

**Communication**

Microorganisms' Killing: Chemical Disinfection vs. Electrodisinfection

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Abstract: Chlorination is the most used technique of killing microorganisms' in water through the potable water industry. Some outbreaks of water born disease, the definition of chlorine as a source of greatly toxic disinfection by-products (DBPs), and the appearance of recalcitrant microbes have conducted to revised regulation for the elimination of microorganisms and DBPs from potable water. Therefore, researching new disinfection techniques has been developed. Electrochemical disinfection or electrodisinfection (ED) has appeared as one of the more valuable alternatives to chlorination. Research employing a range of cell designs has illustrated ED to be efficient towards an interval of microbes. Nevertheless, in several cases, killing pathogens' performance seems to be linked to the production of chlorine species. The obvious dominance of chlorine in the form of the pathway of killing microbes' emerges the interrogation if ED is really more beneficial than chlorination in a matter of its demobilization performance and risk to generate DBPs. Convenient ED devices must be designed and monitored sophisticatedly since the present state of non-monitored use of ED devices is not favorable in terms of hygienic and health risks considerations. Great works remain to be performed.

Keywords: Electrodisinfection (ED), Chlorination, Monochloramination, Ozonation, Drinking Water, Disinfection By-products (DBPs), Reactive Oxygen Species (ROSs)

1. Introduction

To protect humans from water-borne diseases through pathogenic microbes, killing microorganisms is a crucial potable water treatment stage [1-6]. Ordinary disinfection processes comprise chlorine, monochloramine, chlorine dioxide, and advanced disinfection techniques concern ozone, hydrogen peroxide, UV and electrochemical treatment [7-10]. Considered as the most used method through all ages until now, chlorination is also well known for its disinfection by-products (DBPs) formed [11-17]. As an alternative technique, monochloramine may keep the chlorine disinfecting features and decrease the production of DBPs [18, 19]. Considered as a powerful oxidant, ozone may deactivate several types of

microbes efficiently [20] with the reduced generation of DBPs [21, 22]. On the other hand, electrochemical methods in both chloride and chloride-free electrolytes were established to be greatly performant in killing microorganisms; they were also found successful for *Cryptosporidium parvum* oocysts and *Clostridium perfringens* spores, which were resistant to chlorine [23-25]. The electrochemistry-based techniques are interesting thanks to the powerful oxidants formed during the electrolysis [26]. Deactivating performances were particularly astonishing with the boron-doped diamond (BDD) anode, whose scarce feature had been previously illustrated in degrading organic compounds [27-30].

It is crucial to comprehend the pathways of various deactivation techniques; this understanding would be useful to define the main phases in killing pathogens and to describe more

efficient procedures in optimizing real disinfection [31, 32]. Comparing various disinfection methods has been already mentioned in different reviews [33]. Monochloramine is an oxidant with comparatively low reactivity regarding most chemical functional groups [34]. Its killing microorganisms' capacity is much smaller than chlorine [35]. Ozone was found to be more powerful than chlorine and monochloramine in deactivating *Cryptosporidium parvum* oocysts [20, 36] as well as *Bacillus subtilis* (BST) spores [37, 38]. Researchers examined the killing *Escherichia coli* performances of ozone and free chlorine [39]. It was proven that ozone caused the most important degree of surface demolition, at the same time the weak destruction through free chlorine. Ozone possesses a greater oxidizing capacity to enter in reactions with the organic constituents of the cell membrane before entering inside the cell plasma; these routes generate important surface demolition. For chlorine, with a less significant oxidizing capacity, entering in reactions with the organic compounds [40] of the cell envelope is restricted and deactivating microbes was achieved by its mutual actions with intracellular constituents [39]. Several illustrations have been mentioned that disinfected water electrochemically possess a bigger disinfecting activity relatively to usual hypochlorite solutions found in the market [41, 42]; however, it is not easy to compare between them because of the adjustment of equal starting conditions like initial oxidant concentration. As an illustration, researchers [43, 44] considered *E. coli* as the indicator microbe in the electrochemical disinfection (or electrodisinfection, ED). They compared ED with ozonation and chlorination. Their scanning electron microscopy (SEM) observations following various methods of killing microorganisms as well proposed that microbes in the electrochemistry-based process employing chloride as supporting electrolyte [45] were probably eliminated through the chemicals with germicidal capacities identical to that of ozone and much powerful than that of chlorine. Nevertheless, emphasizing the identical situations for comparing purpose was not easy. Some researchers affirmed that electrolyzed water (50 mg/L Cl⁻) possess an elevated lethal capacity than Ca(OCl)₂ of the identical determined active chlorine concentration [46]. In fact, it is not convenient to compare them not taking into account all oxidative species in the medium. Probably the gap in killing microbes' performance aroused from particular features of the present species in different techniques.

As seen above, *E. coli* is hugely used as an indicator microbe for bacteriologic water quality. Nevertheless, deactivating *E. coli* has not the possibility to symbolize mechanically all types of microorganisms' killing. Researchers established that a total elimination of a bacterial population in water after 5 min of electrolysis was realized using 20 mA in the condition of *E. coli*; however, employing 30 mA when treating *Legionella* (*Legionella pneumophila*) [47, 48]. Thus, *E. coli* appears to be more sensitive to killing microbes' electrochemically comparatively with *L. pneumophila*. Bergmann *et al.* [46] worked on three microbes for chlorination, ozonation and ED. They found that *E. coli* cells were more vulnerable than BST and *Saccharomyces cerevisiae* Kolin cells. In addition, it is mentioned that Gram-positive *Staphylococcus aureus* shows more powerful drug-resistance than Gram-negative colon *Bacillus* because of their varying peptidoglycan configurations [49]. Tyrrell *et al.* [50] experimented five indigenous populations (fecal coliforms, enterococci, *C. perfringens*, male-specific bacteriophage and somatic coliphage) in secondary sewage effluents employing Cl_{2(g)} and O_{3(g)}. Deactivating result changed enormously with each other and it is established that only one bacterium indicator, like the fecal coliform, is not convenient for predicting microbe responses in killing microorganisms' processes [24, 51].

On the other hand, the reactive oxygen species (ROs), mostly comprising [•]OH formed during electrolysis were suggested to contribute importantly in ED in the absence of chloride [41, 43]. Nevertheless, comparing electrochemistry-based technology with different disinfection techniques in terms of removing various types of microorganisms was not largely investigated. Even if killing bacteria electrochemically is valuable comparatively with the remaining disinfection methods, it would be helpful to show and establish the fundamental action of [•]OH in electrolysis's route.

Li *et al.* [3] employed four bacteria (*E. coli*, *S. aureus*, BST, an isolated *Bacillus*) as the indicator microorganisms to compare four disinfection methods comprising chlorination, monochloramination, ozonation and ED. They focused on the difference in deactivating results and the SEM analysis for the four bacteria and verified the particular contribution of [•]OH existing in the BDD anode system as a free state in ED without chloride (Figures 1-4). They found that:

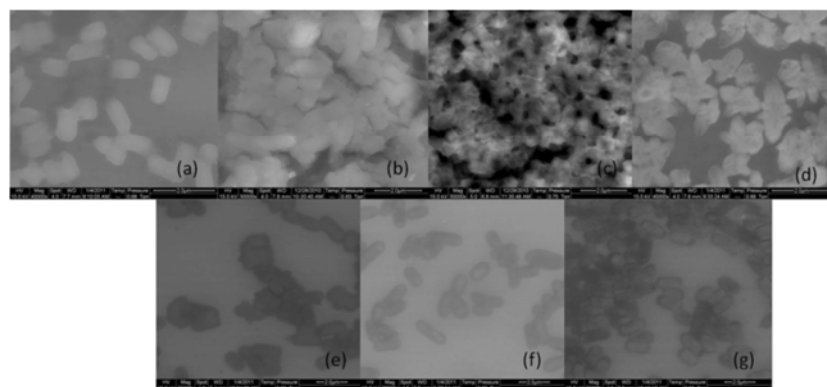


Figure 1. SEM observations of *E. coli*. (a) Untreated cells; (b) cells after 30 min electrolysis; (c) cells after 60 min electrolysis; (d) cells after 150 min electrolysis; (e) cells after ozonation; (f) cells after chlorination; (g) cells after monochloramination [3].

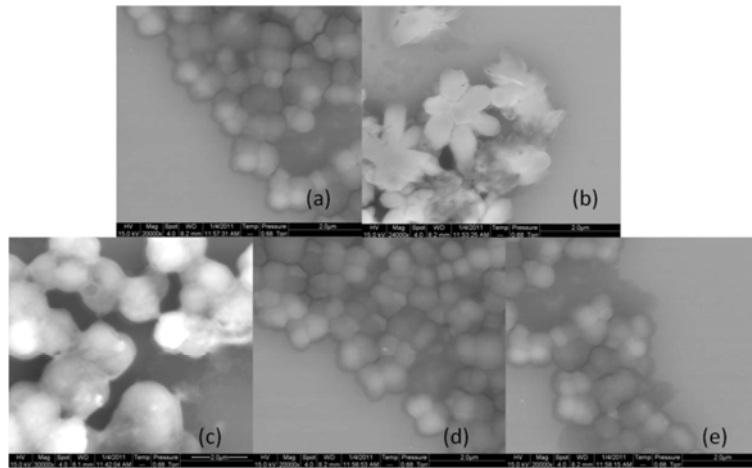


Figure 2. SEM observations of *S. aureus*. (a) Untreated cells; (b) cells after electrolysis; (c) cells after ozonation; (d) cells after chlorination; (e) cells after monochloramination [3].

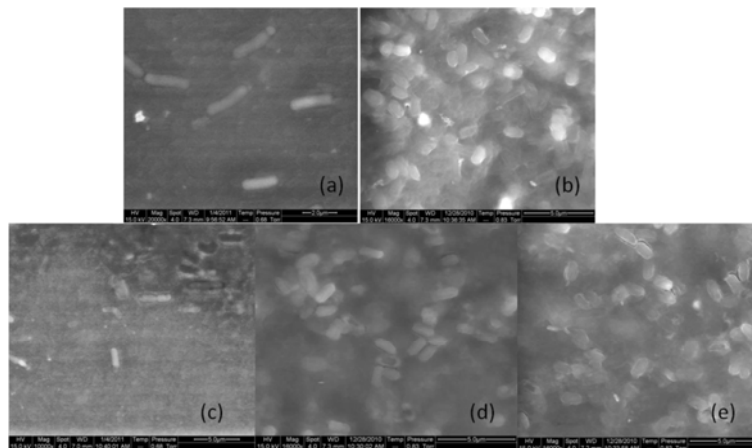


Figure 3. SEM observations of *BST*. (a) Untreated cells; (b) cells after electrolysis; (c) cells after ozonation; (d) cells after chlorination; (e) cells after monochloramination [3].

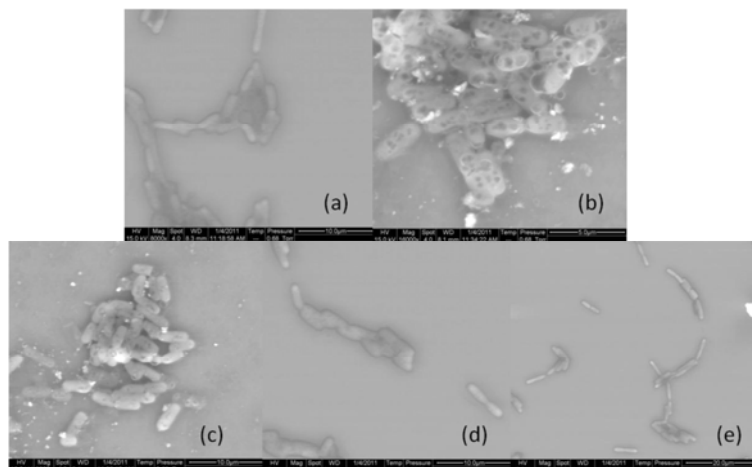


Figure 4. SEM observations of *Bacillus*. (a) Untreated cells; (b) cells after electrolysis; (c) cells after ozonation; (d) cells after chlorination; (e) cells after monochloramination [3].

(1) Killing microorganisms' capability was classified as follow: electrochemical process > ozonation > chlorination > monochloramination when tests were realized at their respective optimal conditions, following the oxidation-reduction potentials of the four

types of disinfectants: hydroxyl radical (2.70 V) > ozone (2.07 V) > chlorine (1.36 V) > monochloramine (1.13 V). Thus, the particular contribution of the hydroxyl radical employing BDD anode in ED had been affirmed.

- (2) Killing microbes' performance was identical for the chosen four bacteria in the electrochemistry-based method employing BDD anode, thanks to the nonselective and powerful-oxidative hydroxyl radical in a free state in the role of disinfectant. At the same time, deactivating BST and *Bacillus* was much slower than *E. coli* and *S. aureus* in the remaining three disinfection techniques. This is because the harsh characters of BST and *Bacillus*, and the weak oxidation capacity of the three remaining disinfectants. The free hydroxyl radical imposed the non-selectivity of ED and its crucial impact in the pathway of BDD anode disinfection system was more explicitly concluded.
- (3) Concerning the hydroxyl radical with non-selectivity and powerful oxidizing capacity implied in the route of ED with BDD anode, cell surface deterioration was more apparent there into than that after ozone application; however, the safety of cells was not influenced in weak oxidizing chlorine and monochloramine for *E. coli*, *S. aureus*, BST and *Bacillus*.

This review focuses on the new trends of ED towards the green chemistry.

2. The Bridge Link Between ED and the Green Chemistry

Green chemistry (GC) [1, 13, 29, 32] is the key to sustainable development since it will conduct to viable

solutions to present difficulties. In addition, it will provide opportunities for fresh methods and products and at its heart is scientific and technological innovation. Following the establishment of the 12 Principles of GC, there has been a steady growth in the vision on what GC signifies. Moreover, there are large perspectives about the greening of chemical water treatment, especially in terms of ferrate(VI) [32, 35] adding, as oxidant/disinfectant/coagulant in the same time, and microchannel reactors which would be considered as promising devices for water treatment due to their proved advantages.

On the other hand, as discussed above, the high efficiency of ED may be associated to the short-lived and energy rich intermediate products with a more performant destruction ability [43]. These agents evidently include free radicals, like OH^\bullet and $\text{O}_2^{\bullet-}$ [12, 55]. By their SEM examination (Figure 5), Diao. *et al.* [43] given more evidence of the hypothesis about the crucial role of OH^\bullet radicals in ED. Cell samples treated by OH^\bullet radicals of the Fenton reaction had a rather comparable look as those after ED remediation. There was important degeneration and decomposition of the cells following from both the Fenton reaction and ED. Liberated cellular materials were collected on the filters, which was remarkable to a lesser amplitude for the samples of ozonation and narrowly remarkable for the samples of chlorination. Consequently, in addition to electro-chlorination, *E. coli* cells during ED remediation were probably deactivated by the intermediate products with an oxidizing strength comparable to that of free radicals and much powerful than that of chlorine [43].

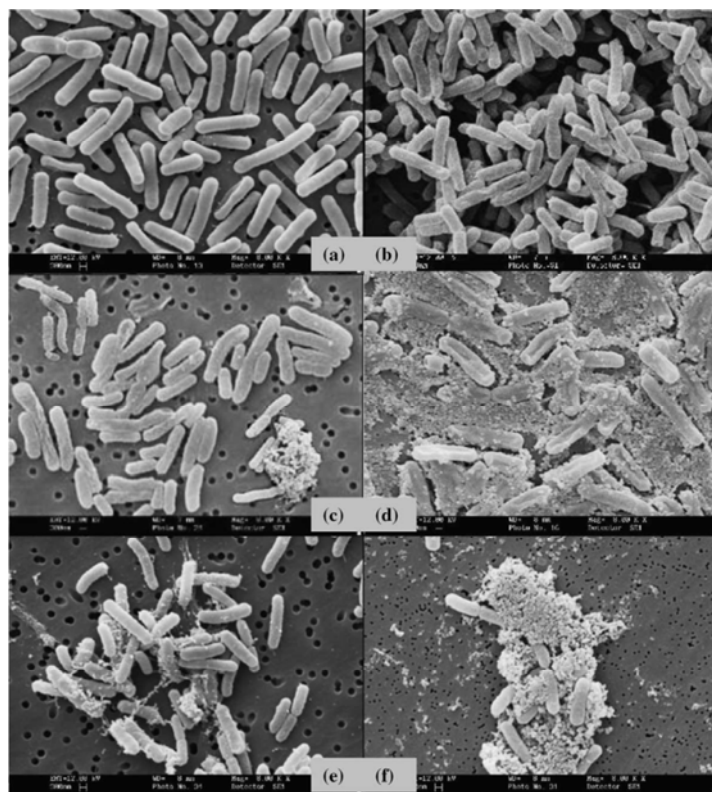


Figure 5. SEM photographs of *E. coli* cells in (a) fresh culture and after (b) chlorination at 5 mg/l for 30 min, (c) ozonation at 10 mg/l for 5 min, (d) the Fenton reaction with 8.5 mg/l H_2O_2 and 0.85 mg/l Fe^{2+} at pH 4 for 10 min, (e) ED at 16 mA/cm² for 2 min and (f) ED at 25 mA/cm² for 2 min [43].

Consequently, the large use of ED remains blocked by many technical issues such as chlorine by-products (CBPs) produced species [7]. Indeed, during ED these carcinogenic products may be generated following the electrode material and applied voltage.

Previously, authors [12, 33] have examined the relationships of CBPs formed species with the electrode material and applied charge in the course of ED treatment. Authors [12, 33] concluded that the usage of electrodes forming highly reactive species has to be more cautiously controlled in hygienically and environmentally oriented using. Following this direction, Pt and BDD anodes have been shown more convenient than other electrodes. The valuable ability of a BDD anode to form ROSs and other oxidizing species during the electrolysis allows establishing a chlorine-free disinfection process [12, 33].

3. Conclusions

ED has known astonishing focus as an option for classical potable water treatment because of its elevated performance and environmental harmony. The most frequent technique of ED is the usage of electro-generated oxidants, such as active chlorine and ROSs, as disinfectants.

Different pathways have been proposed to explain the deadliness of ED method, including (1) oxidative stress and cell loss of life because of electrochemically produced oxidants, (2) irreversible permeabilization of cell membranes by the placed electric field, (3) electrochemical oxidation of vital cellular constituents during exposure to the electric current, and (4) electrosorption of negatively charged *E. coli* cells to the anode surface followed by direct electron transfer reaction.

Convenient devices must be designed and monitored sophistically. The actual state of non-monitored use of disinfection devices is not favorable in terms of hygienic and health risks considerations. Great works remain to be performed.

At least, incorporating the ED methods in existing water treatment factories has to be realized with a view to prove the techno-economic likelihood.

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