Discharge in the gas channel with liquid walls as generator of non-thermal plasma at atmospheric pressure

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Optical diagnostics of the discharge in the gas channel with liquid walls at atmospheric pressure was made. Boltzmann population distributions of the excited electronic states of hydrogen, oxygen, copper atoms and vibronic $C^3\Pi$ states of nitrogen molecules in the generated plasma were shown. Corresponding electronic (T_e) and vibration (T_v) temperatures were determined from Boltzmann plot and non-isothermal character of the investigated plasma ($T_v<T_e$) was found. The difference between T_e of Cu (material of electrodes) and of H, O atoms can be explained by additional electron-ion recombination mechanism of population of the excited electronic states of Cu atoms. It was shown that obtained temperatures (T_e) almost don't depend on the discharge current (I_d) within experimental inaccuracy.

1. Introduction

Plasma is widely used as a source of highly energy particles to achieve maximum efficiency of the treatment at minimum power input in different energetic, ecological and other plasma-chemical technologies. Discharges, which generate non-thermal cold plasma, provide chemical selectivity and high-energy efficiency, but they are limited by low-pressure operation. On the other hand thermal plasma sources enable to deliver high power at high operating pressure, but they provide low excitation selectivity that limits their use in plasma-chemistry.

There are intermediate systems between thermal and non-thermal discharges, which ensure simultaneously high plasma density, power and operating pressure with high level of non-equilibrium. High electronic and low gas temperatures are typical for their plasma. Such transitional non-thermal plasma generators can provide selective chemical processes. Gliding arc [1,2], gliding arc in tornado [3], arc discharge in transverse blowing gasflow [4,5], secondary discharges supported by plasma flow [6,7] belong to these type of discharges and have been efficiently used for reforming of hydrocarbon fuels [8], generation of nanostructures [9], destruction of toxic substances in water [10]. The least investigated among them is the electrical discharge in gas channel with a liquid wall [11]. Its principal discrepancy from diaphragm [12], capillary [13] discharges and from the gas microdischarge in bubbles between valve metal anode and electrolyte solution [14] is its possibility to operate with DC power supply. The main advantages of this discharge are large ratio of the surface of plasma-liquid contact to the plasma volume and the possibility of external control of plasma-created gas compound, which specifies its large potential opportunities in plasmachemical applications. This work is dedicated to the investigation of plasma parameters of the electrical discharge in gas channel with liquid walls at atmospheric pressure.

2. Experiment

Research of discharge plasma in the gas channel with water wall was carried out in the reactor, which is shown on Fig.1.

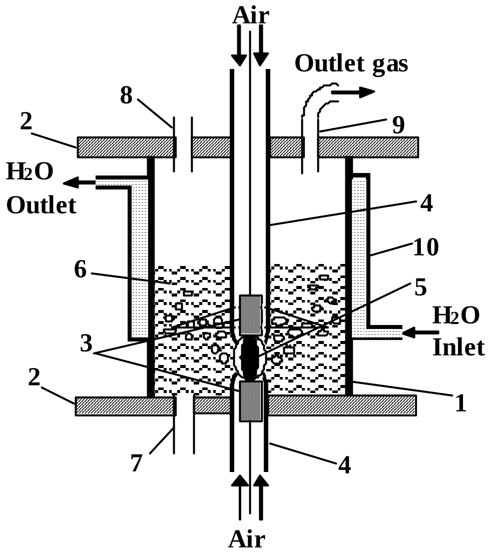

Fig. 1. Experimental schema of the discharge in the gas channel with liquid walls

It consists of quartz cylinder *1* of 50-mm diameter and of 170-mm height. Top and bottom of the cylinder were hermetically sealed by duralumin flanges *2*, in which the system of electrodes was built in. Cylindrical metal electrodes *3* of 3-mm diameter and of 40-mm length were placed inside glass tubes *4* narrowed on the end with outlet of 4-mm diameter. Flows of air directed into these glass tubes along to the top and to the bottom electrodes were colliding and forming stable gas channel, which connects two electrodes. The distance between electrodes was 10 mm. Airflow rate *G* was controlled by rotameter. The liquid *6* was input to the reactor through the tube *7* built into the bottom flange. The level of liquid was kept constant by using the system of communicating vessels. Pressure inside the reactor and communicating vessels was maintained constant by means of tube *8* built in the top flange. Tube *9* built into the top flange was used for outputting gaseous products produced during the plasma treatment of solution.

DC source was used to supply the discharge 5, which burns under atmospheric pressure inside the gas channel with liquid wall. Distilled water was used as working liquid. Volt-ampere characteristic (VAC) of the discharge in the gas channel with water walls is shown on the Fig.2.

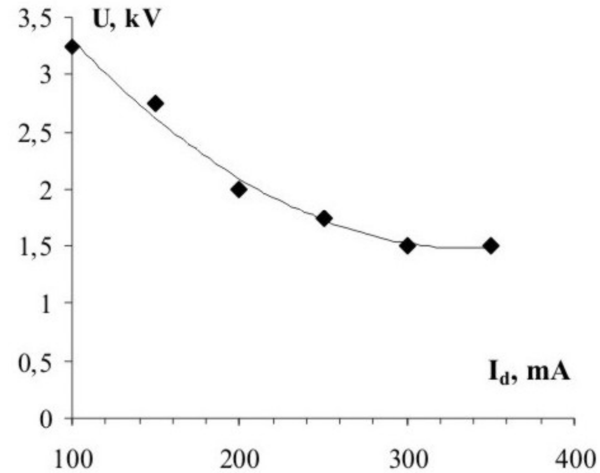

Fig.2. VAC of the discharge in the gas channel with liquid walls.

Emission spectra of plasma were obtained by spectrometer SL-40 based on CCD elements in the range of 200-1100 nm with spectral resolution ~0,75 nm.

Researches of the discharge plasma in the air channel with water wall were carried out for the airflow rate G=110 cm^3/s and for different discharge current I_d changed from 100 up to 350 mA.

3. Results and discussion

Typical emission spectrum of the investigated plasma recorded after two minutes of the discharge burning is shown on the Fig.3.

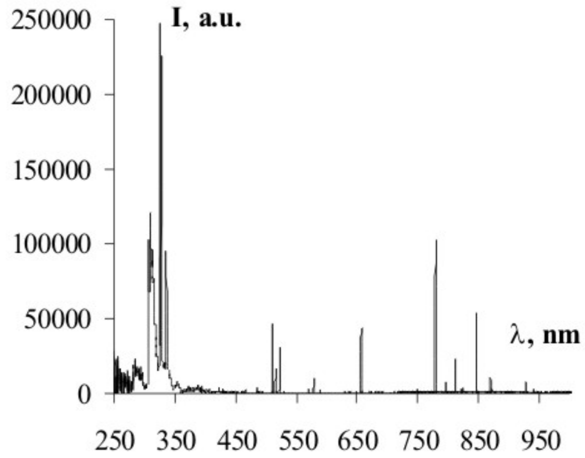

Fig.3. Emission spectrum of plasma of the discharge in the gas channel with water walls obtaned after t=2 min of the discharge burning (I_d=250 mA, G=110 cm^3/s)

It is represented with taking into account the absorption of distillate, which was continuous changing during plasma treatment. Observed spectrum is multi-component and contains UV system of hydroxyl OH ($A^2\Sigma$-$X^2\Pi$: (0-0) 306.4-308.9 nm), 2^+-system of nitrogen N_2 ($C^3\Pi_u$-$B^3\Pi_g$: (0-0) 337.1(0-1) 357.7, (0-2) 380.5, (1-0) 316.0 nm etc.), atomic lines of hydrogen H_α (656.3 nm), H_β (486.1 nm), H_γ (434.05 nm), oxygen OI (777.1, 844.6, 926.6 nm), copper Cu (324.75, 327.4, 465.1, 510.5, 515.3, 521.8, 578.2 nm) etc.

It was shown that excited electronic states population distribution of Cu, O and H atoms in the investigated plasma is close to Boltzmann for used regimes of the discharge. Thus, the temperature, which corresponds to the population distribution of electronic levels (electronic temperature T_e), can be determined from Boltzmann plot:

$$Ln\left(\frac{N_2}{g_2}\right) = Ln\left(\frac{I \cdot \lambda^3}{g_1 f_{12}}\right) - \frac{E_2}{T_e} \qquad (1)$$

Where I– intensity of the emission line; λ– wavelength [μm]; E_2– energy of the upper level; N_2– population of the upper level; g_1 and g_2,– statistical weight of the lower and upper levels correspondingly; f_{12}– oscillator force.

The relative intensities of following emission lines were used for electronic temperature measurements: copper (electrode material) Cu (λ=465.1, λ=510.5, λ=515.3, λ=521.8, λ=578.2 nm); hydrogen H_α, H_β, H_γ (λ=656.3, λ=486.1, λ=434.05 nm); oxygen OI (λ=777.1, λ=844.6, λ=926.6 nm). Choosing of these lines is conditioned by: (i) no overlapping of these lines with other spectral lines and bands; (ii) the value of energy differences between upper excited levels of these spectral transitions, which is high enough to reduce inaccuracy in temperature calculations.

Determination of the temperature of vibration states population distribution (vibration temperature T_v) based on measuring relative intensities of spectral emission bands. It was supposed that vibration temperatures of the ground state and electronic excited states are closed. To calculate vibration states population of the upper electronic state $C^3\Pi_u$ of the 2nd positive system of N_2 (transition $C^3\Pi_u \rightarrow B^3\Pi_g$) the following formula was used:

$$LnN_{C'} \approx LnC + Ln\left(\frac{I_{C'B''}}{g_{C'B''}\nu_{C'B''}^{4}}\right) \qquad (2)$$

Where C is a constant, $\nu_{C'B''}$– frequency of the transition $C^3\Pi_u \rightarrow B^3\Pi_g$, $I_{C'B''}$– intensity of spectral band of corresponding transition, $g_{C'B''}$– Frank-Condon factor of corresponding transition, $N_{C'}$ – population of the upper excited level.

The Boltzmann population distribution of excited vibration states was shown for the 2^+ system of the N_2 (bands (0-0), (1-4), (0-2), (1-3), (2-4)) and vibration temperature of the investigated plasma was determined from Boltzmann plot ($T_v(N_2) \approx 0{,}25$ eV).

It was shown that stabilization time of water temperature in the investigated system is sufficiently large (several minutes). The water temperature is known determines the pressure of vapors. That is why it is interesting to study dependence of energy parameters of plasma on the time of the discharge burning t.

Results of temperature measurements of the investigated plasma for different t and I_d are represented on Fig.4 and Fig.5 correspondingly.

From these figures can be seen that $T_e(\mathrm{O}) \approx 0{,}4$ eV and $T_e(\mathrm{H}) \approx 0{,}35$ eV. These values are noticeably lower than $T_e(\mathrm{Cu}) \approx 0{,}65$ eV. It can be supposed that the main ions in the investigated discharge are ions of copper (material of electrodes). The revealed difference of temperatures can be explained by an additional electron-ion recombination mechanism of excited electronic states population of copper atoms, which is almost absent for the atoms of blowing gas.

The temperature values almost don't depend on the discharge current I_d and time of the discharge burning t within of experimental inaccuracy.

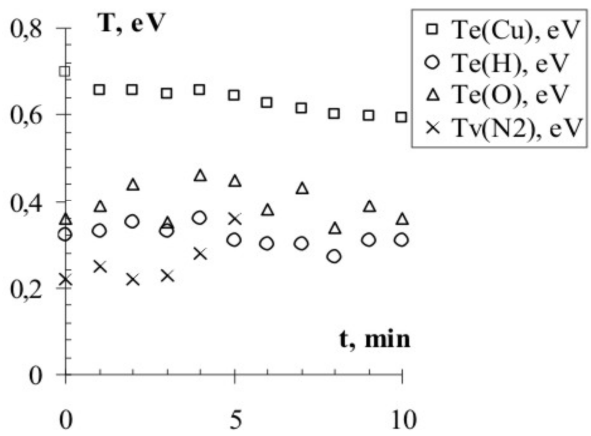

Fig. 4. Temperature dependence on the discharge burning time t for the discharge current I_d=300 mA.

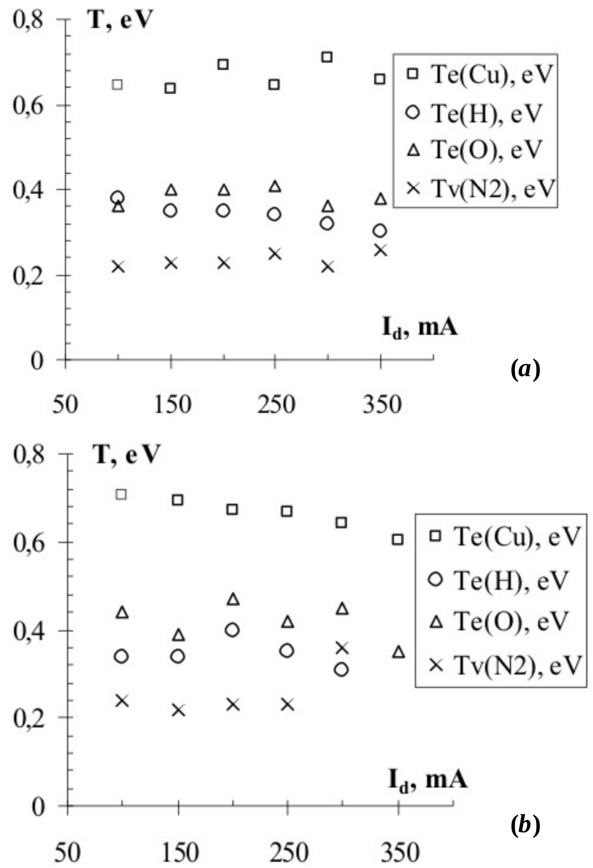

Fig. 5. Temperature dependences on the discharge current I_d for different moments of the discharge burning: *a*– immideatly after turnning on the discharge (t=0 min); *b*– after five minutes of the discharge burning (t=5 min).

4. Conclusions

From the main results of this work the following conclusions can be made:

1. The Boltzmann population distribution of the excited electronic of Cu, O, H atoms and vibronic $C^3\Pi$ states of N_2 molecule was shown and corresponding electronic and vibration temperatures were determined from Boltzmann plot.
2. It was shown that plasma generated by the electrical discharge in the gas channel with liquid walls is strongly non-isothermal ($T_v<T_e$).
3. The difference of T_e temperatures can be explained by an additional electron-ion recombination mechanism of excited electronic states population of copper atoms (material of electrodes).
4. Energy plasma characteristics of the investigated discharge (temperatures of the excited states population distribution T_e, T_v) don't change significantly from the discharge parameters during its burning.

5. Acknowledgement

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