

Atmospheric pressure plasma of dielectric barrier discharges*

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Abstract: The dielectric barrier discharge (DBD) has a number of industrial applications and has been a subject of research for many years. Many studies have been carried out to understand the underlying DBD physics. Despite the fact that much progress has been made, some important issues are still far from being clear. In this work, we summarize the basics of DBD physics and introduce innovative concepts of discharge behavior that were discovered recently.

Keywords: dielectric barrier discharge; streamer; cooperative phenomena; memory effect; regular microdischarge pattern; plasma chemistry.

INTRODUCTION

The first exploitation of dielectric barrier discharges (DBDs) was started in Europe 100 years ago. It was used for ozone production for the treatment of drinking water [1,2]. Since then, the number of industrial applications of this type of discharge started to grow. Now, DBDs are successfully applied to pollution control and to polymer surface treatment in order to promote wettability, printability, and adhesion [3,4]. With the latest invention of atmospheric pressure glow (APG) discharge that also is based on DBD, the fundamental understanding of barrier discharges becomes important.

The DBD possesses essential advantages in surface processing and plasma chemistry. DBD is a low-temperature discharge, usually working at atmospheric pressure. DBD plasma is typically obtained between two parallel electrodes separated by a gap of some millimeters and excited by alternating current (ac) voltage with frequency in the range of 1–20 kHz. The dielectric barrier can be made from glass, quartz, ceramics, or polymer—materials of low dielectric loss and high breakdown strength [5]. As an example, steel tubes coated by an enamel layer can be effectively used in the DBD. The DBD proceeds in most gases through a large number of separate current filaments referred to as microdischarges. These microdischarges have complex dynamic structure and are formed by channel streamers that repeatedly strike at the same place as the polarity of the applied voltage changes, thus appearing to the eye as bright filaments. The extinction voltage of the microdischarges is not far below the voltage of their ignition. Charge accumulation on the surface of the dielectric barrier reduces the electric field at the location of a microdischarge, which results in current termination within tens of nanoseconds after breakdown. The short duration of current in microdischarges leads to low heat dissipation, and the DBD plasma remains strongly nonthermal.

*Paper based on a presentation at the 16th International Symposium on Plasma Chemistry (ISPC-16), Taormina, Italy, 22–27 June 2003. Other presentations are published in this issue, pp. 345–495.

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Recently, there has been interest in characterizing and understanding the diverse phenomena that can be found in atmospheric pressure discharges [6]. The nature of the discharge depends on the gas mixture employed, the dielectric, and the operating conditions. Both glow and filamentary discharge modes were observed at atmospheric pressure, and the experimental conditions leading to ordering or patterning of microdischarges have been reported [6]. However, the development of experimental methods, such as imaging techniques, for quantitative characterization of microdischarges (filaments) and associated cooperative phenomena in atmospheric pressure discharges is still lacking. Furthermore, theoretical models describing cooperative phenomena in these discharges are incomplete.

MICRODISCHARGES AND STREAMERS IN DBDs

In most cases, a DBD is not uniform, but consists of numerous non-steady-state local microdischarges distributed in the discharge volume. The term “microdischarge” refers to a bright, thin plasma filament that is observed in DBDs. The physics of these microdischarges is very sophisticated and is based on the initial avalanche-to-streamer transition that is followed by the streamer formation. The time-line (from left to right) for these processes that result in microdischarge streamer formation is presented in Fig. 1. The avalanche-to-streamer transition and streamer propagation takes about 10 ns in typical DBDs. Streamers are local ionization waves usually moving from anode to cathode to meet avalanches propagating in the opposite direction. The streamer occurs when the applied voltage is high enough that the local electric field caused by charge accumulation in avalanches allows streamer formation. The streamers move very fast (about 10^8 cm/s, an order of magnitude faster than avalanches) and cover the distance between electrodes in nanoseconds. The right side of Fig. 1 shows streamer propagating from the anode to the cathode while attracting additional avalanches. A microdischarge is formed by channel streamers that strike at the same place each time the polarity of applied voltage changes. The persistence of streamers to strike at the same place of their ancestor is due to *memory effect*. The memory effect is associated with charge deposited on the dielectric barrier, as well as on residual charges and excited species in the microdischarge channel. The *memory effect* was described in detail in [7].

The description of microdischarges is based on an understanding of the formation, propagation, and interaction of streamers. The electrons in the conducting channel established by the streamers dissipate rapidly from the gap (in about 30 ns), while the heavy, slowly drifting ions remain in the discharge gap for several microseconds. The deposition of electrons from the conducting channel onto the anode dielectric barrier results in charge accumulation and prevents the formation of new avalanches and streamers nearby, until the cathode and anode are reversed. After the voltage polarity reverses, the deposited negative charge facilitates the formation of new avalanches and streamers in the same spot.

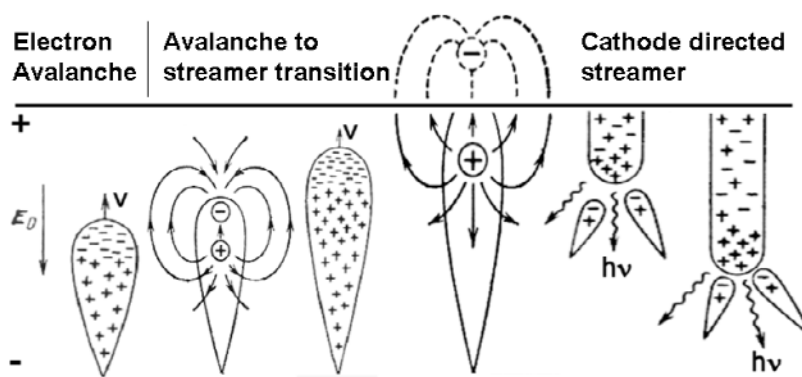


Fig. 1 Evolution of electron avalanche in discharge gap, showing avalanche development, avalanche-to-streamer transition, and streamer propagation.

As a result, a many-generation family of streamers is formed that can be macroscopically observed as a microdischarge that appears to be spatially localized.

EXISTENCE AND IMPORTANCE OF PATTERNS

Streamers were the subject of intensive study over the past century; however, mutual interaction of microdischarges was discovered only recently. This interaction forces microdischarges to arrange themselves into regular patterns, which are similar to Coulomb crystals. Although the patterns' existence was noticed previously [8,9], the physics that leads to the pattern formation was not explained. It appears that the pattern structure has a dramatical influence on DBD performance in most applications. Here, we briefly describe the physics behind the microdischarge interaction and demonstrate its validity by computer simulation of microdischarge patterns and comparison with experiments, referring to the work done at the Drexel Plasma Institute and at Eastman Kodak Company [10,11].

An innovative model that simulates dynamics of filamentary discharge mode, taking into account the memory effect, was first developed in [10,11]. This fundamental model significantly improved the understanding of microdischarge pattern behavior in DBDs. The model is based on the assumption that the space charge of the microdischarge channel will decrease the electric field and prevent streamer formation in the vicinity of this channel. The plasma channel that forms after the streamer strike has a net positive charge because the electrons leave the gap much faster than ions. This residual positive charge influences the formation of the nearby streamers and therefore of the neighboring microdischarges (families of streamers). The mechanism of the influence is the following: positive charge intensifies the electric field in the cathode area near the channel and decreases the electric field in the anode area. Since the avalanche-to-streamer transition depends mostly on the electric field near the anode, where a new streamer originates from, the formation of neighboring streamers and microdischarges is actually prevented, and so, microdischarges effectively repel each other. The quasi-repulsion between microdischarges leads to the formation of short-range order that is related to a characteristic repulsion distance between microdischarges, and in some cases, it results in microdischarge self-organization into regular patterns.

Observation of this cooperative phenomenon depends on several factors, including the number of the microdischarges occurring and the operating frequency (which affects the strength of *memory effect*). For example, when the number of microdischarges is not large enough, then no significant microdischarge interaction is observed. In all cases, filaments stay separated at least by a distance corresponding to the length scale of the microdischarge interaction. If the applied electric field is high enough, it will cause microdischarges to develop in all of the unoccupied spaces, so that the discharge area becomes completely filled, and as all of the microdischarges are separated by approximately the same distance, regular patterns of microdischarges are formed.

Before describing the experiments that were performed at Eastman Kodak Company [10,11], it is reasonable to talk about the patterns imaging. Various techniques for imaging microdischarges have been described in the literature, one of the oldest methods being the Lichtenberg method [12,13]. Photographic images generated by this technique are known as Lichtenberg figures. In this method, light emitted from a microdischarge is imaged using a piece of photographic film, which may be placed underneath a transparent insulator such as glass or quartz, to record the plasma's optical emission, or it may be exposed directly to the discharge. The information obtained from such images includes the physical dimension of the "footprint" of the microdischarge, as well as an estimate about microdischarge density in terms of strikes per unit area. Images of electric phenomena, regardless of how they are generated, are generally referred to as Lichtenberg figures. The work reported in [10,11] demonstrates the use of imaging plates containing photostimulable phosphor for characterization of DBDs in air through the use of the Lichtenberg-type figures. Photostimulable phosphors are also known as "storage" phosphors. This unique class of inorganic materials has the remarkable property of "storing" the energy from ionizing radiation in the form of radiation-induced lattice defects, whose energy can be lib-

erated by selective photostimulation. So, this type of unique storage phosphor imaging plate was used to detect streamer “footprints”. The plate was placed on the electrode to be directly exposed to DBD plasma, and the result is shown in Fig. 2a. The sharper black dots correspond to streamers that occur repeatedly in the same location (i.e., microdischarge). A very important feature that can be easily seen on the picture is that the microdischarges (black dots) are surrounded by white areas, which represent the absence of the streamer “footprints” in the vicinity of microdischarges. This leads us to the conclusion that there are locations in the discharge where no streamers can strike (leave a “footprint”), these “prohibited” places being mostly located around the microdischarges. The analysis of the experimental imaging plates brought us to our idea regarding microdischarge interaction.

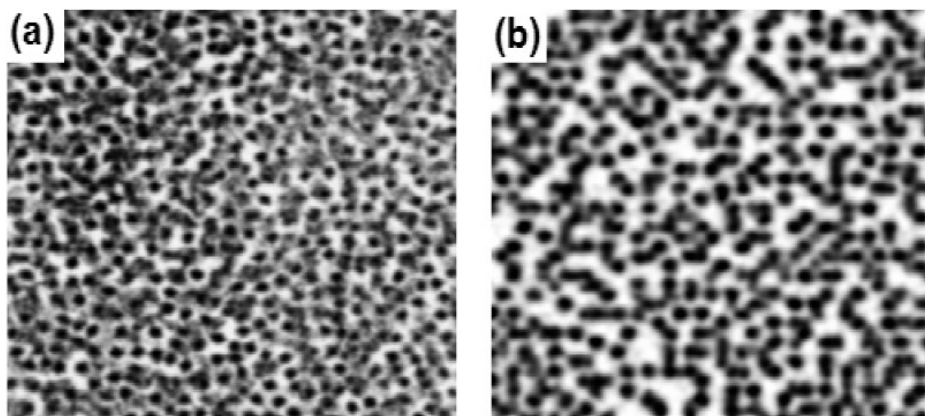


Fig. 2 Streamer “footprints” images in a DBD gap in air for 10 excitation cycles at 20.9 kHz and a discharge gap of 0.762 mm: (a) original experimental image; (b) result of simulation (enlarged).

To validate this idea, the extended cellular automata (CA) [14] model of DBDs was employed. In this model, the area of influence of each streamer was calculated, based on the assumption of microdischarge interaction. The position of each streamer was calculated using a Monte Carlo process which takes into account the areas of influence of all previous streamers. Thus, the position of any given streamer will not be random because it depends on the location of the previous streamers. The stronger the effect of microdischarge interaction, the less random streamer strikes will be. The strength of this effect is seen from Figs. 3b and 3c, where we compared two identical cases with and without microdischarge interaction. Figure 3 represents Voronoi tessellations of microdischarge patterns. The Voronoi tessellation is an elegant way to show the area of the influence of each microdischarge. The influence of microdischarge is cumulative influence of all streamers that constitute the microdischarge. On a Voronoi tessellation, the shades of gray represent the number of the sides in each cell. Most of the cells in the experimental picture (Fig. 3a) and the simulated one (Fig. 3b) have six sides (hexagonal). The hexagon patterns were also observed in an air–argon mixture [15].

Thus, this interaction between microdischarges appears to be a strong effect and a dominant feature of the relatively powerful DBD, where regular microdischarge patterns were observed experimentally and also were simulated.

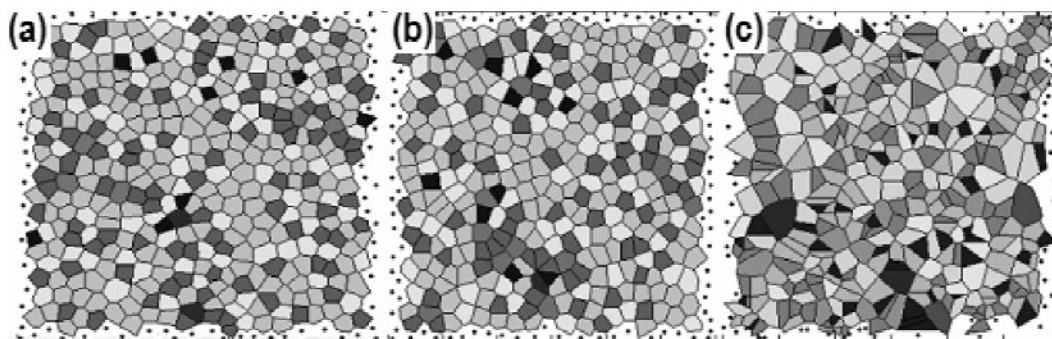


Fig. 3 Voronoi tessellations of the microdischarge patterns: (a) for experimental image (Fig. 2a); (b) for simulated image; (c) for the random microdischarge distribution. Cells are colored to the number of sides. The cells on the images obtained experimentally and after simulation are mainly six-sided cells.

HOMOGENEOUS AND NONHOMOGENEOUS DBD MODES

Barrier discharges can exhibit two major discharge modes: streamer (with bright microdischarges) and glow. The glow mode of DBDs is not necessarily uniform, but can be filamentary also. These glow filaments are formed by avalanches rather than streamers (microdischarges are initiated by Townsend, not a streamer breakdown) [16,17]. The glow mode by DBDs is usually referred to as the APG (this term was introduced by S. Okazaki [18]). The filamentary mode was a topic for active investigations in the past several years. Much experimental and theoretical work has been done in this area. Most industrial DBD applications utilize filamentary discharges because the specific power of filamentary discharge is typically higher [19–21]. However, for homogeneous treatment of surfaces, or for the deposition of thin films, the glow mode of discharge has obvious advantages over the filamentary one. Glow discharges with average power densities comparable to those of filamentary discharges will find a lot of applications, if reliable control could be achieved.

Organization of the APG discharges in DBDs is one of the most challenging problems. The APG was first observed and empirically analyzed by S. Okazaki [18]. This work immediately stimulated enormous interest and found numerous practical applications. However, the fundamental understanding of the phenomenon is still elusive, although some progress in the analysis was made [22].

Unfortunately, it is still difficult to reliably control homogeneous glow discharges at atmospheric pressure, especially in electronegative gases. For instance, changes of the electrode configuration, or small variations of the amplitude or repetition frequency of the applied voltage, or changes in the gas composition, can cause a transition into a more stable filamentary discharge mode. For industrial applications, this could be a severe drawback compared to filamentary discharges. Lack of fundamental understanding of the glow discharge mode makes its stabilization and control over the process difficult.

In order to provide robust control of the behavior of the DBD and the transition from the filamentary to the glow mode, a theoretical model should be developed. Several research groups are now actively working on development of a theoretical model of DBD that will allow understanding and predicting transition from glow to filament mode.

Most of the APGs were implemented in helium, or in other nonelectronegative gases. The real challenge is to sustain APG in electronegative gases without the presence of helium, for example, in air. It is interesting that S. Okazaki was able to successfully operate the APG in the air using a special electrode structure, but the physical explanation is still missing. Understanding of the physics of the APG organization in electronegative gases, and first of all in the air, will result in the optimization of the discharge procedure and will undoubtedly lead to further applications of the discharge.

DBD CHEMISTRY

DBD is the most popular nonequilibrium atmospheric pressure plasma system used for chemical synthesis. For more than 100 years, it's been used for industrial production of ozone (O_3) from air or oxygen, and because the more efficient technology has not been discovered yet while the need for ozone is constantly rising, DBD still remains in a leading position among plasma-chemical reactors.

From the plasma chemistry point of view, the most important characteristic of DBD is dynamics of electron energy distribution function (EEDF) in microdischarge. The electric field in the streamer head can be much higher than the applied electric field, making streamers in DBDs effective in production of high-energy electrons. Plasma chemistry kinetic processes in DBDs are initiated by direct electron impact during streamer strike. The effectiveness of such processes in the discharge (e.g., direct electron impact dissociation) determines the efficiency of plasma chemical synthesis.

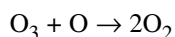
Synthesis of nonorganic compounds in DBD plasma

Numerous attempts were undertaken to use DBDs for synthesis of various nonorganic compounds, most of them did not succeed in economically feasible processes. The challenges with all plasma discharges are low production rates and high energy cost compared to the classical chemical synthesis technology. For instance, it is possible to use DBDs for synthesis of ammonia $N_2 + 3H_2 \rightarrow 2NH_3$, but a classical catalyst-based process (Gabor process) is more efficient. However, in the case of ozone synthesis, very high efficiency in DBDs can be achieved.

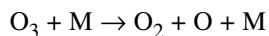
The mechanism and kinetics of ozone electrosynthesis have been deeply studied, and by now a huge amount of experimental data containing the dependence of O_3 production as a function of pressure, temperature, inlet mixture composition, and electrical parameters has been collected [23]. Nevertheless, in spite of a large amount of experimental data and numerical modeling attempts, the researchers still cannot develop a fully adequate theoretical description of O_3 synthesis and substitute complicated and time-consuming experiments by simulation model for parametric study and optimization of plasma chemical process. The chemical kinetics in microdischarge channels is rather complicated, even in the simplest case of oxygen as a working gas, the reaction mechanism contains up to 100 elementary reactions, including reactions where the constant rates depend not only on gas temperature, but also on electric field. However, if we limit ourselves to consideration of only the reactions which define O_3 formation and destruction rates, we can choose a relatively small amount of elementary reactions. The formation of O_3 molecules in microdischarge in oxygen and air happens mainly due to the following reaction:



(where $M = O_2$ or $M = N_2$). And reactions responsible for O_3 destruction are:

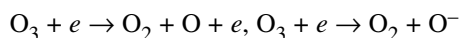


(destruction in microdischarge channel)



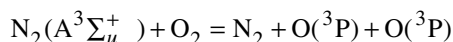
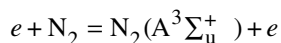
(where $M = O_2$, $M = N_2$, or $M = \text{wall}$; homogeneous or heterogeneous destruction in the reactor volume and on the surface of the electrodes).

Constant rates of the last two reactions greatly depend on temperature, and, therefore, effective cooling of ozonators is a necessary condition for obtaining high O_3 yield. If the concentration of O_3 reaches high values (more than 1 % volumetric fraction), the electron-collision reaction becomes important and should be added to the reactions responsible for O_3 destruction:

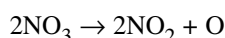
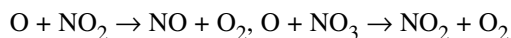
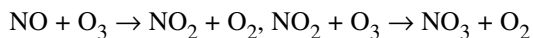


(destruction in microdischarge channel).

Ozone synthesis from air is a more complicated process than from oxygen. The presence of nitrogen results in decreased reaction rate of direct electron–oxygen collision reaction. On the other hand, owing to the presence of excited molecules of N_2 , another additional channel of O_2 dissociation takes place and cannot be neglected:



It is interesting that the contribution of this channel in O-atoms generation is comparable with O_2 dissociation by electron collision. Also, the appearance of essential concentration of NO_x (i.e., hundreds of ppm) in DBD plasma results in a dramatic drop in O_3 yield. The reason of this drop is the existence of catalytic cycles of destruction, for example:



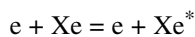
That is why O_3 synthesis from air takes place in the conditions providing minimal NO_x yield (i.e., in dry air with relatively low specific discharge power and effective cooling).

Thermodynamic energy cost of one ozone molecule is 1.5 eV (for the process $3\text{O}_2 \rightarrow 2\text{O}_3$), that corresponds to production rate of ozone 1.2 kg/kW·h. Practical ozonators that operate in pure oxygen can have maximum production rate as high as 0.4 kg/kW·h (4.4 eV/molecule or G factor is 22); in air maximum achievable rate is 0.2 kg/kW·h [24,25].

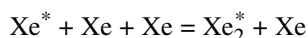
Excimers formation in DBD plasma

Interest in the excimer synthesis in DBD plasma has been increased mostly due to the excimer-laser development, which provides the generation of UV radiation. Developed on the basis of DBD technology, excimer lamps are able successfully to substitute lasers in all corresponding technologies where the coherent radiation is not required. Excimer lamps are easy to manufacture, they do not require an expensive power supplies and are able to provide high radiation intensity which can be easily controlled by changing frequency of the applied voltage.

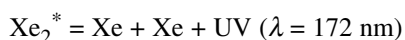
A typical example of excimer synthesis and generation of radiation in DBD is related to the formation and destruction of Xe_2^* (where Xe is the working gas). This process takes place in microdischarge channels and consists of three consequent stages:



(direct electron-impact excitation)



(excimer formation)



(excimer destruction and generation of UV radiation).

As was shown experimentally, the reaction rates of these three reactions are so high that the duration of radiation generated by single streamer does not exceed the corresponding duration of current

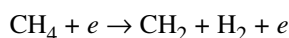
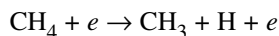
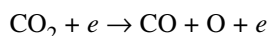
pulse, which is on the level of several ns. The efficiency of an excimer-laser is less than 1–2 %, while the efficiency of excimer DBD lamps can reach 10–15 %.

SYNTHESIS OF ORGANIC COMPOUNDS

The first attempts to apply DBDs for synthesis of organic compounds were performed in the 19th century when the reactions of dehydrogenation and polymerization of hydrocarbons (methane, ethane, ethylene), formaldehyde synthesis, formic acid and complex oxygen-containing organic products (from mixtures H₂+CO₂ and H₂+CO) were produced in DBDs. Since then, many attempts were made to find a worthwhile DBD application for different organic syntheses. However, the results of extensive research in this direction still cannot offer any industrial-scale technologies of organic synthesis in DBDs which are able to compete with already existing chemical methods of organic synthesis (usually catalyst-based). There are two main plasma chemistry challenges that limit use of DBDs for organic synthesis:

- complexity of chemical reaction mechanisms with a high number of parallel channels resulting in relatively low selectivity;
- low product yield per unit energy. For example, in formaldehyde synthesis (from mixture H₂+CO) in DBD, the maximum product yield is only 15 g/kW·h.

Even for the simple mixture (CH₄+CO₂) plasma chemical kinetics in DBDs can be quite complex [26,27]. Reaction mechanism initiated by dissociation of CH₄ and CO₂ in streamers.



After initiation chemistry proceeds in different concurrent directions: polymerization (formation of higher hydrocarbons), dehydration (formation of unsaturated hydrocarbons like alkenes, alkynes (ethylene, acetylene), hydrocarbon oxidation, and surface interaction (polymer films formation).

INTERACTION OF CHEMICALLY ACTIVE PLASMA WITH POLYMER MATERIALS: SURFACE MODIFICATION

Usually, polymer materials have low values of surface energies, low wettability, and low adhesion. One of the most perspective and advanced methods of polymer surface modification is a low-temperature plasma treatment. DBDs were used for polymer surface modification since the 1970s. This treatment allows altering considerably the properties of polymer surfaces and extending the areas of their applications [3,4].

Ecologically clean, modern low-temperature plasma methods have considerable advantage over the chemical modification techniques that require using aggressive agents such as acids, hydroxides, alkaline-earth metals, and their derivatives.

The most important feature in plasma-chemical polymer modification is that only the surface and very thin near-surface layer (from 100 Å and up to some μm) are subject to modification. The rest of the polymer remains unchanged, retaining mechanical, physical, chemical, and electrophysical properties of the original material.

FUTURE TRENDS OF DBD APPLICATIONS IN PLASMA CHEMISTRY

Strong thermodynamic nonequilibrium and simple design—these distinctive properties of DBDs allow us to hope for expansion of its applications in low-temperature atmospheric pressure plasma chemistry.

DBDs have a big potential to be a prospective technology of exhaust cleaning from CO, NO_x, and volatile organic compounds [28]. The successful use of DBDs reported in recent research on plasma-assisted combustion may result in new applications [29]. Also, the applications of DBDs to the problems of cleaning and etching in microelectronic fabrication are in sight.

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