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Article

Nanostructured Conductive Composite Filter Electrodes for Water Sterilization by Application of Low Electrical Current

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Abstract:

There is a crucial need for the development of inexpensive technologies for water sterilization enabling access to safe drinking water for more than one billion people in the developing countries. Water sterilization can be attained by chemical, electrochemical or electrical means. Electrical methods for water sterilization are considered environmental friendly because they use ‘‘electrons’’ as the nontoxic reaction mediator. This paper reports the preparation of electrically conductive composite membranes (ECCMs) for water sterilization. The composite membranes were prepared by two stages dip coating of the porous cotton fibers into conductive graphite and various conductive silver nanostructures. The prepared ECCMs were utilized as filter electrodes for fabrication of pathogen inactivation device, with an applied voltage of -20 and +20 V, respectively. At flow rates of 10 mL/ min, the fabricated device inactivated > 99.9999% E. coli bacteria in the infected water samples having nominal bacterial density in the range of 10^7-10^8 CFU/mL

Keywords: water sterilization; graphite, silver nanostructures; electrical inactivation of bacteria; Electroporation

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1. Introduction

Waterborne disease-causing bacteria continue to be a major problem in the developing world where more than one billion individuals lack access to safe drinking water [1]. Water sterilization can be achieved by chemical, electrochemical or electrical methods. Chemical methods inactivate pathogen by using proper doses of chemicals such as chlorine. The disadvantages of the chemical treatment are three folds, (a) unfavorable taste and odor, (b) generation of potentially toxic or mutagenic products such as trihalomethanes [2], and (c) transportation and storage of dangerous chemicals such as chlorine gas. In case of electrochemical water sterilization methods, disinfection agents are generated electrochemically through redox reactions such as electrolytic generation of free chlorine from brine solution. Electrochemical disinfection can result in formation of toxic byproducts such as ClO_2, ClO_3, and ClO_4 [3].

Electrical methods for water sterilization such as pulsed electric fields (PEFs) [4] and direct electric current [5], are regarded environmental friendly because they use ‘‘electrons’’ as the nontoxic reaction mediator [6]. The bactericidal action of electrical field can be attained by applying potential or electrical current through electrically conductive filter electrodes. The resultant electrical potential formed across the bacterial cell membrane leads to bacteria inactivation, presumably due to cell lysis and leakage of intracellular contents by irreversible electroporation mechanism [7].

Regardless the fact that pathogen inactivation by PEFs requires the application of high voltages (1-100 kV/cm), the entire cellular membrane breaks when the transmembrane potential reaches a critical voltage of $\approx$ 1 V [8]. High voltage PEF has been utilized as non-thermal process for bacteria inactivation in liquid foods [9]. Application of such high voltages requires a high capital investment cost of power supply systems and holds safety concerns during operation. However, weak electrical currents have been utilized effectively for inactivation and detachment of bacteria adhered on conductive surfaces as a strategy for biofouling control [6].

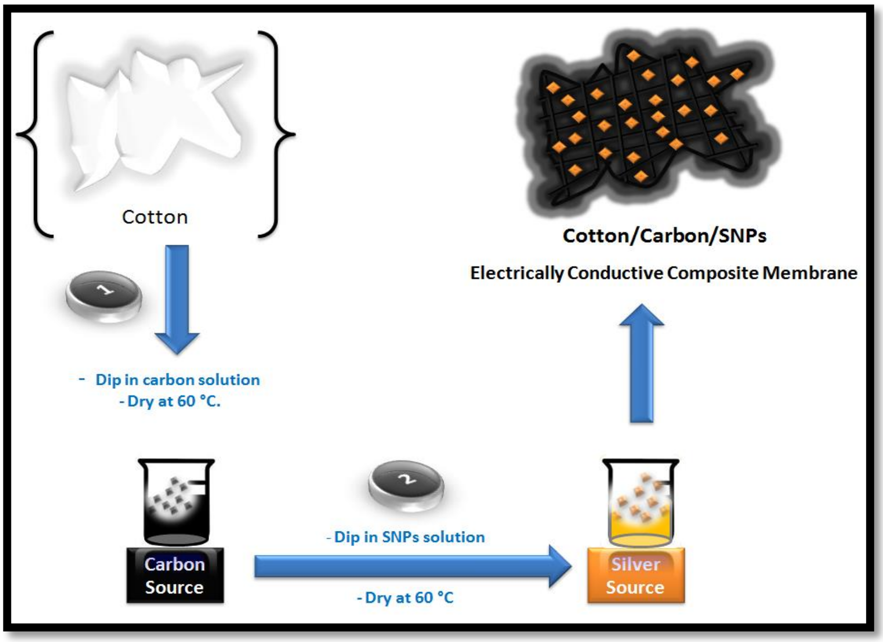
Recently, a new alternative approach was invented for high speed water sterilization utilizing the application of weak currents through one-dimensional nanostructured electrically conductive composite membrane (ECCM) [10]. The reported ECCM is a composite membrane made from cotton, multiwall carbon nanotubes (MWCNTs) and silver nanowires (SNWs). One of the important shortcomings of such ECCM is the use of very expensive and toxic MWCNTs. In this paper, we successfully replaced the conductive MWCNTs component with various inexpensive carbon-derived materials such as conductive carbon black, synthetic graphite and natural graphite. The carbon materials are also used in conjunction with various silver nanostructures including silver nanowires SNWs, silver nanoparticles SNPs or mixture thereof.

2. Results and Discussion

2.1. Fabrication of ECCMs:

In the current study, ECCMs were prepared from cotton, graphite and various forms of silver nanostructures such as SNWs, SNPs or mixture thereof. Cotton fibers possess features such as high porosity, inertness and biodegradability. The high porosity of cotton fibers enables high speed filtration and avoids issues such as bioaccumulation and filter clogging. Graphite represents the second component of the prepared ECCMs. Due to its high electrical conductivity [11], potential antibacterial activity [12], inertness, non-toxicity, chemical stability at anodic potentials and affordability at low cost from natural and synthetic resources, graphite is one of the most suitable carbons for preparation of highly conductive filter electrodes for water sterilization applications. The high electrical conductivity of graphite is an essential component for fabrication of highly conductive ECCMs to enable good electrical contact between active bacteria and the electrode surface during the high speed filtration process. The low cost of graphite and its non-toxicity are key advantages over the expensive and potentially toxic carbon nanotubes (CNTs) [13]. Moreover, graphite can be easily incorporated into different materials by simple dip coating or blending procedures. Addition of silver in the form of SNWs, SNPs or mixture thereof, provides *highly conductive connection points* inside the ECCM and improves electrical charge transport through the conductive filter electrodes. Silver nanostructures are expected to provide higher contact surface area with pathogen being passed through the filter electrode and, hence, higher inactivation efficiencies can be achieved during such high speed filtration process.

Figure 1 Schematic representation for the preparation steps of the ECCM.



As shown in figure 1, the non-conductive cotton fibers were converted into conductive composite filter electrodes by simple two steps procedure. The first step includes dip coating in selected carbon source until constant electrical conductivity is reached. The second step includes dip coating of the dried cotton/carbon composite in a colloidal silver dispersion.

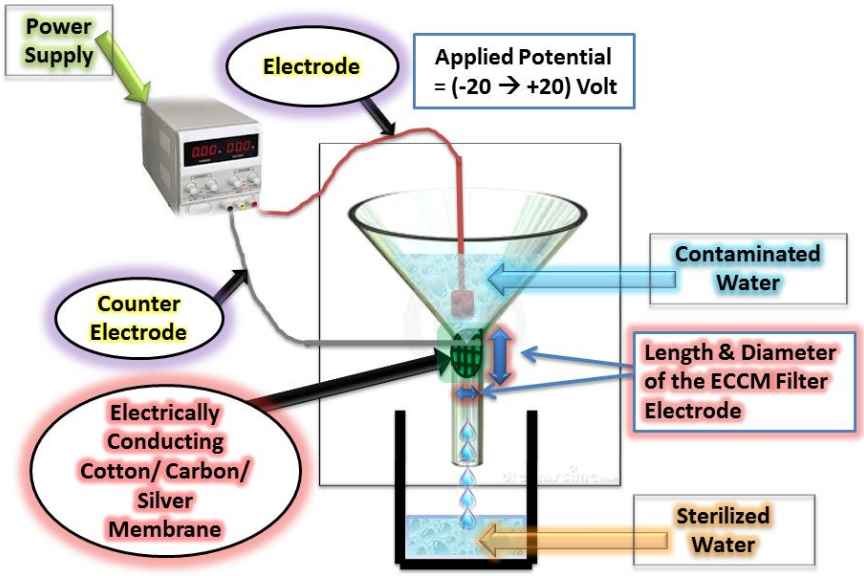
2.2. Electrical Resistance of Fabricated ECCMs

Carbon, synthetic graphite and natural graphite have been investigated for preparation of electrically conductive membranes (ECCMs). The final electrical resistance of the prepared ECCMs varied according to the type of the employed carbon source and silver nanostructures. Generally, in order to obtain ECCMs with enough high conductivity to be used as an electrode material, the specific resistivity of the prepared electrodes shall be ≤ 10 Ohm.cm [14]. In the current study, the electrical resistance of all prepared ECCM sheets was ≤ 1 Ohm/Sq.

2.3. Fabrication of inactivation device

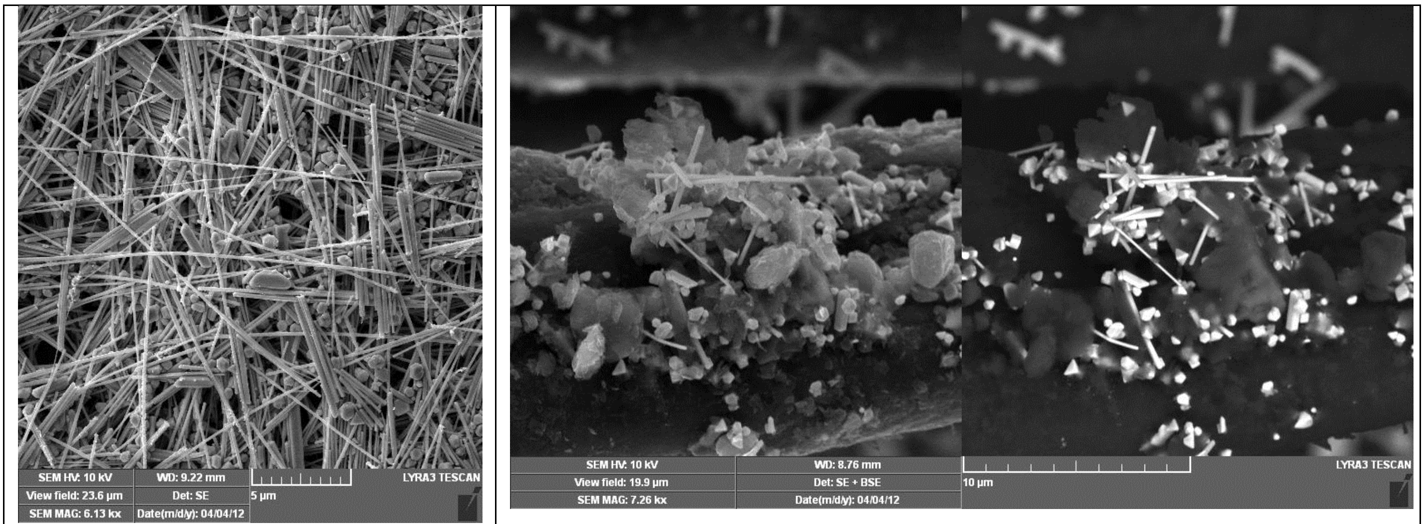
Figure 2 represents a sketch of the fabricated membrane test assembly. ECCMs were prepared from graphite and a mixture of SNPs and SNWs (figure 3). The prepared ECCM was placed in plastic funnel with a stem of an internal diameter of 5 mm and a length of 3 cm. The effect of the applied voltage and filtration speed on the obtained inactivation efficiencies are presented in figure 4.

Figure 2 Schematic illustration of the fabricated bacteria inactivation device



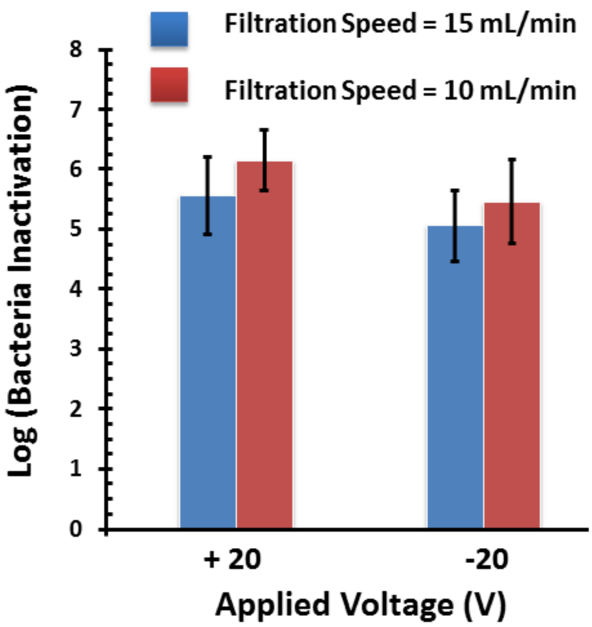
In all cases, E. coli bacteria were inactivated with log reduction values in the effluent of over log 5. The highest inactivation efficiency of > 99.9999% (log 6 reductions) was obtained at flow rate of 10 mL/min and an applied potential of 20 V. In comparison, the obtained inactivation efficiencies at higher flow rates of 15 mL/min were slightly lower by an order of magnitude, presumably due to the lower bacterial residence time inside the filter electrode. These results open the possibility of using various forms of silver nanostructures, without extra purification steps, as a component of the composite filter electrodes for water sterilization by application of low electrical current.

Figure 3 SEM image of: (a) synthesized silver nanostructures containing a mixture of SNPs and SNWs, and (b) a cross section of the ECCM treated with SNPs and SNWs.



(a) (b)

Figure 4 Log reduction of E. coli bacterial count after gravity filtration through nanostructured ECCMs, at different applied voltages. Error bars represent standard deviation. The ECCM was prepared from cotton, graphite and a mixture of SNPs and SNWs, with sheet resistance of < 1 Ohm/ Sq.



3. Experimental Section

Graphite samples with different grades were obtained as a gift from Asbury Carbons, New Jersey-USA. A mixture of SNWs and SNPs was synthesized by modified polyol process [15]. ECCMs were prepared by two steps procedure. In brief, ≈ 100 mg cotton sample was dipped into water solution containing primary artificial graphite material. This process was repeated until constant resistance was achieved. Then, the prepared sample was dried in oven at 60 °C for one hour. After that, cotton/ graphite composite was dipped into water solution containing the prepared mixture of SNWs and SNPs. The resulted ECCM was dried at 60 °C for one hour.

Bacteria inactivation device was fabricated as illustrated in figure 2. Contaminated water samples containing nominal E. coli bacterial density of 10^7-10^8 CFU/mL, were flowed through the ECCM filter with adjusted rates between 10 and 15 mL/min. In each run, 100 mL water sample was allowed to flow through the device and the treated solution was diluted 1000 times from which 100 µl was plated. The device was operated with an applied voltage of -20 V or +20 V.

4. Conclusions

The current study demonstrates a simple and effective water sterilization methodology by the application of low electrical current through electrically conductive filter electrodes. The filter electrodes were prepared from cotton, graphite and a mixture of SNWs and SNPs. The fabricated bacteria inactivation device operated with flow rate of 600 mL/min and an applied voltage of 20 V provided 6 log reductions of E. coli bacteria from contaminated water samples with nominal bacterial density of 10^7-10^8 CFU/mL

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Conflicts of Interest

The authors declare no conflict of interest.

References and Notes

1. Fenwick, A., Waterborne infectious diseases--could they be consigned to history? *Science* **2006,** 313, 1077-81.
2. Dunnick, J. K.; Melnick, R. L., Assessment of the carcinogenic potential of chlorinated water: experimental studies of chlorine, chloramine, and trihalomethanes. *Journal of the National Cancer Institute* **1993,** 85, (10), 817-822.
3. Martinez-Huitle, C. A.; Brillas, E., Electrochemical alternatives for drinking water disinfection. *Angew. Chem., Int. Ed.* **2008,** 47, 1998-2005.
4. Vernhes, M. C.; Benichou, A.; Pernin, P.; Cabanes, P. A.; Teissié, J., Elimination of free-living amoebae in fresh water with pulsed electric fields. *Water Research* **2002,** 36, (14), 3429-3438.
5. del, P. J. L.; Rouse, M. S.; Mandrekar, J. N.; Steckelberg, J. M.; Patel, R., The electricidal effect: reduction of Staphylococcus and Pseudomonas biofilms by prolonged exposure to low-intensity electrical current. *Antimicrob. Agents Chemother.* **2009,** 53, 41-45.
6. Hong, S. H.; Jeong, J.; Shim, S.; Kang, H.; Kwon, S.; Ahn, K. H.; Yoon, J., Effect of electric currents on bacterial detachment and inactivation. *Biotechnol. Bioeng.* **2008,** 100, 379-386.
7. Weaver, J. C., Electroporation theory: Concepts and mechanisms. *Methods Mol. Biol. (Totowa, N. J.)* **1995,** 47, 1-26.
8. Sale, A. J.; Hamilton, W. A., Effects of high electric fields on micro-organisms. 3. Lysis of erythrocytes and protoplasts. *Biochim Biophys Acta* **1968,** 163, 37-43.
9. Wan, J.; Coventry, J.; Swiergon, P.; Sanguansri, P.; Versteeg, C., Advances in innovative processing technologies for microbial inactivation and enhancement of food safety - pulsed electric field and low-temperature plasma. *Trends Food Sci. Technol.* **2009,** 20, (Copyright (C) 2012 American Chemical Society (ACS). All Rights Reserved.), 414-424.
10. Schoen, D. T.; Schoen, A. P.; Hu, L.; Kim, H. S.; Heilshorn, S. C.; Cui, Y., High Speed Water Sterilization Using One-Dimensional Nanostructures. *Nano Letters* **2010,** 10, (9), 3628-3632.
11. Pandolfo, A.; Hollenkamp, A., Carbon properties and their role in supercapacitors. *Journal of Power Sources* **2006,** 157, (1), 11-27.
12. Liu, S.; Zeng, T. H.; Hofmann, M.; Burcombe, E.; Wei, J.; Jiang, R.; Kong, J.; Chen, Y., Antibacterial activity of graphite, graphite oxide, graphene oxide, and reduced graphene oxide: membrane and oxidative stress. *ACS Nano* **2011,** 5, (9), 6971-6980.
13. Jain, A. K.; Mehra, N. K.; Lodhi, N.; Dubey, V.; Mishra, D. K.; Jain, P. K.; Jain, N. K., Carbon nanotubes and their toxicity. *Nanotoxicology* **2007,** 1, 167-197.
14. Beckley, D. A. Continuous filament graphite composite electrodes. US4369104, 1983.
15. Korte, K. E.; Skrabalak, S. E.; Xia, Y., Rapid synthesis of silver nanowires through a CuCl- or CuCl2-mediated polyol process. *Journal of Materials Chemistry* **2008,** 18, (4), 437-441.