

Low-Cost Dielectric Barrier Discharge Plasma Hand Sanitizer Using Air and Tap Water Enriched by Hydrogen Peroxide

M. El Shaer,* M. Mobasher, & A. Zaki

Plasma & Energy Applications Research Laboratory, Zagazig, Faculty of Engineering, Zagazig University, Zagazig, Egypt

*Address all correspondence to: M. El Shaer, Department of Engineering, Physics, and Mathematics, Faculty of Engineering, Zagazig University, 44599, Zagazig, Egypt; Tel.: +20-100-695-0077; Fax: +20552304987, E-mail: melshaer@link.net

ABSTRACT: A low-cost plasma hand sanitizer with minimum capital and running costs, combining flexibility in operation and high efficacy in treating common pathogens, is proposed for sanitizing the hands of medical doctors and nurses. Hands are exposed to a mist that is generated using tap water and activated by plasma coaxial dielectric barrier discharge (DBD) in ambient air. A common ignition transformer operating in the kilohertz range is used for the power supply, and a commercial ultrasonic nebulizer generates the mist. To enhance complete hand sanitization and reduce exposure time without adding oxygen gas to the discharge, solutions of commercial hydrogen peroxide in small concentrations are added to the water that is introduced in the plasma in the form of a mist. The active species produced by DBD during operation as O_3 and OH is measured in the treatment zone near the end of the discharge tube. By adding increasing amounts of hydrogen peroxide in the water, we find that OH radicals increase, and measured ozone concentrations in ambient air decrease. This can be considered a beneficial chemical transformation, reducing possible harmful effects of ozone and enhancing sanitization efficacy. Sanitizing effects are reached with a low hydrogen peroxide concentration of 3 mg/L to avoid any harmful effects on the skin of the hands.

KEY WORDS: DBD, sterilization, *E. coli*, ozone, hydroxyl radicals, hand hygiene, mist

I. INTRODUCTION

Plasma sterilization has proven its effectiveness in killing large numbers of varied pathogens in rather short time intervals.¹ Hand hygiene is a critical issue in the prevention of many infections and diseases, especially in medical professions.² Hand sanitization using plasma under the flow of a water mist has been already proposed.³ Adding water mists of different relative humidity ratios can influence plasma's killing effect on most types of bacteria.⁴ Plasma discharges at atmospheric pressure occurring in air or in different working gases produce various behaviors, depending on the relative humidity ratios in the discharge medium.^{5,6} Discharge electrical characteristics and modes of operation are also affected by varying relative humidity ratios.

It has been found that beyond a certain relative humidity level, discharge can operate in a complete diffuse mode without microdischarges at maximum power.⁷ To increase the efficacy of treating microorganisms by plasma under the flow of a water mist, many

authors have proposed the addition of small amounts of oxygen to the discharge gas.⁸ Because our objective is to perfect a practical hand sanitizer, we reasoned that the introduction of oxygen gas could complicate device operation in unequipped user facilities. We thus propose the addition of hydrogen peroxide solutions to the tap water that is used to produce plasma-activated mist. We examine bactericidal properties of mist enriched by hydrogen peroxide introduced in plasma to improve sanitizing effects and shorten necessary treatment time for bacteria elimination. This effect was reported by Yamamoto et al.,⁹ who investigated the injection of H_2O_2 droplets in a corona discharge. Gołkowski et al.¹⁰ indicated that the efficacy of treatment with dielectric barrier discharge (DBD) is proportional to the amount of hydrogen peroxide present during treatment. Kang et al.¹¹ related the concentration of added hydrogen peroxide to the amount of active species generated in plasma, especially ozone and hydroxyl radicals. Here, we study the effect of adding small amounts of hydrogen peroxide to the water used to produce the mist that is introduced in atmospheric pressure DBD plasma to enhance plasma-sanitizing effects and reduce the time necessary for hand sanitization.

II. MATERIALS AND METHODS

A. Experimental Setup

Plasma was generated by a coaxial DBD in ambient air under the flow of a mist from tap water. The setup of the experiment can be seen in Fig. 1. The power supply was a common ignition transformer (Satronic [Honeywell]; model ZT 931) that delivers a voltage

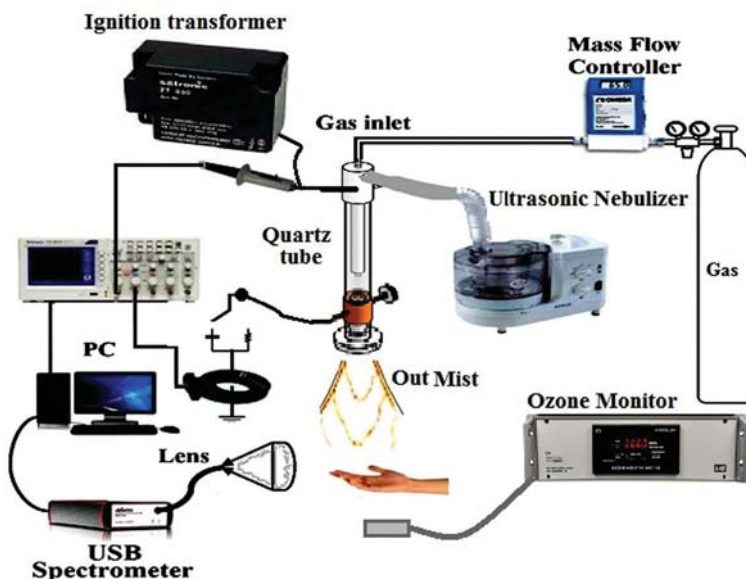


FIG. 1: Experimental arrangement of plasma hand sanitizer

of 14 kV and a root mean square current of 40 mA at a frequency of 20 kHz, for a duty cycle of 3 min. High voltage was measured using a high-voltage probe with a ratio of 1000:1 (Tektronix, Beaverton, OR; model p6015A, 75 MHz) and current was recorded with the use of a Rogowski coil (Pearson Electronics, Palo Alto, CA; model 4100, 1 V/1 A). The discharge gas was ambient air, but in some cases, helium could be used with small additives of oxygen. Gas or simply ambient air was mixed with a water mist that was delivered by a commercial ultrasonic nebulizer (Jiangsu Folee Medical Equipment Co., Jiangsu, China; model W001), producing 30- μ m-diameter water droplets. To increase the efficacy of treating microorganisms by plasma under a flow of water mist, we added small amounts of commercial solutions of H₂O₂ of differing concentrations to the tap water that filled the nebulizer tank. The concentration of hydrogen peroxide in water was determined using peroxides test sticks (Quantofix [Sigma-Aldrich]) with different concentration ranges in milligrams per liter. At the exit area of the discharge tube, we measured ozone in ambient air using a low-concentration ambient ozone monitor (BMT MESSTECHNIK GMBH, Stansdorf, Germany; model BMT 932) with a range of 0–10 ppm and a resolution of 0.001 ppm.

Discharge occurred between two copper cylindrical electrodes, situated inside and outside of a quartz tube that acted as a dielectric barrier. The quartz tube measured 200 mm long, with an inner diameter of 20 mm and wall thickness of 1.5 mm. The inner electrode had an outer diameter of 15 mm and was connected to a high-voltage–high-frequency power supply. The outer electrode had an inner radius of 23 mm and was connected to the ground. The air gap of the cylindrical DBD was 2.5 mm. A water mist was introduced in the space between the two electrodes through a hollow Teflon tube, assuring adequate mixing between mist and injected gas or inlet ambient air. Details of the discharge tube are shown in Fig. 2.

III. RESULTS

To identify the sanitizing effect that occurs after adding different concentrations of hydrogen peroxide solutions to the tap water that is used to generate plasma-activated mist, surviving *Escherichia coli* subjected to the mist were assessed using a colony-counting technique. For comparison purposes, *E. coli* cultures are first subjected to mist from water alone, without plasma or any other additives; no effects were observed on bacteria cultures. By adding small concentrations of hydrogen peroxide solution to the tap water in the nebulizer, bacteria survival rates were moderately reduced. When the plasma was turned on with the introduction of the mist enriched by hydrogen peroxide, bacterial survival rates considerably decreased. Figure 3 shows bacteria effects after adding 3-mg/L hydrogen peroxide to the tap water used in mist production at different plasma exposure times. We observed a large reduction in bacteria after 90 s of plasma exposure.

Figure 4 shows the rates of *E. coli* colony-forming units (CFUs) in three different conditions. *E. coli* cultures were first treated by mist alone that was generated using tap water. Next, the bacteria were treated with mist from the water solution that contained

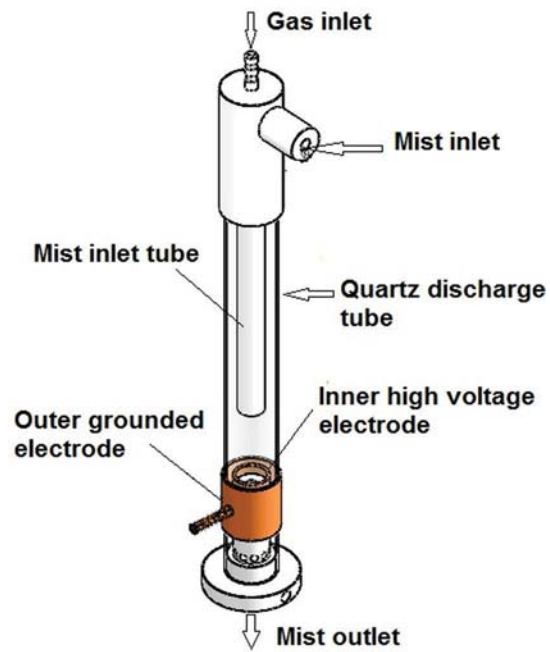


FIG. 2: Discharge tube details

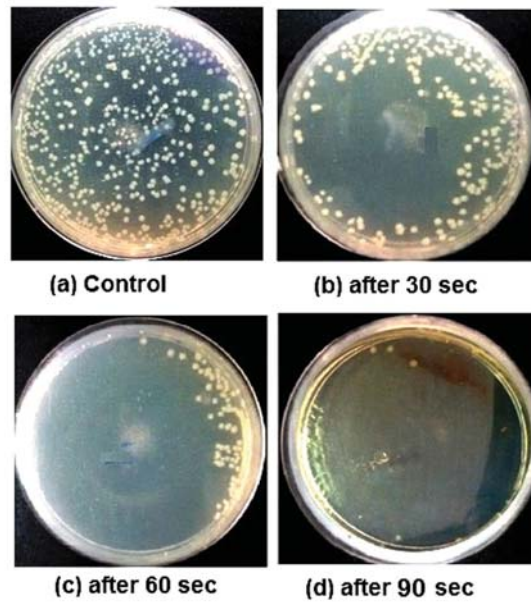


FIG. 3: Petri dishes of *E. coli* cultures at different treatment times. (a) Control bacteria culture; bacteria culture treated by plasma with mist enriched by 3-mg/L H_2O_2 after (b) 30 s, (c) 60 s, and (d) 90 s

3-mg/L hydrogen peroxide. Finally, *E. coli* were treated using mist with hydrogen peroxide in the presence of plasma discharge. The rates show that bacteria survival rates are moderately reduced when hydrogen peroxide is added to the mist without plasma but are largely reduced when plasma is turned on during the injection of hydrogen peroxide-enriched mist.

Figure 5 shows the percentage of reduced *E. coli* CFUs at 90 s of treatment with mist alone, mist with H₂O₂, and mist with H₂O₂ and plasma. One can see that a greater

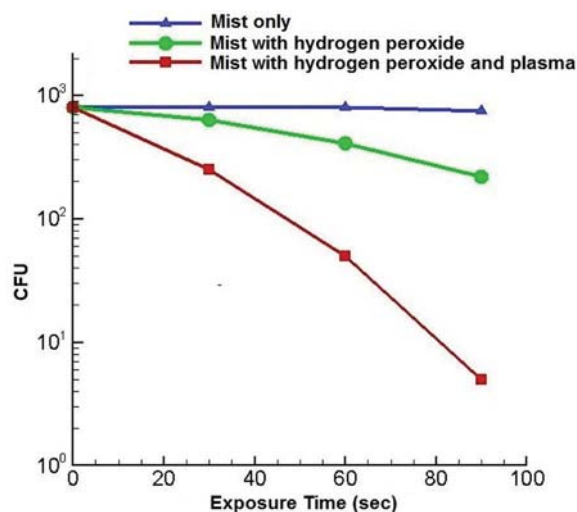


FIG. 4: CFU values of *E. coli* as a function of exposure time at different treatment conditions

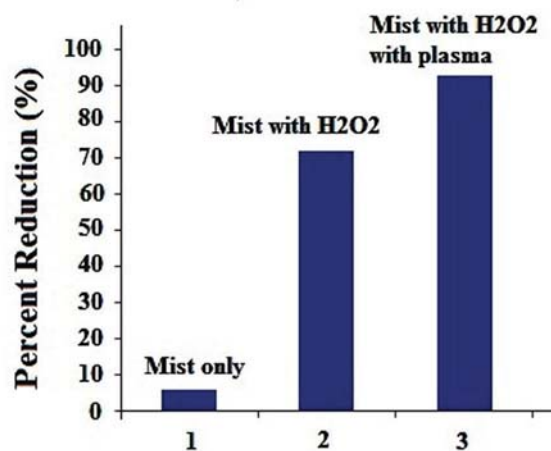


FIG. 5: Percentage of reduction in *E. coli* CFUs at 90-s plasma treatment time for different treatment conditions

reduction occurs when hydrogen peroxide is added at even a small concentration (3 mg/L) to the tap water used to generate plasma-activated mist. Because ozone produced by plasma discharge has a very important role in the sterilization process, ozone concentration was measured in ambient air near the discharge tube, 5 cm apart from the lower end of the tube's output. Results of ozone measurements as a function of discharge time are shown in Fig. 6 for different hydrogen peroxide concentrations in the water used for mist production in the presence of plasma. To clarify the relationship between ozone concentration produced during plasma discharge time and hydrogen peroxide concentration added to the water, we greatly increased hydrogen peroxide concentrations in the water solution and measured the corresponding ozone concentrations in ambient air at different discharge times. In Fig. 6, it is evident that for different hydrogen peroxide concentrations, ozone concentrations gradually increased with discharge time. For certain discharge times, we can observe that concentrations of ozone decreased with increasing concentrations of hydrogen peroxide added to the water used for mist production.

Optical emission spectroscopy in the ultraviolet-visible range is usually used to identify atomic and molecular species produced during discharge.¹² In our study, spectroscopic measurements were taken with a fiber optics spectrometer (AVANTES, Apeldoorn, The Netherlands; AvaSpec-ULS2048 StarLine) with a wavelength range of 200–1100 nm and a resolution 0.9 nm. We investigated emission spectra of plasma produced in ambient air that had been injected with mist produced using hydrogen peroxide-enriched water. Figure 7 shows the N_2 and OH lines for plasma with mist produced by a solution of 256-mg/L hydrogen peroxide in water.

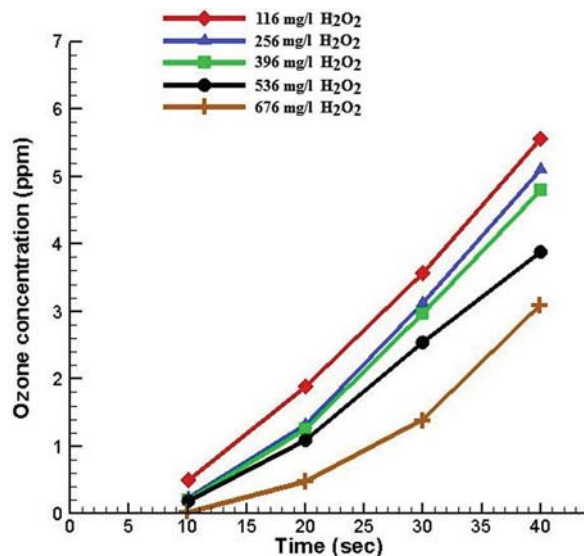


FIG. 6: Ozone concentration in ambient air as a function of plasma discharge time for different concentrations of H_2O_2 in the water solution

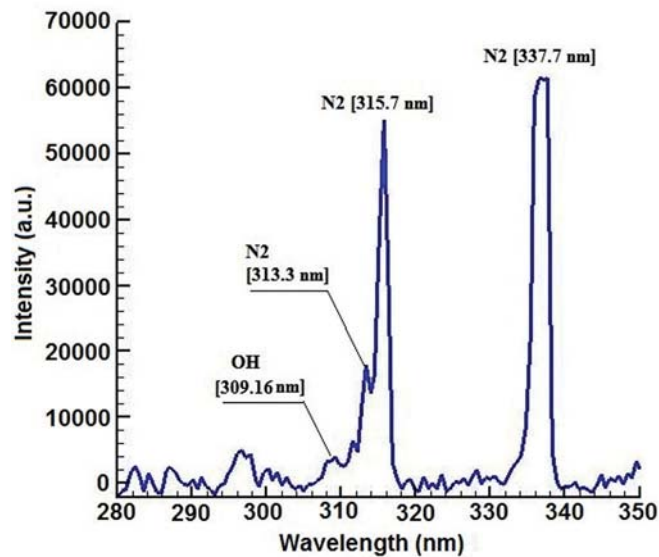


FIG. 7: Measured optical emission spectra for plasma with the mist produced by a solution of 256-mg/L hydrogen peroxide in water

Details of spectral line intensities around the OH line of 309.16 nm are shown in Fig. 8 for different concentrations of hydrogen peroxide in the water used for mist production in the presence of plasma. The spectral line intensity of OH seems to increase with greater hydrogen peroxide concentrations.

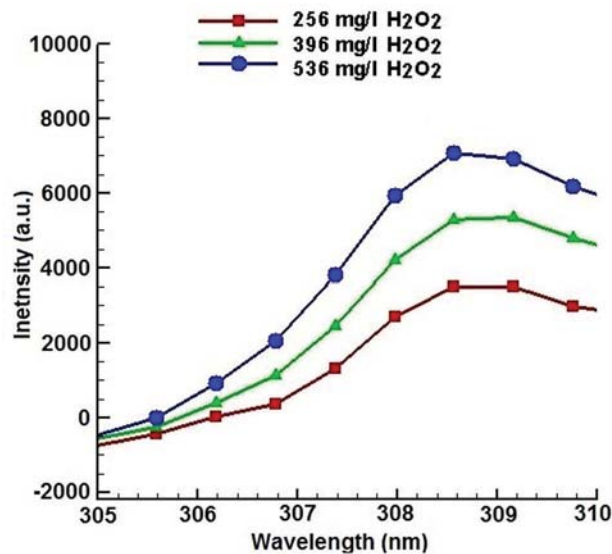
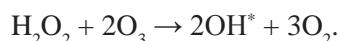


FIG. 8: Details of measured optical emission spectra at 309.16 nm OH spectral line for plasma produced by adding different hydrogen peroxide concentrations in the water used for mist production

IV. DISCUSSION

In biomedical sanitization, it is common to use ozone and hydrogen peroxide as disinfectants for cleaning surfaces. Atmospheric plasma is found to produce atomic N, O, and H radicals as well as NO and OH that initiate plasma chemical processes, leading to the formation of interesting species, including O_3 , H_2O_2 , and OH, for decontamination purposes. Such species have the ability to destroy a large variety of pathogens. Improvement of plasma sanitization at atmospheric pressure can be achieved with the introduction of mists in the plasma discharge, which may enhance the production of reactive species. By adding small amounts of hydrogen peroxide to the water solution used in mist production, we can enrich the plasma with extra reactive species such as O_3 and H_2O_2 . These are effective agents for oxidative degradation of nutritive materials in bacterial cells. OH radicals are one of the strongest oxidative elements. Reduction in ozone concentration and increases in OH radicals can be achieved using the following chemical reaction:



The production of hydroxyl radicals, via the addition of hydrogen peroxide, increases the killing of bacteria and reduces ozone production, which can cause skin to be compromised during hand sanitization. Only a small concentration of hydrogen peroxide (3 mg/L) is needed to eliminate most *E. coli* bacteria in 90 s. This can be considered as an acceptable scheme for practical hand sanitization for medical purposes.

Figure 9 summarizes the effect of increasing hydrogen peroxide concentrations on ozone and OH production at a 40-s discharge time. By increasing hydrogen peroxide

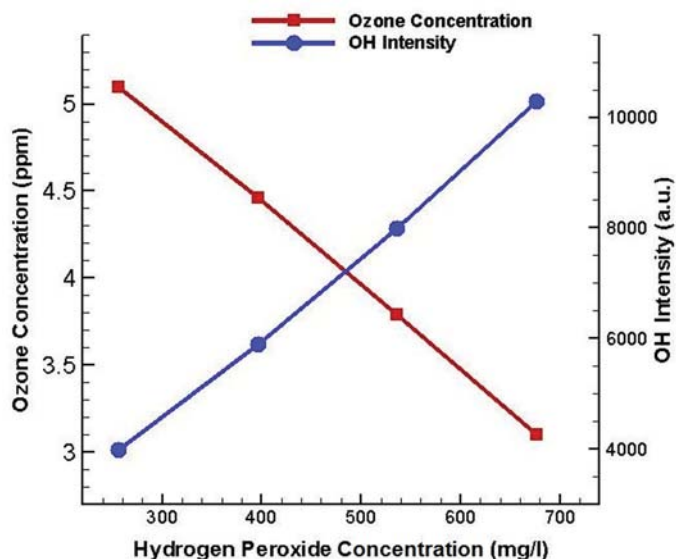


FIG. 9: Ozone and OH concentrations as a function of hydrogen peroxide concentration in water, measured at 40-s plasma discharge time

concentrations, we observe that the reduction in ozone concentration is accompanied by increased OH intensity. Adding H₂O₂ in small concentrations to the water used to produce the mist injected in plasma may reduce ozone and increase OH radicals. Because ozone generated by atmospheric plasma can be toxic and carcinogenic, its chemical reaction with added hydrogen peroxide may reduce its content in the mist and simultaneously increase short-lived nontoxic OH radicals. This will result in more effective and safer hand sanitization.

V. CONCLUSION

Combining the effects of O₃, H₂O₂, and OH during plasma discharge can achieve complete and safe sanitization of hands in medical activities. Adding a small amount (3 mg/L) of hydrogen peroxide to the tap water solution used to produce plasma-activated mist safely eliminates *E. coli* bacteria after ~90 s of treatment. During DBD with hydrogen peroxide-enriched mist, we measured a reduction in undesirable ozone along with a desired increase in OH formation. By-products of such reactions are oxygen and hydrogen, which ensure bacteria inactivation with minimum toxic or carcinogenic side effects. Further research is needed to study all active species produced and define the points of operation in hand sanitization to reduce undesirable species on and increase strong and safe sanitization. It is important to maintain a low as possible amount of added hydrogen peroxide to avoid any harmful effects on hand skin.

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