FILAMENTARY AND DIFFUSE BARRIER DISCHARGES

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1   Introduction

Barrier discharges, sometimes also referred to as dielectric-barrier discharges or silent discharges, are characterized by the presence of at least one insulating layer in contact with the discharge between two planar or cylindrical electrodes connected to an ac power supply (Fig. 1). The main advantage of this type of electrical discharge is, that non-equilibrium plasma conditions in atmospheric-pressure gases can be established in an economic and reliable way. This has led to a number of important applications including industrial ozone generation, surface modification of polymers, plasma chemical vapor deposition, excitation of CO_2 lasers, excimer lamps and, most recently, large-area flat plasma display panels [1-4]. Depending on the application, the width of the discharge gap can range from less than 0.1 mm to about 100 mm and the applied frequency from below line frequency to several gigahertz. Typical materials used for the insulating layer (dielectric barrier) are glass, quartz, ceramics but also thin enamel or polymer layers.
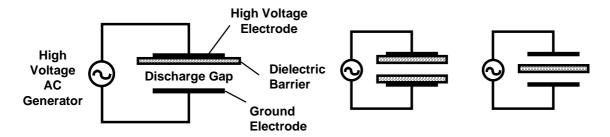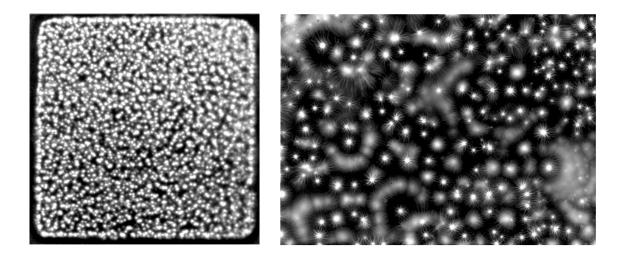

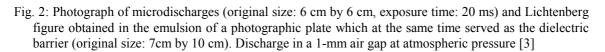Fig. 1: Typical barrier discharge configurations

Many investigations showed that at atmospheric pressure electrical breakdown in such an electrode configuration occurs in a large number of short-lived current filaments referred to as microdischarges. Most of the industrial applications of barrier discharges operate in this filamentary mode. In 1968 Bartnikas showed that helium ac discharges between closely spaced plane parallel electrodes, metallic or covered with a dielectric layer, can exhibit also pulsed or pulseless glow, or what he termed "pseudoglow" character [5]. Three years later he reported similar observations in air and nitrogen discharges [6]. In 1976 Kevin Donohoe at Caltech [7, 8] showed that he could obtain a diffuse plasma in a repetitively pulsed barrier discharge in a mixture of helium and ethylene at much wider electrode spacing (several cm). Also the SD (silent discharge) CO_2 laser first proposed by Mitsubishi researchers in 1981 [9-13] is characterized by a diffuse plasma region between dielectric-covered electrodes of wide spacing (4-5 cm). Again, a large fraction of helium was used and, in addition, a reduced pressure. In the mean time many researchers have obtained diffuse plasmas in barrier discharges in different atmospheric-pressure gases including helium, neon, argon nitrogen, oxygen and air. Beginning in 1987 extensive work in this field was published by the group of S. Okazaki [14-18] at Sophia University in Tokyo, Japan, by F. Massines and coworkers [19-

24] in Toulouse, France, and by J. R. Roth and his group [25-28] at the University of Tennessee, USA. Today we are led to believe that, depending on the gas mixture, the surface properties and the operating conditions, different discharge modes including filamentary, patterned, or completely diffuse barrier discharges can exist.

2   Filamentary discharges and microdischarge properties

2.1   Formation of an isolated microdischarge channel

In 1932 Buss noticed that electrical breakdown of a narrow air gap between parallel planar electrodes covered with a dielectric occurred in a large number of constricted bright current filaments [29]. Roughly 10 years later Raether [30, 31] and Loeb and Meek [32] independently presented their concepts of the mechanism of discharge initiation at high pressure, the streamer breakdown (Kanaldurchbruch), which is quite different from the mechanism Townsend had proposed for the initiation of a low pressure glow discharge [33].

Fig. 2: Photograph of microdischarges (original size: 6 cm by 6 cm, exposure time: 20 ms) and Lichtenberg figure obtained in the emulsion of a photographic plate which at the same time served as the dielectric barrier (original size: 7cm by 10 cm). Discharge in a 1-mm air gap at atmospheric pressure [3]

At atmospheric pressure, due to the high collision rates, a growing electron avalanche can generate appreciable charge density at its tip, already after traveling a short distance. The local "eigenfield" due to charge separation resulting from the difference in drift velocities of electrons and ions is superimposed on the applied field. Collisional ionization in the high field region at the streamer head leads to fast propagation of the ionization region and the formation of a bright plasma channel. The streamer velocity is in the range 10^7-10^8 cm/s, the radius of the propagating streamer head and the resulting ionized channel is about 10^{-2}-10^{-1} cm. The reduced field E/N at the streamer head can reach 500 to 800 Td (1 Townsend or Td corresponds to 10^{-17} Vcm^2), a value that is several times higher than the reduced field at breakdown (about 100 to 200 Td). In atmospheric-pressure air the thickness of the propagating ionizing region is only 0.02 cm. Ultraviolet photoinization of the gas ahead of it is the most likely process to provide seed electrons for the propagating streamer.

The early phases of breakdown in a barrier discharge are similar to those without dielectric. In this phase concepts of streamer propagation between metal electrodes as, for example, recently reviewed in detail by Babaeva and Naidis [34], are helpful. Streamer

propagation in a given electrode configuration is normally simulated by solving 2D hydrodynamic equations for neutral and charged particles coupled with Poisson's equation for the determination of the electric field. To check certain assumptions, like for example the widely used "local field approximation", assuming electrons in equilibrium with the local field, also more detailed Monte Carlo simulations have been performed. Also simple analytical approximations can yield reasonable results about streamer formation and propagation that agree with numerical 2D models [35].

A far as barrier discharges are concerned charge deposition on the dielectric surface becomes a dominant feature when the streamer has bridged the gap. This necessitates the introduction of special boundary conditions, different from those of free streamer propagation. Models treating the formation of a microdischarge in a gas gap bounded by a dielectric surface were formulated by different authors [36-51]. In these models a feedback mechanism at the cathode surface (metal or dielectric) is incorporated. Impinging ions and photons produce secondary electrons that are fed into the discharge channel. At the time the cathode directed streamer reaches the cathode a cathode fall region is established within a fraction of a nanosecond. At atmospheric pressure the thickness of this high field region is only a few micrometers [43, 44, 51]. The current peak in a microdischarge is reached at the time of cathode layer formation. Immediately afterwards charge accumulation at the dielectric surface leads to a local collapse of the electric field in the area defined by the surface charge. This area covered by the surface discharge is considerably larger than the channel diameter. The self-arresting effect of the dielectric barrier limits the duration of a microdischarge to a few ns, the transferred charge to typically 100 pF and the dissipated energy to a fraction of a μJ (for a 1-mm gap at 1 bar). The limited energy dissipation limits the rise of the gas temperature due to a single microdischarge to a few Kelvin, while the electrons in the microdischarge channel have mean energies of a few eV. In the channel electron densities of 10^{14} cm^{-3} and current densities of j ≈ 1000 Acm^{-2} are reached [37, 40]. These values correspond to what one would expect if typical current densities of low pressure glow discharges were scaled to atmospheric pressure by the similarity relation j/p^2 = const. [52]. The presence of a high field region at the cathode, the typical current density and the non-equilibrium plasma conditions justify the characterization of these microdischarges as transient high-pressure glow discharges.
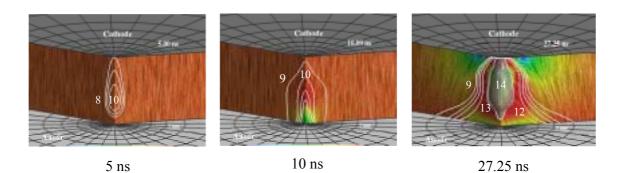

| 5 ns | 10 ns | 27.25 ns |

Fig. 3: Two-dimensional numerical simulation of microdischarge formation in a mixture of 80% H$_2$ and 20% CO$_2$ at atmospheric pressure in a 1-mm gap bounded by 1 dielectric at the anode (bottom) [4]

Fig. 3 shows three stages, at 5ns, 10 ns and 27.25 ns, of the early phases of microdischarge development in a gap of 1 mm width in a mixture of 80% H$_2$ and 20% CO$_2$ at 1 bar and 300 K. The metal cathode is located at the top and a dielectric anode at the bottom. Closed isolines indicate the electron density per cm^3 in powers of 10. The electric field lines are also indicated. Initial electrons were released at the cathode in an originally homogeneous field of

34 kV/cm. The growing electron avalanche travels towards the anode, soon reaches the streamer state, initiates a cathode directed streamer, and finally results in the formation of a conductive microdischarge filament. At this stage maximum electron densities of 10^{14} cm^{-3} are reached in a channel of about 0.2 mm diameter. The widening of the area of the surface discharge is clearly seen in the picture to the right. Model calculations suggest typical charge densities of the order 10-20 nC/cm^2. For an isolated microdischarge on an extended dielectric surface the area of the surface discharge will adjust its size to accommodate the total current (conduction and displacement current) at all times. Its area determines the effective capacitive ballast of the microdischarge.

2.2 Experimental determination of microdischarge properties

Many fundamental microdischarge properties were determined from Lichtenberg figures, image converter recordings [53-55], current measurements [37, 43, 52, 56, 57] and charge measurements [58]. Coogan and Sappey [59] imaged the distribution of OH radicals in a barrier discharge operated in an Ar/O$_2$/H$_2$O mixture by recording their emission in the wavelength range 308-312 nm and laser induced fluorescence at 281.9 nm. OH radicals could be detected only within the 170-μm diameter channels formed by transient microdischarges. Running the discharge at a moderate power density of 0.1W/cm^2 about 10^6 microdischarge channels per cm^2 per second were counted. A significant increase in the density of microdischarges (more than two orders of magnitude) was observed when additional UV radiation used. More recently also in situ electron densities [60], atom and radical densities, excited species concentrations and gas temperatures have been determined by spectroscopic techniques [61-69]. The spatiotemporal evolution of different species in a microdischarge channel can also be revealed by cross correlation spectroscopy, a powerful diagnostic tool first proposed by Kozlov and coworkers in 1995 [70, 71]. The experimental difficulty is to stabilize a single microdischarge sufficiently to do repetitive measurements or to arrange a linear array of microdischarges on a knife edge electrode. In many cases pulsed experiments are preferred for such measurements. Some questions remain whether an isolated microdischarge is representative for what happens in a barrier discharge and whether the conditions in a pulsed filament are identical to those of a filament initiated by a slowly rising voltage.

2.3 Collective phenomena of microdischarges in a barrier discharge

The reality in a barrier discharge is, of course, quite different from that of an isolated microdischarge. In a typical discharge situation individual microdischarges compete for the available surface area of the dielectric to deposit their charge patterns. They also encounter residual charges from previous discharge phases. First attempts of treating multistreamer dynamics by numerical simulation have recently been put forward [72]. Despite of the complexity of the problem there exists extensive practical experience, e. g. from investigations on industrial ozone generation and on pollution control. The number density of microdischarge locations at the electrode surface rises with power density, which depends on the applied frequency and the peak voltage, but not on the form of the applied voltage. During the rising part of the external voltage additional microdischarges are initiated at new locations, because the presence of residual charges on the dielectric has reduced the electric field at positions where microdischarges have already occurred. On voltage reversal, however, the next microdischarges will form at old microdischarge locations. Since the voltage has collapsed at these locations it takes less external voltage swing to reach the breakdown field in

the following half period of opposite sign. Consequently, high voltage/low frequency operation tends to distribute the microdischarges on the available dielectric surface, while low voltage/high frequency operation tends to re-ignite the old microdischarge channels every half period. This memory effect due to charge accumulation on the dielectrics is a dominant feature in all barrier discharges.

Over a wide range of parameters individual microdischarge properties are independent of the external circuit. This is no longer the case when the voltage rise time becomes so short that so many microdischarges are started simultaneously, that there is not enough available surface area on the dielectric for their surface discharges. Extremely fast rising voltage pulses can also result in a synchronization of the microdischarges and an overshoot of the stationary breakdown voltage. Additional changes occur when the repetition frequency is so high that there is not enough time between consecutive half periods to allow for complete removal of charged and excited species in the microdischarge channels before the next ignition.

3   Pattern formation in barrier discharges

In a confined geometry microdischarges with their surface discharges can be quite regularly arranged. The simplest configuration is a 1D geometry that was recently proposed by M. Walhout's group at Calvin College, Grand Rapids, MI, USA [73]. This group succeeded in producing 1D arrays of regularly spaced microdischarges in a glass tube of 2 mm inner diameter, 7.5 mm outer diameter and 30 cm length. Linear strips of silver paint, 3 mm wide, were applied on diametrically opposed portions of the outer surface and connected to an ac source delivering a sinusoidal voltage at 16 kHz. The discharge was operated in a He/Ar mixture at 1 bar. At low voltages regularly spaced microdischarges were obtained with about 8 mm spacing (Fig. 4). When the peak voltage is raised to the range $610V<V_0<940V$ additional microdischarges are striking in between, thus effectively cutting the spacing in half. At higher voltages every second microdischarge appears blurred and above 1200V the whole discharge appears blurred. The authors conclude that in the blurred regions the individual filaments strike randomly at apparently uncorrelated positions, which may be suggestive of spatiotemporal chaos.

Fig. 4: Array of microdischarges in a 1D barrier discharge configuration in Ar/He [73]

Regular patterns can also be observed in 2D barrier discharge configurations. In 1982 Boyers and Tiller [74] published a paper on plasma bubble domains obtained in atmospheric-pressure helium confined between large-area, closely spaced, plane-parallel, dielectric-covered electrodes. Typical discharge gap widths were 50 to 150 μm, the operating frequency was 125 kHz. The authors found that the discharge can form either a dim uniform Townsend discharge, an assembly of regularly spaced glow discharge plasma domains, or a bright uniform normal or abnormal glow discharge. More recently, the phenomenon of pattern formation in barrier discharges was studied in detail by the group of H.-G. Purwins at the University of Münster, Germany [75-78]. About 1991 we also observed diverse regular

discharge patterns in a planar 5 mm wide discharge gap in a round quartz cell of 100 mm diameter with parallel walls of 5 mm thickness [2]. The discharge structure could be observed through a transparent ITO (indium tin oxide) electrode deposited on one of the outside walls. When in a Xe/Cl$_2$ gas mixture the mixing ratio and pressure was varied, regular, fairly stationary patterns could be recorded (Fig. 5). The operating frequency was about 1 kHz. The picture at the right looks very much like a photograph of Bénard convection cells published by Koschmieder [79]. Quite recently, the formation of regularly patterned glow regions in barrier discharges was also reported by Miralaï et al. [80]. It appears that the formation of discharge patterns in barrier discharges is a manisfestation of spatiotemporal pattern formation in a system driven away from equilibrium. Convection instabilities and magnetic bubble domains are other examples. An excellent extensive general review of the subject was published by Cross and Hohenberg [81].

Fig. 5: Two-dimensional discharge patterns obtained in 5-mm quartz cell with planar walls filled with xenon/chlorine mixtures at different pressures (visible diameter: 8 cm)

4 Diffuse barrier discharges

It has repeatedly been demonstrated that also diffuse discharges can be obtained in barrier discharge configurations, even at atmospheric pressure and wide gap spacings up to several centimeters. S. Okazaki and coworkers did extensive work in this field using sinusoidal feeding voltages in different gases with and without additives [14-18]. They proposed the term APG, standing for atmospheric pressure glow. To generate uniform discharges at atmospheric pressure in helium, argon, air, oxygen and nitrogen, even when using a 50 Hz sinusoidal feeding voltage, they used an electrode configuration consisting of two metal foils covered with a special metal mesh and thin ceramic plates or Mylar foils serving as dielectrics [18]. These surprising experiments have been repeated by different groups with similar results [82-84]. More detailed investigations clarifying the mechanism of this discharge mode and the development of 1D models for homogeneous discharges followed by F. Massines and her group [19-24]. Apparently independently of these investigations, the group around J.R. Roth proposed what they called the OAUGDP (one atmosphere uniform glow discharge plasma) and investigated a number of different industrial applications [25-28, 85-87]. Novel applications for mild diffuse discharges are especially expected for biological and medical purposes [88-91]. In many publications the appearance of a single current peak per half period or a single step in the charge measurement is taken as an indication for obtaining a diffuse discharge.

More than 70 years ago German researchers observed already glow discharges between metal electrodes in atmospheric-pressure gases [92, 93]. Intermediate glow-like discharge phases were also observed in the early stages of high-pressure breakdown in nitrogen and hydrogen [94-96]. The subject became a major issue when transversely excited atmospheric pressure lasers (TEA lasers) were investigated in N$_2$, in CO$_2$/N$_2$/He mixtures, and in excimer forming gas mixtures [97-100]. It was found that sufficient preionization by x-rays, UV

radiation, electron beams or by double-discharge techniques helped to establish a uniform glow phase that could be extended up to 1 μs [101-104]. Necessary requirements of a minimum initial electron density were formulated by Palmer [105] and by Levatter and Lin [106, 107]. One requirement for establishing a volume-stabilized diffuse discharge is that the preionization electron density is large enough to cause appreciable overlap and coalescence of primary avalanche heads and smoothing of space-charge field gradients at the stage when streamer formation would otherwise occur. The subject of filamentation in high pressure glow discharges between metal electrodes for laser applications was reviewed by Bötticher [108].

In 1997 Brenning and coworkers [109] formulated more detailed conditions for obtaining homogenous high-pressure pulsed avalanche discharges. They stress the importance of an additional minimum preonization rate just prior to and during breakdown. The most important quantity is the effective primary ionization coefficient α_{eff} (including one and two step ionization, and all attachment and detachment processes) at the moment of breakdown or, more precisely, its partial derivative with respect to the reduced field: $\partial(\alpha_{eff}/n)/\partial(E/n)$. This quantity is strongly influenced by impurities, gas additives and the presence of metastables and residual electrons or ions.

In dielectric-barrier discharges we can make use of residual species from the previous half period. Thus, in addition to the already mentioned memory charges on the dielectric surface(s), we also have a memory effect in the gas volume if the repetition frequency is properly chosen. The mechanism of diffuse barrier discharges in He and in N_2 is now fairly well established. Combining electrical current/voltage measurements, short time (10 ns) exposure photographic recordings and spectroscopic measurements with extensive numerical modelling Massines and her coworkers came to the conclusion that in helium barrier discharges residual electrons, trapped in the low field region of the positive column, are still present when the subsequent discharge is initiated. In nitrogen a different mechanism, based on the presence of residual metastables, generates sufficient seed electrons at the beginning of the next half period. In both cases the time between consecutive discharge periods has to be short enough to ensure sufficiently high concentrations of these precursors. Impurities, originally already present in the gas or introduced by etching material from the surface of the dielectric, have a strong influence on the life time of metastables [110, 111] and on the surface conductivity of the dielectric [112]. It has been demonstrated that 500 ppm O_2 or 2500 ppm H_2 in nitrogen can completely change discharge behaviour from a diffuse to a filamentary appearance [113-115].

Also special dielectric properties can help to establish a homogeneous volume discharge. Tepper et al. [83,84] found that electret dielectrics are capable of accumulating appreciable amounts of charges on the surface. Supported by the applied voltage the charges are trapped uniformly on the surface. When the electric field changes its polarity and exceeds a certain threshold value, the charge carriers are expelled spontaneously from the surface and initiate a homogeneous high-current-density barrier discharge ($j \approx 300$ mA/cm^2). In this high current mode the single current peak per half wave lasts only a few nanoseconds while in the normal diffuse mode 3 μs are recorded in He and about 200 μs in nitrogen discharges [116]. The respective current densities are ≈ 5 mA/cm^2 and ≈ 0.5 mA/cm^2.

Active control of the deposited charges by changing the surface or bulk conductivity of the dielectric layer and control of the feed back mechanism by selecting materials with a high coefficient of secondary electron emission has also been attempted [80]. Interesting effects have also been obtained by using semiconducting materials for the electrodes or by using perforated dielectrics which result in a large number of capillary jets that merge into a diffuse large volume discharge [117]. Also with high-resistivity materials on the electrodes diffuse discharges were obtained at atmospheric pressure [118].

5   Conclusions

Barrier discharges can exhibit a variety of different discharge modes. In the filamentary mode current densities are reached that compare favorably with the highest current densities reached in pulsed glow discharges between metal electrodes [119]. These conditions, however, are restricted to the microdischarge channels and last only for a few nanoseconds.

Also diffuse discharges can be obtained in dielectric barrier configurations if special conditions are met. They depend on a minimum repetition frequency and on the presence of metastables or residual charge carriers. Apparently homogeneous, diffuse discharges can exist at vastly different current densities. Not all of them qualify as glow discharges, which are characterized by a localized high-field cathode fall region. Low current diffuse barrier discharges are more like Townsend discharges in which the charge density is so low that there is has practically no influence on the applied electric field. The observed stable discharge patterns obtained under certain conditions presumably present an intermediate discharge mode in which a number of smaller diffuse discharge areas are arranged in a regular pattern.

As far as industrial applications are concerned, except for the SD CO_2 laser, barrier discharges are operated in the filamentary mode. Diffuse barrier discharges would provide interesting new aspects if reliable control can be achieved and if average power densities can be achieved comparable to those of filamentary discharges. The prospects of tuning the repetition rate of a homogeneous discharge to the speed of a gas flow and to the rates of chemical reactions occurring in the plasma are very attractive. Also, for a homogeneous treatment of surfaces or for the deposition of thin films diffuse discharges have obvious advantages over filamentary discharges.

6   References

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