Breakdown and discharge regimes in standard and micrometer size dc discharges

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Abstract. In this paper, an overview of our recent experimental studies of the breakdown and operation of non-equilibrium discharges in centimetre and micrometer size geometries is presented. In the centimetre size geometries, we focused on elementary processes and phenomenology in gases used for some of the currently most attractive applications of low temperature plasmas - fluorocarbon gases (CF₄, CHClF₂) and water vapour. Measurements were performed at electrode separation $d = 1.1$ cm, in a pressure range from 0.1 to 5 Torr. In the case of micro-discharges, the emphasis was on testing the validity of standard scaling laws and proper determination of scaling parameters $pd$ and $j/p^2$. This was done at electrode gaps of 200 and 500 $\mu$m and pressures of 50 and 20 Torr in argon. The investigation is based on measurements of breakdown potentials (Paschen curves) and Volt-Ampere characteristics, supported by simultaneous ICCD imaging of the discharges.

1. Introduction
Owing to many unique properties, low temperature non-equilibrium discharges have been used in a large number of applications. Fundamental investigations lead to new applications but, recently, in many cases, the needs of applications led to numerous fundamental results. On all accounts, the complexity of these discharges call for full understanding of discharge processes in order to make efficient and reliable devices. Moreover, along with numerous non-equilibrium discharge sources in centimetre size [1,2], many sources with micrometre characteristic dimensions have been introduced recently [3,4]. Hence, an ongoing direction of research about discharges in molecular gases and gas mixtures of interest for applications [5] has been appended with new challenges of discharges taking place between electrodes separated by few tens or hundreds of micrometers – micro-discharges [6]. Additionally, a new type of non-equilibrium discharges taking place in or near liquids has emerged [7], bringing new possibilities for applications together with a whole new set of issues to resolve.

Accordingly, results presented in this paper are divided in two sections. The first one deals with measurements conducted with cm-size chamber. Using previously gathered experience on standard size non-equilibrium discharges in rare gases at low pressures [8-12], our research is continued with experimental measurements in gases of interest for applications: fluorocarbons (CF₄ and CHClF₂) which are widely used for plasma etching (e.g. [13]) and water vapour that is present as working environment of numerous commercial devices [14]. Measurements in these gases aim to provide reliable and systematic data sets required for plasma models that may be extended to applications involving those gases.

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In the second part, results obtained in case of micro-discharges, were used for comparison with the existing data on breakdown and discharge regimes of cm-size Ar discharges [15]. Our knowledge on basic processes in standard size discharges provided us with a possibility to investigate basic properties of micro-discharges from the point of view of scaling laws [16,17]. This type of non-equilibrium discharges with rising number of applications is inaccessible for most standard diagnostic tools due to its small size. Scaling laws, in general, should provide comparison of discharges with different dimensions or electrical characteristics [18]. Therefore, an approach through the analysis of scaling laws and the use of existing knowledge of processes in standard size discharges becomes valuable way of diagnostics of micro-discharges.

2. Experimental setup
Discharge was ignited in two different chambers, with centimetre and micrometer size electrode gaps. In both cases, two parallel-plate electrodes are placed inside a tightly fitting tube made of dielectric. For micrometer-size discharges, stainless steel cathode and a transparent anode with conductive ITO (Indium Tin Oxide) film were separated by \( d = 200 \mu m \) and \( d = 500 \mu m \) distances. Radial profiles are recorded from an end-on view, through the transparent anode. In the cm-size discharge chamber, copper cathode and transparent anode, with a conductive film of platinum deposited on a quartz window, were tightly surrounded by a quartz tube. In this way, along with radial discharge profiles, axial emission profiles could be recorded. Diameter of electrodes was \( D = 5.4 \) cm, while the gap for all measurements presented in this paper was \( d = 1.1 \) cm. Emission from the discharge, integrated in visual spectral range and in time, were recorded by ICCD camera (Andor iStar DH720-18U-03).

Details of electrical circuit and technique of measurement are presented in more details elsewhere [e.g. 11,15]. Apart from establishing low-current discharge, the electrical circuitry of experiment is able to impose a current pulse of desired length in addition to the running dc discharge. This technique allows us to avoid long breakdown delay times as well as gas and cathode heating and electrode conditioning in high-current discharge regimes [10]. Camera gating is synchronized with the development of voltage and current signals from the discharge in order to record emission from the quasi steady-state part of the pulse.

Before measurements, both vessels were pumped down to low pressure (10^{-6} Torr). In order to create the same cathode surface conditions each time [10], in cm-size chamber, the cathode was treated by moderately high-current discharge in hydrogen (30 \( \mu A \)) until a stable breakdown voltage is achieved. The same treatment but using Ar was done in the case of micrometer chamber since hydrogen discharge may damage ITO film on the anode [19].

3. Properties of standard size discharges in molecular gases important for applications
One of the fundamental properties of the dc discharge is the dependence of breakdown voltage (\( V_b \)) on the product of gas pressure and electrode gap (\( pd \)), i.e. Paschen curve. In Fig. 1 we show Paschen curves for some of the gases that we studied: water vapour, CF4 and CHClF2. For comparison, Paschen curve for Ar is also shown. All curves exhibit typical, sharply rising voltages with \( pd \) values decreasing to the left from the Paschen minimum and slow increment of breakdown voltages in the right-hand side branch. Curve for Ar has minimum around \( pd = 1 \) Torr cm with breakdown voltage of around 210 V, well below the ones for molecular gases shown here. The minimal breakdown voltages for CHClF2 and CF4 are 390 V and 445 V respectively. In water vapour, minimal value required for breakdown is around 530 V. Moreover, positions of minimum of the curves for all molecular gases investigated are shifted towards lower \( pd \) values in comparison with Ar. This feature has been noticed in case of other electronegative discharges (SF6, CO2, NO, air) [1].

At every point of the Paschen curve a corresponding axial emission profile of the low-current discharge was recorded (\( i = 1 \mu A \)). Spatial emission profiles recorded for different \( pd \), i.e. reduced electric field (\( E/N \)) values, are shown in Fig. 2a for CF4 and CHClF2 and Fig.2b for water vapour. All profiles are normalized so that peak intensities near the anode are the same. In this way, profile slopes can be compared easily.
Generally, profiles of emission in Townsend regime of discharge exhibit an exponential increase from the cathode towards the anode, when processes of excitation and ionization are predominantly induced by electrons. This kind of behaviour is exhibited in all gasses covered by this study at low $E/N$. Slope of the exponential increase is directly connected to the ionization coefficient [12].

**Figure 1.** Paschen curves for water vapour – squares, CF$_4$ – empty circles, CHClF$_2$ – full circles and Ar – triangles. The cathode is made of copper. Dashed lines show characteristic values of reduced electric field ($E/N$) parameter.

**Figure 2a.** Low-current discharge profiles in CF$_4$ (full lines) and CHClF$_2$ (dashed lines) for different values of $pd$ parameter. Corresponding $E/N$ values: CF$_4$ 0.16 Torr cm / 10 kTd, 0.35 Torr cm / 4 kTd, 0.9 Torr cm / 1.8 kTd; CHClF$_2$ 0.11 Torr cm / 13 kTd, 0.22 Torr cm / 5.5 kTd, 0.54 Torr cm / 2.6 kTd.

**Figure 2b.** Low-current discharge profiles in water vapour for different values of $pd$ parameter. Corresponding $E/N$ values: 0.19 Torr cm / 23 kTd; 0.36 Torr cm / 4 kTd; 0.8 Torr cm / 2 kTd; 2.4 Torr cm / 850 Td.
However, for sufficiently high $E/N$, heavy particles, such as positive ions and fast atoms, can also contribute. These processes are revealed through the additional peak of emission near the cathode [11]. As it can be observed in Fig. 2a, heavy particles contribute to the breakdown in CF$_4$ only at the highest $E/N$-s (~10 kTd), with fast F atoms and positive F$^+$ ions produced in CF$_4$ dissociation as probable candidates [20]. On the other hand, CHClF$_2$ and H$_2$O show notable contribution of heavy particle excitation starting from $E/N$ ~2 kTd. For highest reduced electric fields, heavy particle excitation becomes even dominant over electron induced excitation – in the case of CHClF$_2$ this is achieved at lower $E/N$ values (around 13 kTd) in comparison with H$_2$O (around 20 kTd) [21]. The apparent candidates for heavy particles that induce significant excitation and ionization in these two gases are hydrogen ions and fast atoms. Further spectrally resolved measurements are taking place to determine exact processes that participate in breakdown of these gases [22].

Figure 3. Volt-Ampere characteristics in freons, water vapor and Ar in the chamber with copper cathode. Electrode gap is $d = 1.1$ cm.

In Fig. 3 we show Volt-Ampere characteristics of dc discharges in fluorocarbons (CHClF$_2$ and CF$_4$), water vapour and Ar, for comparison. All Volt-Ampere characteristics were recorded for $pd$ values at the minimum of Paschen curves of particular gases, where the production and losses of the charged particles are well balanced. In all cases the electrode distance was $d = 1.1$ cm. The voltage is shown as the difference between the discharge and the breakdown voltage for the sake of comparison. Characteristics have a standard shape for low pressure dc discharges, where different regimes of the discharge can be clearly distinguished [23]. In the low-current range, small negative slope of the characteristics can be observed. The slope is larger in Ar discharge in comparison to discharges in molecular gases. At higher currents, fluorocarbon and water vapour discharges enter the current range where oscillations occur (gap in the descending part of Volt-Ampere characteristics). On the other hand, at these conditions, oscillations in Ar discharge exist only in a very small current interval. After the regime of oscillations, decrease in discharge voltage continues as it changes regime of operation to normal glow. Comparison in Fig. 3 shows different efficiencies of discharges in different gases. The highest difference between working and breakdown voltage is achieved in water vapour discharge (around 90 V), followed by discharge in fluorocarbons while in Ar difference does not exceed 20 V. However, one should bear in mind that these are relative differences. Positive slope of the characteristics, when a discharge voltage increases with current, is the property of discharge running in a high-current regime, abnormal glow.
4. Properties of micrometer size discharges – scaling laws
Almost all applications of micro-discharges are based on the capability to produce non-equilibrium plasma at atmospheric pressure. So far, non-equilibrium plasma has been comprehensively studied at low pressures and in standard size devices [11]. Since mutual correspondence between cm-size and micro-discharges is indubitably present, it is expected that the employment of scaling laws and data comparison should provide an insight to properties of micro-discharges. To test the applicability of scaling laws and provide description of basic processes governing breakdown and different discharge regimes of micro-discharge, we performed measurements on micro-discharges ignited between parallel electrodes. Although this configuration is more prone to instabilities, compared to hollow cathode-like discharges [3], establishing micro-discharge in plane parallel electrode geometry makes an advantage when it comes to the analysis and interpretation of the results. All measurements presented here were done in pure Ar.

First, we tested the validity of $pd$ scaling. Generally, the dependence of breakdown voltage on $pd$ parameter, i.e. Paschen curve, should be the same for all electrode gaps unless processes that void scaling take place. In micro-discharges, apart from well known processes that cause violation of scaling (gas and electrode heating, stepwise processes and three-body collisions at high pressures), one may expect processes such as field emission and quantum tunnelling of electrons [24].

![Paschen curves of micro-discharge (triangles) and standard size discharge (line) with stainless steel cathode in pure Ar.](image)

In Fig.4 Paschen curves of micro-discharges with $d = 200 \mu m$ and $500 \mu m$ electrode gaps are shown (triangles). For comparison, Paschen curve recorded in a standard size discharge (electrode gap $d = 1.8$ cm, electrode diameter $D = 4.4$ cm) is also shown (line). Breakdown voltages in micro-discharges at different gaps have small differences (up to 20 V) but the curve minimum and shape of the curves agree well in whole $pd$ range investigated. In the left hand-side branch of the 200 $\mu m$ curve the discrepancies are probably due to small differences in cathode surface conditions for the two discharge set-ups. In addition, this branch of Paschen curve is generally more sensitive to variation in gas pressure, due to the steep slope of the curve. The minimum of micro-discharge curves agrees well with the minimum of cm-size curve although the curve taken in cm-size discharge has less steep increase of breakdown voltage in the right-hand branch. There is a very good agreement between Paschen curves of micro-discharges and cm-size discharge. Hence, for the electrode gaps examined no new processes appear in the micro-discharge, i.e. the phenomenology is the same as in standard size discharges. Departures from the standard shape of Paschen curves for this range of electrode gaps, which exists in the literature, in most cases are influenced by discharge ignition at distances larger than electrode gap i.e. by the long path breakdown [15].
The second scaling that has been tested is \( j/p^2 \), which is connected to the effect of space charge [17]. It was shown that for standard size discharges both \( pd \) (and \( E/N \)) and \( j/p^2 \) scalings are valid in parameter region around Paschen minimum [11]. However, it is important to note that one needs to know realistic value of the current density i.e. to take into account effective area covered by the discharge, which may be considerably smaller than the area defined by the electrode diameter [25]. Micro-discharges operate at pressures higher than pressures used in standard size discharges, so that the occurrence of constriction in micro-discharge is even more pronounced [24]. Therefore, it is essential to record radial profiles of emission.

Scaled Volt-Ampere characteristics of micro-discharges for two electrode gaps and \( pd = 1 \) Torr cm are shown in Fig. 5. As before, the voltage is given as the difference between the discharge and breakdown voltages. For comparison, the same dependence obtained from the standard size discharge with copper cathode is also shown. Radial profiles of appropriate regimes (indicated by numbers in Fig. 5) for both electrode gaps of micro-discharges are shown in Fig. 6.

Negative slope of the Volt-Ampere characteristics in low-current regime is significantly smaller in the case of micro-discharge, as the negative differential resistivity scales with \( d^2 \). Profile of micro-discharge at \( d = 500 \) μm is diffuse in this regime (Fig. 6, no. 1). Still, at higher pressure at the distance of \( d = 200 \) μm, radial profile is very constricted and discharge occupies only a part of the electrode area. Sometimes, apart from larger area with emission, smaller emission area detached from the main discharge could be observed. The explanation of the second discharge channel's existence due to a high pressure and consequently small diffusion length seems valid [26].

![Figure 5. IV characteristics of micro-discharges (squares and circles) and standard size discharge (crosses) in Ar. Numbers correspond to images in Fig. 6.](image-url)

At higher currents, discharge profiles at higher pressure (shorter gap) stays constricted while at lower pressure (larger gap) micro-discharge takes up almost the entire electrode area (Fig. 6, no. 2). In this current range, the value of \( j/p^2 \) parameter is smaller for 200 μm discharge although the current is twice as higher than for 500 μm case because of smaller effective discharge area and higher pressure. Discharges operate in normal glow. After oscillations (interval without points in Fig. 5), both characteristics have positive slopes. In both cases, at high-current regime, discharges occupy the whole electrode area (Fig. 6, no. 3). This current range corresponds to the abnormal glow. Existing discrepancy between micrometer and centimetre size characteristics may be the consequence of different cathode materials but also more pronounced edge effects in micro-discharges.
\[ pd = 1 \text{ Torr cm} \] (1) Townsend discharge \[ i = 2.9 \mu A, j/p^2 = 0.35 \] (2) Normal glow \[ i = 469 \mu A, j/p^2 = 39 \] (3) Abnormal glow \[ i = 1172 \mu A, j/p^2 = 93 \] \[ d = 500 \mu m \]

\[ i = 1.78 \mu A, j/p^2 = 0.26 \]
\[ i = 938 \mu A, j/p^2 = 24 \]
\[ i = 4010 \mu A, j/p^2 = 51 \] \[ d = 200 \mu m \]

Figure 6. Radial profiles of micro-discharges for two different electrode gaps at different points of IV characteristics (shown in Fig.5).

5. Conclusion
Measurements of breakdown voltages and electrical and emission properties of different discharge regimes were performed in simple plane-parallel standard size discharge with gases used in applications. Simultaneous recordings of Paschen curves and emission profiles in low-current limit allowed the analysis of dominant processes governing breakdown in CF\(_4\), CHClF\(_2\) and H\(_2\)O gases. It was shown that at higher reduced electrical field values heavy particles have important and even dominant role. Comparison of Volt-Ampere characteristics recorded in aforementioned molecular gases and in Ar revealed differences in electrical properties of operating regimes and efficiencies of discharges in different gases. Further on, we presented results obtained in parallel plate geometry discharge with electrode gap in micrometer range - micro-discharge, that are used in many applications as non-equilibrium discharge sources at high or atmospheric pressures. Since diagnostic of these discharges is difficult due to their small dimensions, we investigated possibility of using \( pd \) and \( j/p^2 \) scaling laws for comparison with cm-size low-pressure discharges. Comparison of the results show the validity of both scaling laws for micro-discharge electrode gap domain investigated. However, it was shown that a special care should be devoted to the proper determination of scaling parameters.

Present results could be extended with temporal development of the profiles which were also recorded [27]. Besides, in case of micro-discharges metastable densities were recorded [28]. Measurements in oscillatory and steady-state regimes of Volt-Ampere characteristics indicate compatibility of in micro-discharges results [29] with the secondary electron yield model [8]. This provides a support to the expansion of the model to RF plasmas [30]. In addition, it would be interesting to expand the measurements to RF breakdown and thus test the existing theories [31]. Finally, one could also pay closer attention to the Townsend regime where absolute calibration may provide data to model heavy particle effects at high \( E/N \) [32] or provide data on ionization coefficients [12] and kinetics of non-local discharges [33] and runaway electrons [34], even data required to study streamers and ionization fronts [35].

6. References
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