New phenomenology of gas breakdown in DC and RF fields

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Abstract. This paper follows a review lecture on the new developments in the field of gas breakdown and low-current discharges, usually covered by a form of Townsend’s theory and phenomenology. It gives an overview of a new approach to identifying which feedback agents provide breakdown, how to model gas discharge conditions and reconcile the results with binary experiments and how to employ that knowledge in modelling gas discharges. The next step is an illustration on how to record volt-ampere characteristics and use them on one hand to obtain the breakdown voltage and, on the other, to identify the regime of operation and model the secondary electron yields. The second aspect of this section concerns understanding the different regimes, their anatomy, how those are generated and how free running oscillations occur. While temporal development is the most useful and interesting part of the new developments, the difficulty of presenting the data in a written form precludes an easy publication and discussion. Thus, we shall only mention some of the results that stem from these measurements. Most micro discharges operate in DC albeit with complex geometries. Thus, parallel plate micro discharge measurements were needed to establish that Townsend’s theory, with all its recent extensions, is still valid until some very small gaps. We have shown, for example, how a long-path breakdown puts in jeopardy many experimental observations and why a flat left-hand side of the Paschen curve often does not represent good physics. We will also summarize a kinetic representation of the RF breakdown revealing a somewhat more complex picture than the standard model. Finally, we will address briefly the breakdown in radially inhomogeneous conditions and how that affects the measured properties of the discharge. This review has the goal of summarizing (rather than developing details of) the current status of the low-current DC discharges formation and operation as a discipline which, in spite of its very long history, is developing rapidly.

1. Introduction: Townsend’s theory and low-current DC discharges - 100 years ago and now
With the development of basic phenomenology and theory of gas breakdown, Townsend’s theory was forged some 100 years ago [1-3]. In this paper we shall give a review of how in the past 20 years the basic Townsend’s theory and phenomenology have been revived, extended, revitalized and put in perspective of modelling higher current technological discharges and plasmas. This was primarily done by the groups of Art Phelps and the Gaseous Electronics Laboratory in Belgrade.

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Recent advances in diagnostics and modelling of complex plasma systems opened an opportunity to revisit the breakdown in gases both in DC and RF fields and also for micro gaps. We shall first discuss the experimental techniques to determine the breakdown voltage. Typical errors, such as neglecting the long-path breakdown on the left-hand side of the Paschen minimum, conditioning of the electrode and measuring properties within an unstable (oscillating) regime, will be covered briefly. In addition, we shall describe a proper methodology to establish volt-ampere \((V-I)\) characteristics and how to use those with the goal of determining the breakdown voltage and the secondary electron yields.

Time-resolved imaging [4] provides us with information on the development of the anatomy of the discharge and its different modes. Using the spatial profile one may decide which of the mechanisms dominate the discharge. The Townsend regime is the low-current diffuse discharge with exponential growth towards the anode [5]. It is necessary to observe such a profile to ascertain that the discharge operates in the Townsend regime where Townsend’s theory may be used to establish the condition for breakdown and effective secondary electron yield. The temporal development of the normal glow or abnormal glow following the breakdown reveals transient multi-regime operation that requires a new paradigm.

When considering volt-ampere characteristics, one may first observe the negative differential resistance in the Townsend regime which may be explained by a combination of space-charge effects and an energy-dependent secondary electrons yield. Thus, \(V-I\) characteristics should be used in addition to the Paschen curve to determine the secondary electrons yields. With the newly found field of discharges in and above liquids, we have also analyzed breakdown in water vapor and ethanol \([6,7]\).

RF and microwave breakdowns have a different phenomenology as the secondary ions production of electrons at the cathode may not be necessary. Yet the RF breakdown is prone to phenomena not often observed in DC breakdowns, like S shaped (double valued) Paschen-like curves, frequency/gas number density scaling and additional mechanisms like multipactors. The principal experimental problem with the RF breakdown is the magnitude of the displacement current that thwarts the measurement of the conduction current making it difficult to ascertain the initiation of the discharge.

### 2. Gas breakdown, feedback mechanism, secondary electron yields and how to use them

Traditionally, the gas breakdown is characterized by Paschen curves. In figure 1 we show one such example of Paschen curves for the topical water vapor and ethanol vapor \([6,7]\).

While most people will argue (including the present authors) that the Paschen law itself is developed with limiting assumptions

\[
V_b = \frac{Bpd}{\ln(Apd) - \ln\left(1 + \frac{1}{\gamma}\right)},
\]

the breakdown equation from the Townsend’s law is in principle exact if the assumptions of the primary feedback through ions are correct:

\[
y(E/N)(e^{a(E/N)d} - 1) = 1.
\]

Different forms of analytical laws have been employed for the DC breakdown in order to provide some insight \([8,9]\), but one needs to be aware of the approximate nature of some of the basic formulae. Still, the same limitations that enter the analytical form of Paschen law may enter the implementation.

![Figure 1. Paschen curves for water [6] and ethanol vapor [7].](image-url)
of equation (2). Over the years, a large amount of data on $\gamma$ have been accumulated, yet seldom those have been compared and systematically collected. Even more importantly, comparisons to binary collision (beam-surface) experiments [10] have been almost avoided in the literature presumably due to poor agreement (both qualitative and quantitative). One example where both systematic collection of data and comparisons to binary experiments have been made is the paper of Phelps and Petrović [3].

In figure 2 we show a comparison of the secondary electron yields obtained from Paschen curves in argon for a range of experiments [3]. These should be compared with a constant yield of 0.08 obtained by beam experiments with an atomically clean surface [10] or to the dot-dash line obtained for the same conditions of the cathode as found in discharge experiments. Discharge results are one order of magnitude too high at higher $E/N$, one order of magnitude lower at moderate $E/N$ and two orders of magnitude higher at the lowest $E/N$. Phelps and Petrović managed to reconcile the binary collision data and the discharge results by including the following:

- ionization coefficient fitted in a wide range of $E/N$;
- a region close to the cathode where electrons gain energy and become in equilibrium with the local field, the simplest way of representing this being using a delay distance $d_0$;
- ion-induced yields at the cathode, modified to represent surfaces that are not atomically clean;
- back-diffusion, i.e. the return of newly emitted electrons back to the cathode;
- secondary electron production by metastables;
- secondary electron production by fast neutrals;
- secondary electron production by resonant radiation – the photo effect;
- trapping of resonant radiation;
- secondary electron production by fast neutrals;
- secondary production due to molecular emission.

The solid and dashed lines in figure 2 indicate the model predictions based on binary collision data (for two limits of possible contributions by molecular radiation). These two lines encompass most of the available experimental data. The good agreement with the experiments shows that all pertinent processes have been included. It also shows that the process of secondary yields modelling may be quite complex and quite challenging due to the need for a wide range of data.

The fact that Townsend’s theory could associate all the yields with the flux of ions is because all fluxes are proportional to the electron flux and the system is linear in the breakdown-Townsend discharge phase. On the other hand, one cannot expect linearity to hold for higher current modes, such as glow discharges. For those, one is left with fitting the experimental data. Thus, we compared the fitting procedure with one based on the breakdown data from Phelps and Petrović [3]. We found [11] that fitting of the glow discharge is well represented by the procedure recommended for the breakdown data [3], except when $pd$ is quite low and fast neutral effects become dominant. Yet, for RF discharges, for example, or for some more complex geometries, one needs to provide a clear guidance as to how the secondary electron yield may be modelled. Also, we need a considerable effort to provide the data for a number of relevant gases, as argon is the only gas covered so far by the detailed analysis.
3. Volt-ampere characteristics and spatial emission profiles

There are two ways to determine the breakdown voltage. The first option is very accurate albeit very difficult – one can extrapolate pre-breakdown currents. The second one, favored by our group, is to establish a self-sustained discharge in the low-current diffuse, i.e. Townsend, discharge and then extrapolate the measurements to zero current. All other techniques suffer from arbitrariness, either induced by the long statistical time lags or by a direct transition to the glow regime. The Townsend’s regime is recognized by an exponential growth peaking at the anode and a normally broad diffusion-determined profile over the entire surface [5,12], as can be seen in figure 3.

In conducting such measurements, we (re)established that in the Townsend regime one has a negative differential resistance. As the effective resistance of the discharge is negative, sometimes, coupled with the external circuit, the overall loop resistance may become negative and oscillations may occur [13,14]. As it turns out, the space charge due to ions increases the field in front of the cathode, which increases the electron production allowing a lower field elsewhere and thus the overall voltage is reduced [14,15]. Thus, the dependence of $\gamma$ on the mean energy is the reason for the negative differential resistivity, brought about by space charge induced electric field and represented as a current dependence of $\gamma$. As the discharge approaches constriction, non-linearities become important [16] and a sudden transition eventually takes place. It has also been shown that if $\gamma$ were constant, the slope of the $V$-$I$ characteristics in Townsend’s regime could become positive [16]. Thus one may conclude that for the full representation of the secondary electron yield one needs to fit not only the Paschen curve but also the $V$-$I$ characteristics. The realm of oscillations often precludes us from achieving stable operation in Townsend’s regime but it is also a source of information on important processes. Thus, fitting of the induced damped or free running oscillations may reveal identity of the dominant ionic species, multiplication and may be related to basic transport properties of relevant particles.

![Figure 3. Spatial profile of the Townsend’s regime low-current diffuse discharge [12].](image)

![Figure 4. Axial profiles of higher current discharge regimes in argon, at (a) $pd = 1.1$ Torr cm (close to the Paschen minimum); (b) $pd = 0.3$ Torr cm (in the left-hand branch of the Paschen curve).](image)
Another important aspect that stems from figure 3 is that spatial profiles not only give us information on the regime of operation (see figure 4 – the peak in front of the anode is for Townsend’s low current discharges, the peak in the bulk corresponds to the glow discharge) but also (if put on an absolute scale) a basis to establish absolute cross sections and even profile of the field. It also shows whether and to what extent is equilibrium (with the field) developed or whether fast neutral excitation is important as recognized by the peak right in front of the cathode.

4. Scaling of the basic properties of micro discharges

Micro discharges were basically developed to take advantage of the non-equilibrium plasma that is formed around the Paschen minimum, but at a much higher pressure. Atmospheric pressure would require a 10 µm gap. On the other hand, to achieve a stable operation at high pressures one needs to use complex geometries as it proved very difficult to operate parallel plate micro discharges. Yet, many authors have assumed parallel plate geometries with narrow strips crossing at small distances and assumed that the breakdown occurs at the shortest distances. This has led to a number of papers where the left hand side of the Paschen curve showed no or little variation that could be erroneously interpreted as the onset of field emission (that was predicted to occur only for \( d < 10 \) µm [9]). We have made an effort to perform measurements in well defined and contained parallel plate discharges, to test the applicability of Townsend’s phenomenology at small gaps [17].

Before proceeding to any modeling, we needed to test the laws of scaling, which for low pressure collision dominated discharges are \( E/N, pd \) and \( jd^2 \) (and also \( \omega/N \) and \( B/N \) for time varying fields and for magnetic fields). The critical scaling is due to the current density \( j \). Rarely are the \( V-I \) characteristics represented through \( j \) (as it should be) and even then it is not stated that \( j \) is actually determined by dividing the current by the entire area of the electrodes. In reality, however, constriction dominates in the glow regime. Even in the Townsend’s and in the abnormal glow regimes the radial profile is quite different and so is the effective area. Taking advantage of transparent yet conducting materials for electrodes and also of ICCD cameras, we were able to record \( V-I \) characteristics and scale them to the real current density even in cases of complex constricted modes. First, we established that in the normal size discharges, when the current density is properly determined, it remains constant throughout the glow regime (so it is represented by a single point in \( V-I \) characteristics [18].

Secondly, we have been able to show that for discharges bordering on micro discharges (0.5 and 1 mm gaps) the \( jd^2 \) scaling works and finally we have extrapolated those findings (by using \( pD \) scaling where \( D \) is the diameter of the constricted region) to smaller gaps where we could not easily record the current profiles [17]. It was found, as can be seen in figure 5, that a Townsend type of scaling holds at those gaps until perhaps some smaller geometries where field emission really comes into play [19]. We have even shown that the spatial profiles scale well so that the ionization coefficient could be determined rather accurately from the axial emission profile of micro discharges [20].

It was also shown that the Paschen curves obtained for micro discharges (and sometimes even for the standard size discharges) that do not show a change of voltage on the left hand side beyond the minimum are due to an incorrectly assumed shortest distance for the gap when long path breakdown was allowed [21]. Reducing the chances for the long path breakdown brings back the Paschen curve to agreement with that obtained for standard dimensions/pressures.

The fact that we proved scaling and also were able to obtain accurate readings of ionization coefficients proves that it is possible to apply Townsend’s phenomenology and even to some degree theory to micro discharges. The \( jd^2 \) scaling allows Townsend regime to operate at considerably higher current densities.

5. Time resolved measurements

A preliminary publication of our results on time dependent recordings of the development of the breakdown and DC discharge regimes was given in [9,22], while the majority of the data remain unpublished. The results may be summarized as follows. During the initial stage, the discharge passes through the Townsend regime and, as the current increases, glow and abnormal glow regimes are visited. During the minimum of oscillations, the discharge almost immediately returns to the
Townsend regime and then changes occur again and again following oscillations in current. Studies such as this one performed for a parallel plate geometry are needed to provide the background for understanding developments in more complex geometries [23] and different scales.

Figure 5. $V-I$ characteristics for standard size and micro discharges obtained by using the $jd^2$ scaling and $pD$ scaling for the size of the constricted regions. One should not pay attention to the actual vertical scale, normally it is taken care of by subtracting the breakdown voltages in each case, but in the case of micro discharges we could not make accurate measurements as it was not possible to achieve stable operation in the Townsend regime. The vertical scale variations are due to different conditions on the electrodes surface [17].

6. Discharges with inhomogeneous cathodes
The usual assumption in the low current limit is that the discharge is uniform over the entire surface and the radial profile is the solution to the diffusion equation. Only in the higher current (glow) regime a constriction develops and only when the field ceases to be constant along the axis. This has been questioned recently for micro discharges when it was found that the scaling laws are maintained only when an assumption of localized modes is made, the modes that have a dimension proportional to the diffusion length for the given pressure [17]. The problem is hindered further by the high pressure that reduces the diffusion length and by the fact that the ratio of gap to radius becomes very small in practical experiments [17].

We have, however, observed localized discharges in Townsend regime for standard size discharges (1 cm) and moderately low pressures [24]. In figure 6 we show one such example in nitrogen where the discharge is limited in the radial direction by the region of deposited material due to numerous pulses in the high current mode. It is clear that the deposited region has a lower secondary electron yield $\gamma$ and thus the discharge cannot be self-sustained over that area for the given voltage. In order to overcome this region of reduced $\gamma$ the volt-ampere characteristics have to be quite different as compared to the one with pristine cathode. Even a positive differential resistance is observed [24].

The characteristics of such a discharge may be a prototype for the breakdown in pulsed DC discharges where numerous discharges change the properties of the cathode considerably. In addition, one may construct cathodes of different materials and design desired characteristics. For example, cathodes coated by a semiconductor [25, 26] have been often used for achieving some properties that are not easily accessible by conducting electrodes. The high resistance of the semiconductor which is in the innermost circuit assures a broader range of stable operation [14]. Combining conducting and semiconductor materials may help reduce the breakdown potential while achieving a $V-I$ characteristic that is less prone to oscillations.
7. RF breakdown

When rate of change of the field is such that ions cannot complete their trajectories, then it is possible to operate in conditions when electrons may be the only particles sustaining the discharge. The feedback is provided by returning electrons in the second half of the period and thus a full circle is achieved. It has been assumed in old textbooks and papers [27] and in more recent papers [28, 29] that an optimum breakdown condition is achieved when the average electron can cross the gap in one half period. In fact, the breakdown condition has been used to obtain experimental values of the drift velocities for RF fields and convert them to DC values by assuming that in RF fields the drift velocity is a sinus function peaking with the DC drift velocity and having no delay [29]. We modelled the RF breakdown using a detailed Monte Carlo

Figure 6. Townsend regime discharge in N₂ with Cu cathode and for \( d = 0.8 \text{ cm}, \ D = 2 \text{ cm}, \ p = 2 \text{ Torr} \) and \( V_b = 310 \text{ V} \). The axial profile shows exponential growth peaking at the anode while the radial profile is very narrow covering only a small part of the diameter (\( D \)).

Figure 7. Development of density and ionization rate for the breakdown of RF swarms. Solid curves represent mean velocity (different colours are for better contrast), while the dotted sinusoidal lines represent inversed AC field. Both curves are for 0.2 Torr; the top curves are for the minimum breakdown voltage (93 V) while the bottom curves are for the maximum breakdown voltage (447 V) [30].
representation of all the collisions and field dependence on time. Figure 7 shows the temporal development of the concentration of electrons and of the ionization rate [30]. As the quasi-Paschen curve for the RF breakdown has double values for breakdown voltage at the same \(pd\) (for a range of \(pd\)) we show results at the lower and at the higher breakdown point. Outside these borders the discharge cannot be ignited. At the lower point, the breakdown condition is achieved by merely matching the production without much of the electrodes overlapping with the swarm. At the higher end, however, the swarm overlaps considerably with the electrodes thus representing significant losses to the electrodes. At the same time, the peaking ionization must compensate for all the losses.

We found that, while the standard explanation of the RF breakdown is very good, still some fine tuning needs to be done by a full kinetic representation (such as a Monte Carlo simulation) to be able to describe all the intricacies. In addition, one needs to extend our model by the contribution of ions and fast neutrals as well as photons. These will modify the Paschen like curve and hopefully make it more realistic but for a wide range of conditions one may find that electrons dominate in sustaining the plasma. We also have to include the secondary electrons formed by the electron impact on electrodes. This effect will lead to the so-called multipacting modes.

One should also note that numerous attempts to use analytical and semi analytical models based on simplified expressions for the basic properties have migrated from DC discharges to the RF breakdown. These results provide insight into pertinent processes [31-34] but require some form of fitting to provide quantitative agreements.

8. Conclusions
The oldest chapter in the book on plasma physics, the breakdown and low current discharges, has been changed tremendously in the past twenty years. The main agent facilitating the feedback needed to achieve DC breakdown, the ions colliding with surfaces, have been changed to include photons, fast neutrals, metastables as well as ions. By including all these processes, as well as back-diffusion [3,35] one was able to predict effective yields and Paschen curves based on the binary collision data [3].

The field of low pressure DC discharges proved to be a fertile ground for both fundamental studies and for obtaining and testing the applicability of the fundamental data that would eventually be used for modelling of more complex systems. Extensions to micro discharges and RF breakdown have been made, together with first attempts to model RF plasmas with secondary electron yields [36] as obtained in a more detailed and recent analysis.

With the new drive for benchmarking plasma modelling systems [37], we believe that the best strategy would be to start from the swarm benchmarks and then use the negative differential resistance of a DC Townsend discharge as a benchmark for space charge effects and also use some additional breakdown properties. This would provide clear and simple experimental observables that may be modelled exactly and independently and provide the next step for more complex plasma benchmarks.

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