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REVIEW ARTICLE

Barrier Discharges in Science and Technology Since 2003: A Tribute and Update

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Abstract
An update to the article "Dielectric-barrier Discharges: Their History, Discharge Physics, and Industrial Applications" by Ulrich Kogelschatz from 2003 is given. The research and applications of barrier discharges of the last decades are summarized. In particular, the latest developments in ozone generation, radiation sources, environmental applications and surface treatment are discussed. Topics, which appeared with growing attention after 2003, such as plasma medicine, carbon dioxide chemistry, liquid treatment and airflow control, are also summarized to provide an outlook into the coming years.

It can be stated, that this type of gas discharge is still of high scientific and technological relevance. Its wide range of applications made the research more inter- and cross-disciplinary while modern diagnostic and modeling enabled deeper insights in the complex physical and chemical processes. In this sense, the contribution of Ulrich Kogelschatz, who introduced and inspired several generations of researchers in the field, cannot be overstated.

Keywords Nonthermal plasma · Dielectric barrier discharge · Plasma technology · Ozone · Environmental applications · Surface treatment · Plasma medicine

Introduction

The article "Dielectric-barrier Discharges: Their History, Discharge Physics, and Industrial Applications" by Ulrich Kogelschatz in 2003 [1] was one of the most comprehensive reviews about this type of gas discharge at that time and has received an overwhelming reception

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since then. In 2003, the author looked back on a more than 30 years of work devoted to low-temperature atmospheric pressure plasmas at the Brown, Boveri & Cie (BBC) research center in Baden, Switzerland. BBC became ASEA Brown Boveri (ABB) in 1988 through a corporate merger. Together with his long-time collaborator Baldur Eliasson he was involved in the development of electrostatic precipitators (based on corona discharges) before he started to study the fundamentals of Dielectric Barrier Discharges (DBDs), which led to the development of ozone generators in the 1980's and excimer lamps in the 1990's. The article is one of several excellent reviews by Ulrich Kogelschatz about this subject. It can be seen as one of the starting points for the book "Non-Equilibrium Air Plasmas at Atmospheric Pressure" edited by him together with Kurt H. Becker, Karl-Heinz Schoenbach and Robert J. Barker [2]. Published in 2005, it was one of the first monographs that paid tribute to DBDs and their multi-faceted applications.

It is noteworthy to state that at the end of the 1990's there was an increasing interest in atmospheric pressure plasmas and in DBDs in particular. Kogelschatz describes this as a "renaissance" of an "in principle rather old gas discharge", triggered by the increased market penetration of DBD-based industrial processes, the availability of modern, cost efficient and well-matched power electronics, and a better fundamental knowledge about discharge formation and the ensuring plasma chemistry, enabled by the progress in modeling, simulation, and diagnostic. He also predicted: "No doubt, this trend will continue" and "these recent achievements will open roads to additional new applications".

In fact, the interest in DBDs and related plasma sources has been even increasing since that time. The topic of "microplasmas" developed in the mid-1990's. Further applications like aerodynamic flow control and analytic detection devices as well as life science applications, gas cleaning and the deposition of tailored layers of materials with specific properties have been studied. All areas are continuously growing. Even the conversion of greenhouse gases such as carbon dioxide and methane - also in combination with catalysts - experienced a revival since 2010 in the context of power-to-gas and power-to-fuel technologies. Plasma catalysis is a highly prominent research topic in the low temperature community today. Furthermore, the generation and modification of nanoparticles is under research and DBD plasma sources and applications explore agriculture now.

The 20th anniversary of the article by Kogelschatz to be celebrated with a special issue in this journal, is an occasion for us to look back on the research and applications of the last decades and to provide an outlook into the coming years. This paper is not intended as a classical review paper, but rather to provide an overview of the developments in the field. Therefore, most citations refer to review articles or summary papers rather than the original literature. The reader is kindly referred to the reference list of these publications for further information.

History and Fundamental Understanding of DBDs

In fact, already in 1777, Lichtenberg generated gas discharges against the surface of insulators to create the famous dust figures, later named "Lichtenberg figures" after him. The article by Kogelschatz mentions von Siemens as the inventor of the first discharge device, the ozoniser in 1857 [3]. However, it has come to light recently that du Moncel already used a DBD a few years earlier [4]. Du Moncel reported several experiments with so-called

Ruhmkorff coils (nowadays known as induction coils) as an AC power source. In some of his studies, he used planar metal electrodes covered by glass plates and separated by a small air gap for gas discharge generation [5, 6]. It is not known whether von Siemens, a German engineer, was aware of these experiments (published in French). In any case, he does not cite this work, his device has a completely different geometry and his paper focuses exclusively on ozone generation. A little bit later, in 1894 Tesla patented an "inert gas discharge tube" [7], which become the basis for the invention of the "plasma globe" by Falk and Parker [8]. These plasma globes and also the "crackle tubes" are low pressure DBDs and have been sold in millions since the 1990's.

In fact, ozone generation was the main application of DBDs for many decades to follow. Therefore, this type of discharge was known as the "ozoniser discharge" and was also commonly named "silent discharge". This term was introduced by Andrews and Tait in 1860, because it creates significantly less banging noise than sparks. This term was used until the 1990's. The first textbook exclusively dedicated to DBDs "Physical Chemistry of the Barrier Discharge" by Samoilovic, Gibalov and Kozlov from 1989 (1st edition in Russian) and 1997 (2nd edition in English) focusses on ozone synthesis and mentions that Jeromin used the term "barrier discharge" ("discharge having dielectric barrier") in the 1960's [6]. The first attempts to use DBDs for other chemical syntheses than ozone can already be dated back to the 1870's. For example, Brodie et al. demonstrated that a DBD in a mixture of either carbon dioxide or carbon monoxide and hydrogen forms formaldehyde and methane [9].

In the 1950's, Eisby invented the surface activation of plastics, commonly called "corona treatment", although most of these devices are based on DBDs in air [10]. Simultaneously, DBDs were also investigated for deposition applications, surface activation and the treatment of textiles.

For ozone generation and surface activation, the studies of DBDs were mostly devoted to air, oxygen or other molecular gases. This changed significantly in the 1960's with the development of the early plasma displays and DBD-based lamps, where noble gases, pure or with molecular admixtures, were used. In the 1970's, the CO_2-laser was developed and later many studies of removal of contaminants from polluted air (e.g. volatile organic compounds, nitride oxides, greenhouse gases) were conducted as well. Furthermore, the importance of so-called partial discharges in insulated high voltage systems, in fact special, "undesired" DBDs, motivated more and more research from the 1960's onward [11]. This work is still ongoing and demanded due to the trend towards smaller insulated systems and circuits. All developments and research activities mentioned above happened (more or less) in parallel and in slightly different communities. Generally, the interest in atmospheric-pressure plasma has increased and a wide variety of such plasma sources exists today [12].

In the last 20 years, DBDs appeared in a much wider range of applications [13–16]. After 2000, more and more activities focusing on plasma decontamination were carried out [17, 18]. Plasma jets, some of them using the DBD concept [19–21], became very prominent plasma sources and found particularly broad acceptance in the new field of plasma medicine [22, 23]. At the same time, DBDs found their way into miniaturized analytical devices, e.g. as ionization sources [24]. Since plasmas are a source for oxidizing species, they enable the breakdown of organic and inorganic compounds in water. Pulsed plasmas are the main method for generating plasmas in liquids, but DBDs with liquid electrodes are considered also [25]. After 1995, microplasmas, e.g. gas discharges where at least one spatial dimension is in the submillimeter range, started to gain prominence [26]. These microplasmas include microarcs and –sparks, laser generated plasmas, microhollow cathode discharges, guided ionization waves in plasma jets, microwave-sustained microplasmas, but also single filament (diameters of about 100 µm) or microscopically DBDs, either as single spots or in regular arrays [27–30]. In so-called gas confined DBDs, an insulating gaseous layer is used as the barrier instead of a solid or liquid dielectric. Therefore, a gaseous layer with a relatively lower breakdown field strength (i.e. argon, helium) is confined by one or two gaseous layers of a gas with a higher breakdown field (i.e. air, carbon dioxide) [31]. The plasma formation appears in the gas with the lower breakdown field while the other gas still acts as the insulator.

The Fig. 1 summarizes some selected milestones of DBD research, most of them already mentioned in Kogelschatz's article [1]. As shown and already mentioned there, in the 1930's researchers became aware of the filamentary nature, i.e. the plasma consists of many individual ionized channels. The study of single filaments, which are mostly formed by the streamer breakdown mechanism [32, 33], was a very active research subject in the last 30 years [34, 35], and it still is. These investigations require special electrode arrangements, e.g. semi-spherical dielectric bodies with embedded electrodes, pin-to-plate electrode configurations, or pins embedded in an insulator as single coplanar DBDs. Such geometries enable sufficient localization and stabilization for dedicated studies. These studies were made possible because of sensitive and fast imaging and spectroscopic devices such as intensified charge-coupled device (ICCD), streak cameras and time-correlated single photon counting [36–38] as well as fast current measurements [39], optical emission spectroscopy and laser diagnostics [40–45], and molecular beam mass spectrometry [46–48]. Furthermore, measurements of surface charges on the dielectric barriers by the Pockels-effect succeeded [49–55]. Today, we have a much better understanding of the discharge formation. The detection of key species such as radicals (e.g. O, OH, H, N), metastable excited species (e.g. $N_2(A)$) and ionic species (pure and hydrated cations and anions) and their physico-chemical kinetics, and the measurement of the reduced electric field strength are possible with established methods today [56].

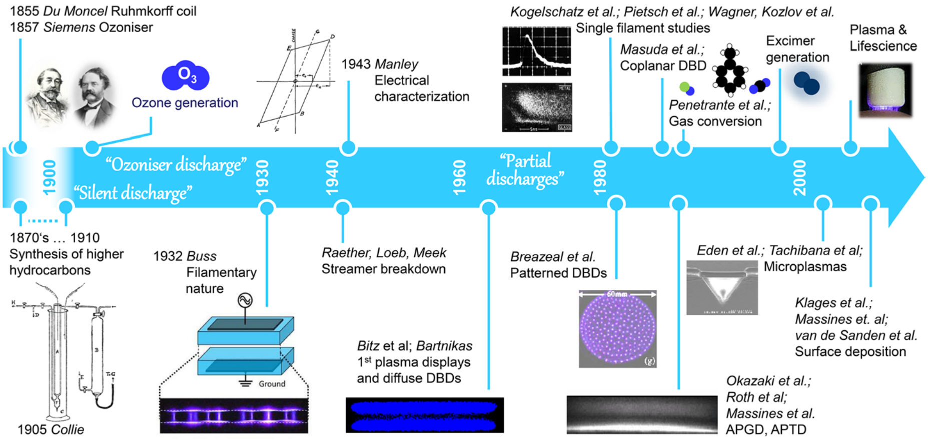

Fig. 1 Selected milestones of research on DBDs. Parts of the figure are publicly available or from references [62, 71–78]

In particular, we have gained a much better knowledge of the role of volume and surface memory effects on the phenomenology and breakdown mechanism of DBDs. Pre-ionization provided by residual species in the volume or surface charges on the dielectrics influence the breakdown mechanism significantly. Even non-filamentary DBDs, i.e. plasmas covering the electrode cross section almost entirely and uniformly, the so-called diffuse DBDs, were obtained. The first diffuse discharges were obtained in the 1960's during the research of plasma displays by Bartnikas [57] and later by Okazaki et al. [58, 59] and Massines et al. [60–63] for surface treatment applications. In the same context, distinct regular and irregular discharge pattern were studied, e.g. by Wertheimer et al. [64, 65] and Stollenwerk and Purwins et al. [66–68] and reviewed by Kogelschatz in 2002 [69] and 2010 [70]. Today, researchers have succeeded in generating them in a stable and reproducible manner and the insights into how streamer formation can be inhibited has been gained. However, the mechanisms are complex and their role is determined by the specific conditions such as the gas composition, electrode geometry and barrier material properties. Penning-ionization, secondary electron emission by metastable species impact, associative ionization, electron detachment from negative ions, and electron desorption from the dielectric surface are all possible processes. Gaining deeper insights into the interplay of plasmas and surfaces in the different gas compositions is still an exciting research topic.

Progress in the field would not be possible without the numerous efforts afforded by plasma modeling and simulation. The models are able to calculate the temporal behavior of the electric field distribution in the discharge gap coupled with the dynamics of the charged species. Different kinds of modeling approaches, e.g. fluid models, Monte Carlo simulations and particle-in-cell models, also in combination, are used [79–82]. In particular, the understanding of streamers as rapidly developing, multi-scale phenomena is at the cutting edge of current research efforts [83, 84].

The review article of Kogelschatz 2003 also addressed the electrical characterization of DBDs, an approach based on the work of Manley in 1943 [1, 71]. For many purposes, it is still sufficient to describe the discharge by overall quantities such as the applied frequency f, the applied voltage amplitude V_0 and the mean discharge voltage U_D in the gap, at which discharge activity is obtained. Since 2003, there have been a number of further developments on this aspect, which become necessary when discharge geometries deviate from the classical arrangement in ozone generators. In particular, surface DBDs, volume DBDs with non-uniform discharge gaps and packed bed reactors desire an extension of the classical equivalent circuit model. Furthermore, the approach has been extended from a purely macroscopic view to the characterization of individual filaments or structured discharge regimes [85].

The simplest electrical equivalent of a DBD is a lumped element equivalent electric circuit consisting of two capacitances, resembling the barrier and the gas gap (C_d and C_g), respectively. Its linear arrangement results in a total capacitance C_{cell}. The applied voltage amplitude V_0 must exceed a certain threshold V_{min} to cause discharge ignition. A time-dependent current source or resistor $R(t)$ in parallel to the gap capacitance then represents the discharge [86–88]. The investigation of voltage-vs.-charge (V–Q) plots does not require high bandwidth probes and oscilloscopes, in contrast to the recording of current waveforms. Several quantities can be obtained from the V-Q plots, namely V_{min}, C_{cell}, the effective dielectric capacitance ζ_d and the total plasma energy per high voltage period W as well as the plasma power P (from the area of the V-Q plot).

The equivalent circuit shown in Fig. 2 (a) is an extended version, which takes into account parasitic capacitances [89] as well as the partial surface discharging effect [90, 91]. Parasitic or stray capacitances, C_{par}, are caused by surrounding capacitances (e.g. cables, high voltage throughputs) or additional dielectrics in the discharge gap (e.g. packed bed particles, spacers). Its presence is practically unavoidable, but they are often overlooked in the interpretation of *V-Q* plots. The partial surface discharging effect appears at low over voltages ($V_0 \approx V_{min}$), in non-uniform discharge gaps or in case of edge effects (i.e. local enhancement of electric field strength) on the electrodes. To account for this, the DBD arrangement is divided into a non-discharging areal fraction with the factor α and a discharging areal fraction with factor β (with $\alpha+\beta\equiv 1$).

Only the discharging fraction implements the resistor (or current source). The schematic *V-Q* plot next to the equivalent circuit in Fig. 2 (a) shows the impact of the additional capacitances, namely on the slopes of the parallelogram. This also affects the determination of the discharge voltage as given by the equations in the figure [92]. The derivation of the power equation for the extended equivalent circuit results in the following equation:

$$P = 4f\beta\left(C_d - C_{cell}\right)V_{min}\left(V_0 - V_{min}\right) \tag{1}$$

The equation contains only measureable parameters and includes the partial discharging factor β. The parasitic capacitance does not influence the plasma power. For $\beta = 1$ the Eq. (1) coincides with the Manley-equation as it is given in the review of Kogelschatz 2003.

In reality, *V–Q* plots can diverge from the parallelogram shape. In fact, a parallelogram is obtained only, when the voltage across the gap U_g is almost constant during the active subperiods of the discharge (namely at a well-determined value, U_D). For pulsed DBDs this is not entirely the case, since the applied voltage changes on the typical timescale of discharge inception (nanosecond range). The influence of experimental errors on the capacitances C_d and C_{cell} was discussed in detail for pulsed DBDs in [93]. Deviations from the parallelogram are also obtained when the barrier becomes conductive by heating of the plasma, pulsing action of the microdischarges or when the plasma expands gradually along the gas-dielectric interface as it is the case in surface and coplanar DBDs. In these geometries, the capacitances of the discharge arrangement strongly depend on the operation conditions (V_0 in particular), similar as in the partial discharging DBD model discussed above. The corresponding *V-Q* plots can show an almond-like shape in case of sinusoidal operation, as shown in the Fig. 2 (b), which is caused by the alternation of the reactor capacitance during the active discharge phase. In the equivalent circuit of the surface DBD proposed in Fig. 2 (b) again a parallel connection of capacitances C_g and variable resistors *R(t)* is employed, but here multiple identical and sequential elements consisting of C_g, *R(t)* and C_d describe the discharge expansion. For a particular applied voltage amplitude the whole expansion length of the discharge *x* consists of *n* basic elements, which are terminated by the capacitance *C'* [94]. The power consumption of surface DBDs is still a matter of debate. The equivalent circuit in Fig. 2 (b) results in a cubic relationship between power and voltage amplitude [94].

$$P \propto fV_0^2\left(V_0 - V_{ign}\right) \tag{2}$$

Other authors suggest a time-varying effective dielectric capacitance and a quadratic behavior between *P* and V_0 [95]. These examples demonstrate that electrical characterization is

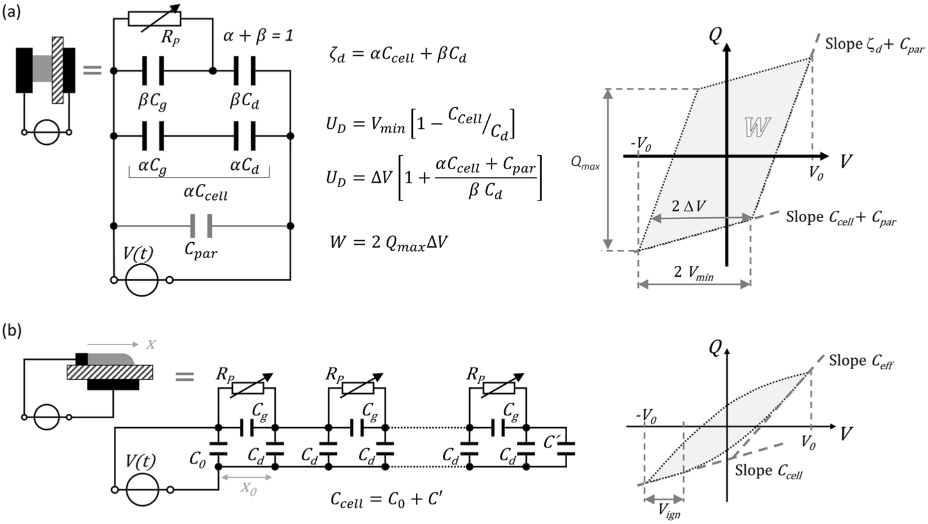

Fig. 2 Equivalent circuits and corresponding V-Q plots for **(a)** a partially discharging volume DBD and **(b)** a surface DBD.

still an active field of research because it provides useful insights for the design, optimization and control of DBD devices and reactors.

Ozone Generation

Ozone (O_3) is a strong oxidizing agent and widely used in applications such as purification of drinking water and industrial wastewaters, cooling towers, air purification of gas emissions from industrial plants, pulp bleaching for chlorine-free production of paper, organic and inorganic synthesis and other advanced oxidation technologies. It is applied in aquaculture and fish farming, and food processing. It is also used for surface treatment/cleaning, e.g., in the fabrication of semiconductors as well as in medicine and esthetics ("ozonotherapy"), surgery, dental care, optical care, manufacture of pharmaceuticals and beauty products. Besides electrolysis and UV lamps, corona discharges and DBDs are still the dominating technology [96]. The largest facility (as far as we know) is located in Texas and produces 800 kg/h for the purification of drinking water for 1.6 million customers [97]. The market for ozone generators is still growing and further growing is expected for the coming years. It should also be noted that ozone can damage the tissues of the respiratory tract, causing inflammation and irritation. Thus, human exposure to ozone may cause headaches, coughing, dry throat, shortness of breath, a heavy feeling in chest, and fluid in the lungs. Higher exposure levels can lead to more severe symptoms and chronic exposure may lead to asthma.

Ozone is formed in a 2-step process. The first step is the electronic dissociation of molecular oxygen in the microdischarge channels followed by a three-body collision of O, O_2, and a third molecule. High-pressure environments such as e.g. atmospheric-pressure DBDs, are required for three-body collisions to occur at a significant rate. When pure oxygen is used as the feed gas, the third collision partner is also an O_2 molecule. The time scale for O_3 formation in atmospheric-pressure oxygen is a few microseconds. The generated ozone then diffuses out of the volume of the microdischarge. An increase of ozone concentration is essentially the result of the action of a large number, typically thousands of microdischarges before the ozone reaches the end of the DBD reactor. Ozone is readily destroyed also by collisions and by UV photons. Ozone formation is affected strongly by the gas temperature. Ozone is not heat stable, but more important, a higher temperature slows down the three-body kinetics of ozone formation and increases the degradation kinetics, e.g. with NO_x formation in air. Thus, the efficient conversion of O_2 into O_3 requires an environment that maximizes O_3 formation while, at the same time, minimizes O_3 destruction. For example, in large-scale industrial ozone generators the discharge gap is kept small to enable an efficient heat removal and the electrodes are cooled down by water.

Plasma chemistry in air fed ozone generators is a little bit more complex. Electronic excited states of nitrogen not only contribute to oxygen atom formation (so-called Penning-dissociation), but also induce the formation of N_xO_y. Nitrous oxide, N_2O, results from collisions of nitrogen metastables, $N_2(A)$ with oxygen molecules during the early stages of microdischarge development while the intermediate species nitric oxide and nitrogen dioxide, NO and NO_2, are further oxidized to the highest oxidation state dinitrogen pentoxide, N_2O_5, under dry conditions. The raising of N_xO_y concentrations is called “discharge poisoning” as it leads to a significant break down of the O_3 generation at the discharge outlet.

Arguably, the ubiquitous use of ozone is in water treatment applications and DBDs have been by far the most commonly used ozone generators in these applications [98]. They represent a mature, fairly reliable and fairly economical technology that can be employed on a large industrial scale. However, the efficacy is not very high, about 20 vol% of ozone at the DBD exit, if pure oxygen is used as the feed gas and significantly less, if air is used. There are some remaining open science and engineering issues under investigation in an effort to improve the conversion efficiency. These include the effects of impurities in the feed gas mixture on the discharge performance as well as materials issues and by-product formation.

Up to about 20 years ago, the standard design of an ozone generator for water purification purposes was an assembly of long, single DBD tubes of uniform diameter. This design resulted in DBDs with filaments that were, more or less, distributed uniformly along the entire tube. Subsequent research revealed that the efficacy of ozone formation depends critically on the amount of charge transported by the microdischarge or by the energy density deposited in the microdischarge channels. The optimum ozonizer design provides the appropriate power induction to raise the amount of energy deposited in the feed gas to tailor the necessary number of microdischarges resulting in higher ozone concentrations, i.e. a tailored degree of discharge filamentation or density of microdischarges at low and high ozone concentrations along the discharge tube. This design concept, commonly referred to as “intelligent gap design”, in which a tailored degree of filamentation or the ability to control the number of microdischarges is obtained by segmenting the long discharge tube into 4 parts (Fig. 3). In the first segment, strong microdischarges quickly lead to the generation of about 80% of the final ozone concentration at the end of the tube. In the subsequent segments, weaker microdischarges will gradually increase the ozone concentration further, while minimizing the reactions that lead to the destruction of ozone. The control of the strength of the microdischarges is obtained primarily by changing the width of the discharge gap.

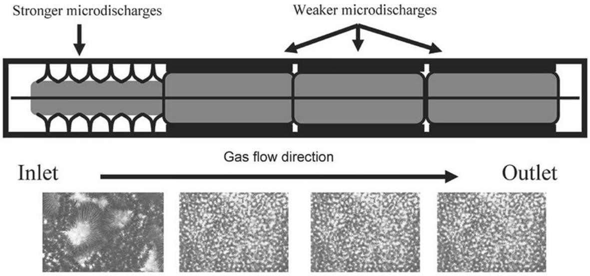

Fig. 3 Schematic of the Intelligent Gap System introduced by Degrémont Technologies (Ozonia), Ltd. in 2007 (not to scale) [98]. A short, high-load inlet region with a lower degree of filamentation is followed by long regions with a high degree of filamentation and weaker microdischarges. The degree of filamentation is illustrated by the microdischarge footprints (so-called Lichtenberg figures) shown at the bottom of the electrode centers

While the plasma physics of the DBDs in ozone generators is well understood, there are open issues around the plasma chemistry and, associated with it, the coupling of the plasma physics to the plasma chemistry. For instance, the impact of by-product formation in DBD-based ozone generators remains an active field of research, especially because of the increased use of lower grade liquid oxygen (LOX) containing higher trace concentration of hydrocarbons, in particular of methane, CH_4. A complex plasma chemical reaction sequence leads to the formation of nitric acid, HNO_3 instead of the above mentioned N_xO_y. It was observed that the amount of HNO_3 formed depends on the concentration of N_xO_y formed in the air fed plasma and the amount of CH_4 that is converted, but not on the O_3 concentration. The kinetics of by-product formation in a DBD plasma depends on three major factors: gas mixture, discharge operating conditions (gas pressure, gas temperature, mode and frequency of applied voltage, electrode arrangement, power density, etc.), and the electrode and dielectric materials.

The most comprehensive earlier experimental investigations into the kinetics of NO_x formation in air-fed, DBD-based ozone generators were carried out by Kogelschatz and co-workers in the 1980's [99, 100], 20 years before the publication of his 2003 seminal review [1]. One of the key issues was whether trace hydrocarbon contaminants significantly affect or change the plasma chemistry or if, in sufficiently low concentrations, they simply act as a "spectator" without impacting the plasma chemical reaction pathways that govern the ozone generation. Recent experimental work provides evidence that methane (CH_4) is more than a mere "spectator" because of the chemically reactive nature of its dissociation fragments (e.g. methyl radicals CH_3).

Two possible reactions leading to the formation of HNO_3 emerged from the earlier studies.

$$N_2O_5 + H_2O \rightarrow 2HNO_3 \tag{2}$$

$$NO_2 + OH \rightarrow HNO_3 \tag{3}$$

with the rapid radical reaction (4) assumed to be the main route to HNO_3 formation. Very recent experimental studies using electrical and optical analysis techniques conclusively showed that the main by-products measured in the gas stream emanating from an oxygen/nitrogen-fed DBD ozone generator were N_2O_5 and N_2O. Furthermore, higher concentrations of CH_4 admixtures were found to result in the almost immediate formation of droplets in the discharge tube in conjunction with the rapid deposition of a “dirt” layer on the ceramic coated electrode, a change in the operating characteristics of the ozone generator, and arcing inside the discharge tube. The exact location of the deposited “dirt” layer and its temporal growth pattern after ignition of the ozone generator yielded useful information about the kinetics of by-product formation. The analysis of the deposits in conjunction with spectroscopic data suggested two scenarios concerning the methane reactions in the discharge process, one for a feed gas consisting of pure O_2 and a CH_4 admixture and one for a feed gas consisting of O_2 admixed with N_2 and CH_4. Admixing small amounts of CH_4 to pure oxygen did not result in droplet formation and deposits in the first 1/3 of the length of the discharge tube. For the remaining 2/3 of the length of the tube, the deposition of the “dirt” layer was balanced by vaporization (via sputtering), which resulted in a thin and rather uniform layer of deposits. When N_2 was present, even in minute concentrations, deposits appeared on the enamel coated electrode very close to the entrance to the discharge tube. As before, CH_4 was converted and H_2O and CO_2 were formed, but in the presence of N_2, NO_x compounds were formed and HNO_3 was produced. Vaporization via sputtering was insufficient to remove all deposits and a permanent “sticky” layer containing HNO_3 begins to form and grow on the ceramic coated electrode. Its thickness increased along the length of the electrode. Once the deposit dries, there were no other means to restore the electrode than by physical cleaning. The amount of N_2O_5 formed was found to depend on only two parameters, the ozone concentration and the amount of the nitrogen admixture. In the presence of hydrocarbons such as CH_4, the by-products differed significantly in oxygen-fed generators as compared to the by-products of oxygen/nitrogen mix-fed generators. The first case produced just humidity, the second one HNO_3.

An interesting observation on ozone generators supplied with high-purity oxygen is the so-called zero ozone phenomenon, a temporal decrease in the ozone concentration at its outlet under constant high discharge power operation. The most recent explanation for this is a thermal decomposition of the produced ozone at the electrode surfaces. The increase of electrode surface temperature is related with the oxidation and powder formation at the stainless steel electrode. The addition of trace of nitrogen avoids this phenomenon as its supports the formation of a passivation layer on the surface [101].

Although a rather mature application, large-scale ozone generation using DBDs has continued to progress and improve as a technology throughout the years. One of the reasons for this is the emergent global demand for environmentally safe and sustainable water treatment technologies due to the increasing global population. The continued drive to better understand the fundamental physics of microdischarges and the plasma chemical reactions will continue to improve this technology.

The development of cost and energy efficient power electronics coupled with the electrical engineering of matching power supply units to tailor DBD properties will also aid further developments. Future progress in large-scale ozone generators will come from ultrashort nanosecond high voltage pulses, advanced dielectric materials, and the continued better understanding and control of ozone producing microdischarges.

Environmental Applications and Greenhouse Gas Conversion

The decomposition of nitrogen oxides (NO_x) and sulphur oxides, volatile organic compounds (VOCs) and other hazardous air pollutants (e.g. hydrogen sulfide, ammonia) by DBDs is an active research topic since more than 40 years [15, 102–104]. Beside chemical compounds, nonthermal plasmas and DBDs can attack other contaminants such as particulate matter, microorganisms and viruses. The efficacy and selectivity of the chemical processes are affected by the humidity, temperature, and the chemical composition of the gas mixture. Due to the increasing environmental standards, there is still need for exhaust as well as indoor air cleaning technologies. Therefore, the research resulted in pilot- and large-scale installations. In particular, the application to large industrial gas flows is realized for the deodorization, e.g., in food production, where gas flows up to 80,000 m_n^3/h are treated [105]. Due to the low ionization degree and plasma density in DBDs the technology is limited to slightly contaminated gas flows (i.e. inlet concentrations $<$ 1000 ppm). However, this makes them attractive for the indoor air cleaning and air hygiene.

The degradation of chemical compounds in DBDs is based on the collision of the pollutant molecules with free radicals and ions generated in the discharge. UV radiation in air DBDs is quite low and therefore photolysis cannot play a significant role. If oxygen and/or water vapor are present in the gas, oxidizing reactions are dominant. Organic molecules, such as hydrocarbons also react with the oxidizing species formed in the plasma, e.g., O, OH, HO_2, O_3. Radical chain reactions involving hydrocarbon-radicals lead to efficient removal and can also enhance the oxidation of NO to NO_2. These reactive species can diffuse outside of the microdischarge channels. Thus, the chemical processes are not spatially restricted to the places where the polluted gas is exposed directly to the active plasma and, so-called indirect processes can also be used. Indirect treatment means, that only clean air is plasma treated and then injected into the polluted gas. This approach is successfully used for deodorization and so-called low-temperature oxidation for the removal of pollutants from waste gas streams from boilers and furnaces. In the latter, the non-water-soluble compounds (e.g. NO and NO_2) are converted to soluble ones (e.g. N_2O_5) by reactions with ozone end then scrubbed or precipitated. Gas streams with up to 600,000 m_n^3/h are treated and more than 16,500 tons of NO_x per year are removed in the USA nowadays. Captured nitrogen oxides result in dilute nitric acid (HNO_3), a valuable compound for the production of fertilizers. This example shows that a non-thermal plasma can be combined with other processes, enabling several synergies. Another example is the plasma enhanced selective catalytic reduction. Here, NO_2 formed from NO in the plasma is converted into solid salt particles, which can also be used as fertilizers, by the addition of ammonia and a catalyst. Other combined processes are adsorption, absorption, catalytic reactors, and biological treatment. In particular, with catalysts there are several synergies possible [106, 107].

The principles and characteristics of plasma-based air cleaning and depollution are not only suitable for large facilities in industry. Small and compact systems for the deodorization of exhaust gas or recirculating cooking stove air have entered the market. Systems for gastronomy as well as private households are established. Furthermore, indoor air cleaning devices are commercialized already. In these applications the DBD is often combined with an active carbon filter. These filter elements destroy the ozone, but also adsorb hydrocarbons. Then ozone can lead to the chemical removal of the adsorbate [108, 109].

The DBD configuration inspired the development of the Capillary Plasma Electrode (CPE) discharge (Fig. 4 (a)). While the CPE discharge design looks very similar to a DBD, the novel design employing capillaries in the dielectric that covers one of both electrodes results in basic properties and an operating principle quite different from a DBD [110]. The CPE discharge exhibits a mode of operation not observed in DBDs, the so- called "capillary jet mode". Capillaries with diameters from 0.01 to 1 mm and length-to-diameter ratios of about 10:1 serve as plasma sources, which produce jets of high-intensity plasma at atmospheric pressure under certain operating conditions. The plasma jets emerge from the end of the capillaries and form a "plasma electrode" for the main plasma in the space between the dielectrics and a stable uniform discharge can be achieved for the right capillary geometry, dielectric material, and exciting electric field. The placement of the tubular dielectric capillaries in front of the electrode(s) is crucial for the "capillary jet mode".

The CPE discharge displays two distinct modes of operation when excited by pulsed DC or AC, a diffuse mode below a critical frequency and/or input power and a mode above that frequency/input power where the capillaries "turn on" and intense plasma jets emerge from the capillaries. This transition is manifested in a drastic increase in the electron density as shown in Fig. 4 (b). When many capillaries are placed in close proximity to each other, the emerging plasma jets overlap and the discharge appears uniform. This "capillary mode" has been characterized in a rudimentary way for several laboratory-scale research discharge devices in terms of its characteristic electric and other properties: peak discharge currents of up to 2 A, current density of up to 80 mA/cm^2, E/p of about 0.25 V/(cm. Torr), electron density n_e above 10^{12} cm^{-3}, power density of about 1.5 W/cm^3 in He and up to 20 W/cm^3 in air.

The CPE discharge was used in a variety of applications. Here we mention the destruction of environmental contaminants present in trace concentrations in respirable atmospheres [111]. Aliphatic and aromatic VOCs, such as n-heptane, pure toluene, and a BTEX mixture of benzene, toluene, ethylbenzene, and xylene at concentrations of up to several hundred parts per million, were used as prototypical compounds. Parameters studied included the reactor volume, the species residence time, the specific energy input, and the influent contaminant concentration. Moreover, the dependence of the overall destruction efficiency on specific energy, contaminant type, and presence of other contaminants in well-defined contaminant mixtures was evaluated. For example, complete removal of benzene was achieved.

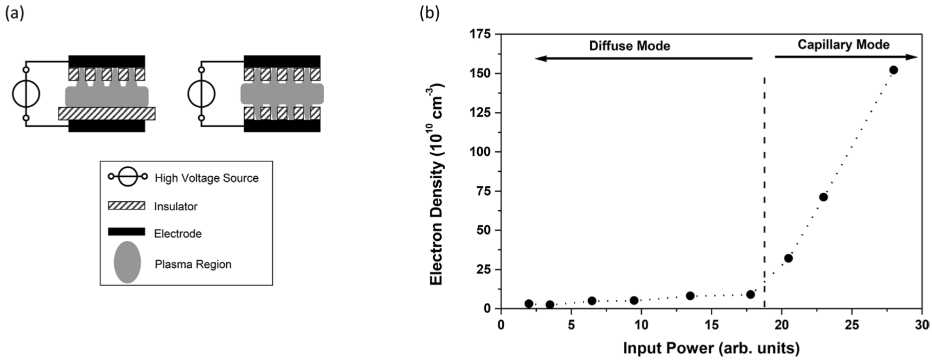

Fig. 4 **(a)** Schematic diagrams of capillary plasma electrode (CPE) discharge with at least one perforated dielectric; **(b)** Electron density in a CPE in helium as a function of power input. The transition of the discharge from the diffuse to the capillary mode with a concurrent drastic increase in the electron density is clearly apparent [110]

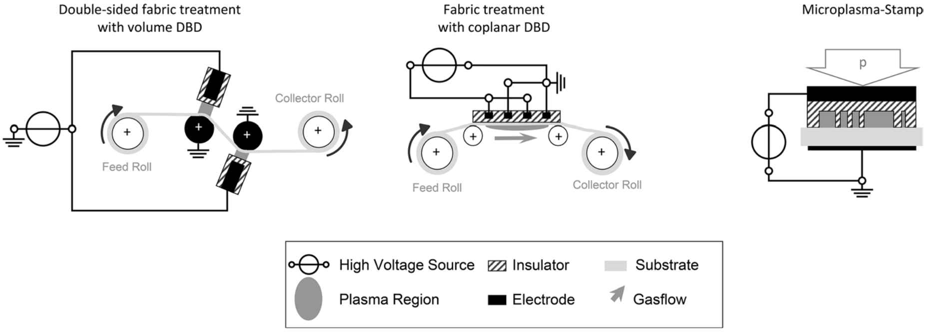

Fig. 5 Surface treatment by means of DBDs: Double-sided fabric treatment (left), coplanar DBD (middle) and microplasma-stamp (right)

However, we note that in some cases, the destruction efficiency leveled off at less than 100%, e.g. at about 80% for toluene. Back reactions leading to the formation of toluene in the plasma are likely responsible for that. By-product formation during the plasma chemical destruction was assessed by monitoring the concentration of gaseous nitrogen and carbon oxides, namely NO, NO_2, CO, and CO_2. Carbon mass balances were used to assess the possibility of complete contaminant destruction leading to mineralization. The results of these studies can be summarized as follows. (1) VOC destruction efficiency increases with specific energy, but tends to level off at values of the specific energy that are compound dependent; (2) as initial VOC concentration increases, destruction efficiency increases; (3) VOC destruction efficiency increases significantly with residence time initially, but tends to level off for higher values of the residence time that are compound dependent; (4) VOC destruction efficiency increases with reactor volume as long as the plasma density does not decrease below a characteristic threshold; (5) for chemically similar compounds, VOC destruction efficiency is inversely related to the ionization energy and directly related to the degree of substitution; this suggests that chemical substitution sites may be the places of high plasma-induced chemical activity; (6) destruction efficiency of individual components (i.e., toluene) in VOC mixtures (i.e., BTEX) is reduced in comparison to the destruction efficiencies obtained when the contaminant is treated alone.

An important advantage of nonthermal plasmas, and DBDs in particular, is that they can be used “on-demand.” Thus, the power consumption can be adapted to the specific, and possibly variable, contamination level of the gas to be purified. This feature makes DBD attractive for so-called power-to-gas or power-to-fuel approaches. Power-to-X means the conversion of intermittent energy from renewable sources (wind, solar) to gaseous or liquid energy carriers that can be stored or processed further. Converting CO_2 in plasmas for carbon capture and utilization (CCU) one could realize closed carbon cycles. The review of Kogelschatz mentions methanol generation and CO_2 reforming of methane. Greenhouse gas conversion was the last topic of his and Eliasson’s activities at the ABB Corporate Research Center in Baden/Switzerland. However, the work stopped around 2000 with his retirement, and for a long time, there was very little work done on this topic. This changed around 2010 with the emerging challenges of the changes in energy policy and the requirement of power-to-X technologies. Since then, there has been a huge amount of work on CO_2 conversion by means of plasmas, in particular in The Netherlands, Belgium, United Kingdom, Germany, China and Japan.

Methanol and carbon monoxide (CO) are of interest as readily storable synthetic fuels, platform chemicals, or chemical feedstock for hydrocarbon and fuel synthesis, respectively. DBDs were studied for CO_2 splitting, hydrogenation, and the dry reforming of methane and methanization. Many studies are devoted to packed-bed DBDs as they enable to the inclusion of catalysts for increasing the energy efficiency and/or selectivity. DBDs are considered as insufficient for industrial application, while quasi-thermal discharges, like microwave-generated plasmas and gliding arcs, but also radio-frequency-generated plasmas show much higher energy efficiencies and conversion degrees [112]. For CO_2 splitting, the energy yields in DBDs are in a range of 3 to 30 g CO/kWh, about 4-fold lower than in microwave plasmas and about one order of magnitude below the chemical limit. However, DBDs feature a simple and robust design, can be easily scaled up to higher gas flows, can operate at pressure above 1 bar, do not require sophisticated high-voltage power supplies and are not prone to significant problems of electromagnetic compatibility (EMC). Therefore, they could be attractive for decentralized power-to-X approaches where cheap electrical energy is available. Finally yet importantly, only DBDs allow the study of the mechanisms for direct plasma-catalyst interaction.

CO_2 splitting in DBDs proceeds by direct electronic dissociation. Its efficacy was found to be determined by the electrode design and geometry, as well as by the operation parameters. The material of the dielectric barrier has no distinct influence on conversion [113]. The effects of dielectric barrier thickness and gap width are discussed controversially. Some authors report a decrease in conversion and energy efficiency for thicker dielectric barriers [114] and shorter discharge gaps [115], while others state that the geometric parameters of the DBD configuration influenced the effectiveness of carbon dioxide splitting but not the efficacy. The energy yield of CO formation is not changed by the reactor construction (discharge gap, thickness and material of the barrier), the pressure or the frequency or duty cycle of the applied high voltage [92, 115]. The discharge zone length seems to have a significant effect, depending on discharge power or gas-flow rate [114]. A higher conversion is achieved in asymmetric coaxial DBDs than in symmetric reactors [116]. The position of partial chemical equilibria also depends on reactor parameters and operating conditions (i.e., power, pressure, and gap) [117]. Adding a packed-bed material in the discharge space usually increases the reactor performance. For specific input energies below 10^4 J/L this quantity is a universal scaling parameter, while for larger values, a more distinct effect of the gas residence time in the reactor is obtained. A distinct dependence of the energy yield of CO formation on the mean reduced electric field strength cannot be concluded solely from the experiments.

Surface Treatment

Similar to ozone generation, surface activation - the second "classical" DBD application - is also enjoying a steadily growing market, since there is a constantly growing need for the treatment of plastic materials and surfaces to make them receptive to bonding, printing, coatings or adhesives or to clean them from organic compounds. Besides plastics, wool and textiles are treated for functionalization and finishing. The pretreatment of insulated wires and cables is known and photoresist layers are removed in lithographic process chains. When using gases other than air, the incorporation of atoms such as nitrogen, fluorine and silicon into the surface layer is possible. In the case of precursor gas admixtures, thin layers deposit in the materials. Polymer and SiO_x coatings, and even hard diamond-like carbon films have been obtained with Atmospheric-Pressure Plasma Enhanced Chemical Vapor Deposition (APPECVD).

On industrial scales, continuous (in-line) treatment of web material is realized by roll-to-roll systems utilizing the volume DBD. High throughput speeds up to 10 m/s and about 10 m wide roll widths are achieved today [118]. The main issues for the improvement of this technology addresses intelligent power deposition to avoid the so-called pinholing (i.e. puncture of plastic substrates by strong microdischarge channels) and more efficient and adaptable high voltage power supplies. For example, microcomputer-controlled automatic adjustment of the operation frequency of the power supply to the resonance frequency of the electrical circuit can be accomplished.

For the in-line treatment of textiles and tissues that are more delicate (e.g. ultrathin plastics, porous fabrics), Cernak et al. developed a coplanar DBD arrangement in the 2000’s [119–121]. In this arrangement, both electrodes are embedded in the dielectric, the plasma is formed about 100 µm above the dielectric and the fibers to be treated are pulled along the surface through the plasma. Such plasma-activated fabrics have been successfully applied to post-plasma grafting of polyacrylic acid, immobilization of chitosan and nanopowders, and electrodeless metal plating. Meanwhile, a roll-to-roll system with curved electrodes has been developed for 28 cm wide materials and treatment speeds of up to 100 m/min [122].

An area-selective activation or functionalization of surfaces has become available by microplasma-stamps developed by Klages and Thomas et al. [123–129]. Here, the plasma is generated in defined cavities in a soft dielectric body on the high voltage electrode, which is compressed onto the substrate. The microplasma spots being formed have a size in the range of some hundred µm and are arranged in customized, predefined pattern.

Since DBDs operate at atmospheric pressure, they become more and more attractive for deposition applications as expensive and maintenance-intensive vacuum technology is avoided. Other advantages are higher deposition rates than in most low-pressure plasmas and compliance with continuous flow processes operating at high speeds. The plasma sources can also be utilized to clean surface prior to the deposition step by changing the gas composition and amazing capabilities to grow patterned or nano-structured surfaces were obtained [130]. Up to now the plasma polymerization of silicon-based precursors, the deposition of epoxy-group-containing plasma-polymer films and aldehyde-group-containing plasma-polymer films as well as copolymerization were studied [131]. Silicon oxide, silicon nitride, metal oxide, sulfonated, hydrocarbon and fluorocarbon films were deposited. A great potential is in the surface engineering of biomaterials [132, 133]. For example, the inclusion of a biomolecules during deposition has been demonstrated [134] and sensitive compounds such as enzymes are not affected [135]. Gases as well as aerosols can be used as precursors [136]. In addition to solid surfaces and fabrics, the deposition of even more complex, porous objects such as foams has been demonstrated [137, 138].

Challenges include the non-homogeneous plasma treatment, the gas flow dynamics and the avoidance of gas contamination, the formation of clusters and powders, arcing, the deposition of the electrode which may limit the lifetime of the plasma source and the fact that chemical pathways as well as the interplay between discharge physics and plasma chemistry are not yet fully explored.

The control of the discharge uniformity (filamentary or diffuse) has a distinct influence on the film properties and growth. For example, higher deposition rates per coated area unit compared to filamentary modes are achieved in the atmospheric-pressure glow discharge (APGD) regime, while the deposited power per area unit can remain below 10 W/cm^2. The role of the different operation parameters, substrate properties as well as plasma and power source designs on the deposition result is discussed in detail in the reviews by Merche et al. [139] and Massines et al. [140].

Using high voltage frequency in the range of a few 100 kHz, a dynamic power matching, and a discharge stabilization network another type of uniform volume DBD, the atmospheric pressure glow-like DBD (GLDBD) is generated [141]. This form of the DBD has a higher peak power than the atmospheric-pressure Townsend discharge (APTD) and the APGD while the uniformity of the deposited layers is higher than for filamentary DBDs. The deposited SiO_2-like layers deposited on large area polymer webs in roll-to-roll reactor were found to be highly smooth and uniform. They reproduced the surface texture characteristics of the polymeric substrate. More recently, van de Sanden et al. were able to improve the plasma uniformity together with the input power by applying an extra 13.56 MHz radio frequency voltage on the usual low frequency voltage (200 kHz) [142].

As all plasmas DBDs show great potential for the generation of nanoparticles on surfaces, nanofiber treatment and nanocomposite coatings with a plethora of applications [143–147]. The in-flight coating of nanoparticles in volume DBDs was one of the last scientific activities of Kogelschatz after his retirement at ABB, when he stayed at the University of Sherbrooke [148, 149].

DBDs for Photonics

Gas lasers and excimer lamps are still largely based on DBDs and the market for these light sources is still growing. Gas lasers are a key element in modern lithography technologies and for material processing; excimer lamps are the first choice for curing and other photochemical processes. The UV-light generation is based on the formation of exited molecules (dimers and trimers), such as XeCl* being formed in the plasma by three-body collisions between metastable excited states and the gas particles [150]. In the 1990's and 2000's several manufacturers offered flat, mercury free lamps for commercial light systems, backlight illumination and photochemistry. Today, these are now in competition with light-emitting diodes (LEDs). Research on this topic did not only provide the basics for efficient and mercury-free light generation, but allowed also very useful insights in the electrical characterization and the development of more efficient power supplies.

The first ideas for plasma displays were described in 1930's, but it took about half a century to bring them to the market. Mass production started in the late 1990's and plasma displays became the most popular flat television screens around the year 2000. Meanwhile, its production stopped and plasma displays have been replaced by more energy efficient LED and organic light-emitting diode (OLED) screens.

DBDs are still under research and development for special lamp arrangements. The ability provided by micro- and nano-fabrication techniques to control the geometry of cavities, including details such as the contour and roughness of surfaces, enabled the construction of new plasma devices [151]. One of the earliest geometries of such microcavity plasma was the inverted square pyramid. Eden et al. demonstrated the fabrication of arrays of nearly identical microcavities being essentially atomically smooth [77, 152, 153]. The inverted square pyramid shape was etched into silicon (Si) by a wet, anisotropic process. While the doped Si-bulk served as one of the electrodes, the dielectric barrier was a polyimide film deposited on the etched structure. A nickel film on top was the second electrode. Later, the microcavities were also fabricated in a wide variety of dielectric materials, including ceramics and glasses. The example in Fig. 6 (a) shows a portion of a large array of microcavity plasmas with a parabolic cross section. The upper circular aperture has a diameter of 150 µm. The cavities are built on an aluminum (Al) foil by a wet electrochemical process. Aluminum oxide (Al_2O_3) film with a thickness of about 20 µm generated by anodization of the Al foils serves as the dielectric barrier. Linear arrays of cylindrical plasma microcavities with diameters of 127 µm were also fabricated in green ceramic tape by buried layers of cured silver paste.

The microplasma operation does not strictly follow Paschen's law (i.e. scaling with the product *pressure x characteristic dimension* at given temperature); streamer-free discharges in the abnormal glow regime are readily generated in rare gases at pressures between 67 mbar and several atmospheres in cavities with characteristic dimensions ranging from 30 to 500 µm. Electron densities in these plasmas are in the range 10^{12}–10^{15} cm^{-3}, but can exceed 10^{17} cm^{-3} at elevated pressures in case of pulsed applied high voltage. Thus, the local specific energy can reach values larger than 1 MW/L_n. With these properties, microplasmas can enable new applications, such as VUV and EUV light sources, electromagnetic wave control, micro-total analysis systems and lamp-driven atomic clocks. The Fig. 6 (b) shows a photo of a commercialized microcavity plasma lamp emitting at 172 nm [154]. To date, emission intensities beyond 300 mW/cm^2 with peak emission powers of 600–800 W in a bandwidth of 9 nm have been achieved. With an active surface area of about 100 cm^2 only, such lamps are capable of generating extraordinary average radiation powers above 30 W.

Microplasma arrays have also demonstrated to control, manipulate, or detect electromagnetic radiation in the microwave and THz spectral range. These devices are called plasma photonic crystals or plasma metamaterials [155]. The control of transmission, reflectance, polarization and other electromagnetic wave properties is based on the variable dielectric permittivity in spatially periodic arrays of non-thermal plasma spots.

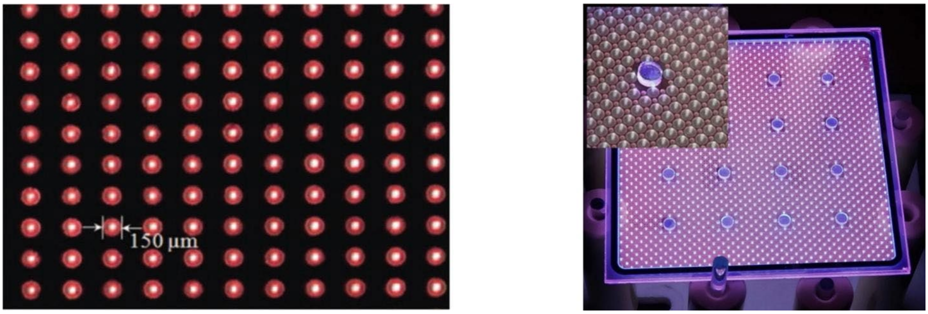

Fig. 6 Optical micrograph of an 11×10 segment of a 200×100 array of Al/Al_2O_3 parabolic microcavity plasma device, operated in neon at 667 mbar by sinusoidal high voltage at 20 kHz (left) and a Xenon lamp emitting at 172 nm (Xe_2* dimer radiation) [154]

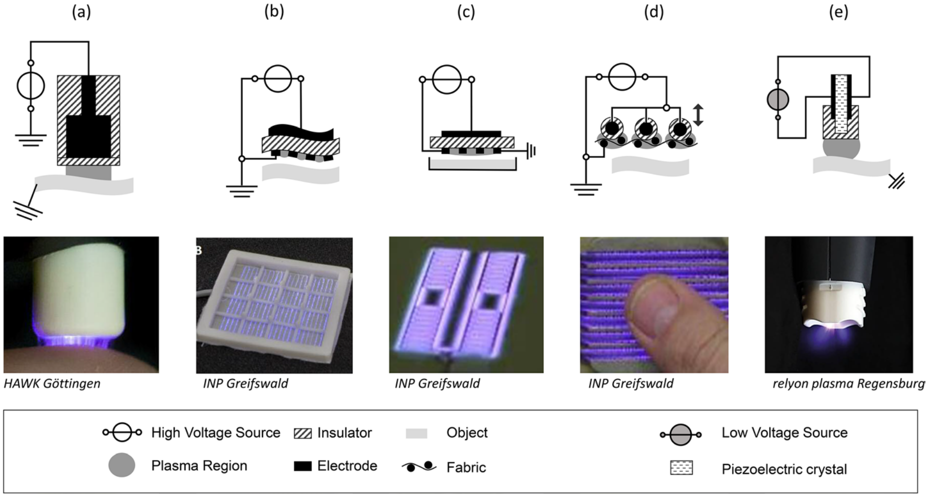

Fig. 7 DBD in plasma medicine: **(a)** Floated Electrode DBD [172]; **(b)** surface DBD on flexible barrier material with a fabric electrode [168]; **(c)** surface DBD on dielectric plate [173]; **(d)** surface DBD realized with tubes or wires [173]; **(e) piezo-electric driven DBD** [174]

Plasma Medicine

Although the first approved plasma-medical tool was a plasma jet [156], life-science plasma applications utilized and investigated DBDs from the very beginning and finally found their way in preclinical and clinical research as well biological decontamination. When Roth et al. demonstrated the biological decontamination of surfaces by means of atmospheric pressure plasmas for the first time a DBD was used [17, 157]. Already at the end of the 19th century and until the 1930's DBD-based plasmas (called "vacuum electrodes" or "violet rays") were applied on humans for "high frequency electrotherapy" [158]. Around 2005, the therapeutic use with focus on the treatment of chronic wounds, infected skin diseases, and dermatitis started [4, 159–163]. Meanwhile, the inactivation of a broad spectrum of microorganisms, including multidrug resistant pathogens by plasma jets and DBDs is confirmed and the stimulation of cell proliferation and angiogenesis with lower plasma treatment intensity has been demonstrated [23]. The inactivation of cells by initialization of cell death with higher plasma treatment intensity could enable the application in cancer therapy [164–167]. The biological plasma effects are mainly mediated via reactive oxygen and nitrogen species influencing cellular redox regulated processes [168–171].

Since plasma jets are more suitable for small spot treatment, DBDs are capable to treat extensive surfaces. Thus, beside the research on plasma jets, e.g. by Stoffels et al. [175] and Weltmann et al. [156, 173, 176–178], the concept of the floated electrode DBD (FEDBD) was introduced by Fridman et al. [179–181] and Viöl et al. [172, 182–185]. The FEDBD is a volume DBD ignited with the body serving as the second electrode (see Fig. 7 (a)). Therefore, the powered electrode is embedded in a dielectric body. The distance between dielectric and body surface is in the range of some millimeters. Due to the insulating properties of the dielectric around the powered electrode, the setup is safe to be used for human body treatment. It was designed for human skin treatment. The plasma is operated in ambient air, but not limited to this gas mixture. Fridman et al. demonstrated biological decontamination in-vivo and skin sterilization of live mice without damage of the living tissue [179, 186, 187]. It is able to control methicillin-resistant *Staphylococcus aureus* in planktonic form and biofilms. Blood coagulation and promotion of apoptotic behavior in melanoma skin cancer cell lines by the FEDBD were shown as well. Numerical simulations showed a significant electric field penetration into intracellular structures of the treated tissue, which could enable electroporation [188].

In particular, for biological decontamination and medical therapy the DBD concept demonstrated its superior versatility. Many different and innovative DBD arrangements have been designed and studied [18, 34, 176]. For example, a cascaded DBD enables the combined action of plasma-generated (V)UV radiation and reactive species [189, 190]. Therefore, it consists of a high voltage powered electrode, a sealed glass chamber filled with excimer-forming gas for UV-light generation, and an air flushed gas gap for the generation of the reactive species faced to the grounded electrode with the objet on it. The treatment of packed goods by a surface DBD arrangement is possible by a label with bonded electrodes on [191–193].

The in-package treatment and decontamination of food can be realized also in large-gap volume DBDs in air with up to 4 cm discharge gaps [194–196]. High voltage amplitudes up to 70 kV_{rms} are required at these conditions. A commercial device for the cleaning and sterilization of liquid transfer devices such as pipette tips, cannulae and pin tools ignites DBDs between a central pin electrode and the inner wall of the tips/cannulae which are serving as the dielectric barrier [197]. Furthermore, DBDs can be generated in the biopsy channels (length of several meters) in endoscopes by equidistantly twisting two electrodes around and along the channel tube, which serves as the dielectric barrier [198]. The “plasma gun” is another plasma source for endoscopy, which is based on a gas flushed capillary with successive propagation of a primary plasma from the inside of the dielectric tube towards a target [199, 200]. These plasma sources open up new opportunities in both clinical applications and medical device disinfection, so-called “plasma endoscopy” [201].

Instead of an insulator a highly resistive material layer (resistivity of a few MΩ/cm or higher) which acts as distributed resistive load can be used. This so-called resistive barrier discharge enables plasma operation not only by DC high voltage, but also shows self-pulsing action [202, 203]. While the plasma appears uniform in helium, microdischarges are obtained in air.

The yet commercialized plasma-medical DBD devices are surface DBDs, i.e. there is no direct contact of the active plasma with the target to be treated, see Fig. 7 (b - d). The plasma is generated at an individually designed electrode structure, which is isolated from a counter electrode and brought in close vicinity to the surface [204–209]. Flexible materials, such as sketched in Fig. 7 (b), or arrays of movable electrodes enable the adaptation to curved geometries such as parts of the body, like extremities [210]. Alternatively, insulated conductor wires in a woven arrangement (fabric-type electrode) can even construct a sticking ‘plasma-plaster’.

For surface treatment and life-science applications the piezo-type DBD sketched in Fig. 7 (e) was developed [174, 211–216]. Mechanical, thermal, or electrical forces applied to a piezoelectric crystal alter its structure and changes the magnitude of its polarization. Resonant electrical-mechanical vibrations induced by an AC voltage with low amplitude can enhance the surface potential at the edge of the crystal to be sufficient for gaseous breakdown. Thus, it enables the operation of a DBD with a low voltage power supply.

As the current state of knowledge about biological non-thermal plasma effects refers mainly to the action of reactive oxygen and nitrogen species supported by electrical fields and UV radiation, for future plasma-based therapeutic systems it is necessary to improve, optimize, and enlarge the spectrum of medical applications and plasma sources as well as their safety. Furthermore, one must identify and define reliable and objective monitor and control parameters.

Since plasma medicine developed to an own-standing field of research and development, we can refer to the several textbooks and review articles and do not go into more detail here.

Other new Topics and Current Trends

The current needs for improved water treatment and the fact, that wounds are partially wet environments inspired a tremendous amount of research about the **treatment of liquids** using non-thermal plasmas [25]. The generation of plasma in liquids is mainly by means of pin-to-pin or pin-to-plane arrangements by fast pulsing high voltage. In contrast, DBDs enable plasma generation at gas-liquid interfaces at much lower power density. In case of surface DBD such as shown in Fig. 7 (c), the discharge is in vicinity to the liquid. The active species of the plasma diffuse into the liquid [217] and cause a complex redox chemistry involving solvated electrons, acidification, and the degradation of pollutants and microorganisms.

Since water and many other liquids are conductive they can be an active part of the discharge, too. Liquid electrodes can be utilized for the treatment of outer and inner surfaces of tubes and other hollow dielectric bodies [218–220]. In water falling film reactors or rotating drum reactors, thin water films flow laminar over the grounded electrode (to avoid charging) and serve as the counter electrode against the dielectric covered high voltage electrode [34, 221, 222]. The development of microfluidic devices for enabling process intensification is an emerging field [223–225]. The main challenge of this approach is the increase of the throughput to reach industrial scales [225]. The direct interaction between gaseous discharge and liquid film includes numerous processes and mechanisms like ionization, excitation, formation of negative ions, sputtering and electrolysis. Its distinct role depends on the both the characteristics of the discharge in the gas phase as well as the properties of the liquid. Evaporation of water during discharge activity changes the gas composition and the surface conductivity of the barrier. This affects the discharge physics and the chemistry. Due to the capacitive load in DBDs, the discharge mostly stops in the streamer regime and sparking is avoided. During the breakdown on the nanosecond time-scale the water mainly behaves as a dielectric. However, due to the polarity of the water molecules the water surface forms funnel-like protrusions under the electric field. In analogy to electrospraying and –spinning these protrusions are called Taylor cones [226]. They decrease the discharge gap, resulting in lower breakdown voltages while its shape and size are independent of voltage polarity [227].

Since about 70 years non-thermal plasmas are under research for active flow control [228]. The idea behind this is the formation of ions and thus, an electric (or ionic) wind to accelerate the airflow tangentially and in close vicinity to the wall of a wing. The airflow profile within the boundary layer is modified which controls laminar–turbulent transitions, prevents or induces separation of gas flows, reduces drag and enhances lift of airfoils, and stabilizes or mixes airflows. These prospects are of high importance for aeronautics and other industries where internal and external airflows must be handled. For example, drag reduction on aircraft wings decreases fuel consumption, enables higher speed and longer flight range. So-called plasma actuators utilize corona discharges or surface DBDs. In contrast to mechanical actuators they do not contain mechanical parts, can be implemented in the airplane wing surface and enable real- time at high frequency due to short response times. First attempts to use surface DBD plasma actuators were stated in the 1990's by Roth et al. [157] and from 2000 on, there is an increasing number of publications about this subject.

Comprehensive reviews about the topic were published by Moreau et al. [228, 229], Corke et al. [230], and Kriegseis et al. [231]. Depending on the operation parameters and the discharge geometry (e.g. thickness of the dielectric barrier, size of the electrodes) single surface DBD strips induce an airflow of up to about 8 m/s at a distance of few 100 µm from the wall. The body force per plasma power created by the ion motion is about 0.15 mN/W. In subsonic airflows up to 30 m/s and for Reynolds number of about 10^5 significant and efficient control could be achieved. Plasma actuators are also investigated for aerodynamic noise reduction and anti-icing of airplane wings [232–234]. Challenges of these approaches are the low efficiency of energy conversion and the degradation of the electrodes and the dielectric due to air plasma exposure [235, 236].

DBDs are also used for analytical applications as dissociative source for optical emission spectrometry and for ambient-ionization techniques in mass spectrometry and ion mobility spectrometry [24, 237–239]. For the latter, DBDs are able to ionize polar and nonpolar compounds with almost any fragmentation, so-called soft ionization which proceed through protonation, charge transfer and cluster ions. Gaseous as well as liquid samples can be analyzed [240]. Planar, microhollow electrode based and coaxial systems have been investigated, as summarized by Franzke et al. [24]. DBDs are well-suited for "lab-on-chip" solutions and "micro total analysis systems" because of small size, low power consumption and heat generation as well as low contamination of the analyte due to dielectric barriers covering the electrodes. These DBDs are mostly capillary based (diameters in the 100 µm-range) and operated in helium or other inert gases [241].

A DBD detector for gas chromatography is commercialized already. It uses the photoionization of the sample molecules via VUV-radiation from helium (or argon) excimers He_2* (Ar_2*). The ionization via helium metastable species (energy about 20 eV) is also mentioned [242]. Since most substances have a lower ionization potential than the metastable energy of helium, this detector is considered as universal. The detection limit is given as below 0.1 ppm (or 0.1 pg/s) and amounts in the order of mmol to µmol can be detected. Reduced dynamic range and nonlinear response to carbon mass are seen as disadvantages, but the interest on this detector is steadily growing since 2012 because of its universality and linear sensitivity, as well as the signal-to-noise ratio, which is 100 times higher than for other detectors [243].

Nanosecond pulsed surface DBDs were demonstrated to be an efficient tool for plasma-assisted ignition and combustion at elevated pressures [244]. Spark-ignition is an established process, but non-thermal plasmas in general may enable to change the energy branching from mainly heating and excitation of low-energetic vibrational degrees of freedom to the excitation of high-energetic electronic states and ionization. Even very low amounts of even relatively small amounts of atomic species and radicals (fractions at about 10^{-3} and even lower) can shift equilibria in the chemical system and initiate combustion. Therefore, plasma assisted ignition and combustion could improve engine performance, increase lean burn flame stability, reduce emissions, or enhance low temperature fuel oxidation. Over the last decade, significant progress has been made [245, 246]. The understanding of the fundamental chemistry and dynamic processes has been improved and applications of plasma in engines has been performed. The pulsed surface DBD in a C_2H_6/O_2 mixture at ambient initial temperature and 1 atm showed a more uniform ignition than a pin-to-pin spark nanosecond discharge.

Since about 10 years, plasma processes are considered for agriculture [103]. The ongoing growth of world population demands for increased food production, while adverse effects of the agricultural production on environment and natural resources such as water, soil and air should be kept to a minimum. The quality and safety of food and other agricultural products must be enhanced as well. All this requires the exploration of new technologies in all sectors of the production chain, "from farm to fork".

DBDs are established in water and air cleaning. They provide even more options, e.g. the chemical treatment and decontamination of crops, seeds, and soil. Plasma can also be used for treatment of food [247]. Reactive species, charged species, electric fields, and ultraviolet radiation from the plasma contributes to seed germination, seed disinfection, plant growth, insect control, retention of quality of agricultural products, and soil remediation. Furthermore, chemical and biological decontamination of processing tools and packaging is possible. DBDs can be used to treat surfaces and gases directly, but the plasma-treated gas can also be applied remotly. Water treated with DBDs directly or flushed with plasma treated air shows the capture of atmospheric nitrogen. The plasma activated water acts similar to conventional nitrogen-based fertilizers. Thus, an on-demand local fertilizer production is enabled. Seeds and soil can be treated in packed bed DBD reactors or by transporting them through a surface or coplanar DBD. In fact, many experiences gained in plasma medicine will be very useful for the future developments of plasma sources in this field and the interpretation of the plasma effects on the majority of objects. To get more insights into the topic, we refer to the reviews of Puac et al. [248], Atti et al. [249], Ranieri et al. [250], and the textbook by Misra et al. [251].

Summary

The Fig. 8 presents some selected milestones of applications of DBDs. All of them have been addressed in this contribution. The ozone generation is the oldest and most established application of DBDs, followed by surface activation and photonics. The figure emphasizes also the wide range of applications of this type of discharge, which raised significantly within the last three decades.

Seeing this and having in mind that the research has become more inter- and cross-disciplinary it can be stated, that the "renaissance of the DBD" is still underway. The reasons for this are manifold. First, the diagnostic methods and modeling/simulation tools enable an increasingly deeper insight in the complex physical and chemical mechanisms. This provides a good basis for the discovery, development and optimization of applications and for

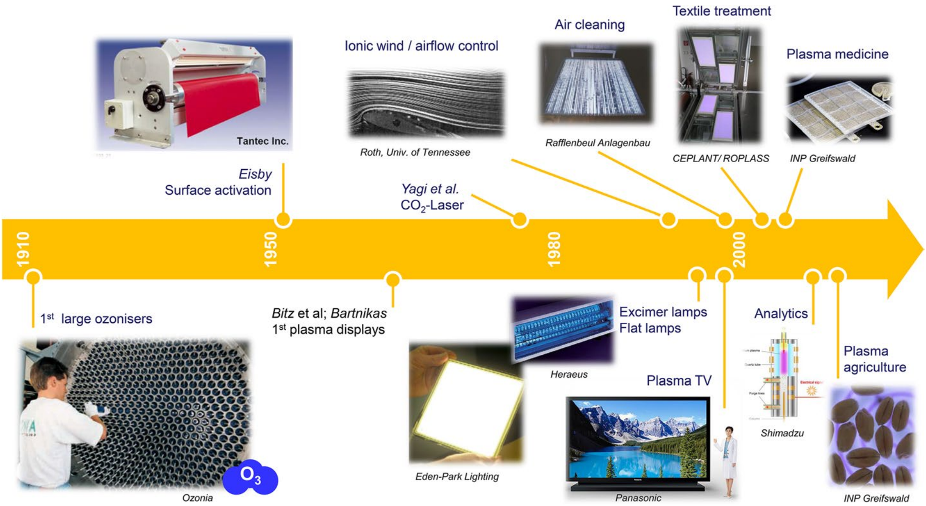

Fig. 8 Selected milestones of applications of DBDs. Pictures are publicly available from the institution being mentioned or reference [157]

the transfer into industry. Second, DBDs provide reliable, robust, reproducible, cost efficient, and stable non-thermal atmospheric-pressure plasma sources that are well scalable. From these points of view, we can still agree with Ulrich Kogelschatz and his statement that the trend of substantially increased market penetration of DBDs will continue, even 20 years after the publication of his review article.

Research and development require inventive freedom, but on the way to the publication of results and the technology transfer, however, standards and quality assurance are then crucial. Innovative linking and integrated analysis of data from laboratory and simulation experiments and the implementation of data principles, standards, workflows and methods for uniform research data management are now under development. Data science methods for automated and AI-supported analysis of data as well as the use of machine learning methods and neural networks to accelerate the development will open up new possibilities in the field.

Ulrich Kogelschatz contributions were pioneering in this regard, as they provided many researchers with an excellent introduction into the subject. Uli, as he was called by many of his friends, understood unlike few others how to combine fundamental research and applications. Many colleagues, who met Uli, will remember that he was aware of nearly every publication in the field, reading it with high curiosity and considering it with high respect and a broad and open mind. To work as a scientists and to discuss scientific concepts with fellow scientists was one of the greatest joys for him.

We miss his scientific insights, his humor, and his humanity.

Author Contributions RB., KHB. and KDW prepared the outline of the manuscript. RB, KHB and KDW wrote the introduction and summary together. RB prepared the figures, wrote the first draft of main text for Sects. 2 and 6 and parts of Sects. 5 and 8. KHB wrote the first draft of main text for section Sects. 3 and 4. KDW wrote the first draft of main text for Sect. 7 and parts of Sects. 5 and 8. RB, KHB and KDW reviewed and edited the whole manuscript.

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