Comparison of Direct and Indirect Effects of Non-Thermal Atmospheric-Pressure Plasma on Bacteria

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Direct and indirect influence on microorganisms of non-thermal atmospheric-pressure DBD in air is compared in terms of bacterial inactivation rates. It is demonstrated that direct influence of the plasma, where charged particles contact bacteria directly, produces inactivation much faster than the indirect treatment, where plasma afterglow is delivered to the bacteria with a gas flow through the plasma region. This leads to an important conclusion that the effect of charged particles on plasma plays the essential role in interaction with living organisms, although synergy with longer living active molecules and atoms as well as UV radiation generated in plasma and at the surface of tissues may also play a role in inactivation.



Introduction

Over the past few years, new non-thermal atmospheric pressure plasma (APP) devices have emerged with several important applications in medicine. These applications involve possibly selective inactivation of unhealthy cells,[1–8] blood coagulation,[5,8–10] and killing of parasites, bacteria, fungi, and viruses on living tissue.[5,7,11–23] Another attractive application is the sterilization of various surfaces.[2,7,11,15,24–29] Two different approaches have been suggested for atmospheric-pressure non-thermal plasma-based sterilization of living tissues. These two approaches can be classified as indirect and direct. Indirect treatment uses a jet of products (plasma afterglow) generated in remotely located plasma discharge. Direct treatment, by contrast, uses the tissue itself as an electrode that participates in creating the plasma discharge, as illustrated in Figure 1. Plasma in this case is contained between the quartz surface of the powered high

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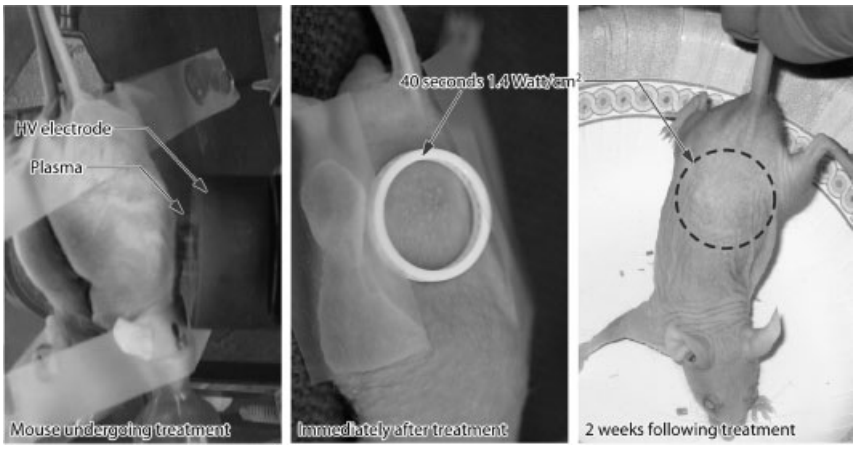


Figure 1. Non-damaging room temperature and pressure plasma for the treatment of living tissue: animal treated for up to 10 min remains healthy and no tissue damage is observed visually or microscopically immediately after and up to 2 weeks following the treatment.

voltage electrode and the surface of bacteria or tissue being treated. It is important to stress that the distinction between these two approaches is not only related to the proximity of the plasma and the tissue, but that a direct contact with plasma brings charged energetic particles to the plasma/tissue interface. By contrast, no charged particles are usually taken out of the plasma region by a jet even if the plasma region is located only a fraction of a millimeter away. The question arises: Is there a substantial difference between the two types of treatments? In other words, is the transport of charged particles to the surface being treated as important or is the effect of plasma due mainly to long-living active molecules and longer-wavelength ultraviolet (UV) radiation (as short-wavelength "vacuum" UV is absorbed by air at atmospheric pressure within some μm).[28,30–33]

In this paper, we compare the effectiveness of direct and indirect sterilization treatment by non-thermal APP generated using the same discharge setup, and demonstrate that the direct treatment can achieve sterilization much faster without any thermal effects. Specifically, we employ the same dielectric-barrier discharge (DBD) to compare both types of treatments. To do that, we carried out two types of experiments. In one set of experiments we employed a surface covered by bacteria as a DBD electrode, placed a second smaller area DBD electrode over it and compared the areas of sterilization with air flow parallel to the surface and without the air flow. In another set of experiments we again employed a surface covered by bacteria as one DBD electrode and compared the rate of sterilization to the case when this surface was separated from the DBD discharge by a grounded mesh electrode (see Figure 2).

Results and Discussion

Careful sample preparation is required before carrying out the plasma experiments. Since our interest is primarily in the sterilization of skin of an animal or human, bacterial samples were collected from deidentified skin samples from human cadavers at the Hahnemann Hospital (Philadelphia, PA). Identification of the bacterial species was performed using standard methodology at the Hahnemann Hospital's Clinical Microbiology Laboratories. Bacteria from skin samples consisted of *Staphylococci*, *Streptococci*, and *Candida* species of yeast. To collect these microorganisms, six skin samples were swabbed twice each by sterile cotton-tipped applicators pre-wetted with 10× phosphate- buffered saline (PBS). These applicators were then transferred into 5 mL of 10× PBS and placed into an ultrasonic bath for 10 min to lift off the bacteria from the swab. The resulting solution was transferred onto the blood agar plate (Trypticase™ Soy Agar with 5% sheep blood) and incubated in air at 37 °C for 24 h. The resulting colonies were collected and reinoculated onto fresh blood agar plates and again incubated for 24 h. These steps were

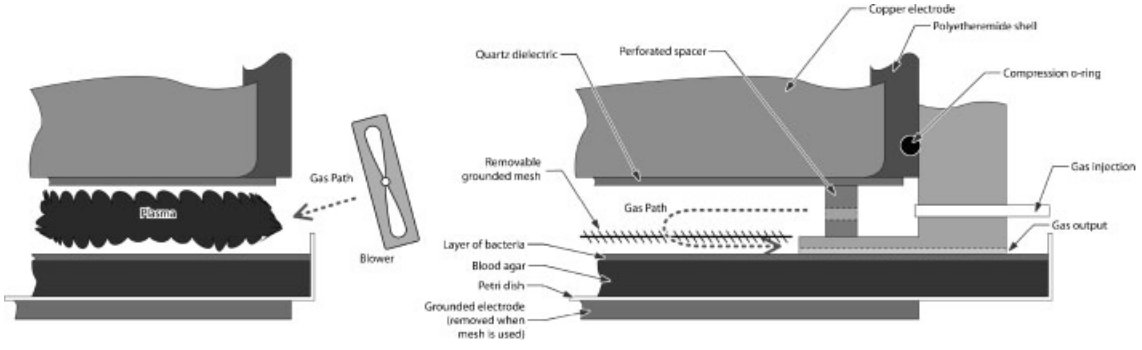


Figure 2. Direct and indirect plasma experimental setups: grounded mesh is removed for direct plasma treatment and placed back for indirect treatment (left) or air is blown through plasma to carry out long-lived species (right).

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repeated to obtain large quantities of bacteria. In the end of this preparation, bacteria were collected from the plates and diluted in 10× PBS to approximately 10^9 colony-forming units (CFU) per mL. Bacterial concentration was assessed by a standard dilution assay.[34,35]

Since the results of the plasma treatment are characterized below in terms of appearance of the blood agar surface after the treatment, it is important to normalize the results by establishing how the agar surface would look for different concentrations of CFUs on its surface. To do that, we exposed an agar-covered Petri dish with bacteria cultured on its surface from samples prepared as described above[5] to the DBD plasma for different times at fixed plasma power levels. The exact plasma setup, surface power density, and other plasma parameters used for these normalization experiments are the same as those used in the experiments described later that compare direct and indirect treatment.

To quantify the sterilization efficiency, 20 μL drops of 10^9 cfu · mL^{-1} bacteria were placed on agar surface, left to dry for 5 min, and finally treated by plasma. This volume was selected as it spread to ≈1 cm^2 over the agar surface; thus, the area covered by the bacterial sample drop was entirely within the area covered by the insulated plasma electrode. Following the treatment, the drop was spread over the entire agar surface and incubated in air at 37 °C for 24 h. Bacterial colonies (CFUs) were then counted and the results are presented in Table 1. There is a clearly visible difference in agar appearance between untreated, partially sterilized, and completely sterilized agar. Based on this difference, it is reasonable to classify the appearance of the bacterial surface into five categories: (1) untreated (10^9 cfu · mL^{-1}), (2) partially disinfected (10^9–10^7 cfu · mL^{-1}), (3) disinfected (10^7–10^4 cfu · mL^{-1}), (4) partially sterilized (1 000–10 cfu · mL^{-1}), and (5) completely sterilized (0 cfu · mL^{-1}). Note that the definitions of sterilization and disinfection follow those that are accepted in the literature.[2,5,15,27,28,36,37]

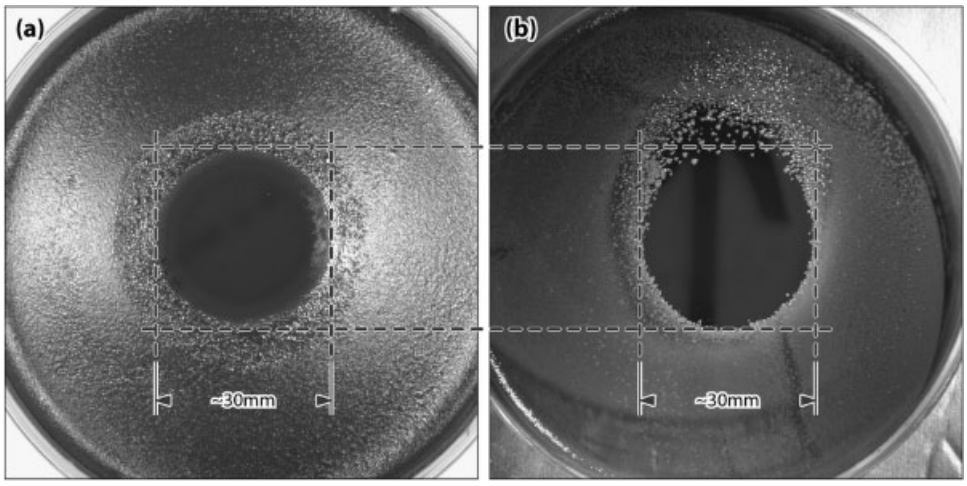


Figure 3. Using a blower to shift the sterilization region does not affect plasma and shows a little effect on the afterglow: 15 s of treatment with (a) blower off and (b) blower on (air flows up).

Table 1. Bacteria sterilization results (all values in cfu · mL^{-1}).

Original concentration	5 s of FE-DBD	10 s of FE-DBD	15 s of FE-DBD
10^9	850 ± 183	9 ± 3	4 ± 4
10^8	22 ± 5	5 ± 5	0 ± 0
10^7	6 ± 6	0 ± 0	0 ± 0

To quantify the extent of sterilization, 1 mL of 10^9 cfu · mL^{-1} was poured over the entire agar surface. These samples were left to dry for 3 h and then treated by plasma. Here plasma only covered a portion of the Petri dish. Following the 24 h incubation period, the extent of sterilization was clearly visible - areas where bacteria were killed looked like uncontaminated agar while areas that received no treatment changed color and appearance significantly as the bacteria grew there. The complete sterilization area is easiest to identify - it corresponds to the agar area that is completely clear from bacteria (e.g., see Figure 3 or ref.[5]). Partial sterilization is also relatively easy to define since the number of CFUs is relatively small and can be counted. Disinfection is more difficult to assess because of the difficulty of counting the large number of CFUs. As can be seen from Figure 3, the complete sterilization gradually fades into untreated areas, forming a "gray-scale" fade that gradually increases from 0 cfu · mL^{-1} in the sterile zone to 10^9 cfu · mL^{-1} in the untreated zone much like in the quantitative experiments described in the previous paragraph.

We now describe the first of the two types of experiments that compare direct and indirect plasma treatments. The setup is illustrated in Figure 2. To test the sterilization treatment, we used an agar-covered Petri dish with bacteria spread on its surface from the earlier prepared samples of 10^9 cfu · mL^{-1} concentration. Specifically, 1 mL of such bacterial sample was transferred onto the blood agar and spread over the entire plate. The plates were dried for 3 h in a Class II biological safety cabinet (SterilchemGard™ SG-4TX, The Baker Company) prior to

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the plasma treatment. After the plasma treatment, the samples were incubated in air at 37 °C for 24 h to assess the ability of the DBD plasma to sterilize. Control experiments with gas flow only were performed for all the gas flows shown here. Bacteria were completely unaffected by flowing room air in up to 30 min (maximum time tested) as these bacteria were at the surface of a hydrogel (agar) which takes over 3 d to dry. When exposed to dry air agar surface begins to show signs of drying, beginning at 10 min, but bacteria were unaffected by this even after 30 min of dry air[a].

When placed on a metal substrate, the agar dish with bacteria on its surface acts as one of the DBD electrodes - bacteria are at the plasma/agar interface. The other smaller area DBD electrode is made of 2.5 cm diameter solid copper disc separated from the plasma by a 1 mm thick quartz dielectric. Plasma gap or the distance between quartz/plasma and agar/plasma interfaces was set to 1.5 mm. Alternating polarity sinusoidal voltage of 35 kV magnitude (peak-to-peak) and 12 kHz frequency were applied between the copper electrode and the aluminum substrate under the agar Petri dish. A standard 120 mm high speed fan (Multicomp MC1122HBT, $120 \times 120 \times 25$ mm^3, 40 slps) was placed ≈20 cm away from the discharge and the angle of the fan was adjusted to allow for the maximum air flow parallel to the substrate through the plasma gap. Flow speed for this experiment was measured between the electrode and the substrate, directly outside of the electrode, to be 0.8 ± 0.3 m·s^{-1} (EXTECH Instruments model 407123 Hot Wire Thermo-Anemometer).

The power surface density delivered into the plasma discharge was measured by two methods. One was the more traditional electrical measurement method involving voltage across the discharge and current into it. This measurement was performed using a high speed high voltage probe (PVM-4 1000:1, North Star High Voltage, Marana, AZ) and high speed current probe (1 V·A^{-1} +1/−0% sensitivity, 10 ns usable rise time, 35 MHz bandwidth, Model 4100 Current Monitor, Pearson Electronics, Palo Alto, CA). Signals from the probes were measured and integrated using a high speed oscilloscope (500 MHz bandwidth, 5×10^9 samples·s^{-1}, TDS5052B, Tektronix, Inc., Richardson, TX). These electrical measurements were confirmed using custom-built calorimetric measurement setup. Both electrical and calorimetric measurements indicated the power of 0.8 ± 0.2 W·cm^{-2}.

When there is no air flow parallel to the surface, the area directly under the second smaller electrode is exposed simultaneously to charged particles, UV, and the plasma afterglow containing longer living active plasma components such as ozone (O_3), nitric oxide (NO), hydroxyl radicals (OH), and other excited molecular and atomic species. In the presence of the airflow, the area under the second smaller electrode experiences practically the same conditions as without the flow. However, in the presence of flow the plasma afterglow will also impinge on the areas of the substrate that are not directly under the second smaller electrode.

It should be stressed that, even in the absence of any flow, the sterilization region is not confined to the diameter of the copper electrode. With increase in treatment time, this region extends several millimeters outside the electrode. There are two reasons for this: first, due to the existence of surface discharge near the base of DBD filaments, the "footprint" of a microchannel is substantially larger than its diameter.[5,38] Second, since the field lines originating near the edge of the insulated electrode tend to spread out slightly, the discharge filaments also spread out beyond the area of the top electrode. The flow, however, is not expected to change the shape of the plasma discharge region because it is not capable of moving the filaments further than a fraction of a millimeter, as was observed with high speed photography (not shown) as well as theoretically.[38,39] However, such effects are expected to be confined to within millimeters from the edge of the copper electrode, while the longer living active species and radicals can spread much further with the gas flow.[27,28,38–41]

The results comparing the sterilization with and without flow are illustrated in Figure 3. As can be seen from this figure, complete sterilization by direct plasma treatment under the insulated electrode is achieved in 15 s. If air is passed parallel to the substrate through the plasma region, the disinfection area shifts by several millimeters, but a "tail" of disinfection appears. Even though a complete sterilization is not observed in this "tail", different degrees of disinfections are evident. Locations closer to the plasma region have greater degree of disinfection. This suggests that plasma "afterglow", or the gas carried through and outside of plasma, does contain active species and radicals that have bactericidal effects, although direct plasma is substantially more potent as a sterilizing agent. Complete sterilization, however, did not extend to the region reachable by the plasma afterglow in as much as 5 min of treatment. This clearly demonstrates that longer living active species generated in plasma are much less effective than the direct plasma where charged particles participate in the process.

In the first type of experiments described above, effects of plasma "afterglow" were separated from the effects of charged particles and from the UV radiation. In the second type of experiments described below, plasma afterglow

[a] Also of note is that the humidity in the incubator is maintained at >80% so if the agar dries outside, once placed in the incubator, bacteria get all the moisture and nutrients necessary for their proliferation.

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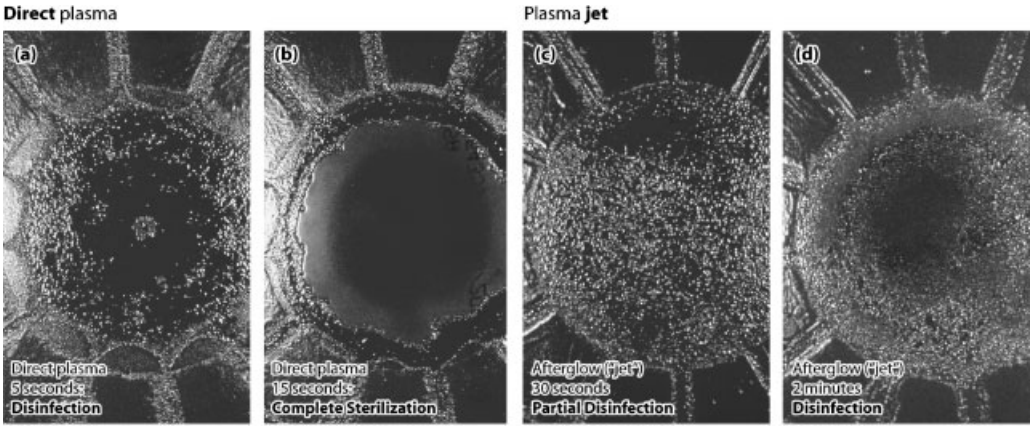


Figure 4. Direct application of plasma yields to better sterilization efficiency than the treatment by plasma afterglow: (a) 5 s and (b) 15 s of direct plasma compared with (c) 30 s and (d) 2 min of plasma jet.

together with UV is compared against the direct plasma, also involving charged particles. The direct treatment is obtained similarly to the first type of experiments described above. However, instead of creating airflow parallel to the substrate, provisions were made to allow the introduction of air at different flow rates into the discharge gap (Figure 2). To obtain an indirect treatment including UV effects, agar surface covered by bacteria was again employed as one DBD electrode; however, this surface was separated from the DBD discharge by a mesh electrode, as illustrated in Figure 2 on the right. With a grounded mesh in place, bacteria are no longer at the plasma interface, as plasma is bounded by the grounded mesh. The mesh (22.04 wires per cm, 0.1016 mm wire diameter, 0.351 mm opening, 60% open area, weaved mesh) is placed 1 mm above the surface of the agar, which permits only the passage of UV radiation and longer wavelength photons (not vacuum UV), gas, radicals, and other longer living particles created in the plasma gap between the mesh and the quartz-insulated electrode above it. The second electrode used with the mesh was the same electrode that was employed in the direct treatment setup illustrated in Figure 2 on the left. The gap between the mesh and the second electrode is set at 1.5 mm, just as in the plasma gap of the direct treatment setup. The setup used to introduce air into the discharge gap at three different flow rates (1, 5, and 10 slpm, corresponding to flow speeds of 0.078, 0.393, and 0.853 $m \cdot s^{-1}$ through the mesh) is illustrated in Figure 2 on the right. The air was introduced into the discharge gap to enhance the extraction of the plasma afterglow through the mesh. In both direct (without the mesh) and indirect (with the mesh) situations the same air flow rates were employed and the discharge was calibrated to the same plasma power for both cases.

To ensure that both setups, with and without the mesh, produce the same UV radiation, we correct for the UV transparency of the mesh using separate measurements. Regular paper is placed over the light source in these measurements to diffuse the light and a spectrum is measured using TriVista Spectrometer System with Princeton Instruments PIMAX intensified CCD camera with only paper, and with paper and mesh. It turns out that the mesh cuts off roughly ≈40% of the visible light and ≈20–40% of UV light, depending on the wavelength of UV (data not shown).

The results comparing direct (without the mesh) and indirect (with the mesh) sterilization are illustrated in Figure 4[b]. It is clearly visible that plasma that comes in direct contact with bacteria is able to sterilize significantly faster than afterglow or jet. Only 5 s of direct plasma treatment results in the appearance of a sterilization region (spot near the center). Complete sterilization occurs within 15 s when direct treatment is employed and only partial disinfection can be achieved within the same time frame with indirect treatment. Over 5 min of indirect treatment is required to achieve sterilization results similar to the direct treatment obtained within 15 s. Therefore, even with substantial UV radiation, indirect plasma treatment is substantially weaker than the direct plasma which includes bactericidal effects due to charged particles.

[b] Note: The spider web-like pattern appears on agar because the electrode is "standing" on the agar and the gas flow through plasma escapes through the 12 equally spaced channels on the bottom of the electrode. These channels are marked "gas output" on Figure 2 and are responsible for the pattern.

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Conclusion

We have carried out a sequence of experiments has been carried out designed to compare direct sterilization by the non-thermal atmospheric pressure DBD plasma in air with indirect effects of various plasma components that can exist outside the plasma region. The results indicate that the direct sterilization is more than one order of magnitude faster. Heating of the bacteria-bearing surface was not observed - the agar would have melted at temperatures around 50 °C, and we did not observe indications of the agar surface melting. We therefore attribute the effectiveness of the direct plasma sterilization to charged particles and fields, microthermal effects, and vacuum UV radiation created by them locally at the bacteria surface.

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