STUDY OF NON-ISOTHERMALITY OF ATMOSPHERIC PLASMA IN TRANSVERSE ARC DISCHARGE

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Properties of non-equilibrium plasma in the transverse arc discharge that occurs in air and argon jets at atmospheric pressure have been investigated by optical emission spectroscopy. The temperatures of population of excited electronic (T_e^*), vibrational (T_v^*), and rotational (T_v^*) states of molecular and atomic components in plasma have been determined, and their distributions along the gas flow have been studied. A high level of plasma non-isothermality, i.e. $T_r^* < T_v^* < T_e^*$, was found to be caused by different scales of characteristic times of elementary kinetic processes in the dynamic gas-discharge system. The revealed difference between the values of T_e^* for copper atoms CuI (the electrode material) and for oxygen OI, hydrogen HI, and argon ArI atoms (the components of a blowing gas) was explained by the availability of an additional electron-ion recombination mechanism of populating the excited electronic states of copper atoms, which is almost inactive for the atoms in the blowing gas.

1. Introduction

Generation of non-equilibrium low-temperature plasma in gas discharges at atmospheric pressure acquires recently the more and more importance, because it makes possible to enhance the power efficiency of plasmochemical processes and to expand the opportunities of using this plasma in various plasma technologies, in particular, in the fabrication of semiconducting structures, deposition of thin-film coating, synthesis of nanopolymer materials, modification of high-molecular compounds, purification and sterilization of environment, as well as in other power-, eco-, and biotechnological applications [1–7].

An important feature of non-equilibrium gas-discharge plasma is related to the fact that the population temperatures of excited electronic, vibrational, and rotational energy levels in molecules and atoms do not coincide with one another [8]. The factor of non-isothermality, when the temperature of electrons considerably exceeds the characteristic temperature of the heavy component (ions, atoms, molecules, radicals), has fundamental significance for the kinetics of non-equilibrium plasmochemical processes. Therefore, the determination of the excitation character and the research of the spatial distribution of non-isothermality in plasma is a very challenging problem [9].

Among electric discharges which can generate high-pressure non-equilibrium plasma (a spark, a corona, and so on), an interesting object for researches is a transverse arc, which is blowing by the gas flow [10]. Unlike the Czernichowski gliding arc [11, 12], the blown transverse arc has a fixed length of the current channel and the convective cooling of the plasma column. Intensive transverse ventilation of plasma enhances its ionization non-equilibrium and non-isothermality. Different setups functioning on the basis of self-sustained and secondary discharges with the use of a transverse arc have been successfully tested by us for the plasma treatment of various gaseous and liquid substances [13–20]. However, in spite of certain achievements in applications, there still exist a lot of unresolved issues in understanding the physical processes which occur in

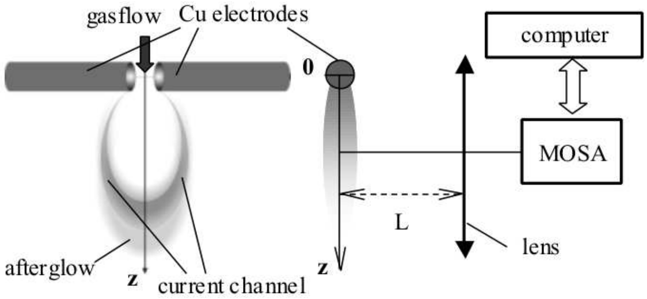

Fig. 1. Schematic diagram of a transverse arc discharge

plasma generated by similar discharges and which require further studies.

The traditional method of diagnostics of non-equilibrium plasma is optical spectroscopy [21]. The emission spectra of arc plasma in an air flow are multicomponent and simultaneously contain atomic lines, molecular bands, and recombination continua of air plasma components, as well as atomic lines given by the products of electrode emission, which frequently overlap; the latter circumstance considerably complicates their identification, interpretation, and treatment [22, 23]. Even the spectral devices with high resolution capability do not always solve this problem completely. Therefore, as an alternative approach, spectral measurements are used in combination with the computer simulation of synthetic spectra, which takes the parameters of the instrumental functions of real devices into consideration. Just such an approach to diagnostics – in particular, the numerical simulation of spectral bands with an unresolved rotational structure [24–26] and multiplet lines with a poorly resolved structure [17] in order to determine the population parameters for vibrational-rotational excited states of molecules and excited electron levels of atoms – was used in this work for studying the character of plasma non-isothermality in a blowing transverse arc.

2. Experiment

Experiments were carried out using a plasmodynamic setup with a transverse arc discharge [20]. The schematic diagram of the experiment is presented in Fig. 1. Cylindrical copper water-cooled electrodes with the diameter $d = 6$ mm were used. The electrodes were arranged coaxially one against another; the interelectrode gap was $\delta = 1.5$ mm. A steel nozzle with the internal diameter $\varnothing = 1$ mm was installed at the distance $H = 20 \div 30$ mm between the electrodes and perpendicularly to them. The nozzle was used to supply a compressed gas (air or technical argon). The gas flow rate G was varied within the range $40 - 180$ cm^3/s. The arc discharge was powered by a high-voltage dc power supply with dropping output voltage-current characteristics, using a ballast resistance. The discharge current I_d was varied within the interval of $100 - 400$ mA.

Optical diagnostics of plasma was carried out by approved methods of emission spectroscopy [16–18]. Plasma radiation was registered by a high-speed multichannel optical spectrum analyzer (MOSA) based on the PC-operated SOLAR SL40-2-USB diffraction spectrometer with a 3648-pixel CCD-detector sensitive in the range of 200–1100 nm (the spectral resolution was about 0.7 nm). Measurements were carried out in various transverse cross-sections of the arc and the afterglow along the axis of the gas flow z; in so doing, the point of intersection of the z-axis with the axis of the electrode system was taken as the zero coordinate, $z = 0$. The image of the arc, which was located at a five-fold focal distance L from the quartz lens, was focused onto the entrance slit of the spectrometer (see Fig. 1).

3. Methodology

The population of the excited electronic levels of atoms and the vibrational-rotational levels of molecules in plasma were determined on the basis of the relative intensities of the emission lines of copper Cu, oxygen O, hydrogen H, and argon Ar atoms and molecular bands of nitrogen N_2; these are the main components in the multicomponent spectrum emitted by plasma in the transverse arc with copper electrodes in the flow of air and argon.

In order to determine the population of the excited electronic levels of atoms, we used the well-known expression for the intensity of a non-reabsorbed spectral line I [22] which is radiated by the nonhomogeneous transparent layer of plasma with thickness l and is registered by a spectral device with the range of vision larger than l:

$$I = C_1 \frac{g_1 f_{12}}{g_2 \lambda^3} N_2,$$

$$N_2 = \int_0^l N_2^* dx, \qquad (1)$$

where C_1 is a numerical constant; g_1 and g_2 are the statistical weights of the lower and upper levels, respectively; f_{12} is the oscillator strength; λ is the wavelength; and N_2^* is the concentration of atoms in the upper level.

To determine the population of the excited electronic levels of oxygen atoms, the relative intensities of the emission lines of multiplets were measured. The intensities of the resulting lines of multiplets with a poorly resolved structure were calculated by the following expression:

$$I_0 = B\frac{(g_1 f_{12})_{\text{eff}}}{\lambda_m^3}\frac{N_2}{g_2},$$

$$(g_1 f_{12})_{\text{eff}} = K\sum_i (g_1 f_{12})_i\,, \qquad (2)$$

where B is a numerical constant, λ_m is the wavelength of the most intensive component in the multiplet, N_2 is the concentration of atoms in the upper level, K is the proportionality factor, and i is the component order number in the multiplet. The spectroscopic constants (the wavelengths, energies of upper excited levels, statistical weights, and oscillator strengths) were taken from the NIST electronic database [27].

In the case where spectra are registered by devices, whose resolution capability allows one to observe the occurrence of multiplet structure, a certain coefficient K should be introduced for correction of the sum of products "the statistical weight times the oscillator strength" of the corresponding multiplet components. In work [17], the simulation for oxygen multiplets was fulfilled taking an instrumental function of the Gaussian type into account, and the dependence of the coefficient K on the instrumental function halfwidth w_i was studied in the range from 0.2 to 2.0 nm. It was shown that this correction coefficient K must be taken into consideration, if the spectra are registered by devices, whose halfwidths of instrumental functions are of the order of the spectral distance between multiplet components ($wi \leq \delta\lambda_i$), i.e. while observing a poorly resolved multiplet structure.

In the general case, the character of the population of excited levels in non-equilibrium plasma can differ from the Boltzmann law, with the corresponding temperatures population differing from one another [8]. Therefore, the Boltzmann distribution of the population of excited levels over energy has to be verified. Provided that there is an exponential dependence for the population variation of the corresponding excited levels N_2/g_2, one can formally introduce the definition of the corresponding population temperature T^* for excited levels in non-equilibrium plasma, i.e. the electronic, T_e^*, vibrational, T_v^*, and rotational, T_r^*, temperatures.

The method for the determination of the temperature of population of excited vibrational levels T_v^*, which use the relative intensities of emission vibrational bands of nitrogen molecules, proceeds from the assumption that the vibrational temperatures in the ground and electronically excited states are close to each other [28]. The expression for calculating the population of vibrational levels in the upper electronic state $C^3\Pi_u$ of the second positive system of molecular nitrogen, which arises owing to the optical transition between electronic states $C^3\Pi_u$ and $B^3\Pi_g$, is as follows:

$$\ln N_{C'} \approx \ln C + \ln\left(\frac{I_{C'B''}}{g_{C'B''}\cdot \nu_{C'B''}^4}\right), \qquad (3)$$

where C is a numerical constant, $\nu_{C'B''}$ is the frequency of transition, $I_{C'B''}$ is the intensity of the vibrational band, $N_{C'}$ is the population of the upper excited level, and $g_{C'B''}$ are the Franck–Condon factors which determine the relative probability of the electronic-vibrational transition.

The technique applied for the determination of the population temperature for excited rotational levels T_r^*, which uses the relative intensities of emission molecular bands with unresolved vibrational-rotational structure, is based on the comparison of the numerically calculated spectral bands with experimental data [24–26]. As the main object of the simulation, band $(1-4)$ of the second positive system of molecular nitrogen (the transition $C-B$) was selected. Such a choice was caused by the fact that this band is free from the overlapping with other spectral lines. A only shortcoming of using this band is its low intensity with respect to other bands of the system concerned. It is widely adopted that, in the ground electronic state of molecules, the distribution function over rotational levels is the Boltzmann one, with the temperature equal to that of the translational degree of freedom [1]. It is also known that, for excited electronic states of molecules as well, the distribution function over rotational levels is close to the Boltzmann one [8]. This results from the fact that, when electrons and molecules collide, the momenta of molecules change insignificantly.

For the determination of the population temperature of the rotational levels of nitrogen molecules, the relative intensities of the vibrational-rotational bands were

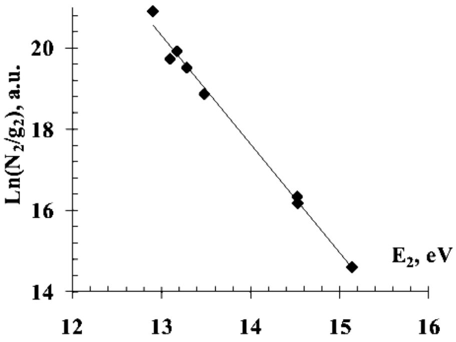

Fig. 2. Population distribution of excited electronic levels of argon atoms in the transverse arc plasma

used:

$$I_{J'J''} = C\nu^4 \exp\left(-\frac{F(J')}{T_r^*}\right), \tag{4}$$

where $I_{J'J''}$ is the intensity of a rotational line for the transition $J' \to J''$; C is a numerical constant; $F(J')$ is the energy of rotational term for the components $^3\Pi_2$, $^3_1\Pi$, and $^3_0\Pi$ of the excited electronic state; J' and J'' are the rotational quantum numbers of the upper and lower states, respectively; and $S_{J'J''}$ are the Höln–London factors which govern the relative probability of the vibrational-rotational transitions.

The magnitudes of rotational terms for the components of an excited state of nitrogen molecules and the values of Höln–London factors were calculated for all components of the energy states by making use of the expressions taken from work [28].

4. Results and Discussion

In this work, we determined the population temperatures T_e^* of excited electronic levels of copper atoms available in plasma using the data on the relative intensities of emission spectral lines with the following wavelengths: λ = 465.1, 510.5, 515.3, 521.8, and 578.2 nm. Again, for hydrogen atoms, we used data for λ = 656.3 and 486.1 nm; for argon atoms, data for λ = 415.9, 603.2, 667.7, 763.5, 852, and 912 nm; and for atomic oxygen, data for multiplets with λ = 777.4, 844.6, and 926.6 nm. This very choice of spectral lines was caused (i) by their sufficient intensity, (ii) by the absence of the overlapping between the majority of those lines and other spectral bands, and (iii) by the largest difference ΔE_2 between the energies of upper excited levels of corresponding spectral transitions that is important for the diminishing of the temperature determination error given by this technique. In the calculations of level populations, we used the expressions obtained from Eq. (2):

$$\frac{N_2}{g_2} = B\frac{I_0 \cdot \lambda^3}{g_1 f_{12}} \exp\left(-\frac{E_2}{T_e^*}\right), \tag{5}$$

$$\ln\left(\frac{N_2}{g_2}\right) = \ln A - \frac{E_2}{T_e^*}, \tag{6}$$

where B and A are numerical constants, E_2 is the excitation energy of the upper level, and T_e^* is the population temperature of excited electronic levels.

The character of the population distribution over energy in the excited electronic levels of argon atoms in the transverse arc plasma is shown in Fig. 2. From this figure, one can see that the dependence is close to a linear one, which made it possible to determine, by the slope angle of the straight line, the population temperature $T_e^*(\mathrm{Ar}) \approx 0.35$ eV. We also tested the Boltzmann character of the population distribution of excited electronic levels in oxygen and hydrogen atoms (the components of the gas flow) and copper ones (the material of electrodes). The following values for the population temperatures were obtained: $T_e^*(\mathrm{O}) \approx 0.35$ eV, $T_e^*(\mathrm{H}) \approx 0.5$ eV, and $T_e^*(\mathrm{Cu}) \approx 0.8$ eV. It should be noted that the temperature determination error for $T_e^*(\mathrm{H})$ was larger than those for $T_e^*(\mathrm{O})$, $T_e^*(\mathrm{Cu})$, and $T_e^*(\mathrm{Ar})$, because the population temperature of excited electronic levels in hydrogen atoms was determined using the data for only two emission lines H_α and H_β (the line H_γ was not observable in the spectra due to its very low intensity).

The spatial distributions of those temperatures in plasma along the gas-flow (coordinate z) are shown in Fig. 3.

Since the basic ions in low-current arc are ions of the electrode material, an additional mechanism of populating the excited electronic levels in copper atoms, besides the excitation by electrons, can appear owing to electron-ion recombination; this mechanism is inactive for the atoms of a blowing gas. Our researches of the distributions of the intensity of copper emission lines along the gas-flow coordinate z revealed a non-monotonous character of those dependences. A reduction of the intensity in the interval $z = 11 \div 20$ mm and the simultaneous growth of the population temperature of excited electronic levels of copper atoms in this z-interval (Fig. 3,*b*) can testify to the emergence of the recombination mechanism of their population. In our

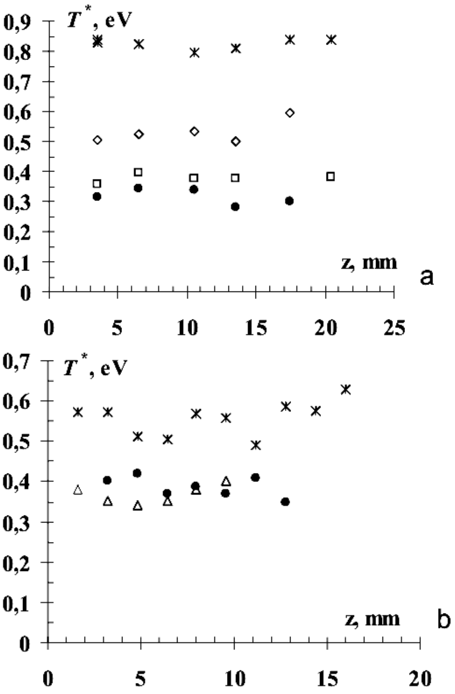

Fig. 3. Population temperature distributions of excited electronic levels of copper, oxygen, hydrogen, and argon atoms and excited vibrational levels of nitrogen molecules along the flow coordinate z in the transverse arc plasma: (a) argon (I_d = 400 mA, G = 75 cm^3/s), (b) air (I_d = 480 mA, G = 40 cm^3/s): $*$ – T_e^*(Cu), ◇ – T_e^*(H), □ – T_e^*(Ar), • – T_e^*(O), △ – T_v^*(N$_2$)

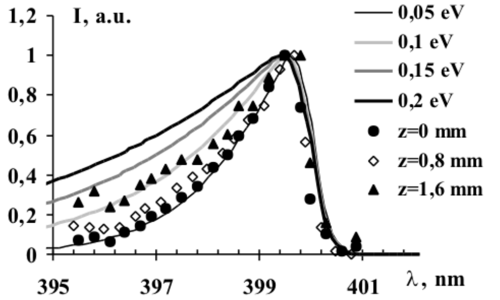

Fig. 4. Theoretically calculated $(1-4)$ band for N$_2$ (the transition $C-B$). Markers denote experimental data for air plasma at various z (H = 30 mm, I_d = 480 mA, G = 150 cm^3/s)

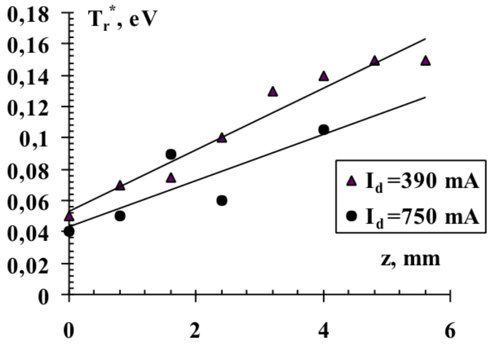

Fig. 5. Distributions of the rotational temperature T_r^* of nitrogen molecules along the flow coordinate z in air plasma for two regimes of discharge current I_d (H = 30 mm, G = 150 cm^3/s)

opinion, it is this circumstance that can explain the difference revealed between the values of population temperature for excited electronic levels in the atoms of a blowing gas and in copper atoms.

In order to determine the vibrational temperature of molecules in plasma, we tested the Boltzmann character of the population of excited vibrational levels in the second positive system of molecular nitrogen (bands $(0-0)$, $(1-4)$, $(0-2)$, $(1-3)$, and $(2-4)$). From the slope of the linear section in the population distribution of excited vibrational levels, the vibrational temperature T_v^* was determined. The spatial distributions of population temperatures of the excited electronic and vibrational levels in plasma along the flow (coordinate z) are depicted in Fig. 3,b.

The results of numerical simulations of the emission spectrum of vibrational-rotational band $(1-4)$ of the second positive system of molecular nitrogen depending on the rotational temperature T_r^* are presented in Fig. 4.

By imposing experimental data on the theoretically calculated grid of temperatures T_r^* for band $(1-4)$, the values $T_{r1}^* = 0.05 \div 0.1$ eV was determined. The obtained data are somewhat lower than those calculated for band $(0-0)$ ($T_{r2}^* = 0.1 \div 0.15$ eV). It is related to the fact that band $(0-0)$ of an N$_2$-molecule (transition $C-B$ with the wavelength $\lambda = 337.1$ nm) is overlapped by intense band $(0-0)$ of NH (transition $A^3\Pi - X^3\Sigma$, branch Q with the maximum at $\lambda = 336.01$ nm). Therefore, it is expedient to determine the rotational temperature in the transverse arc plasma in the air flow on the basis of the relative intensity of band $(1-4)$ of molecular nitrogen (transition $C-B$).

In this work, we also studied the spatial distribution of the rotational temperature T_r^* in plasma along the flow [the dependence on the distance z from electrodes (see Fig. 5)]. The increasing character of this dependence may testify that the gas heating occurs owing to the convective heat exchange between gas particles and the current channel at a periphery of the transverse arc. A more detailed explanation of the revealed distribution of T_r^* requires further researches.

Near values for the population temperature of electronic levels of argon atoms and for the vibrational and rotational temperatures of molecular nitrogen ($T_{\rm exc}$=(4110 ± 230) K, $T_{\rm vib}$=(2100 ± 150) K, and $T_{\rm rot}$=(1100 ± 100) K, respectively) were obtained in work [26] for the non-equilibrium plasma of a gliding arc at atmospheric pressure. Supposing that our blowing transverse arc is similar to the gliding one – because both discharges are realized in the transverse gas flow, – the comparison of the results obtained for similar discharge parameters seems quite correct.

5. Conclusions

In this work, the population temperatures of excited electronic, T_e^*, vibrational, T_v^*, and rotational, T_r^*, levels of molecular and atomic components in atmospheric plasma of transverse arc discharge have been determined, and the dynamics of their spatial distribution along the gas flow has been studied. It has been demonstrated that all regimes under investigation are characterized by a high degree of plasma non-isothermality.

The revealed difference between the values of T_e^* for copper atoms (the electrode material), on the one hand, and oxygen, hydrogen, and argon atoms (the gas flow components), on the other hand, has been explained as a manifestation of an additional electron-ion recombination mechanism which is active in populating the excited electronic levels of copper atoms, but inactive for the atoms of a blowing gas.

The inequality of the temperatures, $T_r^* < T_v^* < T_e^*$, and the different characters of their distributions along the flow – in particular, a gradual growth of the rotational and vibrational temperatures with the distance from the electrodes, in contrast to the practically constant electronic temperature – are caused by different ratios between the characteristic times of transport processes in the dynamic gas-discharge system and the characteristic times of excitation of the corresponding energy levels in different components of plasma. But this effect demands further researches.

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ДОСЛІДЖЕННЯ НЕІЗОТЕРМІЧНОСТІ АТМОСФЕРНОЇ ПЛАЗМИ ПОПЕРЕЧНОГО ДУГОВОГО РОЗРЯДУ

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Резюме

Методами оптичної емісійної спектроскопії досліджено властивості нерівноважної плазми поперечного дугового розряду в потоці повітря та аргону атмосферного тиску. Визначено температури заселення збуджених електронних T_e^*, коливальних T_v^* та обертальних T_r^* станів молекулярних і атомарних компонентів плазми, а також вивчено розподіл цих температур вздовж газового потоку. Показано високий рівень неізотермічності плазми, $T_r^* < T_v^* < T_e^*$, зумовленої різними масштабами характерного часу перебігу елементарних кінетичних процесів у динамічній газорозрядній системі. Виявлену різницю між значеннями T_e^* для атомів міді CuI (матеріал електродів) та для атомів кисню OI, водню HI, аргону ArI (компоненти газового потоку) пояснено наявністю додаткового електрон-іонного рекомбінаційного механізму заселення збуджених електронних рівнів атомів міді, який практично відсутній для атомів обдуваючого газового потоку.