

Electrocoagulation Process Intensification for Disinfecting Water – A Review

Djamel Ghernaout^{1,2,*}, Nouredine Elboughdiri^{1,3}

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

²Chemical Engineering Department, University of Blida, Blida, Algeria

³Chemical Engineering Process Department, University of Gabes, Gabes, Tunisia

Email address:

djamel_andalus@hotmail.com (D. Ghernaout)

*Corresponding author

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Abstract: In the field of disinfecting water, if there is a process that has attracted huge attention from water treatment specialists it is the electrocoagulation (EC) process. Indeed, during the last two decades and thanks to its techno-economic benefits, this electrochemical technology has been the subject of many hundreds of researches and patents published throughout the entire world. The generally accepted tendency concerning the usage of the EC technique is to employ it as an integrated step with additional processes. In the field of killing pathogens, EC process is frequently inserted as a pre-stage before electrooxidation (EO) method in the treatment train. For such a combination, more important virus reduction is possibly reached via the collective actions of physical removal by coagulation/filtration, ferrous iron-based disinfection, and EO disinfection. In this context, much more research needs to be realized to distinguish among the electric field and cohesion contributions. Furthermore, more investigation has to be pointed on evaluating the more and more probable production of the hydroxyl radical (OH) during the EC technology. On the other hand, like in the chemical water disinfection, identical problems such as disinfection by-products generation have also appeared in the EC applications. More research needs to be pointed into such directions.

Keywords: Electrocoagulation (EC), Electrodisinfection (ED), Electric Field (EF), Electro-Fenton (E-F), Boron-doped Diamond (BDD)

1. Introduction

More and more, municipally treated wastewater recovery has deserved increasing attention because of the extremely crucial water shortage in many areas through the World [1-4]. In nations where it is hard to ensure the potable water supply, this condition becomes more serious. For this cause, wastewater recovery may be considered as a valuable water resource [5]. As a result, various techniques have been proposed to remedy municipal wastewater [6-11]. Such technologies are conceived via taking into account that for secured usage of this water, microorganisms have to be deleted. Usually, two indicators are viewed to be the principal objectives: the fecal pathogen *Escherichia coli* (*E. coli*) and the colloids (turbidity) [4].

In order to treat such water, physicochemical methods

have been largely investigated and applied [4, 12, 13]. On the other hand, the significant augmentation observed in the conductivity of the treated waste is viewed as an extremely crucial inconvenient and suggests electrocoagulation (EC) as a viable choice [14-16]. This electrochemical process includes the in situ formation of coagulant agents from the electrochemical dissolution of a sacrificial aluminum or iron [17] anodes [18, 19]. EC differs from the chemical coagulation in the fact that during EC the reagent formed is the metal hydroxide directly and not a salt of a multivalent cation (like FeCl₃ or Al₂(SO₄)₃) that remains to be afterward reduced [20, 21]. Consequently, electrochemical technologies are viewed cleaner and environmentally friendly [22-25].

More than EC, additional electrochemical technologies,

like the Electrochemical Advanced Oxidation Processes, have as well been suggested for wastewater recovery [26]. Such methods are founded on the generation of oxidants with high disinfection potential. As an illustration, the Conductive-Diamond Electrochemical Oxidation (CDEO) [27] concerns the formation of killing agents throughout electrooxidizing ions inherently present in used water employing anodic oxidation via diamond electrodes [4]. Researchers established that it is easy to achieve the total disinfection of the wastewater without the necessity to inject chemical products [28]. Moreover, there is no doubt that EC and CDEO may be integrated successfully, conducting to a technique (known as electrodisinfection-electrocoagulation, ED-EC) able to reduce the concentration of *E. coli* hold in real municipal treated wastewater and to diminish its colloids at the same time [29, 30].

Until now, it is fundamental to remember that the main part of the electrochemical techniques for wastewater recovery has been tested at bench scale [31-33]. Consequently, it is required to investigate the scale-up of such methods to guarantee appropriate running with a huge quantity of used water. In this direction, researchers [27, 30] have mentioned killing pathogens in municipal treated wastewater via electrolysis and photo-electrolysis employing diamond anodes at pilot scale, discovering a performant elimination of *E. coli* at moderate working parameters [4].

2. Suggesting a Combined ED-EC Method for Wastewater Recovery

In their recent research, Cotillas et al. [4] worked on the scale-up of a combined electrodisinfection-electrocoagulation (ED-EC) technique, particularly conceived for the recovery of real municipal treated wastewater, furnished with boron-doped diamond (BDD) anodes and iron bipolar electrodes. The set-up runs in continuous mode and in the prototype the anode area was augmented three times (anodic oxidation) and the bipolar electrode area fifteen times (EC) concerning the device employed at bench scale. Their findings prove that it is feasible to obtain the total and concurrent disinfection and turbidity elimination via implementing current densities inside the domain 5–10 A/m². Free and combined chlorine species were electrogenerated from the chloride included in the effluents (no chemicals were introduced) being these species responsible for the elimination of microbes [34]. In addition, iron coagulant species originating from the electro-dissolution of the anodic side of bipolar electrodes raise turbidity decrease. In the scaled-up prototype, a more important turbidity diminution was attained due to the augmentation in the bipolar electrode area. Finally, it was established that for electric charges below 0.07 kAh/m³ the recovery of municipal treated wastewater may be obtained, bypassing the generation of hazardous chlorates and perchlorates even at current densities more important than 7 A/m².

3. Routes of Virus Attenuation Via Iron Electrocoagulation

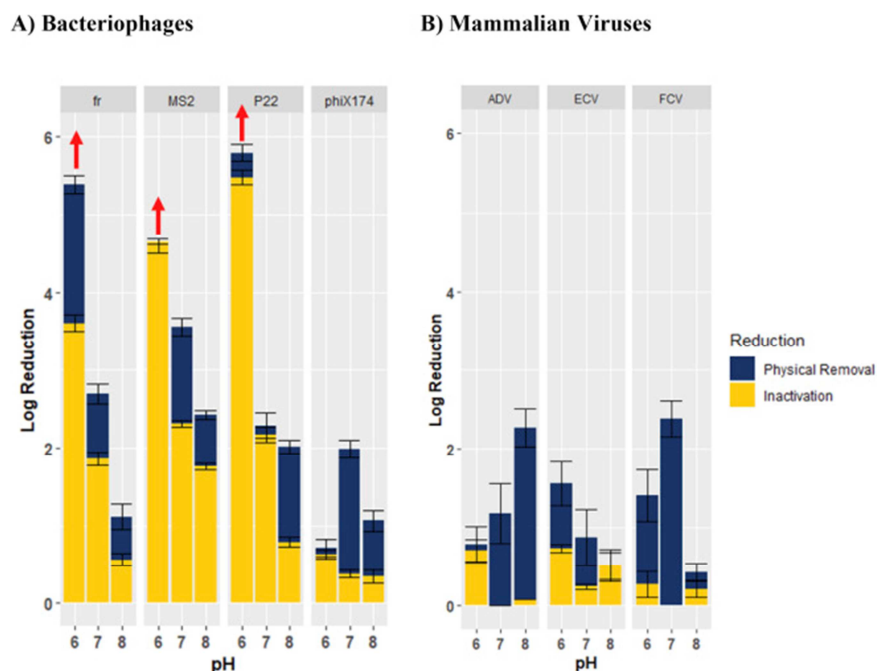


Figure 1. Impact of pH on inactivation and physical removal of A) bacteriophages and B) mammalian viruses due to EC. Upward arrows indicate log reduction beyond the countable limit, so values shown are the limit of quantification. Error bars represent standard error of the mean of triplicate tests [35].

Rising water treatment methods employing ferrous and zero-valent iron offer encouraging virus removal via both deactivation and adsorption. Heffron et al. [35] studied iron EC for virus attenuation in potable water throughout

bench-scale batch trials. They studied comparative participation of physical elimination and demobilization, as determined by recovery via pH 9.5 beef broth elution, for three mammalian viruses (adenovirus, echovirus, and feline

calicivirus) and four bacteriophage surrogates (fr, MS2, P22, and FX174). Although no one bacteriophage totally represented the reduction of the mammalian viruses in all water matrices, bacteriophage FX174 was the exclusive surrogate that presented total elimination comparable to that of the mammalian viruses (Figure 1). They found that bacteriophages fr, MS2, and P22 were all more vulnerable to deactivation than the three mammalian viruses, augmenting worries concerning the appropriateness of these frequent surrogates as parameters of virus reduction. They examined pathways of bacteriophage reduction with a view to deciding why various bacteriophages were specifically vulnerable to demobilization. Physical elimination was firstly attributed to the embodiment in flocs, during the time that demobilization was mainly linked to ferrous iron oxidation. They suggested more important electrostatic attraction, virus aggregation, and capsid durability as causes for virus vulnerability to ferrous-based deactivation. Their findings proposed that total treatment declaration founded on bacteriophage reduction for any iron-based technique must be carefully adopted because of more increased vulnerability of bacteriophages to demobilization throughout ferrous oxidation [36].

4. Consecutive Electrocoagulation-Electrooxidation for Virus Reduction

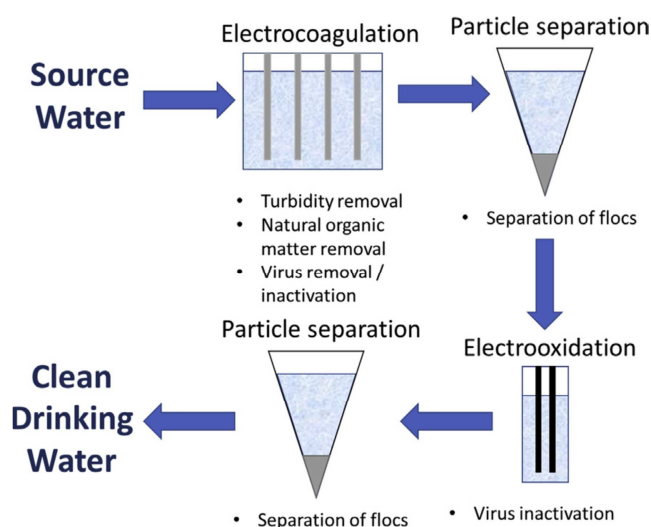


Figure 2. Schematic of electrocoagulation-electrooxidation treatment train and hypothesized treatment effects for each stage [37].

Researchers [37] examined the reduction of viruses employing EC as a pretreatment before electrooxidation (EO) treatment employing BDD electrodes. Their work is the first to test a consecutive EC-EO treatment setup for virus elimination. They employed bench-scale and batch reactors to assess reduction of viruses in changing water quality by EO and a consecutive EC-EO treatment train. They found that EO of two bacteriophages, MS2 and FX174, was prevented by natural organic matter (NOM) and colloids, showing the potential necessity for pretreatment.

Nevertheless, the EC-EO treatment train was useful exclusively in the model surface waters tested (Figure 2). In model groundwaters, EC by oneself was as useful or better than the integrated EC-EO treatment train. Mitigation of human echovirus was importantly lower than one or both bacteriophages in all model waters; however, bacteriophage FX174 was a more representative surrogate than MS2 in the existence of NOM and colloidal particles. Juxtaposed to traditional treatment via ferric salt coagulant and free chlorine disinfection, the EC-EO device was less performant in model surface waters but more efficient in model groundwaters. Consecutive EC-EO was advantageous for some usages; however, functional regards may presently outbalance the advantages.

5. Sequential “Electrochemical Peroxidation-Electro-Fenton” Process for Sludge Treatment

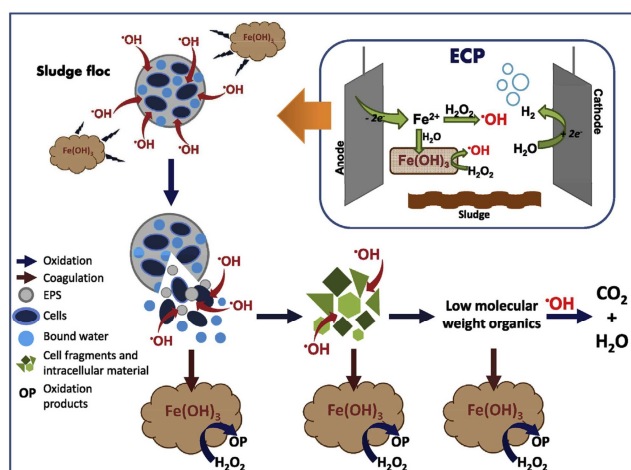


Figure 3. Degradation mechanisms taking place during ECP of anaerobic sludge. Inset panel: production of coagulants and oxidants during ECP using mild steel electrodes [38].

Olvera-Vargas et al. [38] exhibited a consecutive electrochemical technology for complete remediation of anaerobic sludge. They integrated electrochemical peroxidation (ECP) and electro-Fenton (E-F) (Figure 3). In the first stage, ECP (comprising H_2O_2 -assisted EC with Fe electrodes) was used as a processing and stabilizing technique. The synergistic EC/Fenton oxidation contributions greatly decreased the COD, TOC and total suspended solids (TSS) by 89.3%, 75.4%, and 85.6%, respectively, under regulated parameters (initial pH of 5, $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ dose ratio of 5, 15.38mA/cm² and 2h treatment). In addition, total coliforms were totally eliminated during the first hour of application. In the second stage, E-F was efficiently exercised to mineralize the remaining organic portion in the liquid effluent after dewatering, attaining 91.6% and 87.2% of COD and TOC elimination, respectively, following 4h of treatment under regulated parameters (pH 3 and 25mA/cm²), at the same time almost total COD and TOC elimination was obtained in 8h.

6. Consecutive Electro-Assisted Coagulation-Photocatalytic Oxidation for Drug Effluent Treatment

Lalwani et al. [39] suggested a similar approach as above. These researchers mentioned that the rising pollutant cefixime, an active pharmaceutical ingredient (API), was successfully removed using a consecutive two-stage EC and photocatalytic oxidation treatment. Their work is outstanding thanks to dealing with elevated strength crude drug effluent, with total organic carbon (TOC) of 7395mg/L, with a view to decrease the organic load with total removal of the pre-existing microbial population. Remediation techniques were performed in batch reactors. EC was realized using two electrodes (Al and Fe). By regulation, at 10V and 24V for Al and Fe electrode, TOC was found to be diminished by 14% and 22%, respectively. Throughout Fenton's reaction employing Fe electrode, 41% TOC removal was reached, with a decrease of cefixime to 0.01mg/L. Via step two EC with H_2O_2 , an additional 2% TOC reduction facilitated in the decrease of cefixime to 0.001mg/L. The consecutive EC treated-diluted effluent, subjected to TiO_2 and H_2O_2 assisted photocatalytic oxidation via natural sunlight and UV source, separately, conducted to more TOC decrease of 30% and 33%, respectively. EC application successfully diminished the API below 6mg/L, defined by antimicrobial activity and EC-Fenton's reaction allowed the removal of pre-existing bacteria in the effluent, below 0.5 colony forming units (CFU)/mL. As a result, these workers showed the importance of non-biological treatments, i.e., the electrochemical technology, of drug effluents to avoid microbial drug resistance in nature (Figure 4).

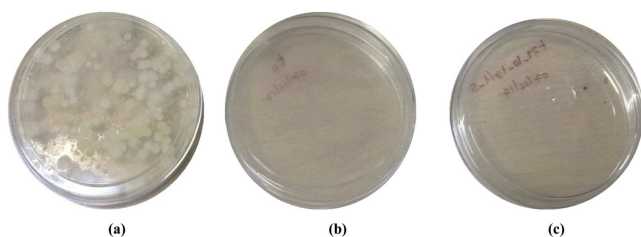


Figure 4. Effluents spread on agar surface (24 h incubation). (a) Raw effluent, (b) Two-step EC treated effluent, and (c) $UV/TiO_2/H_2O_2$ treated effluent [39].

7. Enhancing Electrocoagulation Process Via Ozonation

Barzegar et al. [40] suggested that EC may be improved via applying ozonation for greywater remediation. They focused on the impacts on chemical oxygen demand (COD) and total organic carbon (TOC) eliminations from greywater. They showed that 85% of COD and 70% of TOC were decreased by 60 min electrolysis time, at pH = 7.0, 47.4mg/L ozone and 15mA/cm² current density. In addition, EC employing Fe electrode presented an elevated catalytic activity for ozone activation in contrast with Al

electrode. On the other hand, ozone possessed an elevated efficiency as compared with many chemical oxidants (peroxodisulfate, peroxymonosulfate, and hydrogen peroxide) in integration with the EC method for greywater remediation. Moreover, UV irradiation increased the efficiency of EC/ozone greatly; at the same time, ultrasound could not touch the EC/ozone technique. If UV irradiation is existent, 95% of COD and 87% of TOC were removed. Furthermore, 4 logs of total coliform and 96% of *E. coli* were eliminated upon EC/ozone/UV method (Figure 5). Consequently, EC/ozone/UV method is a performant and practicable method for treating and disinfecting greywater.

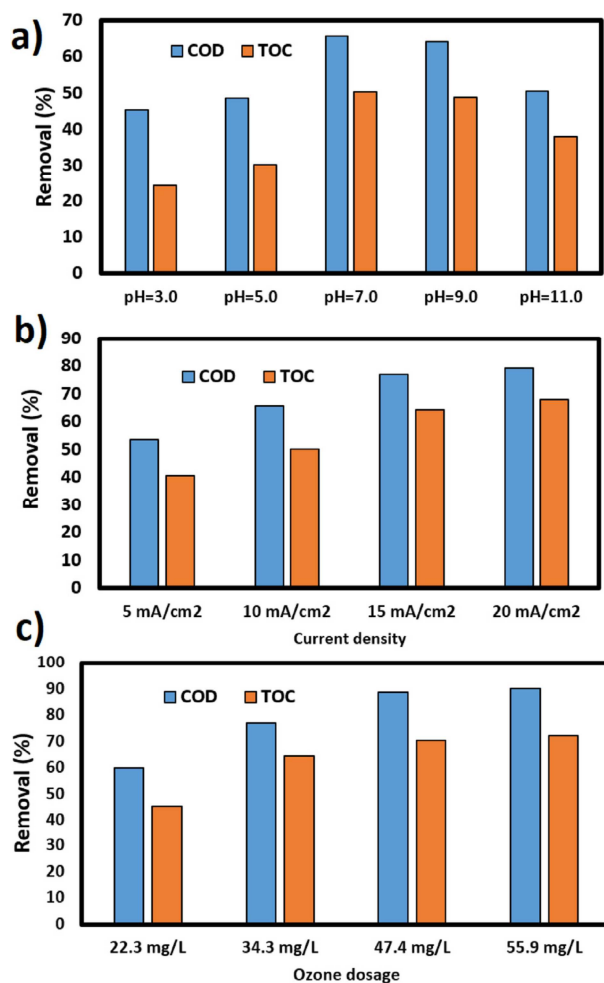


Figure 5. (a) Impact of pH on COD and TOC removal from greywater (ozone dosage = 34.3mg/L, current density = 10mA/cm² and 60 min electrolysis time). (b) Effect of current density COD and TOC removal from greywater (pH = 7.0, ozone dosage = 34.3mg/L and 60 min electrolysis time). (c) Effect of ozone dosage on COD and TOC removal from greywater (pH = 7.0, current density = 15mA/cm² and 60 min electrolysis time).

8. Credible Key Microorganisms' Removal Route Via EC

Govindan et al. [41] discussed the probable key microbes' mitigation pathway via EC. They concluded that microorganism' removal route using EC runs in the next three main mechanisms: (i) Biomass destabilization via

electrochemically formed fresh flocs; (ii) Demobilization of biomass through electrochemically produced reactive oxygen species and germicidal chemicals throughout electrolysis; (iii) Direct application of electric field (EF) [42] on microbes that may demobilize the cell membrane of the pathogens (Figure 6).

Decreasing microbes' pollution from water was greatly reached employing easy EC process with sacrificial Fe, Al and Cu electrodes [41]. The effect of abiotic indicators (pH, time, TSS and COD), electrode materials, supporting electrolyte [43] and NOM [44] on microbes' elimination or demobilization methods were greatly assessed. Appreciable pathogens' removal was obtained via regulated parameters in various kinds of water like industrial wastewater, surface water and synthetic fresh water.

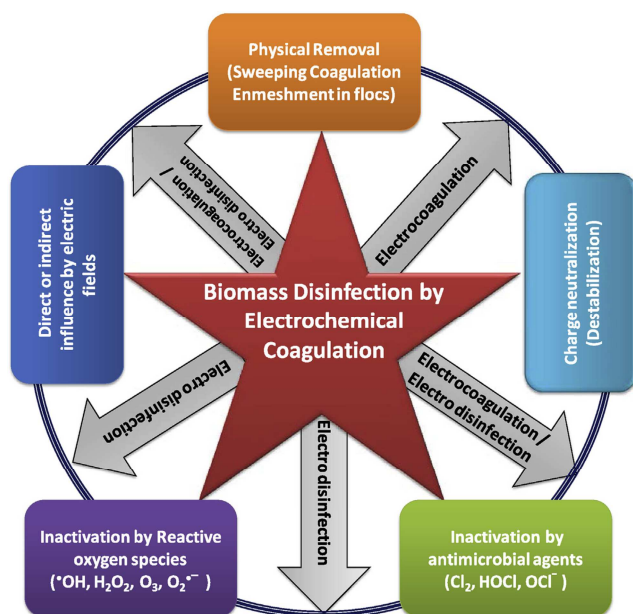


Figure 6. The probable mechanistic pathway of microorganisms' killing throughout EC technique [41].

9. Defiance Facing ED and EC Process Intensification

Employing mixed metal oxide anodes has been proved in disinfecting water, especially the formation of chlorinated disinfectants [45]. In fact, two crucial issues must be resolved to overpass this defiance. The first one is to define how to avert the generation of poisonous by-products like chlorates or trihalomethanes. Chlorates are produced via oxidation of hypochlorite or throughout its disproportionation, which is a natural phenomenon that as well happens throughout aging of the disinfected water. This chemical is linked to dangerous health troubles as it is established to touch the nervous system. The second kind of toxic reagents is even more polemic. Chlorinated chemicals are produced from the integration of organic matter with energetic forms of chlorine, and these chemicals are linked to cancer and different extremely grave illnesses. These chemicals are not special of the electrochemical technology since they are as

well generated throughout the usage of the traditional chlorination process. The additional defiance is to change the mixed metal oxide anodes, like diamond-like coatings, able of not only oxidizing chloride ions but also forming more performant disinfectants, comprising hydroxyl radicals. If employing such electrodes, issues related to the formation of toxic reagents may be exacerbated, due to the well-encountered formation of perchlorates upon oxidation of chlorates. These novel electrode materials unlock the perspective of encouraging the role of various oxidizing reagents like ozone and peroxosalts to assist in eliminating reluctant microorganisms. Suitable residence time among water and the anodes in the electrolyzer, an enough big specific current and the cathodic generation of hydrogen peroxide to avoid additional oxidation of chlorine to chlorates and perchlorates are between techniques rated to bypass issues [45].

Like ED and as an electrochemical technique, EC remains subjected to such issues even if at a less level. Indeed, in EC process, the generation of cationic species from the anode may reduce the formation of such toxic chemicals mentioned above in ED.

Bruguera-Casamada et al. [46] focused on the benefits of E-F over EC for disinfecting dairy wastewater. They disinfected raw dairy wastewater using a consecutive remediation comprising an EC stage with a Fe|Fe cell followed by E-F or UVA-assisted photoelectro-Fenton (PEF). The two latter techniques were executed employing an air-diffusion cathode for H_2O_2 production and either a BDD or a RuO_2 -based anode. They evaluated the demobilization of heterotrophic and lactic acid bacteria, *E. coli* and enterococci. The organic charge was not eliminated efficiently in all situations; at the same time, the microbes were weakly reduced via the flocs produced in EC but hugely demobilized in E-F and PEF. Moreover, E-F was as well beneficial because it avoided the generation of poisonous sludge carrying active bacteria, in contrast to EC. Heterotrophs were the most stable bacteria, whilst the others were completely demobilized in most situations. In the consecutive EC/EF technique implying a BDD anode in the latter stage, the demobilization rate for the lactic acid bacteria was more significant at pH around 7, thanks to the huge capacity of formed active chlorine to oxidize the cell membrane molecules. Employing a RuO_2 -based anode also conducted to a rapid demobilization at pH 3.0. A more important efficiency was obtained if PEF took the place of EF, disregarding the anode, because of the improved bacterial deactivation via UVA radiation. Treating the raw dairy wastewater at natural pH 5.7 upon single E-F stage employing a RuO_2 -anode also produced a more rapid elimination of lactic acid bacteria, *E. coli* and enterococci as compared to BDD, with always remaining small quantities of suspended active heterotrophs in water.

The same researchers applied previously a similar approach with more different microorganisms and with more interesting findings [47].

With regard to EC method conception, the interest has to be addressed to intensify the EC apparatus in terms of contact

period and near exposure chances among water contaminants and electrodes surface [48]. Laminar vs. turbulent regime must be more investigated to better augment the metallic cations' release from the anode and avert or decrease the passivation of the electrodes. The emergence of hydrogen from the cathode and oxygen from the anode must be well regulated. In addition, chlorine generation from the anode has to be bypassed or diminished to obviate disinfection by-products formation [49].

In our previous work [50], we have discussed what should be performed to reduce the gap between the EC process (and ED) and the Green Chemistry [51-54].

10. Conclusions

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

1. Through the literature reviewed, EC technology is largely adopted as an encouraging choice for treating actual wastewaters, as important purification performances are reached. Nevertheless, pre or post-treatments are required following the EC usage. Indeed, new methods have been suggested taking into account the integration of EC with additional secondary or tertiary treatments to satisfy the environmental legislation to discharge the treated effluents.
2. Despite the fact that the EC-EO treatment setup suggested by Heffron et al. [37] was not helpful in all water matrices, the enhanced virus reduction obtained by EC-EO in model surface waters attracts more interest. The advantage of EC-EO was possibly not attributed to iron improved oxidation. As an alternative, more important virus mitigation found in the EC-EO treatment train was probably reached by the collective contributions of physical elimination by coagulation/filtration, ferrous iron-based disinfection, and EO disinfection.
3. The original and efficient approach presented by Olvera-Vargas et al. [38] proves that electrochemical technology may be also successful for treating anaerobic sludge. Such interesting efficiencies may be easily obtained provide that the electrochemical techniques are well integrated as hybrid processes with convenient methods and well-designed stages upon optimized conditions. In the same direction, Lalwani et al. [39] established the importance of non-biological treatments, i.e., the electrochemical technology, of drug effluents to avoid microbial drug resistance in nature.
4. Excellent efficiency of EC process is encountered during a huge analysis of related references. The EF action persists basic in dealing with pathogens [55]. In addition, adsorbing microbe onto Fe/Al hydroxides is viewed as a fundamental step in disinfecting water via EC [56]. Much more research requires to be realized to qualitatively and quantitatively choose among EF and cohesion roles. More study has to be pointed on evaluating the more and more likely formation of the hydroxyl radical (OH) during the EC technology [57, 58].

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