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Nanosecond repetitively pulsed glow discharges in atmospheric pressure air at 300 K

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Abstract: Non-equilibrium air plasmas with high densities of reactive species and low gas heating are produced at 1 atm, 300 K with nanosecond repetitively pulsed glow discharges. The domain of existence of the glow regime is explored. Optical and chemical diagnostics are used to characterize the densities of ozone and nitric oxides in the post-discharge.

Keywords: nanosecond repetitively pulsed discharges, air plasmas, glow regime, cold plasmas, atmospheric pressure, surface treatment, ozone, nitric oxides, absorption spectroscopy

1. Introduction

Plasma treatment of thermosensitive materials or biological systems is usually performed with nonequilibrium microplasma discharges (see for example Ref. [1], [2], [3]) working with noble gases such as helium or argon, with small additions of oxygen, nitrogen, or water to increase the reactivity of the gas. Replacing noble gases with air would considerably simplify the implementation of these treatment methods as this would eliminate the need for compressed gas cylinders and reduce operating costs. However, producing atmospheric pressure air plasmas with high densities of reactive species and temperatures remaining close to 300 K has been a great challenge for many years ([4], [5]).

In recent years, Pai et al. [6] proposed a method for producing glow discharges in atmospheric pressure air using high voltage Nanosecond Repetitive Pulses (NRP), and a criterion for the existence of a diffuse, low temperature regime called the NRP glow regime. Based on these results, Rusterholtz [7] successfully demonstrated the existence of the NRP glow regime in air at 300 K and 1 atm, using high voltage pulses (duration 10 ns, amplitude 15 kV, repetition frequency 1–30 kHz) applied across two pin electrodes with a gap distance of 2.5 mm.

The first objective of this work is to explore the domain of existence of NRP glow discharges in atmospheric air as a function of key electrical and geometric discharge parameters. The second objective is to measure the production of active species such as ozone and nitric oxides in these discharges.

2. Domain of existence of the NRP glow in ambient air

The existence of the NRP glow regime is highly sensitive to the choice of a few key parameters, namely the pulse voltage amplitude, pulse repetition frequency, pulse duration, flow velocity, gap distance, and sharpness of the electrodes. The reactor consists of two pin

electrodes with adjustable gap distance (2–15 mm) placed in a transverse air flow of variable velocity up to 10 m/s. High voltage pulses of duration 20 ns and amplitude between 4 and 30 kV are applied at a repetition frequency ranging from 10 to 100 kHz.

The domains of existence of the NRP glow are investigated experimentally for different sets of input parameters. Figure 1 shows the results for the case of a discharge applied at 10 kHz in air flowing at 3.4 m/s. The measured upper and lower limits of the domain shown in the figure match the glow-to-spark and corona-to-glow transitions, respectively, predicted by Pai et al. [6].

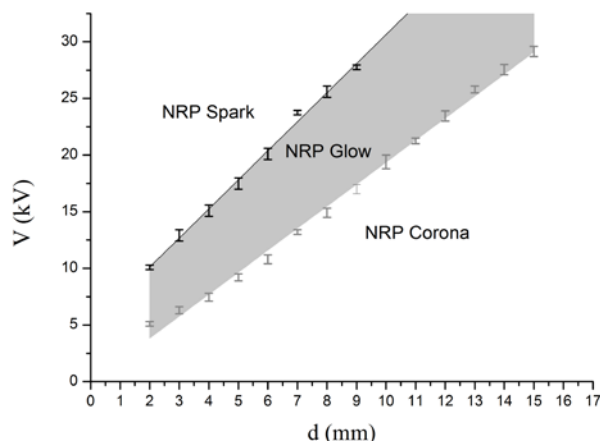


Fig. 1. Domain of existence of the NRP glow regime for a given set of parameters ($v = 3.4$ m/s, $f = 10$ kHz).

The slope of the upper limit of the domain (glow-to-spark transition) is equal to the spark breakdown electric field. In Fig.1, this value is about 26 kV/cm. Similarly, the slope of the lower limit corresponds to the field required for the corona-to-spark transition, and its value in Fig.1 is about 19 kV/cm. A parametric study was conducted to assess the influence of the repetition

frequency (1-100 kHz, results on Fig. 2) and of the air flow velocity (0.3-10 m/s) on these two transition electric fields.

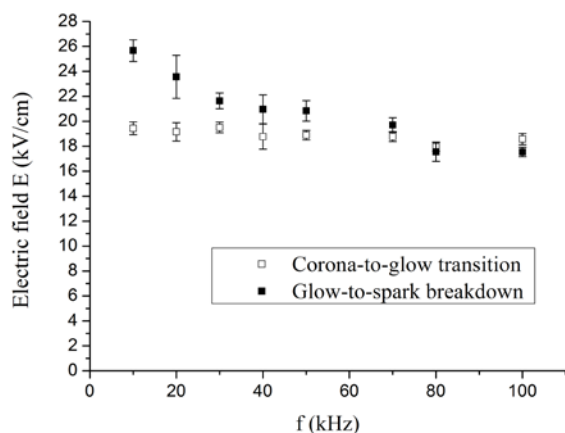


Fig. 2. Transition electric fields (Corona-to-glow and Glow-to-spark) as a function of repetition frequency. Air velocity is kept constant at 3.4 m/s.

The corona-to-spark transition field is independent of these two parameters. In contrast, the spark breakdown electric field E_{GS} is found to decrease with the repetition frequency and to increase with the flow rate. This can be explained as follows: as shown for instance by Pai et al. [6], the reduced electric field $(E/N)_{GS}$ is independent of the temperature. The gas density N is inversely proportional to the temperature. Thus, the decrease of the spark breakdown field E_{GS} with the repetition frequency reflects an increase of the gas temperature. This gas temperature increase is the result of higher energy addition to the gas at high repetition frequency. Conversely, higher flow rates limit gas heating, hence they reduce the spark breakdown electric field.

3. Post-discharge analysis

The concentrations of reactive species in the post-discharge of the NRP glow regime are studied by optical and chemical analysis. In particular, the concentrations of NOx and ozone are investigated, and ozone production in the NRP glow and spark regimes are compared. NOx (NO and NO₂) production is monitored by an Ecom J2KN Pro NOx analyzer. The ozone concentration is measured with UV absorption spectroscopy in the Hartley band (200-310 nm), using an OceanOptics Maya2000 Pro spectrometer. A Deuterium lamp is used as the light source. The reactor consists of the pin-to-pin configuration used above, with fixed repetition frequency of 30 kHz, gap distance of 6 mm, and air flow velocity of 1.2 m/s.

Electrical diagnostics are used to measure the energy deposited in both the glow and the spark regimes. The voltage amplitude and current through the plasma are monitored with high voltage probes (Lecroy PPE20kV) and a Pearson coil current monitor (model 6585). The

electric signals are synchronized at the sub-nanosecond scale and recorded with a Lecroy 7100 WavePro digital oscilloscope (1 GHz). We determined the reactor capacitance to isolate the conduction current from the displacement current corresponding to the charging and discharging of the reactor capacitance.

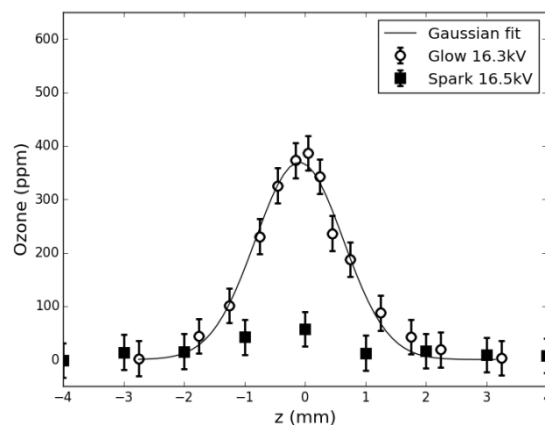


Fig. 3. Radial distribution of ozone density in the post-discharge (17 mm downstream of the electrodes) in the NRP glow (16 kV) and spark (16.5 kV) regimes. Spatial resolution: 300 μ m.

In the post-discharge of the NRP glow regime, the NOx production reaches a maximum of 70 ppm (20 mm downstream of the electrodes). The ozone concentration is found to increase linearly with the voltage and the energy deposited. A peak of 400 ppm in the post-discharge (17 mm downstream of the electrodes) is detected at a voltage amplitude of 16 kV (Fig 3). In the spark regime, a significant drop in ozone production is observed although the energy deposited increases by more than a factor of 10 (from less than 100 μ J to 1 mJ). This drop is explained by the higher stability of ozone at lower temperature, and the very low heating in the NRP glow discharge compared to the NRP spark regime. Complementary temperature measurements are conducted to assess this hypothesis. Gallium thermometer measurements in the post-discharge (20 mm downstream of the electrodes) evidenced a heating of $3 \pm 2^\circ\text{C}$ in the glow regime, and a heating of $30 \pm 5^\circ\text{C}$ in the spark regime. Emission spectroscopy of the second positive system of nitrogen is also used to measure the rotational temperature of N₂(C) in the discharge.

4. Conclusion

The domain of existence of the NRP glow regime in air at 300K is explored. It is shown the discharge can be sustained over a broad range of geometric and electric parameters, with gap distance of up to 15 mm, frequencies of 100 kHz and air flow velocity of 10 m/s. Chemical and optical diagnostics are used to characterize the densities of nitric oxides and ozone in the post-discharge. A high density of ozone (400 ppm) is

measured and is explained by the high reactivity and low heating of the NRP glow discharge. Temperature measurements confirm the expected negligible heating of the air flow in the NRP glow post-discharge.

5. References

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