Published in final edited form as:

J Phys D Appl Phys. 2014 June 25; 47(25): 252003–. doi:10.1088/0022-3727/47/25/252003.

Uniform and non-uniform modes of nanosecond-pulsed dielectric barrier discharge in atmospheric air: fast imaging and spectroscopic measurements of electric field

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Abstract

In this study, we report experimental results on fast ICCD imaging of development of nanosecond-pulsed dielectric barrier discharge (DBD) in atmospheric air and spectroscopic measurements of electric field in the discharge. Uniformity of the discharge images obtained with nanosecond exposure times were analyzed using chi-square test. The results indicate that DBD uniformity strongly depends on applied (global) electric field in the discharge gap, and is a threshold phenomenon. We show that in the case of strong overvoltage on the discharge gap (provided by fast rise times), there is transition from filamentary to uniform DBD mode which correlates to the corresponding decrease of maximum local electric field in the discharge.

Keywords

dielectric barrier discharge; electric field measurement; atmospheric air DBD uniformity; uniformity analysis

1. Introduction

Dielectric barrier discharge (DBD), as an easy and simple way of generation of non-thermal plasma, has found a number of application in various fields [1]. However, development of a homogeneous, or uniform, DBD that would operate at atmospheric pressure conditions in atmospheric air, would open a number of new applications in various fields from thin film coatings to plasma medicine. Currently, there is no adequate model of the uniform dielectric barrier discharge development in atmospheric air. Although extensive studies have been performed on understanding of the nature of the pulsed DBD uniformity (see review in [2]), until now there is still little understanding of the mechanism of the DBD transition from the filamentary mode to uniform mode. One of the reasons for this is that development of streamers, and later – filaments, occurs on the sub-nanosecond and nanosecond time scales, and therefore requires imaging and other diagnostic techniques with corresponding speed of registration. For example, Rahel et al [3] investigated the visually uniform DBD discharge in air by registering time-resolved emission with a photomultiplier and recording discharge current, however no fast imaging was reported to directly verify the discharge uniformity: at

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The results presented in the manuscript are novel, and would be interesting for the readers of JPhysD. The results are significant for the fields of non-thermal plasma, especially atmospheric pressure materials processing, plasma medicine, etc.

a high density of non-stationary streamers the discharge may actually look diffuse-like [4–7]. It is important to note that here we are focusing on the development of a single-pulse DBD, which is qualitatively different from the physics of high repetition frequency nanosecond discharges (NRP, [2]): streamer-less “glow” discharge may be formed in the case of high preionization levels which can be achieved at sufficiently high repetition frequencies. In the case of traditional single-pulsed (or low repetition frequency) discharge, Townsend regime is changed by streamer regime when Meek criterion is satisfied (i.e. when critical electron density of $\sim 10^{13} \text{ cm}^{-3}$ is reached). Traditional recipes for avoiding the transition into streamer regime include [2, 8–15] a) conditions when Meek criterion is not reached, i.e., low plasma densities (by decreasing gas pressure or heating up the gas for example); b) preionization (for example, by increasing repetition frequency), or c) slowing down ionization by using certain gas mixtures.

In this study, we have been able to perform imaging of the discharge development in nanosecond time scales, and show that DBD uniformity strongly depends on applied electric field in the discharge gap. We show that the discharge uniformity may be achieved in the case of strong overvoltage in the discharge gap (provided by fast rise times), when anode-directed streamers are formed. We show that in the case of strong overvoltage on the discharge gap, there is transition from filamentary to uniform DBD mode. This, in turn, allows not only safety of the discharge, but also provides controllable chemistry in the vicinity of the floating electrode – treated tissue. Independence of the nanosecond-pulsed uniform FE-DBD on the second electrode is another, if not the most important and attractive quality of this type of plasma.

2. Experimental Setup and Methodology

To initiate the uniform DBD discharge in atmospheric air we used a pulsed power system. The pulse generating, monitoring and triggering was performed as described in [16, 17]. Briefly, the power supply (FID Tech Company) generated pulses with +15.5 kV pulse amplitude in 50 Ohm coaxial cable (31 kV on the high-voltage electrode due to pulse reflection), 10 ns pulse duration (90% amplitude), 2 ns rise time and 3 ns fall time. Pulses were delivered to the electrodes via 100 ft of RG 393/U high voltage coaxial cable. Due to the length of cable the pulse shape was expected to change, and assuming a linear change in amplitude and rise/fall rate per unit length of the cable, the effective amplitude change was < 5 % and the estimated voltage rise/fall time at the end of the cable was 4 ns / 5 ns respectively. A return current shunt was mounted 22 ft from output of the power supply and was used for pulse monitoring as well as triggering. The discharge cell had a sphere-to-plate geometry with a plane high-voltage electrode diameter of 2.4 cm covered with 1 mm thick quartz. The grounded bronze sphere diameter was 5 cm, and the inter-electrode distance was 0.5–4 mm.

The discharge imaging experiments were performed using 4Picos ICCD camera from Stanford Computer Optics. The camera had an 18 mm diameter multi-alkaline photocathode with a spectral response from 180 to 750 nm. The camera's typical spectral response was 250 – 750 nm. Camera was positioned perpendicularly to the discharge gap and focused using 25-mm quartz lens (Newport). Typical field of view was 5x8 mm. Discharge optical

emission spectrum was obtained using a fiberoptic bundle (Princeton Instruments-Acton, 10 fibers – 200 μm core) connected to the spectrometer (Princeton Instruments – Acton Research, TriO Vista TR555 spectrometer system with PIMAX digital ICCD camera, Trenton, NJ) calibrated using Hg (Ar) spectral calibration lamp, Newport, and 63978 NIST Traceable Quartz Tungsten Halogen Calibrated Source, Newport. Electric field measurements were performed based on the method described by Starikovskaya et al [18, 19]. Briefly, ratio of emission from 0–0 vibrational transitions of the second positive (337.1 nm) system of molecular nitrogen and of the first negative (391.4 nm) system of molecular nitrogen ions was measured and electric field value was calculated using the technique described in [18, 19].

The synchronization system for the experiments was built on the basis of a Tektronix AFG-3252 Arbitrary/Function Generator (Figure 1). The generator has synchro-output and two adjustable channels with a signal rise/fall time of less than 2.5 ns and a typical jitter (RMS) less than 20 ps with a delay time resolution of 10 ps.

3. Experimental results and discussion

3.1. Integral images of nanosecond-pulsed DBD and image analysis

The equivalent circuit of the DBD cell one may represent as shown in Figure 2, with C_g and C_d being the equivalent capacitances of gap and dielectric layer, respectively, and R – plasma resistance. As shown in, for example, [20], the initial voltage in the gap may be calculated using the following expressions (assuming no memory charge accumulation):

$$V_g = \frac{c_d}{c_d + c_g} V_0, C_d = \frac{\epsilon_0 \epsilon_d S}{d_d}, C_g = \frac{\epsilon_0 S}{d_g},$$

where V_0 is the applied voltage generated by the power supply. Below, we therefore recalculate the applied electric field in the gap as

$$E_g = \frac{c_d}{c_d + c_g} \times \frac{V_0}{d_g}.$$

Discharge images (false color) with the exposure times of 20 ns obtained for different discharge gaps and applied electric fields with the applied voltage of 23.36 kV are shown in Table 1. As the electric field if the discharge gap increases, the discharge appears to be more uniform. Already at the electric field of ~ 400 Td, almost uniform discharge is observed.

Chi-square test [21–23] is adopted for uniformity exam of the first pulse (20 ns exposure) images. In the case of filamentary discharge, the brighter pixels correspond to the position of streamers (non-uniformities), while in the image of uniform discharge brighter pixels are often spread evenly across the discharge area. Here the dependency of brighter pixels to its position is tested as a measurement of uniformity. For that, the profile emission intensity is recorded and divided into several groups according to their position along the discharge

area. And every pixel which is brighter than the average intensity is count as a “bright” pixel.

Obtained distributions are tested to see how well they fit with a chi-square distribution:

$$f(x, k) = \begin{cases} \frac{x^{(k/2)-1} e^{-x/2}}{2^{k/2} \Gamma(\frac{k}{2})}, & x \geq 0; \\ 0, & otherwise \end{cases}$$

Where x is the variance of the data, and k is the number of degrees of freedom (number of regions minus one). The acceptance threshold is chosen to be 0.05 [24]. If $f(x; k)$ is greater than 0.05 then it is safe to conclude that the bright spot distribution is random and free of its position. Figure 3 shows how $f(x; k)$ value changes when the gap distance increases. It is clear that with 1 mm gap, the discharge image is uniform.

3.2. Time-resolved imaging of DBD development and corresponding maximum local electric field measurements

Time-resolved imaging of the discharge initiation reveals fundamentally different behaviour depending on the value of applied electric field in the gap (Table 2). It should be noted, that the higher light intensity originates from the area corresponding to the head of the streamer (higher electric field) [19]. Generalizing, it is clear that at lower electric field value (2 mm, ~400 Td) the discharge develops as a conventional cathode-directed streamer, while at higher values of overvoltage almost uniform light emission is observed in the gap. We hypothesise that this is related to the change of the discharge development mechanism, namely transition to anode-directed streamers. Initial avalanches, due to high electric field in the gap, transition to streamer mode ($ad > 20$) at the very beginning of the discharge ignition, before they reach the anode. This time can be roughly estimated on the basis of avalanche development. Given the applied voltage of about 18.4 kV over a 1 mm gap, the reduced electric fields (E/n , where n is the gas concentration) is about $\sim 7 \times 10^{-15} \text{ V cm}^2$. This gives an electron drift velocity (v_d) in air of $\sim 10^8 \text{ cm s}^{-1}$ and a time of $\sim 10^{-9} \text{ s} = 1 \text{ ns}$ to bridge a 1mm gap. If the voltage rise time is shorter than the time needed to bridge the gap and the maximum voltage significantly exceeds the critical breakdown voltage (discharge develops under high overvoltage conditions) the discharge develops uniformly due to the high electric field in front of the ionization wave. The high value of the reduced electric field E/n means: (1) suppression of the instabilities by saturation of the ionization coefficient; (2) fast expansion of the plasma channels and their overlapping; (3) generation of VUV radiation and photoionization of the gas ahead of the ionization front; (4) generation of run-away electrons and pre-ionization of the gas. Thus the criteria for the uniform discharge development could be formulated as simple relations:

$$(U/n)/d \gg (E/n)_{crit} \quad (1)$$

$$\text{and } \tau_{rise} \ll d/v_d \quad (2)$$

where U is the pulse voltage, n - the gas density, d - the discharge gap length, τ_{rise} - the voltage pulse rise time and v_d - the electron's drift velocity in critical electric field.

The transition of the DBD from uniform to non-uniform regime was also confirmed by measurements of electric field. The results shown in Figure 4. From the measured values of the electric field as a function of applied electric field in the gap, one may notice that two distinctive modes of the discharge exist: high electric field mode (non-uniform regime, conventional cathode-directed streamers) and low electric field (uniform regime).

3.3. Plasma medical applications of nanosecond-pulsed DBD in atmospheric air

One of the most promising and exciting new application of atmospheric air plasmas is medicine. In the area of plasma medicine, a variety of different atmospheric pressure non-equilibrium plasma systems have been employed by researchers. They differ in uniformity of plasma produced, degree of ionization, plasma temperature, gasses employed, and ultimately in the amount and identity of active species and charged particles that can influence biological interactions. Plasma itself can be influenced significantly by living tissues or biological medium when it is produced in direct contact with them. While plasma produced in plasma jet systems can sometimes be better controlled, the flux of active species from it depends on distances from biological media and flow rates. In order to study the mechanism of interaction of plasma with biological systems it is critically important to be able to produce plasma that, on the one hand, is capable of delivering all possible active agents (radicals, electric fields, charges) to the biological targets and, on the other hand, can be well-characterized and controlled during the treatment process. Nanosecond-pulsed dielectric barrier discharge is uniquely suited for that purpose because it can be applied directly to the biological target delivering all active species that non-equilibrium plasma can produce and, on the other, as we show in this study, it produces highly uniform plasma independently of the features of the biological target which permits effective characterization and control of the plasma. One of the important aspects that arises while dealing with plasmas for medical applications is safety. Non-uniformity of plasma, i.e. high current density of the filaments, and therefore temperature, presents a danger of damaging the treated tissues and cells. This threat may be eliminated again by usage of uniform, i.e. controlled and measurable, dielectric barrier discharge. Unfortunately, at atmospheric pressure, a uniform DBD can be easily transformed into a filamentary dielectric DBD; therefore some serious issues arise, such as gas heating due to strong discharges in the random microdischarge channel and non-uniform energy distribution, which adversely affect applications. These issues traditionally are solved by use of an appropriate working gas composition, an alternating current driving frequency, lowering of gas pressure, etc [10–12]. The transitions between discharge modes in the same experimental conditions have been thoroughly investigated in nitrogen, rare gases and their mixtures with air and other gases [2, 8, 13, 15]. In the case of plasma medicine, these methods, especially those related to gas composition and pressure, obviously may not be applied in a convenient manner. Here we show that by controlling the applied (global) electric field in ns-pulsed DBD, it is possible to control the uniformity of the discharge. In addition, this offers better controllability of the discharge chemistry due to local changes of electric fields and therefore electron energy distribution function. Measurements of production of reactive oxygen and nitrogen species

in nanosecond-pulsed DBD as a function of applied and local electric field (uniform and non-uniform modes) are underway.

4. Conclusions

The development of nanosecond-pulsed dielectric barrier discharge in atmospheric air has been studied by means of fast ICCD imaging and spectroscopic measurements of local electric fields. The major results can be summarized as follows:

- It is shown that the discharge operates in two distinctively different modes which appear as “uniform” and “non-uniform” regimes. Qualitative uniformity analysis of the discharge images is performed using chi-square test.
- It is shown that measured maximum local electric field in the discharge is in a good agreement with these modes. We hypothesize that these results can be qualitatively explained by the absence of individual streamers in the uniform mode due to their overlapping and corresponding decrease of the maximum local electric field to the value of average electric field if the discharge.

Due to a strong coupling between discharge physics, and reduced electric field in particular, and plasma chemistry (which in turn determines applications of plasmas) [25], possibility of controlling discharge basic parameters together with its uniformity by simply changing applied voltage or distance between electrodes offers unique and exciting opportunities in a wide range of applications, from treatment of biological tissues to energy applications.

Acknowledgments

This work was supported in part by NIH Grant 1 R01 EB013011-01, specifically focused on understanding of DBD applicability in orthopedics via stimulation of stem cell differentiation and the understanding of the underlying mechanisms, and W.M. Keck Foundation Grant focused on understanding of nanosecond-pulsed DBD plasma physics, and its effects on biochemical processes.

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Figure 1.
Experimental setup

Figure 2.
The DBD electrode configuration and equivalent circuit.

Figure 3.
Chi-square test results for DBD uniformity.

Figure 4.

Electric field evolution after the beginning of the voltage pulse in nanosecond-pulsed DBD in atmospheric air (left, 2 ns exposure time, 100 accumulations), and average measured electric field in DBD as function of applied electric field (20 ns exposure time, 100 accumulations).

Table 1

Nanosecond-pulsed DBD images obtained using fast ICCD camera at different applied electric fields

Electrode gap, mm	Applied voltage on the gap, kV	Applied electric field, kV/cm	Applied electric field, Td	False-color image (20 ns exposure, no accumulation) and corresponding distribution of emission intensity viewed at mid-gap
1	18.4±0.1	185	711	
2	20.6±0.1	103	397	

Electrode gap, mm	Applied voltage on the gap, kV	Applied electric field, kV/cm	Applied electric field, Td	False-color image (20 ns exposure, no accumulation) and corresponding distribution of emission intensity viewed at mid-gap
3	21.5±0.1	72	275	
4	21.9±0.1	54.8	211	

Table 2

Fast ICCD images of DBD development in atmospheric air. Grounded cathode – on top, powered anode – on bottom. 200 ps exposure, 100 accumulations.

Time step	1 mm, applied electric field 185 kV/cm	2.5 mm, applied electric field 84 kV/cm
0.1 ns		
0.2 ns		
0.3 ns		
0.4 ns		