

Short Communication

Virus Removal by Electrocoagulation and Electrooxidation: New Findings and Future Trends

Djamel Ghernaout^{1,2*}

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

²Chemical 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, Tel./Fax: +213-25-433-631; E-mail: djamel_andalus@hotmail.com

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Abstract

Electrocoagulation (EC) process is a highly performant technique for treating water particularly in terms of killing pathogens, especially viruses. This paper discusses briefly some recent advances in removing viruses from water. Routes of bacteriophage reduction due to EC were discussed throughout the literature. Physical elimination was mainly attributed to embodiment in flocs; however, demobilization was firstly linked to ferrous iron oxidation. Removing bacteriophage for any method working on iron has to be seriously viewed because of more important vulnerability of bacteriophages to demobilizing through Fe^{2+} oxidizing. Through juxtaposing the traditional treatment via $\text{FeCl}_{3(s)}$ as coagulant and $\text{Cl}_{2(g)}$ as disinfection agent, the EC-electrooxidation (EO) approach was observed less performant in typical surface waters but more performant in typical groundwaters. In many usages, consecutive EC-EO was useful, still real aspects such as free radicals remaining in water risk to overpass the combined process inherent advantages. More research concerning both virus removal and pathogenic bacteria remains to be performed in the perspective of a large industrial usage of EC process.

Keywords: Electrocoagulation (EC); Disinfection; Electric field; Electrooxidation (EO); Viruses; Microorganisms

Introduction

Treating water electrochemically remains promising like a viable choice especially for conventional approaches depending on chemical coagulation and disinfection particularly in small-scale water devices [1-4].

Electrochemical oxidation, or electrooxidation (EO), employs stable electrodes to directly oxidize pollutants at

the electrode exterior and/or produce oxidants in water [5]. Usually, boron-doped diamond (BDD) electrodes are utilized in EO laboratory experiments thanks to BDD's elevated resistance to chemical and thermal decomposition and low inclination to react with solvents [6,7]. EO using BDD is able to kill organisms via either the generation of reactive oxygen species (ROS) from electrochemical water decomposition or free chlorine and chlorine dioxide formed from oxidation of chloride [8-12]. If chloride is not present, hydroxyl radicals at the electrode exterior are the main oxidant species, and disinfection occurs via pathogen transport and sorption to the electrode exterior [13,14]. The efficiency of BDD disinfection augments frequently with the concentration of chloride in the solution [11,12,15,16]. Augmented disinfection in the existence of Cl^- ions can imply that chlorine species are more significant to BDD treating juxtaposed to ROS. On the other hand, chlorine can possess an interactive impact on ROS formation, with more ROS produced in high chloride matrices [12].

Researchers [17,18] employed an integrated technique utilizing EO with BDD accompanied by electrocoagulation (EC) for *Escherichia coli* removal. EC is the in-place production of coagulant species in the water thanks to oxidation of a sacrificial anode, usually Al or Fe [19-21]. EC has been applied as a pretreatment method for elimination of colloids and natural organic matter in diverse usages [22-26]. Moreover, EC is a performant technology of virus removal [27]. The main route of EC is frequently viewed to be identical as conventional coagulation, i.e., physical elimination through charge neutralization or sweep flocculation [27-29], even if researchers such as Ghernaout et al. [1,30-32] and Boudjema et al. [33] mentioned the huge contribution of the electric field. Nevertheless, EC may as well demobilize viruses and

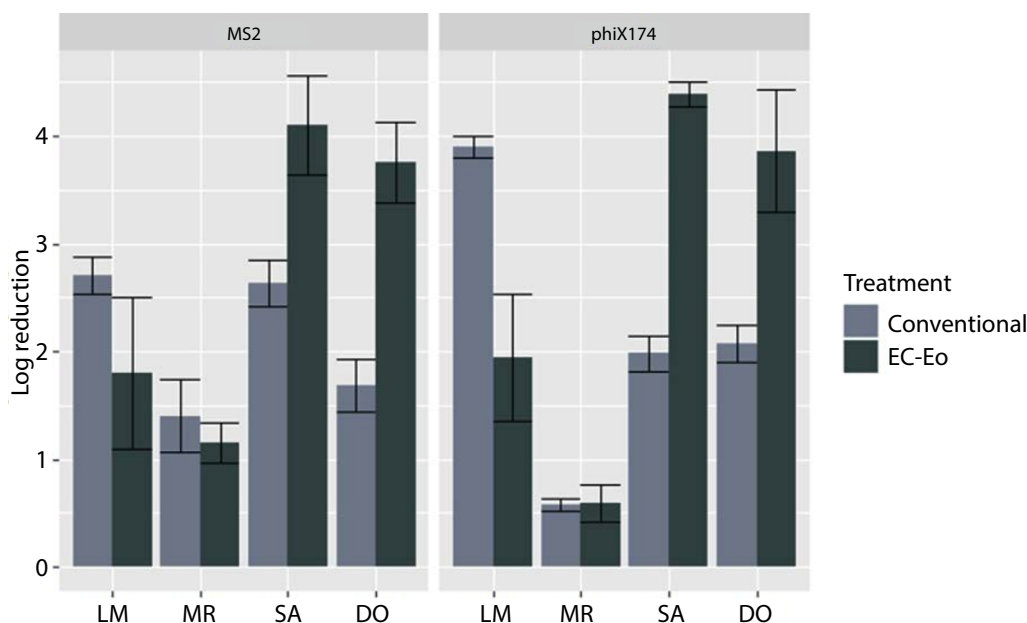


Figure 1: Comparison of traditional coagulation/chlorination treatment train to the EC-EO treatment train for removing bacteriophages MS2 and FX174. “Traditional” treatment composed of FeCl₃ conventional coagulation (22 mg/L Fe) assisted by NaOCl disinfection (6 mg-min/L Cl₂). EC-EO was performed around the optimal division of 150 C/L in the EC-EO treatment train (25% EC for LM and MR, 50% EC for DO and SA). LM ¼ Lake Michigan, MR ¼ Mississippi River, SA ¼ Sandstone Aquifer, and DO ¼ Dolomite Aquifer model waters. Each column shows the mean values of triplicate trials with ± 1 standard error presented by the error bars [4].

bacteria through the formation of free chlorine or Fenton-like reactive intermediates because of Fe²⁺ oxidation [34-39]. Fe-EC produces Fe²⁺ in solution via oxidizing a zero-valent Fe anode [40,41]. Oxidizing ferrous ions to ferric (Fe³⁺) may produce intermediate oxidants capable of demobilizing viruses [36,37]. Virus deactivation because of Fe-EC is more dominant in moderately acidic waters (~pH 6); however, real elimination is the prevalent fate of viruses in Fe-EC for pH>7 [35]. In a hybrid EO-EC reactor, researchers [18] observed that Fe electrodes were more efficient in removing *E. coli* compared to Al electrodes. They explained the greater elimination showed with Fe electrodes by the generation of a passivation film on Al electrodes, even if the probability of *E. coli* demobilization because of Fe oxidation was not tested [4].

Disinfecting water using EO has been largely studied with a view to removing microbes [12,13,15,17,42-44]; however, virus elimination using EO has attracted relatively insignificant focus [45-47]. Both bacteriophage MS2 and recombinant human adenovirus have been observed to be more reluctant to electrochemical disinfection juxtaposed to *E. coli* and *Enterococcus* [47]. In fact, microbes cannot be exact measures of virus removal through EO, so the shortage of data about virus removal by means of EO is a crucial failure. In addition, EC stage in front of EO can present benefits for virus elimination, but it has not until now completely evaluated.

This short communication focuses on the recent findings of Heffron et al. [4] work published this year. Their other new and excellent researches [27,35,36] need more deep

discussion to get better comprehension of the EC and EO usage as water treatment technology especially in terms of pathogens removal.

EC and EO for Killing Viruses

Heffron et al. [4] performed an excellent investigation in which they assessed Fe-EC as a pretreatment for killing waterborne viruses through BDD-EO. Firstly, they analyzed the impacts of pH, natural organic matter, and colloids on virus removal via EO. They examined the effect of Fe²⁺ on EO with a view to designing an efficient remediation series employing consecutive EC and EO. Moreover, they tested a consecutive EC-EO treatment train for removal of two bacteriophage surrogates and echovirus in four synthetic water matrices representing a range of source waters. They compared the EC-EO device to a more traditional treatment train including conventional coagulation and Cl_{2(g)} disinfection (Figure 1).

Heffron et al. [4] defined both a foundation for employing a new, consecutive EC-EO treatment train for potable water and minutely assessed treatment efficiency for two bacteriophages and a human waterborne virus. Despite the fact the EC-EO treatment setup suggested in their investigation was not useful in all water matrices, the enhanced virus removal attained by EC-EO in model surface waters attracts more focus. The advantage of EC-EO was possibly not attributed to iron improved oxidation. On the other hand, bigger virus removal detected in the EC-EO treatment train was possibly obtained via the complementary influences of real elimination by coagulation/filtration, ferrous iron-based disinfection, and EO disinfection.

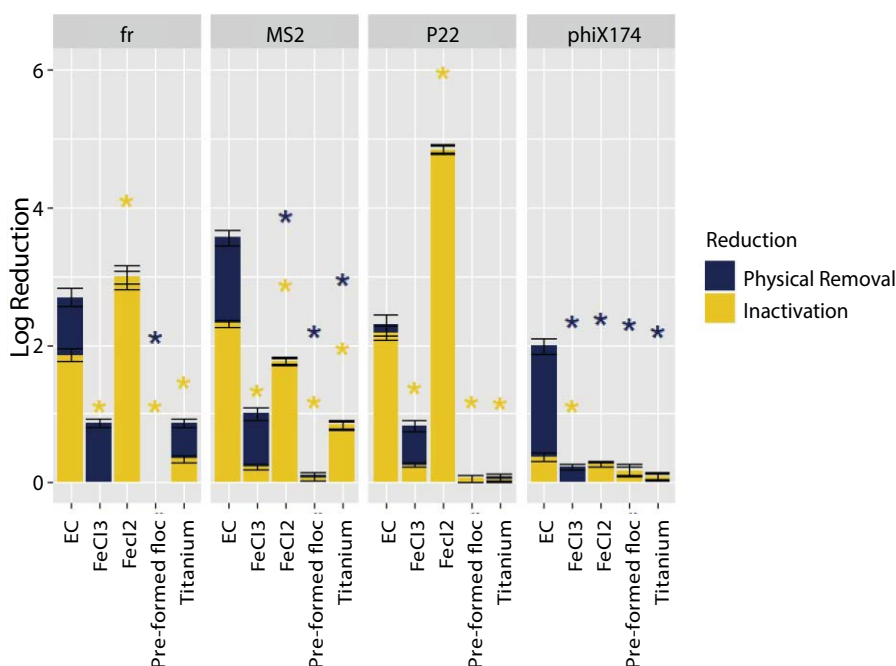


Figure 2: Routes of bacteriophage reduction thanks to EC, conventional coagulation, adsorption and EO. Demobilization and physical elimination were compared between EC, chemical coagulation with ferric chloride (FeCl₃), chemical coagulation with ferrous chloride (FeCl₂), flocs produced *via* EC prior to the addition of bacteriophages (pre-formed flocculation), and EO with inert titanium electrodes (Titanium). Asterisks indicate a significant difference in log reduction from EC due to physical removal (blue asterisk) or inactivation (yellow asterisk). Error bars represent standard error of the mean of triplicate tests [35].

Mechanistic Insight into Virus Removal During EC

In order to ascertain why several bacteriophages revealed deactivation thanks to EC, Heffron et al. [35] studied the routes of bacteriophage reduction. The comprehension of the cause why Fe²⁺ demobilizes various bacteriophages can assist in the choice of the better virus surrogates or distinguish more sensitive pathogen targets (Table 1). As illustrated in Figure 2, ferric chloride coagulation and ferrous chloride coagulation fairly divided if demobilization or physical elimination was the controlling bacteriophage destiny in EC, while adsorption to preformed flocs and EO were not crucial routes. Former works [36,37,48] have established a relationship among oxidation of Fe²⁺ and bacteriophage demobilization. For that reason, traditional coagulation with FeCl₂ was predicted to attain deactivation, while the already oxidized ferric coagulant (FeCl₃) should obtain only physical removal.

More details may be found here [35]. These researchers performed an excellent investigation on the pathways of virus removal and acceptability of bacteriophages as surrogates in potable water treatment employing Fe-EC. However, a deep study taking into account both real surface water parameters and sophisticated microscopies instruments would be very useful.

Following the current intensity as a function of applied voltage variation and the pertinent literature, three mechanisms have been proposed for acid, neutral and

alkaline pH [49-51]. For pH 2, Mechanism #1 explains Fe(OH)_{2(s)} formation; for pH 7, Mechanism #2 concerns both the varieties Fe(OH)_{2(s)} and Fe(OH)_{3(s)} production; and for pH 12, Mechanism #3 is characterized by Fe(OH)_{3(s)} apparition (Table 1). From these results, it can be seen that there is an extremely high dependence of iron species on pH in EC system.

As mentioned above, virus removal due to the fact that Fe-EC is more dominant in moderately acidic waters (~pH 6); however, real elimination is the prevalent fate of viruses in Fe-EC for pH>7 [35]. Considering Table 1, for pH 2 there is Fe(OH)_{2(s)} formation and for pH 7 the Fe(OH)_{2(s)} and Fe(OH)_{3(s)} production is considered.

For the future researches, it may be suggested here to follow pH interval one by one (pH 1; 2; etc. until pH 12) to better distinguish the pH impacts on disinfection.

EC future trends

As seen above, EC is an option to traditional coagulation, during which the coagulant is given by dissolving sacrificial electrodes under an applied electric field [52]. The easiness of process and the secondary phenomena implying the formation of gas bubbles are the main benefits. Even if the laboratory design of an EC cell is extremely easy, its scale-up is not as simple especially for large water treatment plants. It is not frequently ready to employ tank cells with sheets of Fe and Al, and there is a necessity to employ cheap materials as sacrificial electrodes. Employing low-quality Fe or Al may enable bipolar electrode configurations to be used.

Fe mechanisms	
<i>Mechanism #1 (pH 2)</i>	<p style="text-align: center;">Anode:</p> $2\text{Fe}_{(s)} - 4e^- \rightarrow 2\text{Fe}^{2+}_{(aq)} \quad (E^\circ = +0.447 \text{ V}) \quad (1)$ $2\text{H}_2\text{O}_{(l)} - 4e^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)} \quad (E^\circ = -1.229 \text{ V}) \quad (2)$ <p style="text-align: center;">Solution:</p> $2\text{Fe}^{2+}_{(aq)} + 4\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{2(s)} \quad (3)$ <p style="text-align: center;">Cathode:</p> $8\text{H}^+_{(aq)} + 8e^- \rightarrow 4\text{H}_{2(g)} \quad (E^\circ = 0.000 \text{ V}) \quad (4)$ <p style="text-align: center;">Total:</p> $2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow \text{O}_{2(g)} + 4\text{H}_{2(g)} + 2\text{Fe}(\text{OH})_{2(s)} \quad (5)$
<i>Mechanism #2 (pH 7)</i>	<p style="text-align: center;">Anode:</p> $2\text{Fe}_{(s)} - 4e^- \rightarrow 2\text{Fe}^{2+}_{(aq)} \quad (E^\circ = +0.447 \text{ V}) \quad (1)$ $\text{Fe}^{2+}_{(aq)} - e^- \rightarrow \text{Fe}^{3+}_{(aq)} \quad (E^\circ = -0.771 \text{ V}) \quad (6)$ $\text{Fe}_{(s)} - 3e^- \rightarrow \text{Fe}^{3+}_{(aq)} \quad (E^\circ = +0.037 \text{ V}) \quad (7)$ <p style="text-align: center;">Solution:</p> $\text{Fe}^{2+}_{(aq)} + 2\text{OH}^-_{(aq)} \rightarrow \text{Fe}(\text{OH})_{2(s)} \quad (3)$ $2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)} \quad (8)$ <p style="text-align: center;">Cathode:</p> $8\text{H}_2\text{O}_{(l)} + 8e^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)} \quad (E^\circ = -0.828 \text{ V}) \quad (9)$ <p style="text-align: center;">Total:</p> $3\text{Fe}_{(s)} + 8\text{H}_2\text{O}_{(l)} \rightarrow \text{Fe}(\text{OH})_{2(s)} + 2\text{Fe}(\text{OH})_{3(s)} + 4\text{H}_{2(g)} \quad (10)$
<i>Mechanism #3 (pH 12)</i>	<p style="text-align: center;">Anode:</p> $2\text{Fe}_{(s)} - 6e^- \rightarrow 2\text{Fe}^{3+}_{(aq)} \quad (E^\circ = +0.037 \text{ V}) \quad (7)$ <p style="text-align: center;">Solution:</p> $2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)} \quad (8)$ <p style="text-align: center;">Cathode:</p> $6\text{H}_2\text{O}_{(l)} + 6e^- \rightarrow 3\text{H}_{2(g)} + 6\text{OH}^-_{(aq)} \quad (E^\circ = -0.828 \text{ V}) \quad (9)$ <p style="text-align: center;">Total:</p> $2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)} + 3\text{H}_{2(g)} \quad (11)$
Al mechanism	
<i>Mechanism (pH 7)</i>	<p style="text-align: center;">Anode:</p> $\text{Al}_{(s)} - 3e^- \rightarrow \text{Al}^{3+}_{(aq)} \quad (E^\circ = +1.66 \text{ V}) \quad (12)$ $2\text{H}_2\text{O}_{(l)} - 4e^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)} \quad (E^\circ = -1.229 \text{ V}) \quad (2)$ <p style="text-align: center;">Solution:</p> $\text{Al}^{3+}_{(aq)} + 3\text{OH}^-_{(aq)} \rightarrow \text{Al}(\text{OH})_{3(s)} \quad (12)$ $\text{Al}(\text{OH})_{4(aq)}^- \rightarrow \text{OH}^-_{(aq)} + \text{Al}(\text{OH})_{3(s)} \quad (13)$ <p style="text-align: center;">Cathode:</p> $4\text{H}_2\text{O}_{(l)} + 4e^- \rightarrow 2\text{H}_{2(g)} + 4\text{OH}^-_{(aq)} \quad (E^\circ = -0.828 \text{ V}) \quad (9)$ $\text{Al}_{(s)} + 4\text{OH}^-_{(aq)} - 3e^- \rightarrow \text{Al}(\text{OH})_{4(aq)}^- \quad (14)$ <p style="text-align: center;">Total:</p> $2\text{Al}_{(s)} + 8\text{H}_2\text{O}_{(l)} \rightarrow 5\text{H}_{2(g)} + 2\text{Al}(\text{OH})_{3(s)} + \text{O}_{2(g)} \quad (15)$

Table 1: EC mechanisms using Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [49-51].

Integrating EC with free radical assisted processes (e.g., EO) remains an encouraging method to promote its implantation at full scale [52].

Conclusions

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

Few investigations have evaluated EC and EO one by one for virus removal, and Heffron et al. [4] have studied an EC-EO method for virus removal. They determined a foundation for employing a fresh, consecutive EC-EO treatment train for potable water and minutely assessed treatment efficiency for two bacteriophages and a human waterborne virus. Though their EC-EO treatment device suggested was not helpful in all water matrices, the enhanced virus removal obtained by EC-EO in model surface waters attracts additional interest. The advantage of EC-EO was possibly not due to Fe improved oxidation. Rather, bigger virus removal detected in the EC-EO treatment train was probably attained

by the additive impacts of physical elimination through coagulation/filtration, ferrous iron-based disinfection, and EO disinfection.

Integrating EC with free radical assisted processes (e.g., EO) remains an encouraging method to promote its implantation at full scale. This due to the fact that the contribution of each process is complementary to the other one.

More research concerning both virus removal and pathogenic bacteria remains to be performed in the perspective of a large industrial application of EC process.

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