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# Non-thermal atmospheric pressure plasma jet for the bacterial inactivation in an aqueous medium

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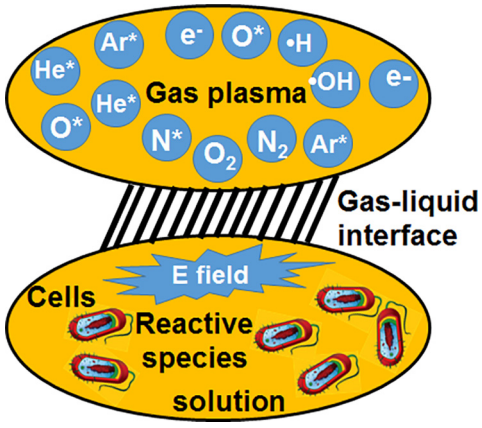
## HIGHLIGHTS

- Non-thermal APPJ has been tested for the inactivation of bacteria.
- The active species such as $H_2O_2$, $HNO_3$, •OH and $O_3$ were quantified.
- Gaseous mixtures followed the order: Ar + air > He + air > air > Ar > He.
- The solution pH plays a prominent role in the bacterial inactivation.
- Addition of $Fe^{2+}$ salt to the plasma treated water improves the efficiency.

## GRAPHICAL ABSTRACT

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## ABSTRACT

This study reports the potential of non-thermal atmospheric pressure plasma jet for the bacterial inactivation in an aqueous medium. All experiments were conducted in a reactor containing aqueous solution i.e., water, pre-inoculated with bacterial suspensions and after plasma exposure solution is inoculated in Petrifilm to know the viable count. The plasma jet exposure to the bacterial aqueous solution was carried out under various gases such as helium, argon, air and also in combination as Argon + Air and Helium + Air. In each case, the oxidizing species such as hydrogen peroxide, nitric acid, hydroxyl radicals and ozone formed in the reactor during the plasma exposure were quantified. The effect of applied voltage and gas flow rate were studied to optimize the conditions for its efficacy. The solution pH plays a prominent role in bacterial inactivation, as the process is effective at low pH exhibiting 7 log reduction of bacterial population. The bacterial inactivation is efficient at below the critical pH (<4.7) and the inactivation process becomes less effective if the pH goes above 4.7. Plasma treatment of deionized water produces some reactive species such as hydrogen peroxide and nitrates, this plasma treated water is used to test the bacterial inactivation. Addition of $Fe^{2+}$ salt to the plasma treated water improves the efficacy by converting hydrogen peroxide to hydroxyl radicals, which serves to be a major contributor to the bacterial inactivation. Especially, Non-thermal plasma offers an alternative way to sterilize vacuum sensitive and thermo-labile living tissues.

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## 1. Introduction

Sterilization of biomaterials during tissue transplantation is of great importance to avoid the transmission of infection to susceptible recipients. Most conventional sterilization techniques make use of chemical agents, ionizing radiations and exposures to high thermal/pressure to get rid of microbial contamination. However these techniques pose a great threat to biomaterials by damaging the biomaterials, making them less suitable for the recipients. (Marsit et al., 2016). And also some sterilization procedures involving UV exposure can lead to photoreactivation of microbial cells which cause serious infections (Guo et al., 2012). The need for appropriate sterilization technique caused a breakthrough in testing of some advanced techniques such as advanced oxidation processes (AOPs) via sonolysis (Feng et al., 2018), photo-Fenton (Guo et al., 2017), exposure to ultraviolet radiation (Ferro et al., 2016) or oxidative gases (Miranda et al., 2016) and a combination of these techniques (Ferro et al., 2016) for the purpose of complete bacterial inactivation. Plasmas have been known to have great potential to inactivate microorganisms due to the ease of mild and ambient operation conditions, whose heat or UV is harmless to living cells (Ikawa et al., 2010). Recently, non-thermal plasmas (NTPs) (Ma et al., 2015), dielectric barrier discharge (Liao et al., 2018), atmospheric pressure plasma jet (APPJ) (Uhm et al., 2007) and needle plasma (Jiang et al., 2017) are employed for this purpose. Plasma is a soup of electrons, free radicals, ions, neutrals, electric field and UV light (Gaunt et al., 2006; Herrmann et al., 1999). NTP based electrical discharges generated at the gas-liquid interface during plasma treatment offers an electric field, UV light, electrons, ions, free radicals, and a wide range of active species like •OH, $H_2O^+$,$•O_2^-$,•H, $NO_3^-$, O, HOO•, $NO_2^-$ and $O_3$ (Gaunt et al., 2006; Herrmann et al., 1999). It is known that, plasma treatment process in solution, superoxide anion radicals ($•O_2^-$) are converted into hydroperoxyl radicals (HOO•), which can easily penetrate into the bacterial cell wall and destroy the intracellular components (Korshunov and Imlay, 2002) and makes the bacterial inactivation process efficient. Last two decades, NTPs created at near ambient temperature has been emerging as a potential reaction medium for various applications such as wastewater treatment (Chandana and Subrahmanyam, 2016; Zhang et al., 2016), surface modification (Keating IV et al., 2018), biomedical (Jiang et al., 2017; Xu et al., 2018; Laroussi, 2009) and food packaging industry (Pankaj et al., 2014). In addition, NTP offers an alternative way to sterilize vacuum sensitive and thermo-labile living tissues (Ikawa et al., 2010). The atmospheric plasmas were first used for sterilization of contaminated matter by Laroussi (1996) which lead to the development of many plasma devices for microbial inactivation on a solid surface (Kayes et al., 2007). Inactivation of *Escherichia coli* (*E. coli*) with plasma-generated atomic oxygen (O) and $•O_2^-$ in the gas phase was reported by Uhm et al. (2007). The effect of $N_2/O_2$ composition on *Escherichia coli* inactivation by discharge plasma at the gas-solution interface (Ke et al., 2017), the biological and physical scenarios of bacterial death induced by non-thermal plasma were reported (Lunov et al., 2016). The efficiency of the cold argon oxygen plasma jet to reduce *Escherichia coli* and *Streptococcus pyogenes* (Mortazavi et al., 2016) were reported recently as advancement in plasma treatment in bacterial inactivation.

Many of the reported applications on NTP based sterilization were focused under dry conditions, whereas sterilization by APPJ under wet conditions is found to be more prominent for the complete destruction of the microorganisms (Ikawa et al., 2010). The treatment mechanism involved in dry conditions significantly differs from the wet conditions. Microorganisms are destroyed physically due to the highly energetic electrons, electric field and UV light in dry conditions, whereas in wet conditions microorganisms were present in water and were killed by reactive species generated in water during plasma treatment. NTP sterilization under wet conditions is relatively tedious due to the dielectric nature of water as in the aqueous medium and also UV, electron/ion interaction with bacteria is limited. Under wet condition, plasma was created at the gas-liquid interface, the gas phase reactive species like Ar*, N*, O* and NO* are transferred to liquid phase to generate reactive oxygen species such as HOO•, $•O_2^-$, hydroxyl radicals (•OH), ozone ($O_3$) and reactive nitrogen species like nitrates ($NO_3^-$) and nitrites ($NO_2^-$) etc. Various attempts have been made for the sterilization of microorganisms under wet conditions, but the quantification of reactive species and optimum conditions for the sterilization process are largely limited (Ikawa et al., 2010; Liu et al., 2010).

In this article, we have reported experimental results for inactivation of bacterial species *E.coli* DH5 alpha, *E.coli* BL21 and *E.coli* K-12. The major objective of the present study was to investigate the bacterial inactivation with APPJ under wet conditions. Long-lived species $H_2O_2$ and $HNO_3$, short-lived species •OH were quantified and conditions favoring their formation were optimized. The influence of various parameters like applied voltage, flow rate of the gaseous mixture, various feed gases and solution pH were considered. The antibacterial activity was performed with plasma treated water (PTW) and PTW containing ferrous salts ($Fe^{2+}$).

## 2. Experimental

### 2.1. Experimental setup

Prior and post to the experiments, the reactor (as shown in Fig. 1a, Chandana and Subrahmanyam, 2016) was well cleaned with commercially available laboratory glassware cleaning detergents, rinsed twice with sterile deionized water and further placed in autoclave for 15 min at 121 °C. The plasma was created between the high voltage electrode (stainless steel rod) and a stainless steel mesh, which also serves as the ground electrode. A high voltage probe was connected to the inner electrode to measure the applied voltage (12–16 kV) and the voltage (V)-charge (Q) wave forms were recorded with an oscilloscope (Tektronix TDS 2014B). The applied voltage was plotted against the charge to get a Lissajous figure, whose area was multiplied by the frequency gives the power dissipated in the discharge (Reddy et al., 2014). The maximum power dissipated in the voltage range 12 to 16 kV was 0.6 W, 0.8 W and 1.0 W respectively, with (Ar + air) gaseous mixture (Fig. 1b).

### 2.2. Reagents and methods

Terephthalic acid (TA), 2-hydroxy terephthalic acid (HTA), hydrogen peroxide ($H_2O_2$), Titanium dioxide ($TiO_2$), sulfuric acid ($H_2SO_4$), hydrogen chloride (HCl) sodium hydroxide (NaOH) and Ferrous chloride ($FeCl_2$) are analytical grade chemicals purchased from Merck, India. The nutrient broth (peptone 10 g/L, beef extract 10 g/L, sodium chloride 5 g/L and pH adjusted to 7.3 ± 0.1) purchased from Himedia, India and 3 M™ Petrifilm™ *E. coli*/Coliform is purchased from 3 M, USA for bacterial quantification. Aalborg mass flow controller (MFC) was used to adjust the gas flow rate of $1.6 \times 10^{-6}$, $3.3 \times 10^{-6}$ and $5 \times 10^{-6}$ $m^3/s$. The pH variations during the discharge process were measured with a pH meter (μ pH System 361 Systronics, India) whereas the changes in conductivity were measured by using a conductivity meter (Systronics, Conductivity Meter 306, India). The concentration of nitrate ions was analyzed by DIONEX ICS-2100 ion chromatography (IC), where 0.38 mL/min flow rate, 25 mM KOH eluent, 24 mA current and 10 μL injection volume was used and the column temperature was maintained at 30 °C. The influence of the various parameters like applied voltage (12–16 kV), flow rate of the gaseous mixture ($1.6 \times 10^{-6}$, $3.3 \times 10^{-6}$ and $5 \times 10^{-6}$ $m^3/s$), various feed gases (He, Ar, Air, Ar + Air, He + Air), solution pH (4.0–7.0) and the plasma treatment time (30 s to 150 s) were studied. In this work, bacterial inactivation was carried out in two different ways: one is with direct plasma treatment of pre-inoculated bacterial suspension. After plasma exposure solution is inoculated in Petrifilm to know the viable count. The other one is with plasma treated water-plasma treatment of deionized water produces

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