



# Disinfection By-Products (DBPs) Control Strategies in Electrodisinfection

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**How to cite this paper:** Ghernaout, D. and Elboughdiri, N. (2020) Disinfection By-Products (DBPs) Control Strategies in Electrodisinfection. *Open Access Library Journal*, 7: e6396.

<https://doi.org/10.4236/oalib.1106396>

**Received:** May 3, 2020

**Accepted:** May 17, 2020

**Published:** May 20, 2020

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## Abstract

Chemical water treatment problems such as disinfection by-products (DBPs) generation have urged on the search for better water treatment technologies such as electrochemical water technologies that have been applied successfully in different water/wastewater pollutants removal. However, their large expansion is hindered by similar DBPs troubles. Throughout the electrochemical process, such carcinogenic substances can be produced depending on the electrode material and applied voltage. This work aims to discuss recent advances recorded in dealing with DBPs formation in electrochemical devices. Numerous sophisticated techniques are lately suggested such as an interesting method employing carbon felt cathodes in which DBPs are less formed, and another judicious method utilizing boron-doped diamond anodes in which perchlorate production is decreased. Many action plans for removing halides from water to reduce DBPs are also listed. Combining electrochemical processes and their merging with nanotechnologies for better efficiency in dealing with pathogens and DBPs removal are suggested. Secure multi-barrier techniques, like distillation, granular activated carbon, and membrane processes have proven their excellent effectiveness in eliminating pathogens and pollutants. Employing those invincible technologies, thanks to their relatively low costs and ease of applications, is an encouraging domain of research with a perspective to bypass the DBPs formation during the efficient electrochemical processes.

## Subject Areas

Chemical Engineering & Technology, Electrochemistry

## Keywords

Disinfection by-Products (DBPs), Chlorinated by-Products (CBPs), Electrodisinfection (ED), Carbon Felt Cathode (CFC),

## 1. Introduction

Distributed small-scale water treatment units have been generally employed both in residents' buildings and in public facilities (like hospitals, schools, and malls), to meet the search for high-quality potable water via eliminating heavy metals, hardness, toxins, etc. [1] [2] [3]. Such cleaners do not every time work in a continuous manner, because of work schedule or vacation leave [4] [5] [6]. Discontinuous functioning of the apparatus leads to the occurrence of water stagnation and then augments the microbiome development and pathogenic communities, making possible biological hazards to consumers [7] [8] [9]. However, such distributed cleaners are habitually out of the range of inspection thus they were not paid sufficient notice to [10] [11] [12]. Cleaners behave like microbial contamination sources at a certain level and re-disinfection procedures are urgently required [13] [14] [15]. Re-disinfection remains a professional procedure; however, it is not handy for residents [1] [16] [17]. Therefore, it arises a problem that cleaners could ameliorate water quality whereas intermittent water supply leads to possible microbial pollution [18] [19] [20]. Electrolysis is encouraging to resolve the problem at the last meter of drinking water supply [1] [21] [22].

Investigations on disinfection via electrolysis in the potable water treatment field were mentioned rarely [1] [23] [24]. Electrolysis has been considered as a substitutional technique for disinfecting potable water [25] [26] [27]. Electrolysis is not recently born [28] [29] [30]; however, it is less utilized or less current, since power was so costly when electrolysis first came out to the world and electrode materials were also an obstacle [31] [32] [33]. Lately, cost of power diminished obviously and green power such as solar gradually expanded [34] [35] [36], and more innovative electrode materials have been established well efficient in resistance to corrosion and in reducing disinfection by-products (DBPs) precursors [37] [38] [39]. For instance, platinum anode depicted elevated efficacy in forming more active chlorine [40] [41] at lower current densities and that their lifetime was not diminished via regular polarity reversal [42] [43] [44]. Stainless steel was usually selected as a cathode for hygiene and anti-corrosion performances [45] [46] [47]. Such signs of progress are improving the competitiveness and technical maturity of electrolysis technology [48] [49] [50]. Further, the most implemented event was to furnish electrolyzed sodium hypochlorite in more distributed water supply systems for chlorination lately [21] [51] [52]. Researchers [53] suggested the disinfection techniques of "liquid-liquid" via moderately acidic electrolysis [54] [55] [56]. Nevertheless, there is a lag phase (10 - 50 min) in the phenomenon of electrolysis disinfection or chlorination [26] [37] [57]. This implicates that the consumer has to wait for at least 10 min to drink safe water biologically, even if he commonly directly collects water and drinks it at the same time, making the hazard of absorbing pathogenic microbes [58] [59]

[60]. Conserving electrolyzed water safe biologically in the intermittent system is very important [29]. Despite the fact that much effort for enhancing electrolysis utilization in potable water disinfection has been performed until now, more researches require to be dedicated to improving electrolysis disinfection in the intermittent drinking water supply to remove microbial pollution provoked by water stagnation [1] [29] [61].

This work discussed many advances recorded in dealing with DBPs formation in ED devices. DBPs control procedures in distributed potable water supply using electrolysis are presented. An interesting technique employing carbon felt cathodes (CFCs) in which DBPs are less formed is also discussed. Another judicious method utilizing boron-doped diamond (BDD) anodes in which perchlorate production is decreased is presented. Reducing DBPs generation via cathodic  $H_2O_2$  production through electro-peroxone (E-peroxone) technique is briefly reviewed. Several action plans for removing halides from water to reduce DBPs are also listed. Combining electrochemical techniques for treating water is proved efficient in reducing DBPs, an illustration of such integrations is discussed.

## 2. Distributed Potable Water Supply Employing Electrolysis: DBPs Monitoring

Recently, Chen *et al.* [1] investigated and juxtaposed the anti-bacterial efficacy and DBPs potential in various stagnated water samples with techniques of electrolysis and chlorination. They found that electrolysis must be performed during at least 30 min to reduce heterotrophic plate counts under the hygiene standard (<500 colony-forming units (CFUs)/mL), which was identical to chlorination and was better than hydraulic cleaning. They suggested a viable but non-culturable calculation model to estimate the viability of bacteria following electrolysis and chlorination. Electrolysis depicted better effectiveness than chlorination in reducing the fraction of viable but non-culturable bacteria and opportunistic pathogens. Moreover, electrolysis participated in DBPs monitoring. They employed differential spectra to estimate DBPs potential, and the estimates of DlnA350 and DA272 were smaller in electrolyzed water than those in chlorinated water. They utilized standard queueing theory and multi-objective programming to regulate electrolysis. Reducing the waiting time of disinfection and the total running time were the main objectives of optimization. Circulation flow electrolysis and pulse chlorination can keep outlets from distributed purifiers safe biologically in full time, and decrease the waiting period for consumers to collect safe water. The ideal running and pausing time were 0.5 h and 2 h, to diminish the mathematical expectation of waiting time to none [1].

## 3. Electrochemical Remedy of Urban Wastewater via Carbon Felt Cathodes (CFCs)

Cotillas *et al.* [62] used electrolysis with CFCs for the remedy of real effluents from urban wastewater treatment facilities (WWTFs) in integration with nu-

merous anode materials (dimensionally stable anodes-DSAs, conductive diamond anodes-CDAs and iron-Fe). The efficiency of electrodisinfection (ED) with CDA and DSA was evaluated, proving that total removal of *Escherichia coli* could be obtained at applied electric charges under  $0.03 \text{ Ah/dm}^3$ , and that the disinfection technique is more efficacious when employing CDA. In addition, the generation of hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) on CFCs restricts the concurrence of DBPs (chlorates, perchlorates, and organic chlorinated by-products (CBPs)), an important finding that widens the potential of CDA for the restoration of domestic wastewater. Utilizing Fe anodes illustrates that it is easy to reach the total elimination of microbes with analogous performance to that of CDA (thanks to the participation of Fenton's response) and that it is probable to fully eliminate the turbidity of the effluent when running at current densities from  $12.50 \text{ A/m}^2$ . Further, Fe is established as the most efficacious anode material (lowest power consumption) at low current densities and CDA is the most suitable one at current densities bigger than  $5 \text{ A/m}^2$ . The anode-cathode CDA-carbon felt and Fe-carbon felt pairs seem to be the most encouraging electrode materials to be utilized in wastewater remedy technologies.

#### **4. Formation of Perchlorate throughout Water Electrolysis Employing Boron-Doped Diamond (BDD) Anodes**

Bergmann *et al.* [63] performed electrochemical investigations to assess the possibilities of perchlorate production in potable water disinfected via direct electrolysis. They employed BDD anodes in the laboratory and commercially obtainable cells at  $20^\circ\text{C}$ . They varied the current density from 50 to  $500 \text{ A/m}^2$ . Further, additional anode materials like platinum and mixed oxide were also tried. They noted that BDD anodes have a thousand fold higher perchlorate formation potential juxtaposed to the other electrode materials that were verified. In long-term discontinuous tests, all the chloride finally reacted to produce perchlorate. The identical finding was obtained when probable oxychlorine intermediates ( $\text{OCl}^-$ ,  $\text{ClO}_2^-$ ,  $\text{ClO}_3^-$ ) were electrolyzed in synthetic waters in the ppm domain of levels. The trend to generate perchlorate was affirmed when the flowrate of potable water was changed between 100 and  $300 \text{ L/h}$  and the temperature augmented to  $30^\circ\text{C}$ . In a continuous flow mode of operation, a higher chloride level in the water resulted in a lower perchlorate production. This would be interpreted via reaction competition of species near and on the anode surface for trials both with synthetic and local potable waters. They concluded that the usage of electrodes forming highly reactive species must be more carefully restrained in hygienically and environmentally oriented uses.

#### **5. Chlorine Dioxide Generation in the Electrodisinfection (ED) of Drinking Water**

Bergmann and Koparal [64] focused on issues of chlorine dioxide generation and responses throughout and following electrolysis. They worked on synthetic

and actual potable waters employing titanium anodes with IrO<sub>2</sub>/RuO<sub>2</sub> coatings. They followed the impact of chloride level up to 250 mg/L, current density (up to 500 A/m<sup>2</sup>), and additional variables on ClO<sub>2</sub> production. The ClO<sub>2</sub> generation is proportional to the Cl<sup>-</sup> level. Important effects of pH and rotation rate could not be found until now studying the electrochemical method. They showed an electrochemical pathway of ClO<sub>2</sub> formation possibly from chloride or active chlorine.

## 6. Reducing DBPs Generation via Cathodic H<sub>2</sub>O<sub>2</sub> Production through E-Peroxone Technique

Yao *et al.* [65] studied the generation of CBPs during surface water treatment by a recent suggested electrochemical advanced oxidation process (EAOP), the electro-peroxone (E-peroxone) process, which combines ozonation with *in situ* electro-generation of H<sub>2</sub>O<sub>2</sub> from cathodic oxygen reduction. Because of the improved ozone (O<sub>3</sub>) transformation to hydroxyl radicals (<sup>•</sup>OH) via electro-generated H<sub>2</sub>O<sub>2</sub>, the E-peroxone process significantly increased the reduction of ozone-refractory micropollutants like clofibric acid and chloramphenicol in the chosen surface water confronted to traditional ozonation. Further, the cathodically produced H<sub>2</sub>O<sub>2</sub> 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 reactions of HOCl with dissolved organic matter (DOM) was negligible through the E-peroxone process, and comparable concentrations of TCM and CAAs were mostly detected in the traditional ozonation and E-peroxone treated water. In contrast, important quantities of HOCl can be produced from the anodic oxidation of chloride and then gathered in the surface water throughout the traditional electrolysis method, which conducted to considerably higher levels of TCM and CAAs in the electrolysis treated water. Such findings propose that the E-peroxone method could overcome the main restriction of traditional electrochemical techniques and furnish an efficient and secure EAOP choice for micropollutant removal via water treatment.

## 7. Combining Sonochemical and Electrodisinfection (ED) with DSA Anodes

Cotillas *et al.* [66] worked on the disinfection of real domestic wastewater via merging ultrasound (US) irradiation and ED with DSAs. They examined the inactivation of *E. coli* throughout the sonochemical disinfection at elevating US power. It was not easy to obtain total disinfection, even at the highest US power (200 W) dosed by the experimental setup employed. Disinfecting electrochemically with DSA anodes at various current established that it was required a minimum current density of 11.46 A/m<sup>2</sup> to attain the total disinfection. A merged sono-electrodisinfection process depicted an interactive effect when integrating US irradiation with DSA ED, with a synergy coefficient greater than 200% of the

disinfection rate obtained for the highest US power utilized. During such a technique, hypochlorite and chloramines were found as the major products for the disinfection process (neither chlorate nor perchlorate was detected), and the occurrence of trihalomethanes (THMs) was far under admissible values. Establishing such interactive influence with DSA anodes suggests new efficacious disinfection technology, restricting the production of toxic DBPs.

## 8. Action Plans for Eliminating Halides from Water to Diminish DBPs

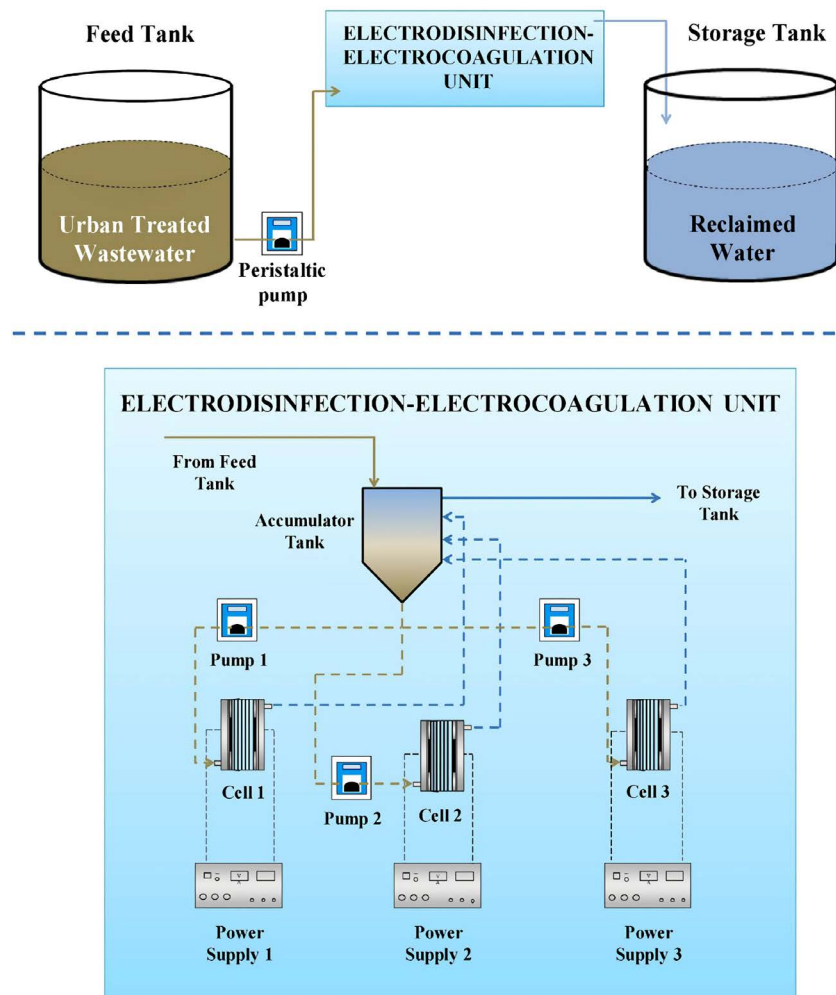
The occurrence of bromide ( $\text{Br}^-$ ) and iodide ( $\text{I}^-$ ) in source waters conducts to the generation of brominated and iodinated DBPs, which are frequently more poisonous than their chlorinated analogs. Watson *et al.* [67] published a summary of research into bromide and iodide reduction from potable water sources. Bromide and iodide elimination methods have been generally categorized into three classes: membrane [68] [69] [70], electrochemical and adsorptive technologies. They discussed reverse osmosis (RO) [71] [72], nanofiltration (NF), and electro-dialysis membrane processes. The electrochemical methods examined are electrolysis, capacitive deionization, and membrane capacitive deionization [73]. Investigations on bromide and iodide removal employing adsorptive methods included layered double hydroxides (LDHs), impregnated activated carbons, carbon aerogels, ion exchange resins, aluminum coagulation [74] [75] [76]. They juxtaposed halide removal techniques have been compared, and defined areas for future study. Research trends may be summarized as follows [67]:

1) Numerous fields in which more investigation is required to evaluate the bromide and iodide removal potential of treatment engineering have been determined. The major fields related to DBPs control in potable water via reducing halide precursors are optimization of techniques like electro-dialysis reversal, electrolysis, LDHs, silver-doped aerogels, and resins for the water treatment plant scale; adaptation for commercial production/application of resins, soils and aerogels; diminishing the impacts of competing anions and natural organic matter (NOM) [28] [77] [78] on adsorptive methods for halide reduction; enhancing energy efficiency of membrane processes, etc. [67].

2) There is a global demand for more expansion of halide reduction technologies able to be utilized in industrial water treatment implementations [79] [80] [81]. Most halide reduction researches have examined reductions of particular anions in isolation from other halides and other potentially competing anions. There is a demand to evaluate halide removal techniques towards both bromide and iodide, as well as competing anions. Eliminating both halides and NOM at the same time constitutes the ideal solution for DBP monitoring via precursor elimination [82] [83] [84]. Further potential is in the expansion of hybrid technologies from the integration of less efficient/limited methods, to attain this optimum objective, of efficacious NOM and halide elimination for DBP minimization [67] [85] [86].

## 9. Electrodisinfection-Electrocoagulation (ED-EC) Process for Wastewater Reclamation

Cotillas *et al.* [87] suggested the scale-up of a merged electrodisinfection-electrocoagulation (ED-EC) technique, particularly adapted to the recuperation of real domestic treated wastewater, equipped with BDD anodes and Fe bipolar electrodes. The setup works in continuous mode and in the prototype the anode area was augmented three times (anodic oxidation) and the bipolar electrode area fifteen times (EC) with respect to the system employed at bench scale (**Figure 1**). It is feasible to reach the full and concomitant disinfection and turbidity elimination through applying current densities within the domain 5 - 10 A/m<sup>2</sup>. Free and combined chlorine species were electrogenerated from the chloride contained in the effluents (no reagents were injected) being these species in charge of killing pathogens. Also, Fe coagulant species coming from the electro-dissolution of the anodic side of bipolar electrodes support turbidity elimination. In the scaled-up prototype, a more performant turbidity reduction was attained, thanks to the



**Figure 1.** Electrodisinfection-electrocoagulation (ED-EC) pilot plant for wastewater reclamation [87].

augmentation of the bipolar electrode area. It was established that for electric charges under  $0.07 \text{ kAh/m}^3$  the recuperation of domestic treated wastewater can be obtained, averting the generation of toxic chlorates and perchlorates even at current densities bigger than  $7 \text{ A/m}^2$ .

## 10. Conclusions

From this work, the following conclusions can be drawn:

1) Transforming traditional ozonation to the E-peroxone method will not greatly augment the production of CBPs throughout the treatment of chloride-containing water [65]. Even with comparatively elevated  $\text{Cl}^-$  levels and an anode that possesses elevated electrocatalytic activity for chlorine formation, the rate of chlorine generation at the anode was much slower than that of  $\text{H}_2\text{O}_2$  formation from  $\text{O}_2$  reduction at the cathode. Because of the quenching of HOCl via  $\text{H}_2\text{O}_2$ , residual chlorine was not observed throughout the E-peroxone treatment of the chosen surface water. Consequently, identical concentrations of TCM and CAAs were usually detected in the water treated via traditional ozonation and the E-peroxone method. Further, thanks to the acceleration of  $\text{O}_3$  decay and quenching of HOBr by electrogenerated  $\text{H}_2\text{O}_2$ , the E-peroxone method considerably decreased the generation of bromate throughout the treatment of bromide-containing water juxtaposed to traditional ozonation. Such findings show that unlike traditional electrochemical techniques, the productions of halide-derived by-products are not a main worry for the E-peroxone process throughout water treatment.

2) The killing pathogens rate of a sonochemical method augments with US power irradiated but it is not feasible to obtain the total disinfection of the effluent in the span of US power utilized by Cotillas *et al.* [66]. ED employing DSA anodes is an efficacious technique to neutralize microbes in wastewater. Synergies bigger than 200% of disinfection rate are noted when merging ultrasound irradiation and ED with DSA anodes. Chlorate and perchlorate were not found, being hypochlorite and chloramines the major responsibility of the disinfection method. It was established that the US ameliorates mass transfer phenomena and thus, it enhances the formation of disinfectant species in the solution. Also, the US favors the suppression of the agglomeration of *E. coli* cells in the bulk, permitting an efficacious attack of disinfectant species to pathogens. Such findings open large perspectives of suggesting excellent disinfection systems that avert the DBPs formation.

3) Employing granular activated carbon post-treatment could greatly reduce the concentrations and poisonous effects of DBPs. Moreover, secure multi-barrier techniques, like distillation and membrane processes, remain to be suggested, tested, and industrially encouraged. Despite their limitations, both adsorptive techniques and membrane processes persist to be an encouraging domain of research thanks to their relatively low costs and ease of applications [88] [89] [90].



## Acknowledgements

This research has been funded by the Research Deanship of University of Ha'il, Saudi Arabia, through the Project RG-191190.

## Conflicts of Interest

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

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