



Electrocoagulation as a Pretreatment of Electrooxidation for Killing *Escherichia coli*

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Abstract

Although the literature mainly reports on the inactivation of bacteria by various electrochemical disinfectants, the impact of process variables and reactor design on bactericidal performance is not fully understood. This review concentrates on recent achievements of electrocoagulation (EC) and electrooxidation (EO) in killing pathogens such as *Escherichia coli*. Lynn *et al.* [1] [2] showed that in addition to EC alone, EC-EO enhanced *E. coli* reduction only after pH adjustment. They proposed that additional process optimization may lead to further improvements, such as adjusting the iron dosage for natural organic matter (NOM) removal, which would limit the effectiveness of oxidant scavengers. Additionally, more efficient filtration techniques (e.g., granular filtration) will reduce NOM and total iron content in the EO feed-water, decreasing the need for oxidants. Furthermore, continuous EC-EO treatment requires more elevated EO current densities to improve *E. coli* removal. Investigating the pathways of demobilizing *E. coli* in drinking water at high iron concentrations in the EO range will also provide deep insights into ongoing setup design. This review provides crucial, reliable, safe, and versatile alternatives to the widespread trouble of human drinking water pollution. Using and propagating the EC-EO technique will diminish health risks related to water quality, economic burden, lost labor time, import washout to the national economy, and natural resource management. Commercial-scale deployment of EC-EO technology will undoubtedly increase the socio-economic burden on local communities via secured water supply and result in a reduction in government health expenditures.

Subject Areas

Chemical Engineering & Technology

Keywords

Pathogens, Chemical Coagulation (CC), Electrocoagulation (EC), Electrophoretic Mobility, Natural Organic Matter (NOM)

1. Introduction

Potable water is critical to prevent significant outbreaks of microorganisms and chemicals in water that can lead to illness and death [1] [2] [3]. Potable water treatment factories help safeguard humans by reducing microbes and viruses in water [4] [5] [6] as well as essential pollutants such as disinfection by-products (DBPs) [7] [8]. Public potable water treatment factories must comply with wastewater standards set by the United States Environmental Protection Agency (US EPA), such as the Safe Drinking Water Act (SDWA) of 1974. More identified regulations are derived from the SDWA, like the Total Coliform Regulation (TCR) of 1990 (revised in 2013) and the Disinfectants and Disinfection By-products Regulation (DBPR) of 1996. These regulations are intended to reduce levels of chemical and microbiological pollutants (e.g., DBPs and *Escherichia coli*) in drinking water. Since *E. coli* is found in the digestive tract of mammals, it is an indicator of fecal contamination. As mentioned in TCR, it can also be detected quickly.

Compared with traditional water treatment technology, electrochemical technology has various benefits and is especially suitable for small water treatment plants [1] [9] [10]. Advantages include the absence of corrosive chemicals, small footprint, no alkalinity consumption, ease of operation and automation, and portability for water treatment in emergency situations and remote locations [11]. Benefits comprise the absence of corrosive chemicals, small footprint, no alkali consumption, ease of application and automation, and portability for water treatment in emergency situations and remote locations [11]. Two frequent electrochemical methods are electrocoagulation (EC) and electrooxidation (EO). They can potentially mitigate (including physical elimination and demobilization routes) various pollutants, comprising natural organic matter (NOM) and microorganisms such as *E. coli*.

EC uses direct current (DC) to generate coagulant *in-situ* employing corrodable metal electrodes (usually Fe or Al) [12] [13]. Liberating metal ions produces hydroxide flocs that can be physically removed from the solution by flotation (known as electroflotation, EF [14] [15]), sedimentation, or filtration [16]. EC has been shown to kill microorganisms like *E. coli* through the *in-situ* generation of metal cations, which can then flocculate with the microorganisms and be filtered out of suspension [17] [18] [19] [20]. Delaire *et al.* [19] reported 2-4 log magnitude removal of *E. coli* employing EC with Fe plaques, with greater attenuation when increasing coagulant dosage or adjusting pH. Furthermore, EC can remove NOM from water, thus reducing the generation of poisonous DBPs [11]

[21] [22]. To further improve NOM removal, it may be helpful to enhance EC as in chemical coagulation (CC) by augmenting the coagulant dosage or lowering the initial pH [1] [16].

EO employs DC power and non-reactive electrodes, like mixed metal oxides (MMOs) and boron-doped diamond (BDD), to reduce contaminants directly or indirectly by producing oxidants in the solution [23] [24]. Following the water matrix or electrode nature, EO may oxidize Cl^- to form free chlorine species [25] [26]. Consuming suitable electrodes, EO could also generate reactive oxygen species (ROSs) like hydroxyl radicals ($\cdot\text{OH}$) [27] [28] [29]. Demobilizing bacteria, like *E. coli*, could take place during EO usage via reactions with the generated oxidants in water [28] [30] [31] [32]. Like conventional disinfection processes [33], EO will produce DBPs when the oxidants and NOM enter in interactions [25]. Even if EO remains a promising technique, the occurrence of DBP precursors needs pretreatment to retain NOM [1] [34] [35].

Several scientists have examined EC and EO techniques for drinking potable water and used water [17] [18] [19] [36] [37] [38], and combined such methods for treating industrial and domestic used waters [36] [39] [40]. Nonetheless, killing *E. coli* in potable water by continuous EC-EO has not until now been tried.

In this context, Lynn [1] examined the continuous EC-EO efficiency for reducing *E. coli* in synthetic surface and groundwater matrices. Lynn [1] suggested that the use of EC-EO could reduce *E. coli* populations in surface waters that contain NOM and whose oxidant requirements may interfere with disinfection methods. EC is expected to reduce high NOM concentrations (thus reducing the need for oxidants), thus improving the elimination of *E. coli* by EO. Also, Lynn [1] concentrated on using enhanced EC (by pH control) as a preliminary stage to EO to retain *E. coli* since a basic pH was suggested to increase NOM removal using EC and ameliorate disinfection by yielding a more significant part of free chlorine in the more effective HOCl form during EO. Fe liberated during EC was anticipated to consume oxidants (e.g., free chlorine) during EO and ameliorate *E. coli* retention via Fenton-like phenomena.

This review provides crucial, reliable, safe, and versatile alternatives to the widespread trouble of human drinking water pollution. Using and propagating the EC-EO technique will diminish health risks related to water quality, economic burden, lost labor time, import washout to the national economy, and natural resource management. Section 2 overviews conventional and electrochemical treatments for killing *E. coli*, focusing on EO and EC. Section 3 evaluates the alternative disinfection technologies, such as O_3 and UV. Sections 4 and 5 give insights into the EO and EC pathways for killing *E. coli*, respectively. Finally, the Conclusion lists the main points drawn from this review.

2. Conventional and Electrochemical Treatments for Killing *Escherichia coli*

Potable water plants aim to produce potable water by eliminating pollutants. In

this regard, the maximum contaminant level (MCL) goal for total coliforms comprising *E. coli* is zero. Microorganisms, e.g., *E. coli* O157, and chemicals in public waters have sometimes conducted to illness or death worldwide [1].

The purpose of a drinking water system is to produce drinking water by removing contaminants. In this regard, the maximum contaminant level (MCL) for total coliforms, including *E. coli*, is zero. Pathogens, such as *E. coli* O157, and chemicals in public waters sometimes cause illness or death worldwide [1].

2.1. Conventional Treatment

A conventional drinking water treatment factory could involve grit screening, CC, flocculation, sedimentation, granular filtration, and disinfection [41] [42]. Even if every separation stage could decrease pollutants, the main part of *E. coli* retention frequently occurs throughout disinfection. As an illustration, chlorination is well known to kill *E. coli* [41] [43] [44]. A plant could not have more than 5% positive total coliform samples in the treated effluent per month [1]. To satisfy such a need, a *Ct* level (Concentration of disinfectant (mg/L) × residence period (min)) is employed to guarantee that the wanted degree for microorganisms' demobilization is reached. Following water's temperature and pH, *Ct* values correlate to some level with microbial inactivation. For example, a *Ct* of 15 mg·min/L for free chlorine leads to a ~ 4-logs *E. coli* inactivation at pH 7 and 22°C [1] [45].

The temperature and pH of the suspension can also be considered when estimating *Ct* [44]. To illustrate this, we created a mathematical model to predict the inactivation of *Giardia lamblia* cysts following chlorine injection [46] [47]:

$$Ct = 0.9847C^{0.1758} pH^{2.7519} T^{-0.1467} \quad (1)$$

where: *C* = chlorine injection ($C \leq 4.23$ mg/L), *t* = time to demobilize 99.99% of the cysts, pH range is 6 - 8, and *T* = temperature range is 0.5°C - 5.0°C.

Also, **Table 1** gives the *Ct* value of a segment of microorganisms [48]. Resistance to chlorine occurs in the following order: protozoan cysts > viruses > non-sporulating bacteria [46].

Table 1. Inactivation of microorganisms by chlorine: *Ct* level (T = 5°C; pH = 6.0) [48].

| Microorganism | Chlorine concentration (mg/L) | Deactivation period (min) | <i>Ct</i> |
|------------------------------------|-------------------------------|---------------------------|-----------|
| <i>E. coli</i> | 0.1 | 0.4 | 0.04 |
| Poliovirus I | 1.0 | 1.7 | 1.7 |
| <i>Entamoeba histolytica</i> cysts | 5.0 | 18 | 90 |
| | 1.0 | 50 | 50 |
| <i>G. lamblia</i> cysts | 2.0 | 40 | 80 |
| | 2.5 | 100 | 250 |
| <i>G. muris</i> cysts | 2.5 | 100 | 250 |

Also, designing and operating water treatment plants should focus on forming DBPs [49]. Literature elaborated on how coagulation technology [50] could lessen DBP precursors (like NOM) before disinfection [16] [41] [51]. The purpose of enhanced coagulation is to achieve a greater removal of NOM by increasing the coagulant dosage or lowering the pH of the water [52]. Particle removal techniques (e.g., CC) could retain some microorganisms. The use of iron-based coagulants reduces *E. coli* by an average of 2 logs during CC/flocculation/decantation processes [1].

Conventional treatment techniques remain suitable for retaining some pollutants from potable water but have hazards and restrictions [1]. Killing pathogens with chlorine touches the odor and taste of the water and adds corrosive chemicals such as sodium hypochlorite, which is hazardous for transportation, handling, and storage [25] [45] [53] [54] [55]. Moreover, demobilizing pathogens with chlorine is less performant against chlorine-resistant microbes [56] like *Cryptosporidium* [45] [57] [58] [59].

CC is also restricted as introducing a coagulant could affect overall water quality by depleting alkalinity, reducing buffering potential, and leading to more introduced chemicals in downstream treatment [60] [61] [62]. Sulfates and chlorides added with Fe or Al salts could also cause downstream corrosion [1] [52] [63].

2.2. Electrochemical Treatment

The electrochemical treatment utilizes certain electrode material to produce *in-situ* ions in the solution, aiming for physicochemical pollutants elimination [25] [55] [64] [65]. Electrochemical techniques (e.g., EC and EO) are more advantageous than traditional processes. The EC does not need handling and storage of dangerous chemical products [14] [66] [67], no alkalinity consumption [1] [63], is easy to use during emergencies [11] [68], and has lower DBPs formation relatively to chlorination [38] [69]. Furthermore, electrochemical treatment could be more cost-effective than traditional treatment (Table 2) [70], meaning that such methods are helpful for small potable water setups [11] [55] [71] [72]. Nonetheless, the elevated electricity demand by electrochemical technology should be considered [1].

The current density (CD) mainly contributes to the electrochemical process performance [1]. The CD is the electric current applied over the electrode's submerged surface area. It determines the product's generation rate and the electrode performance [25] [73] [74]. Designing the reactor influences functionality following the electrodes' number and the reactor's shape [25] [73]. The level of electrolytes (such as chloride) in the water, known in technical terms as supporting electrolytes [26], is critical in determining whether the technology will thrive, especially when determining which oxidants (e.g., free chlorine or $\cdot\text{OH}$) may be formed [25] [29] [38].

Another critical aspect of electrochemical process is the electrode material,

Table 2. A comparison of disinfection technologies operating costs [70].

| Disinfection method | Cost (£/m ³) |
|--|--------------------------|
| Chlorine | 0.0022 - 0.01 |
| Ozone | 0.01 - 0.06 |
| Electrochemically generated mixed oxidants | 0.001 - 0.024 |
| Medium pressure ultraviolet (UV) | 0.0019 |
| Electrocoagulation (EC, electrochemical) | 0.014 |
| Photodynamic disinfection | 0.0256 |
| Pulsed electric fields | 0.0266 |
| Advanced low-pressure UV | 0.204 |
| Titanium dioxide photocatalysis | 0.26 |
| Irradiation | 0.38 |
| Solar-titanium dioxide photocatalysis | 0.45 |
| Hydrodynamic cavitation | 0.85 |

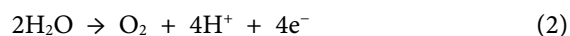
which determines the products generated [1]. EC uses iron or aluminum electrodes to create the same products as CC [73] [75] [76]. Also, EO electrode materials are selected based on the oxidant required for disinfection (e.g., MMO) promotes higher free chlorine production, while BDD is employed for higher ROS production [25] [28] [55].

2.2.1. Electrooxidation (EO)

EO is a disinfection technique employed in treating water electrochemically [31] [55]. For small potable water setups, EO has many benefits over conventional chlorine disinfection, including ease of use, environmental protection, and cost-effectiveness [27] [31] [55]. Throughout EO, the electrolysis phenomenon could reduce numerous biological and chemical pollutants through direct oxidation (DO) and indirect oxidation (IO) methods [27] [38] [55]. Also, EO significantly reduces the generation of DBPs because it generally produces lower levels of free chlorine than traditional disinfection techniques [1] [38].

1) *Electrooxidation (EO) pathways*

In reducing waterborne pollutants by EO, two significant routes have a crucial contribution: DO and IO [25] [77]. DO happens when water is absorbed on the anode surface and is oxidized to generate $\cdot\text{OH}$ [77], which will directly oxidize contaminants in contact with the electrode surface [25] [31] [78]. IO happens after water electrolysis at the anode and cathode [1], as defined by Equations (2) and (3), respectively:



EO has a well-known secondary electrolysis phenomenon: the oxidation of Cl^- to generate free chlorine, a famous killing agent [27] [55]. Free chlorine generation (hypochlorous acid or hypochlorite ion, following pH) constitutes a similar

killing mechanism to traditional method [1] [31]. Additional oxidants (e.g., O₃ or H₂O₂ [79]) could also be produced [37] [55]. However, concentrations of these oxidants remain lower, and they are not considered as crucial contributors to disinfection relative to free chlorine and •OH [1] [37].

Producing •OH presents an additional indirect pollutant reduction pathway for EO, particularly in water without chloride [1] [25] [27] [28] [69]. Hydroxyl radicals break down efficaciously NOM and pathogens [27] [28] [78].

A supplementary likely IO method is Fenton's reaction, which is oxidizing Fe²⁺ with H₂O₂ at an acidic pH (<4), leading to the hydroxyl radical's generation [36] [80] [81] [82]. However, the added metallic salt makes such a reaction unwanted [25] [55] [78].

2) *Electrooxidation (EO) anode*

The nature of metals or coated metals utilized as electrodes is crucial to the EO efficacy [28] [55] [77]. For EO, MMOs and BDD are the most frequent electrode kinds employed [1] [25] [83]. MMO electrodes are typically titanium (Ti)-based electrodes and can be better categorized following the metal coating on the electrode surface, such as iridium oxide (IrO₂) or ruthenium oxide (RuO₂) [28] [84]. Adding metal coating elevates the MMO electrode performance by augmenting its surface area, thus enhancing electrolysis yield and reaction kinetics [78]. The anode material dictates the critical route of the EO process [55] [77] [78]. MMO electrodes are dictated mainly by producing free chlorine, besides the coating affecting the chlorine formation rate [28]. On the other hand, BDD electrodes possibly generate ROSs [25] [85].

3) *Electrooxidation (EO) for reducing E. coli*

Demobilizing *E. coli* using EO could be attained by either MMO [28] [38] [55] [69] or BDD [28] [37] [55] anodes. Demobilizing *E. coli* is mainly affected by CD, water properties, and electrode type [1]. Higher CDs are related to elevated rates of *E. coli* inactivation [27] [28] [37] [69]. Scientists [38] utilized MMO platinum (Pt) electrodes to determine the effect of specific electrolytes on demobilizing *E. coli* inactivation. They proved that Cl⁻ has a leading role to inactivation. Most waters carry chloride, and several researchers affirmed that the MMO-Ir/O₂ electrode has the most significant Cl₂ production rate and called it the most performant electrode for killing microorganisms using Cl₂ [28] [86].

4) *Electrooxidation (EO) restriction*

EO produces DBPs via oxidation reactions with NOM present water [25] [69] [83] [85]. Retaining NOM by EO needs long application periods or high DCs to reach elimination [1] [83]. MMO and BDD electrodes form DBPs throughout EO treatment [69] [85]. Importantly, through the disinfection of *E. coli* using MMO electrodes, DBPs were formed, namely total trihalomethanes (TTHMs). Lower CDs produced lower levels of TTHMs, as anticipated, since less free chlorine was formed. Scientists [85] observed 30 mg/L chlorate formation following a 10-min residence period employing a BDD anode. Producing DBPs via EO requires some pretreatment to diminish NOM in the water before EO. As anticipated, lower CD results in lower TTHM values because less Cl₂ is formed.

Scientists [85] observed the formation of 30 mg/L of chlorate after a 10-minute residence period at the device employing BDD anode. Producing DBPs via EO requires some pretreatment to reduce NOM in the water prior EO.

2.2.2. Electrocoagulation (EC)

Identical to CC, EC has several merits over CC [68]. Chemicals (e.g., $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ and FeCl_3) provoke many troubles in treatment industry [1]. CC could reduce pH and consume alkalinity, leading to more chemical injections to reestablish water neutrality before distribution [87]; however, EC does not consume alkalinity [63] [88].

CC has additional trouble dealing with sludge, and EC usually has lower sludge creation [63] [68] [89] [90]. Relative to CC, EC (Figure 1) is also more performant in removing smaller particles thanks to electrophoretic mobility [68] [76] [89] [91] [92]. Such small particles can include dissolved compounds (like hydrophilic acids, a part of NOM, Figure 2) that are not easy to retain by CC [1] [63] [93]. Eliminating hydrophilic acid could happen if pH is acidic (Figure 3), promoting charge neutralization (CN) and precipitation [3] [94] [95] [96] [97]. Further, EC may form $\text{H}_{2(g)}$ at the cathode, inducing an EF phenomenon by pushing flocs to rise to the surface [14] [14] [68] [89].

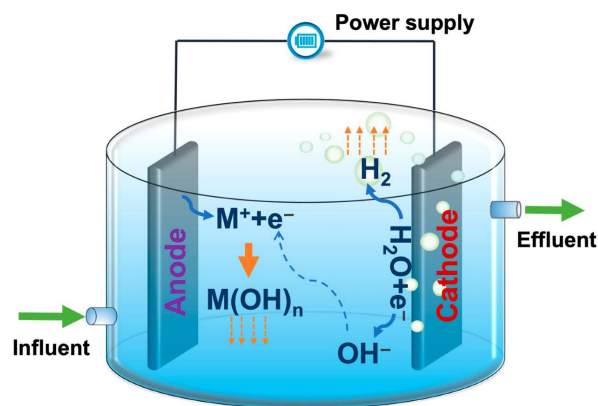


Figure 1. Schematic of the electrocoagulation (EC) technique [90].

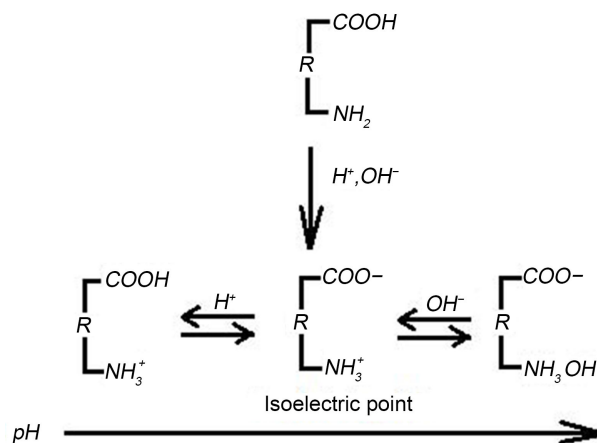


Figure 2. Principal charges of hydrocolloids depending on pH [50].

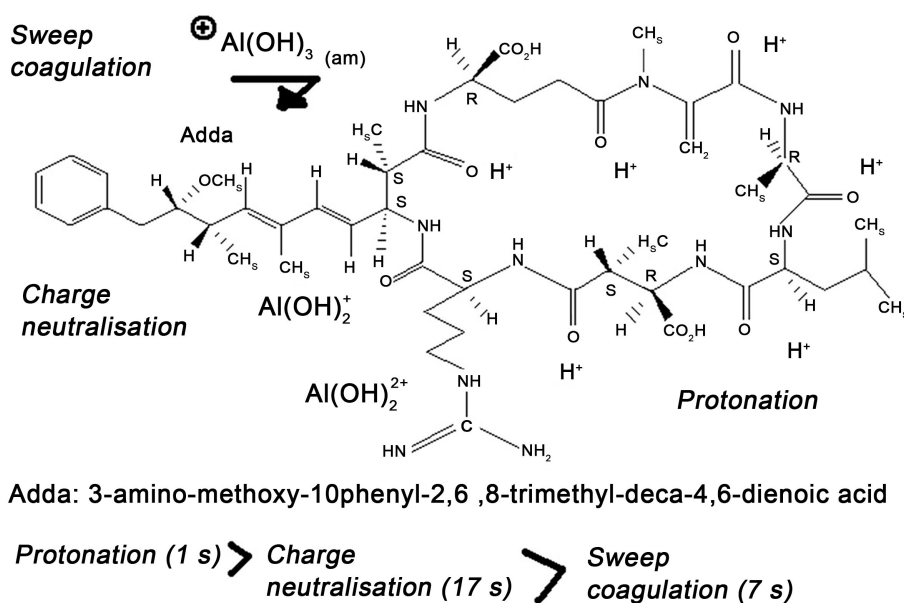


Figure 3. Coagulation mechanisms for microcystin-LR removal [3].

A decade ago, we suggested the hydrophilic/hydrophobic ratio as a function of removing the hydrophilic and hydrophobic content by the coagulation process [93]. It is well-established that the coagulation process could reduce dissolved organic carbon by around 30% - 60% by increasing the coagulant dose and optimizing reaction pH, in which large organic molecules with hydrophobic properties were removed preferentially. Furthermore, the literature affirmed that the more excellent removal of UV-absorbing substances indicates that alum coagulation preferentially removed the hydrophobic fraction of the total organic carbon. The hydrophobic fraction needs to be removed entirely without transforming it into hydrophilic fractions by a coagulation process avoiding pre-chlorination/pre-oxidation due to the risk of fragmentation of organic molecules [7]. Determining the exact numerical values of the hydrophilic/hydrophobic ratio for raw water and treated water at different stages of the treatment processes in a water treatment plant would help more focusing on OM control and removal.

Ten years ago, we suggested that the hydrophilic/hydrophobic ratio is a function of the removal of hydrophilic and hydrophobic parts during coagulation [93]. It is known that by augmenting the coagulant injection and optimizing the solution pH, the coagulation method can reduce dissolved OM by about 30-60%, mainly retaining large hydrophobic organic substances. Also, better elimination of UV-absorbing species has been demonstrated showing that Al coagulation mostly retains the hydrophobic portion of OM. The hydrophobic part must be completely eliminated without transforming it into the hydrophilic part through the coagulation technique, averting pre-chlorination stage because of the possible fragmentation of organic substances [7]. Assessing accurate levels for the hydrophilic/hydrophobic ratio of raw and treated

water at various steps of the treatment in water treatment factories will help place more significant emphasis on the control and removal of OM.

1) *Electrocoagulation (EC) pathways*

During the EC process (same as CC), particles are eliminated through physicochemical pathways [1]. The main difference is the *in-situ* liberation of coagulating metal cations during the EC application [25] [76]. The injected concentration, C (g/L) (Equation (4)), can be calculated according to Faraday's law (Equation (5)), which determines the dose of metal coagulant ions ($[Al^{3+}/Fe^{2+}]$) liberated into the device, according to a certain CD and time [1] [89]:

$$C = \frac{m}{V} \quad (4)$$

$$m = \frac{I \times t \times M_w}{z \times F} \quad (5)$$

where: m is the amount of dissociated metal (g), I is the electric current (A), t is the residence period (s), M_w is the molecular weight of the metal, z is the number of electrons (for Fe^{2+} , $z = 2$), F is Faraday's constant (96,485 C/mol), and V is the volume of the treated water (L).

Figure 4 presents numerous Fe and Al species, which could be present depending on the pH and metal level. Identical to CC, the M^+ ions interact with OH^- to generate numerous polymeric hydroxide complexes [25] [65] [68] [98]. Based on the pH, the polymeric hydroxides interact with negatively charged particles (such as NOM and *E. coli*) via CN [41] [76] [89] [94]. In addition, metal hydroxide precipitate could be instantaneously generated, causing the aggregation of smaller particles via differential settling flocculation [19] [41]. As the pH decreases, CN will have a more considerable role, especially at lower coagulant dosages [41].

The cathode generates $H_{2(g)}$ and OH^- simultaneously, simultaneously auguring pH during EC [25] [76] [89]. The resulting precipitates will be retained via EF or additional flocculation and sedimentation [11] [25] [89]. Flocculation pathways such as diffusion, advection, or differential settling let EC retain diverse particles from water, comprising pathogens, NOM, and inorganics [17] [18] [19] [65] [67] [99] [100].

During the EC process, the cathode produces $H_{2(g)}$ and OH^- , simultaneously auguring the pH [25] [76] [89]. The resulting precipitate is retained by EF or additional flocculation and sedimentation [11] [25] [89]. Flocculation mechanisms like diffusion, advection, or differential sedimentation enable EC to retain a variety of particles in the water, including pathogens, NOM, and minerals [17] [18] [19] [65] [67] [99] [100].

2) *Electrocoagulation (EC) for removing Natural Organic Matter (NOM)*

Researchers [83] discussed the literature on removing NOM (a DBP precursor) using EC, noticing that in many investigations, the NOM removal was more significant than 70% from synthetic solutions and actual water. As a rule,

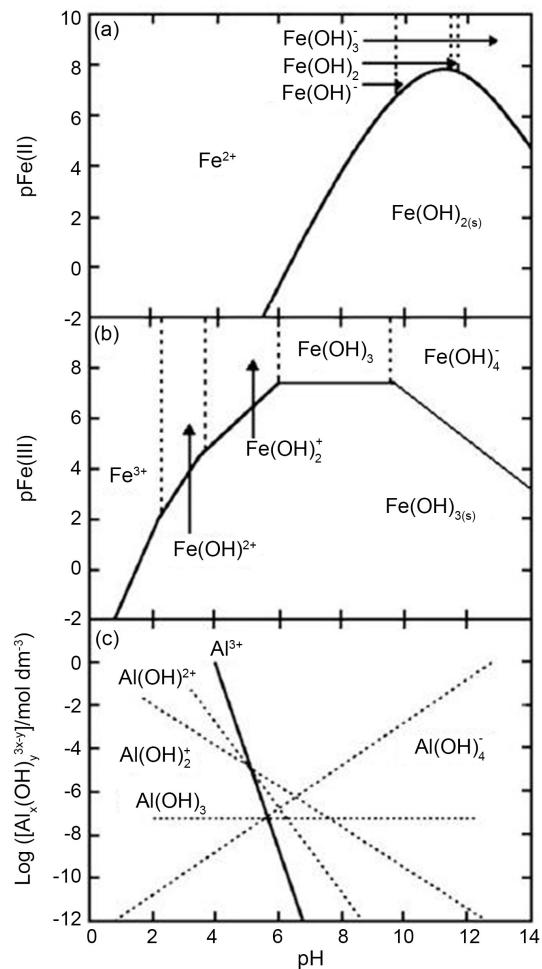


Figure 4. Aqueous species diagram for (a) Fe(II) and (b) Fe(III). Below the solid line represents the insoluble iron species at equilibrium and the dotted lines represent the dominance limits of the soluble iron species. (c) Al(III) solubility vs. pH diagram [1] [25].

NOM reductions are identical, even if moderately less employing EC than CC [11]. CDs equal to or less than 10 mA/cm² depicted the more significant elimination of NOM, with more extensive CDs illustrating no more amelioration [101]. Scientists [95] noticed that the electrical charge (applied current × reaction time normalized to the reactor's volume (C/L)) and the pH were the pivotal variables for eliminating organic substances [1].

In addition, temperature moderately influenced NOM reduction efficacy, possibly because of the dependence on the production rate of the metal coagulant dose [1] [95]. A crucial parameter needing more estimation is the metal ions speciation in solution, particularly in iron EC [102]. Researchers [22] [103] proved that unique iron species formed throughout EC could retain NOM at varying levels. Fe²⁺ or Fe³⁺ can generate complexes with NOM prior producing metal hydroxides, reducing ideal floc creation [11] [71] [104].

3) Electrocoagulation (EC) for removing *E. coli*

Eliminating *E. coli* using EC could equal or surpass achievements reached in CC with decantation [1] [19] [105]. Scientists [17] discovered that augmenting CD results in a more critical reduction of *E. coli*, similar to the phenomenon depicted for NOM retention (Figure 5). Electric current (Faraday's law, Equation (4) is responsible for the level of coagulant produced in EC device, and more significant coagulation formation could generate more flocs [106] for the physical elimination phenomenon (Figure 6) [17] [18] [107]. Increasing the current charge rate by adjusting the application time could also affect the dosage, even at fixed CD, improving the killing of *E. coli* [108] [109] [110]. Researchers [19] demonstrated enhanced killing of *E. coli* in synthetic groundwater as the EC iron injection increased. Scientists [18] noted that 2.84-logs of *E. coli* bacteria were killed utilizing EC run at 2.0 A during 10 min (~110 mg/L Al) in contaminated natural river water, even if a more critical injection could affect the EC performance via forming more considerable sludge mass and wearing the electrodes [25].

The main elimination pathway of *E. coli* reduction throughout EC is adsorption via flocculation [111]-[117]. Secondary practicable pathways involve

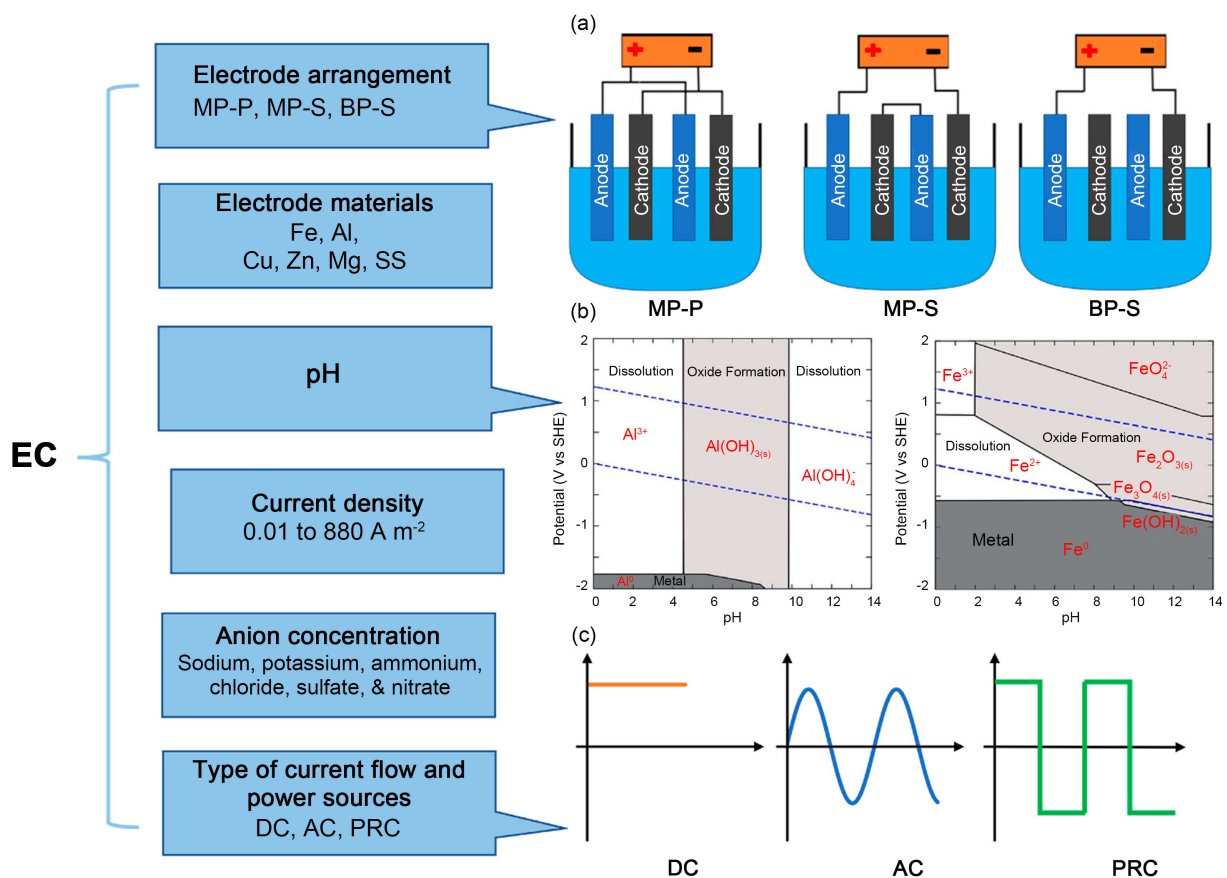


Figure 5. Parameters affecting electrocoagulation (EC)'s reduction efficacy (a); Electrode configuration, monopolar parallel (MP-P), monopolar series (MP-S), and bipolar series (BP-S) [5]; (b) Eh-pH diagrams at 25°C of Al and Fe in solution, the dashed lines show the electrochemical stability span of water; (c) Current flow type for EC-direct current (DC), alternating pulsed current (AC), polarity-reversal current (PRC) [90].

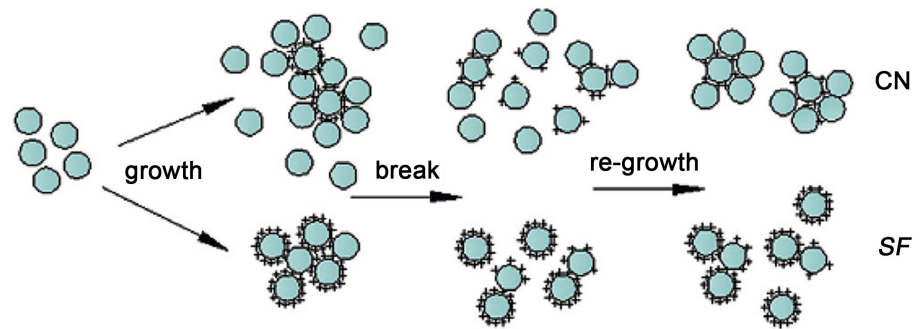


Figure 6. Pathway of flocs generation, breakage and regrowth in charge neutralization (CN) and sweep flocculation (SF) [106].

direct harm to the cell or proteins by passage via the electrical field [18] [91] [92] [118] or reaction with oxidants generated through electrolysis [1] [19] [119] [120].

2.2.3. Electrocoagulation-Electrooxidation (EC-EO)

Regarding demobilizing *E. coli*, EO and EC have merits and disadvantages. Deactivating *E. coli* using EO is exceptionally efficient, even if DBPs may be formed due to reactions between NOM and oxidants [25] [69] [121] [122]. As seen above, EC has been depicted to remove NOM at rates equal to CC and could reduce some *E. coli* [19] [123]. Consequently, taking advantage of the merits of both techniques in successive applications, identically to that of a traditional multi-barrier treatment factory, could lead to satisfying *E. coli* killing and suitable NOM retention to meet water quality requirements [1] [39].

Scientists [39] implemented EC-EO in industrial wastewater, facilitating the degradation of pollutants such as chemical oxygen demand, color, turbidity, and coliforms. They attained 99% elimination of such pollutants, reducing the residence time from 21 hours employing EO alone to 2 hours using EC-EO. Integrating EC with electro-Fenton [80] [124], which oxidizes Fe^{2+} to form $\cdot\text{OH}$, has also depicted efficiency. Scientists [36] established that combining Fe EC and electro-Fenton with BDD electrodes plus air diffusion in domestic wastewater can eliminate numerous microorganisms at a neutral pH. They noted that successive implementation of EC and electro-Fenton (CDs of 20 and 33 mA/cm^2 , respectively) for 30 min led to a more significant reduction than either method alone. Several authors utilized an integrated electrochemical cell, in which EC and EO processes occur in the identical recipient. An integrated electrochemical setup can considerably reduce *E. coli* concentrations in domestic wastewater, even at CDs below 2 mA/cm^2 [1].

Although combined electrochemical techniques have proved efficient *E. coli* reduction in industrial and domestic wastewater, the use of successive EC-EO to demobilize *E. coli* in drinking water sources was not noted until the Lynn work (Figure 7) [1]. Lynn [1] focused on assessing the efficacy of a successive EC-EO in reducing *E. coli* in variable-quality drinking water.

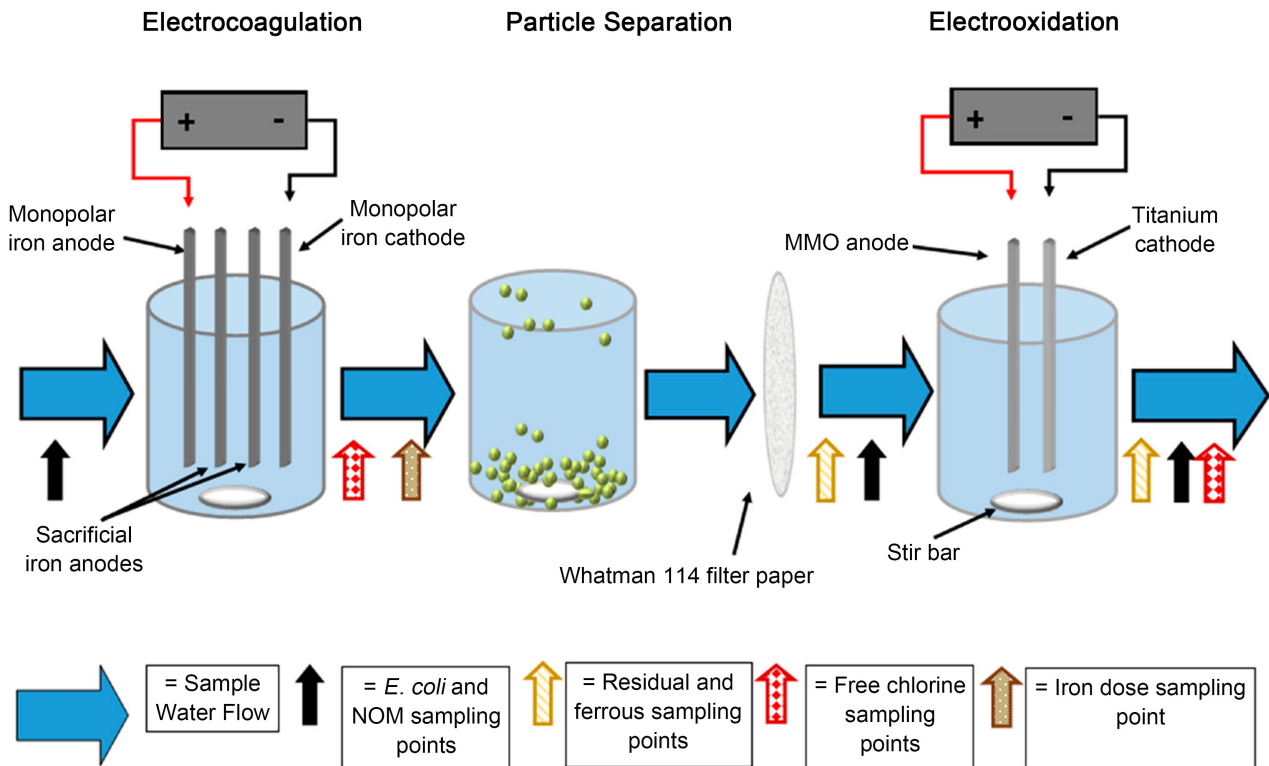


Figure 7. Schematic representation of the laboratory-scale electrocoagulation-electrooxidation (EC-EO) treatment technique and related sampling locations. Each method works in a batch mode [1] [2].

Lynn [1] estimated continuous EC-EO for reducing *E. coli* in four model drinking waters and demonstrated that EC was not an efficacious pretreatment technique for EO to remove *E. coli* in groundwaters. Injecting Fe using EC possibly limited ameliorations in groundwater due to the depletion of oxidants and high total remaining Fe concentrations following filtration. Alternately, EO single was sufficient for decreasing *E. coli* in groundwaters, assuring 4-log and 5-log decrease in the model shallow and model deep aquifer, respectively, utilizing CDs less than those requested for EC. The energy efficiency per order (EEO) of *E. coli* removal for EO single was less than that of either EC or the continuous procedure [1].

EC retained 64% of NOM from the model river water, which is considerable because NOM is a DBP precursor. Nonetheless, the retention of NOM by EC did not enhance *E. coli* removal by EO. Potential reasons for the absence of increased *E. coli* decrease could be a high final pH, depletion of free chlorine by Fe, or insufficiently implemented EO CD [1].

More studies utilizing improved EC-EO with pH regulation to deal with surface waters depicted that a lower pH for EC-EO moderately ameliorates *E. coli* removal. Both surface waters noticed moderate enhancements in retaining NOM at pH 6 or below. Still, the remaining NOM would scavenge oxidants, thus limiting ameliorations in reducing *E. coli* using EC-EO. At the same time, as additional regulation of NOM removal remains requested, the EEO for *E. coli* re-

moval using each process enhanced when pH was below 6, proving the interest in pH regulation [1].

Decreasing pH increased EC's residual Fe concentrations, which would also deplete free chlorine. However, the residual Fe concentrations following EC-EO decreased, possibly due to the oxidation of $\text{Fe}(\text{OH})_{2(s)}$ to $\text{Fe}(\text{OH})_{3(s)}$, offering more efficacious floc settling. Oxidizing Fe^{2+} could also induce Fenton's reaction. A moderate correlation was depicted between EC's remaining Fe levels and *E. coli* reduction throughout EO (after EC step). Besides, Fe^{2+} was transformed to Fe^{3+} throughout EO, establishing that Fenton-like reactions may take place, leading to improved *E. coli* demobilization. When decreased initial pH enhanced *E. coli* demobilization for EO after EC, the difference was unimportant for any case, showing that the EO CD was too low to form the oxidants requested for increased *E. coli* demobilization [1].

3. Evaluating the Alternative Disinfection Technologies

Kerwick *et al.* [70] assessed many alternatives to chlorination following the literature. **Table 3** recapitulates the findings for each technique following numerous criteria employed in the discussion. The findings clarify many of the uncertainties and restrictions required to be controlled prior such techniques could be seen as appropriate alternatives to chlorination. Techniques like O_3 and UV [125] [126] disinfection function and provide established secured disinfection without residual capabilities, even if such techniques could, nonetheless, have competition by two alternative techniques, *i.e.*, direct electrochemical disinfection and mixed oxidant generators, which possess the capacity to offer both primary and residual disinfection. Their chlorine production possibility can hamper the application of such techniques; however, optimizing the methods could limit or prevent chlorine species' generation [25]. More studies remain requested before implementing such processes.

4. Insight into the Electrooxidation (EO) Pathway for Killing *Escherichia coli*

Generating powerful oxidants (e.g., O_2 , O_3 , or OCl^-) in the anode via water electrolysis is the primary driving force behind the electrochemical killing phenomenon occurring in the EO step [129]. Such oxidants are formed when plunging electrodes use electric current to aqueous microorganisms' waters. When the water comprises Cl^- , electrolysis has a span of oxidants, comprising H_2O_2 and O_3 when O_2 is existing, as well as free chlorine and ClO_2 when Cl^- is existing (**Figure 8**). Nonetheless, the oxidant type is a function of the applied current, electrolyte solution, and anode kind [25]. As an illustration, anodes employed in killing pathogens electrochemically by hypochlorite ions, such as Pt electrodes, must possess a low overpotential toward $\text{Cl}_{2(g)}$ evolution, even if pure Pt anodes are not utilized industrially due to their elevated prices and the alternatives for $\text{Cl}_{2(g)}$ evolution are PbO_2 electrodes [129].

Table 3. Evaluation of the alternative technologies to chlorination [70] [110] [127] [128].

| Disinfection technology | Inactivation efficiency | | | | | | Scalability | | | | |
|---------------------------------|-------------------------|-----------------|----------------|----------------|----------------|----------------|-------------|-------|-------|-----|----------------|
| | Bacteria | Viruses | <i>Crypto</i> | DBPs formation | Toxicity | Aesthetics | Costs | Bench | Pilot | Ops | Residual |
| Ozone | √ | √ | √ ^a | √ ^c | √ | √ | √ | √ | √ | √ | × |
| Ultraviolet (UV) light | √ | √ | √ | √ | √ | √ | √ | √ | √ | √ | × |
| Direct electrochemical | √ | # | ? | √ ^e | ? ^d | √ ^e | √ | √ | √ | ? | √ ^e |
| Mixed oxidant generators | √ | √ | √ | √ ^e | √ | √ ^e | √ | √ | √ | √ | √ |
| TiO ₂ photocatalysis | √ | √ | # | √ | ? | ? | × | √ | √ | ? | × |
| Irradiation | √ | √ | # | √ | √ | √ | × | √ | √ | ? | × |
| Pulsed electric fields | √ | # | # | √ | ? | ? | × | √ | √ | ? | × |
| Sonication | √ | # | # | √ | √ | √ | × | √ | ? | ? | × |
| Metal ions (Au/Ag/Cu) | √ | √ ^{ab} | ? | √ | √ ^d | × ^a | ? | √ | √ | ? | √ ^c |
| Ferrates | √ | √ | ? | √ | √ | ? | ? | √ | ? | ? | × |

√Satisfies the criteria. ×Does not satisfy the criteria. ?Not noticed in the literature. #Only a few examples noted in the literature, effective dosing range is not clear. ^aHigh $C \times T$ (disinfectant concentration \times contact time) requested for efficacious kill. ^bDoses may be above the maximum contaminant levels (MCLs) if employed alone, when utilized in integration this is not a trouble. ^cResidual capacity scavenged by organics. ^dDependent on electrode material. ^eDependent on if Cl⁻ is employed in the electrolyte.

Killing pathogens from water may be realized in small electrochemical setups using oxygen gas produced from the anode where the chlorine species formation is not wanted [130], the most employed electrodes for generating oxygen remain stainless steel (SS) and graphite electrodes [129].

Hellal *et al.* [129] utilized graphite as the anode and SS as the cathode for destructing *E. coli* due to the generation of oxygen derivatives induced by the applied electric field, which is by its nature poisonous to cells [114], provoking irreversible permeabilization of cell membranes. Hellal *et al.* [129] noticed it was impossible to produce chlorine compounds for disinfection with the short contact time and suggested the scheme in **Figure 8**.

Killing microorganisms may happen because of oxidation by $\cdot\text{OH}$ produced from the water oxidation at the anode. The demobilization phenomenon happens inside the surrounding area of the electrode and solution interphase (**Figure 8**). Powerful oxidant species are consequently formed from electrolyte EO at the anode surface, leading to the DO phenomenon of the microorganism (**Table 4**). Following the application of CD, the oxidation of microorganisms happens by direct electron transfer in the potential region prior $\text{O}_{2(\text{g})}$ evolution through electrogenerated $\cdot\text{OH}$. Also, the water oxidation reaction to generate $\cdot\text{OH}$ competes frequently with the secondary reaction of anodic dissociation of these radicals in oxygen and the oxygen evolution reaction, as depicted in Equation (2) [129].

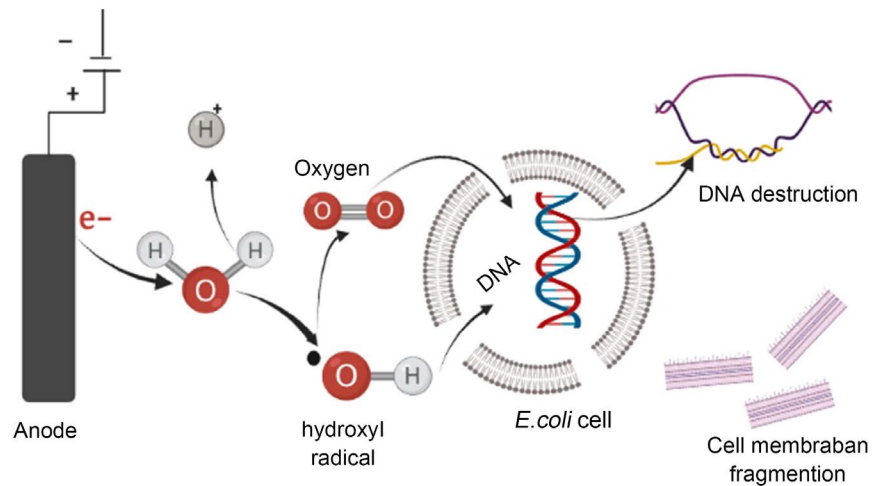


Figure 8. Suggested pathway of *E. coli* demobilization in the electrooxidation (EO) unit [129].

Table 4. Major routes proposed explaining the electrochemical technique deadliness [114].

| Oxidants | | Electric field | |
|---|--|---|--|
| Oxidative stress and cell loss of life. | Irreversible permeabilization of cell membranes. | Electrochemical oxidation of vital cellular constituents. | Electrosorption of negatively charged <i>E. coli</i> cells to the anode surface + direct electron transfer reaction. |

Since the $\cdot\text{OH}$ species are suggested to be the significant killing agent of bacteria in the EO technique, scientists established a direct link between their anode's surface and the radical activity thanks to their high oxidation potential. Graphite as the non-active anode is advantageous thanks to its high oxygen overpotential, attaining the oxidation of microorganisms by an electrochemical procedure mediated by physisorbed $\cdot\text{OH}$.

This suggested demobilization route was also proved with a gas chromatographic analysis of the solution following disinfection with EO at such conditions for detecting any chlorinated by-products due to the formation of chlorine chemicals and free chlorine determination in the solution. The findings depicted no detection of any halogenated or chlorinated chemical in the solution. Consequently, the inactivation route of *E. coli* in these conditions is the electrochemical killing using $\text{O}_{2(\text{g})}$ [129].

5. Insight into the Electrocoagulation (EC) Pathway for Killing *Escherichia coli*

Physical removal and chemical demobilization mechanisms are proposed for microbes' elimination phenomena during the EC technique utilizing Fe/Al anodes (**Figure 9**): 1) entrapping microorganisms in flocs, 2) destabilizing negatively charged microorganisms via sweep flocculation (SF), and 3) deactivating microorganism cell envelopes upon electrochemically produced ROSs or direct

influence of the electric field [116].

Table 5 shows the EC reactions using Fe and Al electrodes.

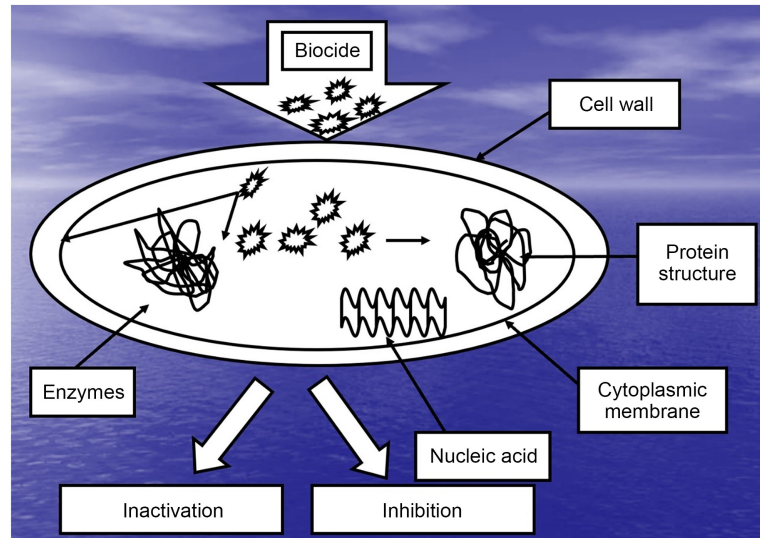


Figure 9. Target sites of biocides in microbial cells [114].

Table 5. Electrocoagulation (EC) pathways employing Fe (pH 2, 7, and 12) and Al (pH 7) electrodes [123].

| Fe Mechanisms | Medium | Reaction | | |
|---------------------------------|--|--|--------------------------------|-------|
| <i>Mechanism # 1</i> (pH 2) | Anode | $2\text{Fe}_{(s)} - 4e^- \rightarrow 2\text{Fe}^{2+}_{(aq)}$ | $(E^\circ = +0.447 \text{ V})$ | (1') |
| | | $2\text{H}_2\text{O}_{(l)} - 4e^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)}$ | $(E^\circ = -1.229 \text{ V})$ | (2') |
| | Solution | $2\text{Fe}^{2+}_{(aq)} + 4\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{2(s)}$ | | (3') |
| | Cathode | $8\text{H}^+_{(aq)} + 8e^- \rightarrow 4\text{H}_{2(g)}$ | $(E^\circ = 0.000 \text{ V})$ | (4') |
| | Total | $2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow \text{O}_{2(g)} + 4\text{H}_{2(g)} + 2\text{Fe}(\text{OH})_{2(s)}$ | | (5') |
| <i>Mechanism # 2</i> (pH 7) | Anode | $2\text{Fe}_{(s)} - 4e^- \rightarrow 2\text{Fe}^{2+}_{(aq)}$ | $(E^\circ = +0.447 \text{ V})$ | (1') |
| | | $\text{Fe}^{2+}_{(aq)} - e^- \rightarrow \text{Fe}^{3+}_{(aq)}$ | $(E^\circ = -0.771 \text{ V})$ | (6') |
| | | $\text{Fe}_{(s)} - 3e^- \rightarrow \text{Fe}^{3+}_{(aq)}$ | $(E^\circ = +0.037 \text{ V})$ | (7') |
| | Solution | $2\text{Fe}^{2+}_{(aq)} + 4\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{2(s)}$ | | (3') |
| | | $2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)}$ | | (8') |
| Cathode | $8\text{H}_2\text{O}_{(l)} + 8e^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ | $(E^\circ = -0.828 \text{ V})$ | (9') | |
| Total | $3\text{Fe}_{(s)} + 8\text{H}_2\text{O}_{(l)} \rightarrow \text{Fe}(\text{OH})_{2(s)} + 2\text{Fe}(\text{OH})_{3(s)} + 4\text{H}_{2(g)}$ | | (10') | |
| <i>Mechanism # 3</i> (pH 12) | Anode | $\text{Fe}_{(s)} - 3e^- \rightarrow \text{Fe}^{3+}_{(aq)}$ | $(E^\circ = +0.037 \text{ V})$ | (7') |
| | Solution | $2\text{Fe}^{3+}_{(aq)} + 6\text{OH}^-_{(aq)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)}$ | | (8') |
| | Cathode | $8\text{H}_2\text{O}_{(l)} + 8e^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ | $(E^\circ = -0.828 \text{ V})$ | (9') |
| Total | $2\text{Fe}_{(s)} + 6\text{H}_2\text{O}_{(l)} \rightarrow 2\text{Fe}(\text{OH})_{3(s)} + 3\text{H}_{2(g)}$ | | (11') | |
| Al Mechanism (pH 7) | Anode | $\text{Al}_{(s)} - 3e^- \rightarrow \text{Al}^{3+}_{(aq)}$ | $(E^\circ = +1.660 \text{ V})$ | (12') |
| | | $2\text{H}_2\text{O}_{(l)} - 4e^- \rightarrow \text{O}_{2(g)} + 4\text{H}^+_{(aq)}$ | $(E^\circ = -1.229 \text{ V})$ | (2') |
| | Solution | $\text{Al}^{3+}_{(aq)} + 3\text{OH}^-_{(aq)} \rightarrow \text{Al}(\text{OH})_{3(s)}$ | | (13') |
| | | $\text{Al}(\text{OH})^+_{4(aq)} \rightarrow \text{OH}^-_{(aq)} + \text{Al}(\text{OH})_{3(s)}$ | | (14') |
| | Cathode | $8\text{H}_2\text{O}_{(l)} + 8e^- \rightarrow 4\text{H}_{2(g)} + 8\text{OH}^-_{(aq)}$ | $(E^\circ = -0.828 \text{ V})$ | (9') |
| | $\text{Al}_{(s)} + 4\text{OH}^-_{(aq)} - 3e^- \rightarrow \text{Al}(\text{OH})^+_{4(aq)}$ | | (15') | |
| Total | $2\text{Al}_{(s)} + 8\text{H}_2\text{O} \rightarrow \text{O}_{2(g)} + 5\text{H}_{2(g)} + 2\text{Al}(\text{OH})_{3(s)}$ | | (16') | |

Ten years ago, we discussed [29] the possibility of advanced oxidation process (AOP) phenomena occurring in the EC technology. AOPs (e.g., EO) have been broadly defined as near-ambient temperature treatment processes based on highly reactive radicals, particularly $\cdot\text{OH}$ as the primary oxidant. In principle, as water-containing colloidal particulates, oils, or other pollutants move through the applied electric field, ionization, electrolysis, hydrolysis, and free-radical generation could change the physicochemical characteristics of water and pollutants. Also, when the electrochemical units work at an elevated cell potential and an anodic phenomenon happens in the potential region of water discharge, $\cdot\text{OH}$ is produced. Ultrasound during EC may be beneficial to reach more possibilities of free radical production in EC. Further, the EC method at $\text{pH} < 3$ has a higher probability of forming $\cdot\text{OH}$. There is no evidence of the occurrence of AOP reactions inside the EC apparatus, and more investigations are required here concentrating on free radical generation [29].

In the context of the present review, which focuses on the influence of EC pretreatment on *E. coli* killing employing EO, the EC process would be powerfully efficient in killing *E. coli* at $\text{pH} < 3$ and in the presence of ultrasound acting during EC.

6. Conclusions

Although the literature mainly reports on the inactivation of microbes using various electrochemical disinfectants, the impact of technique parameters and device design on bactericidal efficiency is not fully understood. This review concentrated on recent achievements of electrocoagulation (EC) and electrooxidation (EO) in killing pathogens such as *Escherichia coli*. From this review, the essential drawn points are:

- 1) Lynn [1] demonstrated that EC-EO did not enhance *E. coli* removal more than EC alone, even if more technique regulation could produce ameliorations (e.g., regulating the Fe injection for eliminating natural organic matter (NOM) would diminish the influence of oxidant scavengers). Also, a more performant filtration technique (e.g., granular filtration) would decrease NOM and Fe concentrations in EO influent, reducing the consumption of free chlorine due to these components. Higher EO current densities (CDs) must be implemented in the continuous treatment to enhance the removal of *E. coli*. Considering the increased oxidant production as a function of CD, a larger reduction in *E. coli* is expected. In addition to assembly regulation, further studies are requested to identify specific pathways for *E. coli* elimination in drinking water during EO when high iron levels are present, and Fe speciation must also be considered in addition to electrostatic interactions between Fe and pathogens.

- 2) Lynn *et al.* [2] showed that *E. coli* reduction with EC-EO was greater than that with EC single only following pH regulation and suggested that ongoing technique control may lead to more ameliorations, such as regulating the Fe dosage in NOM reduction would reduce the effect of oxidant scavengers. Also,

more performant filtration techniques (e.g., granular filtration) will diminish NOM and total Fe contents in the EO feedwater, thereby reducing the need for oxidants. Furthermore, sequential EC-EO treatment requires the application of higher EO CDs to improve *E. coli* removal. An in-depth study of the demobilization pathways of *E. coli* in drinking water at high FE concentrations in the EO will also help ongoing setup design.

3) A methodology has been devised to evaluate alternatives to chlorination for disinfecting drinking water [70]. Seven criteria were identified to measure the technique's acceptance as an option for chlorination, including demobilization efficacy, risk for generation of disinfection by-products (DBPs), toxicity, aesthetic water quality, cost, scalability, and residual maintenance. Such criteria are evaluated regarding significance to the water utility and are linked to water quality regulations. The methodology was assessed employing ultraviolet (UV) disinfection and the results depicted that it satisfied all criteria except assuring remaining disinfectant. UV light is a permissible option to chlorination when remaining disinfectant is not requested or chemical residues can be employed. Several other options for chlorination were evaluated employing the suggested methodology [70], and the uncertainties and restrictions of each technique were determined. Two processes (direct electrochemical disinfection and mixed oxidant generators) have been recognized as possible ongoing options for Cl₂.

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Conflicts of Interest

The authors declare no conflicts of interest.

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