

Disinfecting Water with the Carbon Fiber-Based Flow-Through Electrode System (FES): Towards Axial Dispersion and Velocity Profile

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Abstract

Electrochemical technology for the killing of pathogens has been largely investigated. Lately, Ni et al. [1] published excellent research on the disinfection efficiency of a carbon fiber-based flow-through electrode system (FES) versus Gram-negative bacteria (Escherichia coli and fecal coliform) and Gram-positive bacteria (Enterococcus faecalis and Bacillus subtilis) in normal saline over a large span of applied voltages (1 - 5 V) and hydraulic retention times (HRTs) (1 - 10 s). They established that the Gram-negative microbes were more susceptible to FES for their thinner cell walls and over 6.5 log reduction (no live bacteria found) was obtained at the applied voltage of 2 V and HRT of 2 s; however, Gram-positive microbes were demobilized at slightly bigger voltages (3 V, 2 s) or longer HRTs (2 V, 5 s). Demobilizing microorganisms was related to the alteration and laceration of cell membranes mostly via anode direct oxidation in the absence of bacterial regrowth. Further, the disregarding formation of the free chlorine at low voltages (≤2 V) could avert the production of possible chlorinated disinfection by-products. Therefore, FES could furnish an undeveloped substitute to traditional disinfection processes for eliminating pathogens in water. This work concludes that focusing on axial dispersion and velocity profile inside anode will be very useful in comprehending the transport phenomena and proposing a fresh model that merges the axial dispersion and velocity profile for the FES. Such a research trend will more encourage the FES implementation at the large industrial level for disinfecting water.

Subject Areas

Chemical Engineering & Technology

Keywords

Carbon Fiber Felt (CFF), Electrodisinfection, Gram-Negative Bacteria, Gram-Positive Bacteria, Axial Dispersion, Velocity Profile

1. Introduction

Disinfecting water efficaciously remains vital for treating water and wastewater. Traditional disinfection techniques comprise a separate or joint application of chlorination [1] [2], ultraviolet (UV) radiation [3] [4], and ozonation [5] [6]. Nevertheless, the preceding technologies stay alas followed by the generation of unwanted chlorinated disinfection by-products (DBPs) for chlorination [7], the formation of bromate and high-energy consumption for ozonation method [8], and the photoreactivation or dark repair for UV radiation [9]. Therefore, a low-energy consumption, high-efficiency, and chlorine-free demobilization technology has to be suggested as mohair to classical demobilization techniques.

Eliminating microorganisms via electrochemical technology has been largely investigated [10]. Such researches mostly focused on the level of disinfection efficiency [11], electrolyte composition [12], and the demobilization pathway of indirect oxidation with electro-generated oxidants (chlorine and reactive oxygen species) [13], or the direct oxidation of microbes [14]. Nevertheless, the indirect oxidation phenomenon via free chlorine unavoidably elevates the hazards of the production of unwanted DBPs [15] [16]. Consequently, killing microorganisms electrochemically via a direct oxidation technique at low voltage has attracted augmenting interest not long ago [1].

In terms of electrochemical engineering, the device arrangement remains substantial and mostly categorized as flow-by and flow-through setup [17] [18] [19]. Employing flow-by devices with plate electrodes stays restricted because of its small exposure area, which requests more plate electrodes number or longer residence period to attain efficacious killing [20]. Alternatively, the flow-through setup constitutes a typical reaction area, in which the polluted water goes along the internal zone of electrodes to ameliorate efficaciously the mass transfer phase [21]. In such a device, three-dimensional (3D) materials, like metal or carbonaceous porous electrodes, remain largely tested. As an illustration, a CuO-nanowire-modified copper foam with an elevated conductivity was tried as the electrode to remedy an influent (\sim 10⁷ colony-forming units (CFU)/mL bacteria in normal saline) at the operating voltage of 1 V applied by direct current (DC) power supply. It attained nearly 7 log elimination of bacteria (*i.e.*, no live microbes observed) at the hydraulic retention time (HRT) of 7 s [22], which was low juxtaposed with chlorination [23], and flow-by electrodisinfection [24]. Nevertheless, the chemical in stability of metal material would limit the service life of electrodes, which restricted the large usage of metal materials [1] [25].

The 3D carbonaceous materials, like carbon nanotube (CNT) sponge [26] and carbon fiber felt (CFF) [14], remain broadly employed as anode and/or cathode thanks to their elevated specific surface area, excellent electric conductivity, and chemical stability. Indeed, the CNT sponge attained outstanding killing pathogens' activity at the HRT of 5 s and voltage of 2 V [26]. Nevertheless, the intricate manufacture and elevated material price of CNT sponges limit the large-scale utilization. For the CFF, it can be maturely manufactured and commercially purchased with wanted thickness and elevated porosity, which is very advised to work as electrodes [1].

Liu *et al.* [14] established that the flow-through electrochemical technique with the CFF electrode may ensure the considerable reduction of *Escherichia coli* and low energy-consumption at the applied voltage under 3 V [14]. Nevertheless, there are numerous microbes in the water medium, comprising Gram-negative and Gram-positive bacteria, which differ mostly in the structure and composition of the cell membrane [27] [28], and possess various resistance to the disinfection technique [29]. Therefore, the dissimilarities of disinfection efficiency among Gram-negative and Gram-positive bacteria require to be more studied consistently. Moreover, it also requests to prove the regrowth/reactivation event of bacteria following electrodisinfection.

2. Carbon Fiber-Based Flow-Through Electrode System (FES) for Killing Gram-Negative and Gram-Positive Bacteria

Ni *et al.* [1] constructed a carbon fiber-based flow-through electrode system (FES) (**Figure 1**). They tested consistently the physicochemical properties of CFFs), like the morphology, electrical resistivity, void space portion, elemental composition, and specific surface area. They realized disinfection trials of FES against Gram-negative bacteria (*E. coli* and fecal coliform) or Gram-positive bacteria (*Enterococcus faecalis* and *Bacillus subtilis*), and free chlorine analysis in effluent at low applied voltages (1 - 5 V) and short HRTs (1 - 10 s). They determined the modification of cell membranes before and after disinfection using scanning electron microscopy (SEM) analysis. They assessed the route of FES and the action of CFF cathode and anode via an *in-situ* sampling test. Further, the storage trial was performed to more clarify if the microorganisms would regrow after FES application.

The carbon fiber-based FES furnished efficient demobilization of bacteria (>6.5 log reduction, no live bacteria found) at low applied voltages and short HRTs (**Figure 2**). Most importantly, Gram-negative bacteria (*E. coli* and fecal coliform) were totally demobilized at the applied voltage of 2 V and HRT of 2 s; while bigger voltages (3 V, 2 s) or longer HRTs (2 V, 5 s) were required for the demobilization of Gram-positive bacteria (*E. faecalis* or *B. subtilis*) with comparatively thicker cell walls. The route of FES disinfection was anode direct oxidation,



Figure 1. Construction and characterization of the flow-through electrode system (FES). (a) The schematic diagram of the FES including Plexiglas coaxial electrode holders, a filter paper as the insulating layer, and two CFF electrodes connected with the DC power supply. (b) Scanning electron microscopy (SEM) image of the micro-structure of CFF electrodes. (c) SEM image of the carbon fibers under high magnification [1].

conducting to the modifications and rupture of cell membranes. There was no important influence on bacterial demobilization via anode indirect oxidation of generating free chlorine at low voltage, which can avert the production of potential chlorinated DBPs. Further, it was as well illustrated that the bacteria were deteriorated irreversibly, and were unable to regrow throughout the storage procedure [1]. The global outstanding effectiveness of the carbon fiber-based FES presented a potential solution for emergency and rapid water disinfection.

From the disinfection findings aforesaid, it was mentioned that the Gram-positive bacteria were less defenseless to FES disinfection than Gram-negative bacteria. The distinction in elimination performance among two sorts of microbes may be related to the constitution of cell walls (peptidoglycan and teichoic acids) [1] [30] [31] [32] [33]. The peptidoglycan film of Gram-positive bacteria was much thicker, which was linked to their resistance [34]. However, the cells of Gram-negative microorganisms were not as robust as Gram-positive microorganisms and effortless to be demobilized for the thin peptidoglycan membrane with an outer membrane consisting of lipopolysaccharides and lipoproteins [27]. Moreover, FES depicted better efficiency towards B. subtilis rather than *E. faecalis* probably due to the cell size of *B. subtilis* (about $2.7 \pm 0.2 \mu m$ in length) was much bigger than that of *E. faecalis* (about $0.9 \pm 0.3 \mu m$ in length) determined by the SEM image (Figure 3(c), Figure 3(d)), which furnished much more contact area with the electrode surface for electrodisinfection. Further, the high-efficiency disinfection performance can remain for at least seven days [35].



Figure 2. The disinfection performance of the carbon fiber-based FES towards four bacterial strains without and with the applied voltages of 1 - 5 V at the HRTs of 1 s (a), 2 s (b), 5 s (c), and 10 s (d) [1].

3. Carbon Fiber-Based Flow-Through Electrode System (FES) Disinfection Pathway

Figure 3 illustrates representative SEM images of *E. coli*, fecal coliform, *E. faecalis*, and *B. subtilis* before and following FES treatment. Most importantly, cell membranes of Gram-negative bacteria after a 2 V, 2 s treatment were dramatically dehydrated and broken juxtaposed to the untreated cells; however, the dead Gram-positive bacteria following a 3 V, 2 s treatment maintained relatively intact cell membrane structures, particularly for *E. faecalis*. The distinction between Gram-positive and Gram-negative microbes proposed that Gram-negative bacteria were relatively liable to electrodisinfection for the thinner cell walls. As a consequence, it was established that the route of bacterial death below FES treatment was the demolition of cell membranes [1].



Figure 3. Morphology of live and dead bacteria. (a) The live and dead *E. coli* cells after a 2 V, 2 s treatment. (b) The live and dead fecal coliform cells after a 2 V, 2 s treatment. (c) The live and dead *E. faecalis* cells after a 3 V, 2 s treatment. (d) The live and dead *B. subtilis* cells after a 3 V, 2 s treatment [1].

Further, Ni *et al.* [1] focused more on the influent with $\sim 10^6$ CFU/mL *E. coli* to ascertain the pathway of FES disinfection technique. In an effort to recognize the role of anode and cathode, they performed the in-situ tests to reach the cells elimination following cathode treatment and successive cathode-anode treatment below 2 V and 3 V, respectively.

As illustrated in Figure 4(a), the *E. coli* in the effluent samples was demobilized efficiently following consecutive cathode-anode treatment, and the disinfection effectiveness was enhanced with the elevation of the applied voltages and/or HRTs. Nevertheless, the reduction proportion (\sim 31%) following only cathode treatment below 2 V was little and there was no evident role for cathode demobilization (\sim 33% reduction) with the augmentation of voltage (3 V) or HRT (10 s). Consequently, it is shown that the reaction at the anode controlled the disinfection performance of FES treatment, which was in conformity with former investigations [14] [36].

The likely route for anode electrochemical operation comprised direct oxidation and indirect oxidation via forming reactive species like hydroxyl radical ('OH) and free chlorine [10] [37] [38] [39] [40] [41]. Nevertheless, the small applied voltage of 2 V for *E. coli* demobilization was incapable to produce 'OH which requested much bigger anode potential [42] [43], while the free chlorine with standard evolution potential of 1.15 V vs Ag/AgCl was fit to take place in the FES treatment [1].

On the other hand, Ni *et al.* [1] followed the level of free chlorine produced throughout demobilization at changed applied voltages and HRTs (Figure 4(b)).



Figure 4. (a) The log reduction of *E. coli* in cathode-treated influent and effluent at diverse HRTs below applied voltages of 2 V and 3 V. (b) The concentration of free chlorine in effluent at different applied voltages and HRTs [1].

There was no considerable formation of free chlorine (lower than the detection limit, 10 μ g/L) at the applied voltage under 2 V no matter how long the residence period was. Further, the level augmented with a bigger residence period at the higher applied voltages (over 3 V).

Considering the findings aforesaid, Ni *et al.* [1] concluded that demobilizing *E. coli* was mostly related to the anode direct impact at the applied voltage of 2 V, instead of indirect oxidation via 'OH and free chlorine generation. It also averted the production of potential chlorinated DBPs, which was apt to begin to be an encouraging substitutional for chlorination.

4. Axial Dispersion and Velocity Profile for the FES

As shown in **Figure 1**, influent enters the FES. It passes across the CFF cathode, an insulator layer, and CFF anode. In terms of electrochemical engineering, the main reactions occur in the CFF cathode and CFF anode. As mentioned above, bacterial demobilization was related to the alteration and rupture of the cell membrane mostly through anode direct oxidation without bacterial remobilization [1]. Therefore, what happens inside the anode via direct oxidation constitutes the main stage of electrodisinfection process. However, before the electrochemical reaction taking place, wastewater has to be put in contact with the surface anode. In other words, there is a mass transfer phase.

Indeed, the residence time distribution (RTD) remains a crucial feature in the exploration of non-ideal reactors [44]. In micro-reactors that work mostly below small Reynolds numbers, the axial dispersion is fundamental [44]. Because the channel length or residence time of fluid is frequently not long enough (from 2 to 5 s [1]), the axial dispersion model cannot anticipate an exact RTD of the fluid in micro-reactors [44]. With a view to controlling such a problem, Fazli-Abukheyli and Darvishi [44] suggested a fresh model that integrates the axial dispersion and velocity profile employing the parallel tanks-in-series compartment model. The model includes two parameters, involving the velocity profile exponent and Peclet number. It may be employed for Newtonian and non-Newtonian fluid flow in the devices with a large span of Reynolds number and in all domains of flow regime, comprising plug, laminar, perfectly mixed, and different profiles among such regimes. Following validating the model, Fazli-Abukheyli and Darvishi [44] assessed the impacts of velocity profile and Peclet number on the RTD of fluid in the tube for m-laminar and y-laminar velocity profiles.

Rastegar and Gu [45] proposed a novel correlation for the axial dispersion coefficient employing experimental information in the literature for axial dispersion in fixed-bed columns packed with particles. The Chung and Wen correlation, the De Ligny correlation are two famous empirical correlations. Nevertheless, the former lacks the molecular diffusion term and the latter does not take into account bed voidage. Their novel axial dispersion coefficient correlation is founded on additional experimental data in the literature by taking into account both molecular diffusion and bed voidage. It is more comprehensive and accurate. The Peclet number correlation from the new axial dispersion coefficient correlation on the average leads to 12% lower Peclet number values compared to the values from the Chung and Wen correlation, and in many cases much smaller than those from the De Ligny correlation. Investigations, as performed by Fazli-Abukheyli and Darvishi [44] and Rastegar and Gu [45], will be consequently helpful in understanding the transport phenomena and suggesting a novel model that integrates the axial dispersion and velocity profile for the FES. For instance, Malayeria *et al.* [46] focused on modeling of gas-phase heterogeneous photocatalytic oxidation reactor in the occurrence of mass transfer limitation and axial dispersion. Such research would initiate the investigation field suggested here for coming works.

5. Conclusions

From this work, the following conclusions can be drawn:

1) Pure water supply remains one of the most vital and precious requests for human beings to continue, and the shortage of supply of potable water has been a grave problem during the last decades. The existence of pollutants like pathogens has the tendency to be the main worry for keeping nature and human life. There are several techniques being done on dealing with the polluted water; however, an optimum technology stays to be discovered. Nanotechnology has been earning currency in numerous fields especially in the implementation of water and wastewater remediation. The exceptional features of the nanomaterials composite formed are apt to give leap forward chances to metamorphose water and wastewater handling. Many of the nanotechnologies have shown their stunning characteristics in the water treatment industry. Additional amendment on such nanomaterials to enhance the features and decrease the effective price depicts an encouraging outlook of such carbon-based nanomaterials in the water and wastewater treatment field [47].

2) An investigation like performed by researchers [44] [45] will be very useful in comprehending the transport phenomena and proposing a fresh model that merges the axial dispersion and velocity profile for the FES. Such a research trend will more encourage the FES implementation at the industrial level for disinfecting water.

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Conflicts of Interest

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

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