SOURCES OF NONEQUILIBRIUM PLASMA AT ATMOSPHERIC PRESSURE

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The electrical arc discharge in a transverse blowing gas flow [transverse arc – (TA)] and the discharge in the gas flows immersed into the liquid [discharge in the gas channel with a liquid wall – (DGCLW)] are studied as the sources of nonequilibrium plasma at the atmospheric pressure. Diagnostics of both discharges is made by optical emission spectroscopy. The population distribution temperatures of excited electronic and vibrational states (electronic T_e^* and vibrational T_v^* temperatures, respectively) are determined from Boltzmann plots. Rotational temperatures T_r^* are determined by comparison of some nitrogen and hydroxyl experimental spectral bands with results of their computer simulations. Measurements are carried out for different currents I_d and gas flow rates G. Plasma parameters of TA and DGCLW were compared with other known sources of nonequilibrium plasma such as a gliding arc (GA) and the gliding arc in a tornado (GAT). It is concluded that TA and DGCLW could generate a high-pressure non-thermal plasma stationary in time. At the same time, the level of non-equilibrium of the TA plasma is higher than that in DGCLW.

1. Introduction

Non-equilibrium atmospheric-pressure plasmas are widely used in different fields of plasmochemistry, energy-efficient and ecology technologies such as the plasma-assisted reforming of hydrocarbon fuel, purification of water and air, production of nanoparticles, *etc.* The most interesting sources of high-pressure non-thermal plasma, which can provide a high plasma density and a high level of nonequilibrium, are GA [1, 2], GAT [3, 4], DC transverse glow discharge [5], TA in the blowing gas flow [5–8], and DGCLW [9]. In the present work, we study TA and DGCLW. Diagnostics of plasma parameters of both investigated discharges is carried out by optical emission spectroscopy (OES). The characteristic temperatures, which correspond to the population distribution of the excited electronic states of atoms (electronic temperature T_e^*), vibration and rotational levels of molecules (vibration T_v^* and rotation T_r^* temperatures) in the plasmas under study, are determined by OES methods [8].

2. Experiment

2.1. Arc in the transverse blowing gas flow

The arc in the transverse blowing gas flow differs from a non-stationary gliding arc of the Czernichowski type [1,2] by the fixed arc length. It has also a convective cooling of the plasma column by the airflow but without conductive heat losses at walls, since it is a free arc jet. An intense transverse ventilation of the arc plasma increases its ionization non-equilibrium and non-isothermality. The schematic diagram of a TA discharge is shown on Figure 1,*a*.

The atmospheric-pressure gas (air or argon) flowed from the nozzle across two horizontal opposite electrodes and formed a bright crescent-shaped electric arc. The rod copper electrodes with the diameter $d = 6$ mm were mounted with the nominal gap $\delta = 1.5$ mm. The axisymmetric stainless-steel nozzle with the inner diameter $\varnothing = 1$ mm was centred between the electrodes at the distance $L = 20$ mm above the electrode axis. The gas flow rate G was regulated from $G = 0$ to 110 cm^3/s. There was the enough high gas-dynamic pressure in the flow to blow out the electric arc downstream.

The TA discharge was powered by the DC power supply through the ballast resistance $R = 2k\Omega$ in the circuit. Due to the absence of the cooling of electrodes, the electric discharge energy was transferred totally to the plasma flow. The discharge current I_d during the experimental run was kept constant: typical values were $I_d = 400$ mA in a case of argon flow and $I_d = 480$ mA in the case of air flow. The current-voltage characteristics of a TA discharge in the air flow are shown in Figure 1,*b*.

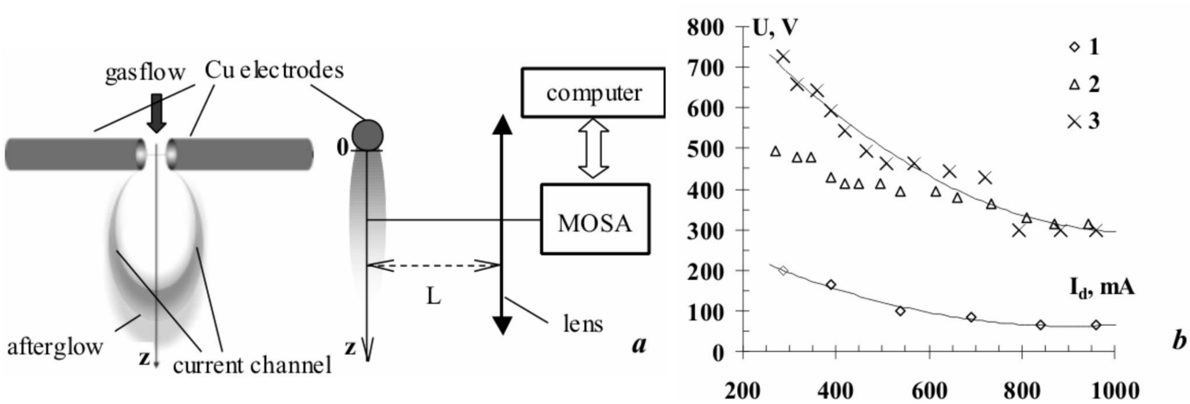

Fig. 1. Arc discharge in the transverse blowing gas flow: *a* – scheme of the OES diagnostics of TA; *b* – current-voltage characteristics of TA in air for different gas flow rates G= 0 (*1*), 38 (*2*), and 75 cm^3/s (*3*)

The OES diagnostics was conducted at different cross-sections along the arc and in the afterglow with a spatial resolution of ∼ 0.1 mm. A portable PC-operated CCD-based spectrometer SL40-2-3648 with a spectral resolution of ∼ 0.7 nm was used for the registration of spectra within the range of 210–1100 nm. The images were focused on the entrance slit of a spectrometer by a quartz lens at the bench 5-focus distance from the arc. The standard mercury, xenon, deuterium, and tungsten band lamps were used for the spectral calibration.

2.2. Discharge in a gas channel with liquid wall (DGCLW)

The discharge in the gas channel with a liquid wall has basic distinction from the diaphragm and capillary gas-liquid discharges operating in the DC mode. Its main advantages are: (i) large ratio of the plasma-liquid contact surface to the plasma volume; (ii) wide variation of gas discharge reactivity; (iii) selectivity of plasmochemical processes during the treatment. The schematic diagram of a DGCLW reactor is shown in Figure 2,*a*.

It consists of a quartz cylindrical tank *1* of 50 mm in diameter and 170 mm in height. The top and bottom of the tank were hermetically sealed by duralumin flanges *2*, in which the electrode system was built in. Cylindrical copper electrodes of 3 of *3* mm in diameter and 40 mm in length were placed inside glass tubes *4* ending by 4-mm-diameter outlet nozzles. The tubes with electrodes were installed coaxially nozzle-to-nozzle. The gas (air) was directed through the tubes along the top and bottom electrodes and formed gas channel *5* connecting both electrodes. The nominal gap between electrodes was 10 mm. The working liquid (distilled water or a solution) filled up the reactor through tube *7* built at a bottom flange. The level of the liquid was kept constant by using the system of communicating vessels. The pressure inside the reactor and communicating vessels was maintained by means of tube *8* built in a top flange. Tube *9* built at the top flange was used for the output of gaseous products created during the plasma-treatment of the working liquid. The reactor walls were cooled by water-cooling system *10*.

The discharge was powered by the DC power supply. The current I_d varied from 100 up to 400 mA, the airflow rate G during the experimental run was kept constant: the typical value was G = 110 cm^3/s. The current-voltage characteristics of DGCLW in the air-water system are shown in Fig. 2, *b*. The OES diagnostics for DGCLW was made.

3. Results and Discussion

3.1. Optical diagnostics of the transverse arc

It was found that, in the TA plasma, the character of the population distributions of excited electronic states of argon, oxygen and copper atoms is close to the Boltzmann law [8]. Thus, the corresponding T_e^* values were determined from the Boltzmann plots. For this purpose, the relative intensities of the following emission lines were analyzed: (1) ArI lines: λ = 763.5, 842.5, 852, and 912 nm; (2) OI multiplets: λ = 777, 844, and 926 nm; (3) CuI lines: λ = 465.1, 510.5, 515.3, 521.8, and 578.2 nm. The choice of these lines is conditioned by: (i) no overlapping of these lines with other spectral lines and bands; (ii) energy differences between the upper excited

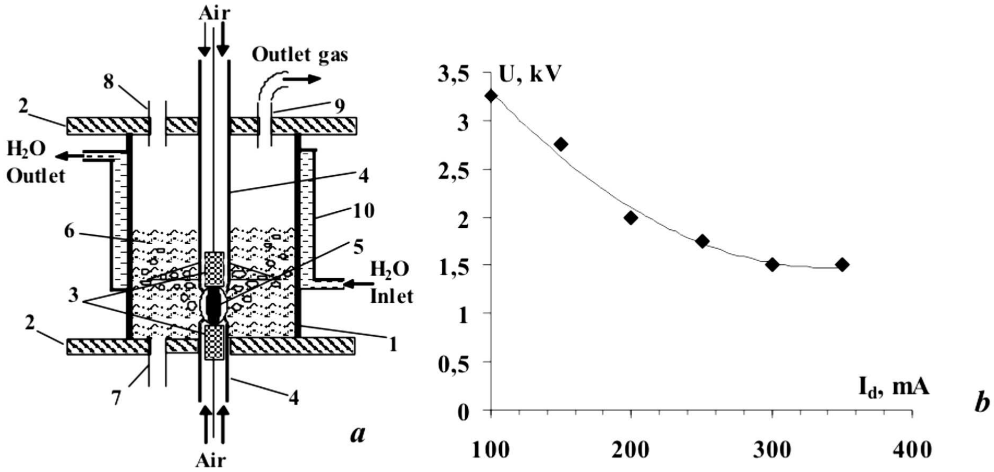

Fig. 2. Discharge in the gas channel with liquid wall: *a* – scheme of a DGCLW reactor; *b* – current-voltage characteristics of DGCLW in the air-water system for the gas flow rate $G = 110$ cm^3/s

levels of these spectral transitions, which are high enough to reduce the inaccuracy in temperature calculations.

The Boltzmann population distribution of excited vibrational levels of nitrogen molecules in the TA plasma was shown for the 2^+-system of N_2 (electronic transition $C^3\Pi_u$–$B^3\Pi_g$: vibrational bands (0–0) $\lambda = 337.1$ nm; (0–1) $\lambda = 357.7$ nm; (1–4) $\lambda = 399.8$ nm; (0–2) $\lambda = 380.5$ nm; (1–3) $\lambda = 375.5$ nm; (2–4) $\lambda = 371.0$ nm), and corresponding vibrational temperatures T_v^* were determined from the Boltzmann plots. The rotational temperatures T_r^* were evaluated from the comparison of experimental spectra with the results of numerical spectral simulations for the N_2 ($C^3\Pi_u$–$B^3\Pi_g$) vibrational-rotational band (1–4) at $\lambda = 399.8$ nm. Typical values of T_e^*, T_v^*, and T_r^* temperature distributions in the TA plasma are shown in Figure 3,*a*. The inaccuracy of temperature measurements does not exceed 15%.

From these data, one can see that T_e^* of Cu atoms (material of electrodes) is slightly higher than T_e^* of O and H atoms (products of the discharge). Since the main source of ions in the plasma of low current arcs are ions from a material of electrodes, besides the direct electron excitation, the additional population of upper excited electronic states of Cu atoms can occur due to the electron-ion recombination. Thus, the revealed difference between T_e^* values can be explained by the additional electron-ion recombination mechanism of population of the upper excited electronic states of Cu atoms, which is almost absent for O and H atoms in the blowing gas.

3.2. Optical diagnostics of the discharge in a gas channel with liquid wall

Emission spectra of the DGCLW plasma contain the UV system of OH ($A^2\Sigma$–$X^2\Pi$: 306.4–308.9 nm), 2^+-system of N_2 ($C^3\Pi_u$–$B^3\Pi_g$: 337.1, 357.7, 380.5, 316.0 nm), atomic lines H_α (656.3 nm), H_β (486.1 nm), H_γ(434.05 nm), OI (777.1, 844.6, 926.6 nm), CuI (465.1, 510.5, 515.3, 521.8, 578.2 nm) *etc.* It was shown that the population distributions for the excited electronic states of H, O, and Cu atoms and for excited vibration levels of N_2 molecules in DGCLW are also close to the Boltzmann law for the used regimes of a discharge. Thus, corresponding electronic and vibrational temperatures were determined from the Boltzmann plots.

The T_e^* and T_v^* temperature dependences in the DGCLW plasma are shown in Fig. 3,*b*. As can be seen from these curves, the electronic temperatures are almost independent of the discharge current I_d for the investigated regime of the airflow rate G. It can be related to the exponential dependence of the ionization rate on the electron's temperature (effect of the electric field strength E/N).

3.3. Comparison of various sources of nonequilibrium atmospheric-pressure plasma

The comparative analysis of the plasma parameters of both investigated electric discharges, TA and DGCLW, with other known sources of non-equilibrium atmospheric-pressure plasma was made, and the main results are presented in the Table.

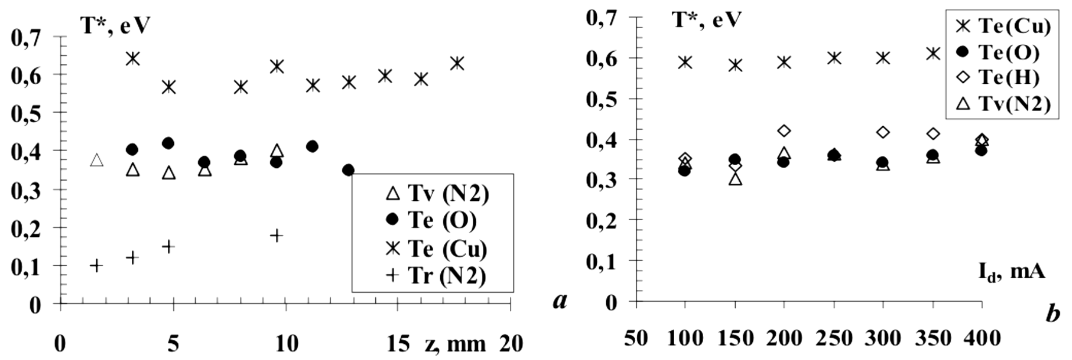

Fig. 3. Characteristic temperatures in the investigated discharge plasma: *a* – temperature distributions along the gas flow z in TA (I_d = 480 mA, G = 38 cm^3/s); *b* – temperature dependences for different discharge currents I_d in DGCLW (G = 110 cm^3/s)

Parameters of non-equilibrium atmospheric-pressure plasma sources

Type	Electric power, W	Gas flow rate G, cm^3/s	T_e^*, eV	T_v^*, eV	T_r^*, eV
GA [2]	200–1000	$(2 \div 50) \times 10^3$	*T: 0.52 **NT: 0.86	*T: 0.27–0.34 **NT: 0.17–0.26	*T:0.2–0.34 **NT: 0.07–0.18
GAT [4]	90–300	$(0.5 \div 2.5) \times 10^3$	> 0.9		0.17–0.34
TA [6,8]	220–330	$(0.04 \div 0.2) \times 10^3$	in air: (Cu) 0.6 (O) 0.35 (H) 0.35 in argon: (Cu) 0.9 (O) 0.3–0.35 (H) 0.4–0.55 (Ar)0.3–0.4	(N_2) 0.35	(N_2) 0.1–0.2
DGCLW [This work]	260–300	0.11×10^3	(Cu) 0.6 (O) 0.35 (H) 0.35	(N_2) 0.3–0.35	(OH) 0.35–0.4

*T – thermal regime of GA, **NT – non-thermal regime of GA [2]

4. Conclusions

It is found that the TA discharge plasma in the investigated regimes is highly non-thermal: $T^*_{r(\mathrm{N_2})} < T^*_{v(\mathrm{N_2})} < T^*_{e(\mathrm{O,H,Ar})} < T^*_{e(\mathrm{Cu})}$.

It is revealed that the electronic temperature T_e^* of Cu atoms (material of electrodes) in the plasma is higher than that for Ar, O, and H atoms due to the additional electron-ion recombination mechanism of population of excited electronic states of Cu atoms that is almost absent for Ar, O, and H atoms in the blowing gas.

It is shown that, due to the effective heat and mass transfer at the plasma-gas and plasma-liquid interface, TA and DGCLW could generate non-thermal plasmas. At the same time, the level of non-equilibrium in the plasma of TA is higher than that in DGCLW.

In comparison with GA and GAT sources, both TA and DGCLW discharges operate with the gas flow rates, which are much less than that used in GA and GAT under a similar electric power, and can generate a non-equilibrium plasmas stationary in time.

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ДЖЕРЕЛА НЕРІВНОВАЖНОЇ ПЛАЗМИ АТМОСФЕРНОГО ТИСКУ

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Р е з ю м е

Електричний дуговий розряд, що поперечно обдувається газовим потоком (поперечна дуга – ПД), і розряд у газових потоках, затоплених рідиною (розряд у газовому каналі з рідкою стінкою – РГКРС) вивчені як джерела нерівноважної плазми атмосферного тиску. Діагностику обох розрядів виконано за допомогою оптичної емісійної спектроскопії. Температури заселення збуджених електронних та коливальних рівнів (електронні T_e^* та коливальні T_v^* температури відповідно) визначені з графіків больцманівських розподілів. Обертальні температури T_r^* визначено шляхом порівняння деяких експериментально виміряних спектральних смуг азоту і гідроксилу з результатами їх комп'ютерного моделювання. Вимірювання виконані для різних значень струмів розрядів I_d та швидкості потоку газу G. Параметри плазми ПД та РГКРС порівнювались з іншими відомими джерелами нерівноважної плазми, такими, як ковзаюча дуга та ковзаюча дуга у торнадо. Зроблено висновок, що ПД та РГКРС можуть генерувати неізотермічну стаціонарну у часі плазму високого тиску. В той же час рівень нерівноважності плазми ПД вищий, ніж у РГКРС.