

# From ozone generators to flat television screens: history and future potential of dielectric-barrier discharges\*

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**Abstract:** Dielectric-barrier discharges (silent discharges) combine the ease of atmospheric pressure operation with nonequilibrium plasma conditions suited for many plasma chemical processes. In most gases at this pressure the discharge consists of a large number of randomly distributed short-lived microdischarges. Their properties are discussed in detail. Traditionally mainly used for industrial ozone production, dielectric-barrier discharges have found additional large volume applications in surface treatment, high-power CO<sub>2</sub> lasers, excimer ultraviolet lamps, pollution control and, most recently, also in large-area flat plasma display panels. Future applications may include their use in greenhouse gas control technologies. Historical aspects, properties and applications of dielectric-barrier discharges are discussed.

## HISTORICAL BACKGROUND

In 1839 Schönbein identified the odor rising from the anode during the electrolysis of water as an attribute of a new chemical compound which he gave the name *ozone* [1]. A few years later, in 1857, Siemens, proposed a novel type of electrical gas discharge [2] that could generate ozone from atmospheric-pressure oxygen or air. This discharge was initiated in an annular gap between two coaxial glass tubes. By using coaxial external electrodes a radial electric field was applied by an alternating voltage of sufficiently high amplitude to cause electrical breakdown in the flowing gas. Since the electric current is forced to pass through the glass walls acting as dielectric barriers the discharge is commonly referred to as the *dielectric-barrier discharge (DBD)*. Historically it was originally called the *silent discharge*, an expression used already by Andrews & Tait [3] in 1860. It is still used today. Some oxygen molecules in the air flowing through the discharge gap between the glass tubes were converted to ozone. It was soon found out that also traces of other compounds were generated and much effort was devoted to the identification of the different discharge products: N<sub>2</sub>O, NO, NO<sub>2</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, in humid air also HNO<sub>2</sub> and HNO<sub>3</sub>. These compounds can interfere with ozone formation. When they reach a certain concentration ozone generation completely breaks down [3]. This *ozoneless mode* of the discharge is also referred to as the state of *discharge poisoning*. Also the use of DBDs for the decomposition of different gaseous compounds like NH<sub>3</sub>, H<sub>2</sub>S and CO<sub>2</sub> was already reported in the last century. Industrial large-scale production of ozone for the treatment of drinking water started in Europe around 1900. M.-P. Otto [4] in France and H. Becker [5] in Germany made important contributions to the development of industrial ozone generators in the first part of this century. In 1943 T.C. Manley [6] derived the famous power formula for DBDs which relates the dissipated power to the operating frequency, the applied peak voltage and some discharge properties. First applications of silent discharges to the generation of VUV excimer radiation were published by Tanaka [7] in 1955 and first applications to the generation of infrared radiation in CO<sub>2</sub> lasers were presented by Yagi & Tabata [8] in 1981. The year 1996 finally is characterized by major investments in production facilities for flat television screens using ac plasma

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display panels [9]. This novel application of DBDs is expected to reach market volumes that correspond to at least 10 times the original ozone market.

## DISCHARGE PROPERTIES AND MICRODISCHARGE MODELING

Dielectric-barrier discharges are very attractive for industrial applications because they can provide nonequilibrium plasma conditions at about atmospheric pressure. The classical DBD configurations (Fig. 1) utilize planar or cylindrical arrangements with at least one dielectric layer placed between the electrodes.

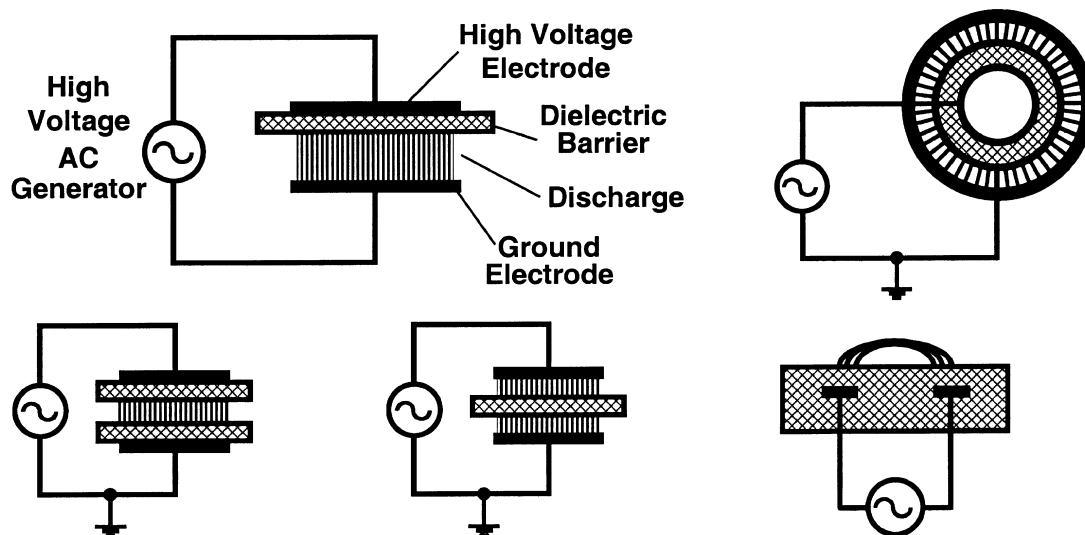
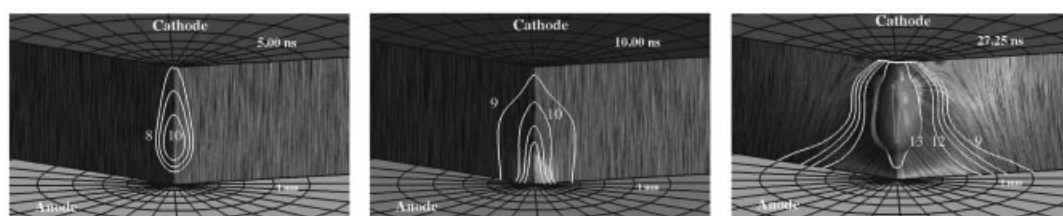


Fig. 1 Common planar and cylindrical dielectric-barrier discharge configurations.

Typical dielectric materials used are glass, quartz, ceramics or also polymers. Typical discharge gap spacings range from 0.1 mm in plasma displays over about 1 mm in ozone generators to several cm in CO<sub>2</sub> lasers. The most interesting property of DBDs is, that in most gases at about atmospheric pressure, breakdown is initiated in a large number of independent current filaments of nanosecond duration. This filamentation of atmospheric-pressure air breakdown in DBD configurations was first observed by Buss [10] in 1932 and since then investigated by many authors. Extended lists of references can be found in [11–16]. The plasma parameters in these breakdown channels, now frequently referred to as microdischarges, can be predicted by model calculations and can be influenced by external parameters. Advanced plasma diagnostics and computer modeling has put us in a position to investigate microdischarge properties in detail and to tailor them for any desired application.

The microdischarge filaments can be characterized as weakly ionized plasma channels with properties resembling those of transient high-pressure glow discharges. Due to charge build-up at the dielectric surface, within a few ns after breakdown, the electric field at the location of a microdischarge is reduced to such an extent that the current flow at this position is interrupted. Because of the short duration and the limited charge transport and energy dissipation this normally results in little gas heating. In a microdischarge a large fraction of the electron energy can be utilized for exciting atoms or molecules in the background gas, thus initiating chemical reactions and/or emission of radiation. In DBDs bursts of such microdischarges are observed at a repetition frequency corresponding to twice the driving frequency. The frequency of the applied voltage can range from line frequency to several hundred kHz. At higher frequencies different phenomena are observed because there is not sufficient time between consecutive voltage half-waves to allow the decay of plasma conductivity in a microdischarge filament. In recent years it was also found that under certain conditions homogeneous glow discharges can be obtained in DBD configurations at atmospheric pressure [17–22].

The theoretical description of microdischarge formation and the simultaneous treatment of the chemical reactions in the plasma filaments has evolved only during recent years. Fast computers and efficient numerical codes are required to treat the equations describing excitation, dissociation, ionization, attachment and recombination processes, the influence of the generated charge on the electric field, the electron energy distribution, and the chemical reactions. Two-dimensional modeling of streamer formation has become an important research tool. After the first early attempts to model the formation of a microdischarge in a DBD configuration by Gibalov *et al.* [23] in 1981 different authors have published 2D simulation results on microdischarges in oxygen, air, xenon, flue gases and greenhouse gas mixtures [15,24–33]. Quite recently, even the interaction of adjacent microdischarges and the self-organization of filaments in DBD glow discharges has been treated [34–35]. Figure 2 shows a numerical simulation of 3 stages of microdischarge development in an atmospheric-pressure mixture of 80% hydrogen and 20% CO<sub>2</sub> at 1 bar and 300 K [36].



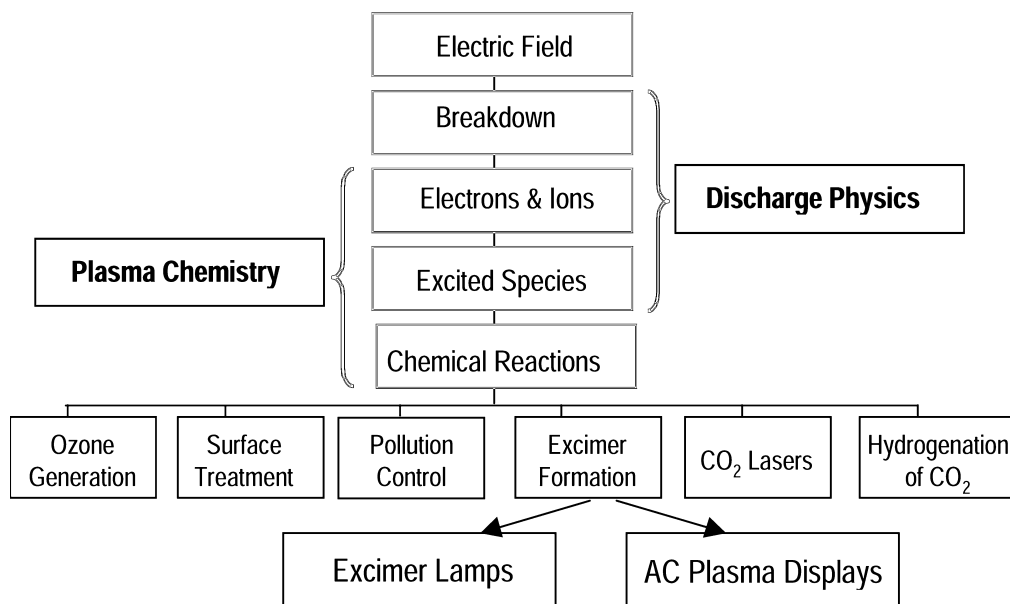
**Fig. 2** Three stages of microdischarge development in a mixture of 80% H<sub>2</sub> and 20% CO<sub>2</sub> ( $t = 5$  ns, 10 ns, 27.25 ns).

The closed isolines indicate the electron density per cm<sup>3</sup> in powers of 10. The other lines indicate the direction of the electric field. The computation was started by releasing initial electrons at the cathode (top) in an initially homogeneous field of 34 kV/cm. The anode (bottom) is 1 mm away and is covered by a dielectric ( $\epsilon = 3$ , thickness: 0.8 mm). After the initial electron avalanche propagates to the anode a cathode directed streamer develops which finally causes an extremely fast formation of a cathode fall region. The maximum current of the microdischarge coincides with the establishment of the cathode fall region. During this stage the electron density is of the order  $10^{14}$  cm<sup>-3</sup>, the lateral extension of the order 0.2 mm. The electron energy inside the conductive filament corresponds to a few eV, while the gas temperature stays close the temperature of the background gas, which is determined by the dissipated average power in the discharge and the heat removal by the cooling circuit [12,13,37]. The energetic electrons in the microdischarge collide with molecules and atoms of the background gas and generate ions, excited species, as well as atoms and other molecular fragments that initiate the ensuing chemical reactions. This combination of nonthermal discharge physics and plasma chemistry had led to a number of important industrial applications of DBDs that are summarized in Fig. 3.

## INDUSTRIAL OZONE GENERATION

Ozone can be generated from oxygen, air or from other N<sub>2</sub>/O<sub>2</sub> mixtures. The first step towards ozone formation in gas discharges is the dissociation of O<sub>2</sub> molecules by electron impact and by reactions with N atoms or excited N<sub>2</sub> molecules, if nitrogen is present [12–14,16,37–43]. Ozone is then formed in a three-body reaction involving O and O<sub>2</sub>. Undesired side reactions pose an upper limit on the temperature and atom concentration in the microdischarge channels. If the oxygen atom concentration is too high recombination to O<sub>2</sub> becomes dominant. If the temperature is too high ozone formation is drastically reduced and thermal ozone decomposition becomes important. Another important aspect is that energy losses to ions must be kept at a minimum because ionic reactions essentially do not contribute to ozone formation. Discharge modeling helped to identify optimum discharge conditions with respect to gap width, dielectric properties, operating pressure, voltage wave form and driving frequency [12,13,25,37].

Many technical ozone generators [13,37] use cylindrical discharge tubes of about 20–50 mm diameter and 1–3 m length. In a traditional ozone generator Pyrex (Duran) glass tubes are mounted inside stainless steel tubes to form annular discharge gaps of about 0.5–1 mm radial width. Conductive coatings (e.g. thin



**Fig. 3** Schematic diagram summarizing the general principle of dielectric-barrier discharges and major applications.

aluminum films) inside the glass tubes are connected to the high voltage supply via individual HV fuses. Other dielectrics, like for instance ceramic tubes or special dielectric coatings on steel tubes, are also increasingly used. High-performance ozone generators now use nonglass dielectrics and narrow discharge gaps. Large ozone generators use several hundred discharge tubes. The outer steel tubes serve as ground electrodes. They are cooled on the outside by a transverse water flow in a classical heat exchanger configuration. Traditionally, ozone generators were operated at line frequency or using motor generator sets. Modern high-power ozone generators take advantage of power semiconductors to generate square-wave currents or specially formed wave trains with a repetition rate of 0.5–5 kHz. Higher operating frequencies can deliver the desired power density at much lower operating voltages, now < 5 kV compared to typically 20 kV in the past. The lower voltage results in less electrical stress on the dielectrics. Power conditioning with modern high power thyristors or IGBTs (insulated gate bipolar transistors) resulted in superior process control and power factor compensation.

Based on optimized discharge conditions and modern power electronics, in recent years considerable progress was made with respect to attainable ozone concentrations and energy consumption. Ozone concentrations up to 5 wt% from air and up to 18 wt% from technical oxygen can now be reached [44]. Large ozone generating facilities produce several hundred kg ozone per hour at a power consumption of several megawatts. The capacity of a big ozone generator is up to 100 kg/h. With modern technology ozone can be produced for less than 2 US\$/kg. The main applications are in water treatment and in pulp bleaching. Applications in organic synthesis include the ozonation of oleic acid and the production of hydroquinone, piperonal, certain hormones, antibiotics, vitamins, flavors, perfumes and fragrances. (Figs 4 and 5).

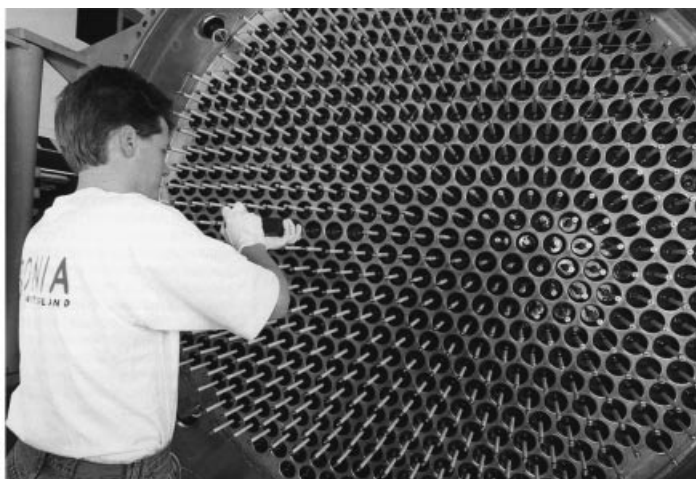
## GENERATION AND APPLICATIONS OF ULTRAVIOLET AND INFRARED RADIATION

### The formation of excimer complexes in dielectric-barrier discharges

When DBDs are operated in high-pressure rare gases or rare gas/halogen mixtures the plasma conditions in the microdischarge channels resemble those in pulsed excimer lasers. Each microdischarge can act as an intense source of ultraviolet (UV) or vacuum ultraviolet (VUV) radiation. Excimer formation is favored by high collision rates at elevated pressure and efficient excitation or ionization of precursor



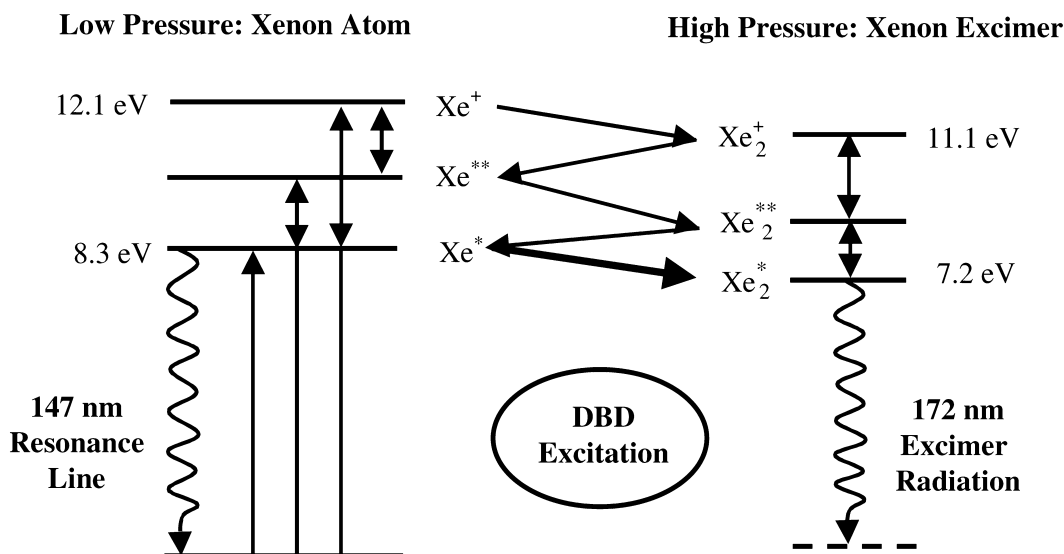
**Fig. 4** Ozone generators for water treatment (by courtesy of Ozonia Ltd).



**Fig. 5** Large ozone generator producing 60 kg ozone/h (by courtesy of Ozonia Ltd).

species, which requires nonequilibrium discharge conditions. DBDs conveniently combine these two requirements. Typical examples are the formation of  $\text{Xe}_2^*$ ,  $\text{KrCl}^*$  or  $\text{XeCl}^*$  excimer complexes, radiating at 172 nm, 222 nm or 308 nm, respectively. In *excimer lamps* such excimer forming gas mixtures are sealed in quartz vessels whose walls act as dielectric barriers. Transparent or perforated external electrodes are used through which the UV radiation can exit. These *electrodeless* lamps can be operated at high power densities and operating frequencies of a few hundred kHz. A considerable fraction of the discharge power can be converted to UV or VUV radiation. Excimer UV sources provide high intensity narrow-band UV radiation at various specific wavelengths [45–54]. Since, under special discharge conditions,  $\text{Xe}_2^*$  excimers reach theoretical VUV conversion efficiencies of 50–80% [55–57] and large volume applications are foreseen in VUV sources and in plasma displays the system is discussed briefly (Fig. 6).

Excitation of xenon atoms by electron collisions to the  $\text{Xe}^*(^3\text{P}_1)$  state gives rise to 147 nm resonance line radiation. At low current densities and low pressure ( $p < 100$  Pa) this process is very efficient ( $\eta \approx 70\%$ ). The intensity, however, is very limited due to self-absorption in the gas. If, on the other hand, the pressure is raised to the order 1 bar the excited  $\text{Xe}^*$  atoms react in a three body collision to form the  $\text{Xe}_2^*(^1\Sigma_u^+, ^3\Sigma_u^+)$  excimer complexes which radiate at 172 nm (2nd excimer continuum). This excimer



**Fig. 6** Xenon resonance and excimer VUV radiation from dielectric-barrier discharges.

system can be pumped at very high power densities ( $> 1 \text{ MW/cm}^3$ ) and is not subjected to self-absorption because the excimer has no stable ground state [46,58]. Thus, powerful VUV sources in the form of excimer lamps can be designed and sufficient radiation can be generated in the tiny volume of a plasma display pixel cell.

### Fluorescent lamps and plasma display panels

Efficient generation of VUV excimer radiation can also be utilized in fluorescent lamps and in plasma display panels (PDPs). With the aid of phosphors the UV radiation can be converted to visible light. It is now possible to produce mercury-free fluorescent lamps with obvious advantages for the environment. Excimer fluorescent lamps filled with a mixture of xenon and neon are already in use in copying machines. Also flat planar large-area excimer fluorescent lamps have been developed for applications like back-illumination in liquid crystal displays. As far as the future market potential of DBDs is concerned the most important use will be in ac plasma display panels used for flat, large-area, wall hanging TV sets with 1 m to 1.5 m picture diagonal. These displays combine the original idea of the plasma display proposed by Bitzer & Slottow [59] in 1966 with the more recent experience about efficient generation of excimer radiation. In tiny addressable DBD cells of  $100 \mu\text{m}$  electrode spacing and  $200 \mu\text{m}$  width xenon VUV radiation is converted to red, green or blue image points by activating internally applied phosphor layers. Gas mixtures containing 5–10% Xe in Ne or He are used at pressures of 50–70 kPa. The operating voltage is only 200–300 V. Multi-billion dollar investments in production facilities for these flat television screens in Japan and South Korea have recently led to a new dimension of large-scale industrial dielectric-barrier applications. In 1998 already about 50 000 1 m PDPs were sold. The market volume is expected to increase to 5 million sets in the year 2005 [60].

### Applications of excimer UV radiation

Intense ultraviolet radiation can initiate photophysical and photochemical processes by breaking molecular bonds and by modifying surface properties [16,47,61–67]. It can e.g. be used for UV curing applications to induce photo-polymerization of special paints, printing inks, varnishes and adhesives. Important novel applications, especially for the shorter UV and VUV wavelengths, were found in materials processing, surface modification [68,69], thin film deposition [70–74] and in pollution control [75,76]. Photo-induced structured metal deposition, and the formation of semiconducting or insulating thin films have become important processes. In addition to the well-established oxidation of polymers also low temperature oxidation of silicon could be achieved with the aid of VUV radiation from an

excimer lamp [77]. These intense novel ultraviolet sources offer an enormous potential for industrial materials processing.

### High power SD CO<sub>2</sub> lasers

In Japan dielectric-barrier discharges are also extensively used to pump high power CO<sub>2</sub> lasers [8,78,79]. Nearly diffraction limited infrared radiation at 10.6  $\mu\text{m}$  is obtained at power levels up to 10 kW with an efficiency surpassing 10%. The main application of these SD CO<sub>2</sub> lasers (SD stands for silent discharge) is high speed welding and cutting of metal plates and other materials. Water-cooled plane metal electrodes, forming a discharge space of 20–50 mm height, are covered with glass or alumina dielectrics. A high-velocity transverse gas flow passes the discharge gap at a speed 50–80 m/s for efficient heat removal and discharge stabilization. The discharge is operated at a frequency of about 200 kHz in a CO<sub>2</sub>/N<sub>2</sub>/He mixture at the reduced pressure of about 10 kPa. Under these conditions the discharge looks fairly homogeneous.

### SURFACE TREATMENT AND POLLUTION CONTROL

Polymer surfaces are often smooth and difficult to wet. It is difficult to apply glues, to paint or print on them or to deposit adherent metal coatings. It is possible to *activate* such surfaces by a DBD treatment in air and certain other gases at atmospheric pressure. The original approach to use dc corona discharges with bare metal electrodes [80] has been replaced by DBD configurations in which at least one of the electrodes is covered by a dielectric [16,65,81]. This way much higher specific powers can be applied. Plastic foils up to 10 m wide are treated at speeds up to 10 m/s. Typical powers of the order 100 kW are required in such applications. Preferred operating frequencies are in the range 20–50 kHz. Also the deposition of thin coatings by using reactive gas mixtures and the removal of photoresist layers in DBDs has been investigated [82,83]. Recent investigations concentrate on the treatment of wool and textiles with DBDs.

Applications of DBDs to the destruction of poisonous compounds and to pollution and odor control in general have received growing attention [16,84–86]. An increasing number of investigations are devoted to the decomposition of nitrogen and sulfur oxides in flue gases, and of volatile organic compounds (VOCs) emanating from various industrial processes. Many hazardous organic compounds are readily attacked by excited species, free radicals, electrons, ions and/or UV photons generated in DBDs. For some applications it is advantageous to fill the discharge space with dielectric pellets [87–90]. These pellets can in addition be coated with special catalysts that promote the destruction. Also the combination of DBDs with conventional catalytic processes is investigated.

### CONCLUSION AND OUTLOOK

Intensive research activities originally oriented towards a better understanding of the ozone formation process in dielectric-barrier discharges and their fundamental properties have resulted in a number of novel, often quite unexpected, DBD applications. These are now predicted to reach annual market volumes of several billion US\$. No doubt, further interesting DBD applications will develop in the future. Among the future oriented research topics investigated currently are the conversion of the greenhouse gases CO<sub>2</sub> and CH<sub>4</sub> to syngas or liquid fuels [91–94] and to the generation of H<sub>2</sub> and elemental sulfur from H<sub>2</sub>S.

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