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Review

Design Considerations for Low-Temperature Plasma Production in Air Using Pulsed Dielectric Barrier Discharges: A Review

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Abstract

Low-temperature atmospheric plasma (LTP) is widely used in industrial processes, such as disinfection, surface modification and wastewater treatment. The dielectric barrier discharge (DBD) is regarded as one of the most robust and reliable methods for generating LTP in ambient air. Compared to conventional AC excitation, pulsed powering offers several advantages (i.e., lower energy use and heat production). The present trend is to use short and fast pulses (in the nano- and picosecond range). In this review, the key design parameters of a DBD (barrier thickness, relative permittivity and gap distance) are discussed. Material-specific phenomena like surface charging and degradation are analyzed. The complex interactions between the pulse source and DBD are examined. By mapping the interdependencies, this review aims to support the rational design and optimization of pulsed DBD systems, and to facilitate their broader industrial use.

Keywords: low-temperature; cold atmospheric plasma; pulsed excitation; dielectric barrier discharge; design optimization; material degradation

1. Introduction

Low-temperature atmospheric plasma (LTP) has a long history in both laboratory and industrial environments. As early as 1785, van Marum reported an ‘odor of electricity’, later identified as ozone produced by his spark discharge setup. In 1857, von Siemens constructed the first dielectric barrier discharge (DBD) ozoniser using a coaxial design [1].

Since then, numerous LTP applications have been established. In air, LTP enables inactivation of pathogenic aerosols (viruses, bacteria) [2,3], odor removal [4], volatile organic compound (VOC) breakdown [5] and CO₂ conversion [6]. On surfaces it is applied for: seed disinfection [7,8], seed priming [9] and surface treatment [10]. In water it facilitates wastewater treatment [11], removal of PFAS [12], breakdown of pharmaceutical residues (such as antibiotics) [13] and inactivation of pathogenic bacteria [14]. LTP is highly versatile: it produces a wide range of reactive oxygen and nitrogen species (RONS), electrons, ions and photons in atmospheric air at near ambient temperature (hence the term ‘cold atmospheric plasma’).

Since the pioneering work of von Siemens, the DBD technology made a lot of progress. DBDs are considered reliable devices for generating LTP in air. Recent research aims at



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optimizing the scale, efficacy and power efficiency of DBD devices. Extensive overviews of DBD designs are given by Brandenburg et al. [15,16].

The electrical powering has also advanced considerably. The transition from corona discharge to DBDs marked a shift from DC to AC power. The AC frequencies for DBDs increased substantially. For example, a recent innovation is the resonant piezoelectric transformer (RPT) which could replace more bulky AC powering systems to produce plasma with high voltage AC powering (with a frequency of tens of kHz) [17]. In recent years, pulsed powering has emerged, as promising alternative, offering many advantages [18]. Nowadays, there is a focus on pulses with steep rise times (nano- or picosecond range) for enhancing power efficiency. Also, the pulse shape (square, bipolar, multi-stage waveforms) can be tailored for a better performance [19]. Some authors used AC powering in burst mode, through lowering of the duty cycle. This lowers DBD energy use, resulting in lower heat generation (which is discussed in Section 2.4). However, pulsed operation remains superior. Compared to (burst) AC powering, it results in lower heat generation, higher RONS formation, lower electrode damage and lower energy use [20]. Meropoulis & Aggelopoulos achieved a very high energy yield for dye degradation in water using a nanopulsed DBD [21]. Klymenko et al. were able to breakdown perfluor- and fluoroalkyl substances (PFAS) in water using nanopulsed plasma [12]. Fan et al. effectively removed pathogenic microorganisms from melon seeds using a nanopulsed DBD, without harming the seeds [22]. Sun et al. reported a two-step process involving a nanopulsed DBD to produce ammonia in water [23]. Zhang et al. used a pulsed DBD for air disinfection and found that a pulsed source produced a more intense and homogeneous plasma compared to AC powering [24].

Despite this progress, upscaling DBD systems to an industrial scale remains quite challenging. For DBD reactors to be viable, they must be reliable, durable, power efficient, effective, safe and scalable. A DBD-based LTP system can only be assessed properly when the whole system is considered, from power circuit to DBD to ambient conditions. This integrated system has many interdependent variables as shown in Figure 1. For example, the pulse source influences the DBD and vice versa. The DBD influences the ambient conditions and vice versa. The system under consideration thus evolves over time across electrical, chemical, thermal and material domains.

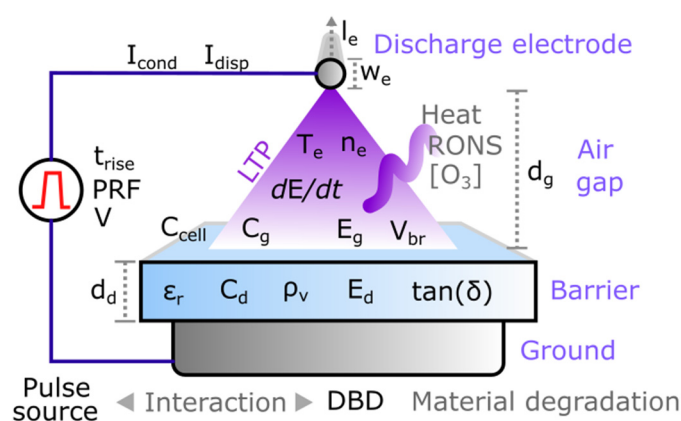


Figure 1. Schematic of a DBD. It shows the main design parameters discussed in this review. The meaning of the symbols is given in the list of abbreviations.

This review gives an overview of the design parameters for pulsed DBD operated in ambient air. It aims to facilitate the design of novel and optimized DBD systems that may advance the industrial use of LTP in ambient air. First, a short survey of streamer dynamics is presented (Section 2.1). Then, the material selection (for both the electrodes and the dielectric barrier) is discussed (Sections 2.2 and 2.3). The main mechanisms

causing DBD heat generation are summarized in Section 2.4. Next, the main design variables controlling the performance of pulsed DBDs are examined. These variables include: dielectric barrier thickness and relative permittivity, electrode shape, gap distance, dielectric loss factor and DBD capacitance (Sections 2.5–2.7). The factors influencing ozone yield are outlined in Section 2.8. Finally, Section 2.9 covers measurement challenges and the interrelation between DBD and pulse source. A literature search was performed (2022–2025) using the Semantic Scholar research tool (www.semanticscholar.org), by searching on terms such as ‘DBD optimization’, ‘DBD ozone/heat production’, ‘influence of dielectric thickness/relative permittivity’, ‘pulse rise time’. The tool allows to easily dive deeper into the topics by linking to newer studies that cited the used papers, giving an up-to-date overview. A total of >1000 papers was read during the course of the research period, and main findings per paper were systematically categorized in an overview document. From here, the most important findings were selected and summarized in this review. Also, several books on the topic of (pulsed) low-temperature plasma were consulted in the Delft University of Technology library, the Netherlands ([25–28]). The trends in this review can be applied to a wide range of DBD geometries (coaxial, wire-plate, needle-plate, micro-hollow, grating, surface and volume DBDs in Figure 2) unless otherwise specified.

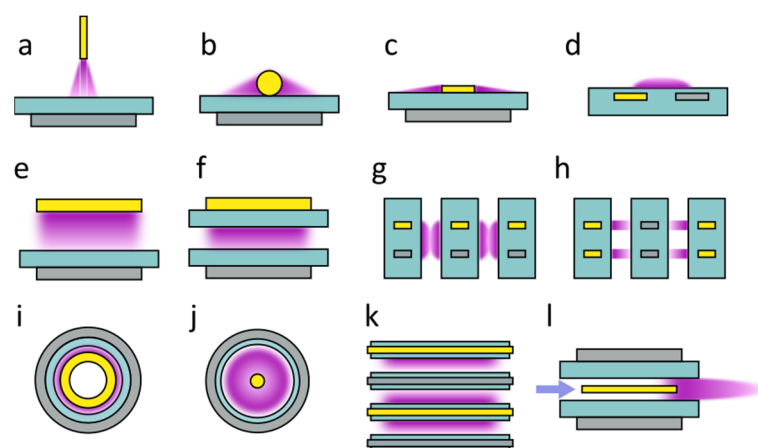


Figure 2. Schematic side view of commonly used DBD geometries for pulsed plasma production in air. Yellow: high-voltage electrode. Blue: dielectric barrier. Gray: ground electrode. Purple: plasma formation. (a): Needle–plate DBD [29]. (b): Wire–plate DBD [30]. (c): Surface DBD [31]. (d): Coplanar DBD [32]. (e,f): Volume DBDs [33]. (g,h): Multi-hollow DBDs [34]. (i,j): Coaxial DBDs [35]. (k): Grating DBD [2]. (l): Plasma jet (arrow depicts direction of air flow) [36].

2. Design Considerations for Pulsed DBDs in Air

2.1. Streamer Dynamics

A low-temperature plasma generated in atmospheric air usually is a streamer discharge (also called filamentary discharge). This discharge consists of many microdischarges (or filaments or streamers), initiated by electron avalanches. When avalanche formation was suppressed by applying a narrow gap or very short pulses (<10 ns in [37]), a diffuse discharge (or glow/homogeneous discharge [16,38]) could be formed [39]. Sustaining a diffuse discharge in atmospheric air is difficult [16,40–42], because electron avalanches develop quickly. Several seemingly diffuse discharges turned out to be streamer discharges upon closer inspection [43–45]. When the plasma intensity increases (e.g., with a higher voltage), a diffuse discharge will turn almost always into a streamer, which is a preferential ionization path [46]. It is an ‘ionization wave that propagates from a high voltage electrode toward the grounded electrode’ [28].

Positive streamers arise when a positive high voltage is applied to the discharge electrode (anode). They propagate from the anode toward the cathode (ground electrode). As negative streamers have different dynamics, this review focuses solely on positive streamers (the differences between both are discussed in [47–49]). Positive streamers are advantageous in practice due to lower breakdown voltage (V_{br}) [50], higher electron density (n_e), charge deposition (Q) [47,51] and enhanced RONS production [50]. Many studies have been carried out on DBD streamer dynamics. In this review a brief overview of the main processes associated with positive streamers will be given. For a detailed description of streamer dynamics we refer to the comprehensive review of Komuro [52]. Some important streamer observations are summarized in Table 1.

Kogelschatz et al. summarized that typical single streamers could last 1–10 ns, have a diameter of 100 nm and a length of a few mm [53]. Streamer evolution is divided in a primary streamer phase, secondary streamer phase and a post-discharge period. The primary streamer and secondary streamer are stages of the same streamer event, They differ from the primary and secondary discharge discussed in Section 2.2. In a positively pulsed DBD, the primary discharge corresponds to a positive streamer, while the secondary discharge (or back discharge) appears more diffuse [27].

A streamer head creates a very high local electric field and propagates towards the cathode. It forms when the electric field in the air gap (E_g) exceeds the dielectric breakdown strength of air (~ 3 kV/mm). Background seed electrons start the process by ionizing neutral gas molecules through inelastic collisions. Additional electrons are created due to these collisions and detachment from gas molecules (known as electron avalanche). The streamer head propagates with a certain velocity through air, while ionizing molecules in its trajectory.

In positive streamers, the streamer head propagates in opposite direction to the electron drift and is supported by photoionization (photons emitted by excited gas molecules create seed electrons in front of the streamer head) [49,54]. Its ‘slipstream’ is called the streamer channel. In this region the electric field is below the ionization threshold. Here, the electron density decreases due attachment to gas molecules (particularly in humid air) [52]. Most energy in the streamer channel is transferred to vibrational excitation of air molecules [52].

When the streamer head reaches the cathode (or the barrier on the cathode), a cathode fall (or surface streamer) develops near the surface [29,54]. The strong local electric field in the thin layer separating the streamer head from the barrier, will initiate secondary electron emission. At this stage, a conductive channel connects the anode to the cathode (the streamer head disappears). The electric field between the electrodes is redistributed. Now, a secondary streamer can develop at the anode [29,50]. The electron density in the secondary streamer strongly depends on the applied electric field [52]. After the secondary streamer phase terminates, the post discharge period begins. During this stage various processes such as recombination, relaxation and diffusion occur.

Therefore, when optimizing LTP for production of a specific RONS (such as ozone), one must consider both the discharge and the post-discharge period. In repetitive pulsed operations in air, positive and negative ions remain after the first pulse. These ions may disturb the subsequent pulses (‘pre-ionization’) [47,52,55].

Table 1. Important streamer observations in literature.

Observation	Refs.
An increase in pulse rise rate increases discharge current, streamer velocity and diameter.	[56]
A decrease in the air gap decreased the streamer diameter (volume DBD).	[57]

Table 1. Cont.

Observation	Refs.
Higher streamer velocity, diameter and current observed for positive pulses.	[49]
The electrical field magnitude and streamer diameter influence radical yield.	[52]
Streamer length is independent of d_d , decreases with increasing ϵ_r (surface DBD).	[58]
Streamer velocity increases when ϵ_r increases (needle-plate DBD).	[59]
A shorter gap length reduces the streamer duration.	[52]
The length of the secondary streamer increases linearly with applied voltage (volume DBD).	[29]
A positive streamer has a lower onset voltage and a higher velocity.	[60]
The primary streamer length and velocity decreased by increasing the PRF (surface DBD).	[61]
Primary streamer velocity and diameter increase by increasing the electric field in the gap (E_g).	[62]

2.2. Charge Trapping in and on Barriers

In a pulsed DBD the total current consists of a displacement current (I_{disp}) and a conduction current (I_{cond}) [27,63]. The displacement current originates from the charging and discharging of the DBD which behaves as a capacitor. It does not contribute to LTP generation [64], and is proportional to the DBD capacitance (C_{cell}) [1]. The DBD capacitance typically is a series combination of gap capacitance (C_g) and dielectric capacitance (C_d), as shown in the equivalent circuit in Figure 3. For voltages $V(t)$ below the breakdown voltage V_{br} both capacitances remain constant. Once breakdown takes place, C_g changes, because plasma is formed in the gap (the resistance of the gap $R_g \rightarrow 0$) [45]. Then, a conduction current appears either superimposed on the displacement current or slightly delayed [27,65,66]. It happens when the electric field in the air gap (E_g) is sufficiently strong to produce microdischarges. The conduction current results from electron avalanches in the air gap. It corresponds to the collective current of all microdischarges. A higher conduction current indicates a larger plasma volume, emerging from more streamers and/or longer streamers.

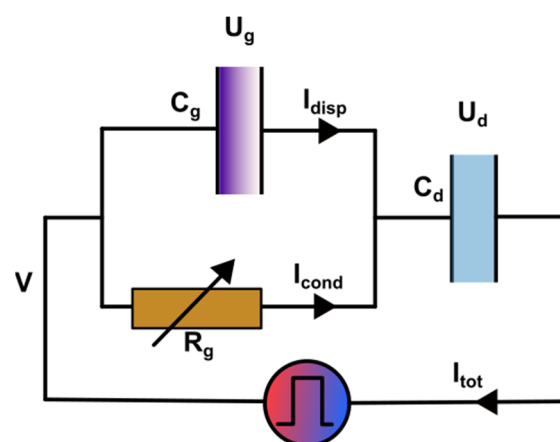


Figure 3. Equivalent circuit of a pulsed DBD. V : Applied voltage. C_g : Gap capacitance. C_d : Dielectric capacitance. I_{disp} : Displacement current. I_{cond} : Conduction current. I_{tot} : Total current. U_g : Voltage over the gap. U_d : Voltage over the barrier. R_g : Variable resistance of the gap.

During operation, the dielectric barrier accumulates surface charges. It leads to the characteristic secondary discharge in nanopulsed DBDs, where charge (Q) temporarily stored on the dielectric surface due to the primary discharge is released at the end of the voltage pulse, causing a secondary or back discharge [34,67]. In Figure 4, both discharge events are displayed. A positive pulse, with a peak voltage of 4 kV and a duration of 600 ns

produces two distinct current peaks, one during the rising edge and one during the falling edge of the pulse. Each discharge is accompanied by optical light emission detectable with a photomultiplier tube (PMT).

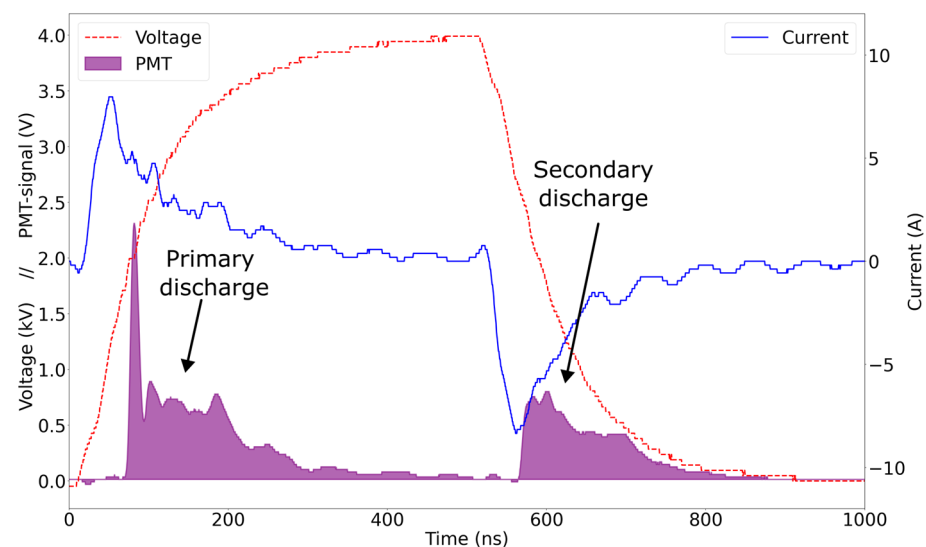


Figure 4. Primary and secondary discharge observed during a 600 ns positive pulse applied to a DBD in ambient air (experiments performed in our laboratory).

Surface charging shows up for repetitive positive, negative and alternating pulses [51,68]. It can be (partially) removed between pulses by using alternating pulses [51] or using slightly conductive/resistive barriers [69,70]. Although the plasma formation benefits from the secondary discharge, the residual surface charge between pulses suppresses the electric field of the next pulse [71]. When the PRF was increased from 0.1 to 5 kHz, the primary streamer length decreased ~15% and its velocity decreased ~30% in a surface DBD, attributed to higher the residual charge on the surface [61]. Investigations in [72] (based on [51]) identified two decay time scales for surface charge in a surface DBD: around 1 μ s, caused by volumetric charge transport around the electrode, and around 100 μ s, corresponding to 'charge transport (redistribution) over the dielectric surface'. But redistribution does not imply complete neutralization. Volumetric charge transport occurs only within a few mm of the high-voltage electrode [72].

Some materials can store charges for a long time, not only on the surface, but also within their bulk. The dielectric barrier then becomes charged, and this charge can be trapped for a long time (days, weeks) [16,47,55,73,74]. It changes the local electric field distribution and typically lowers the conduction current. Other effects were reported such as a higher number of seed electrons [55], longer streamer channels [55] or dielectric breakdown [74]. Pipa et al. also observed a change in V_{br} caused by charges persisting for >13 h [35]. Some materials behave as an electret under continued plasma exposure (PTFE, epoxy resin, silicone rubber, PP, PI, alumina) [67,73–78].

Three mechanisms are responsible for surface charge decay: bulk neutralization, gas-phase neutralization (recombination with ions in the air) and surface conduction [74]. Bulk neutralization means the neutralization of surface charges through migration of opposite charges (ions or electrons) from within the material (the 'bulk') to the surface. Gas neutralization appears to dominate the charge decay in DBDs, as surface conduction and bulk neutralization are often low for thin, insulating barriers [74]. Large amounts of ions (10^6 – $10^8/\text{cm}^3$) are present in the air gap of nanopulsed DBDs [47,55,74]. The higher the ion density in the air volume surrounding the barrier, the faster the surface charge decays [74]. Charge decay through surface conduction varies per barrier material. Silicone

rubber shows faster surface charge decay than PTFE and epoxy resin, possibly due to shallower traps and higher polymer chain mobility in silicone rubber at room temperature (associated with its low glass temperature) [74,77].

Ren et al. identified three time constants ruling the decay of surface charges, namely the ion recombination time τ_r (in air, estimated 10–100 ns [47]), the diffusion time for charges along the dielectric surface τ_d and the surface charge relaxation time τ_s (which depends on the volume resistivity ρ_v and relative permittivity ϵ_r since $\tau_s = \epsilon_0 \times \epsilon_r \times \rho_v$) [47,73]. This implies that lower ϵ_r and ρ_v promote a faster charge decay [74]. However, these parameters are constrained by the barrier material chosen (to satisfy oxidation resistance etc.). For some barriers with moderate resistivity the discharge may operate as resistive barrier discharge [69,79]. But these materials (such as silicone) are usually less resistant to electron bombardment and can break down at high voltages. Ren et al. estimated that in atmospheric air $\tau_s \gg \tau_d \gg \tau_r$ [47]. This means that the surface charge decay is mainly dictated by ϵ_r and ρ_v . Since ρ_v is a bulk property, the barrier can be thin. A low ϵ_r would accelerate surface charge decay. However, charge decay is a complicated phenomenon, which can hardly be entirely predicted by a laboratory test setup, as noted by Molinié [80].

The voltage waveform also affects charge accumulation. AC driven discharges give a net positive surface charge, due to asymmetries between positive and negative microdischarges [72]. Modelling shows that bipolar pulses can potentially increase the charge deposited per pulse by about 30%, as the surface charge enhances the electric field during the opposite polarity pulse [47]. This applies also to alternating pulses, where the charge per pulse may increase by a factor 4 [68]. Leonov et al. used Kapton tape as a barrier and found even larger differences in charge per pulse for unipolar and alternating pulses, namely a factor 5 [51].

From a practical perspective, the hardware to create bipolar/alternating high voltage nanopulses is more complex than that of unipolar pulses. For example, a push-pull circuit needs two DC sources (a positive and a negative) instead of one. Simeni Simeni et al. used a custom-made magnetic pulse compression circuit [68], first described in [81]. Successful production of alternating and bipolar high voltage pulses was achieved using a cascaded H-bridge in [82,83], a solid-state Marx generator [84], and a combination of these [85].

As surface charging can negatively impact LTP production, dielectric barrier materials must be carefully chosen. Polymers appear to be particularly vulnerable to surface charging. They also have the ability to retain charge in and on the surface, forming electrets.

2.3. Electrode and Barrier Degradation

Many materials deteriorate when exposed to plasma due to chemical, mechanical and thermal damage (summarized in Table 2). High concentrations of RONS can cause chemical damage (oxidation of barriers and electrodes). Ozone is one of the strongest oxidizers found in nature (2.07 V, next to fluorine and 1.5× that of chlorine) [86–88]. In humid air, other powerful oxidizing agents such as the hydroxyl radical ($\cdot\text{OH}$) are formed as well [20,21,89,90]. Electron and ion bombardment can gradually erode the barrier (eventually leading to breakdown), thereby increasing the air gap and changing the surface properties (e.g., increase hydrophilicity or induce semi-permanent charging) [76,91]. When a DBD is operated with a high intensity for a long time, heat damage may occur [92,93]. Excessive heating even caused melting damage on a high voltage nickel electrode in [92].

Plasma degradation of the following dielectric barrier materials has been investigated: FR-4 (epoxy-fiberglass used as substrate for most printed circuit boards) [94,95], PVC [91,96], PMMA [96], polyimide ('Kapton') [95,97–99], polypropylene [99], silicone polymer [100], silicon nanowires [101], PTFE ('Teflon') [76] and natural quartz [102]. No degradation

was noticed in boro(alumino)silicate glass [96,99,103], alumina [97,99,104], mica [105] or man-made quartz [97,99].

Electrode degradation has been reported for a range of metals: silver/platinum [106], silver/palladium (shown in Figure 5) [106,107], copper [103,106,108], (photolytic deposited) gold [109], aluminum [108], silver [97,100,110], tungsten [103,111,112], nickel [92] and stainless steel [93,111,112]. Typical degradation patterns include oxidation, crater-forming, melting and erosion. Gold wire performed well in air as it did not oxidize [110,111].

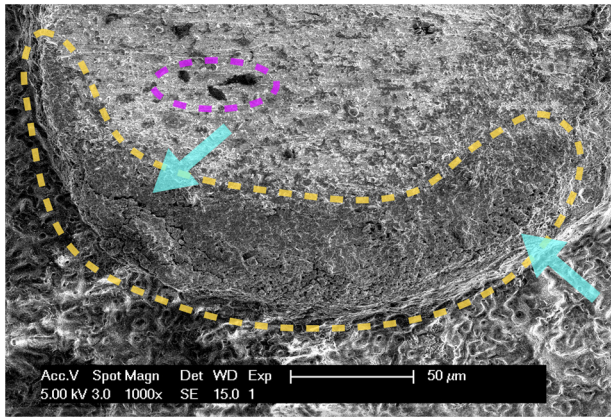


Figure 5. SEM image of erosion of an Ag/Pd electrode after long nanopulse operation. The arrows show the cracks. Black spots are encircled in purple, the eroded area in yellow, from [107].

The type of powering also affects electrode degradation. Nguyen-Smith et al. experienced melting of the nickel electrode in a pulsed surface DBD when $t_{\text{pulse}} = 12 \mu\text{s}$, whereas this did not occur when $t_{\text{pulse}} = 340 \text{ ns}$ [92]. This effect could be attributed to the much shorter duty cycle of nanopulses, leading to lower electrode heating. In addition, nanopulses produce a more homogeneous discharge [24,92,113], preventing the formation of local ‘hot spots’ on the electrodes.

The electrodes can be protected against plasma by embedding them in a dielectric. Embedded electrodes were employed in coplanar DBDs [7,114,115], grating DBDs [2], multi-hollow DBDs [34,116,117] and volume DBDs [118]. However, embedded electrodes are still subjected to (rapidly changing) high electric fields and may heat up, without the possibility of convectional cooling by air (flow). Another drawback is that part of the applied voltage is ‘lost’ over the barrier. This lowers E_g in the air gap. To compensate for this loss, higher operating voltages are required compared to DBDs with an uncovered electrode. It cannot be excluded that plasma forms in microscopic voids between electrode and adjacent barrier (called parasitic discharges in [1]), which can eventually damage electrode and barrier.

Concluding, electrodes and barriers are prone to degradation during DBD operation. The best material for electrodes appears to be gold, whereas the best materials for barriers appear to be ceramics, quartz and glass.

Table 2. List of materials that were damaged through LTP exposure.

Degraded Material	Observed Effect
FR-4 (epoxy/fiberglass)	Whitening, increased hydrophilicity [94], epoxy erosion [95]
PVC	Increased hydrophilicity [91], discoloration [96]
PMMA (acrylate)	Cavities, increased roughness, powder particle formation [96]

Table 2. Cont.

Degraded Material	Observed Effect
Polyimide (Kapton)	Erosion [95], structural change polymer chain, whitening, etching [99], hole formation [97], roughening [98]
Polypropylene (PP)	Breakdown, structural change polymer chain, whitening [99]
PTFE (Teflon)	Charging [76]
Silicone rubber	Erosion [100]
Silicon nanowires	Fracturing of nanowires [101]
Natural quartz	Microsoftening, decreased hydrophilicity [102]
Silver/Platinum	Oxidation, deformation [106]
Silver/Palladium	Oxidation, deformation [106], erosion, blackening [107]
Silver	Erosion [97], blackening [100]
Gold (photolytic deposited)	Erosion, melting [109]
Aluminium	Oxidation, microcraters [108]
Copper	Erosion [103], oxidation [108], blackening [106]
Copper with Ni/ Au coating	Rough edges, holes [94]
Tungsten	Oxidation [111,112], erosion [103]
Nickel	Erosion, melting [92]
Stainless steel	Release of particles [93], oxidation [111], erosion [112]

2.4. Heat Production and Dielectric Loss Factor

DBD heat production is generally undesirable as it reduces the electrical efficiency, can damage the DBD and lowers ozone production (as discussed in Section 2.8) [26,92]. The heating can be limited by using (nano)pulsed powering instead of conventional AC [60,113,119–121]. Zhu et al. showed that ozone production increased 28% when nanopulsed powering was applied instead of AC powering [121]. The maximum temperature of their surface DBD, reached 59 °C when powered by nanopulses, but 73 °C when using AC powering. Two mechanisms contribute to this effect.

Firstly, due to a faster changing electric field (dE/dt), most energy is transferred to the electrons (T_e), while less energy is used for vibrational and rotational excitation (T_{vib} , T_{rot}) of the gas molecules (O_2 , N_2 , CO_2 , H_2O). In any LTP, $T_e \gg T_{vib} > T_{rot} > T_{gas}$ [4,119,122,123]. But, the relative increase in T_e is larger for nanopulsed excitation than for AC (whether the AC is continuously operating or in burst mode) [119]. Excited gas molecules release heat when relaxing back to their ground state [52]. The local temperature can rise several hundreds of degrees in a few microseconds [52].

Secondly, the relatively long intervals between repetitive nanopulses, allow the air to cool down [31,124]. Another hypothesis is that the streamer channel exists for a shorter time under nanopulsed driving. The electric field in a streamer channel is too weak to ionize air molecules, and most energy goes into vibrational excitation [52]. Shortening the streamer channel duration therefore limits the vibrational heating of the gas [26]. However, the streamer channel might be important in radical formation, as Ono and Oda showed that it could contain the highest ozone densities (up to 150 ppm in their pulsed needle-plate DBD) [29].

Hatamoto et al. recognized three mechanisms for heat generation in DBDs, namely Joule heating, ion flux heating and dielectric heating by time-varying electric fields [125]. Joule heating is caused by the conduction current in the air. Exothermic reactions involved in the formation of RONS, such as ozone, also heat the surrounding air [1]. Hot air warms

the barrier by forced convection (depending on barrier's thermal conductivity). When Joule heating is low, convective transfer to the dielectric barrier is limited. Additional cooling can be achieved through a gentle airflow. Ion flux heating derives from ions which heat the air and the dielectric by collisions. Although ion motion (due to Brownian motion and recombination) cannot be avoided once a plasma is formed, the flux of ions is low under nanopulsed excitation. The dielectric barrier will absorb energy from high frequency electric fields. This interaction is controlled by the dielectric loss factor (dlf) [1,125]. Hatamoto et al. found that Joule heating gives the largest temperature rise for AC powering [125]. However, in nanopulsed DBDs, it is the relative contribution of the dielectric loss factor which becomes more significant.

The dielectric loss factor is defined by [1]:

$$\text{dlf} = \epsilon_r \cdot \tan(\delta) \quad (1)$$

where $\tan(\delta)$ is the dielectric loss tangential, which depends on the material and the operating frequency. Some values for common barrier materials are listed in Table 3 below:

Table 3. Tangential loss factor, relative permittivity and dielectric loss factor of commonly used dielectric barrier materials.

Material	$\tan(\delta)$	ϵ_r	dlf	Refs.
Quartz	1×10^{-3} – 1×10^{-4}	3.8–4	3.7×10^{-3} – 3.7×10^{-4}	[126,127]
Alumina	1×10^{-2} – 1×10^{-3}	9.6–9.7	9.5×10^{-3} – 9.8×10^{-2}	[104,126]
Borosilicate glass	1×10^{-3} – 1×10^{-4}	4.6–7	4×10^{-4} – 5×10^{-3}	[126,128]

A higher dlf leads to more heating of the dielectric, as more electromagnetic energy is dissipated by dipolar relaxation processes [1]. Once heated, thermal diffusion spreads the heat through the barrier, and this process is faster in materials with higher thermal conductivity [126]. Barrier materials with low ϵ_r and low $\tan(\delta)$ are most suitable for minimizing dielectric heating, which is beneficial for formation of certain RONS (discussed in Section 2.8).

2.5. Relative Permittivity (ϵ_r), DBD Capacitance (C_{cell}), Dielectric Thickness (d_d) and Electrode Length (l_e)

The dielectric properties of the barrier strongly influence DBD performance. A low ϵ_r decreases the electric field in the air gap (E_g) and the charge deposition (Q). This reduction can be compensated, by lowering the barrier thickness d_d , further discussed in Section 2.6 and [66,129,130].

For instance, Correale et al. reported a 33% increase in energy per pulse (E_p) when the barrier thickness was reduced by 50% [129]. This can be attributed to an increased DBD capacitance (C_{cell}). Leonov et al. observed that increasing C_{cell} from 33 to 55 pF, achieved by increasing electrode overlap, led to a 63% rise in the charge Q per pulse [51,131]. It doubled the discharge efficiency in their setup. Reducing d_d brings several benefits: it increases the conduction current, while lowering the displacement current [66]. It also shortens the delay between the voltage pulse and plasma ignition [66]. Hulka and Pietsch found that Q increased 3.6 times in their coplanar DBD, by changing the dielectric material from quartz ($\epsilon_r = 2.3$) to float glass ($\epsilon_r = 8.4$) [130]. While they used AC powering, the same effect is expected upon pulsed operation, by the fact that E_g will be magnified in the same manner. When they decreased the electrode distance from 2 to 1 mm (effectively lowering d_d and d_g), the number of visible microdischarges doubled. Similarly, when Zhang et al. reduced the dielectric barrier thickness of their pulsed surface DBD from 1 to 0.5 mm, the

measured current peak increased from 4 to 8 A [66]. It resulted in a doubling of the E_p . Patel et al. found that E_p increased from 0.7 to 2.8 mJ when d_d was lowered from 9 to 2 mm in a pulsed volume DBD [131]. We thus observe a similar trend in various DBD setups: the energy deposited in the plasma is increased either through increasing ϵ_r or decreasing d_d .

Both a low d_d and a high ϵ_r increase C_{cell} [131], which in turn increases the pulse rise time [85,126]. In nanopulsed DBDs, not only E_g (set by the pulse voltage), but also the rate of field change (dE/dt , controlled by t_{rise} , t_{fall}), plays a crucial role in RONS production [50,58,132,133] and plasma uniformity [134,135]. A higher dE/dt enhances the streamer-head velocity and the electron energy (T_e) but shortens the duration of a high local E_g [136]. These effects could oppose each other in the formation of specific RONS, such as ozone [57].

Taken together, these observations suggest that nanopulsed DBDs benefit from a moderate or low ϵ_r . It will lower dlf and increase dE/dt by reducing C_{cell} . Once C_{cell} is optimized, it can be further matched to the nanopulse source by adjusting the electrode length l_e (schematically shown in Figure 6). This will increase C_{cell} which in turn will increase the deposited charge Q (and thus the conduction current I_{cond} per pulse) [66,81,137] and RONS production [123,138]. Zhang et al. increased l_e from 10 to 50 mm in a pulsed surface DBD. As a result, the measured current increased from 1 to 7 A [66].

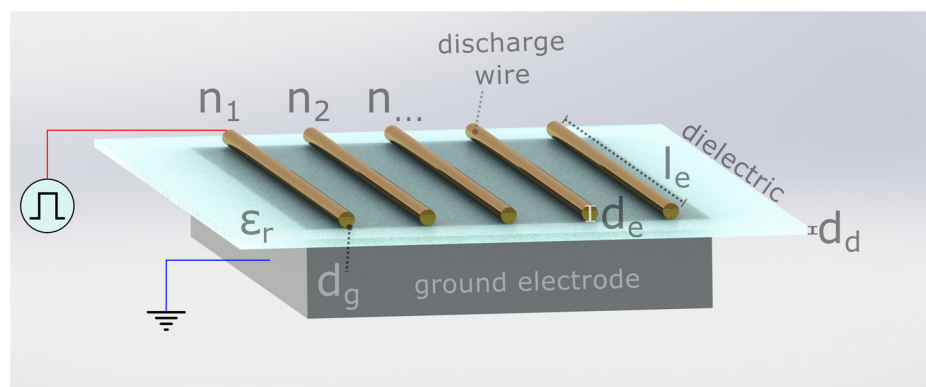


Figure 6. Wire-to-plate DBD, with dielectric barrier between the wires and the flat ground electrode. The high voltage pulses are applied to the thin discharge wires. Several parameters can be varied: the number of wires (n), ϵ_r , d_g , d_e , l_e and d_d .

Experimental results support this design strategy. Sokolova et al. found that the microdischarge length increased from 6 to 11 mm when ϵ_r decreased from 15 to 7 in a pulsed surface DBD, while the number of microdischarge channels remained nearly constant [58]. A similar finding was reported in [139], where the discharge length more than doubled when ϵ_r decreased from 9.8 to 2.4. For positive nanopulses, d_d itself did not influence the microdischarge length, reinforcing the preference for thin barriers with modest relative permittivity. Portugal et al. showed that, in a surface DBD, ozone production increased when the ratio d_d/ϵ_r declined (from 1.0 to 0.1), under otherwise identical conditions [140]:

$$\frac{dO_3(t)}{dt} \propto \frac{1}{\ln(d_d/\epsilon_r)} \quad (2)$$

This implies an equal contribution of d_d and ϵ_r to ozone production. It can be explained by a higher power deposition in the plasma, leading to a higher number of energized electrons, which increases the possibility of nonelastic collision with oxygen molecules, leading to ozone formation (as further discussed in Section 2.8). Concluding, a DBD capacitive load (C_{cell}) should stay within bounds for the pulse source to perform well. Strategies to lower C_{cell} include lowering the barrier relative permittivity, increasing the

barrier thickness, or decreasing the electrode area. However, to effectively deposit energy in the plasma, a low dielectric thickness and a long electrode length are beneficial. This conflict could be best resolved by choosing a thin barrier with a low to moderate ϵ_r , while using a thin discharge electrode, with the length l_e scaled so that C_{cell} is within bounds for the pulse source to operate reliably. C_{cell} unavoidably changes during plasma formation. It increases when surface charge remains on the barrier after a microdischarge event (see Section 2.2). Mitigation strategies include lowering the PRF (giving the remaining charge more time to decay), using bipolar pulses and choosing barrier materials with fast charge decay (such as silicone).

2.6. Electric Field Strength (E_g) Enhancement

The applied voltage strongly affects streamer development in DBDs. With the pulse voltage, the streamer length and width can be varied [50]. The voltage controls the magnitude of E_g , and also the duration of the high field period. A higher field allows the streamer head to propagate further along the barrier surface, thereby increasing the discharge length. Ultimately, E_g will define the RONS composition as it controls the electron density n_e , T_e and I_{cond} [135,141,142].

An effective way to enhance E_g is to reduce d_d rather than raising ϵ_r . This strategy was applied by Park, Eden et al., who achieved a volume DBD with a long electrode length and a thin alumina barrier of several hundreds of μm through an oxidation process [118]. As a result, ozone was formed at a very low applied voltage of 1.1 kV_{RMS}. V_{br} depends on both d_d and ϵ_r . V_{br} decreases by lowering d_d (and/or increasing ϵ_r) [66]. Soloviev et al. showed that increasing ϵ_r from 10 to 100 reduced the breakdown voltage by ~43% [143]. The dielectric thickness also influenced V_{br} as $V_{\text{br}} \propto \sqrt{d_d}$, provided that surface roughness is negligible [143]. The following relation holds between V_{br} , ϵ_r and d_d [143]:

$$V_{\text{br}} = 2.3 \times \left(1 + \frac{1}{\pi \times \epsilon_r} \right) \times \sqrt{d_d} \quad (3)$$

where V_{br} is in kV and d_d in mm (for ϵ_r between 3–10). The relationship between E_g and V_{br} is important: a decrease in V_{br} implies a stronger E_g and more efficient plasma production.

Zhang et al. modelled the effect of ϵ_r and d_d on the discharge energy (mJ/cm^2 , which is the energy per pulse E_p divided by the plasma area) [144]. They found that the discharge energy relies much more on d_d than on ϵ_r . Specifically, when d_d was lowered 6 times, the discharge energy increased 15 times. When ϵ_r was raised 3.7 times, the discharge energy increased 4.9 times. In this model, dE/dt was held constant, so the effect of changing C_{cell} (either through ϵ_r or d_d) on the voltage rise time was not accounted for. They further reported that increasing ϵ_r from 2.7 to 10 raised E_g by 9%, n_e by 83% and T_e by 1.4%. Reducing d_d from 0.9 to 0.15 mm also raised E_g by 2.7%, n_e by 143% and T_e by 0.9%. Experimental studies also confirm the dominant effect of barrier thickness. Using the Lissajous method for an AC-driven volume DBD, Yang et al. found that increasing ϵ_r from 3.9 (quartz) to 9.6 (alumina) increased the discharge power by ~54%, whereas decreasing d_d from 3 to 1 mm increased the power > 400% [145].

Kogelschatz proposed that the charge Q is proportional to ϵ_r/d_d , implying equal contributions from both parameters [146]. Peeters also observed an increase in charge per pulse with increasing ϵ_r/d_d , although the magnitude depended strongly on the material [147]. The increase was four times stronger for quartz (0.08 nCmm) than for alumina (0.02 nCmm). Zhang et al. found that a DBD with a quartz barrier showed higher microorganism disinfection efficacy per energy input compared to other barriers (alumina, PEEK) [2].

In practice, not only ϵ_r and d_d contribute to Q (and thus I_{cond}). Other factors like the surface roughness also play a role. Alumina surfaces are rougher than quartz surfaces [126].

Rougher surfaces cause microscopic deviations in the electric field distribution, and lead to localized microdischarges [41,52]. This effect becomes more pronounced for thin electrodes and barriers [143]. It leads to ‘hot spots’, locations where microdischarges more frequently occur. This local heating may lead to faster material degradation. Other material properties such as water absorption, secondary electron emission and shallow traps also affect the discharge characteristics [39,52,148]. Shallow traps are structural defects where electrons with a low energy T_e (<1 eV) can be temporarily stored [39]. These provide seed electrons for subsequent discharges.

An alternative route to enhancing RONS formation involves the application of a magnetic field in the air gap [149–154]. Electrons in the air gap follow helical trajectories (Larmor precession), thereby increasing the probability of collision with gas molecules [151]. Although beyond the scope of this review, this mechanism suggests that adding magnets to a DBD may further increase the RONS yield. A summary of the influence of all these DBD variables is given in Table 4. In general, a thin dielectric enhances the electric field in air, similarly to a high relative permittivity of the barrier. Lowering the barrier thickness appears to be the most effective strategy to this end, however, as ϵ_r often is not free to choose.

Table 4. Influence of the dielectric barrier relative permittivity (ϵ_r), dielectric thickness (d_d), electrode length (l_e), electrode width (w_e) and air gap (d_g) on various plasma parameters. E_g : electric field strength in air gap. E_d : electric field strength in dielectric barrier. V_{br} : breakdown voltage, dlf: dielectric loss factor, C_{cell} : DBD capacitance (plasma off), t_{rise}/t_{fall} : rise and fall time of the voltage (nano)pulse, n_e : electron density, T_e : electron temperature (electron energy), I_{cond} : conduction current, RONS: reactive oxygen and nitrogen species, Q : deposited charge (per pulse), E_p : energy per pulse, η_{O_3} : ozone yield.

	$\epsilon_r \uparrow$	$d_d \downarrow$	$l_e \uparrow^*$	$w_e \downarrow$	$d_g \downarrow$
E_g	$\uparrow$	$\uparrow$	-	$\uparrow$	$\uparrow$
E_d	$\downarrow$	$\uparrow$	-	$\uparrow$	$\downarrow$
V_{br}	$\downarrow^{**}$	$\downarrow$	-	$\downarrow$	$\downarrow$
dlf	$\uparrow$	-	-	-	-
Barrier heating	$\uparrow$	$\uparrow$	$\downarrow$	$\uparrow$	$\uparrow$
C_{cell}	$\uparrow$	$\uparrow$	$\uparrow$	$\downarrow$	$\uparrow$
t_{rise}, t_{fall}	$\uparrow$	$\uparrow$	$\uparrow$	$\downarrow$	$\uparrow$
dE/dt	$\downarrow$	$\downarrow$	$\downarrow$	$\uparrow$	$\downarrow$
I_{cond}, n_e, T_e	$\uparrow^{***}$	$\uparrow^{***}$	$\uparrow^{***}$	$\uparrow$	$\uparrow^{***}$
RONS	$\uparrow$	$\uparrow$	$\uparrow$	$\uparrow$	$\uparrow$
Q, E_p	$\uparrow^{****}$	$\uparrow$	$\uparrow$	$\downarrow$	$\uparrow$
η_{O_3}	$\uparrow$	$\uparrow$	-	$\uparrow$	$\uparrow$

$\uparrow$: Increase of parameter value. $\downarrow$: Decrease of parameter value. * Provided that the change in resistance is negligible with the increase in electrode length (for example by using bypasses). ** V_{br} is more dependent on d_d than on ϵ_r [143]. *** An increase in E_g increases this variable, but a decrease in dE/dt decreases this variable. We here assumed that the effect of E_g is dominant over that of dE/dt , based on [2,34,62]. **** Magnitude varies per material [147].

2.7. Electrode Width (w_e) and Gap Distance (d_g)

The geometry of the electrode strongly influences the electric field in the discharge region. Electrodes with sharp or narrow features are commonly employed in LTP systems because they enhance E_g due to their small radii of curvature (compared to a flat or wide electrode) [46]. As a result, V_{br} will then also become lower [107]. Therefore, many DBD setups use thin wires, needles, sharpened edges or narrow electrodes, with small electrode

width w_e [155–157]. Using sharp or narrow electrodes produces several beneficial effects. It promotes faster streamer head initiation, increases streamer density and leads to a more uniform plasma [60]. It boosts the ionic wind (thrust) [155] and enhances ozone yield [158]. Lowering w_e also decreases C_{cell} (by lowering area A in the equation $C = \epsilon_0 \times \epsilon_r \times A/d$), allowing a higher dE/dt and a lower displacement current I_{disp} [62].

The gap distance d_g also plays a crucial role. It was explained in [62] that a small gap distance increases E_g , which boosts the primary streamer head velocity, the streamer diameter, and oxygen radical production in air. It was shown that as d_g increases, E_g will weaken, causing the conduction current to drop sharply [159,160]. The conduction current peak dropped from ~2 to 0.6 A when the gap distance in a pulsed corona discharge went up from 3.5 to 4.5 cm [159]. It follows from Paschen's law that V_{br} increases when d_g increases (this occurs for gaps $> 7.5 \mu\text{m}$ in atmospheric air, depicted in Figure 7) [161]. Small gaps ($< 1 \text{ mm}$) decrease the energy per individual microdischarge [162]. Then, charge deposition will become more homogeneous (given that l_e is sufficient to accommodate multiple microdischarges simultaneously [53]). This behaviour was observed for gap distances of 1 and 4 mm in [162]. A short gap distance was for example applied in the multi-hollow DBD (by Homola et al. in [116]), where $d_g = 0.6 \text{ mm}$, promoting ozone production with a yield of up to 205.5 g/kWh in air Aerts et al. found that lowering d_g (from 3.3 to 1.8 mm) increased CO_2 conversion in their DBD from 20 to 30% [163]. They related this to the smaller gap having a higher power density, a higher electron density and a stronger electric field in the air gap. Buntat et al. found that a decrease in d_g from 3 to 1.5 mm resulted in an increase in ozone production (500 vs. 1200 ppm) in a volume DBD [33]. Additionally, a small d_g appears to suppress the production of low energy electrons that would otherwise contribute to ozone dissociation through electron-impact processes [1,164]. A small electrode width and gap distance could thus, in summary, facilitate plasma formation in air.

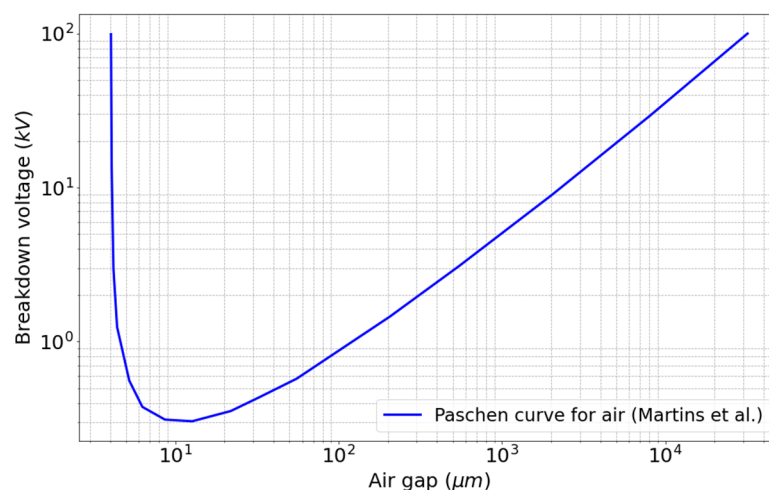


Figure 7. Paschen curve for atmospheric air. Plotted based on the data given in [161].

The combined findings in Sections 2.5–2.7 are summarized in Table 4 and Figure 8.

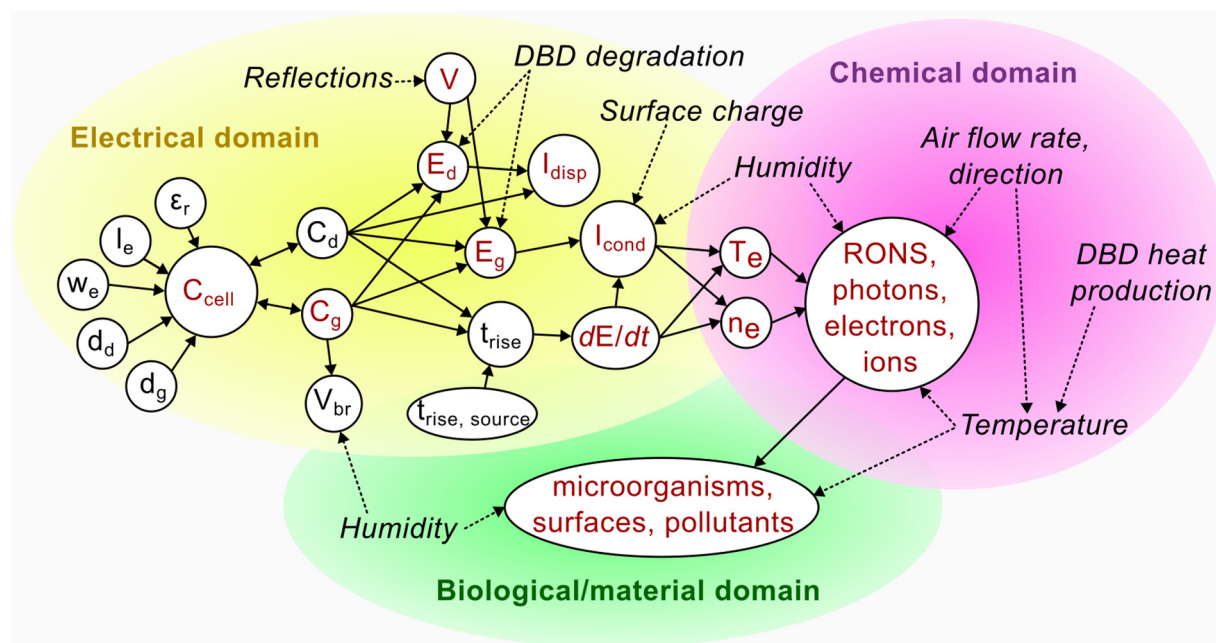


Figure 8. Many variables could be considered when designing pulsed DBD systems for treating a material or biological substrate in ambient air. These variables could be categorized in the electrical domain (pulse source, DBD parameters), the chemical domain (RONS and heat production, ambient temperature, humidity and presence of air flows) and the biological/material domain (interaction of the plasma with the object to be treated). The main variables are displayed in this figure. Black arrows point to a variable that is influenced by another variable (i.e., E_g influences I_{cond} , which then influences n_e). Red terms are varying in time (during plasma operation), and black terms are constant over time. Dashed arrows: External and ambient factors influencing DBD performance.

2.8. Ozone Production and Decomposition

Ozone production is one of the most important applications of nanopulsed DBDs. LTP is regarded as one of the most efficient technologies to produce ozone on an industrial scale [165]. Ozone is used for gas cleaning, odour removal, disinfection and water treatment [3,4,12]. The production of ozone in air depends strongly on microdischarge channel temperature, ambient temperature and humidity, air flow, electric field strength and voltage pulse rise time.

2.8.1. Formation, Decomposition and Transport of Ozone

Ozone formation can be lowered, when the ambient temperature and/or microdischarge channel temperature increase [32,166–170]. This effect, called ‘quenching’ [171–173] or discharge poisoning [15,26,174], occurs when O atoms rapidly react with nitrogen molecules before they can form ozone [26]. Under such conditions, the discharge shifts from the O_3 mode (producing mainly O_3 and some N_2O , NO_2 , N_2O_4 , N_2O_5 and HNO_3) to the NO_x mode (producing mainly HNO_3 , NO , N_2O_5 , NO_2 and N_2O) [117,174–176]. Active cooling can delay this shift by lowering the temperature rise [20,26]. Similarly, excessive voltages and pulse repetition frequencies can quickly heat the plasma (by a too high power density or specific input energy (SIE)) and also trigger a transition from O_3 to NO_x mode [177].

Some ozone formation and dissociation reaction rates are plotted against air temperature in Figures 9 and 10.

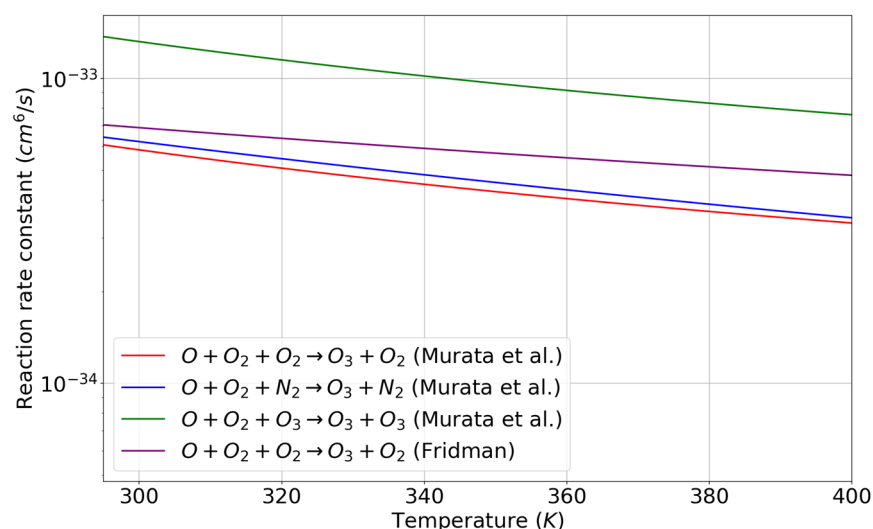


Figure 9. Ozone formation reaction rate constants versus air temperature, based on the rate coefficients provided in [26,32].

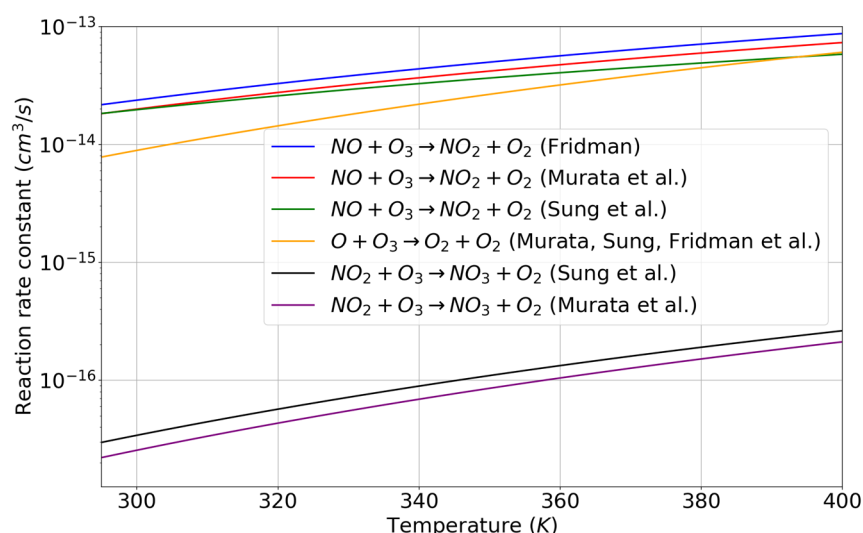


Figure 10. Ozone dissociation reaction rate constants versus air temperature, plotted by using the rate coefficients given in [26,32,168]. Note that the curves given by the authors for the same dissociation reaction differ somewhat.

Ozone can decompose, once formed, by several ambient factors. Air flow can be useful to transport ozone away from the plasma area. However, high air flow can promote ozone decomposition by mechanical force [20,117,176,178,179]. Increasing the humidity also reduces the ozone yield (Equation (5)) [20,94,117,178], because atomic oxygen (O) participates in competing reaction pathways, leading to HO_2 and hydroxyl radicals production, rather than ozone [26,89]. A high humidity also affects the plasma dynamics by lowering streamer radius and propagation speed [52], increasing the surface conductivity of the barrier and modifying the electric field in the air gap [26,165]. Charged water clusters can be formed in humid air, which capture ions and electrons, further limiting ozone formation [52,180].

Ozone production in air relates closely to the electric field in the air gap [181]. The parameter commonly used is the reduced electric field E/N (in Townsend units, Td), where E equals E_g and N is the number density of neutral particles ($N = 2.46 \times 10^{25}$ in air). E/N can be estimated using optical emission spectroscopy (OES, shown in Figure 11) [182–185], the EFISH [73], SHG [68] or Pockels technique [186]. But, this is not simple, as E/N varies both in time (by AC or pulsed powering, and by streamer propagation) and in space

(especially in the case of asymmetric and/or sharp electrodes). OES therefore uses time- and space integration. With a correction factor (CF), the streamer head field (E_s) and the average field (E_e) can be estimated (with $E_s = 1.4 \times E_e$ in atmospheric air) [187]. Surface charging may alter E/N markedly in a DBD (see Section 2.2). Even though E/N is not precisely known, it is well established that for optimal ozone production $V(t)$ must be moderate, only slightly above V_{br} (displayed in Figure 12) [26,181]. Buntat et al. showed that ozone production in a volume DBD clearly increased by increasing the air flow rate (0.2–1 L/min) [33]. But, at each rate they found the highest ozone yield at the same voltage, underscoring the importance of E_g in ozone production.

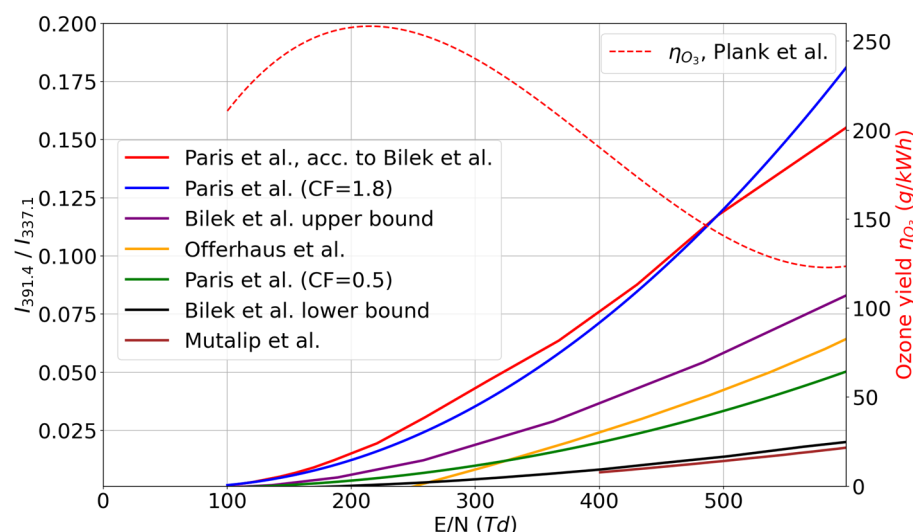


Figure 11. OES wavelength intensity ratio (391.4 vs. 337.1 nm) versus reduced electric field and ozone yield. The equations given in [181,182,184,188,189] were plotted. Notice that the authors make different predictions of the course of the intensity ratio vs. E/N .

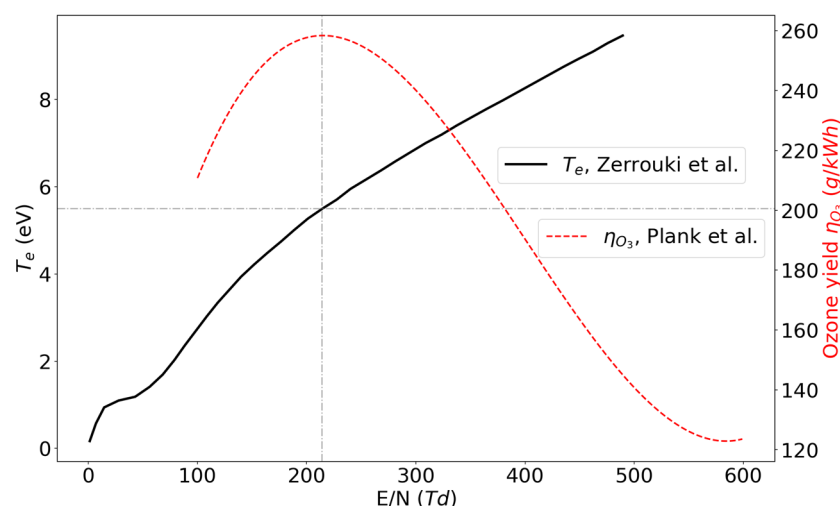


Figure 12. Electron energy and ozone production yield versus electric field strength in air, based on [141,181]. The value of T_e is marked where the ozone yield is maximal.

Figure 13 shows how the electron energy is separated among different processes in air [25]. Plank et al. and Haacke et al. found maximum ozone production at $E/N = 214$ Td, which equals ~ 53 kV/cm in ambient air [181,190]. At this point the largest fraction of electron energy goes into O_2 vibrational excitation, which drives O_2 dissociation, forming atomic oxygen. Atomic oxygen next forms ozone (shown in Figure 9 and [26,181,187]). The

optimal electron energy for ozone production is ~ 5.5 eV [26,141,191], while the threshold for O_2 vibrational excitation is 4.63 eV [192], easily reachable with pulsed DBDs.

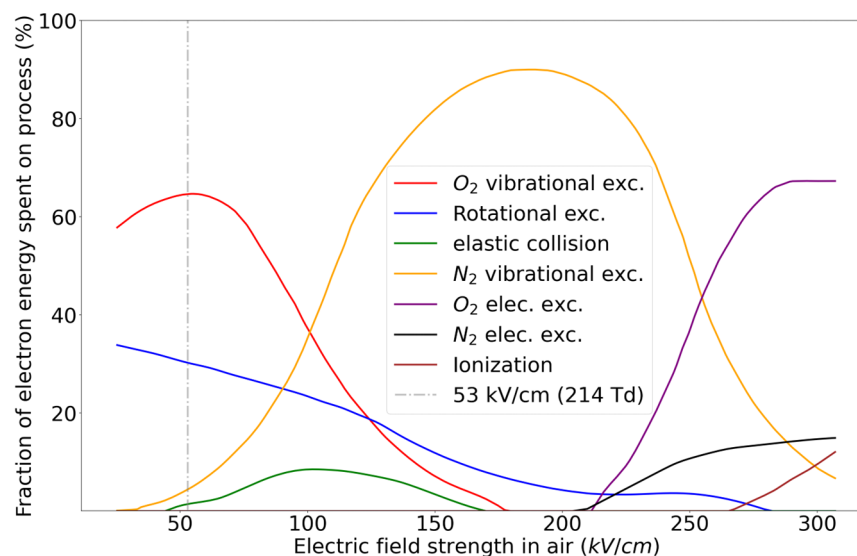


Figure 13. Fractions of electron energy spent on different chemical processes versus electric field strength in air. Plotted based on data given in [25].

Ozone diffusion cannot be neglected. When ozone is formed, it should travel from the plasma region to mix with the surrounding air. After a microdischarge, ozone appears in 8–10 μs via the ‘third body reaction’ $O + O_2 + M \rightarrow O_3 + M$ (various molecules can act as M, as displayed in Figure 9) [193]. The total reaction time to form ozone through all pathways may take up to 100 μs [26,29,136,194]. Therefore, pulse or AC frequencies of >10 kHz can interfere with ozone production by limiting the post-discharge period needed for proper ozone formation and diffusion. For example, Park et al. found that the ozone concentration more than doubled in their surface DBD by using positive nanopulses with a PRF of 1 kHz compared to 3 kHz [31]. They also hypothesized that a higher PRF promoted ozone quenching, which is probable as it increases the gas temperature due to a higher number of microdischarges over time. Diffusion of ozone out of the plasma region and subsequent mixing with the background gas could take up to 100 ms (depending on geometry [195]) [52,90,194,196]. The diffusion coefficient for ozone is about 0.1–0.2 cm^2/s [165]. Pressure waves originating from microdischarges create an additional air flow, as observed by Schlieren imaging in [45,129,197].

2.8.2. Influence of Pulse Rise Time on Ozone Production

The density of atomic oxygen increases with pulse rise rate, accelerating streamer head propagation [62]. However, it is unclear whether this really produces more ozone, as not only reactions in the streamer head, but also those in the streamer channel, cathode fall, secondary streamer and post-discharge period contribute to ozone formation and decomposition [136]. Ono and Oda observed that, in a pulsed wire-plate DBD, the secondary streamer (which contained the highest ozone density) elongated at higher pulse voltages [29]. But, excessively long secondary streamers may cause gas heating, and potentially provoke ozone dissociation [198].

Sun et al. found that the ozone concentration increased when the pulse rise time increased from 125 to 225 ns in a bipolar pulsed surface DBD, possibly due to a lower electrode temperature [175]. They calculated that faster rise times increased the vibrational N_2 excitations, raising NO production and decreasing O_3 . Iza et al. found that pulses with 40 ns rise times produced higher mean electron energies than those with a rise time of 20,

10 or 1 ns [37]. Jin et al. observed only a moderate ozone yield rise (from 25 to 35 g/kWh) when the rise time was lowered greatly from 250 to 50 ns in a pulsed multi-hollow DBD [34]. However, the specific input energy (SIE, regulated by the pulse voltage) had a much larger impact on ozone yield in their study. Zhang et al. found that the peak total current increased from 3.3 to 18.2 A when the pulse rise time was shortened from 500 to 50 ns [24]. However, even higher peak currents were obtained by simply increasing the pulse voltage from 12 to 16 kV (ozone was not measured). Rao et al. confirmed this finding by showing that higher light emission occurred when the pulse rise time decreased from 1239 to 132 ns in a volume DBD [132]. Thus, the conduction current, responsible for plasma formation, can be increased effectively by lowering the pulse rise time, but also by increasing the pulse voltage. Komuro et al. concluded that the average applied electric field during streamer propagation is more important than the rise time for oxygen radical production [62,198]. The conduction current depends on E/N as well [135]:

$$I_{\text{cond}} \sim n_e \times \mu_e \times (E/N) \sim (E/N)^{0.88} \quad (4)$$

where μ_e is the electron mobility.

2.8.3. Ozone Yield

Ozone yield (in g/kWh) is often calculated as [34,116,173]:

$$\eta_{\text{O}_3} = \frac{60 \times C \times Q_s}{P} \quad (5)$$

where C is the ozone concentration (in g/Nm³) and Q_s the gas flow rate (in standard litre per minute). P is the discharge power, calculated by integrating the voltage-current product over a single pulse, multiplied with the PRF. In AC reactors, it is the area of the Lissajous figure [34,116]. Ozone yields of ~200 g/kWh in air are considered high [4,60,116]. Cimerman et al. reported that a shift from ozone mode to NO_x mode occurred when the specific input energy ($\text{SIE} = 60 \times P/Q_s$) was 1000–1100 J/L in dry air and 600–450 J/L when the relative humidity increased from 20 to 80%, using AC power [117]. Absorption spectra showed that NO₂ and N₂O are the most prominent species in NO_x mode. Jin et al. used a similar DBD with nanopulsed power [34]. They found that ozone concentration increased with SIE up until 300 J/L (with minimal NO₂ and N₂O concentrations). At higher SIE, NO₂ and N₂O started to rise up to 3 and 35 ppm, respectively, at the expense of ozone [34].

Several factors complicate the use of Equation (5). First, the impact of ambient temperature and humidity on ozone production is not included [20,117,170]. Some authors tried active cooling or air drying to increase ozone production [20,199]. The extra power needed for cooling and drying is not added to P . Second, air flow patterns are complex and affect ozone formation and dissociation. Ozone yield differs for laminar and turbulent flow, it is also affected by the flow direction and interaction with the electrodes and reactor walls [32,36,135,142,170,179]. For example, an air flow of 60 m/s over the electrode improved discharge homogeneity in a pulsed surface DBD [135]. Xie et al. reported a ~30% increase in η_{O_3} when the air flow direction changed from lateral to transversal towards the electrodes [179].

The calculation of P is prone to errors [116]. A time delay between voltage probe and current coil changes the outcome of the voltage-current integral. The current measurement may be distorted by EM interference coming from the cables powering a nanopulsed DBD (called ‘loop inductance’ in [200]). To minimize this effect, the cable loops must be kept short. Voltage probes and current coils should be calibrated by measuring the V-I relationship of a resistor driven by a waveform generator. The voltage and current values then obey Ohm’s law. Time delays between probe and coil are also made visible by

this method. Lastly, reflections of the voltage pulse may cause additional distortions (see Section 2.9). An detailed error analysis is given in [116].

In general, ozone yield will maximize when microdischarge temperature is low, as formation is then promoted, and dissociation lowered. It can be achieved by various strategies: applying moderately high voltages (slightly above V_{br}), moderate PRFs (100–1000 Hz) and short pulses (in the range of several hundreds of ns with a fast rise time). Additional air flow can increase ozone yields, but this depends on air flow velocity and direction. As such, it can heavily complicate yield calculations. A low ambient humidity promotes ozone yield. DBD heating, which promotes ozone decomposition, could also be partially mitigated by choosing thin dielectric barriers.

2.9. DBD—Pulse Source Interaction

Interactions between the DBD and the pulse source occur virtually in all systems, yet are seldom discussed in detail. Some important effects are summarized below.

A voltage pulse, fed to a plasma reactor, can be (partially) reflected back into the pulse source. Such reflections disturb the pulse shape [201], reduce power efficiency and may damage the pulse source [124,129,191,202]. One way to prevent this is by matching the pulse source and plasma device impedances [158,203–205]. This approach is used in an impedance matched Marx generator (IMG) and is equally necessary for single-line and Blumlein-line pulse sources [85,133,191,206]. These pulse sources operate efficiently only when connected to a predetermined load impedance (and thus DBD size). Reducing the barrier thickness from 1 to 0.5 mm decreased the fraction of reflected energy from 72 to 51%, by lowering the DBD impedance [129]. It should be noted that plasma formation temporarily increases the capacitive load, complicating matching [50,204,207]. A high pulse source impedance further increases the characteristic time for potential variation of the HV electrode [72]. This decreases the (conduction) current and thus plasma formation.

C_{cell} and the resistance of the electrode both increase the pulse rise time in push-pull nanopulse switches [208–210]. Barrier surface roughness can also distort the applied waveform [39]. C_{cell} limits the operational range of the switching device, as its maximum operational power is the product of DC input voltage, PRF and capacitive load [209,211]. The pulse source output voltage is influenced by the effective impedance of the load, which may change due to electrode erosion, barrier degradation and surface charging. Likewise, some pulse sources, particularly spark-gap switches, erode during long-term operation. This may affect the pulse source output voltage characteristics [85,206].

3. Summary & Outlook

This review investigates the key parameters for the design and operation of a pulsed DBD for low-temperature plasma production in ambient air. In general, low-temperature plasma production using a pulsed DBD benefits from a thin dielectric barrier, with moderate to high relative permittivity, a small gap distance (in the order of tens-hundreds of μm) and the use of sharp, long electrodes. Regarding electrode material, gold appears to perform very well, as it is oxidation resistant. Barrier materials such as ceramics, glass and quartz are clearly superior to polymers, as polymers show signs of (semi)permanent charging and plasma damage (heat, etching, oxidation). For optimal performance of a pulse source, the DBD capacitance (C_{cell}) should ideally be as low as possible (or match the source impedance for some pulse sources). This allows for high voltage pulses with short rise times and low reflections. Short pulses (in the range of several hundreds of ns or shorter) limit electrode and barrier degradation.

The operation of a pulsed DBD in air is largely dependent on ambient factors such as temperature, humidity, air flow direction and speed. One also has to consider if the

DBD should operate in ozone mode or NO_x mode. Heat production varies largely per DBD geometry (i.e., electrode shape) and powering (waveform, repetition frequency). Therefore, optimal settings varied among different studies.

The electrification of the power grid provides opportunities for upscaling of pulsed DBD systems. This is supported by the fact that high voltage components (such as MOSFET switches) are becoming less costly at increasing voltage ratings. The market demand for such systems is expected to increase, based on regulatory pressure (for example the outlawing of pesticides) and the trend towards lowering energy costs.

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Abbreviations

The following abbreviations are used in this manuscript:

LTP	Low-temperature atmospheric plasma
RONs	Reactive oxygen and nitrogen species
ϵ_0	Vacuum permittivity
ϵ_r	Relative permittivity
w_e	Electrode width
d_g	Gap distance
d_d	Dielectric barrier thickness
l_e	Electrode length
C_{cell}	DBD capacitance
C_d	Dielectric barrier capacitance
C_g	Air gap capacitance
U_g	Voltage over the air gap
U_d	Voltage over the dielectric barrier
R_g	Variable resistance of the air gap
t_{rise}	Rise time of voltage pulse
t_{fall}	Fall time of voltage pulse
$t_{rise,source}$	Rise time of the pulse source without load
E_g	Electric field strength in the air gap
E_d	Electric field strength in the dielectric barrier
dE/dt	Change of electric field strength over time
N	Number density of neutral particles
E/N	Reduced electric field
T_{gas}	Gas temperature
T_{vib}	Vibrational temperature
T_{rot}	Rotational temperature
T_e	Electron energy (electron temperature)
n_e	Electron density
μ_e	Electron mobility
V	Applied (peak) voltage

V-I	Voltage-current
V_{br}	Breakdown (ignition) voltage
I_{cond}	Conduction current
I_{disp}	Displacement current
Q	Charge per pulse
E_p	Energy per pulse
P	Discharge power
PRF	Pulse repetition frequency
C	Ozone concentration
η_{O_3}	Ozone yield
SIE	Specific input energy
Q_s	Standardized air flow
AC	Alternating current
$\tan(\delta)$	Tangential loss factor
dlf	Dielectric loss factor
PMT	Photomultiplier tube
OES	Optical emission spectroscopy
EFISH	Electric field-induced second harmonic
SHG	Second harmonics
IMG	Impedance-matched Marx generator
τ_r	Ion recombination time
τ_d	Space charge diffusion time on dielectric surface
τ_s	Surface charge relaxation time
ρ_v	Material volume resistivity
Anode	Electrode with positive voltage applied
Cathode	Grounded electrode
PFAS	Per- and polyfluoroalkyl substances
PTFE	Polytetrafluoroethylene (Teflon)
PI	Polyimide (Kapton)
PP	Polypropylene
PMMA	Polymethylmethacrylate (Plexiglas)
PVC	Polyvinylchloride
PEEK	Polyether ether ketone

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