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25 YEARS OF MICROPLASMA SCIENCE AND APPLICATIONS: A STATUS REPORT

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Abstract. Microplasmas gained recognition as a distinct area of research within the larger field of plasma science about 25 years ago. Since then, the activity in microplasma research and applications has continuously increased. This paper provides a summary of some of the pertinent developments of microplasma sources that have contributed to making this field an exciting and rapidly growing area of interest, both in terms of scientific challenges and technological opportunities.

Keywords: microplasma, discharge, plasma, microhollow cathode.

1. Introduction

Microplasmas, defined as plasmas with at least one dimension in the submillimeter range, include microarcs and microsparks, which are generated by electrical breakdown in gases and liquids. Research into nonthermal glow-type microplasmas with gas temperatures much below electron temperatures has undergone an enormous growth in the past 25 years based on the fact that such discharges can be operated stably at high gas pressures, in rare gases as well as in molecular gases, in a direct current (dc) mode as well as in pulsed dc and alternate current (ac) mode over a wide range of frequencies. Modeling and improved diagnostics have allowed us in the past decade to gain more insight into the physics of nonthermal microdischarges. Simultaneously, the range of applications has grown at a rapid pace.

For glow discharges, high pressure and small size are mutually consistent based on similarity laws, which hold for breakdown voltage (Paschen law) as well as sustained glow discharges. The minimum axial dimension of a glow discharge between two parallel-plane electrodes is given approximately by the cathode fall length of a normal glow discharge.

Shorter lengths are possible, but require a drastically increased voltage. According to the similarity laws, the cathode fall length is inversely dependent on the pressure, p. Minimizing the axial dimension of a normal glow discharge thus requires maximizing the pressure. Assuming an average value of 1 Torr.cm for the product of cathode fall thickness and pressure, reducing the thickness of the plasma to 100 µm requires a pressure of about 100 Torr, for 10 µm to more than atmospheric pressure [5]. This assumes that the electron generation at the cathode is due to ion impact. Studies on microgaps [1, 2] showed that for gaps in the micrometer range, the breakdown voltage actually decreases with decreasing electrode spacing instead of increasing, as expected from the Paschen law 1. This is likely caused by pure field emission and, in the transition region (up to about 10 µm), through

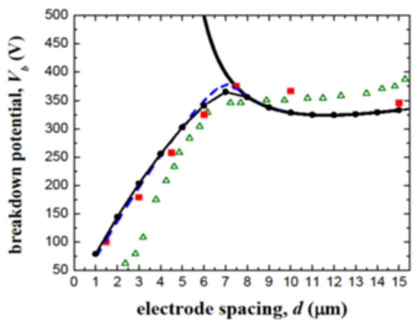

Figure 1. Breakdown potential vs. electrode spacing. Solid curve: Paschen curve for atmospheric pressure air (secondary emission coefficient Îş = 0.0075). Experiments [1] show a strong deviation from Paschen's law at very small gaps, which is supported by recent calculations [2–4].

ion-enhanced field emission caused by the presence of positive ions near the cathode surface [2].

Whereas the minimum axial dimension of a glow discharge is likely determined by the surface roughness of the electrodes, the limitations in the transverse direction is determined by the transition from a normal glow discharge to a subnormal glow discharge. Reducing the current in a normal glow discharge causes a reduction in the diameter of the cathode fall area. When the diameter reaches values on the order of the cathode fall length (dark space at the cathode), the discharge voltage rises [6]. This increase is due to the increased radial losses of charged particles. Based on these considerations, and assuming that the thickness of the cathode fall region is determined by values of pD in the range of 0.5 to 1 Torr . cm, the minimum size of an atmospheric pressure glow discharge in the lateral direction is about 10 µm. Over the past 25 years, several types of microdischarges were developed and characterized and are used in many technological applications. Below is a summary of some commonly used microdischarge configurations.

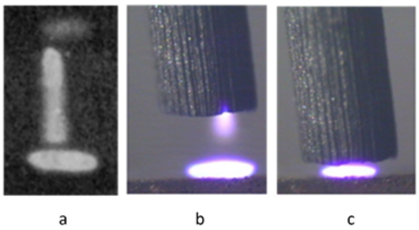

Figure 2. (a) Glow discharge in air between cooled Cu electrodes 2 mm apart (anode: top, cathode: bottom) [7]. The radially extended luminous area close to the cathode is the negative glow. (b) and (c) Images of a glow discharge in air at (b) 0.5 mm, (c) 0.1 mm electrode spacing [8].

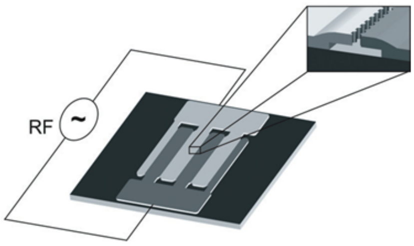

Figure 3. Schematic view of a micro-structured electrode (MSE) array with the electrode gap design shown in detail [11].

2. Microdischarge Configurations

This section gives a brief overview of the various microdischarge source configurations and modes of excitation that have been developed and characterized [5].

2.1. Microdischarges between Parallel-Plate Electrodes

The various geometrical electrode arrangements and power schemes for microdischarges between parallel-plate electrodes include:

2.1.1. DC Micro-glow Discharges between Parallel Plate Electrodes

An image of an atmospheric-pressure "micro" glow discharge in air with a gap of 2 mm is shown in 2(a) [7]. Studies with smaller gaps show the presence of a positive column and a very distinct extended luminous area, the negative glow for gaps larger than 0.1 µm (2(b)) [8]. For gaps as small as 0.1 µm, the positive column has disappeared, and the discharge in axial direction consists of cathode fall and negative glow only (2(c)).

The quadratic increase in current density with pressure is a problem in operating high-pressure micro-glow discharges. The power density in a microdischarge with dimensions of the cathode fall ($\sim$ 100 µm in atmospheric pressure air) and, assuming a cathode fall voltage of 150 V, exceeds 10^5 W/cm^3. This causes strong heating of the plasma and eventually the transition from a glow discharge to an arc (glow-to-arc transition). The earliest attempts to generate (micro) glow discharges between parallel plate electrodes at atmospheric pressure used cooling of the cathode to limit the plasma temperature. Another way to prevent such a transition is to limit the duration of the discharge to times shorter than the time required for the development of such an instability, usually times below one microsecond.

2.1.2. DC Micro-Glow Discharges between a Flat Cathode and Ring-Shaped Anode

By replacing the parallel plate anode with a ring-shaped anode surrounding the negative glow, i.e. separated from the cathode only by a distance on the order of the cathode fall, the negative glow is used as a virtual anode, carrying not only an axial current as in conventional glow discharges, but also a transverse current toward the physical ring-shaped anode. This type of discharge was named cathode boundary layer discharge (CBL discharge) [9, 10].

2.1.3. AC (Microwave) Micro-glow Discharges between Parallel Edge Electrodes

Radiofrequency (rf) sustained microdischarges in planar electrode systems, bonded to dielectric substrates, also allow to generate plasmas in atmospheric-pressure gases. An electrode system (Microstructured Electrode Array) is shown in 3 [11, 12]. The electrode gap has a width of 70 µm. With this electrode gap it was possible, using a 13.56 MHz generator, to ignite microdischarges at voltages below 390 V even in atmospheric pressure air.

Increasing the frequency into the microwave range provides new opportunities for the generation of microdischarges [13]. At low frequencies and/or small gaps, the amplitude of the electron oscillation is larger than the discharge gap, and the electrons are lost to the electrode. This mode of operation is called the Îş-mode. At high frequencies and/or larger gaps, the amplitude of the electron oscillation is less than the gap length and the electrons are trapped in the in the discharge gap (α-mode), where the ionization processes are confined to the bulk of the plasma. For atmospheric-pressure Ar microplasmas in a gap of 100 µm, the frequency needs to extend 250 MHz to operate the discharge in the α-mode.

2.2. From Planar to Microcavity Electrode Structures

The simplest electrode configuration, plane-parallel metal electrodes, does not satisfy the requirement of a positive differential resistance, and is, consequently, inherently unstable. Parallel operation requires a ballast resistor in series to the discharge. A way to stabilize a high-pressure microdischarge is to use a built-in loss mechanism. By confining the plasma in small metallic cavities (the electrodes) and dielectric cavities (between the electrodes), thermal conduction to the electrodes and diffusion to the walls of the dielectric layer between the electrodes damps small plasma fluctuations, which otherwise would lead to instabilities. For high-pressure discharges, these cavity dimensions need to be around 100 µm. This approach allows to extend the pressure and current density range and to generate stable glow discharges at pressures up to and above one atmosphere, even in molecular gases without using ballast resistors.

2.2.1. Microhollow Cathode (Microcavity) Discharges

Stable dc glow discharges in small cavities was first reported by White [14], who studied hollow cathode discharges in 100 Torr Ne with the cathode containing a nearly spherical cavity of 750 µm diameter and a pin anode. In the mid 1990s, stable discharges in air at pressures of 350 Torr were generated by reducing the dimensions of the cylindrical hole in a molybdenum cathode to a diameter of 75 µm [14]]. Since the depth of the cathode hole had only a minor influence on the discharge characteristics, the cathode hole was reduced to a ring-shaped cathode with a thickness comparable to the hole diameter. This “sandwich” micro-hollow cathode (MHC) discharge configuration was subsequently also used by others, but sometimes under a different name, such as 3D Microstructured Electrode (MSE) discharge [15]. In terms of mode of excitation, microcavity discharges can be excited by DC, AC, or pulsed power.

DC operation of microcavity discharges

The phrase “hollow cathode discharge” historically refers to a specific mode of discharge operation, where the cathode fall regions in the interior of the cylindrical cathode are so close that high energy electrons emitted from one side of the cathode can enter the opposite cathode fall, where they are accelerated back towards the axis. This “pendulum” motion of the electrons leads to increased ionization, which appears as a negative differential resistance [16]. For a 200 µm wide cathode hole, hollow cathode discharges can exist for pressures below 100 Torr. In experiments at higher pressures, but similar hole diameters, the luminous plasma develops along the edge of the hole or even outside the hole. Although there is still a range of currents where the V-I characteristic has a negative slope, the underlying mechanism is no longer related to the hollow cathode (pendulum) effect. Modeling showed that under these conditions a positive V-I characteristics at low currents can be attributed to an abnormal glow inside the hole, and a flat part at high currents to a normal glow discharge developing on the flat surface of the cathode outside the hole [17]. The superposition of the two discharges provides a range where the voltage-current characteristic has a negative slope, similar to that obtained in hollow cathode discharges. Since these microdischarges are characterized by a different mechanism than the original microhollow cathode discharges, MHC discharges are often simply referred to as microdischarges.

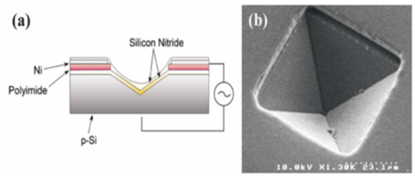

Figure 4. (a) Cross-section of a Si microplasma device with an inverted square pyramid microcavity; (b) SEM of a single device viewed from above. The emitting aperture is $50 \times 50\,\mu\mathrm{m}^2$ *[20].*

Whereas the first microcavity cathodes made from molybdenum were manufactured by mechanical drilling and later by laser drilling, other groups focused early on using microfabrication techniques. Eden and co-workers used silicon as electrode material [18]. The motivation for this approach is the ability to controllably and reproducibly fabricate a wide range of features at the micro- and even nanoscale. With this technology, it was possible to reduce the width of the cathode hole to values of 10 µm (with length of 30 µm), and to operate the discharges in Ne in a pressure range from 700 Torr to 1100 Torr [19].

AC Operation of Microcavity Discharges

Although most basic studies and modeling efforts were performed on DC microcavity discharges, efforts have also been devoted to study AC microcavity discharges. Rf-driven microdischarges in hollow slot electrode geometries were studied by Collins and coworkers [21, 22], and rf microdischarge arrays were investigated in Taiwan [23]. Planar Micro-Structured Electrode (MSE) Arrays operated at rf were successfully used to generate atmospheric-pressure microplasmas [11, 12]. With Si as cathode material coated with a thin film of silicon nitride, large arrays of mcrodischarges can be fabricated [20].

Pulsed Operation of Microcavity Discharges

Operation of single dc MHC discharges with currents exceeding about 8 mA leads to thermal damage even to molybdenum electrodes. However, by operating the discharge in a pulsed dc mode, the current can be increased considerably, often by an order of magnitude [24]. Whereas the use of pulsed dc with pulse durations in the millisecond range allows one to increase the current, ultrashort pulses allows one also to affect the electron energy distribution. By superimposing 10 ns pulses to a dc microdischarge, the electron density could be increased by more than an order of magnitude to values around $5 \times 10^{16}\,\mathrm{cm}^{-3}$.

2.3. Capillary Plasma Electrode (CPE) Discharges

The configuration of the Capillary Plasma Electrode (CPE) discharge (see 5 looks very similar to that of a dielectric barrier discharge (DBD). However, its basic properties and operating principle are based on a novel electrode design [25], which uses dielectric capillaries that cover one or both electrodes of

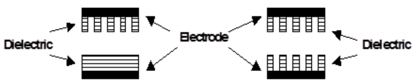

Figure 5. Schematics of a capillary plasma electrode (CPE) discharge with 1 or 2 perforated dielectrics

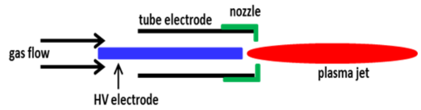

Figure 6. Schematics of a DFE jet

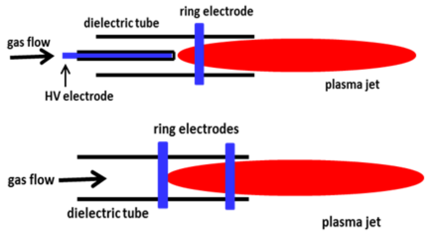

Figure 7. Schematics of two typical DBD jets

the discharge device. The CPE discharge exhibits a mode of operation not observed in a DBD, the so-called "capillary jet mode". Capillaries with diameters from 0.01 to 1 mm and length-to-diameter ratios of a 10:1 serve as plasma sources, which produce high-intensity plasma jets at atmospheric pressure under certain operating conditions. The plasma jets emerge from the end of the capillary and form a "plasma electrode" for the main plasma 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 capillary(s) in front of the electrode(s) is crucial for the "capillary jet mode". The CPE discharge has two distinct modes of operation when excited by pulsed dc or ac, a diffuse mode below a critical frequency and a mode above that frequency where the capillaries "turn on" and a bright, intense plasma jet emerges from the capillaries. This transition is manifested in a drastic increase in the electron density. CPE discharges can operate at atmospheric-pressure in He, Ar, $He - N_2$, He-Air, $He - H_2O$, $N_2 - H_2O$, and air-H_2O gases and gas mixtures and discharge volumes of more than $100\,cm^3$.

3. Plasma jets

Atmospheric-pressure plasma sources, which generate plasma in open space rather than in spatially confined discharge gaps, so-called "atmospheric-pressure plasma jets (APPJs)", have gained much prominence in the past 10 years. The need for such sources was largely driven by the need for plasma treatment of objects, whose size and shape did not allow them to be placed in the discharge gaps for direct place treatment. Conventional sources also pose a challenge for remote plasma treatment because of the short lifetime of reactive plasma species, particularly at atmospheric pressure, where the electron – heavy particle collision frequency is high. Thus, the concentration of species most effective for the plasma treatment is significantly reduced when the species reach the object, which renders the plasma treatment ineffective. Different methods were developed to address these challenges and a number of APPJ concepts have been realized. Notable summaries of APPJs can be found in Refs.[26–29].

While most APPjs operate with rare gases or rare gases with small admixtures of reactive gases, there are some APPJs that can operate in atmospheric-pressure ambient air without causing arcing. APPJs operating in rare gases fall into the categories of dielectric-free electrode (DFE) jets, dielectric barrier discharge (DBD) jets, DBD-like jets, or single electrode (SE) jets [26]. An early version of a DFE jet source is schematically shown in fig. 6 [30]. The DFE jet requires high powers to generate the plasma, which results in higher gas temperatures that make the jet unacceptable for most biomedical applications, yet suitable for materials treatments not adversely affected by high temperatures. Since the electric field in the discharge gap and in the plasma plume is relatively weak, the plasma plume is gas-flow driven and not electrically driven. Most APPJs are DBD or DBD-like jets of different designs. Fig. 7(top) shows a jet generated inside a dielectric tube with two metal ring electrodes connected to AC power operating in the kHz regime. When the operating gas flows through the tube, a cold plasma jet is ignited, which extends beyond the nozzle into the surrounding air. In the source shown in fig. 7(bottom), one of the ring electrodes is replaced by an insulated central pin electrode, which increases the electric in the direction of the plasma plume and leads to spatially elongated jets with a high concentration of active species [26].

The kINPen 09 source (Fig. 8 [27, 29] consists of a hand-held unit for the generation of a plasma jet at atmospheric pressure, a rf power supply, and a gas supply unit. The plasma is ignited at the tip of a electrode inside a capillary and blown out by the gas flow. The kINPen 09 is well suited for focused treatments of small spots. Large-scale treatments can be realized by moving the jet over a selected area. These features make it interesting for the treatment of complex geometries and cavities, e.g. in operative dentistry or dermatology.

Reports of "plasma bullets" appeared in 2005 [30]. Laroussi reported a "bullet-like" cold plasma plume with speeds above 105 m/s, much higher than the gas velocity [28].

4. Summary

Microdischarges can generate stable plasmas at atmospheric pressure. They can be operated in a di-

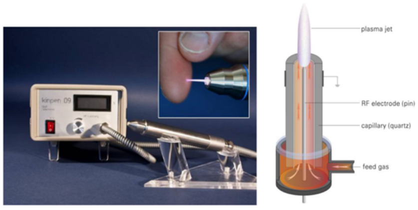

Figure 8. The INP kINPen 09 [27, 29]

rect current mode, at rf and microwave frequencies, and pulsed with pulse durations as short as a few nanoseconds. They can be operated in rare gases and molecular gases, and in gas mixtures. Their electron energy distribution contains a tail of high energy electrons, which cause high excitation and ionization rates. Electron densities can exceed $10^{16}\,\mathrm{cm}^{-3}$ in pulsed microdischarges. Gas temperatures, on the other hand, are low and range from to room temperature in rare gases and to about 2000 K in atmospheric-pressure air. With discharge voltages of a few hundred volts and current densities of $10\,\mathrm{A/cm}^2$, the power densities can reach values of about $10^5\,\mathrm{Wcm}^{-3}$. The nonthermal characteristics of microdischarges together with features such as stable high-pressure operation, high power density, non-Maxwellian electron energy distribution with a significant component of energetic electrons, low gas temperatures, and the possibility to arrange them in arrays without individual ballasting led to numerous microplasma devices for a wide range of applications, incl. large flat 2D microdischarge arrays. More recently, the generation of reactive species in microdischarges, and the ability to transport them in form of plasma jets to biological tissue, has contributed to a rapidly expanding field of research of plasma medicine.

The rapid development of microplasma applications has often occurred by trial-and-error, i.e. without an understanding of the underlying basic science. Plasma applications as diverse as surface modification and functionalization, light sources, ozone generation, pollution control, biological inactivation, sensors, etc. rely on the plasma-initiated generation of chemically reactive species, in particular reactive oxygen and nitrogen species. While we understand some key plasma chemical reaction pathways in certain applications, this insight is lacking in many others, most notably in the field of plasma medicine.

References

[1] G.P. Slade and E.D. Taylor. *IEEE Trans. Compon. Packag. Technol.*, 25(390), 2002.

[2] D.B. Go and A.D. Pohlman. *J. Appl. Phys.*, 107, 2010.

[3] W. Zhang, T.S. Fisher, and S.V. Garimella. *J. Appl. Phys.*, 96, 2004.

[4] G. Tirumala and D.B. Go. *Appl. Phys. Lett.*, 97, 2010.

[5] K. H. Schoenbach and K. Becker. *Europ. Phys. J. D*, 70(29), 2016.

[6] A. von Engel. Ionized gases, 1965.

[7] A. Von Engel, R. Seeliger, and M. Steenbeck. *Z. Physik*, 85(144), 1933.

[8] D. Staack, B. Farouk, A. Gutsol, and A. Fridman.

[9] M. Moselhy and K.H. Schoenbach. *J. Appl. Phys.*, 95, 2004.

[10] K.H. Schoenbach, M. Moselhy, and W. Shi. *Plasma Sources Science and Technology*, 13, 2004.

[11] L. Baars-Hibbe and P. Sichler et al. *J. Phys. D: Appl. Phys.*, 38, 2005.

[12] C. Schrader and L. Baars-Hibbe et al. *Vacuum*, 80, 2006.

[13] J. Hopwood, A.R. Hoskinson, and J. Gregorio. *Plasma Sources Sci. Technol.*, 23, 2014.

[14] K.H. Schoenbach and et al R. Verhappen. *Appl. Phys. Lett.*, 1996.

[15] C. Penache and et al M. Miclea. *Plasma Sources Science and Technology*, 11, 2002.

[16] G. Schaefer and K.H. Schoenbach. *Basic Mechanisms Contributing to the Hollow Cathode Effect in Physics and Applications of Pseudosparks.* Plenum Press, 1990.

[17] J.P. BÅŞuf, L.C. Pitchford, and K.H. Schoenbach. *Appl. Phys. Lett.*, 86, 2005.

[18] J.W. Frame and et al D.J. Wheeler. Appl. phys. lett. 71, 1997.

[19] S.-J. Park and J.G. Eden et al. *Appl. Phys. Lett.*, 85, 2004.

[20] J.G. Eden and S.-J. Park. *Plasma Physics and Controlled Fusion*, 47, 2005.

[21] Z. Yu, K. Hoshimiya, and J.D. Williams et al. 83, 2003.

[22] R. Rahul and O. Stan et al. *J. Phys. D: Appl. Phys.*, 38, 2005.

[23] Y.-B. Guo and F.C.-N. Hong. Appl. phys. lett. 82, 2003.

[24] M. Moselhy and et al W. Shi. *IEEE Trans. Plasma Science*, 30, 2002.

[25] K.H. Becker and B.B. Godfrey et al. *IEEE Trans. Plasma Sci.*, 43, 2015.

[26] X. Lu, M. Laroussi, and V. Puech. *Plasma Sources Sci. Techn.*, 21, 2012.

[27] K.D. Weltmann and E. Kindel et al. *Pure Appl. Chem.*, 82, 2010.

[28] X. Lu and M. Laroussi. *J. Appl. Phys*, 100, 2006.

[29] K.D. Weltmann and E. Kindel et al. *Contr. Plasma Phys.*, 49, 2009.

[30] M. Teschke and J. Kedzierski et al. *J. Engemann, IEEE Trans. Plasma Sci.*, 33, 2005.