Generation of atmospheric pressure non-thermal plasma by diffusive and constricted discharges in air and nitrogen at the rest and flow

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Abstract. Main subject of this paper is low current atmospheric pressure gas discharges powering with DC power supplies. These discharges are widely used for generation of non-thermal or non-equilibrium plasma in air and nitrogen which are much cheaper compared to rare gases like He or Ar. Molecular nitrogen as plasma forming gas has a unique capability to accumulate huge energy in vibration, electron (metastables) and dissociated (atomic) states. Besides, all active species have a long life-time, and they can be therefore transported for a long distance away from the place of their generation. Different current modes (diffusive and constricted) of these discharges are discussed. Experimental and numerical results on generation of chemically active species in the diffusive and constricted mode are presented.

1. Introduction

The weekly ionized non-thermal plasma (NTP) is of great interest for many applications because of its strong non-equilibrium state wherein an average electron energy $T_e$ exceeds markedly gas temperature $T_g$, i.e. electrons in the NTP are strongly overheated compared to neutral gas. Energetic electrons due to frequent collisions with the neutrals excite and dissociate effectively atoms and molecules of the plasma-forming gas that results in a creation of physically-, and bio-chemically active gaseous medium in a practically cold background gas.

The NTP at atmospheric pressure is of considerable interest for practice. A reason is that sustaining the NTP at atmospheric pressure at first allows us to avoid the use of expensive vacuum equipment and second gives opportunity to use the NTP for treatment of the exhausted gases and polluted liquids. The second opportunity cannot be realized at all with use of the NTP at low pressure. Another advantage of atmospheric pressure NTP compared to plasma at low pressure is its ability to intensify plasma treatment that is well suited for fast activation of the subjects to be treated.

At present there are many kinds of plasma sources working at low and atmospheric pressure and using MW, RF, low frequency, pulsed and DC power supplies for NTP generation [1-10]. The subject of this paper is low current atmospheric pressure gas discharges powering with DC power supplies. These discharges are widely used for generation of non-thermal or non-equilibrium plasma in air and nitrogen which are much cheaper compared to rare gases like He or Ar. Molecular nitrogen as plasma forming gas has a unique capability to accumulate huge energy in vibration, electron (metastables) and
dissociated (atomic) states. Besides, all N₂-active species have a long life-time, and they can be therefore transported for a long distance away from the place of their generation.

Low current DC discharges at atmospheric pressure exhibit different current modes (diffusive and constricted). Physical reasons that are responsible for sustention of diffusive and constricted mode of these discharges are discussed. Experimental and numerical results on generation of chemically active species in the diffusive and constricted mode are presented.

2. The most important parameters of atmospheric pressure NTP

The NTP at atmospheric pressure is very complicated subject and characterized by numerous parameters. We would like to point to the most important of them. The measurement and/or calculation of these parameters is to be main concern for the experiment and numerical modeling the NTP.

The most important parameter is the reduced electric field strength \( E/N \) which determines a value of the average electron energy \( T_e \) (\( E \) is the electric field strength, \( N \) is the gas density). Besides the \( E/N \) ratio influences the electron energy distribution function (EEDF) which governs a distribution of the energy released by NTP in between different channels like gas heating, vibration and electronic state excitation, dissociation, ionization, etc.

Other important parameters characterizing the NTP are the number densities of the charged particles: electrons \( N_e \), negative ions \( N_n \) and positive ions \( N_p \). These densities vary over a wide range (\( 10^9 \) – \( 10^{13} \) cm\(^{-3}\)) and determine the intensity of electron and ion collisions providing a transfer of electrical energy to the NTP.

The ratio between electron temperature \( T_e \) of plasma and translation \( T_g \) and vibration \( T_v \) temperatures of the process gas shows the degree of non-equilibricity of the NTP. As a rule, \( T_g \ll T_v \ll T_e \). In such a case, energetic electrons generate intensively numerous bio-chemical active species. High magnitude of \( T_v \) provides effectively the promotion for non-equilibrium plasma-chemical reactions under cold ambient. A magnitude of \( T_g \) determines NTP-applicability for treatment of thermal-sensitive materials.

Bio-chemical reactivity of the NTP is determined by sort and number density of active species generated by the NTP. Because of this, solid data on number densities of the radicals and metastables like O, OH, CH, He*, N₂*, etc formed by particular NTP are needed, and they allow us to judge the applicability and efficiency of this plasma to the solution of any specific problem.

3. On problems referred to sustaining the diffusive atmospheric pressure NTP

The atmospheric pressure NTP can exist in two forms: diffusive (or glow) and constricted. As a rule, diffusive NTP is characterized by lower gas temperature and current density than those in the constricted NTP. Gas discharge with diffusive NTP has a narrow current region for its existence. Development of ionization instability in the diffusive NTP results in its transition to the constricted mode. Let us point to several physical reasons provoking the constriction of diffusive discharge.

In the absence of vortex electric fields, each gas discharge can be roughly divided into three main parts: two near-electrode layers and plasma column. In the course of discharge forming, both cathode and anode layers are going to the states characterized by so-called normal current densities. Cathode/anode normal current density grows drastically (quadratic) with an increase in gas pressure and equals to 100-200 A/cm\(^2\) for discharge in N\(_2\) and air at atmospheric pressure (note a diffusive plasma column is characterized by much lower current density). A tendency of near-electrode layers to come to the states with high current density leads to formation of the cathode/anode current spots which provoke a constriction of diffusive plasma column.

Normal current density in He is lower than that in molecular gases by a factor 100. Due to this, time \( \tau \) needed for normal near-electrode layer formation is much longer compared to that in the most of molecular gases. So, formation of normal spots can be avoid, if period \( T \) of the alternating applied voltage \( T<<\tau \). This condition can be satisfied in DBD with He (or mixture with He) that enables us to generate a diffusive (or homogeneous) NTP at atmospheric pressure [11, 12]. Diffusive DBD can be
obtained also in N₂ at atmospheric pressure but within very restricted region of the experimental parameters [13].

4. Influence of an electrode geometry and gas flow on a discharge stability

Steady-state normal cathode layer is characterized by strong electric field. It is therefore difficult to destroy cathode spot or diminish its current density by any external action. To extend/spread DC gas discharge in transverse to the current direction, cathode is fabricated as sectioned, i.e. the cathode surface has been broken mechanically into numerous small pieces each of them can be ballasted (or not) by individual resistor. In the sectioned discharge, there are many small cathode spots regularly dispersed over the cathode surface. Fortunately, small cathode spots do not provoke a constriction of plasma column. The reason is that the Dark Faraday Space which shields diffusive plasma column from cathode layer with high electric field.

Figure 1a demonstrates the sketch of the electrode system with the sectioned cathode. Every cathode element is ballasted by a resistor of 1MOhm. This system works in atmospheric pressure gas flow and generates diffusive (glow) discharge (Fig.1b). Installation can be easily scale up and enables us to generate diffusive plasma inside big volume. For details see [14-17].

![Fig.1. a) Sketch of the electrode system with the sectioned cathode; b) Image (fragment) of DC diffusive (glow) discharge with the sectioned cathode (below) in airflow at atmospheric pressure. The photo is taken by camera in upwind direction; c) Image of DC gas discharge chamber with the sectioned cathode. Lilac color inside discharge chamber corresponds to the diffusive plasma. Gas flow rate of this installation is 300 m³/hour.](image)

Steady-state normal anode layer is characterized by much lower electric field compared to that in cathode layer. However anode spot is much more dangerous for stability of a diffusive plasma column because the anode spot actively initiates plasma column constriction. Fortunately, owing to low electric field, normal anode spot can be easily destroyed, i.e. transformed into anode layer with low current density which equals close to that in diffusive plasma column. One can possibly avoid a normal anode spot formation by two approaches: with changing the shape of anode and blowing the anode. This idea is illustrated with figure 2.
Fig.2. a) Image of the electrode geometry providing the stabilization of both the anode layer against its transition to normal anode spot and the diffusive plasma column against its constriction (from [18]). Cathode (upper) and anode (bottom) are fabricated with several sharpened spikes and spherical crater. b) Side view of steady-state glow discharge with diffusive NTP in airflow at atmospheric pressure, I=1mA.

5. NTP created by constricted discharges at atmospheric pressure

Diffusive state of the NTP at atmospheric pressure is unstable in regards to development of different ionization instabilities. In the most cases atmospheric pressure NTP exists therefore in a constricted form like filaments or microdischarges (MDs).

Advantages of the constricted NTP: constricted discharges at atmospheric pressure can be realized much easier compared to high pressure diffusive discharges; the constricted NTP is also non-equilibrium plasma and can generate effectively and abundantly different sorts of active species.

Disadvantages of the constricted NTP: filamentation (constriction) of a discharge leads to the reduction of a gas volume activated by NTP; mixing active species of the constricted NTP with non-activated surrounding gas diminishes non-equilibricity of the afterglow; MDs provide intensive local energy deposition that can result in non-homogeneous surface treatment and even cause a local damage in the case of thin films.

5.1. Constricted (filamentary) discharges in transverse gas flow

Filamentary discharge in transverse gas flow is always non-stationary and exists only as pulsing self-repeated discharge. In general, this discharge is similar to well-known gliding arc between two divergent electrodes but not the same to that. The key features of filamentary discharge in transverse gas flow can be illustrated by example of the constricted discharge in ambient airflow between needle and knife (Fig.3). Gas velocity is 50 m/s. Average current is 2.5 mA.

Figs. 3a) and 3b) show front (in upwind direction) and side view of DC constricted (filamentary) discharge in transverse airflow. With the naked eye this discharge looks more or less homogeneous as it seen in Fig.3b. In fact, the constricted discharge exists in the pulsing self-repeated regime the period of which includes a formation of filament, its blowing and decay. Repetition frequency of the current pulses is not constant but varies within 1-10 kHz. A set of the side view shots (#1-4) elucidates a spatial-time behavior of the self-repeated filamentary discharge: #1 – gas gap breakdown or formation of the filament; #2-3 – extension of the filament due to its blowing from discharge gap; #4 – break and decay of the filament. Exposure time of each shot is 5 µs. Time interval between shots #1 and #4 equals to 150 µs.
Fig. 3. a) and b): Front (in upwind direction) and side view (flow from left to right) with the naked eye of DC constricted (filamentary) discharge in transverse airflow. Electrodes are needle (upper) and knife (bottom). Exposure time is 0.1 s. Gas velocity is 50 m/s. Average current is 2.5 mA. Constricted discharge exists in the pulsing self-repeated regime. Repetition frequency of the current pulses is not constant but varies within 1-10 kHz. A set of the side view shots (#1-4) elucidates a spatial-time behavior of self-repeated filamentary discharge: #1 – gas gap breakdown or formation of the filament; #2-3 – extension of the filament due to its blowing from discharge gap; #4 – break and decay of the filament. Exposure time of each shot is 5 µs. Time interval between shots #1 and #4 equals to 150 µs.

Fig. 4 presents experimental evidences of non-equilibricity and high efficiency of the constricted NTP generated by pulsing self-repeated DC discharge in transverse airflow. Parameters of discharge are the same as those in Fig. 3. The coordinate “zero” in the horizontal axis corresponds to the outlet of the gas discharge gap. Spectroscopic measurements allow us to prove the generation of such active species as electronic excited nitrogen molecules N\textsubscript{2}(C), oxygen atoms O and radicals OH. Besides, one can see also a big difference between vibration and translation temperatures.

However spatial behavior of the species formed by high-energetic electrons is opposite to that for vibration temperature determined by collisions of low-energetic electrons with N\textsubscript{2}-molecules. The reason is that increase in the filament length due to its blowing results in diminishing the reduced electric field inside the constricted plasma. These changes in the E/N value influence strongly the EEDF high-energetic “tail” that is responsible for electronic excitation, dissociation and ionization but not so much the EEDF low-energetic part (the EEDF “core”) responsible for vibration excitation.
Fig. 4. a) Averaged in time and normalized spatial distribution of the excited N₂(C), radical OH and atom O in and outside the gap for pulsing self-repeated filamentary discharge in transverse airflow. b) Averaged in time spatial distribution of the vibration and rotation temperatures in and outside the gap. Experimental parameters of the constricted discharge are the same as those in Fig. 3. The coordinate “zero” in the horizontal axis corresponds to the outlet of the gas discharge gap.

5.2. **Constricted (filamentary) discharges in longitudinal gas flow**

Longitudinal discharge was activated inside a glass tube blown out with N₂ flow at atmospheric pressure [19]. The diameter of glass tube and inter-electrode gap were the subjects to variation from 5 to 20 and 10 to 40 mm, respectively. Images of short constricted discharge and its long and wide afterglow are shown in Fig. 5a. Volt-Ampere Characteristics of this discharge is presented in Fig. 5b.

Fig. 5. a): Images of DC steady-state constricted discharge (upper) and its afterglow in longitudinal N₂ flow at atmospheric pressure. Discharge is activated in glass tube with inner diameter of 5 mm. Inter-electrode distance is 15 mm. Radius of the constricted plasma column is about 1 mm. b) Averaged in time Volt-Ampere Characteristics of the constricted DC discharge. Gas flow rate is variable parameter: 1 – 320 cm³/s; 2 - 190 cm³/s; 3 - 0 cm³/s (gas at the rest). Purity of N₂ is 99.999%.
Figure 6a shows a dependence of rotational Trot (or Tgas) and vibrationTvib temperatures inside the constricted discharge in longitudinal N\textsubscript{2} flow vs average discharge current under different gas flow rate of N\textsubscript{2}. For comparison, the temperatures T\textsubscript{r} and T\textsubscript{v} measured in the CGD at rest gas are shown as well. One can see, both temperatures depend on the current, however their dependences are not similar to each other: rotation temperature T\textsubscript{r} grows monotonically with an increase in current but behavior of the vibration temperature T\textsubscript{v} is non-monotonic having maximum about 6000K at the current I\approx 7-8mA. Maximum deviation from thermodynamic equilibrium in the NTP (T\textsubscript{v}>>T\textsubscript{r}) there is in the current region 2mA<I<20mA. With increase in current both temperatures, T\textsubscript{r} and T\textsubscript{v}, tend to approach each to other. It corresponds to gradual transition of the constricted discharge to the state with thermodynamic equilibrium like to that in arc discharge.

Figure 6b presents the distribution of T\textsubscript{gas} and T\textsubscript{vib} along N\textsubscript{2} afterglow formed by plasma jet ejected from the tube with the constricted discharge. Cross-section of afterglow is bigger compared to that of the constricted discharge. Both the expansion of plasma jet on outlet the tube and mixing with ambient non-activated gas result in diminishing of temperatures T\textsubscript{gas} and T\textsubscript{vib} that is seen in Fig.6b.

Optical spectra of the constricted N\textsubscript{2} flow discharge and its afterglow are presented in Fig.7 One can see (Fig.7a) the emission of the first positive system 1\textsuperscript{+} (N\textsubscript{2}(B\textsuperscript{3}Π\textsuperscript{'}→A\textsuperscript{3}Σ)) second positive system 2\textsuperscript{+} (N\textsubscript{2}(C\textsuperscript{3}Π\textsuperscript{'}→B\textsuperscript{3}Π\textsuperscript{'})) and first negative system 1\textsuperscript{−} (N\textsubscript{2}+(B\textsuperscript{2}Σ\textsuperscript{+}→X\textsuperscript{2}Σ\textsuperscript{−})) give the most input in total emission from the constricted plasma column. Besides there are weak bands of CN radical (CN(B\textsuperscript{2}Σ\textsuperscript{−}→X\textsuperscript{2}Σ\textsuperscript{−})). An existence of CN emission from the constricted discharge is attribute to trace level impurities containing inevitably in nitrogen of high purity (99.999%).

Spectrum of emission from afterglow is distinct from that of the constricted plasma – with the naked eye the constricted plasma column looks as red colour but afterglow has purple or lilaceous colour. The emission from afterglow does not contain spectral lines of nitrogen but there is intensive emission of CN, NO and NH. These particles are formed due to secondary reactions of N\textsubscript{2} active species with trace admixtures of ppm-level like O\textsubscript{2}, H\textsubscript{2}O, C\textsubscript{x}H\textsubscript{y}.

Note the rate of primary active species transformation in reactions with the impurities increases with an increase in pressure of “pure” gas: the higher gas pressure, the stronger influence of the impurities on the composition of active species in afterglow and in the discharge as well.
6. Numerical calculations on the composition of reactive species in the NTP

Vast diversity of the collision processes forming the NTP provides, from one side, a variety of plasma properties, but from other side, the complexity in modeling the plasma itself and plasma chemistry as well. The present state in both the designing/invention of plasma sources and NTP-processing is characterized, as a rule, by a “trial and error” strategy. Besides, some of the important plasma parameters not always can be determined by experimentally. In such a case, important role belongs to numerical modelling of both the reactive species generation by NTP and their behaviour in afterglow. Numerical simulation of the diffusive NTP in airflow was done in [15, 16]. Here main attention will be paid to simulation of the constricted NTP.

Structure of the program module that is normally used for simulation of the NTP is presented in Fig.8. In general, such module includes the blocks with electron scattering cross-sections data, plasma chemistry kinetic data and several solvers: Boltzmann equation solver, plasma chemistry solver and gas dynamic and electric circuit solver. The input for calculations are experimental conditions such as gas mixture, temperature and pressure, gas flow rate and velocity, electrode and chamber geometry, voltage amplitude, frequency, parameters of electric circuit, etc. The output of simulations is a composition of the NTP and reactive species.

![Fig.8. Structure of program module that is used normally for simulation of the NTP.](image-url)
6.1. Modeling of the constricted NTP in transverse airflow at atmospheric pressure

The processes taken into account in the simulation of the NTP in airflow are listed below. The results obtained are presented in Fig. 9.

| Chemical Reaction                                      | Chemical Reaction                                      |
|--------------------------------------------------------|--------------------------------------------------------|
| \( O_2 + e \rightarrow O_2(\text{vibr}) + e \)        | \( H_2O + e \rightarrow H_2O(\text{rot}) + e \)       |
| \( O_2 + e \rightarrow O_2(0.98) + e \)               | \( H_2O + e \rightarrow H_2O(\text{vibr1}) + e \)    |
| \( O_2 + e \rightarrow O_2(1.63) + e \)               | \( H_2O + e \rightarrow H_2O(\text{vibr2}) + e \)    |
| \( O_2 + e \rightarrow O + O \)                        | \( H_2O + e \rightarrow H + OH + e \)                 |
| \( O_2 + e \rightarrow O_2(4.5) + e \)                | \( H_2O + e \rightarrow H_2O^+ + e + e \)             |
| \( O_2 + e \rightarrow O + O + e \)                    | \( H_2O + e \rightarrow OH + H \)                     |
| \( O_2 + e \rightarrow O + O(\text{D}) + e \)         | \( H_2O + e \rightarrow H_2 + O \)                    |
| \( O_2 + e \rightarrow O_2^+ + e + e \)               | \( N_2(A3) + H_2O \rightarrow N_2 + H + OH \)         |
| \( O_2 + e \rightarrow O + O^+ + e + e \)              | \( N_2(A3) + O_2 \rightarrow N_2 + O + O \)           |
| \( O_2+e+(O_2)\rightarrow O_2^- + (O_2) \)            | \( N_2(A3) + O_2 \rightarrow N_2O + O \)              |
| \( N_2 + e \rightarrow N_2(\text{vibr}) + e \)        | \( N_2(A3) + N_2O \rightarrow N_2 + N_2 + O \)        |
| \( N_2 + e \rightarrow N_2(\text{E}=1) + e \)         | \( N_2(A3) + NO_2 \rightarrow NO + O + N_2 \)         |
| \( N_2 + e \rightarrow N_2(\text{esum}) + e \)        | \( N + NO \rightarrow N_2 + O \)                      |
| \( N_2 + e \rightarrow N + N + e \)                    | \( N + NO_2 \rightarrow N_2O + O \)                   |
| \( N_2 + e \rightarrow N_2^+ + e + e \)                | \( N + NO_2 \rightarrow NO + NO \)                    |
| \( H_2O + e \rightarrow H_2O(\text{rot}) + e \)       | \( N + O_2 \rightarrow NO + O \)                      |
| \( H_2O + e \rightarrow H_2O(\text{vibr1}) + e \)     | \( NO + O_3 \rightarrow NO_2 + O_2 \)                 |
| \( H_2O + e \rightarrow H_2O(\text{vibr2}) + e \)     | \( O + O_2 + O_2 \rightarrow O_3 + O_2 \)             |
| \( H_2O + e \rightarrow H + OH + e \)                  | \( H_2O + e \rightarrow OH + H \)                     |
| \( H_2O + e \rightarrow H_2O^+ + e + e \)              | \( H_2O + e \rightarrow H_2 + O \)                    |
Fig. 9. Evolution in time of different chemically active species in the constricted discharge zone and in post discharge region. DC constricted (filamentary) discharge in transverse airflow. Experimental conditions correspond to those in Fig.3.

6.2. 1-D modeling of steady-state constricted N₂ glow discharge at atmospheric pressure

Numerical model takes into account the radial transfer of the particles and heat due to diffusion and heat conduction and includes the set of appropriate balance equations for charged particles: electrons Ne and positive ions, N⁺, N₂⁺, N₃⁺, N₄⁺. The charged particle kinetics considers the ion conversion and several types of ionization processes: direct ionization, stepwise ionization, associative and pooling ionization and electron-ion recombination and ambipolar diffusion as well. Electronically excited states N₂(A), N₂(B), N₂(C), N₂(a'); vibration energy for the ground state of N₂(X,v) and gas heating were included also. The needed rate coefficients were taken from literature and calculated with use of Boltzman equation solver developed by I.Kochetov and A.Napartovich. For details see [20].

In the constricted plasma column, gas temperature changes in a radial direction from high to low magnitude (3500 K at the axis and 520 K at the wall). Under such conditions, the impact of ion conversion on discharge radial structure reveals itself the most obviously: main sort of positive ions is atomic N⁺ ion inside hot plasma, and molecular N₃⁺ ion in cold periphery region of discharge (see Fig.10. a) The radial changing in the ion composition in the constricted longitudinal discharge in N₂ flow. I=40 mA. Gas flow rate is 114cm³/s. b) Ionization balance in hot core of the constricted plasma.
Fig.10). The strongest changing in the ion composition occurs within a thin cylindrical layer at the nearest periphery of plasma column \((0.75 \leq r \leq 1.5 \text{ mm})\) where \(14\%\) of the total current is transferred.

Based on information in Fig.10b one can conclude that ionization balance in hot core of the constricted \(N_2\)-plasma column is non-local because it is controlled by ambipolar diffusion but not recombination. From this it amazingly follows that constricted plasma column in \(N_2\) at atmospheric pressure is similar to diffusive glow discharge at low pressure under Schottky regime!

![Graphs showing ion composition changes](image)

Fig.11. Atoms and molecules (a) and radicals (b) number density distribution along afterglow of \(N_2\)-plasma jet containing impurities: 5 ppm \(\text{CH}_4\), 5 ppm \(\text{O}_2\) and 5 ppm \(\text{H}_2\text{O}\). Time = distance from the nozzle/gas velocity. The reduced energy deposition in the constricted discharge is 600 J/liter.

Figure 11 presents numerical results on composition of non-\(N_2\) active species in \(N_2\)-afterglow due to influence of gaseous impurities of ppm-level. The impurities of such level there are in the balloons with “pure” nitrogen (99.999%) delivered by the manufacturer. Non-\(N_2\) active species with number densities about \(10^{14} \text{ cm}^{-3}\) can dramatically change both the paths of plasma chemical reactions and final results of the NTP treatment.

7. Conclusion
Atmospheric pressure NTP can be generated in two forms: diffusive and constricted. Diffusive NTP can be created by AC discharges (for instance, by DBD) and DC discharges with sectioned electrodes in transverse gas flow. Constricted NTP can be generated by AC and DC discharges in transverse and longitudinal gas flow.

In the case of transverse gas flow the constricted NTP is non-stationary (discharge is pulsing and self-repeated), in the most cases of longitudinal gas flow the constricted NTP is steady-state. As a rule, constricted NTP characterized by higher gas temperature nevertheless strong non-equilibricity of plasma can be kept.

In “pure” gases from balloons always there are trace impurities. Plasma chemical reactions of these impurities with primary active species generated by NTP change drastically the composition of active species. This effect has to be taken into account in the use of atmospheric pressure plasma treatment and interpretation of the final results.

Acknowledgement.
Authors thank to the RFBR for partial support of this work (grant No 08-02-00601a).
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