DC DISCHARGES IN ATMOSPHERIC PRESSURE AIR IN THE GLOW AND GLOW-TO-ARC TRANSITION REGIMES

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ABSTRACT

We experimentally investigate DC discharges in air at atmospheric pressure. Spectroscopic and electrical measurements show that at currents below 100 mA, these discharges are of the glow type and generate nonequilibrium air plasmas with high electron densities (~10^{12} cm^{-3}) and relatively low gas temperatures (~2000 K). With increasing current, the gas temperature increases and a gradual transition toward near-LTE conditions occurs. Nevertheless, this glow-to-arc transition is smooth, so stable discharges may be established between the glow and arc regimes, generating still non-thermal but relatively hot plasma. These DC discharges represent interesting sources of non-thermal atmospheric air plasmas with adjustable properties.

INTRODUCTION

Air plasmas at atmospheric pressure present considerable interest for a wide range of applications such as air pollution control, bio-decontamination, material processing, plasma-assisted combustion, aerodynamic flow control, and electromagnetic wave shielding. Generally desirable are plasmas with high electron densities (above 10^{12} cm^{-3}) and relatively low gas temperatures (below 2000 K). These properties can only be achieved in thermal nonequilibrium where the electron temperature, T_e, is higher than the gas temperature, T_g.

Atmospheric DC or AC discharges as sources of nonequilibrium plasmas have received renewed attention during the past few years. Our investigations are aimed at DC discharges in atmospheric pressure air. These discharges do not use dielectric barrier layers, and as such are different from the recently widely investigated atmospheric pressure glow discharges (APGD). The DC discharges presented here have the advantage of producing relatively large volumes of fairly homogeneous air plasmas. In addition, DC operation provides easy control of the current and plasma properties.

The paper synthesizes results obtained at the LPGP, University Paris XI, France [1] and related results obtained at Stanford University, CA, USA, [2]. This discharge type was successfully applied previously at low currents for the abatement of volatile organic compounds (VOCs) [1,3].

EXPERIMENTAL

A sketch of the experimental setup is shown in Figure 1. DC power supplies, placed in series with ballast resistors, were employed to sustain the discharge; Del High Voltage RHVS (10 kV, 1.5 A) was used at Stanford and Del High Voltage RHVS (60 kV, 5 mA) in Paris. The ballast resistor (typically 3-500 kΩ) was used to stabilize the discharge. 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 and voltage signals were processed by digitizing oscilloscopes; 400 MHz Tektronix DSA 602 in Paris and 250 MHz HP 54510A at Stanford.

The discharges can be obtained using electrodes of various materials and shapes. We used rhodium points opposite to copper planes in the original study in Paris, and two platinum pins welded on water-cooled stainless steel tubes at Stanford. Electrodes with small radius of curvature (pins) permit easier discharge ignition. In the recent Stanford experiments we tested cathodes made of thermionic materials: lanthanum chromite ($LaCrO_3$), lanthanum hexaboride (LaB_6) and molybdenum (Mo). Typically, ambient air was flown through the discharges axially with various flow velocities. We also tested swirl air flow injection into the discharges placed in glass or quartz tubes of various diameters.

Spatially resolved optical emission spectroscopy and electrical diagnostics were employed to characterize the discharge properties. A schematic of the two spectroscopic systems used at Stanford is also shown in Figure 1. An Ocean Optics S2000 dual channel spectrometer, fitted with two grating/CCD combinations, provided 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 2000x800 pixel CCD camera SPEX TE2000 (15x15 µm pixel dimension) provided spectral resolution of 0.12 nm, sufficient to resolve the rotational structure of molecular spectra. This second system also enabled to take wavelength-specific CCD camera images of the discharges, which were useful for measuring their diameters. Both spectroscopic systems had two-dimensional scanning capability with a spatial resolution of 250 µm. Absolute intensity calibrations were obtained by means of radiance standards traceable to NIST calibrations. A similar optical diagnostics set-up was previously 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 at Stanford for photo-documentation of discharges.

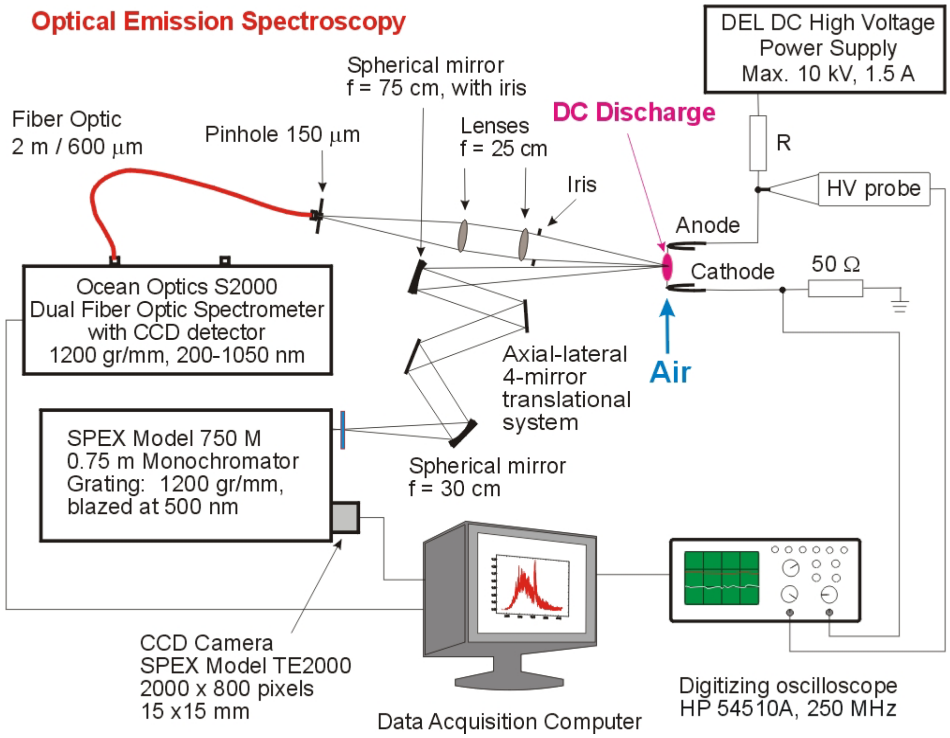


Figure 1. Experimental setup for optical emission spectroscopy and electrical measurements of the DC discharges.

RESULTS AND DISCUSSION

The DC atmospheric pressure air discharges under study are stable continuous discharge types without pulses. They operate with DC currents from 1.6 to several hundreds of mA, and DC voltages from a few kV to a few hundred V. The gap length (interelectrode distance) can be varied from 1 mm to a few cm. The voltage-current characteristic of the discharges is descending, as shown in Figure 2. In contrast, the discharge power increases with the current.

The discharges are ignited by a streamer-to-spark transition, but the ballast resistor immediately limits the spark current. The ballast resistor is chosen 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 3 shows photographs of DC 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 spectra emitted by the DC air discharges in the UV-VIS region for various discharge parameters. The N_2 2nd positive ($C^3\Pi_u$-$B^3\Pi_g$) and NO γ ($A^2\Sigma^+$-$X^2\Pi_r$) spectral systems were 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 was found to vary between 1500 and 2500 K, depending on the current and the gas flow velocity and slightly varying along the discharge axis. Measured T_v was higher (around 4000 K) and therefore suggests that the plasma is in a state of thermal nonequilibrium. At low flow velocities of ambient air through the discharge, T_g increased and T_v decreased with increasing current, as shown in Figure 4. This trend indicates that with increasing current, the plasma conditions smoothly shift from a distinct thermal nonequilibrium toward equilibrium. Such a result agrees with the modeling of an atmospheric air DC glow discharge by Benilov and Naidis who associate the transfer from nonequilibrium to LTE plasma with the change of the dominant ionization mechanism from electron-impact to associative ionization that occurs at currents of several tens of mA (at no gas flow) [4].

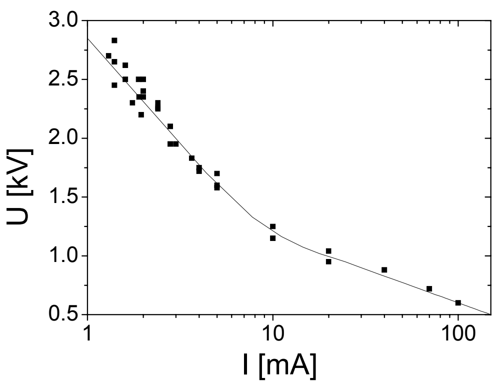
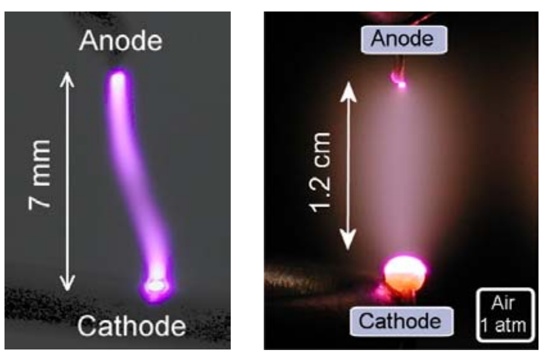


Figure 2. Voltage-current characteristic of the DC discharges in ambient air. Interelectrode distance d = 7 mm, various electrode materials, point-to-plane or pin-to-pin configurations.



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

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

Figure 3. DC discharges in ambient air flow (v = 0.2 m/s) at atmospheric pressure. Platinum pin-to-pin configurations, different exposure times were used.

When the gas flow velocity was increased, less power was deposited in the discharge, and thus we measured lower T_g even at high currents. In summary, two external parameters - current and gas flow velocity - control the gas temperature and consequently the "level" of nonequilibrium in the plasma.

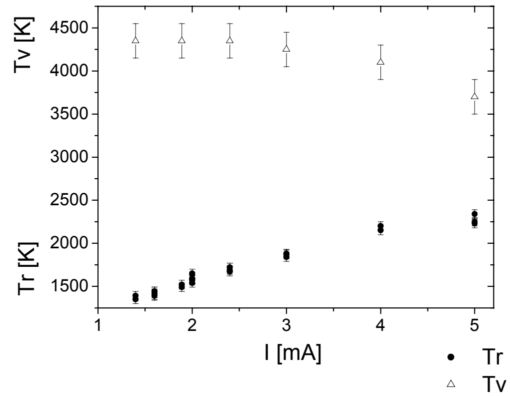


Figure 4. Rotational and vibrational temperatures as functions of the discharge current in ambient air. Flow velocity v ≈ 0.2 m/s, interelectrode distance d = 7 mm, positive point-to-plane configuration.

We measured the radial emission intensity profiles at various wavelengths to determine the diameter of the discharge column. The discharge diameter was 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) enabled us to estimate the plasma conductivity, hence the electron number density n_e, using Ohm's law [2,5]. The discharge diameters were measured from 0.4 to 3.3 mm, depending on the current, the gas flow conditions, and the gas temperature. The corresponding current densities j were between 0.1 and 10 A/cm^2 and the estimated electron densities n_e were in the range of 10^{12}-10^{13} cm^{-3}. The measured current densities are lower than the typical current densities of arcs. The high values of n_e at gas temperatures between 1500 and 2500 K confirm the thermal nonequilibrium, since the 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.

Optical emission diagnostics also provide the spatial distributions of the emission intensity along the discharge axis. At low currents, the axial emission intensity profiles measured at 337 and 358 nm (band heads of N_2 2nd positive system) indicate the stratification into dark and bright layers, typical of low pressure glow discharges [5]. 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. The positive column occupies most of the interelectrode space.

Measurements of the floating plasma potential along the discharge axis by a platinum pin probe enabled us to calculate the electric field strength E with better than 10 % uncertainty. The measured electric field was approximately uniform in the positive column, with values decreasing from 3000 to 200 V/cm as the current increased, thereby leading to a falling voltage-current characteristic (Figure 2). For discharge currents below about 100 mA, the voltage drop across the very thin region adjacent to the cathode was 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 [5]. The observed thermal nonequilibrium, the relatively low current densities, and the cathode fall values of several hundreds of V indicate that the discharges at these conditions are of the glow type.

With thermionic cathode materials ($LaCrO_3$, LaB_6), we observed that the cathode fall was reduced and became a function of the current. Figure 5 shows the discharge voltage as a function of the interelectrode distance at various currents, using $LaCrO_3$ thermionic cathode in atmospheric air discharges. The values where the lines cross the voltage axis correspond to the cathode fall, since at atmospheric pressure the size of the cathode layer is negligible when compared with the size of the positive column. The cathode fall decreases with the current, unlike in regular glow discharges where it only depends on the cathode material and the gas. This is because the cathode temperature increases with the discharge current, which amplifies the thermionic electron emission.

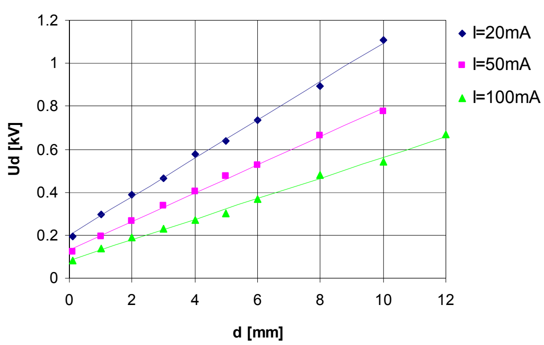


Figure 5. Voltage as a function of interelectrode distance d for various currents of DC discharges with $LaCrO_3$ cathode. The vertical offset on U_d-axis represents the cathode fall.

Thermionic emission typically occurs in arc discharges. Here, we employed thermionic cathodes in discharges that cannot be yet classified as arcs due to thermal nonequilibrium and relatively low current densities. They cannot be considered as glow discharges either, because the cathode fall is too low. These discharges operate in the transition region between glow and arc.

In the most recent series of experiments performed at Stanford we tested swirl flow injection of ambient air into the discharges. With this approach, we obtained stable discharges with gap lengths up to 10 cm or more, which is interesting for plasma volume scaling. The swirling flow in the tube confined the discharges, and thus the current densities increased up to 10-40 A/cm^2 and the electron densities up to 10^{13}-10^{14} cm^{-3}. This represents an enhancement of n_e by 1-2 orders of magnitude with respect to the typical discharges in open air. The electric field was low (200-600 V/cm) in these swirl-stabilized discharges, and T_g often reached 3000 K. At these conditions, the discharges operate at the transition between glow and arc. Nevertheless, the plasma is still non-thermal, since the measured n_e is about 3-4 orders of magnitude higher than the equilibrium value of n_e at 3000 K (~10^{10} cm^{-3}).

CONCLUSIONS

We demonstrated a possibility to maintain stable atmospheric pressure DC discharges in ambient air. At low currents (below 100 mA), the discharge properties are those of the glow type, with a distinct cathode layer and a positive column, and the plasma is in a state of thermal nonequilibrium. With a gradually increasing current, the discharge type smoothly transforms from glow to arc, producing non-equilibrium but relatively hot plasma (2500-3000 K). The gas flow conditions also substantially influence the plasma properties. The higher gas flow velocities cool the plasma and intensify the departure from thermal equilibrium. We found that swirl flows stabilize the discharges and cause them to confine, leading to an enhancement of the electron density.

The studied discharges provide a source of highly ionized nonequilibrium air plasma with electron number densities of the order of 10^{12}-10^{14} cm^{-3}, and gas temperatures between 1500 and 3000 K. The easy DC operation and the volume scalability make them attractive for many applications, including pollution control.

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