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Trends in Decreasing Disinfection By-Products Formation during Electrochemical Technologies

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Abstract

Disinfecting water and wastewater electrochemically is a cost-effective and environmentally friendly alternative for the chemical disinfection. During electrochemical disinfection, the water is passed through an electrolytic cell which is equipped with a set of electrodes. The effectiveness of the process depends upon cell configuration, electrode material, electrolyte composition, microorganism, water flow rate, and current density. One of the main advantages of electrodisinfection is the on-site production of disinfectants; thereby the common drawbacks of chlorination including transportation and storage of hazardous chemicals can be avoided. On the other hand, the high cell voltages due to low electrical conductivity of water and the high capital cost are the main bottlenecks for electrodisinfection. The generation of chlorinated by-products stays the main worry related to electrochemical water treatment processes. This work discusses the main tendencies in dealing with such issues. In some setups, the electrolyte separates the anode and cathode is a proton exchange membrane. This assists to reduce the formation of perchlorate and such conduct is improved in the smallest cell for which the so short contact periods between the electrodes and the water helps to avert the formation of perchlorates when working in a single-pass mode, which becomes a really remarkable point. Other strategies are examined such as developed electrochemical advanced oxidation process, the electroperoxone (E-peroxone) process, which combines ozonation with *in situ* electro-generation of hydrogen peroxide (H_2O_2) from cathodic oxygen reduction.

Electrochemical processes could be merged with nanotechnologies for better efficiency in dealing with pathogens and pollutants removal. In the next future, a hybrid process combining both techniques would be suggested as a part of treatment train for treating water and wastewater.

Subject Areas

Chemical Engineering & Technology

Keywords

Disinfection, Disinfection By-Products (DBPs), Organic Matter (OM), Hydroxyl Radical, Electroperoxone (E-Peroxone), Nanotechnologies

1. Introduction

Disinfection employing electrochemical technologies is beginning to be actually crucial technical dare nowadays [1] [2] [3]. Most of the achievements noticed recently focused on determining the dissimilarities in the disinfection performances obtained among numerous kinds of electrodes [4] [5] [6] [7], which may be employed in such engineering [8] [9] [10] [11]. Many reports worked on averting the generation of disinfection by-products (DBPs) [12] [13] [14] [15] [16]. Some researchers dealt with the design of the electrochemical reactor, which has an extremely fundamental influence on the electrocatalytic conduct of the setup, increasing or decreasing the generation of oxidants [17] [18] [19]. In this context, electrochemical reactors in which the residence period between the electrodes and the water to be disinfected is short could be a good option, since they may participate in avoiding the generation of poisonous species such as chlorate and perchlorate [20] [21] [22] [23] [24]. Such an action plan, which could be realized via employing a single pass of the water to be disinfected across the cell, has been considered as encouraging, even if it is not the definitive manner to deal with such an issue [1] [25]-[30].

Oxidizing chlorides to chlorine is a well-known industrial technique [1] [2] [31] [32] [33] [34]. Chlorine is produced so efficaciously with numerous kinds of electrodes like platinum or mixed metal oxides (MMO) (Equation (1)). While generating chlorine in solution without separation of the anodic and cathodic compartments of the electrochemical setup, chlorine is disproportionated into chloride and hypochlorite (Equations (2) and (3)) [1] [2] [35] [36].

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (1)$$

$$Cl_2 + H_2O \rightarrow HClO + H^+ + Cl^- \quad (2)$$

$$HClO \leftrightarrow H^+ + ClO^- \quad (3)$$

Such a phenomenon is significant when estimating the anodic oxidation of wastewater contaminated with organic matter (OM) since hypochlorite conducts to faster reduction of OM but generating organo-chlorinated species [37] [38]

[39] [40] [41]. This is one of the main barriers to such environmental utilizations presently [1] [42] [43] [44].

Ten years ago, it was observed that chlorine was not the last result of the oxidation of chloride using diamond anodes but, with such electrodes, the response persists until the generation of perchlorates, due to the activity of the hydroxyl radicals produced on their surface [1] [45] [46]. It was an extremely difficult obstacle, as the oxidizing performance reached with such electrodes was much bigger and, indeed, they could form other significant oxidants throughout disinfection (such as ozone, peroxocarbonates or peroxophosphates), which could participate to a more performant technique [1] [47] [48] [49] [50].

Therefore, the dissimilarities between the efficiencies of the electrodes are greatly significant [1]. Oxidizing chloride to chlorine can be directly on the surface of the electrode or mediated by hydroxyl radicals [2] [51]. In the case of the MMO electrode, the hydroxyl radicals produced throughout the oxidation of water are rapidly integrated with the metal oxides of the anode to generate transient higher oxidation state oxides. Consequently, the reaction of chlorinated species ends there and only aging of the hypochlorite solution is in charge of the production of small quantities of chlorate, which had been interpreted in terms of complex chemical pathways. Nevertheless, if employing diamond electrodes, hydroxyl radicals do not interact with the surface of the electrode and they are accessible to merge with hypochlorite, following Equations (4) and (5), leading to the production of perchlorates [1].

$$ClO^- + 2^\bullet OH \rightarrow ClO_3^- + 2H^+ + 2e^- \quad (4)$$

$$ClO_3^- + {}^\bullet OH \rightarrow ClO_4^- + H^+ + e^- \quad (5)$$

As a consequence, to avert the generation of this species, it is so crucial to prohibit the contact between the hydroxyl radicals and the hypochlorite [1].

This work discusses the main tendencies in dealing with such issues. In many setups, the electrolyte that separates the anode and cathode is a proton exchange membrane. This assists to reduce the formation of perchlorate and such conduct is improved in the smallest cell for which the so short contact periods between the electrodes and the water helps to avert the formation of perchlorates when working in a single-pass mode, which becomes a really remarkable point. Other strategies are examined such as developed electrochemical advanced oxidation process, the electroperoxone (E-peroxone) process, which combines ozonation with *in situ* electro-generation of hydrogen peroxide (H_2O_2) from cathodic oxygen reduction. Electrochemical processes could be merged with nanotechnologies for better efficiency in dealing with pathogens and pollutants removal.

2. Averting the Generation of Hazardous Chlorates and Perchlorates through Electro-Disinfection with Diamond Anodes

Isidro *et al*. [1] worked on the prohibition of the production of chlorates and

perchlorates. Their action plan adopted is the reduction in the contact period of the electrode with the electrolyte [52] attempting to avert the interactions of hypochlorite with hydroxyl radicals produced on the surface of the diamond anode. They founded their action plan on former remarks. In juxtaposing the efficiency of two commercial cells fabricated by CONDIAS (Itzehoe, Germany) and equipped with the diamond electrodes, they observed that the formation of perchlorate with one of them was minor or completely inexistent. Such conduct was even more unexpected when juxtaposing the cells as they had precisely the identical fundamental design standards. They wanted to deal with such a dare and to interpret the reasons, focusing on the manner of employing diamond electrodes for killing pathogens without generating perchlorates. As mentioned above, such an objective is important since the electrolytic diamond technique in eliminating microorganisms from water is how to bypass the formation of chlorates and perchlorates throughout the method.

Isidro *et al*. [1] employed a particular kind of commercial cells designed by CONDIAS in two different sizes: the CabECO and the MIKROZON cells (**Figure 1**). In such setups, the electrolyte that separates the anode and cathode is a proton exchange membrane. This assists to reduce the formation of perchlorate and such conduct is improved in the smallest cell for which the so short contact periods between the electrodes and the water helps to avert the formation of perchlorates when working in a single-pass mode, which becomes a really remarkable point. Isidro *et al*. [1] established such excellent efficiency and interpreted the dissimilarities noted in the two cells running with the identical wastewater.

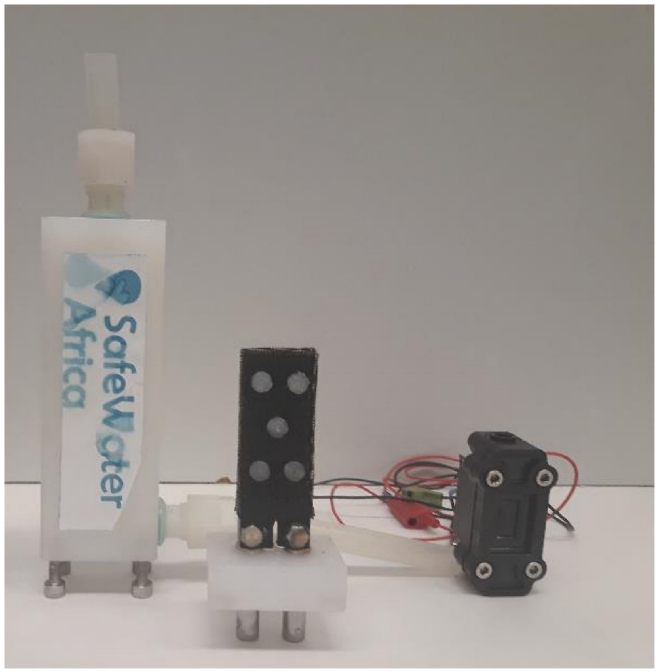

Figure 1. From the left to the right: CabECO (with a detail of its inner part) and MIKROZON cells [1].

Despite utilizing a polymer exchange membrane as electrolyte instead of water, devices consisting of membrane electrode assemblies like the commercial cells CabECO and MIKROZON have the potential to be efficaciously employed for disinfecting water. In fact, such setups were designed to generate ozone in low conductivity water. They could eliminate 3 - 5 logs of CFU with electric charges applied as low as 0.04 Ah/L. The higher the charge applied, the higher is the disinfection level attained [1].

3. Disinfecting Electrochemically Water via Direct and Indirect Oxidation Processes

Rahmani *et al*. [53] studied the electrodisinfection of total coliform (TC) and fecal coliform (FC) with an attention on direct and indirect oxidation utilizing a series of anodes (stainless steel (SS)/lead (Pb) O_2 (SS/PbO_2), stainless steel, titanium (Ti), platinum (Pt), graphite (GP) and Pb/PbO_2). They followed the impact of electrode material, current density (CD), charge passed, initial pH values, different concentrations of NaCl, total dissolved salts (TDSs), electrical conductivity and energy consumption on technique efficiency. The quantity of *in situ* produced active chlorine throughout the method below the best circumstances for each anode was observed to be efficacious in eliminating microbes. The disinfection setup was so performant in killing pathogens in occurrence of 0.01 M NaCl. Augmenting CD from 0.16 to 0.5 mA/cm^2 conducted to a reduction of more than 60% in $\log_{10}$ bacterial load for all electrodes except Pt. Among anodes tried, SS/PbO_2 showed the greatest performance (total demobilization) in 5 min via implementing 0.01M NaCl in CD of 0.5 mA/cm^2 when charge passed was > 15 C. The performance of the anodes in demobilizing TC and FC was in the order of SS/PbO_2 > SS > Ti > Pt > GP > Pb/PbO_2. In spite of the elevated effectiveness of the electrochemical technology in neutralizing TC and FC, indirect oxidation possesses a bigger efficacy thanks to the formation of powerful oxidants like radical hydroxyl and active chlorine (**Figure 2**).

4. Impacts of pH and Reactive Oxygen Species on the Generation of Chlorate and Perchlorate during Electrolysis Utilizing Pt/Ti Electrodes

Jung *et al*. [54] investigated the properties of chlorate (ClO_3^-) and perchlorate (ClO_4^-) generation throughout the electrolysis of water carrying chloride ions (Cl^-). The tests were realized employing an undivided Pt/Ti plate electrode (**Figure 3**) below various pH circumstances (pH 3.6, 5.5, 7.2, 8.0 and 9.0). ClO_3^- and ClO_4^- were produced during electrolysis in proportion to the Cl^- level. The formation rates of ClO_3^- and ClO_4^- below acidic circumstances (pH 3.6 and 5.5) (**Table 1**) [55] were lower than in alkaline pH circumstances (pH 7.2, 8.0 and 9.0). Nevertheless, the pH of the solution did not affect the transformation of ClO_3^- to ClO_4^-. The influences of intermediately produced oxidants on the formation of ClO_3^- and ClO_4^- were detected utilizing sodium thiosulfate

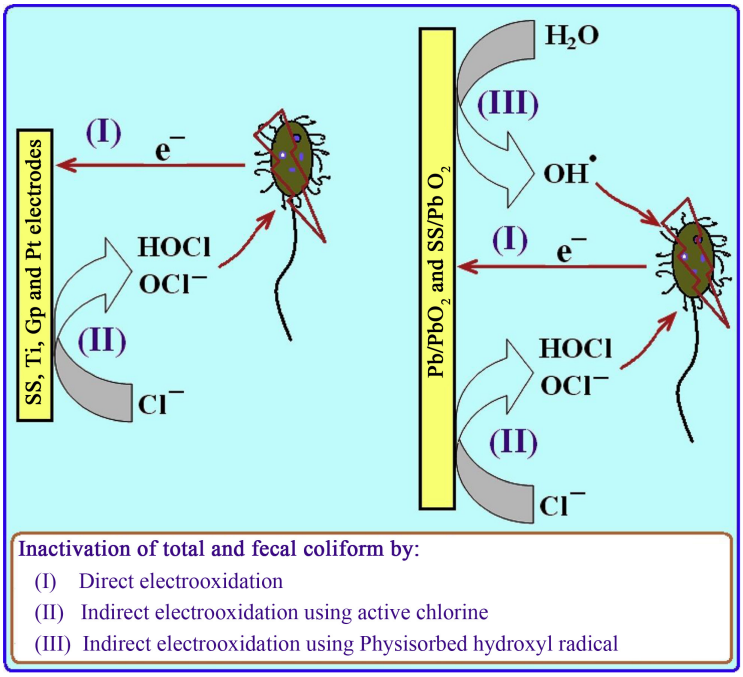

Figure 2. The pathway of TC and FC inactivation by electrodisinfection [53].

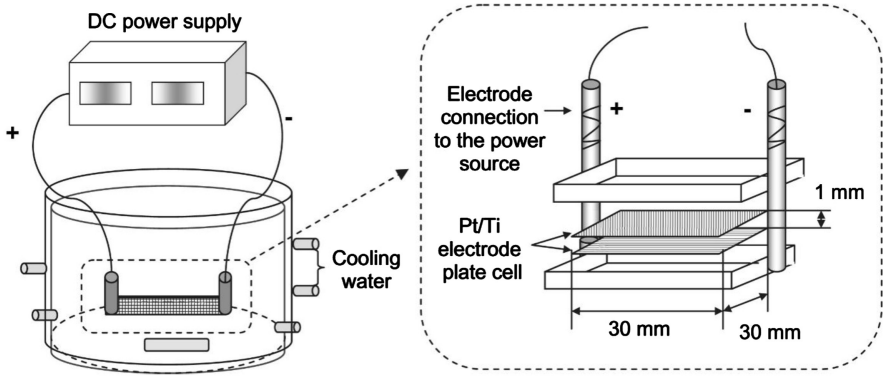

Figure 3. Schematic view of electrolysis device [54].

Table 1. Oxidation power in acidic media of various anode materials used in electrooxidation [55].

Electrode Material	Oxidation Potential (V)	Overpotential of O_2 Evolution (V)	Adsorption Enthalpy of MO_x ($^{\bullet}OH$)	Oxidation Power of the Anode
RuO_2-TiO_2	1.4 - 1.7	0.18	Chemisorption of $^{\bullet}OH$	
IrO_2-Ta_2O_5	1.5 - 1.8	0.25		
Ti/Pt	1.7 - 1.9	0.3		
Ti/PbO_2	1.8 - 2.0	0.5	↑	↓
Ti/SnO_2-Sb_2O_5	1.9 - 2.2	0.7		
p-Si/BDD*	2.2 - 2.6	1.3	Physisorption of $^{\bullet}OH$	

*BDD: Boron-doped diamond.

($Na_2S_2O_3$) as the active chlorine scavenger and tertiary butyl alcohol (*t*-BuOH) as the hydroxyl radical ($^{\bullet}OH$) (**Table 2**) [26] [55] [56] scavenger. Electrolysis reactions that implied active chlorine participated dominantly in ClO_3^- formation (**Figure 4**). The direct oxidation reaction rate of Cl^- to ClO_3^- was 13%. The $^{\bullet}OH$ species that were intermediately produced throughout electrolysis were also observed to importantly touch ClO_3^- and ClO_4^- formation.

5. Nanomaterials for Killing Pathogens

As seen above, chemical technologies of disinfection possess several obstacles and numerous microorganisms have formerly developed resistance versus such traditional methods [57] [58] [59] [60]. Implementing nanomaterials in water treatment and pollutant removal possesses a highly daunting and encouraging future than their conventional counterparts, wiping out restrictions and secondary impacts of classical techniques. Metal oxide nanoparticles, fullerenes, photocatalysts, nanomembranes, and additional similar techniques guarantee a long-term solution for biological contamination of water. Integrating two or more nanomaterials could work as a multipurpose solution to neutralize pathogens as well as dissolved and undissolved impurities. Several of these nanomaterials

Table 2. Thermodynamic formation potentials of several oxidants [26] [55] [56].

Oxidant		Standard Reduction Potential, E^0 (V)
Hydroxyl radical	($H_2O/^{\bullet}OH$)	2.80
Ozone	(O_2/O_3)	2.07
Peroxodisulfate	($SO_4^{2-}/S_2O_8^{2-}$)	2.01
Hydrogen peroxide	(H_2O/H_2O_2)	1.77
Permanganate ion	(MnO_2/MnO_4^-)	1.67
Hypochlorous acid	(Cl^-/HOCl)	1.48
Chlorine	(Cl^-/Cl_2)	1.36
Dichromate	($Cr^{3+}/Cr_2O_7^{2-}$)	1.23
Oxygen	(H_2O/O_2)	1.23
Hypochlorite	(Cl^-/ClO^-)	0.81

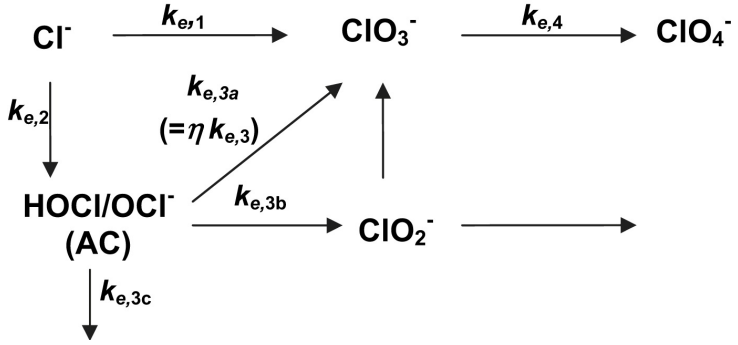

Figure 4. Suggested primary mechanisms of Cl^- oxidation to ClO_4^- throughout electrolysis [54].

possess also increase adsorbent features, which establish new pathways for the development of a hybrid system for water treatment techniques. Many carcinogenic chemicals will be discontinued after their cost-effective establishment as water treatment units. The future of safe drinking water can be thrust upon these "tiny particles." The tailoring nature of these nanomaterials lets the researchers develop more and more sustainable and robust technology. Nevertheless, every technology arrives with some hurdles. As discussed by Ojha [57], there are numerous worries in the nanotechnological utilizations. Such problems require deeper insight and should be duly dealt with. More and more focus on eco-friendly nanomaterials should be made so as to have a perfect technique to combat the water crisis. Methods should be developed with a small cost of production and fewer chemicals required. Biosynthesis and biopolymers or bionanoparticles can play an important role in this regard. Nanomaterials with the least effect on humans and ecology are still to be designed for using them substantially in daily life. Lab to field applications and the studying effect of these nanoparticles in the living system should be taken care of [57].

6. Juxtaposing Carbonized and Graphitized Carbon Fiber Electrodes under Flow-Through Electrode System (FES) for Demobilizing Bacteria

Ni *et al*. [61] assessed the disinfection efficiency of a flow-through electrode system (FES) employing numerous carbonized (C1, C2, and C3) and corresponding graphitized (G1, G2, and G3) carbon fiber felt (CFF) electrodes. They analyzed the physicochemical and electrochemical features to determine the dissimilarities among CFFs. Graphitized CFFs (gCFFs) could attain total demobilization of *Escherichia coli* (>6 log) at the voltage of 3 V and flux of 120 - 3600 L/(m^2 h) for high conductivity and chemical stability, while carbonized CFFs (cCFFs) only reached about 1 log removal with apparent carbon corrosion. For the gCFFs, G1 (>6 log removal) with higher conductivity, better graphite structure, and larger surface area (related to fiber diameter and density) depicted better killing potential at the flow rate of 30 mL/min than G2 (~3 log) and G3 (~1 log). In addition, no regrowth and reactivation of bacteria happened during the storage under visible light illumination after FES treatment. Three parallel FESs with G1 were operated continuously for one week (24 h per day, 7 days) treating the solution with an *E. coli* concentration ranging from 10^6 to 10^7 CFU/mL at the applied voltage of 3 V and the flow rate of 20 mL/min. No live bacteria were detected in the effluent of any of these three FESs (**Figure 5**). In-situ sampling experiments demonstrated that the inactivation of bacteria on anode was the dominant mechanism for FES treatment, which can be attributed to the sequential adsorption, direct-oxidation and desorption process on anode, instead of indirect oxidation by generating chemical oxidants. In addition, hydroxide ion generated from cathode reaction enhanced anode adsorption and inactivation of bacteria by providing alkaline environment. Combining the analysis results of material proper-

ties and disinfection performance, the gCFF-based FES was suggested to be a low-cost, high-efficiency, and safe alternative for future water disinfection [61].

7. Electrocoagulation and Electro-Fenton for Demobilizing Microbiota from Urban Wastewater

Anfruns-Estrada *et al*. [62] focused on juxtaposing the capability of two types of electrochemical technologies, namely electrocoagulation (EC) [63] [64] [65] [66] [67] and electro-Fenton (EF), to disinfect primary and secondary effluents from urban wastewater treatment plants. They tried heterotrophic bacteria, *E. coli*, enterococci, *Clostridium perfringens* spores, somatic coliphages and eukaryotes (amoebae, flagellates, ciliates, and metazoa) as indicator microorganisms. EC with a Fe/Fe cell at 200 A/m^2 and natural pH reached >5 log unit removal of *E. coli* and final concentration below 1 bacteria/mL of coliphages and eukaryotes from both effluents in ~60 min, whereas heterotrophic bacteria, enterococci and spores were more resistant. A bigger reduction was attained for the primary effluent, possibly due to the flocs that eliminate a bigger quantity of total organic carbon (TOC), enmeshing more easily the microbiota [68] [69] [70]. EF with a BDD anode and an air-diffusion cathode that generates H_2O_2 on site was first realized at pH 3.0, with huge or even complete neutralization of pathogens for 30 min. A more efficacious microorganism reduction was obtained as juxtaposed to EC because of $^{\bullet}OH$ produced from Fenton's response. Faster disinfection was

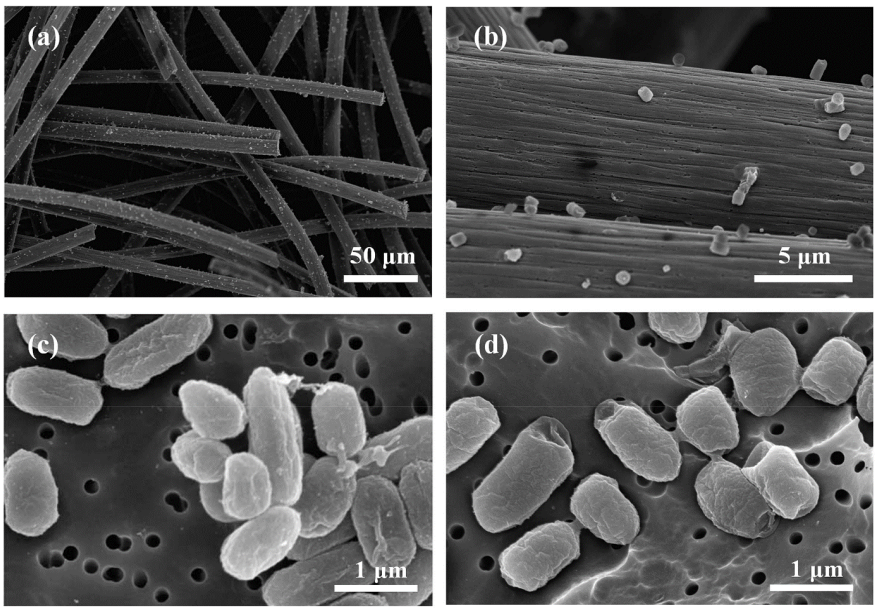

Figure 5. SEM image of the *E. coli* on the G1 fibers and in the water samples. (a) The *E. coli* adsorbed on the G1 fibers at the voltage of 3 V and flow rate of 30 mL/min under 450 × magnification. (b) The *E. coli* adsorbed on the G1 fiber at the voltage of 3 V and flow rate of 30 mL/min under 6000 × magnification. (c) The morphology of live *E. coli* in the influent sample under 25,000 × magnification. (d) The morphology of dead *E. coli* in the effluent sample after 3V, 30 mL/min treatment using G1 as the electrode under 25,000 × magnification [61].

detected for the secondary effluent thanks to its smaller TOC content, letting the attack of bigger amounts of electrogenerated oxidants on microbes. Disinfecting wastewater via EF was also possible at pH ~7, depicting identical removal of active pathogens as a result of the interactive action of produced oxidants such as active chlorine and coagulation with iron hydroxides. A consecutive EC/EF treatment (30 min each) was more efficient for integrated decontamination and disinfection of domestic wastewater (Figure 6).

8. Cathodic Hydrogen Peroxide Production for Reducing Chlorinated By-Products Generation

Yao *et al*. [71] studied the generation of chlorinated by-products throughout surface water treatment via a freshly developed electrochemical advanced oxidation process, the electroperoxone (E-peroxone) process, which combines ozonation with *in situ* electro-generation of hydrogen peroxide (H_2O_2) from cathodic

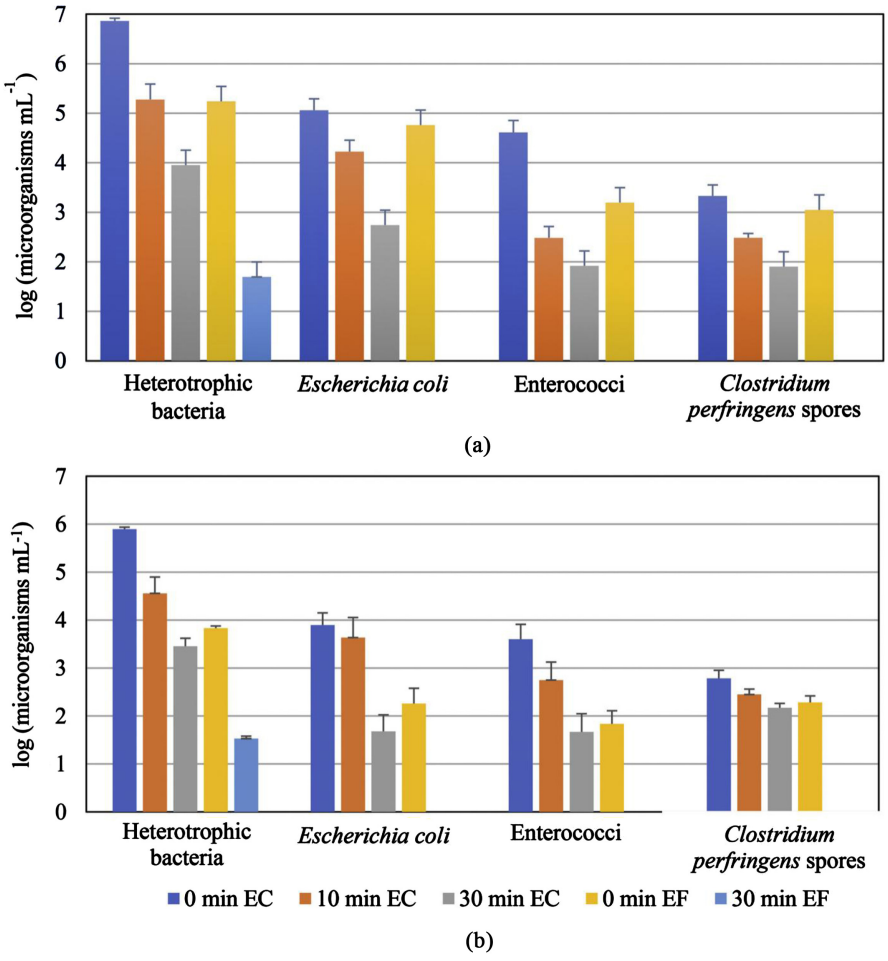

Figure 6. Change of the logarithm of the concentration of the microorganisms upon sequential EC/EF treatment (30 min each). The EC step was applied to 200 mL of (a) primary and (b) secondary effluents at j = 200 A/m^2 and 25˚C, being followed by the EF step with 100 mL of the resulting solution, at pH ~ 7 and j = 333 A/m^2 [62].

oxygen reduction (Figure 7). Thanks to the improved ozone (O_3) transformation to hydroxyl radicals ($^{\bullet}OH$) by electro-generated H_2O_2, the E-peroxone process greatly increased the removal of ozone-refractory micropollutants like clofibric acid and chloramphenicol in the selected surface water juxtaposed to traditional ozonation. Further, the cathodically produced H_2O_2 efficiently quenched hypochlorous acid (HOCl) derived from the anodic oxidation of chloride in the surface water. Consequently, the generation of trichloromethane (TCM) and chloroacetic acids (CAAs) from the responses of HOCl with dissolved organic matter (DOM) was negligible throughout the E-peroxone process, and identical concentrations of TCM and CAAs were usually detected in the traditional ozonation and E-peroxone treated water. In contrast, important quantities of HOCl could be formed from the anodic oxidation of chloride and then accumulated in the surface water throughout the traditional electrolysis process, which conducted to considerably bigger levels of TCM and CAAs in the electrolysis treated water. The findings of such research propose that the E-peroxone process could overtake the main barrier of classical electrochemical techniques and furnish an efficacious and secure EAOP option for micropollutant removal during water treatment.

9. Conclusions

From this work, the main conclusions emerge:

1) Generation of chlorate and perchlorates throughout electrodisinfection could be avoided with the usage of cells with very low contact time between the water and the electrode and a single-pass strategy. This is explained by the fact that further oxidation of hypochlorite to chlorates and perchlorates could only happen by the direct reaction of hydroxyl radicals which takes place mostly in the close proximity to the anodes [1]. Via connecting cells in series, the disinfection obtained could be augmented and more than 8-logs have been reached [1]. In any case the N/N_0 follows the same decay trend with respect to the current charge passed regardless of the number of cells attached. Even increasing residence

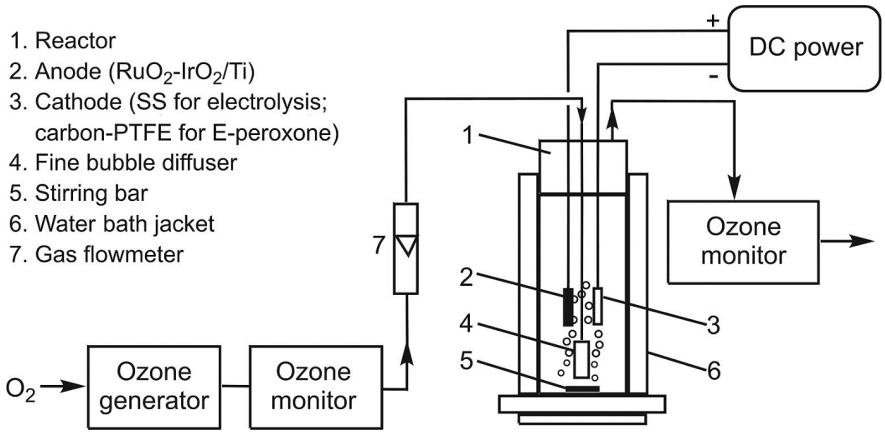

Figure 7. Schematic of the reactor used for electrolysis, conventional ozonation, and the E-peroxone process [71].

time four-fold, the concentration of chlorates is very low and close to the detection limit of the analytical technique used.

2) Employing a Ti/IrO_2 anode was efficient for disinfecting aqueduct water. The disinfection rate corresponded to the applied current density. Concerning the specific charge passed, the highest current density (8 mA/cm^2) depicted the greatest rate of disinfection; this corresponded to the increased levels of active chlorine formation in the highest current density tests. Nevertheless, DBPs generation and energy consumption related to the highest current density were bigger than the lower current densities for a specific degree of disinfection. Efficient disinfection without regulatory exceedances in some DBPs could be reached employing a Ti/IrO_2 anode for handling surface water [36].

3) Electrochemical engineering possesses the capacity to become a so crucial technology for dealing with almost any type of contaminated water. It is a strong method for killing pathogens and decomposing toxic and recalcitrant organics. Nevertheless, electrochemical methods stay on the edge of industrial and commercial breakthrough, and more comprehension on the complicated chemistry in the cell and the engineering perspectives in designing and optimizing an electrochemical treatment unit are required before the technique could be viewed as a mature and ready applicable treatment solution [55]. Nanotechnologies could be merged with electrochemical processes for better efficiency in dealing with pathogens and pollutants removal. Separately, both technologies have shown their efficiency in eliminating microorganisms and degrading organic and inorganic contaminants. In the next future, a hybrid process combining both techniques would be suggested as a part of treatment train for treating water and wastewater [72]. On the other hand, the technique of adsorption and electrochemical regeneration employing the graphite intercalation compound (GIC) adsorbents was observed to be efficient in reducing several bacteria (*P. aeruginosa*, *S. aureus* and *L. pneumoph*), fungi, and yeasts. The aptness of the method was also estimated for the fungal species *A. awamori*. Further, the technique was also discovered to be performant in disinfecting yeasts comprising *S. cerevisiae* and *R. turoloides*. However, the disinfection of *C. parvum* via adsorption and electrochemical regeneration employing the GIC adsorbent was not depicted successfully. In this context, an early examination concerning *C. parvum* proposes that using a chloride-free solution in the cathode compartment and a comparatively high current density can be efficacious [73].

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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