

Communication

Iron Electrocoagulation Process for Disinfecting Water – A Review

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Abstract: During the last two decades, electrocoagulation (EC) process has known a large success as an electrodisinfection (ED) technology especially if iron (Fe) electrodes are used instead of aluminum ones as sacrificial anodes. This review discusses the pertinent findings of the Delaire et al. excellent work who focused on microbes removal via Fe-EC and attracted the attention on the interactions between bacterial phosphate groups and Fe (III) precipitates. Employing the model indicator *Escherichia coli*, physical elimination through enmeshment in EC precipitate flocs was shown the main procedure of bacteria reduction in the existence of HCO_3^- , which importantly prevents demobilization, probably because of a decrease in the lifetime of reactive oxidants. The fixation of EC precipitates onto cell membranes, which leads to microbes' encapsulation in flocs, is directed firstly via interactions among EC precipitates and phosphate functional groups on bacteria surfaces. 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 are crucial to anticipate the efficiency of Fe-EC to remove bacterial pollutants from the water with changing chemical compositions. In spite of the huge advances in studying the EC process as an ED technique, considerable efforts remain to be performed to reach its better acceptance in the industrial world.

Keywords: Electrocoagulation (EC), Electrodisinfection (ED), Iron, Bacteria, Cell Wall, Gram-positive and Gram-negative Cells

1. Introduction

During iron electrocoagulation (Fe-EC) process, an Fe (0) anode is solubilized electrolytically to produce Fe (II), which is oxidized in the presence of solubilized O_2 to form Fe (III) (oxyhydr) oxide precipitates with an affinity for microbial and chemical pollutants [1-5]. Fe-EC may efficaciously eliminate microorganisms in a variety of water matrices [1, 6, 7]. Delaire et al. [1] established that Fe-EC might reduce *Escherichia coli* from synthetic Bengal groundwater (SBGW) without losing arsenic elimination. Two phenomena participated in reducing microbes in Fe-EC: (1) physical elimination, induced by fixing EC precipitates on cell membranes, which conducts to microorganisms enmeshment in Fe (III) flocs and next settlement,

and (2) demobilization through reactive species formed during Fe (II) oxidation via O_2 [8]. Basic details of the routes inherent to these two phenomena stay obscure [9, 10]. As an illustration, the bacterial functional groups and the sort of chemical interactions (electrostatic vs. specific bonding) controlling microbes' enmeshment in flocs are not well-established [11]. Furthermore, the impact of main groundwater components, like HCO_3^- , Ca, Mg, Si, and P, which may overlap with both demobilization and elimination, has not been examined. Also, the effect of microbes' surface construction (Gram-positive vs. Gram-negative, smooth vs. rough Gram-negative) on reduction has not been explained. By treating such understanding lacks, research may enhance importantly Fe-EC efficiency in different water matrices carrying various sorts of bacterial pollution [11].

Above microbe cell membranes, four sorts of surface functional groups are existing at similar densities: hydroxyl ($pK_a \sim 9.0$), amine ($pK_a \sim 9.0$), carboxyl ($pK_a \sim 4.7$), and phosphate groups ($pK_{a1} \sim 3.1$, $pK_{a2} \sim 6.6$) [12, 13]. Hydroxyl and amine moieties do not possess a solid affinity for Fe (III) oxides [14, 15] and consequently they are not predicted to strongly interact with EC precipitates [11]. On the other hand, carboxyl and phosphate moieties possess solid affinities for Fe (III) oxides [16-18]. Researches employing Attenuated Total Reflectance Fourier-Transform Infrared spectroscopy (ATR-FTIR) have illustrated direct bonding of bacterial phosphate and carboxyl groups to hematite and goethite [19-21]. Nevertheless, such investigations were realized in managed laboratory setups and easy water matrices. In addition, they cannot be immediately extrapolated to Fe-EC in groundwater. Indeed, in such medium precipitates and microorganisms interact in an agitated suspension and the existence of bivalent cations (Ca and Mg) and oxyanions (P and Si), which may sorb to bonding sites on microbes [22, 23], and precipitates [24], respectively, and may, therefore, interfere with adhesion [11].

More than the electrolyte constitution, several investigations have proved that the biomolecular configuration of bacterial cell membranes may influence their interactions with mineral surfaces via modifications in surface charge, hydrophobicity, and steric hindrance [25-27]. Since waterborne pathogenic microbes and indicator organisms surpass the domain of Gram-positive, smooth and rough (with and without Oantigen) Gram-negative strains [28], realizing the influence of cell membrane composition on microbes reduction using Fe-EC is fundamental to extrapolate Delaire et al. [11] results to all bacterial species linked to water quality.

In order to investigate bacteria-Fe interactions, spectroscopic techniques like ATR-FTIR, X-ray fluorescence (XRF) and X-ray absorption spectroscopy (XAS) have been employed [19, 29-31]. Nevertheless, such techniques cannot conveniently define bacteria-Fe (III) interactions in setups where Fe (III) is co-precipitated with bacteria in complicated electrolytes comparable to groundwater. As an illustration, Fe-Fe bonds from bacteria-precipitate interactions and from aqueous P sorption to precipitates appear very identical employing ATR-FTIR [19] and would not be discernible with P K-edge XAS [11]. As well, ATR-FTIR is not adequate to examine interactions producing within huge flocs because of the small penetration length of infrared beams in the aqueous medium ($\sim 1 \mu\text{m}$). To avoid such imperfections, Delaire et al. [11] suggested an original method, where macroscopic facts of bacteria reduction in systematically changed electrolytes are merged with ζ -potential assessments to clarify the molecular interactions among bacteria and EC precipitates. Even if such a method may at the last give indirect proof for particular interactions among bacteria and precipitates, it constructs over former spectroscopic investigations. These researches have specified bacteria-Fe oxide bonding phenomena in easy controlled systems [19-21] and configurations of Fe-EC precipitates in complicated water matrices [24], to obtain data concerning bacteria elimination routes in groundwater-like electrolytes.

2. Mechanism of Microorganisms' Removal Via Fe-EC

Delaire et al. [11] established that microbes' demobilization, which may be crucial in the absence of oxidant scavengers, is greatly inhibited by HCO_3^- levels distinguishing of natural waters. Thus, Delaire et al. [11] suggested that physical elimination would be the first procedure of bacteria reduction in most water treatment's usages (Figure 1).

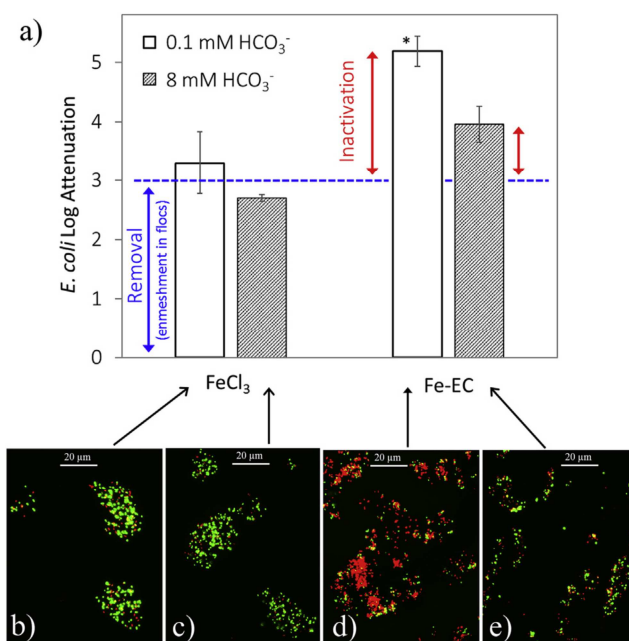


Figure 1. *E. coli* reduction with Fe-EC and FeCl₃, with and without 8 mM HCO₃⁻. Fe dosage was 0.5 mM in all experiments. Panel a shows *E. coli* log attenuations. The asterisk indicates that the detection limit for bacteria attenuation was reached for some of the replicate experiments. Panels b-e show fluorescent microscopy images of live (green)-dead (red) stained *E. coli* cells. The blue dashed line is the average attenuation in all FeCl₃ experiments (with and without HCO₃⁻) and represents removal (blue arrow). *E. coli* log attenuations are compared to this baseline to deduce approximate log inactivations (red arrows). All experiments were conducted at pH 7.0. In 0.1 mM HCO₃⁻ experiments, 2 mM NaCl were added for conductivity [11].

Delaire et al. [11] established that elimination is forced by the interactions of EC precipitates with bacterial phosphate groups, which may link to Fe (III) surfaces directly or via a Ca/Mg bridge to P sorbed on precipitates. These pathways are harmonious with the minimal effect of ionic strength, which is not anticipated to touch greatly these particular interactions. Considering such pathways, the differing influences of P and Si detected by Delaire et al. [11] may be extrapolated to additional strongly- (e.g., arsenate) and weakly- (e.g., borate, arsenite, nitrate) sorbing oxyanions, respectively. Identically, the illustrated effect of Ca/Mg (hardness) may be generalized to metallic bivalent cations that may be existent in wastewater, like Cu²⁺, Cd²⁺, Pb²⁺, and Zn²⁺. Natural organic matter (NOM) has been observed to restrain virus reduction through Fe-EC [2, 32]. Nevertheless, since NOM at most carries carboxyl and hydroxyl moieties [33-38], which possess a weaker sorption affinity for Fe (III) (oxyhydr) oxides than bacterial phosphate

groups, it is not awaited to prevent microorganisms' elimination via Fe-EC hugely, harmonious with Delaire et al. former discovery in synthetic Bengal groundwater [1]. In addition, extracellular polymeric substances (EPS), in which environmental bacteria are typically embedded, are not anticipated to influence the adhesion of EC precipitates importantly, since EPS carry phosphate functional groups that may constitute bonds with Fe (III) oxides [39], identical to bacterial cell membranes [40].

Compatible with the global existence of phosphate groups on microbes' surfaces, Fe-EC is evenly performant towards Gram-positive and Gram-negative bacteria, rough and smooth alike. Delaire et al. [11] robustly proposed that Fe-EC might as well efficiently eliminate all sorts of bacterial pollution from a large domain of groundwater sources. Practical establishment of such encouraging findings, as well as a research of virus reduction, are required to underline the capacity of Fe-EC to replace present disinfection techniques when used in groundwater treatment [11].

3. Impact of the Electric Field on Microorganisms' Removal Via EC

Boudjema et al. [41] applied successfully EC process for treating the Oued El Harrach river water. They established that the electrical current has a basic contribution to the remediation performance and particularly in the demolition of microbes via shaping pores in the cytoplasmic wall, which augments the cell permeability (Figure 2). As a result, the inter-extracellular exchange is not realized by the cell membrane but assists cytoplasm escape followed by the microbial cell death. The bacterial resistance to the electric field changes following the cytoplasmic membrane structure (Gram negative, Gram-positive). This resistance is more important for the spore-forming bacteria.

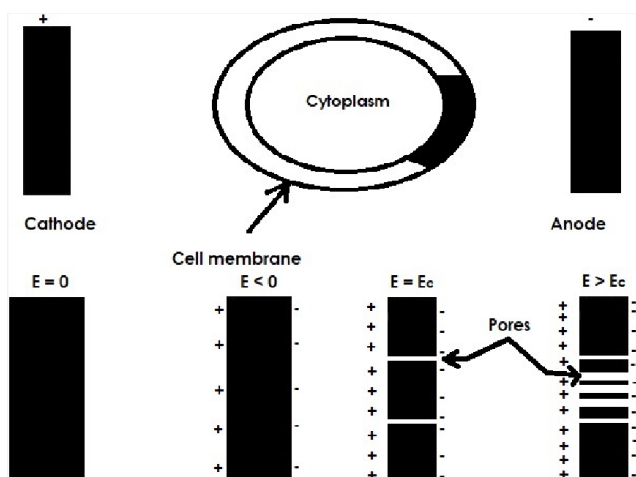


Figure 2. Principle of the lethal effect of an electric field on the cell membrane. (E) Electric field intensity, (Ec) critical electric field intensity, (■) cell membrane [41].

In 2008, the significance of the electric field action in killing *E. coli* was established [6]. Recent reviews have

discussed the importance of the electric field participation in deleting pathogens throughout electrodisinfection (ED) technology in a general manner [42-49] (Figure 3).

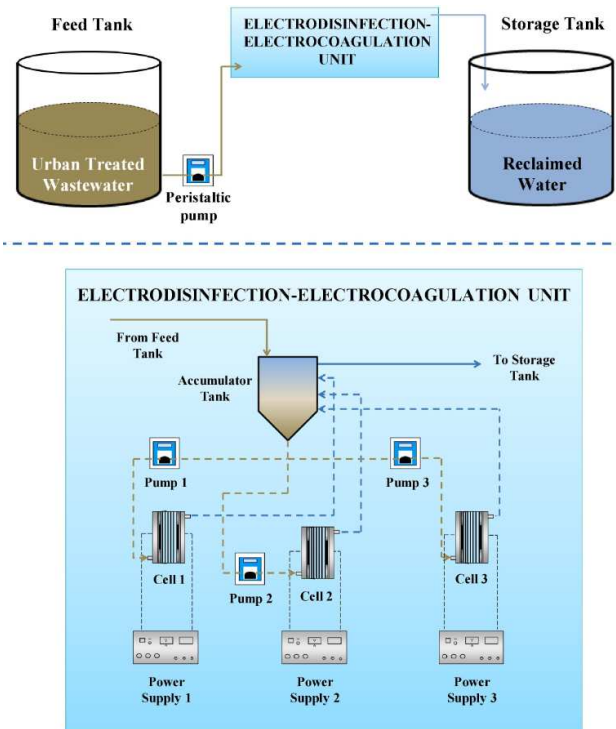


Figure 3. Electrodisinfection-electrocoagulation pilot plant for wastewater reclamation [49].

4. Conclusions

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

Compatible with the global existence of phosphate groups on bacteria surfaces, Fe-EC is equally efficient towards Gram-positive and Gram-negative bacteria, rough and smooth alike. Fe-EC can efficiently eliminate all kinds of bacterial pollution from a large domain of groundwater sources. Practical proof of such encouraging discoveries and profound research of virus removal are required to emphasize the capacity of Fe-EC to replace chemical disinfection for water treatment.

There is no doubt that EC process is a well proven as an ED technique; however, considerable efforts remain to be performed to reach its better acceptance at the industrial level.

Like in chemical water disinfection, similar issues like disinfection by-products (DBPs) generation have driven to the search for better water disinfection techniques such as ED methods. Nevertheless, their wide extension is restrained by DBPs formation. More research needs to be pointed into such direction.

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