GAS TEMPERATURE OF DIFFUSE NEGATIVE CORONA DISCHARGE

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The emission spectra of the second positive system of nitrogen for the diffuse negative corona discharge in ambient air were studied. The rotational structure of spectral lines was analyzed and the spectra were identified. The calculations, using the model of non-rigid rotor was made. The rotational temperature of nitrogen molecules was specified. The dependence of rotational temperature on the applied voltage as well as on the discharge operation mode was shown.

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INTRODUCTION

The widespread use of ozone technologies demands the new cost-effective methods and systems for ozone synthesis. The main issue is improvement of the ozone generator productivity due to reduction of energy consumption for ozone synthesis. Most of ozone generators, which are produced in the world, use a dielectric barrier discharge for ozone synthesis. However, in recent years, much attention was paid to ozone generators which exploit the barrierless gas discharge, in particular, corona gas discharge at atmospheric pressure in the pin-to-plate electrode system [1]. High efficiency of the negative corona discharge for ozone synthesis in air is realized due to low energy consumption [2].

In this paper the spectral characteristics for the diffuse stage of the negative corona discharge in air was investigated.

Negative corona occurs in an electronegative gas with–the pin-to-plate electrode systems when the DC high voltage negative potential is applied to the pin electrode. In air at atmospheric pressure the discharge can operates in self-maintained pulsed mode called Trichel pulse mode [3]. In this mode the discharge current represents a quasi-steady-state sequence of pulses. In the Trichel pulse mode, the optical radiation pulses are observed both in the cathode and anode regions of the short discharge gap [4]. Further increase of the voltage leads to transition from the Trichel pulse mode to a quasi-stationary (diffuse) mode. The direct discharge current and the diffuse glow from the discharge gap are the distinguishing features of the diffuse negative corona discharge.

Stable discharge operation [5] should be provided for correct investigations of the electrodynamic characteristics of the discharge and the spectral characteristics of discharge radiation. To achieve stable discharge operation, both the point and plane electrodes were specially processed before experiments. The criteria for stable discharge operation are the high reproducibility of the Trichel current pulse waveform and the stable pulse repetition rate. These parameters are controlled by the oscilloscope. Also the transition to a spark breakdown should be prevented. To study the spark breakdown transition criteria, the gas temperature in the discharge gap should be measured and monitored, because the spark breakdown mainly depend on the gas temperature [6]. There are different methods to measure the temperature of particles in gas discharge, but the most popular among them are optical diagnostic methods. These methods are widely used as they are non-contact, have no perturbation effects on the object of study and provide high precision measurements. Gas discharge emission spectrometry takes a special place among the optical methods. It allows determining the temperature of gas particles (molecules), as well as observing the temperature dynamics in time. In the current work, the most developed spectrometric method based on measurement of relative intensities in the rotational structure of (0-0)-band emission spectrum of 2^+ nitrogen system ($C_3\Pi_u - B_3\Pi_g$ transitions) was used to measure the gas temperature in discharge. In most practical cases, rotational temperature is similar to the translational temperature (more precision definition of gas temperature requires detailed review of rotational-translational relaxation processes).

1. EXPERIMENT

Experimental study of the emission spectra of the diffuse negative corona within the wavelength range (300-400) nm was carried out using the experimental setup schematically shown in Fig. 1. Spectrometric study of the discharge was carried out using the optical bench based on the monochromator-spectrograph "Solar-Tii" MSDD-1000 with double dispersion. A double diffraction grating with 2400 grooves per 1mm was installed in the monochromator. The reciprocal linear dispersion of the diffraction grating is 0.41 nm/mm. A high-speed photomultiplier tube (PMT) Hamamatsu R9110 with the spectral band of 185…900 nm and signal pulse rise time of $\tau_\Gamma = 2.2$ ns was installed on the output slit of the monochromator. The signal from the photomultiplier was processed by the analog to digital converter (ADC) Velleman PCS 500, which was connected to a computer. The software PC-Lab2000 was used to display the digitized data received from the ADC Velleman PCS 500 on a computer monitor in a real-time graphic mode and to record the digitized data into the computer's memory.

Investigations were carried out when the DC high voltage negative potential was applied to the pin electrode. The voltage on the discharge gap was supplied by a stabilized high voltage DC power supply with the maximum amplitude of the output voltage up to 40 kV. To provide stable diffuse mode of corona discharge the appropriate voltage (from the range of 5…14 kV) was chosen. Stable discharge operation mode was controlled by the digital storage oscilloscope LeCroy WaveJet 324A.

The voltage of the discharge gap was measured using the high voltage probe Tektronix P6015A. In the external electric circuit, the ballast resistor R=10 MΩ was set to limit the discharge current. The average discharge current was measured by a micro-ammeter M906. The waveform of the current signal was controlled by a digital storage oscilloscope. The width of the discharge gap between the pin and plate electrodes was set within the range of 5…15 mm.

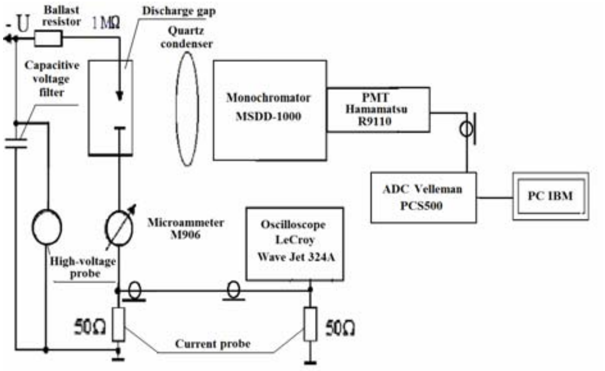

Fig. 1. Schematic diagram of the experimental setup

Temporal characteristics of current pulses were measured using calibrated 50-Ohm current shunts. The signal was processed in the oscilloscope with the bandwidth of 200 MHz and sampling rate of 1 GHz. Temporal characteristics of the current shunts used in the experiments were calibrated by means of the current shunt Tektronix CT1, which has the following characteristics: bandwidth – 25 kHz…1 GHz, pulse rise time – 0.35 ns (10…90% signal), sensitivity – 5mV/mA.

2. RESULTS

The emission spectrum of the discharge radiation within the wavelength range of 300…400 nm were registered. The obtained spectra correspond to the second positive system of nitrogen (transition $C^3\Pi_u$–$B^3\Pi_g$) [7]. The spectra were registered from the cathode region (at ~ 1 mm from the tip of pin electrode) of the discharge. The discharge gap was 5 mm. The absolute humidity of ambient air was 3.4 g/m^3. The width of monochromator slits was 50 µm (output) and 200 µm (input) at the reciprocal linear dispersion of 0.41 nm/mm. The width of monochromator slits, PMT signal amplification factor and the photomultiplier supply voltage were kept up at constant level. The emission spectrum of the discharge within the wavelength range of 300…400 nm is presented in Fig. 2 below.

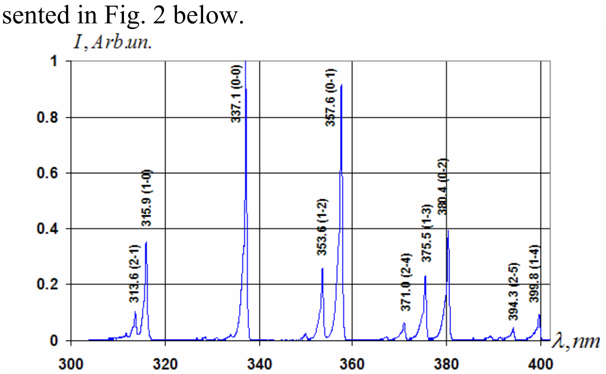

Fig. 2. Emission spectrum of the diffuse negative corona in air at atmospheric pressure. Cathode region of the discharge gap (~1 mm from pin electrode)

Partially-resolved rotational spectrum of the $C^3\Pi_u(0)$–$B^3\Pi_g(0)$ transition was used to determine the rotational temperature. The emission spectrum of the discharge in the wavelength range of 334…338 nm is shown in Fig. 3.

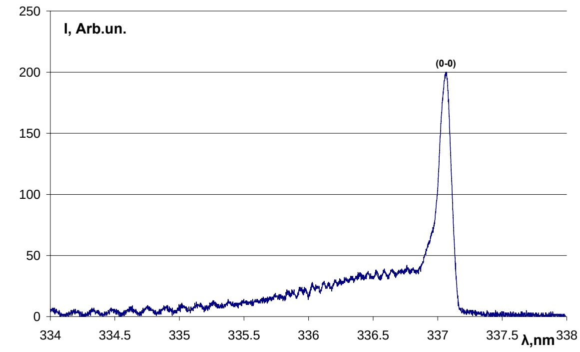

Fig. 3. Emission spectrum of the discharge within the wavelength range of 334…338 nm. ADC sensitivity is 50 mV / div

The fragments of the spectrum within 334…335,8 nm wavelength range were recorded at the maximum sensitivity of the ADC for more detailed investigation of the rotational structure of the spectrum. The fragment is shown in Fig. 4.

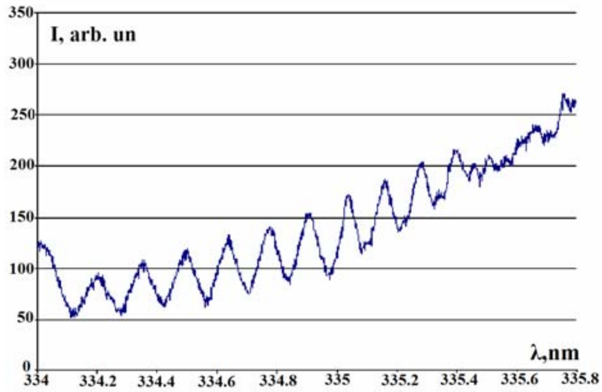

Fig. 4. The fragment of negative corona emission spectra. ADC sensitivity 5 mV / div. The R-branch rotational lines of $C^3\Pi_u(0)$–$B^3\Pi_g(0)$ transition are partially resolved (the range of corresponding rotational numbers is J=20…29)

Theoretical calculation of relative intensities of rotational lines was carried out for more detailed identification of spectral lines and their comparison with experimental results. The calculation was performed using the reference constants [8]. According to the Born-Oppenheimer approach, the term of vibrational-rotational state (J, v) for the given electronic state, is given by [9]

$$E = T_e + G_v + F_J, \qquad (1)$$

where T_e is the electron energy, G_v is the vibrational energy, F_j is the rotational energy.

The vibrational energy (in the first approximation) is given by [9]:

$$G_v = \omega_e (v + 1/2) - \omega_e x_e (v + 1/2)^2, \qquad (2)$$

where v is the vibrational quantum number, ω_e is the energy of vibrational quanta, $\omega_e x_e$ is the anharmonicity constant.

In the non-rigid rotator approach the rotational energy of molecule is given by [9]:

$$F_j = B_v \cdot J \cdot (J+1) - D \cdot \left[J \cdot (J+1)\right]^2, \quad (3)$$

where $B_\upsilon = B_e - \alpha_e(v + 1/2)$ is the rotational constant (in the approach of vibrational-rotational interaction), α_e is the parameter characterizing the dependence of rotational constant on vibrational excitation, J is the rotational quantum number, D is the centrifugal distortion constant.

Using the Fortr diagrams for the linear molecule N_2 at the electronic transition between C $^3\Pi_u$ and $B^3\Pi_g$ states, the correspondence of rotational line wavelength to the specific rotational quantum numbers J was determined. The values of ΔJ = (-1, 0, +1) were allowed according to the selection rules. These rules form P, Q and R branches in the rotational structure of the spectrum. When the wavelength of rotational transitions and its respective rotational quantum numbers are determined, the intensity of rotational lines can be calculated. The radiation intensity of single rotational lines at electronic-vibrational transition is given by [10],

$$I_{J'J''} = \frac{hc}{\lambda} \cdot N_{J'} \cdot A_{J'J''}, \quad (4)$$

where $N_{J'} \sim \frac{B_v'}{kT_{rot}} \cdot (2J'+1) \cdot \exp\left(-\frac{F_j}{kT_{rot}}\right)$ is the population of upper rotational level, T_{rot} – rotational temperature, $A_{J'J''} = \frac{64\pi^4}{3h\lambda^3} \cdot \frac{S_{J'J''}}{2J'+1}$ is the probability of rotational transition (J'-J") [11], λ is the transition wavelength, h is the Planck constant, c is the speed of light , $S_{J'J''}$ is the Henley London intensity factor.

Thus, the intensity of single rotational line is given by the following expression:

$$I(\lambda) \sim \lambda^{-4} \cdot \frac{B_v'}{kT_{rot}} \cdot S_{J'J''} \cdot \exp\left(-\frac{F_j}{kT_{rot}}\right). \quad (5)$$

The calculation of rotational spectra at different T_{rot} showed that there is a spectra range in which the intensity of R-branch lines dominates the intensities of P and Q branches. It was found that only single lines of R-branch with rotational quantum numbers J = (20-29) should be used for analysis of the spectra obtained in the experiment. The fragment of rotational structure for the spectrum of $C^3\Pi_u(0)$–$B^3\Pi_g(0)$ transition is shown in Fig. 5.

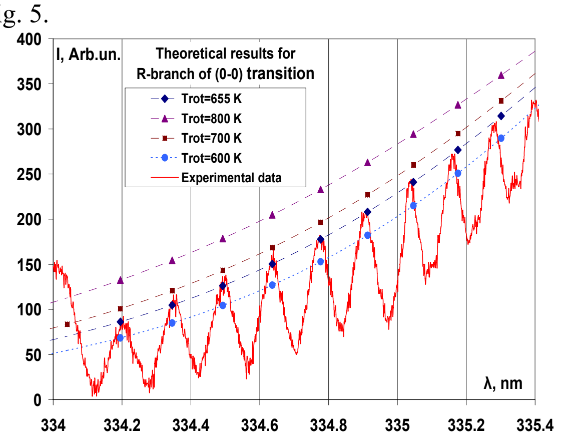

Fig. 5. Rotational structure for the spectrum of $C^3\Pi_u(0)$–$B^3\Pi_g(0)$ electron-vibrational transition (solid line). The calculated distribution of intensities for rotational lines on the R-branch is marked with points

The distribution of intensities for rotational lines on the R-branch is marked with points. The intensity of rotational lines was calculated using different values of rotational temperature T_{rot}.

Analysis of the presented in Fig. 5 spectrum shows a close correspondence of experimental data to theoretical results at T_{rot}=655 K. Fig. 5 also shows that the distribution of intensities for rotational lines is significantly changed together with the rotational temperature value.

There is other approach to determine the rotational temperature from the relative intensities of rotational lines. On the basis of the above mentioned theoretical model, the following equation can be given using (5), and assuming for the R-branch (for sufficiently large J'), $S_{J'J''} \sim J'$ [12]:

$$\ln\left[\frac{I_1(\lambda_1) \cdot \lambda_1^4 \cdot J_2'}{I_2(\lambda_2) \cdot \lambda_2^4 \cdot J_1'}\right] = -\frac{2.618K}{T_{rot}}\left[J_1'(J_1'+1) - J_2'(J_2'+1)\right], \quad (6)$$

where J_1' and J_2' is the rotational numbers corresponding to separate rotational lines, λ_1 and λ_2 is the wavelengths of selected lines, $I_1(\lambda_1)$, $I_2(\lambda_2)$ is the intensities of spectral lines obtained from the experiment.

The T_{rot} can be determined by substituting the measured relative intensities of the rotational lines to the equation (6). The calculated values of nitrogen rotational temperature at different parameters of diffuse negative corona discharge are presented in Table.

Discharge parameters	Rotational temperature, T_{rot}
U=14,2 kV, I=170 μA	623 K
U=14.6 kV, I=215 μA	645 K

CONCLUSIONS

The emission spectra of the nitrogen second positive system at diffuse negative corona discharge were studied. Distribution of emission intensity in electronic vibrational-rotational bands corresponding to $C^3\Pi_u$ - $B^3\Pi_g$ transitions of molecular nitrogen was analyzed. A comparison between experimental data and theoretical results was made in the approach with vibrational-rotational interaction and centrifugal distortion of molecules. The rotational temperature of nitrogen molecules was obtained on the basis of spectral rotational structure analysis. The dependence of rotational temperature on the discharge operation mode, as well as on the applied voltage value was shown. It was shown that rotational temperature of nitrogen molecules is increased together with applied voltage.

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ТЕМПЕРАТУРА ГАЗА В ОТРИЦАТЕЛЬНОМ КОРОННОМ РАЗРЯДЕ ДИФФУЗНОЙ СТАДИИ

О.В. Болотов, В.И. Голота, С.Д. Гуртовой, Ю.В. Сытникова, Д.В. Мошинский

Исследованы спектры излучения отрицательной короны в диффузной стадии горения. Проанализировано распределение интенсивности излучения в электронно-колебательно-вращательных полосах молекулярного азота. Проведен теоретический расчет интенсивности линий в приближении колебательно-вращательного взаимодействия и центробежного растяжения молекул. Определена вращательная температура молекул азота в прикатодной области разряда. Установлена зависимость вращательной температуры как от режима горения разряда, так и от величины напряжения, приложенного к разрядному промежутку.

ТЕМПЕРАТУРА ГАЗУ У НЕГАТИВНОМУ КОРОННОМУ РОЗРЯДІ ДИФУЗНОЇ СТАДІЇ

О.В. Болотов, В.І. Голота, С.Д. Гуртовой, Ю.В. Ситнікова, Д.В. Мошинський

Досліджено спектри випромінювання негативної корони в дифузній стадії горіння. Проаналізовано розподіл інтенсивності випромінювання в електронно-коливально-обертальних смугах молекулярного азоту. Проведено теоретичний розрахунок інтенсивності ліній в наближенні коливально-обертальної взаємодії і відцентрового розтягування молекул. Визначена обертальна температура молекул азоту в прикатодній області розряду. Встановлена залежність обертальної температури як від режиму горіння розряду, так і від величини напруги, прикладеної до розрядного проміжку.