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Disinfection of water using pulsed power technique: Effect of system parameters and kinetic study

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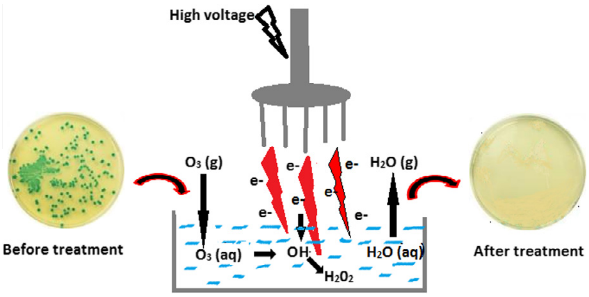
HIGHLIGHTS

- Sequential stress conditions showed better efficiency than continuous stress.
- Effect of voltage, frequency, alkalinity, pH and natural organic matter.
- An empirical model for disinfection was developed using response surface method.
- Kinetic study of bacterial disinfection and kinetics of reactive oxygen species.
- Bacterial inactivation is permanent by adapting pulsed power technique.

GRAPHICAL ABSTRACT



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ABSTRACT

Present study investigated the use of pulsed power technique for disinfection of water under different operating and environmental conditions. Final concentrations of reactive oxygen species (ROS) like hydroxyl radical, hydrogen peroxide, ozone, and superoxide radicals generated in the system were found to be 56, 17, 1 and 18 mg/L, respectively, for an applied voltage of 23 kV, frequency of 25 Hz and a streamer discharge time of 12 min. It was observed that disinfection efficiency was high with sequential stress compared to continuous stress. The disinfection efficiency increased with increasing applied voltage and frequency. Disinfection efficiency was high when pH was less than 7. Presence of alkalinity, natural organic matter and turbidity reduced the disinfection efficiency significantly. For 7 log reduction of *Escherichia coli*, the treatment time was increased from 6 to 10 min, when pH was increased from 4 to 9. Complete disinfection of *E. coli* was achieved in a short treatment time of 4–10 min, with an energy consumption of 0.0056–0.014 kW h for 50 mL of contaminated water. An empirical model for optimum disinfection efficiency was developed using Box–Behnken design (BBD). As per the model, applied voltage, time of treatment and alkalinity were found to be the most significant factors affecting the disinfection efficiency. Model predicted values were in good agreement with the experimental values. Rate constant for disinfection and ROS formation was also evaluated. Rate of disinfection was between 0.59 and 1.68 log(cfu/mL)/min.

1. Introduction

About 80% of the diseases in developing countries are caused due to the consumption of poor quality water [1]. Around 11% of the world population lacks access to safe drinking water and more than 500,000 children die every year due to diarrhoea [2]. Drinking contaminated water leads to several diseases such as cholera, jaundice, typhoid, malaria, dengue, yellow fever and river blindness. Some researchers have projected that almost 135 million people would die by the year 2020 because of pathogen contaminated drinking water [3].

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Chlorination is the most commonly used technique for disinfection. However, application of this technique is reducing because of the formation of carcinogenic disinfection by-products like trihalomethanes, haloacetic acids and organochlorine compounds in water [4–6]. Other disinfection processes like UV irradiation, ozonation and gamma irradiation are expensive. Also, it is reported that these processes are inefficient in inactivating pathogens like *Cryptosporidium* [7]. Among several other methods, solar water disinfection (SODIS) is a simple and low cost disinfection technique, mainly used for the removal of bacteria and viruses [8]. However, this technique is not effective for highly turbid water with high inorganic and organic matter [9]. In addition, it cannot inactivate protozoan cysts and spores of some bacteria [8]. Advanced oxidation processes (AOPs) are emerging as alternative technologies for the treatment of water contaminated with various pollutants. Several AOPs like Fenton, photo-Fenton [10], H_2O_2/UV [11], O_3/UV [12], O_3/UV/Ag-TiO_2 [13], electrochemical process [14,15] and photo-catalysis [16,17] have been applied for disinfection. Most of the AOPs have certain advantages and disadvantages in terms of efficiency and energy consumption [18]. Also, presence of natural organic matter (NOM) like humic acid, bi-carbonates, carbonates and other ions in water affects the performance of AOPs [19]. Therefore, assessing the effects of these water constituents on the performance of AOPs is indeed important.

In recent times, plasma technology has been gaining importance because it exhibits a rapid disinfection potential [20,21]. Pulsed power technique (PPT) is a process by which energy is accumulated over a relatively long time and then it is released rapidly, thereby increasing the instantaneous power. Pulsed high voltage streamer discharge in air and water generates plasma that is a mixture of highly reactive atoms, free radicals, electrons and certain photons [22]. Plasma generated by streamer discharge diffuses into the water and forms reactive oxygen species (ROS), which efficiently degrades the organic pollutants and inactivates the pathogens. Shock waves and UV light generated along with the ROS, in turn, increase the disinfection efficiency [22]. Study of bactericidal effects of the corona discharge was initiated by Kuzmichev et al. [23]. Several studies on the application of plasma for bacterial inactivation have been recently reported [24–26]. Inactivation of vegetative forms and spores of *Bacillus subtilis* using corona discharge was reported by Joubert et al. [27]. Inactivation of viruses such as bacteriophage MS2 by streamer corona [28] and inactivation of *Cryptosporidium parvam* by pulsed UV with pulsed plasma gas discharge have also been reported [29]. The information of the effect of applied pulse, wave characteristics and energy estimates of these plasma processes for water disinfection is limited.

Information on disinfection of water in the presence of high turbidity, alkalinity, NOM and different pH conditions by plasma treatment processes is scanty. It is essential to understand the effect of these parameters on the formation of ROS during the treatment. Optimisation of operating conditions of PPT such as voltage, frequency, and time, along with environmental parameters such as pH, organic content, and alkalinity, for effective disinfection is seldom carried out. The primary objective of the present work is to study the disinfection efficiency of PPT in various environmental conditions and quantification of ROS during the disinfection process. In order to determine optimal conditions for disinfection, an attempt was made to develop an empirical model for disinfection using Box–Behnken design (BBD) by incorporating various operating parameters (voltage, frequency, time) and environmental parameters (pH, alkalinity and organic matters) simultaneously. The effect of intermittent and continuous discharge on disinfection efficiency was also investigated.

2. Materials and methods

2.1. Growth media and bacterial cultures

Escherichia coli was used as the model organism in all bacterial disinfection studies. A single colony of *E. coli* was isolated from tryptone bile glucuronic agar (Himedia, India) plate and inoculated in 100 mL of Luria Bertani (Himedia, India) liquid media in a 250 mL conical flask. The culture was incubated overnight at 35 °C in an orbital shaker at 150 rpm. 10 mL of culture was then taken and centrifuged at 5000×*g* for 10 min at 4 °C. The pellet was washed three times using physiological saline and re-suspended in physiological saline (0.85% NaCl) and diluted to 50 times for performing the experiment. Initial cell concentration of 10^7 cfu/mL was used for all experiments.

2.2. Chemicals

All the chemicals used in the study unless specifically mentioned, were procured from Rankem, India. Bentonite clay was used for the preparation of turbid water. DPD kit (Prerana Laboratories, Pune, India) was used for ozone quantification. Titanium oxy-sulphate and potassium superoxide were procured from Sigma–Aldrich, India. Nitro blue tetrazolium salt (NBT) (Loba, India) and ethanol (Jiangsu Huaxi International, India) were used for quantification of superoxide. Uridine (Loba, India) was used for UV detection.

2.3. Experimental set up and reactor configuration

The experimental set up was divided into two sections: (a) high voltage source (Fig. 1a) and (b) reactor cell (Fig 1b). The discharge free test transformer (100 kV, 5 kVA) was used for the generation of high AC voltage. The generated AC voltage was converted to DC voltage by the use of high voltage diode (140 kV, 20 mA, 100 kΩ). DC voltage was subsequently measured using resistor divider. The constant DC voltage was converted to square pulse using rotating spark gap (RSG). Also, the duration of pulse was controlled by varying the speed of the RSG. Shape and magnitude of generated square wave pulse was recorded using high voltage probe connected to the oscilloscope (HP 54645A, 100 MHz). The measured rise time of the pulse was 0.4 μs and duration of the pulse was 14 ms, 17 ms and 20 ms for frequency of 30 Hz, 25 Hz and 20 Hz, respectively. A typical shape of generated voltage wave is shown in Fig. 1c.

In the present study, streamer discharge was achieved by using needle plate configuration. Multiple needles were attached to the circular plate, which is connected to the high voltage terminal. The circular plane bottom electrode was connected to the ground terminal. The high voltage and ground electrode were fixed in a cylindrical container filled with water. 50 mL of water was used in all studies. The needle was made up of tungsten electrode. The radius of curvature of the tip was about 50 μm. This ensured continuous streamer propagation in the liquid. The gap between needle tip and water surface in the container was adjustable between 3 and 8 mm. The applied voltage was fixed in such a way that the visual streamer propagation was observed during the test. A water jacket was provided to avoid temperature rise and it was maintained between 30 °C and 40 °C for all the experiments. A thermal vision camera was fixed to observe any temperature increase during the test period. Two ports were provided in the top electrode; one for injecting sample and the other for extracting the sample for characterisation.