

# About the importance of volume memory effects in pulsed dielectric barrier discharges in O<sub>2</sub>/N<sub>2</sub> gas mixtures at atmospheric pressure

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This paper presents experimental results on volume memory effects, which allow a control of the breakdown in pulsed dielectric barrier discharges (DBDs) in O<sub>2</sub>/N<sub>2</sub> gas mixtures at atmospheric pressure. The filamentary DBDs were examined by electrical and optical diagnostics in a 1 mm gap while varying the pulse width (time between two subsequent DBDs) and the O<sub>2</sub> concentration in N<sub>2</sub>. By the pulse width, the breakdown can be controlled down to the fundamental level, i.e. changing the breakdown regime from a cathode-directed streamer to a cathode- and anode-directed streamer or no streamer propagation at all. In a certain parameter range, an increase of the pulse width has the same effect as the decrease of the O<sub>2</sub>/N<sub>2</sub> ratio, e.g. an ignition time delay, a higher discharge current maximum in combination with a faster discharge current decay. The volume pre-ionisation was found to have a crucial impact in on the DBD characteristics and can be controlled by the pulse width (shifting the DBD in afterglow of the previous one) or by the O<sub>2</sub> content. A higher O<sub>2</sub> admixture leads to faster electron decay by attachment to oxygen and effectively collisional quenching of excited N<sub>2</sub> species by oxygen.

## 1. Introduction

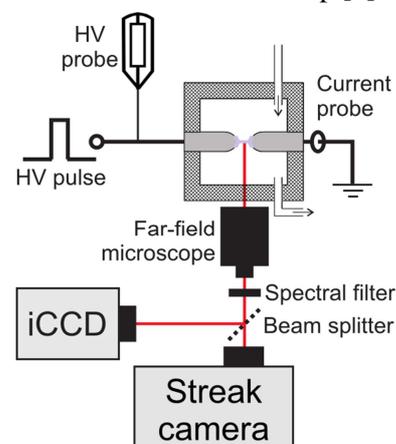
Non-thermal plasmas generated in dielectric barrier discharges (DBDs) at atmospheric pressure feature high electron energies while heavier particles (molecules, atoms, ions) remain at low energies, i.e. the gas temperature stays near 300 K [1]. This allows an efficient generation of active species, which are important for industrial applications such as ozone generation and exhaust treatment [2].

The “tailoring” of a DBD for a specific application is one of the main goals of the current research. This requires a distinct control of the DBD characteristics. The inception and development of the discharge is strongly influenced by the processes in the so-called Townsend pre-phase, i.e. the time before the actual breakdown occurs [3-5]. While in sinusoidal-operated DBDs the pre-phase lasts for at least several hundred nanoseconds, in pulsed-operated DBDs this duration is limited by the rise time of the HV pulse (in our case  $\approx 50$  ns for 10 kV pulse amplitude).

The pulsed-operation, however, offers an additional electrical control parameter (apart from voltage amplitude and repetition frequency): the pulse width [6,7]. In addition to the analysis of pulse width variation, the influence of the O<sub>2</sub> content in O<sub>2</sub>/N<sub>2</sub> gas mixtures was investigated. Furthermore, the interrelations between the DBD control by pulse width and O<sub>2</sub> admixture were evaluated.

## 2. Experimental Set-up

For the investigations a single filament DBD arrangement (double-sided, half-sphere Al<sub>2</sub>O<sub>3</sub> covered electrodes) with 1 mm gap was used. The DBDs were driven by unipolar positive HV pulses with 10 kV amplitude and 10 kHz repetition rate. The pulse width was varied from 50 to 0.2  $\mu$ s for 0.1 vol% O<sub>2</sub> in N<sub>2</sub>. The O<sub>2</sub> concentration in N<sub>2</sub> was changed from 0.1 to 20 vol% at a fixed pulse width. Fast electrical, iCCD and streak measurements are performed to record the electrical characteristics as well as the spatio-temporal DBD development; see figure 1 for an overview of the set-up [8].

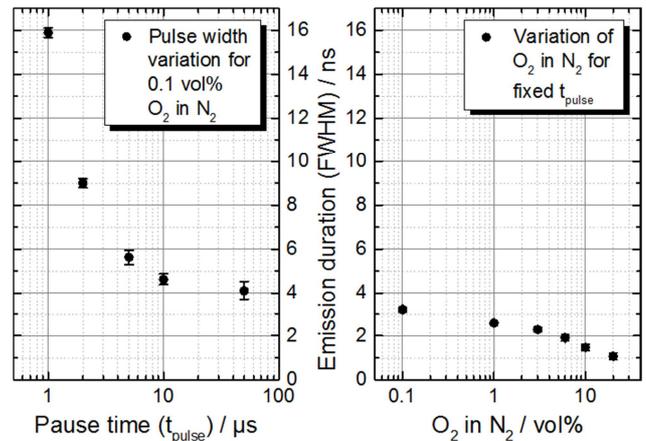


**Figure 1:** Schematic representation of the DBD cell and the applied diagnostics.

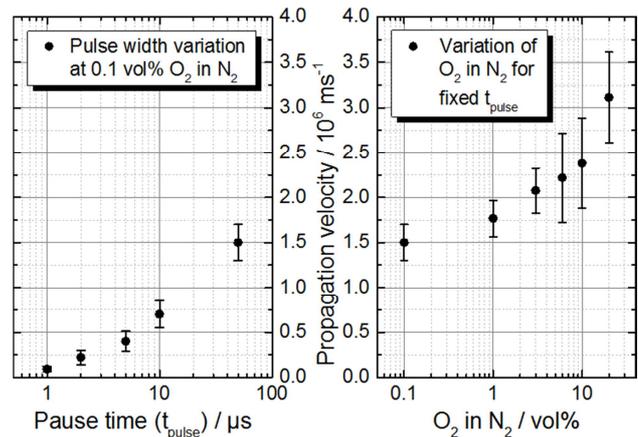
### 3. Results and Discussion

The DBD breakdown characteristics at the falling slope of the HV pulse could be controlled by the pulse width [8,9]. Besides the effects of voltage slope steepness (e.g. amplifying the streamer propagation due to the significant voltage rise during the breakdown), the pulsed operation offers via the pulse width an excellent parameter to set the pre-ionisation by shifting the DBDs in the after-glow of the previous discharge using asymmetrical HV pulse waveforms. The breakdown characteristics of these subsequent DBDs igniting in different pre-ionised conditions defined by the residual charge carrier densities originated from the previous DBD in the gap can be controlled down to the fundamental level. For instance, four different breakdown regimes in single filament DBDs for 0.1 vol% O<sub>2</sub> in N<sub>2</sub> were described and connected to the processes during their pre-phases [10]. The “classical” DBD development (cathode-directed streamer followed by a transient glow discharge) could be controlled in a certain range, followed by a transition to a breakdown regime featuring a simultaneous propagation of a cathode- and an anode-directed streamer, and finally to a reignition of the previous DBDs without any propagation just with reducing the pulse width (time between two subsequent DBDs), i.e. increasing the pre-ionisation level. This manipulation of the pre-ionisation level was found to be the crucial point to affect and consequently control the DBD behaviour.

Changing the O<sub>2</sub>/N<sub>2</sub> ratio has a significant impact on the DBD characteristics, too [11,12]. While the transferred charge in the investigated DBDs depends not on the pulse width, but on the O<sub>2</sub> concentration (higher [O<sub>2</sub>] → lower transferred charge); there are some physical quantities, which show a similar behaviour either for increasing pulse width or increasing O<sub>2</sub> concentration. This is shown in figure 3 for the DBD emission duration and in figure 4 for the maximal velocity of the cathode-directed streamer obtained in front of the cathode using streak camera images. To minimise the effect of the pre-ionisation caused by the previous DBD for the variation of the O<sub>2</sub>/N<sub>2</sub> ratio, the impact of the O<sub>2</sub> concentration was evaluated at the rising slope for 10 μs pulse width (i.e. 90 μs to the previous DBD, which has no significant effect on the pre-conditions). The absolute values do not agree, but the same tendency is clearly visible: an increase of the pulse width as well as the O<sub>2</sub> concentration leads to shorter emission duration and higher propagation velocities.



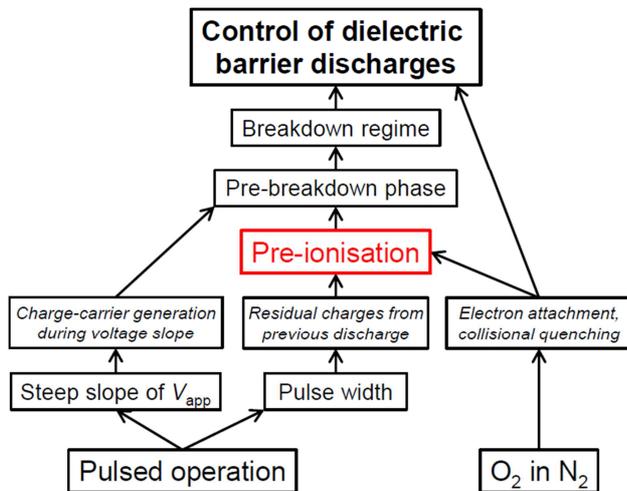
**Figure 3:** Effect of pulse width (falling slope) and O<sub>2</sub>/N<sub>2</sub> ratio variation (rising slope) on the full width at half maximum (FWHM) of the emission duration.



**Figure 4:** Effect of pulse width (falling slope) and O<sub>2</sub>/N<sub>2</sub> ratio variation (rising slope) on the maximal velocity of the cathode-directed streamer (in front of the cathode).

Although, the streamer velocities depend i.a. on the first Townsend coefficient, which changes with the O<sub>2</sub>/N<sub>2</sub> variation and the strong collisional quenching for higher O<sub>2</sub> concentrations, it can be stated, that to some extent, several of the reported effects achieved by varying the pulse width at a fixed O<sub>2</sub> concentration in N<sub>2</sub> (0.1 vol%) were also observed for a fixed pulse width and changing O<sub>2</sub> content. As a consequence of the high electronegativity of oxygen, negative ions are formed, which lead to a decrease in the pre-ionisation. Therefore, an increase of the O<sub>2</sub> concentration leads to similar effects as a prolongation of the pulse width at 0.1 vol% O<sub>2</sub> in N<sub>2</sub> in a specific parameter range of the pulse width and O<sub>2</sub>/N<sub>2</sub> ratio. Hence, the response of the DBD

properties to changing pre-ionisation levels seems to be a general principle of DBD control.



**Figure 5:** Control mechanisms of the DBD breakdown via pulse width and  $O_2/N_2$  ratio.

In figure 5, the possible control mechanisms and their interrelations are summarised underlining the influence of the volume pre-ionisation.

So far, the discussion has not addressed the role of surface charges, especially their temporal development, which is assumed to be on the microsecond time scale [13]. The inclusion of these processes is important to evaluate the correlation between volume and surface memory effects as proposed e.g. in [14].

## References

- [1] A. Fridman, A. Chirokov, A. Gutsol, *J. Phys. D: Appl. Phys.* **38** R1 (2005).
- [2] U. Kogelschatz, *Plasma Chem. Plasma Process.* **23** 1 (2003).
- [3] K.V. Kozlov, H.-E. Wagner, R. Brandenburg, P. Michel, *J. Phys. D: Appl. Phys.* **34** 3164 (2001).
- [4] Y.V. Yurgelenas, H.-E. Wagner, *J. Phys. D: Appl. Phys.* **39** 4031 (2006).
- [5] T. Hoder, R. Brandenburg, R. Basner, K. D. Weltmann, K. V. Kozlov, H.-E. Wagner, *J. Phys. D: Appl. Phys.* **43** 124009 (2010).
- [6] X. Lu, M. Laroussi, *J. Phys. D: Appl. Phys.* **39** 1117 (2006).
- [7] M. Kettlitz, H. Höft, T. Hoder, K.-D. Weltmann, R. Brandenburg, *Plasma Sources Sci. Technol.* **22** 025003 (2013).
- [8] M. Kettlitz, H. Höft, T. Hoder, S. Reuter, K.-D. Weltmann, R. Brandenburg, *J. Phys. D: Appl. Phys.* **45** 245201 (2012).
- [9] T. Hoder, H. Höft, M. Kettlitz, K.-D. Weltmann, R. Brandenburg, *Phys. Plasmas* **19** 070701 (2012).
- [10] H. Höft, M. Kettlitz, M.M. Becker, T. Hoder, D. Loffhagen, R. Brandenburg, K.-D. Weltmann, *J. Phys. D: Appl. Phys.* **47** 465206 (2014).
- [11] K.V. Kozlov, R. Brandenburg, H.-E. Wagner, A.M. Morozov, P. Michel, *J. Phys. D: Appl. Phys.* **38** 518 (2005).
- [12] H. Höft, M. Kettlitz, T. Hoder, K.-D. Weltmann, R. Brandenburg, *J. Phys. D: Appl. Phys.* **46** 095202 (2013).
- [13] M. Bogaczyk, S. Nemschokmichal, R. Wild, L. Stollenwerk, R. Brandenburg, J. Meichsner, H.-E. Wagner, *Contrib. Plasma Phys.* **52** 847 (2013).
- [14] Y. Akishev, G. Aponin, A. Balakirev, M. Grushin, V. Karalnik, A. Petryakov, N. Trushkin, *Plasma Sources Sci. Technol.* **20** 024005 (2011).