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A Review of Dielectric Barrier Discharge Cold Atmospheric Plasma for Surface Sterilization and Decontamination

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***Abstract*— Numerous investigations have shown that non-equilibrium discharges at atmospheric pressure, also known as "cold atmospheric plasma" (CAP) are efficient to remove biological contaminants from surfaces of a variety of materials. Recently, CAP has quickly advanced as a technique for microbial cleaning, wound healing, and cancer therapy due to the chemical and biologically active radicals it produces, known collectively as reactive oxygen and nitrogen species (RONS). This article reviews studies pertaining to one of the atmospheric plasma sources known as Dielectric Barrier Discharge (DBD) which has been widely used to treat materials with microbes for sterilization, disinfection, and decontamination purposes. To advance research in cold atmospheric plasma applications, this review discusses various types and configurations of barrier discharge, the role played by reactive species and other DBD-CAP agents leading to its antimicrobial efficacy, a few collection of DBD-CAP past studies specifically on surface, and emerging applications of DBD-CAP technology. Our review showed that non-thermal/equilibrium plasma generated from DBD could sterilize or disinfect surface of materials without causing any thermal damage or environmental contamination.**

***Index Terms*— Dielectric barrier discharge, cold atmospheric plasma, bacterial biofilm, decontamination, sterilization.**

I. INTRODUCTION

COLD Atmospheric Plasmas (CAP) are weakly ionized gases with charged species including electrons, positively/negatively charged ions, and neutral species (atomic and/or molecular radicals and non-radicals) under electric fields. [1]–[4].These plasmas, which are also known as low temperature non-equilibrium plasmas, can be produced at low pressures 0 between 100 Pa and 100 kPa, and even atmospheric pressure above 100 kPa.

Cold atmospheric plasmas generally have a gas temperature of less than 100 °C and an ionization level of less than 0.1%. Traditionally, the study of low temperature plasma discharges was primarily focused on two areas: (1) Low pressure discharges ($<$ 1 Torr) mainly used in semiconductor industries for surface cleaning, etching, and deposition, and (2) High pressure discharges (up to 1 atm) with applications to ozonizers, electrostatic precipitators, and other devices [5].

Cleaning, sterilization, and decontamination using CAP have been investigated over the past 20 years [6]–[15]. For example, several CAP-based cleaning techniques were developed to treat heat-sensitive components of devices exposed to bio-hazards including food industry vessels [16], contaminated equipment in locations vulnerable to acts of warfare [17], and medical devices [18]. For these purposes, researchers have created various CAP devices including plasma jets, dielectric barrier discharges (DBD), glow discharges, corona discharges, using various sources including sinusoidal kHz AC [19], DC [20], pulsed DC [21], radio frequency (RF) [13], and microwave frequency (MW) [22] to better evaluate the inactivation efficacy and mechanism [23]. The process of inactivation heavily depends on a wide range of parameters including the types of plasma sources, properties of surfaces and environments, distances between the plasma and the treated surface, operational settings, gas composition and flow rate, treatment time, and the type of microorganism. As a result, it is challenging to comprehensively compare the efficiency of all CAP cleaning techniques.

Dielectric barrier discharge (DBD), also known as barrier discharge or silent discharge, constitutes distinct form of AC discharge. It generates a robust non-equilibrium plasma at atmospheric pressure and moderate gas temperatures. This type of discharge is usually achieved through an arrangement comprising two elec-

This work did not involve human subjects or animals in its research.

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trodes, with atleast one of two of them being covered by a dielectric layer strategically placed along their current path between the metallic electrodes. The incorporation of insulating layers on or between the powered electrodes gives the unique method for establishing a non-equilibrium atmospheric pressure discharge. DBD relies on capacitive coupling, necessitating time-varying voltages to drive its operation [24].

Some of the major distinctions between DBD and other known classical discharges are; (i) separation of electrodes and discharge through a dielectric material which effectively prevents electrode corrosion and etching, (ii) DBD's inability to function with DC voltages due to capacitive coupling of the dielectric, thereby demanding alternating voltages to generate a displacement current, (iii) DBD maintenance without the need for gas feed and large surface area of exposure than plasma jets [25], (iv) manifestation of different discharge modes including homogeneous glow and filamentary glow discharges, and (v) distinctive combination of quasi-continuous and non-equilibrium unique behavior exhibited by DBD which has sparked broad interest in its applications and investigations [24]. Many DBD configurations are subjected to AC voltages or pulsed power ranging from 1 to 100 kV, with frequencies spanning through line frequency to several megahertz.

The active species from DBD-CAP that interact with either liquids or air can produce reactive species [26] include reactive oxygen (ROS) and nitrogen (RNS) species. Reactive oxygen and nitrogen species (RONS), such as $^{\bullet}$OH, $^{\bullet}$NO, $^{\bullet}$NO$_2$, $^{\bullet}$O$_2^-$, HNO$_3$, H$_2$O$_2$, and $\mathrm{N}_x\mathrm{O}_y$ has shown to play more important role in chemical reactions and physical modifications on solid surfaces, liquids, and soft matters (including cells and tissues) [27], [28]. For example, Yan et al. [29] investigated if atmospheric plasma can enhance microbial inactivation, wound healing, and cancer treatment of bone implant materials through the release of RONS. Further, to establish a baseline for clinical applications specifically in dental implants, these authors treated titanium with atmospheric low-temperature plasma. Their results show that treated titanium increased the capability of cells stickiness and growth on the surface of the metal.

Although, the precise mechanisms involved in actual inactivation pathways of cold plasma remain unclear [30]–[32], there is a growing consensus that the mechanism depends on the type of gases that are employed to create the plasma. It has also been shown that reactive chemical species, UV light, and electric fields all contribute to the antimicrobial effects of the low-temperature plasmas [1]–[3], [33], [34]. Depending on the techniques used in the plasma formation and whether the sample is subjected to direct or indirect plasma treatment, reactive chemical species may not always be the main inactivation factor to eliminate the target microorganism [35]. However, it is noted that the majority of investigations on the inactivation processes of plasma used bacterial spores. To fully understand the mechanisms driving the inactivation of different bacteria utilizing plasma, and the factors responsible for the decontamination, more research is needed [35].

The motivation for this review article is to offer comprehensive insight into past studies that utilized DBD for different applications related to the sterilization and decontamination of materials. The literature review focuses primarily on the use of dielectric barrier discharge due to its homogeneous, electrical insulation, and energy efficient nature used as a direct plasma source to sterilize material surfaces and perform microbial decontamination treatments. Atmospheric plasma from DBD has frequently been shown to possess high activity against microbes including multidrug resistant bacteria and fungi, on non-living surfaces, and in decontamination and sterilization of biofilms [36]–[41]. For instance, bacteria [42], [43], virus [44], [45], fungi [46], [47], and other pathogens [15] have been treated using atmospheric pressure plasma sources and a combination of inactivation agents i.e., reactive oxygen and nitrogen species, electric field, ultraviolet radiations e.t.c generated when in operation from which the results showed possible complete inactivation.

This review paper is organized as follows. Following the introduction, Section II describes the existing types of barrier discharges in a combination of quasi-continuous and non-equilibrium characteristics to generate atmospheric pressure discharge. Section III discusses various designs, configurations, and types of the DBD system and their working principles. Section III also describes previous studies that demonstrated successful sterilization of various materials and the decontamination of surface microbes using DBD technology. Section III-B reviews treated materials and biofilms, and discusses possibilities of disinfecting biotic as well as abiotic surfaces. Section V presents the technologies that are currently employed to examine surface morphology and to confirm the viability of microorganisms after treatment. Section VI describes the potential of cold plasma technologies in a range of emerging fields and/or applications. Finally, a summary and concluding remarks are given in Section VI.

II. TYPES OF BARRIER DISCHARGES AND AFFILIATED DBD-CAP MICROBIAL INACTIVATION AGENTS

Typically, the DBD can manifest in two primary discharge modes: the filamentary mode, which represents the prevalent form characterized by numerous micro discharges distributed randomly across the surface of elec-

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trode; or the homogeneous glow discharge, also referred to as the atmospheric pressure glow discharge mode due to its resemblance to DC glow discharge [48], [49]. A few types of DBD configuration currently used and the major DBD-CAP agents which may be responsible for the microbial sterilization and decontamination processes on surfaces are briefly discussed in this section.

A. *Types of Dielectric Barrier Discharge*

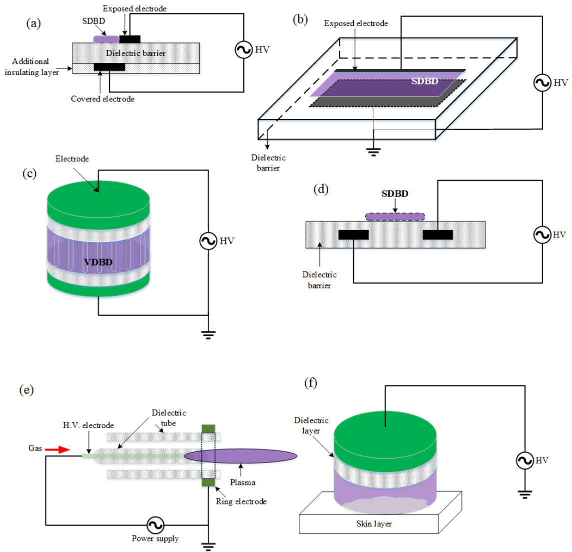

Fig. 1
Some major types of barrier discharges. (a) Cross-sectional view of a linear SDBD arrangement. (b) Linear SDBD arrangement having CAP along the border of the exposed electrode. (c) Dielectric layers covering both electrodes in a VDBD setup. (d) SDBD configuration with both electrodes embedded within the dielectric. (e) Typical plasma jet configuration. (f) Floating-electrode DBD (FE-DBD) with skin tissue as dielectric layer.

1) Surface DBD (SDBD): Surface discharge devices consist of a slender and long electrode positioned on a dielectric surface, accompanied by an elongated counter-electrode on the opposite side of the dielectric. Usually with this arrangement, a distinct discharge is not well-defined, allowing for propagation of discharge along the surface of the dielectric [50]. There is usually no gap between the electrodes and the dielectric barrier. Consequently, the discharges assume an almost two-dimensional distribution across the surface of the barrier and along the exposed edge of the electrode as illustrated in Figure 1(b). Also, the lower electrode is grounded and concealed beneath an additional layer of dielectric material to prevent any potential plasma formation on the side of the SDBD generator. This is clearly depicted in a cross-sectional view in Figure 1(a). Alternatively, another SDBD reactor, shown in Figure 1(d), involves placing both electrodes side by side and covering them with a layer of dielectric material [51]. All of these setup results in the generation of cold plasma over the barrier without direct contact between the electrodes and the surrounding gas.

2) Volume DBD (VDBD): In VDBD, microdischarges are initiated within a three-dimensional region defined by the gap between the electrode, as depicted in Figure 1(c). For this case, either one or both electrodes can be coated with a dielectric material. Volume discharge can basically exhibit the planar or coaxial configurations. For the planar electrode setups, both of the electrodes run parallel, and either one or two dielectric barriers are invariably positioned either on the ground or ground electrode, on both electrodes, or between the two metal electrodes. On the other hand, the coaxial arrangements are also possible for the DBD, it involves one electrode encased within another, followed by atleast one or two dielectric barriers that are positioned either: (i) on the outer surface of the inner electrode or the inner surface of the outer electrode, (ii) on both electrodes facing each other, or (iii) in the gap between the two cylindrical electrodes [24].

3) Co-planar DBD: The co-planar discharge is simply the combination of both volume and surface discharge configurations as seen in the Figure 2 employed in plasma display panels. Co-planar discharge device features sets of long electrodes with opposite polarities embedded in the surface proximity around dielectric bulk [52], [53].

4) Floating Electrode DBD (FE-DBD): In the dielectric isolator FE-DBD system, the process involves bringing the electrode energy close to the surface to be treated at a distance known as the discharge gap, which is less than 3 mm [54]. FE-DBD system was also employed with flexible electrodes to establish the plasma's characteristics managed by adjusting both the dielectric properties and the voltage applied to the flexible powered electrode [25]. As the name suggests, the ground or buried electrode is removed as seen in Figure 1(f), and the high-voltage electrode is left unconnected , or "floating". Under the circumstances, the applied voltage alone isn't potent enough to induce breakdown. So, when an high dielectric constant object, such as skin tissue goes closely to the electrode, it gives rise to a substantial capacitance where

majority of the voltage are concentrated. This, in turn, generates a robust electric field within the gap that may result in breakdown and formation of atmospheric plasma discharge [55].

5) Micro-hollow DBD: Rather than employing the conventional parallel plate configuration of DBD, an integrated have been made featuring coaxial micro-hollow discharges. This approach involves stacking two metals meshes coated with ceramic material, allowing for scalability while maintaining uniform discharge across the electrode area [56]. An additional advantage of this arrangement is its capacity to facilitate gas flow perpendicular to the electrode plane. This configuration enables the effective flow of active product species from the discharge plasma to the surfaces being processed in close proximity to to the electrode surface.

6) DBD plasma jets (DBD jet): The DBD jet amalgamates dielectric barrier discharge (DBD) and atmospheric pressure plasma jet (APPJ) techniques, ensuring a consistent and homogeneous plasma treatment process [57]. It has a specific type of electrode that requires the introduction of an extra gas as shown in Figure 1(e). This gas is subsequently ejected as a low-temperature plasma jet into ambient air. Since the temperature involved remain below 40 °C, making it easier to come into contact with delicate substances, including tissues, without inducing thermal harm [58]. The discharge from DBD jet was initiated by a sinusoidal high voltage in the kHz range and sustained using helium gas directed into a dielectric capillary tube, with one end exposed to the surrounding atmospheric air. In essence, this phenomenon involves the advancement of streamers within a channel created by both a dielectric medium and a noble gas permeating the ambient air. The presence of nobles gas aids ionization due to higher ionization coefficient in comparison to air at the same reduced electric field conditions [59].

B. DBD-CAP Agents responsible for the Mechanism of Microbial Inactivation

The role of CAP agents for the microorganism inactivation process has been the subject of several studies in recent years [3], [6], [7], [60]. An essential question is how DBD-CAP eliminates microorganisms and the role of CAP species in the process. Further, Dobrynin et al. [61] concluded that the interaction of living entities with plasma involves both positive and negative plasma ions. Consequently, the antibacterial effectiveness is believed to be caused collectively, by these active species [62].

Here, we discuss the contributions of five primary inactivation components from dielectric barrier cold atmospheric plasma below:

1) Reactive oxygen and nitrogen species (RONS): RONS are a class of highly reactive molecules that consist of both radical species and non-radical oxidants containing oxygen and nitrogen. These species can arise as either primary products or metabolic byproducts [4]. It is also suggested that the hydroxyl-radical, $^{\bullet}OH$, damages the outer structure of bacterial cells via oxidation process [63]–[66]. The lipid oxidation process involves lipid peroxidation, DNA damage, protein modification, and eventually may lead to cell death [32], [67]. Ozone (O_3) also has same potent bactericidal effect as it interferes with cellular respiration [68].
2) Charged particles (electrons and ions): The charged particle may also be capable of contributing to bacteria inactivation and $^{\bullet}O_2{}^{-}$ may be able to play an important role in the inactivation mechanism as produced by a direct CAP [69]. Therefore, the electrostatic stress in charged particles of cold plasma which refers to the force or pressure experienced by the particles due to the electric fields present in the in CAP discharge is believed to aid microbial inactivation [23]. The bombardment of bacterial cells by charged particles for eradication of microorganisms is expected to be limited under specific conditions [23], [70]. However, the accumulation of these particles may damage the cell as a result of electrostatic stress [71], [72]. Due to their minor contribution in inactivation process, the vast majority of the studies conducted in this field have omitted to look into whether or not electrons and ions play any part in the process of bacterium inactivation [68].
3) Ultraviolet Radiation: Cold plasma produces UV radiation over a wide range of wavelengths, from 100 nm up to 280 nm (vacuum UV), depending on the working gas. Atmospheric pressures prevent the propagation of those wavelengths below 200 nm, even though they generate energetic photons [73], [74]. Many types of UV lights have been used to produce CAP species to disinfect the surfaces of certain equipment and devices. Such UV sources include low-pressure mercury lamps, medium-pressure mercury lamps, and excimer lamps. Several milliwatt-seconds of UV light per square centimeter at wavelengths between 220 and 280 nm (germicidal range) is needed to kill cells [3], [75]–[79]. UV photons also have a crucial role in the production of reactive oxygen species (ROS), which can lead to lipid peroxidation and harm the cell membrane [71]. In most

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cases, target samples for sterilization are situated in a low-pressure vessel, utilizing argon or a N_2/O_2 gas combination, when a wavelength range of this kind is to be employed. Similarly, low-pressure plasmas also can generate UV dosages that are sufficient to render bacterial cells inactive [80]. The power density of UV radiation is less than 50 μW/cm^2 in DBD-CAP, indicating a minor or no significant role in the inactivation mechanism [3], because most of the UV radiation generated gets reabsorbed in cold atmospheric plasma [81].

4) Electric field: A plasma discharge is generated between the electrodes when an electric field of adequate strength to maintain electron-ion pairs in the gas is establish. The ignition of this plasma discharge near the electric field prompt charge particles to accelerate towards their respective electrode. Both the intensity of the electric field and resultant forces produced, can be adjusted by modifying the spatial orientation of the electrodes [82]. A lot of studies have investigated the impact of agents such as reactive species, charged particles, and UV radiation on the antimicrobial effectiveness of DBD-CAP. However, the influence of electric field has relatively been less explored and studied. Furthermore, the charged particles and the electric field could have substantial impact on the DBD-CAP sterilization mechanism. The mechanism responsible for sterilizing and decontaminating surfaces through DBD-CAP is quite intricate, but often regarded as a synergistic outcome involving reactive species, charged particles, and UV radiation [23].
5) Heat: Heat does not play a major role in the inactivation of bacterial cells since non-equilibrium plasma do not generate high temperatures [3]. Temperature studies in various cold plasma sources revealed that the biological samples exposed to the plasma did not significantly raise their temperature [58]. Laroussi and Leipold [3] observed the rotational band structure of the 0-0 transition of the second positive system of nitrogen with stimulative spectra at various temperature. They calculated the gas temperature in their plasma discharge (DBD in air) and also measured the temperature in a sample that was placed 2 cm distant from the discharge using a thermocouple probe. They concluded that thermal impacts do not have a significant contribution because their observations revealed a negligible temperature increase.

III. METHODS TO GENERATE ATMOSPHERIC PLASMA IN DBD

A. *Designs and Operation of DBD Cold Atmospheric Plasma Systems*

The use of DBD treatment generates uniform surface modification [83]. Figure 2 shows different existing configurations of the dielectric barrier discharge which has been used by various researchers classified into planar, cylindrical, co-planar, and surface discharges. Importantly, the effectiveness of DBD plasmas is influenced by the design and operational parameters of the plasma sources [84].

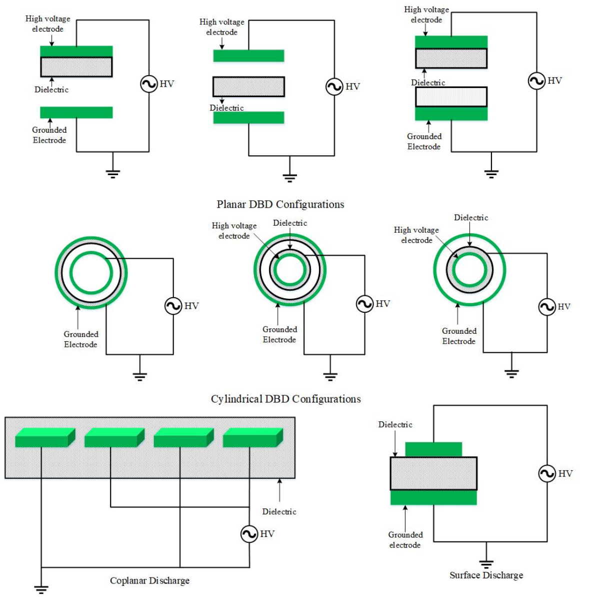

Fig. 2
Basic electrode arrangements of DBD configurations.

In DBD, a secure configuration is made for producing CAP with at least one of two electrodes covered with a dielectric substance including glass, ceramics, silicon, quartz, and polymers because insulating materials can prevent plasma transfer into an arc discharge or spark [85], [86]. One of the electrodes is usually connected to the ground, while the other electrode is supplied with high voltage. The discharge current and arc generated are consequently restricted by the dielectric material around the high voltage electrode [87]. The gap distance between the two electrodes can range from millimeters to several centimeters apart [88], depending on the operating voltage, fed gas, and configuration adopted [7]. When the DBD is operated at atmospheric pressure and surrounded by air, it generates an electrical discharge between two electrodes. Figure 3 (a) demonstrates the schematic design of a DBD setup where Teflon and

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quartz materials are used as the cover dielectric materials. And Figure 3 (b) shows an actual experiment replicating the setup.

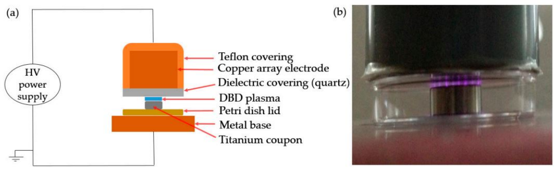

Fig. 3

Typical DBD plasma setup is shown in a schematic design and picture. (**a**) demonstrate the schematic diagram of the normal DBD plasma and (**b**) Shows the actual experimental setup for DBD. Courtesy of Tripti Thapa Gupta. Used with permission [89]. Copyright 2018 IEEE.

B. Utilization of DBD for Materials Sterilization and Microbial Decontamination

This review is centered on bacterial biofilms which have complex structures composed of bacterial cells closeup in a self-produced extra-cellular matrix. This matrix are known to contain proteins, polysaccharides, and DNA, giving protection and structural support. Table III-B below summarizes some CAP bacterial biofilm treatment outcomes specifically using various barrier discharge for surface sterilization and microbial decontamination since they are found on various surfaces, such as medical devices (implants, catheters), teeth, and natural environments. Numerous studies have been conducted using the DBD's direct application of plasma inactivation of a wide range of gram-negative and gram-positive bacteria. Inactivation also occurs to bacterial biofilms, spores, and viruses [35], [89]–[95].

The inactivation effect is usually represented by decimal logarithm of the ratio between the number of viable cells observed in suspension before and after a treatment using DBD CAP [96]. For instance, say an initial bacteria of 10^3 were present before applying the DBD cold atmospheric plasma, upon a decrease of 2 $\log_{10}$ after treatment, the final bacteria count would be 10^1 representing a reduction up to 100 times.

Venezia et al. [104] used an afterglow PlasmaSol device (PlasmaSol Corporation) to sterilize bacteria. This device has a sterilizing container with an embedded plasma-generating electrode powered around 30±1 W. A combination of 1% ethylene, 50% oxygen, and 49% nitrogen at a flow rate of 1 L/min was utilized as the reagent gas. The study demonstrated that a 2 minutes exposure duration of *B. atrophaeus* to plasma achieved a decrease of 5 $\log_{10}$. In an either dry or moist environment, the number of bacteria decreased by roughly 5 $\log_{10}$ after just 10 minutes of treatment.

Eto et al. [105] used a setup to decontaminate samples kept in Tyvek wrapping (approximately 40 × 40 mm). To accommodate any form of Tyvek packaging, they employed an electrode arrangement that adopted a flexible sheet-like structure and this device was used to sterilize the wrapped samples. The DBD was placed close to the Tyvek package by a stainless steel electrode grid at 2.5 kV and 5 kHz. Stainless steel bowl-shaped discs were contaminated and placed inside the Tyvek packing facing the discharge. An initial population of 3×10^6 *Geobacillus stearothermophilus* spores were utilized as test organisms for various combinations of process gases (oxygen and nitrogen). The findings indicated that 15 minutes of exposure can result in complete inactivation of all bacteria, where 50% N_2 and 50% O_2 produced the most effective inactivation. The discharge was thought to be related to the production of ozone, which has concentration of 80 ppm and the reactive species that are produced were seen to be the most efficient for the inactivation.

Hahnel et al. [106] tested the distance effect on inactivation of microorganisms, using a surface dielectric barrier discharge SDBD in ambient air as shown in Figure 4(a). A unique electrode geometry was created while the mean gas temperature of the discharge was held under 300 K using 10 kV sinusoidal ignition voltage, 2 kHz frequency in pulsed mode with 1 Hz, and a maximum of 500 ms plasma on-time. *B. atrophaeus* spores was used at relative humidity to investigate the microbial killing. Their results suggested significant dependence on humidity, having an actual decrease factor of 5 $\log_{10}$ after a 5 minute plasma treatment with a 300 ms on-time per second.

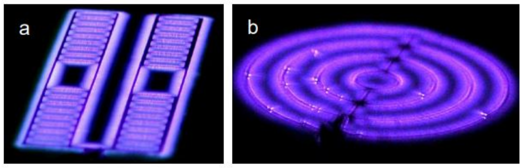

Fig. 4

Circuit board materials were used to make a variety of surface discharge electrodes as test strip. Courtesy of Marcel Hähnel and Katrin Oehmigen. Used with permissions (**a**) [106] (**b**) [107]. Copyright 2010 John Wiley and Sons.

Oehmigen et al. [107] also studied the effects of direct plasma treatment using the surface dielectric barrier discharge shown in Figure 4(b), examined in a solution where a surface DBD of a similar design (as in 4(a)) was made to fit into a petri dish having a 60 mm diame-

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Table III-B: Related CAP Plasma Treatments using Dielectric Barrier Discharge for Materials Sterilization and Microbial Decontamination

Control Strain	Type of DBD-CAP and Parameter (Freq., Voltages, Operating Power, Applied gas, and Rate of Flow)	Inactivation rate	Surface for Biofilm Formation, and Time for Sterilization or Decontamination	Ref.
Staphylococcus aureus	FE-DBD: 0.13 W/cm^2 power and 120 V	All biofilms were sterilized	Coverslip and 96 well plate for $<$ 2min	[97]
Pseudomonas aeruginosa	DBD: 50 Hz and 120 kV	Biofilm reduced to undetectable level	96 well plate and Cover slips for 5 min	[98]
Streptococcus mutans and saliva multispecies	Volume DBD (VDBD): 40 kHz, 10 kV and Ar at 0.05 slm and Hollow DBD (HDBD):37.6 kHz, 8.4 kV and Ar & Ar$_{+O2}$ at 1 and 0.01 slm	5.38 log for S. mutans and 5.67 log for saliva surface biofilm	Titanium disc for 10 min	[99]
Pseudomonas aeruginosa and Staphylococcus epidermidis	Surface DBD (SBD): Structured electrode planar SDBD-A: (13 kV and 20 kHz) and a wire electrode SBD-B: (8 kV and 30 kHz with compressed air at 0.5 slm)	Reduction of 7.1 and 3.8 log using SBD-A and SBD-B in P. aeruginosa and reduction of 3.4 and 2.7 log using SBD-A and SBD-B in S. epidermidis	Polycarbonate disc plate for 10 min	[37]
Escherichia coli and Staphylococcus aureus	DBD: 31 kV peak-to-peak, 1.5 kHz, 10 μ s pulse, and power density of 0.29 W/cm^2	Inactivation up to $>$95% of biofilms	Discs of implant materials for 3 min	[95]
Candida albicans	VDBD: 40 kHz, 10 kV and 16 W power, and HDBD: 37.6 kHz, 9 kV and 9W power, Ar at 6 slm and Ar+(1%)O$_2$ at 0.06 slm	Reduction of 5 log	Titanium disc for 10 min	[66]
Stereptococcus mutans	DBD: 2 W/cm^3 power density, 580 kHz and He at 2L/min	98% eliminated	Tooth slices for 30 s	[100]
Escherichia coli and Grampositive Staphylococcus aureus	DBD: 50 Hz, 15 kV, 6 mm discharge gap, O$_2$ gas at 2 slpm	Relatively faster biphasic nanofiber treatment	Bombyx mori silk fibroin (BMSF) for 10–60 s periods	[101]
Escherichia coli, Cronobacter sakazakii, and Staphylococcus aureus	Underwater DBD 18 kV, 20 kHz, dry air at a rate of 5 L/min	$\sim$ 90% bacteria inactivation at 5.50 log, 6.88 log, and 4.20 log CFU- coupon for E. coli, C. sakazakii, and S. aureus respectively	Stainless steel coupons for 90 min	[102]
Staphylococcus aureus and Escherichia coli	Air-based DBD: 7 kV, 22 kHz, and chamber volume of around 6200 cm^3 with diameter of 8 cm and thickness of 2 cm. Electrode distance was set at 4 mm	Biofilms disruption up to 70% and 85% for S. aureus and E. coli respectively	Biofilm in 6- well plates for 4 min	[90]
Bacillus subtilis	Open-air DBD: micro-discharge current pulses from 7 $\times$ 10^5 to 1.5 $\times$ 10^6 for 9.6-10.8 kV & power density from 10-20 W/cm$^-$3	Log reduction of 2.7 in (5.2 4.3) $\times$ 10^3 CFU ml^{-1} no of bacteria	Polymer foils treated for 180 s	[94]
Bacillus subtilis	Low-pressure (LPDBD): low AC HV below 10kV, 6 and 10mm gap distance, 28% and 25% at 50 and 30 kPa max. discharge rates	No surviving cells were observed at LPDBD 6mm gap	Glass beads surface for 4 min	[103]

ter. *Escherichia coli* and *Staphylococcus aureus* ranging from (10^6 –10^8 cfu/mL) were thoroughly inactivated within 15 minutes, depending on the volume of the liquid sample. Their measurements of pH, nitrate, and nitrite concentrations after DBD plasma or NO gas treatment showed that the primary cause of liquid acidification was the formation of nitric acid due to reactive nitrogen species generated by the plasma.

Leipold et al. [9] attempted to disinfect some spinning cutting tools used for slicing purposes in meat industries. The configuration of their DBD is shown in Figure 5. The cutting tool is a 450 mm diameter disc with shallow cones that rotates at a period of 3.6 seconds per cycle. Two DBD were employed to the opposite sides of the cutter, which serves as the grounded electrode. One 100 × 100 mm electrode size of DBD is placed at a 2 mm to 4 mm distance to the surface of the circular knife. An initial concentration of 5×10^7 CFU/ml of *L. innocua* (a biological indicator for *L. monocytogenes*) in 240 μl was sprayed directly onto the knife. Inactivation increased by roughly 4-5 $\log_{10}$ between 68-170 s and 360 W; compared with the second DBD having an applied power of 1.8 W yielding a decrease of 3.5 $\log_{10}$ for a 300 s procedure but 37 s exposure time.

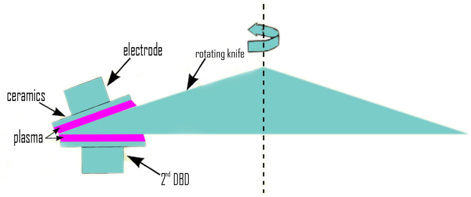

Fig. 5
Experimental setup sketch for plasma surface cleaning of cutting knives used for slicing in meat business. Courtesy of Frank Leipold. Used with permission [9]. Copyright 2010 Elsevier.

Schwabedissen et al. [108] used a different arrangement to disinfect items in a closed packaging as shown in Figure 6. They also employed a similar surface DBD where two powered electrodes operating in the kHz range are attached to the outside of the enclosed package, while a third electrode is connected to the outer electrodes located on the inside wall. A discharge between the inner electrode and the outer powered electrode produced approximately 2-4 W of discharge power at a peak-to-peak voltage of around 8 kV. Ozone, a disinfectant, was produced inside the enclosed packaging when used with ambient air. A paper strip containing *B. subtilis* was placed within a polystyrol petri dish to assess the microbicidal effectiveness of the PlasmaLabel discharge. A 4 $\log_{10}$ decrease after 10 minutes of ozone exposure was obtained.

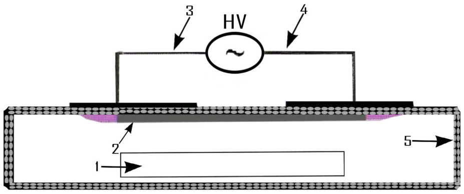

Fig. 6
A schematic design of the PlasmaLabel idea showing the arrangement in a closed packaging. (1) an inner electrode, (4) two powered electrodes outer the package (2 and 3), and the sealed package (5). Courtesy of Axel Schwabedissen. Used with permission [108]. Copyright 2007 John Wiley and Sons.

Ercan et al. [109] also employed floating-electrode (FE-DBD) and reported that contrary to prior speculation, non-thermal plasma treated fluids retain their antimicrobial properties for a longer time (three months by delay time and two years by solution aging). A wide variety of multidrug-resistant bacteria and fungi pathogens, in both their planktonic and biofilm forms, were all inactivated by this fluid-mediated and plasma-based technology, which was quicker than the previously reported 15 min holding time (the time at which the plasma-treated liquid comes in contact with the suspended bacteria). The antimicrobial impact is attributed not only to change in pH, presence of H_2O_2 or nitric acid, but also to the likely formation of supplementary species or products that are accountable for the potent biocidal outcome. Therefore, thorough investigations involving chemical analysis are necessary to validate the existence of these substances or their products.

In terms of power and frequency dependent effects for the antimicrobial inactivation, decrease of 3–7 $\log_{10}$ was observed with DBD plasma ranging 8-13 kV and 20-30 kHz [37], while a reduction of 5 $\log_{10}$ was obtained using a 8–10 kV and 37–40 kHz plasma source [66].

Collectively, CAP's effectiveness to eliminate bacteria is influenced by a number of factors including, 1) its method of production, exposure time, voltage, and frequency, 2) the properties of the target bacteria such as species type, growth stage, 3) the presence of a protective biofilm, and 4) some environmental conditions, such as, the composition of the surrounding material, humidity, and acidity. The amount of bacteria initially present also can be a factor that influences the efficacy of plasma treatment [32].

IV. MATERIALS AND SURFACES TREATED BY DBD-CAP

One of the biological application of low-temperature atmospheric pressure plasmas was to eliminate of microorganisms on material surfaces. Due to it's ability to quickly and safely sterilize a variety of materials including glass, polymers, and human tissue, cold atmospheric plasma are the subject of ongoing studies for cleansing and sterilization. [7], [18], [110]. From the standpoint of contamination control, the in-package generation of plasma reactive species presents a distinct advantage by effectively reducing post-processing contamination and the occurrence of cross-contamination incidents [111].

There have been a need of novel technologies to properly sterilize heat-sensitive medical instruments [112]–[114], and to disinfect wounds. This led to enthusiasm in developing low-temperature plasma to sterilize, decontaminate, or disinfect both biotic and abiotic surfaces [10], [115]–[121]. CAP may be able to offer a safe and efficient approach to address the challenges associated with peri-implantitis treatment. CAP has proven to be efficient in the elimination of bacterial biofilms [14], [37], [39]–[41], [43], [66], [90], [95], [97]–[99], [122], [123].

Recent literature demonstrates that CAP can have positive effects on the skin. This implies that CAP technology has potential in the development of cosmetic treatments that can help prevent aging [124], and may also be effective therapy of various skin diseases [75], [125], [126]. Additionally, CAP has demonstrated anti-tumor effects, enhance stem cells, and also signals potential in enhancing immunotherapy for cancer patients [127]–[135].

V. METHODS TO ASSESS EFFECTIVENESS OF DBD-CAP AFTER TREATMENT

Since the emergence of DBD cold atmospheric plasma, various investigations has been conducted to adequately analyze and interpret the surface modifications of treated materials [88], some of which have been briefly discussed here.

A study showed no difference in the surface roughness as determined by a surface profilometer gauge after Ti discs has been treated with CAP and then subjected to different storage conditions. Contact angle and surface roughness measurements were taken at the end various of time intervals. Optical Emission Spectra (OES) analysis showed the presence of OH and NO as plasma-generated species. The contact angle measurements showed that the samples stored in saline retained their hydrophilicity for a longer duration compared to those stored in room conditions and an inert ambient, while, surface roughness measurements using a profilometer indicated no significant difference compared to the control group (untreated Ti) [136].

Proliferation tests are used to measure the plasma-induced microbicidal effectiveness. There exist many viability assays such as BacTiter-Glo and LIVE/DEADBacLight [7]. These tests entail labeling several bacterial compounds with fluorescent substances, which enables the fluorescence signals to distinguish between healthy and non-living bacteria [137].

Plasma diagnostics also play a crucial role in understanding CAP application on material surfaces. These diagnostic techniques entail gathering essential information about the plasma's properties and behavior, characterization, and optimization of plasma parameters. Typically, Oscilloscopes are employed to conduct the electrical measurements, while the UV-VIS-NIR spectroscopy ranging from 190–2200 nm may be used with a fiber optic spectrometer for their optical properties [129]. Additionally, electron microscopy can provide evidence of ruptured membranes or other surface morphological abnormalities [138] or using an atomic microscopy [106], [139], [140].

VI. OTHER EMERGING APPLICATIONS OF DBD-CAP TECHNOLOGY

Floating electrode dielectric barrier discharge (FE-DBD), a DBD-based plasma generator have also been utilized in modern plasma medicine [141]. Viruses like SARS-CoV-2, which just produced the COVID-19 pandemic, has shown potential of being eliminated using DBD low-temperature plasma [142]–[144].

It is important to recognize that CAP, like that produced by DBDs, can also be a crucial tool to disinfect space probes looking for signs of life on distant planets and moons. Additionally, DBD-CAP may be employed in manned deep space, lengthy trips like in the expedition to Mars, to sanitize and disinfect astronauts' equipment like hand, head, and body wears that may come into contact with alien germs during such lengthy voyages [88].

Sterilization of food packaging is essential in keeping foods safe for consumption. Cold atmospheric plasma generated using DBD may be used as a final decontamination step to remove pesticides from fresh food products and their packaging [14], [16], [46], [143], [145]–[150].

VII. Conclusion and Future Perspective

Cold atmospheric plasma is anticipated to be used as an antimicrobial device in the future. DBD-CAP is highly effective in inactivating multidrug-resistant pathogens and bacterium on biofilms. The inactivation outcomes vary among different types of CAP devices, working gases, treated materials etc. Thus, it is crucial to understand various parameters to effectively design and optimize a CAP device. The variety of operational gas and power input significantly affects effectiveness of CAP treatment, and the inactivation of free radicals and oxidants largely depend on high-voltage electric fields and frequency.

We have collated literature pertaining to the recent advancement in cold atmospheric plasma DBD for sterilization and decontamination of biological and non-biological materials. Reactive nitrogen species, reactive oxygen species, electric field, and charged particles play a major role in the inactivation mechanism of DBD-CAPs, while UV radiation and heat plays a minor or no significant role, making the inactivation mechanisms quite complex. To generally validate DBD-CAP as antimicrobial tool, there is a growing need for uniformity in terms of characterization, optimization, and methodology which will be used as a baseline for future studies in this field.

The reviewed literature have amply demonstrated and proven that CAP, such as the one produced from a dielectric barrier discharge, is a promising technology in the treatment of material surfaces and in other emerging areas already discussed. Although, other CAP devices such as Radio Frequency [13], Corona Discharges [151], and CAP Jets [152] have also been tested to inactivate microbial growth over medical devices, hospital instrument surfaces, and other materials.

Collectively, DBD-CAP has the potential to become a highly effective antimicrobial technology for surface treatment, lessening the burden in the food, health and other sectors, and is expected to be used as antimicrobial therapy.

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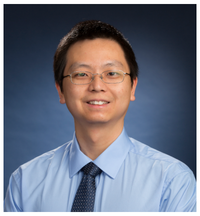

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