Time and space resolved measurement of electric field and electron density in an atmospheric pressure helium/nitrogen dielectric barrier discharge

Niklas Nawrath[1,2], Nikita Bibinov[2], Ihor Korolov[2], Peter Awakowicz[2] and Andrew R. Gibson[1]

[1]*Research Group for Biomedical Plasma Technology, Ruhr University Bochum, Germany*
[2]*Institute for Applied Electrodynamics and Plasma Technology, Ruhr University Bochum, Germany*

Abstract: The characterisation of atmospheric pressure dielectric barrier discharges (DBD) requires high temporal and spatial resolution due to its rapidly changing parameters. In this study, a collisional radiative model and absolutely calibrated images of nitrogen emission with high temporal and spatial resolution were used in order to determine the electric field and the electron density of a helium/nitrogen plasma with electric field and electron density varying from 30 to 80 Td and $0.5 \cdot 10^9 – 10^{10}$ cm^{-3}, respectively.

Keywords: dielectric barrier discharge, emission spectroscopy, collisional-radiative model

1. Introduction

Non-thermal atmospheric pressure plasmas are useful tools in many applications, where they serve as a source of excited species and chemical active radicals. Here, the dielectric barrier discharge (DBD) gained broad application, especially in various biomedical application and surface functionalization [1]. In order to understand the plasma processes and their outcomes, the plasma sources in use have to be characterized in detail. Three important parameters to investigate are the reduced electric field E/N, the electron density n_e and the gas temperature. These parameters are fundamental plasma properties that characterize the discharge and influence plasma processes like excitation, ionization and chemical reactions. In atmospheric pressure DBDs, the former two parameters vary on nanosecond time scales during the ignition pulse. Due to the low mean free path of the electrons, the spatial distribution also varies on micrometre scales. Therefore, to understand the physical and chemical properties of these plasma sources and optimize their applications, diagnostics are needed which provide sufficient temporal and spatial resolution. Previous studies have shown that a combination of emission spectroscopy and numerical simulations can provide sufficient resolution in time and space to describe the distribution of E/N [2], [3] as well as n_e [4]. This combination, called a collisional radiative model, gives access to high-resolution information about the discharge. Briefly, in such a model, the electron excitation rate coefficients are determined in dependency of the reduced electric field using a simulated electron velocity distribution function (EVDF) in kinetic energy scale. Comparison of the measured intensity of two suitable nitrogen emission bands with the calculated excitation rates then leads to the reduced electric field and by using the absolute emission intensities, the electron densities can be determined.

2. Experimental & setup

Fig. 1 shows a schematic of the experimental setup. The discharge is operated in an enclosed vacuum vessel that allows for precise control of operating conditions like gas composition and pressure, which are set to 2 slm helium with admixture of 9 sccm nitrogen and 1 bar, respectively. Pressure and gas flow are controlled using mass flow controllers (El-Flow Prestige, Bronkhorst, Germany) for each gas (5.0 purity, Air Liquide, Germany) and a vacuum pump with a needle valve. The grounded and driven electrodes are covered by a section of silicon wafer and an alumina ceramic with a diameter of 10 mm and a thickness of 1 mm. Both electrodes are separated by a gap of 0.8 mm, where the discharge is located. The discharge is ignited by a high voltage damped sinusoidal pulse sequence at a pulse repetition frequency of 445 kHz and peak-to-peak voltage of maximum 14 kV. The pulse sequence repetition frequency amounts to 1 kHz. This results in a time averaged deposited power of ~1.3 W. Spatially and temporally resolved measurements were performed by a pco DiCam CCD sensor. The sensor in combination with two bandpass filters for the emission of N_2(C-B) and N_2^+(B-X) at 380±5 nm and 390±5 nm, respectively, is calibrated separately to absolute units using a nitrogen microwave plasma source and an absolute calibrated, high-resolution echelle spectrometer, as described in [4]. To estimate the gas temperature, emission spectra were measured using an echelle spectrometer.

3. Methods

In the present study, the reduced electric field and the electron density of an atmospheric pressure dielectric barrier glow discharge with small admixture of nitrogen (0.45 %) is characterized with a temporal and spatial resolution of 40 ns and 14,6 µm, respectively. The

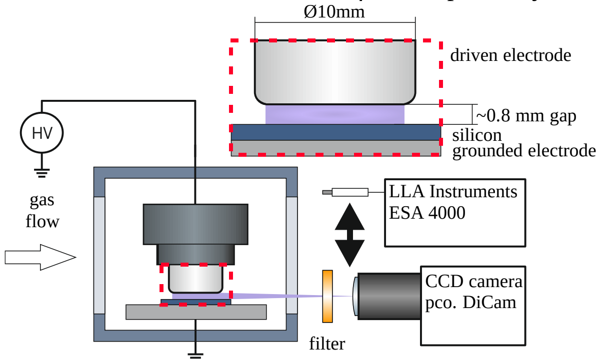

Figure 1: Schematics of the experimental setups for the characterization of dielectric barrier discharge

integrated emission of N_2(C-B, 0-2) and N_2^+(B-X, 0-0) molecular bands is obtained at 380 nm and 390 nm, respectively, and Abel inversion of the images provides cylindrically symmetric radial distribution of the emission. Penning ionization via helium metastable states is assumed to be the main ionization process for the production of N_2^+(B,0). Under this assumption, balance equation for N_2^+(B,0)- and N_2(C,0)-states as well as helium metastable He_{m1et} lead to a set of differential equations, that are solved to obtain a ratio of excitation rate constants $k_{exc}^{He_{met}}/k_{exc}^{N_2(C,0)}$ as a function of reduced electric field. At the same time, this ratio can be calculated with the experimental intensities of N_2(C-B) and N_2^+(B-X) under the above mentioned assumption.

After Abel inversion of the measured images, the reduced electric field for each recorded pixel can be determined by comparing the measured data with the theoretical data applying a polynomial fit. Using the electric field in each pixel, the excitation rate constant for N_2(C, 0) (in cm^3s^{-1}) can be calculated. By combining this rate constant and the measured absolute emission (in photons $\cdot$ $cm^{-3}s^{-1}$), the electron density can then be calculated for each recorded pixel at each time point.

4. Results and Discussion

Figure 2 and 3 shows the radial (r) and axial (z) distribution of the reduced electric field and the electron density in the first 40 ns of the first and the second half-wave (hw) of the damped sinusoidal. The anode and the cathode are located at $z = 0$ mm and $z = 0.8$ mm, respectively, for the first half-wave and exchange positions for the second half-wave. The area outside the lines in figures 2 – 4 indicate the plasma regions that are problematic to analyse in the current data, probably due to a non-optimal optical alignment of the camera to the discharge region or non-reliable assumptions of the applied collisional-radiative model. Future measurements with an optimised optical setup should improve the data in these regions. The emission of the nitrogen states can be measured up to a radius of ~4.5 mm in each case, outside of this region, no significant emission occurs.

The highest electric field can be found at the cathode, where the cathode fall is located. Simulations carried out under similar, but not identical conditions [5], [6], indicate that the cathode fall thickness in a helium/nitrogen glow-like DBD is approximately

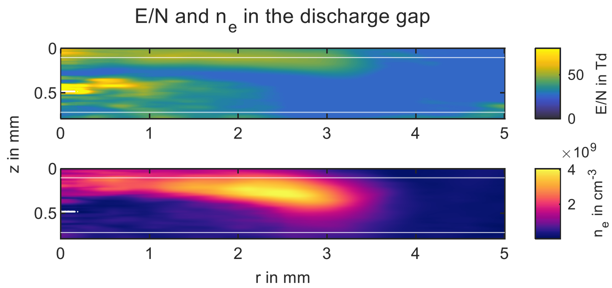

Figure 3: Same as fig. 2 for the second half-wave with reversed polarity. The small white spot at the centre is caused by negative values after Abel inversion.

0.2 mm to 0.6 mm in thickness with decreasing nitrogen admixture from 150 ppm to 50 ppm, while figure 4 shows a cathode fall thickness of approximately 0.6 mm and 0.2 mm for the first and the second half-wave, respectively. The maximum reduced electric field of ~80 Td is also comparable to the simulated values of 40 – 80 Td for 50 – 150 ppm of N_2. The measured electron density is approximately one magnitude lower compared to the simulated electron density. Although the simulated and measured values compare, the nitrogen admixtures differs between 50 – 150 ppm in the simulation and 4500 ppm in the measurement and nitrogen as a diatomic molecule absorbs large amounts of energy in its rotational and vibrational states that, as

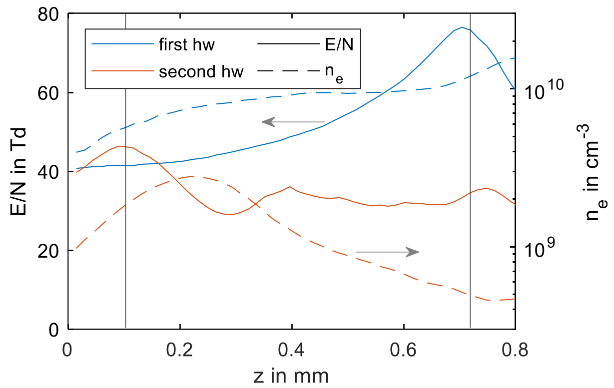

Figure 2: Axial distribution of reduced electric field and electron density in the discharge gap in the first 40 ns of the first (blue) and the second half-wave (red). Note that the polarity of the electrodes changes between both measurements. Outside of black vertical lines, data is possibly non reliable.

a consequence, is not available for ionization processes. For the second half-wave regions can be found that could be classified as a negative glow (0.2 mm to 0.4 mm) with an increasing field and a positive column (>0.4 mm) with nearly constant field.

The distribution of electrons in both half-waves reduces from the cathode to the anode with a peak for the second half-wave at the minimum of the electric field and, for the second half-wave, compares well to the distribution found in the simulations.

The gas temperature was determined to be 317 ± 3 K using the emission of N_2(C-B, 0-0) band at 337.1 nm.

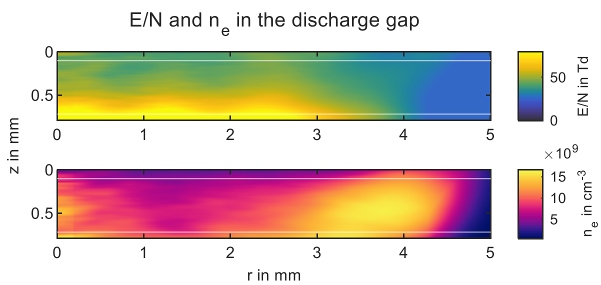

Figure 4: Spatial distribution of reduced electric field and electron density in the discharge gap in the first 40 ns of the first half-wave with the cathode located at the bottom. Outside of white rules, data is possibly not reliable.

5. Conclusion

In this study the application of collisional radiative models is extended to helium with low nitrogen admixtures, where several processes like Penning ionization have to be considered. Further, a high spatial and temporal resolution was acquired for an atmospheric pressure dielectric barrier glow discharge for electron density and electric field measurements. Using a method like this, the discharge dynamics of helium nitrogen plasmas can be investigated with a high level of detail and with small adaptions to this model, variations of nitrogen concentration can be taken into account.

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