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Electrocoagulation and Electrooxidation for Disinfecting Water: New Breakthroughs and Implied Mechanisms

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

Email address:
djamel_andalus@hotmail.com

Abstract: Electrochemical technology may present an option of treating water since they have proved some benefits over conventional techniques, like decreased handling and storage of chemicals and cost-effectiveness. Consecutive electrochemical techniques have yet to be tried for removing *Escherichia coli* in potable waters. In this review, a brief discussion of the work of Lynn [1] is presented. Lynn [1] studied electrocoagulation (EC) and electrooxidation (EO) employing two model surface waters and two model groundwaters to define the performance of consecutive EC-EO for removing *E. coli*. At a current density of 1.67 mA/cm^2 for 1 min, bench-scale EO alone attained 4-logs reduction of *E. coli* in the model shallow aquifer. Elevating the EO current density to 6.67 mA/cm^2 for 1 min presented similar levels of *E. coli* reduction in the model deep aquifer. Employing a current density of 10 mA/cm^2 for 5 min EC attained 1-log or bigger *E. coli* removal in all model waters. No supplementary reduction beyond EC alone was reached employing consecutive EC-EO. Diminutions in the initial pH of the surface waters in order to reach bigger natural organic matter elimination did not improve *E. coli* cells killing with EC-EO comparatively with EC alone. De facto, around 64% of NOM was eliminated regardless of the variation in pH, which possibly restricted *E. coli* removal. More causes for the shortage of enhancement in *E. coli* disinfection may have comprised the existence of iron after EC or deficient EO current density. Diminishing the initial water pH did enhance *E. coli* reduction employing EO when pretreated via EC compared to the baseline water matrix pH. Despite the breakthroughs obtained throughout the Lynn [1] research in both EC and EO processes for disinfecting water in terms of mechanisms and optimization, great research remains to be accomplished with a view to largely accept these electrochemical techniques in the water treatment industry. Finding the correct hybridization and appropriate combination of such methods, and probably introduce other physical techniques like adsorption and magnetic treatment, would open large perspectives in implementing electrochemical engineering in water treatment.

Keywords: Electrocoagulation (EC), Electrooxidation (EO), Disinfection, Electric Field, Electrodes, Microorganisms

1. Introduction

Electrochemical techniques can present many benefits over traditional water treatment [1, 2], rendering those especially useful to small water treatment devices [1, 3]. Benefits comprise avoiding corrosive chemical products, small footprint, no alkalinity depletion, straightforward operation and automation, and portability for water treatment during emergencies and in remote settings [4, 5]. Two frequent electrochemical methods are electrocoagulation (EC) and electrooxidation (EO), which both provide viable removal (comprising both physical elimination and/or demobilization routes) of a collection of pollutants comprising natural organic matter (NOM, the main disinfection by-product (DBP) precursor) and microorganisms such as *Escherichia coli* [6, 7].

EC uses DC power to generate *in situ* coagulants employing corrodible metal electrodes, usually iron [8] or aluminum [1, 9]. Liberating metal ions produces metal hydroxide flocs [10], which may thereafter be physically removed from water employing flotation, sedimentation, or filtration [11, 12]. EC has shown elimination of microbes such as *E. coli* through the production of *in situ* coagulants, which posteriorly flocculate with microorganisms and may be filtered from water [13-16].

Researchers [15] mentioned the elimination of 2-4 logs *E. coli* employing EC with Fe electrodes, with more important reduction as coagulant dose augmented or pH was adjusted. Moreover, EC may eliminate NOM from water, that way decreasing the generation of toxic DBPs [4, 17, 18]. With a view to more increase NOM elimination, enhanced coagulation employing elevated coagulant dose or diminished initial pH may assist [19-21].

EO utilizes DC power and non-reactive electrodes, like mixed metal oxides (MMO) and boron-doped diamond (BDD), to remove contaminants, directly or indirectly via producing oxidants in water [22]. Following the water matrix or electrode kind, EO may oxidize chloride to produce free chlorine species. Employing convenient electrodes, EO possessed the potential of generating reactive oxygen species like hydroxyl radicals [23]. Demobilizing microorganisms, such as *E. coli*, may happen in EO application during reactions with the formed oxidants in solution [24]. Identically to classical disinfection methods, EO will induce DBPs generation if the oxidants react with NOM [24, 25]. If EO was viewed as an encouraging method, the existence of DBP precursors proves the necessity for a pretreatment technique to eliminate NOM [1].

During the last three decades, many scientists have largely tested EC and EO remediation for both potable water and wastewater [13-15, 26-28]. Several investigations surprisingly integrated electrochemical techniques for dealing with industrial and urban wastewaters [26, 29, 30]. Nevertheless, Lynn [1] focused on consecutive EC-EO with a view to deal with *E. coli* in potable water. To attain such objective, Lynn [1] studied the performance of consecutive EC-EO for removing *E. coli* in different potable water matrices. Firstly, Lynn [1] aimed to prove *E. coli* reduction employing consecutive EC-EO to deal with a synthetic surface and groundwater matrices. It was supposed that *E. coli* removal in the surface water would be promoted utilizing EC-EO since surface waters carry NOM, the oxidant demand of which may overlap with killing microorganisms' technologies. EC was expected to eliminate an elevated level of NOM (that way decreasing oxidant demand), so elevating *E. coli* removal via EO. Secondly, Lynn [1] focused on the usage of enhanced EC (employing pH adjustment) as a pretreatment before EO for removing *E. coli*. A lower initial pH was supposed to augment NOM reduction via EC and then promote killing microbes' via producing a higher portion of free chlorine in the more efficient HOCl form throughout EO [1]. The iron injected throughout EC was envisaged to destroy oxidants like free chlorine throughout EO, but also improve *E. coli* removal by Fenton-like reactions.

2. Traditional Treatment

A traditional potable water treatment plant usually contains grit screening, coagulation, flocculation, sedimentation, granular filtration, and disinfection [31, 32]. As each unit process may decrease pollutants, a plurality of *E. coli* reduction frequently happens throughout killing microorganisms' stage [33]. As an illustration, it is well established that chlorination demobilizes *E. coli* [1].

According to the US EPA's National Primary Drinking Water Regulations, a facility cannot have more than 5% positive total coliform samples in the treated effluent per month [34]. With a view to satisfying this demand, a *Ct* level (Concentration of disinfectant × contact time) is employed to confirm that the objective degree for microorganisms' killing is obtained. Usually, *Ct* levels correspond to a definite area of microorganisms' demobilization following water key parameters such as temperature and pH. As an illustration, a *Ct* of 15 mg-min/L free chlorine conducts to approximately 4-logs *E. coli* demobilization at pH 7 and 22°C [1].

More than the aimed demobilization of microbes, water treatment technology's scheme and running have as well to take into account the generation of DBPs. The US EPA Enhanced Coagulation Guidance Manual [35] explained how coagulation techniques might be ameliorated to diminish DBP precursors like NOM before disinfectants injection [18, 36]. Enhanced coagulation fixes on bigger NOM elimination through either augmenting the coagulant dose or diminishing the water's pH [35, 37, 38]. Solids elimination methods like coagulation may as well reduce many portions of microbes [39]. As an illustration, an average of 2 logs reduction of *E. coli* may be attained throughout coagulation/flocculation/sedimentation upon optimized parameters with an iron-based coagulant [1].

Conventional treatment techniques are suitable to eliminate a range of pollutants from potable waters; nonetheless, traditional handling can possess hazards and restrictions [40]. Disinfecting using chlorine has an effect on the odor and taste of the water as well as injecting corrosive chemical products, like sodium hypochlorite, which may be poisonous to transport, handle, and store [41,42]. Disinfecting employing chlorine is as well less performant versus chlorine-resistant microorganisms, like *Cryptosporidium* [1]. Coagulation as well possesses many restrictions. Injecting a chemical coagulant may affect global water quality through consuming alkalinity, that way diminishing buffering potential, which may conduct to more introduced chemical agents in downstream treatment. The sulfates and chlorides injected with ferric or aluminum coagulants may as well provoke corrosion downstream [17].

3. Electrochemical Treatment Techniques

Electrochemical technology concerns the usage of particular electrode material conceived to generate *in situ* ions in the water viewing physical-chemical pollutant elimination [43, 44]. Electrochemical techniques, like EC and EO, possess diverse benefits over classical technology [45, 46]. The EC method does not necessitate the processing and storage of harmful chemical agents [47, 48]. Additional benefits comprise no alkalinity utilization [48, 49], easy availability during emergencies [50, 51], and lower production of DBPs compared to chlorination [52, 53]. In addition, the electrochemical application may be more cost effective than

conventional treatment, rendering such methods useful to small drinking water systems [54, 55]. Nonetheless, the augmented electricity request by electrochemical industries has to be taken into account [1].

In terms of electrochemical technology performance, the current density is viewed as the main parameter [1]. It is described as the current exercised above the submerged surface area of the electrode [56, 57]. The device scheme may as well affect running following the number of electrodes or the device's form [57, 58]. Concentrations of electrolytes such as chloride [43] in the solution have great contributions in if or not the technique is performant, like deciding which oxidants are probably to produce, e.g., free chlorine or hydroxyl radicals [22, 41].

An additional fundamental indicator of electrochemical technology is the electrode material, which dictates the species formed [1]. EC employs Fe or Al electrodes to produce species identical to that of chemical coagulation [59, 60]. On the other hand, EO electrode materials are chosen following the oxidants necessitated for killing microorganisms. As an illustration, MMO boost bigger free chlorine production, whilst BDD are utilized for more important reactive oxygen species formation [22, 41].

3.1. Electrooxidation (EO)

As an electrochemical technology, EO is employed for killing pathogens' purposes [5, 27]. EO possesses many benefits comparatively with classical chlorine-founded disinfection, comprising that it is readily worked, environmentally friendly, and cost-effective [23]. Throughout EO application, diverse biological and chemical pollutants may be removed by direct and indirect oxidation procedures [23, 28]. In addition, generating DBPs is importantly diminished during EO application as it usually forms a lower free chlorine concentration than utilized in classical disinfection methods [1, 24, 28].

EO acts throughout two major pathways in removing microorganisms: direct and indirect oxidation [41, 61]. Direct oxidation happens if water adsorbs to the anode surface and is oxidized to produce hydroxyl radicals [61]. The hydroxyl radicals will directly oxidize pollutants in contact with the electrode surface [61]. Indirect oxidation takes place based on electrolyzing water at the anode and cathode [1], as shown via Eqs. (1) and (2), respectively:

$$2H_2O_{(l)} \rightarrow O_{2(g)} + 4H^+_{(aq)} + 4e^- \qquad (1)$$

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \qquad (2)$$
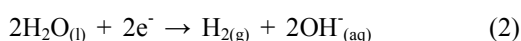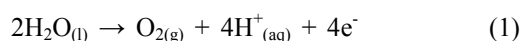
It is well established that a secondary electrolysis process during EO is the oxidation of chloride to produce free chlorine, a frequent killing agent [23, 41]. Free chlorine formation (hypochlorous acid or hypochlorite ion, based on pH) presents an identical demobilization route to conventional disinfection [7, 27]. Different oxidants, like ozone or hydrogen peroxide, may as well be formed [27]. Nonetheless, levels of such oxidants are less important, and they, consequently, are not viewed as main actors in killing pathogens comparatively with

free chlorine and hydroxyl radicals [1, 27].

A different indirect pollutant removal route in EO is the formation of hydroxyl radical, particularly in waters without chloride electrolytes [23, 41]. Hydroxyl radicals are very performant in fragmenting organic molecules and microbes [1, 23].

An additional probable indirect oxidation route is Fenton's reaction, which is the oxidation of ferrous iron with hydrogen peroxide at a low pH (< 4), conducting to the formation of hydroxyl radicals [26, 45]. Nevertheless, the introduced contaminant of metallic salt renders this a non-ideal reaction [1].

Demobilizing *E. coli* via EO may be realized employing either MMO [27, 28] or BDD [27] anodes. The principal parameters that affect the removal of *E. coli* comprise current density, water features, and electrode material [1]. Diverse studies have proved that bigger current densities correspond with elevated results of *E. coli* reduction [23, 27]. Researchers [28] employed MMO platinum (Pt) electrodes to assess the effects of particular electrolyte on *E. coli* demobilization and established that chloride possesses a fundamental contribution in killing microbes. Demobilizing *E. coli* routes are imposed by anode type and supporting electrolytes existing in water [27]. Chloride is usually present in most waters and several studies have shown MMO Ir/O2 electrodes possessing the most important chlorine rate production, rendering them the most efficient electrode for killing microorganisms using free chlorine [27, 41, 62].

On the other hand, it is well-established that EO may form DBPs during oxidation reactions with the NOM present water [24, 41, 63, 64]. It was observed that prolonged treatment periods or elevated currents were needed to obtain the elimination of NOM [64]. As a result, producing DBPs during EO dictates that some pretreatment would be necessary to diminish NOM in the water before EO.

3.2. Electrocoagulation (EC)

Even if EC is identical to the conventional coagulation [65, 66], it possesses several benefits over her. Chemical coagulants such as aluminum sulfate (alum) and ferric chloride induce several problems in treatment plants [67, 68]. As an illustration, traditional coagulation may diminish pH and may waste alkalinity, which conducts to more chemical introduction to re-equilibrate a neutral pH before distribution [69]. On the other hand, EC does not destroy alkalinity [1].

An additional problem with conventional coagulation is treating the sludge waste. Usually, EC usually deals with lower sludge generation [17, 70]. In addition, EC references propose that it can more efficiently eliminate smaller colloids comparatively with classical methods because of electrophoretic mobility [65]. This may comprise dissolved species such as hydrophilic acids (a portion of NOM), which are hard to reduce chemically [17, 71]. Eliminating hydrophilic acid may take place if the pH is acidic, promoting precipitation and charge neutralization [39, 72]. In addition, EC forms hydrogen gas on the cathode, which provokes electroflotation via pushing flocs to rise to the surface [1, 46,

66].

Identically to classical coagulation, throughout EC both chemical and physical reduction routes are involved in solids elimination. The main gap is the *in situ* injection of the coagulant metal dose throughout EC [41]. The concentration introduced, C (g/L) (Eq. (3)), may be determined following Faraday's law (Eq. (4)), which assesses the injection of metal coagulant ion (M^+ [Al^{3+}/Fe^{2+}]) introduced to the water accordingly to a defined current and time [1, 57]:

$$C = \frac{m}{V} \qquad (3)$$

$$m = \frac{I \times t \times M_w}{z \times F} \qquad (4)$$

where m is the mass of liberated metal (g), I is the electric current intensity (A), t is the residence time (s), M_w is the molecular weight of the metal, z is the number of electrons (e.g., in this case, $Fe^{2+} = 2$), F is the Faraday's constant (96,485 C/mol), and V is the volume of the treated water (L).

Figure 1 illustrates diverse iron and aluminum species that may be generated following the pH and metal concentration. Identically to chemical coagulation method, the M^+ ions will react with the hydroxyl to produce different polymeric hydroxide complexes [41, 73]. As a function of pH, the polymeric hydroxides will react with negatively charged molecules and solids, such as dissolved and particulate NOM and *E. coli* cells, via charge neutralization [11, 13]. Metal hydroxide precipitate may as well be in a flash formed, inducing agglomeration of smaller solids throughout differential settling flocculation [15]. Following the pH diminution, charge neutralization will possess a more significant contribution, particularly at lower coagulant doses [1].
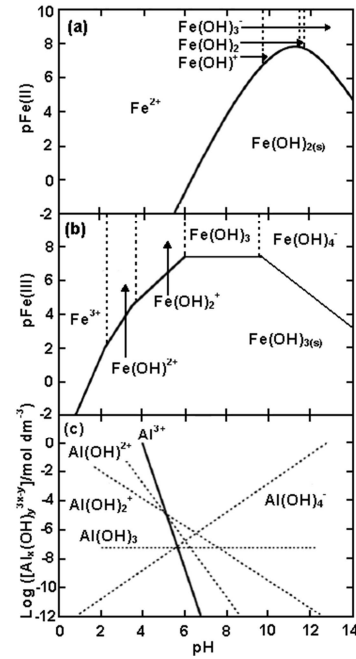
Figure 1. *Predominance-zone diagrams for (a) Fe(II) and (b) Fe(III) chemical species in aqueous solution. The straight lines represent the solubility equilibrium for insoluble Fe(OH)₂ and Fe(OH)₃, respectively, and the dotted lines represent the predominance limits between soluble chemical species [41]. (c) Diagram of solubility of Al(III) species as a function of pH [73].*

Table 1 lists the EC mechanisms employing Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [22, 55, 70]. At the same time, the cathode liberates hydrogen gas and hydroxide ions, which participates in augmenting pH during EC application [41]. As a result, the formed flocs will be eliminated by either electroflotation or further flocculation and settling [41]. Flocculation pathways (such as diffusion, advection, or differential settling) let EC eliminate diverse solids from water comprising microorganisms, NOM, and inorganics [13-15].

Table 1. *EC mechanisms using Fe (pH 2, 7 and 12) and Al (pH 7) electrodes [22, 55, 70].*

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

enhanced *E. coli* removal were an elevated final pH, consumption of free chlorine by iron, or insufficient applied EO current density [1].

More study employing enhanced EC-EO with pH adjustment to treat surface waters proved that a lower pH for EC-EO somewhat elevate *E. coli* reduction, but not important. Little ameliorations in removing NOM at pH 6 or below were observed for both surface waters; however, the residual NOM would scavenge oxidants, that way restricting enhancements in *E. coli* removal via EC-EO. As more optimization of NOM reduction is necessitated, the EEO for *E. coli* reduction employing each method augmented if the initial pH was below 6, illustrating the advantage of pH adjustment [1].

Diminishing the initial pH augmented EC total remaining iron levels, which would as well ingest free chlorine [1]. Nevertheless, the total remaining iron levels following EC-EO diminished, probably because of oxidation of ferrous to ferric hydroxides (Table 1, Mechanism #2) which presented more performant floc settling. Oxidizing ferrous iron may as well provoke Fenton's reaction. A little linkage was observed among EC total remaining iron levels and *E. coli* reduction throughout EO (after EC pre-stage). Ferrous iron was as well transformed into ferric throughout EO, proving that Fenton-like reactions may have happened, conducting to the augmented *E. coli* removal. During the time that diminished initial pH ameliorated *E. coli* reduction for EO after EC, the gap was not crucial for any situation. This shows that the EO current density was too weak to produce the oxidants needed for augmented *E. coli* removal.

4. Conclusions

From this work, the following conclusions can be drawn:

Lynn [1] proved that EC-EO did not enhance *E. coli* reduction further away than EC single. Coming technique regulation can fruit enhancements. As an illustration, regulating the iron injection for NOM elimination would restrict the effect of oxidant scavengers. In addition, a more efficient filtration method like granular filtration would diminish NOM and total iron levels in EO influent, which would reduce consumption of free chlorine because of such constituents. More elevated EO current densities have to be exercised to the consecutive remediation to improve the reduction of *E. coli*. Enhanced *E. coli* elimination would be anticipated following more important oxidant production depending on current density.

After regulating the setup, more research is required to describe the particular routes of *E. coli* removal in potable waters throughout EO if elevated levels of iron are existing. This needs taking into account iron speciation and electrostatic interactions among the iron and microorganisms [1].

Despite the breakthroughs obtained throughout the Lynn [1] research in both EC and EO processes for disinfecting water in terms of mechanisms and optimization, great research remains to be accomplished with a view to largely accept these electrochemical techniques in the water treatment industry. Finding the correct hybridization and appropriate combination of such methods, and probably introduce other physical techniques like adsorption and magnetic treatment [74, 75], would open large perspectives in implementing electrochemical engineering in water treatment.

Abbreviations

BDD: Boron-doped diamond
DBP: Disinfection by-product
EC: Electrocoagulation
EO: Electrooxidation
MMO: Mixed metal oxides
NOM: Natural organic matter

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