacteria from seawater (Figure 1).

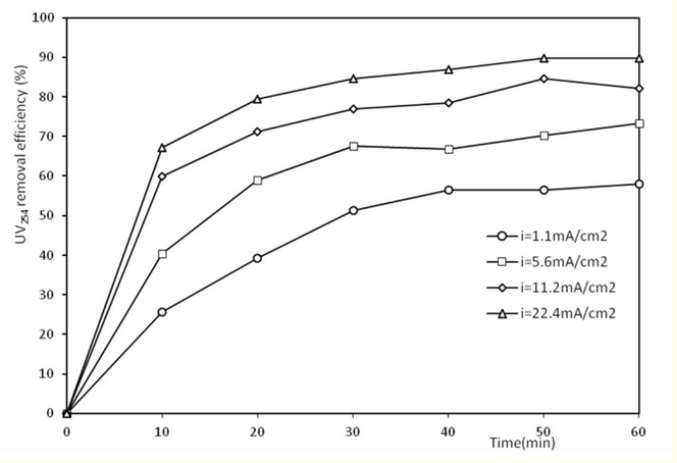

Figure 1: *Effect of current density on Cultivable Heterotrophic Bacteria (CHB) removal efficiency [65].*

Microbial cells elimination through Fe-EC

Delaire., *et al.* [43] performed deep researches on Fe-EC process for disinfecting water and showed that this green technology is a promising treatment option for groundwater containing arsenic and bacterial contamination. In their following work [10], employing *E. coli* as the model indicator, they established that physical elimination through enmeshment in EC flocs is the main pathway of bacteria inactivation in the existence of HCO_3^-, which greatly hinders deactivation, supposedly explained by a decrease in the lifetime of reactive oxidants (Figure 2). They explained that the fixation of EC flocs to cell membranes, which conducts to microorganisms' encapsulation in precipitates, is controlled mainly through interactions among EC precipitates and phosphate functional groups on microbe surfaces. In single solute electrolytes, both P (0.4 mM) and Ca/Mg (1 - 13 mM) blocked the fixation of EC precipitates to bacterial cell membranes, while Si (0.4 mM) and ionic strength (2 - 200 mM) did not affect *E. coli* inactivation. More important, P (0.4 mM) did not influence *E. coli* deactivation in electrolytes carrying Ca/Mg, consistent with bivalent cation bridging between bacterial phosphate groups and inorganic P linked to EC precipitates. Delaire., *et al*. [10] demonstrated that EC precipitate fixation is greatly independent of cell membrane composition, consistent with comparable densities of phosphate functional groups on Gram-positive and Gram-negative cells. Such findings may be crucial to predict the efficiency of Fe-EC to remove bacterial pollutants from waters with various chemical compositions.

In the same direction, Nguyen., *et al.* [71] assessed a fresh pilot scale EC setup for increasing total phosphorus (TP) elimination from municipal wastewater. Their EC device was functioned in continuous and batch operating mode under changing parameters (such as flow rate, initial concentration, electrolysis time, conductivity, voltage) to determine correlative phosphorus and electrical energy consumption. Their findings established that the EC device could efficiently eliminate phosphorus to satisfy present discharge standards of less than 0.2 mg/L during 2 - 5 minutes. This objective was attained in all intervals of initial TP concentrations examined. As concluded by Nguyen., *et al.* [71], EC technology is greatly performant in a relatively simple, easily managed, and cost-effective for wastewater treatment system.

Cotillas., *et al.* [72] combined Fe-EC with UV irradiation (photo-EC) for the concurrent elimination of colloids and *E. coli* from real treated municipal wastewaters. Their findings proved that single EC behaves as an extremely powerful technique even employing low

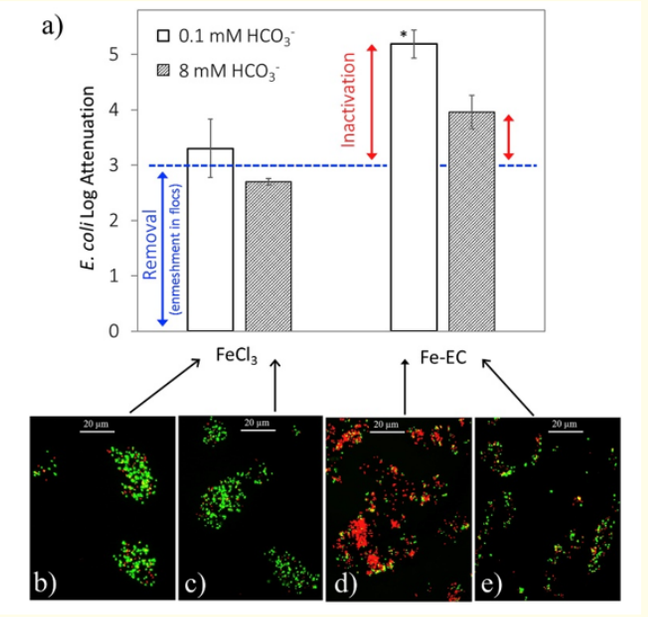

Figure 2: *E. coli removal with Fe-EC and $FeCl_3$, with and without 8 mM HCO_3^-. Fe dosage was 0.5 mM in all trials. Panel a shows E. coli log attenuations. The asterisk shows that the detection limit for microorganisms inactivation was obtained for some of the replicate tests. Panels b-e indicate fluorescent microscopy images of live (green)-dead (red) stained E. coli cells. The blue dashed line is the average deactivation in all $FeCl_3$ tests (with and without HCO_3^-) and represents elimination (blue arrow). E. coli log removals are compared to this baseline to determine approximate log deactivations (red arrows). All tests were performed at pH 7.0. In 0.1 mM HCO_3^- trials, 2 mM NaCl were injected for conductivity [10].*

current densities. The *E. coli* cells are eliminated not only throughout their enmeshment into germinating flocs, but also via the attack of electrochemically formed chlorine disinfectant species. Integrating UV irradiation to Fe-EC enhances technology performance in a matter of eliminating *E. coli* and colloidal particles. They assessed the impact of current density on process performance, discovering a synergistic interaction of both methods at low current density (1.44 A/m^2); however, a counter influence at higher values of current density (7.20 A/m^2). This counter impact is induced by the less performant transmission of UV irradiation to the bulk solution because of the augmentation in the content of colloids.

Future tendencies

Several scientists, such as Govindan., *et al.* [73] and Llanos., *et al.* [74], examined deeply the basic disinfection pathway occurring through the EC technology [74]. They discussed the biomass (bacteria, virus, and algae) elimination procedure via EC following the practical researches realized during these twenty years. Physical elimination and chemical deactivation mechanisms are described for microbes' removal procedure through the EC technique: (i) enmeshment of microbial pollutants upon EC flocs, (ii) sweep flocculation is preferentially for the destabilization of negatively charged microorganisms, and (iii) deactivation of bacteria membranes via electrochemically formed reactive oxygen species or direct interaction of the electric field (Figure 3) [75].

In order to reach a deeper comprehension of virus and algae removal route, more sophisticated research on algae and virus elimination is necessitated [73,76,77]. Heffron., *et al.* [78] performed an excellent research on EC-electrooxidation for virus mitigation in drinking water.

There is a huge tendency of using EC process as hybrid technology combined with physical or advanced oxidation processes such as ozonation [79,80], UV irradiation, membrane processes [81-83], and Fenton (Figure 4) [84].

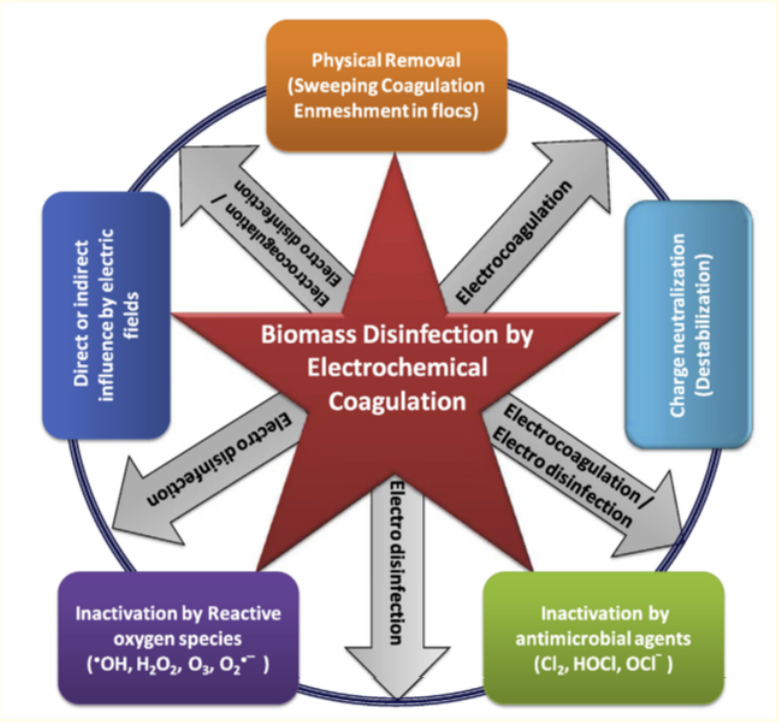

Figure 3: *Viable pathways for microorganisms' killing operation during EC process [73].*

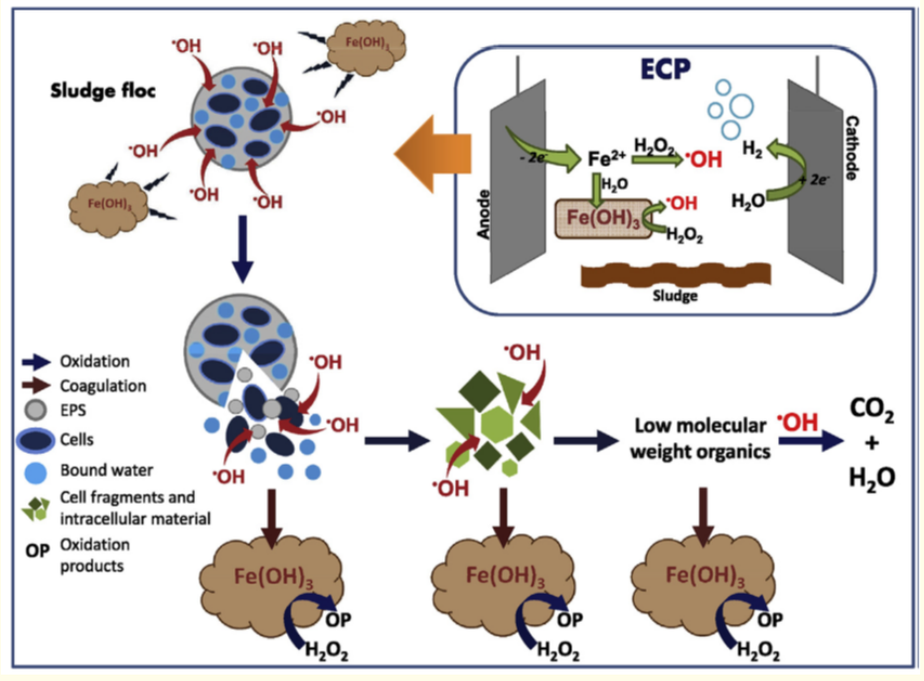

Figure 4: *Degradation mechanisms taking place during electrochemical peroxidation (ECP) of anaerobic sludge. Inset panel: production of coagulants and oxidants during ECP using mild steel electrodes [84].*

As discussed in our previous works [85-87], green chemistry remains indispensable for sustainable development, as it will conduct to fresh solutions to existing problems [88]. There are interesting perspectives concerning the green chemistry implementation in water treatment technology [89], like ferrate(VI) [90-92] injection, as oxidant/disinfectant/coagulant, algae harvesting from the algal biomass for biodiesel production [93], simulation of the open sky seawater distillation [94,95] and water reuse [96-100]. In such perspectives, EC technique should possess its proper place as a single technique or hybrid process [101-103].

Regardless of the considerable development attained in EC technology usage, more research has to be performed to arrive at an elevated degree of industrial satisfaction [104-106]. Humankind future quality is highly dependent on preserving nature from more polluting disasters and treating wastewater for drinking water direct use objectives.

Conclusion

The main points drawn from this work may be given as:

1. If EC was utilized as a killing microorganisms' technique, an elevated disinfection performance was reached with nearly total elimination of bacteria. Even if before seawater desalination, EC was established as a performant choice to chlorination, since the latter shows numerous complications, like its ineffectiveness to avoid biofouling and its reactivity with organic chemicals that may conduct to the generation of carcinogenic organic by-products comprising trihalomethanes, haloacetic acids, and various poisonous disinfection by-products. Moreover, chlorinating feed seawater may induce the breaking of organic macromolecules into smaller pieces readily consumable by microbial cells, that way provoking biofouling.
2. Compatible with the terrestrial existence of phosphate groups on bacteria surfaces, Fe-EC is to the same degree efficient towards Gram-positive and Gram-negative bacteria, rough and smooth alike. Fe-EC, as a technically viable to decentralized arsenic remediation in low-resource settings, may as well efficiently eliminate all kinds of microbial pollution from a large interval of groundwater sources. With a view to proving the capacity of Fe-EC to take the place of present killing microorganisms' techniques when employed to groundwater, field validation of such encouraging findings and research of virus removal are required.
3. Despite EC has been greatly developed for killing microorganisms' with a view to treat wastewater, more researches are needed to expand pilot-scale and industrial usages. More investigation on virus demobilization upon EC is more needed.

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