DC Glow Discharges in Atmospheric Pressure Air

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***Abstract*:** We present experimental investigations of DC glow discharges in atmospheric pressure air with the aim of producing nonequilibrium air plasmas with high electron density (~10^{12} cm^{-3}) and relatively low gas temperature (less than 2000 K). Such plasmas are potentially interesting for many applications, including air pollution control. The discharge of our study is ignited by a streamer-to-spark transition, but thanks to an appropriate ballast resistor, it operates in a pulseless regime with currents from 2 to 500 mA, current densities of 1-10 A/cm^2, and electric fields of 3000-300 V/cm. Spectroscopic and electrical measurements show that the discharge is of the glow type and generates a nonequilibrium air plasma. We also describe an innovative approach where thermionic cathodes and tubes with swirl gas flow are employed. With this approach, electron densities of up to 10^{13}-10^{14} cm^{-3} can be obtained and the production of relatively large plasma volumes is possible.

Introduction

Atmospheric pressure air plasmas present considerable interest for a wide range of applications such as air pollution control, bio-decontamination, plasma-assisted combustion, material processing, surface treatment, and electromagnetic wave shielding. Desirable conditions are high electron densities (above 10^{12} cm^{-3}) and relatively low gas temperatures (below 2000 K). These properties can only be achieved in nonequilibrium plasmas where the kinetic temperature of the free electrons, T_e, is higher than the temperature of heavy species (gas temperature), T_g.

Various types of atmospheric DC or AC glow discharges as sources of nonequilibrium plasmas have received renewed attention during the past few years. The subject of our research is a direct current (DC) glow discharge in atmospheric pressure air. This discharge type does not use dielectric barrier layers, and as such should not be identified with the recently widely investigated atmospheric pressure glow discharges (APGD). APGDs in nitrogen or noble gases can produce large homogeneous plasmas that are suitable e.g. for surface treatment, but they typically become filamentary in air. The DC glow discharges presented here have the advantage of producing relatively large volumes of fairly homogeneous plasma. In addition, DC operation enables an easy control of the current and plasma properties. Examples of atmospheric air discharges most similar to the type presented here are micro-hollow cathode discharges (*1*), discharges with one or both electrodes covered by water (*2*-*3*), and low current DC glow discharges (*4*-*5*).

In the main part of this paper, we provide the general characteristics of a DC glow discharge in ambient atmospheric pressure air obtained by electrical and spectroscopic measurements. Results from the original study performed at the LPGP, University Paris XI, France (*6*) are combined with results obtained at Stanford University, CA, USA, (*7*-*8*). This type of discharge was successfully demonstrated previously at low currents for the abatement of volatile organic compounds (VOCs) (*6*, *9*). In the last part of the paper, we describe a novel method to increase the electron density and to enhance the discharge stability using thermionic cathodes and tubes with swirl gas flows.

Experimental

We used two different DC power supplies, placed in series with ballast resistors, to sustain the discharge: a Del High Voltage RHVS (10 kV, 1.5 A) at Stanford and a Del High Voltage RHVS (60 kV, 5 mA) in Paris. The experimental setup is schematically shown in Figure 1. The ballast resistor was

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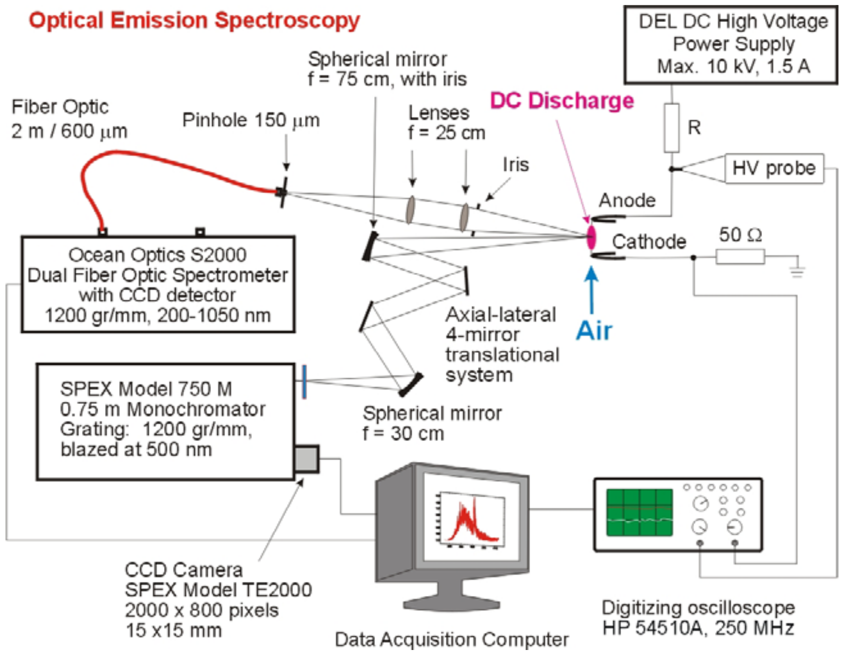


Figure 1. Experimental setup for optical emission spectroscopy and electrical measurements of the DC discharges.

used to stabilize the discharge. Its value was chosen experimentally, typically between 3 and 500 kΩ, depending on the operating current. The discharge voltage was measured with high voltage probes: Tektronix T6015A (1000 x, 3 pF, 100 MΩ) and North Star PMV10 (200 x). The discharge current recorded across a 50 Ω resistor and the discharge voltage measured by the probes were processed by digitizing oscilloscopes (400 MHz Tektronix DSA 602 in Paris, and 250 MHz HP 54510A at Stanford).

Rhodium points opposite to copper planes were used as electrodes in the original study in Paris, while two platinum pins were used at Stanford. The electrode material is not crucial for the discharge mechanism, but inert metals are preferable to prevent electrode corrosion. We used electrodes with small radius of curvature (pins) because they permit easier discharge ignition. In the most recent Stanford experiments described in the last part of this paper, we replaced the metal cathodes with cathodes made of thermionic materials ($LaCrO_3$, LaB_6 and Mo), and we placed the discharges in glass or quartz tubes with swirl injection of ambient air.

Spatially resolved optical emission spectroscopy and electrical diagnostics were employed to characterize the discharge properties. Figure 1 shows a schematic of the two spectroscopic systems used at Stanford. An Ocean Optics S2000 dual channel spectrometer, fitted with two grating/CCD combinations, provides quick but low resolution scans in the 200-500 and 400-1050 nm spectral ranges, with respective wavelength resolutions of 0.41 and 0.88 nm. A 75-cm monochromator SPEX 750M (200-800 nm grating) fitted with a 2000 x 800 pixel CCD camera SPEX TE2000 (15 x 15 μm pixel dimension) provides spectral resolution of 0.12 nm, sufficient to resolve the rotational structure of molecular spectra. Furthermore, this second system provides wavelength-specific CCD camera images of the discharge, which are useful for measuring its radial diameter. Both spectroscopic systems have two-dimensional scanning capability with a spatial resolution of 250 μm. Absolute intensity calibrations were obtained by means of two radiance standards traceable to NIST calibrations. A similar optical diagnostics set-up was used in Paris, using a Jobin Yvon monochromator HR 640 (200-700 nm, best resolution 0.01 nm) combined with a photomultiplier tube Hamamatsu C659S. A digital camera Nikon Coolpix 990 was used for photo-documentation of discharges.

Results and Discussion

The DC atmospheric pressure air discharge under study is a stable continuous discharge regime with no pulses. It operates with DC currents from 1.6 to several hundreds of mA, and DC voltages from a few kilovolts to a few hundred volts. The gap length (interelectrode distance) can be varied from 1 mm to a few cm, depending on the gas flow conditions and the current. The voltage-current characteristic of the discharge in ambient air is descending, as shown in Figure 2. In contrast, the discharge power increases with the current, as shown in Figure 3.

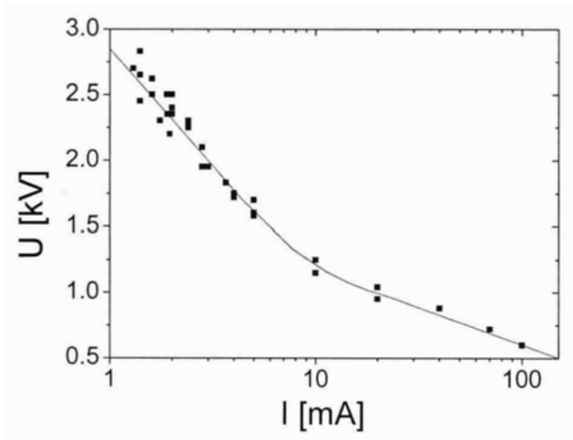


Figure 2. Voltage-current characteristic of the DC discharge in ambient air. Interelectrode distance d = 7 mm.

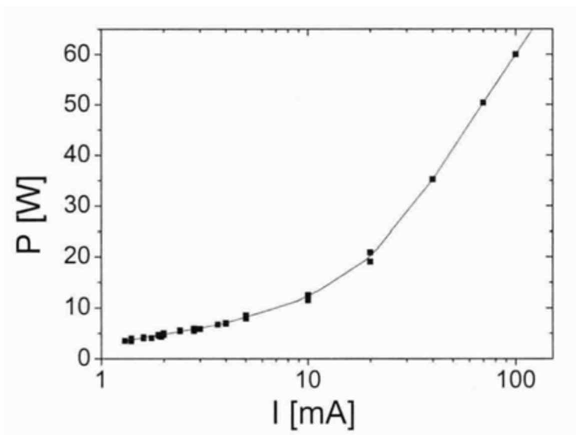
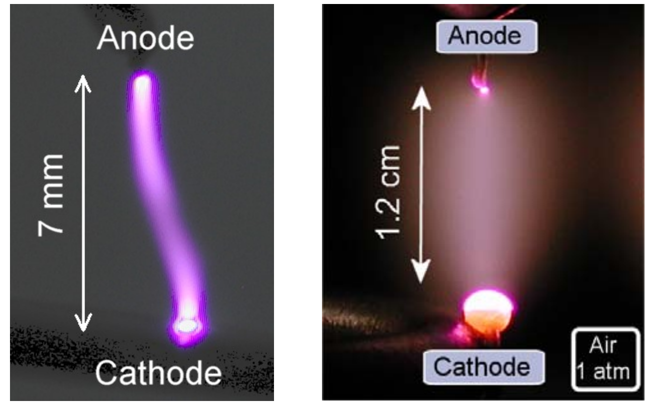


Figure 3. Discharge power as a function of current. Interelectrode distance d = 7 mm.

The discharge is ignited by a streamer-to-spark transition, but the ballast resistor immediately limits the spark current. However, the ballast resistor is chosen experimentally so that the limiting current is large enough that the discharge, after extinction of the initial spark phase, enters a state of permanent conduction. The ballast-limited current then controls this pulseless discharge regime.

Figure 4 shows photographs of DC glow discharges in ambient air at atmospheric pressure operating at low flow velocity (about 0.2 m/s) and currents of 5 mA and 100 mA.

We recorded the spectra emitted by the DC air discharge in the 200-1050 nm region for various discharge parameters. The N_2 ($C^3\Pi_u$-$B^3\Pi_g$) and NO γ ($A^2\Sigma^+$-$X^2\Pi_r$) spectral systems were then used to measure the rotational and vibrational temperatures, T_r and T_v, by comparison with simulated spectra. In atmospheric pressure plasmas, the rotational temperature is close to the gas temperature ($T_r \approx T_g$) owing to fast collisional relaxation. The gas temperature T_g, measured at the centerline of the discharge column,



a)
I = 5 mA
U = 1.7 kV
d = 7 mm

b)
I = 100 mA
U = 1.5 kV
d = 12 mm

Figure 4. DC glow discharges in ambient air flow (v = 0.2 m/s) at atmospheric pressure (different exposure times were used).

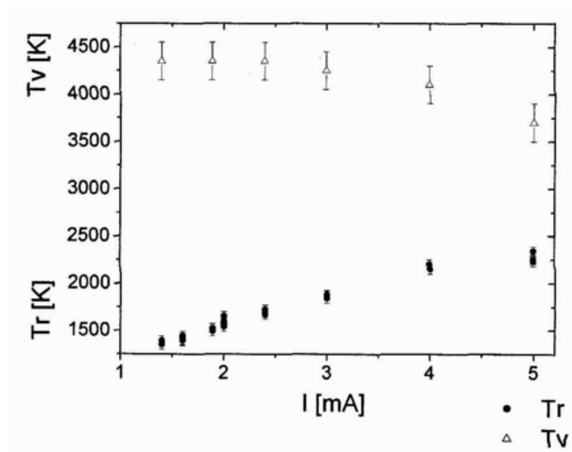


Figure 5. Rotational and vibrational temperatures as functions of the discharge current in ambient air at low flow velocity v ≈ 0.2 m/s, interelectrode distance d = 7 mm.

varies typically in the range of 1500-2500 K. The current and the gas flow velocity control the power density deposited in the discharge, and thus the gas heating. At a given low flow velocity of ambient air through the discharge, T_g increases and T_v decreases with increasing current (Figure 5). The measured vibrational temperatures T_v of the excited states are around 4000 K. These temperatures are higher than the gas temperature and therefore suggest that the plasma is in a state of thermal nonequilibrium.

Besides temperature measurements, optical emission spectroscopy diagnostics also provides the spatial distributions of emission intensity. The axial emission intensity profiles indicate the stratification into dark and bright layers, typical of low pressure glow discharges (*10*). The bright and dark regions near the cathode are reminiscent of the negative glow and the Faraday dark space, although their dimensions seem quite large to identify them without further analysis. Nevertheless, the positive column occupies most of the interelectrode space. Measurements of the floating plasma potential along the discharge axis by means of a platinum pin probe enable us to calculate the electric field strength E with better than 10% uncertainty. The measured electric field is approximately uniform in the positive column. It decreases from 3000 to 300 V/cm as the discharge current is increased, thus leading to a falling voltage-current characteristic (Figure 2). The voltage drop across the very thin region next to the cathode is close to 277 V for the Pt cathode, and 370 V for the Cu cathode. These values are typical of the cathode fall of glow discharges in air with Pt and Cu cathodes (*10*).

The radial emission intensity profiles of N_2 ($C^3\Pi_u$-$B^3\Pi_g$), NO γ ($A^2\Sigma^+$-$X^2\Pi_r$) and OH ($A^2\Sigma^+$-$X^2\Pi_{3/2}$) systems were used to measure the diameter of the positive column. This approach assumes that the emission profiles are representative of the electron density profile. We confirmed this assumption experimentally in an atmospheric pressure nitrogen DC discharge where we have obtained reasonable agreement between various emission profiles of excited N_2 and N_2^+ states, and the electron density profile calculated from the N_2^+ X (v=0) ion concentration measured by cavity ring-down spectroscopy (*11*). Here, in the air discharge, the diameters measured from NO γ and OH are typically about 1.2-1.5 times larger than the diameter measured from N_2. This result can be explained by assuming direct electron impact as the dominant excitation mechanism, and a radially decreasing distribution of electron energies. Electrons with higher energy are needed to excite N_2 C state (11 eV), while energies of only about 5.5 and 4 eV are sufficient to excite NO A and OH A from their ground states. Thus, the spatial extent of the N_2 C-B radiation is narrower than those of NO γ A-X and OH A-X. Nevertheless, the width of the emission profiles of N_2, NO, and OH are within 20-50% of each other and this gives a reasonably accurate estimate of the discharge diameter and related parameters, namely the current density.

The estimated discharge diameter is then used to estimate the plasma volume and the current density j. Knowledge of E, j and the gas density N (calculated from the measured T_g) enables us to estimate the plasma conductivity, hence the electron density n_e, using Ohm's law (*8, 10*). The measured discharge diameters range from 0.4 to 3.3 mm, depending on the current, the gas flow velocity, and the gas temperature. The corresponding current densities j are between 0.1 and 10 A/cm^2 and the estimated electron densities n_e are in the range of 10^{12}-10^{13} cm^{-3}. The measured j is lower than the typical current density of arcs. Moreover, the measured cathode fall is typical of glow discharges. The stable operation of this discharge is supposedly provided by the secondary electron emission from the cathode. The high values of n_e at gas temperatures between 1500 and 2500 K confirm the departure from thermal nonequilibrium. (LTE values of n_e in atmospheric pressure air are about 10^2, 10^6, and 10^9 cm^{-3} for temperatures of 1500, 2000, and 2500 K, respectively).

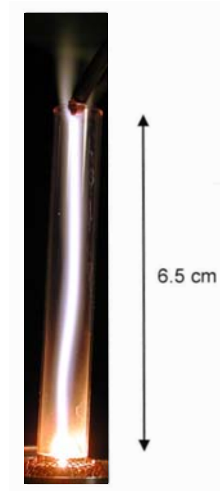


Figure 6. DC air discharge with a $LaCrO_3$ cathode in a glass tube and vortex flow, Q = 110 slpm, tube diameter 8.38 mm, I = 500 mA, discharge diameter 1.8 mm.

Recently we employed thermionic cathodes and swirl flow injection of ambient air to the discharges surrounded by glass or quartz tubes. Various cathode materials, tube diameters and flow properties were tested. With this approach, stable discharges can be obtained with gap lengths up to more than 10 cm, which is interesting for plasma volume scaling. The swirling flow in the tube confines the discharge, and thus the current density increases up to 10-40 A/cm^2 and the electron density up to 10^{13}-10^{14} cm^{-3}. This represents an enhancement of n_e by 1-2 orders of magnitude with respect to our typical discharges with metal cathodes in open air. The electric field is low (200-600 V/cm) with these high current density discharges, and T_g often reaches 3000 K. Under these conditions, the discharge may be classified as a swirl-stabilized regime operating at the transition between glow and arc. Nevertheless, the plasma is still in a state of nonequilibrium, because the measured n_e is about 3-4 orders of magnitude higher than the equilibrium n_e at 3000 K (~10^{10} cm^{-3}). Figure 6 shows a photograph of a 6.5 cm long discharge in a glass tube with a lanthanum chromite ($LaCrO_3$) cathode. Further investigations are required to understand the relative importance of the swirl flow and the thermionic effect.

Conclusions

This work demonstrates that it is possible to maintain stable atmospheric pressure DC discharges in ambient air without arcing. The discharge properties are those of a glow discharge, with a distinct cathode layer and positive column. This type of discharge provides a source of highly ionized non-equilibrium air plasma with electron number densities of the order of 10^{12}-10^{14} cm^{-3}, and gas temperatures of 1500-3000 K. The ease of DC operation and the volume scalability of the generated non-equilibrium plasma make this discharge attractive for many applications, including pollution control.

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