

The optical emission spectroscopy of pulsed and pulse-periodic discharges initiated with runaway electrons

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Abstract. We report on the results of measurements of an electron T_e and a gas T_g temperatures as well as a reduced electric field strength E/N in the plasma of a high-voltage nanosecond discharge initiated with runaway electrons in a gap with a strongly nonuniform electric field distribution. The foregoing plasma parameters were determined with optical emission spectroscopy techniques. The possibility of using the method for determining T_e and E/N in thermodynamically nonequilibrium plasma, which is based on a determination of a ratio of a peak intensities of the ionic ($\lambda = 391.4$ nm) and molecular N_2 ($\lambda = 394$ nm) nitrogen bands, is proved. To measure a gas temperature the optical emission spectroscopy technique based on the measurement of a relative radiation intensity of rotation structure of electronic-vibrational molecular transitions was used, as well.

1. Introduction

Currently a lot of attention is given to study diffuse pulse and pulse-periodic discharges in dense gases preionized with runaway electrons (REP DD) under conditions of an inhomogeneous electric field distribution [1-5]. The main feature of such discharges is a generation in a gap of runaway electrons and X-ray, affected on a breakdown. These processes was shown to provide the formation of diffuse discharges at the excitation of gas medium by high-voltage nanosecond pulses of both polarities.

Determination of the main parameters of REP DD plasma is of both scientific and practical interest. The knowledge of the density N_e and electron temperature T_e , gas temperature T_g , and reduced electric field strength E/N , where E is the electric field strength and N is the particle density, is important for general plasma characterization and verification of theoretical models of the REP DD plasma. Reliable data on these parameters can be used for calculating the rates of different plasma chemical processes and estimating the amplitude-time, spectral, and energy characteristics of the REP DD plasma light emission.

In practice, diffuse discharges at atmospheric gas pressure can be used for generation of chemically active plasma. Such plasma is in demand, for example, in surface modification of solids, surface cleaning and sterilization, decomposition of organic compounds, and waste processing. The data on the plasma parameters are also important for developing other modern technologies based on homo- and heterophase plasma chemical processes.

At present, it is known about several publications concerning the determination of REP DD plasma parameters [6-10]. The results of measurements of N_e in the cathode spot by the method of Stark



broadening of spectral lines were reported in [6]; this method was used in [7, 8] for measuring the values of Ne in the plasma of the diffusive discharge in helium, as well. It should be noted that the values of Ne were measured in [7] in various regions of the discharge volume, the dependence of Ne on a gas pressure and its dynamics were determined. In addition, the value of Te in the plasma of the atmospheric pressure discharge was estimated. The method of Stark broadening of spectral lines was used to Ne measurement in hydrogen [9] and nitrogen [10].

It should be noted that the REP DD plasma is thermodynamically nonequilibrium. This excludes the possibility of using the methods for Te measurement, which are applicable where local thermal equilibrium or partial local thermal equilibrium conditions are fulfilled [11]. In nitrogen and nitrogen mixtures, in particular in air, Te and reduced electric field strength E/N in the REP DD plasma can be determined by means of the procedure based on the radiative-collisional plasma model [12, 13]. Its physical substantiation is that the ratio $R_{391/394}$ of peak radiation intensities of the N_2^+ ion (transition $B^2\Sigma_g^+ (v=0) \rightarrow X^2\Sigma_g^+ (v=0)$, $\lambda = 391.4$ nm) and N_2 molecule (transition $C^3\Pi_u (v=2) \rightarrow B^3\Pi_g (v=5)$,

$\lambda = 394.3$ nm) bands is a function of electron temperature $R_{391/394} = f(Te)$ or reduced electric field strength $R_{391/394} = f(E/N)$. The values of Te , E/N in the discharge plasma can be determined from the ratio of peak radiation intensities in bands of the first negative (1^-) ($B^2\Sigma_g^+ \rightarrow X^2\Sigma_g^+$) and second positive (2^+) ($C^3\Pi_u \rightarrow B^3\Pi_g$) nitrogen systems. The procedure is applicable providing fulfillment of the following conditions:

- (I) Maxwellian electron velocity distribution;
- (II) Excitation of the upper states $N_2^+ B^2\Sigma_g^+$ and $N_2 C^3\Pi_u$ is mainly by direct electron impact from the ground state of nitrogen molecule.

The optical emission spectroscopy technique can be used to measure the gas temperature Tg in plasma of REP DD in nitrogen, as well. It is based on a measurement of a relative radiation intensity of rotation structure of electronic-vibrational molecular transitions [11, 14].

This study aims at determining the values of Te , Tg and E/N in the plasma of REP DD operated in atmospheric-pressure nitrogen in a pulsed and pulse-periodic (2 kHz) modes.

2. Experimental setup and measurement

In the experiments, two setups were used; their block diagrams are shown in figure 1. Both setups comprised a system for pump-out and supply of gases. On setup No. 1 (figure 1a), the integral values of plasma parameters within a pulse were measured. The peaks radiation intensities of the N_2^+ ion (transition $B^2\Sigma_g^+ (v=0) \rightarrow X^2\Sigma_g^+ (v=0)$, $\lambda = 391.4$ nm) and N_2 molecule (transition $C^3\Pi_u (v=2) \rightarrow B^3\Pi_g (v=5)$, $\lambda = 394.3$ nm) bands were recorded with a PI-MAX 2 CCD camera (Princeton Instruments) installed at the exit of a MDR-23 monochromator. In the plane of its rectangular entrance slit, an image of the discharge plasma was formed with a lens, while the monochromator exit slit was demounted. On the matrix of the CCD camera in the exit slit plane, an image of the entrance slit was formed in monochromatic components. One of the directions (along the matrix row) coincided with the dispersion direction. The second direction (vertical along the column) coincided with the direction of the entrance slit, which made possible determination of peaks radiation intensities in sites corresponding to different zones along the vertical line. All the forgoing along with possibility to horizontally shift the image of the discharge plasma with respect to the entrance slit allowed spatial-resolved measurements of plasma parameters in two directions.

The photodetector used on setup No. 2 (figure 1b) was an EMI-9781B photomultiplier mounted on a VM-502 monochromator (Acton Research Corp.). The time behaviour of the peaks wavelength intensity of the N_2^+ ion (transition $B^2\Sigma_g^+ (v=0) \rightarrow X^2\Sigma_g^+ (v=0)$, $\lambda = 391.4$ nm) and N_2 molecule (transition $C^3\Pi_u (v=2) \rightarrow B^3\Pi_g (v=5)$, $\lambda = 394.3$ nm) bands were recorded by the EMI-9781B photomultiplier. An overview radiation spectrum of the plasma was recorded using an EPP2000C-25 spectrometer (StellarNet Inc.) with known spectral sensitivity in relative units in the range 200–850

nm. This ensured that the spectral bands required for calculations was present in the radiation spectrum and was not overlapped by other spectral components.

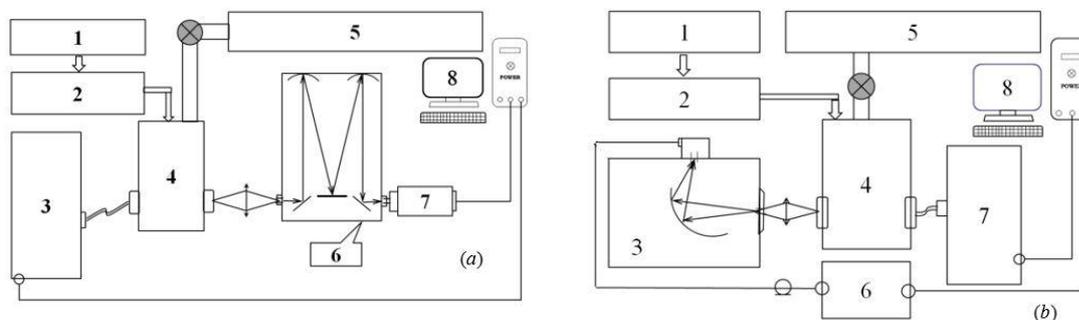


Figure 1. Blok-scheme of experimental setup: (a) - for time-integrated measurements: 1 - master oscillator BNC, 2 – pulser, 3 – spectrometer EPP2000C-25 (StellarNet Inc.), 4 – discharge chamber, 5 – pumping system, 6 – monochromator MDR-23, 7 – CCD-camera PI MAX 2, 8 – PC. (b) - for time-resolved measurements: 1 - master oscillator BNC, 2 – pulser, 3 – monochromator VM-502, 4 – discharge chamber, 5 – pumping system, 6 – oscilloscope TDS-3054B, 7 – spectrometer EPP2000C-25 (StellarNet Inc.), 8 – PC.

For each of the setups, the instrument function and normal slit width were determined using a low-pressure mercury lamp. For setup No. 1 comprising the MDR-23 monochromator with a diffraction grating (2400 grooves/mm), the instrument function was $\sim 0.24 \text{ \AA}$ at a normal slit width of $\sim 10 \text{ \mu m}$. For setup No. 2, the instrument function and normal slit width were $\sim 3.0 \text{ \AA}$ and $\sim 30 \text{ \mu m}$, respectively.

The high-voltage pulsers RADAN-220 and FPG-60 were used for REP DD excitation in pulsed and pulse-periodic modes, respectively. In the idle running regime, RADAN-220 formed voltage pulses in the discharge gap with an amplitude up to $\sim 250 \text{ kV}$ and the half-amplitude duration of $\sim 2 \text{ ns}$ for a matched load with a front duration of $\sim 0.5 \text{ ns}$ in the transmission line. High-voltage pulser FPG-60 formed voltage pulses with an amplitude on a high resistance load up to 60 kV and voltage pulse rise time of $2\div 3 \text{ ns}$ at a pulse repetition rate up to 2 kHz .

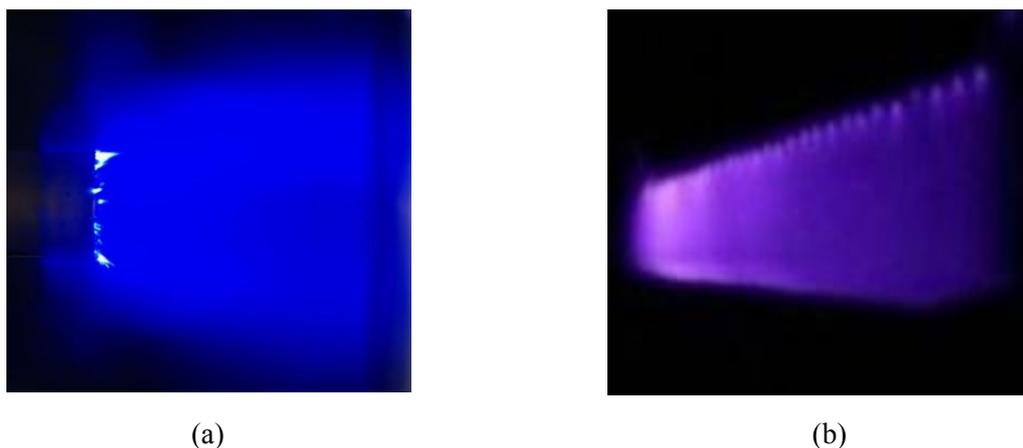


Figure 2. REP DD glowing in nitrogen at pressure of 1 atm at pulsed (a) and pulse-periodic (b) modes.

At the pulsed mode, the electrodes of discharge chamber were a cathode of small curvature radius and a plane anode. The cathode was made of steel foil of thickness $\sim 100 \text{ \mu m}$ in the form of a tube of diameter $\sim 6 \text{ mm}$. The interelectrode gap was $\sim 12 \text{ mm}$. The radiation was extracted through side windows of the discharge chamber. At the pulse-periodic mode, the discharge gap consisted of

electrodes with length of ~ 40 cm. The typical photos of glowing of REP DD plasma in nitrogen at pressure of 1 atm at pulsed and pulse-periodic modes are shown on the figure 2.

3. Experimental results and discussion

To substantiate the possibility of using above-mentioned technique for Te , E/N measurements, it is necessary to prove that the Maxwellian velocity distribution sets in by the instant at which the intensity of nitrogen bands used in this technique is sufficient for recording. In view of the effective energy transfer during the electron–electron collisions, the characteristic time τ of stabilization of the Maxwellian distribution can be estimated as the quantity reciprocal to the electron–electron collisions frequency (1):

$$\tau = \frac{1}{N_e v_e \sigma_{ee}} \quad (1)$$

In (1), N_e is the electron density; V_e is the relative electron velocity; σ_{ee} is the electron–electron collisions cross-section. Estimation showed that at conditions under study, the values of τ are from fractions of nanosecond to several nanoseconds and this suggests that condition (I) is fulfilled. To substantiate that condition (II) is valid for the PER DD plasma in nitrogen, it is necessary to show that the main channel of population of the N_2^+ ($B^2\Sigma_g^+$) and N_2 ($C^3\Pi_u$) states is collision of a nitrogen molecule in the ground state with an electron. It is known that excitation of the resonant $C^3\Pi_u$ state of a nitrogen molecule by direct electron impact prevails over other excitation channels at high electron energies when the reduced electric field strength E/p is close to values optimal for excitation of this state: $E/p \sim 150\text{--}200$ ($V \cdot \text{cm}^{-1} \cdot \text{Torr}^{-1}$), which corresponds to a short phase of voltage drop across the gas discharge gap after its breakdown [15]. The fact that the excitation of the N_2^+ ($B^2\Sigma_g^+$) and N_2 ($C^3\Pi_u$) states by direct electron impact from the ground state of a nitrogen molecule dominates under the experimental conditions is confirmed by calculation and comparison of the excitation rates of the N_2^+ ($B^2\Sigma_g^+$) and N_2 ($C^3\Pi_u$) states due to the direct and stepwise excitation process. The calculation showed that at conditions under study the direct excitation process was markedly higher as compare to the stepwise one.

The measurements of Te and E/N in plasma of REP DD were carried out at nitrogen excitation at pressure of 1 atm both in pulse-periodic (~ 2 kHz) and pulsed modes. In the first case the time average values of Te and E/N was found to be of ~ 2 eV and ~ 270 Td, respectively. As well, dynamics of these parameters was determined. Values of the ratio $R_{391/394}$ monotonically decreased of about two times during ~ 10 ns. It means that values of Te and E/N are reduced by ~ 1.5 times (from 3 to 2 eV for Te , from 400 to 270 Td for E/N).

For discharge excitation in the pulsed mode, the measurements of Te and E/N values were carried out both in time-integrated and time-resolved modes, as well. In the first case, the spectrum of plasma emission radiation in the spectral range of 390–395 nm was registered with a monochromator MDR-23 and CCD-camera PI-MAX 2. The average values of Te and E/N corresponding to this spectrum was found to be of ~ 2 eV and ~ 240 Td, respectively. The dynamic of these parameters was determined by the registration of the time behaviour of N_2^+ ($\lambda = 391.4$ nm) and N_2 ($\lambda = 394.3$ nm) bands' peaks intensity with the monochromator and photomultiplier tube. The values of Te and E/N was obtained to decrease monotonically in the ranges of 3 to 2 eV and of 400 to 260 Td, respectively.

A value of Tg is known to link to a value of rotational temperature Tr due to the relaxation process of inelastic collisions between molecules. The relaxation number, which is the average number of inelastic collision required for the exchange of translational and rotational energy, was shown to be of 4–6 [16]. The mean time of N_2 molecules elastic collisions $\bar{\tau}_{Coll}$ can be calculated according to (2):

$$\bar{\tau}_{Coll} = \frac{1}{[N_2] \cdot \sqrt{2} \cdot \bar{v} \cdot \sigma}, \quad (2)$$

where $[N_2]$, \bar{v} , σ - density, thermal velocity and gas-kinetic cross section of N_2 molecules, respectively. Since the value of $\bar{\tau}_{Coll}$ in atmospheric pressure nitrogen at room temperature is ~ 0.13 ns, the rotational relaxation time is about 0.65 ns. This is more than one order of magnitude shorter

than the duration of REP DD under study. The relation of T_g and T_r of the $C^3\Pi_u$ state is as follows [17]:

$$T_g \approx T_r \cdot \frac{B_e^0}{B_e^*} \approx 1.09 \cdot T_r \quad (3)$$

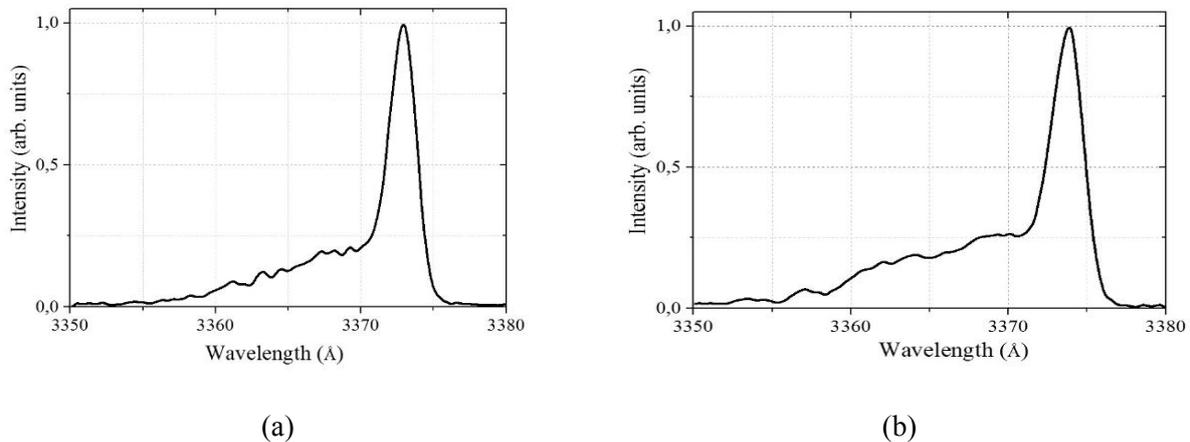


Figure 3. Spectrum of N_2 ($C^3\Pi_u$ ($v=0$) \rightarrow $B^3\Pi_g$ ($v=0$)) transition in atmospheric-pressure nitrogen at pulsed (a) and pulse-periodic (b) excitation modes.

where B_e^0 , B_e^* - rotational constants of $X^1\Sigma_g^+$ and $C^3\Pi_u$ states of N_2 molecule, respectively. The measurements of T_g were carried out both at pulsed and pulse-periodic modes. The typical unresolved rotational emission spectra of N_2 ($C^3\Pi_u$ ($v=0$) \rightarrow $B^3\Pi_g$ ($v=0$)) transition are presented on the figure 3. The value of T_g at pulsed mode was found to be of ~ 380 °K. In the case of pulse-periodic mode, the value of T_g calculated was found to be of 820 °K.

4. Conclusion

Electron T_e , gas T_g temperatures and reduced electric field strength E/N of gas discharge plasma under REP DD excitation of atmospheric-pressure nitrogen at pulsed and pulse-periodic (2 kHz) modes were determined by optical emission spectroscopy techniques. We have performed calculations confirming the applicability of the technique based on the radiative-collisional plasma model [12, 13] to determine T_e and E/N at condition under study.

Acknowledgments

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