

V.V. AZHARONOK, I.I.FILATOVA, V.D.SHIMANOVICH, V.N OCHKIN, L.N.ORLOV, J.I.NECRASHEVICH, V.V.NEVDAKH

PREPRINT



SPECTRAL STUDIES OF SPACE STRUCTURE AND RF FREQUENCY DEPENDENCIES OF PLASMA PARAMETERS IN A SLAB α -DISCHARGE IN CO2+N2+He GAS MIXTURES

SPECTRAL STUDIES OF SPACE STRUCTURE AND RF FREQUENCY DEPENDENCIES OF PLASMA PARAMETERS IN A SLAB α-DISCHARGE IN CO₂+N₂+He GAS MIXTURES

V.V. Azharonok¹, I.I.Filatova¹, V.D.Shimanovich¹, V.N Ochkin², L.N.Orlov³, J.I.Necrashevich³, V.V.Nevdakh³

¹ Institute of Atomic and Molecular Physics, Skaryna prospect 68, Minsk 220072, Belarus

² P.N.Lebedev Physical Institute RAS, Leninski prospect 53, Moscow 119991, Russia

³ Institute of Physics of NASB, Skaryna prospect 70, Minsk 220602, Belarus

Abstract

The spectroscopic parameters of transversal RF discharge in gas mixtures $CO_2 + N_2 + He$ are investigated with space resolution. It has been observed that the width of the shields in a capacity RF discharge in such mixtures is almost independent on the frequency of RF field, so sufficiently differs from the standard theoretical predictions. The spatial distribution of the neutral gas temperature in various gas mixtures has been measured and discussed. The influence of the small additives of water vapour on luminescence intensity and its spatial distribution is also discussed.

I. Introduction

The high-frequency capacitive discharge (RFCD) at average pressure is widely used in a lot of various practical applications. Recently it has been reached a great progress in an engineering of electrical discharge CO₂ lasers with RF pumping as well as in the plasma-chemical reactors designing. For successful development of these devices and other fields of application it is necessary to have an adequate model of RFCD plasma. It, in turn, demands a reception of new experimental data about the features of RF discharge in various conditions; for instance, a spatial structure and spectral parameters of the planar RFCD plasma in molecular gases are not yet satisfactory investigated. In our previous works [2-4] the used technique and the outcomes of our experimental researches of the spectroscopic parameters of RF discharge in gas mixtures $CO_2 + N_2 + He$ are reduced in details. Here we shall stop on the new results of our research of parameters of RF discharge plasma that concern with the problem of shields width in such discharge and with the influence of the small additives of water vapour on a luminescence intensity and its spatial distribution in various gas mixtures.

2. Details of experiment

We have measured the spatial distribution of a luminescence intensity in various gas mixtures by the observing of a discharge structure along the transversal gap *L* of the discharge tube (DT). The discharge was excited at frequencies $f_{RF} = 5,28$, 40 and 81 MHz. The used scheme of a power supply ensured a possibility to excite in DT volumetric stable capacity discharge in mixtures $CO_2 + N_2 + He$ at pressure *P* from 1 up to 50 Torr and discharge currents *i* from 0,5 up to 2,6 A so as voltage drop *U* on electrodes DT was 100 -330V. The discharge was excited in the DT of a planar type with metallic water-cooled electrodes. The area of the electrodes was 40 x 50 mm, the breadth of the gap between electrodes was L = 3 - 5 mm. A sketch of our experimental set-up is shown at Fig. 1.

A structure of a RFCD was determined by the registration of an emission spectra intensity of bands for the (1-) system of N₂⁺, (2+) system of N₂ and by observation of other lines in a range of wavelengths 350-1000 nm. Diagnostics of a discharge plasma was carried out by methods of optical emission spectroscopy. The plasma radiation from the interelectrode gap was focusing with the help of a lens on an entering slot of grating spectrometer DFS-452. A video camera was used as a detector of radiation, that allowed to register two- dimensional distributions of an emission intensity I (L, λ) in coordinates «space (L) - spectrum (λ)» of a RFdischarge plasma with temporal and spatial resolution $\Delta t \sim 25$ ms and $\Delta L \sim 0.1$ mm correspondently. The video signal, generated by the video camera, was introduced through computer controlled videoblaster (as two-dimensional matrix I (L_m , λ_n) with dimension $m \ge n = 256 \ge 256$) into the base computer, where it was processed in view of an amplitude performance and an apparatus function of a spectra registration tract. The registration of survey spectra of a plasma emission was carried out photographically in a range of wavelengths from 250 up to 700 nm with a spectral resolution $\Delta\lambda \sim 0,001$ nm.

3. Results

A lot of emission intensity profiles I(L) are shown in Fig. 2. They are typical for an α - RF discharge with the dark zones of the space charge sheath at the electrodes [1]. In pure nitrogen at low pressure ($P \le 1$ Torr) I(L) distribution have a maximum displaced toward the powered electrode. At P > 5 Torr for all researched mixtures N₂ + CO₂ + He two maxima of a radiation was registered in spatial zones in the vicinity of the powered (RF) and grounded (\perp) electrodes. The visible luminescence intensity in the central «glow» part of the discharge is much lower than that we observed near the electrodes. But only in the central region of the electrode gap lasing conditions could be obtained.

The effect of a plasma luminescence localization in a neighborhood of electrodes could be connected with the preferential spatial localization of the processes of electron energy losing in the electrons - molecules inelastic collisions. Near the electrodes both the ionization and excitation of particles in a discharge happen mainly by oscillating electrons from a plasma column, which are in phase accelerated by a field of an uncompensated positive charge in the shields. At $P \le 0.5$ Torr in N₂ a large part of accelerated plasma electrons penetrates without essential energy losses into the central zone of the gap (because of greater length of electron's free run) and effectively excite many electronic levels of molecules.

The typical observed profiles I(L) (Fig.2) are quite similar to observed in [1] for combustion regime of RFCD.

As a rule they are consisted of the two dark bands near the electrodes (where the electron density is too low), so as two peaks of visible emission (usually connected with the spontaneous decay of the species excited by the electrons which are ejected from the electrodes and accelerated due to the characteristic positive ions shields) and the central «glow» part of the discharge; the visible luminescence intensity here is much lower, but only in this region the lasing conditions could be obtained. In pure nitrogen at low pressure ($P \le 1$ Torr) two peaks of intensity could overlapped and distribution I(L) should have maxima near the center of the interelectrode gap. At P > 5 Torr for all researched mixtures $N_2 + CO_2 + He$ two maxima of a radiation was registered in spatial zones contiguous to high-frequency (RF) and grounded (\perp) electrodes.

The ratio between the radiation intensities of the various components depends on frequency of a RF-field. In spectra of the discharge in mixtures N₂/CO₂/He at f_{RF} =5.28 MHz the bands of (1-) system of N₂⁺ are most intensive while the bands of (2+) system of N₂ practically are absent, except the discharge in mixtures N₂+He at P> 10 Torr in pure nitrogen where a luminescence of (2+) N₂ is intensive enough. For the discharge at f_{RF} =81 MHz the (2+) N₂ bands are reliably registered, but the (1-) N₂⁺ bands are hardly appreciable. The established distinctions are connected, on our sight, to features of the electron energy distribution function $f_e(\varepsilon_e)$ in RF discharges and meanings of electron's kinetic factors for the mixtures N₂/CO₂/He connected with the $f_e(\varepsilon_e)$ distribution in a RF field. In the discharge with f_{RF} =5.28 MHz the num- ber of electrons n_e with energy $\varepsilon_e > 16$ eV, (which are in need for excitation of electronic transitions $B^2\Sigma^+u - X^2\Sigma^+g$ in a N2⁺ molecular ion) is essentially exceeded the appropriate meanings of n_e with such high energy for RFCD with f_{RF} =81 MHz.

The similar effect was observed in [5], where they have measured a delay of a luminescence of bands (2+) in N₂ relative to (1-) bands in N2⁺. The authors of [6] have accounted the constants of excitation for the states N2 ($C^{3}\Pi_{u}$) and N₂⁺($B^{2}\Sigma^{+}_{u}$) in association with the parameter E/ω , where $\omega = 2\pi f_{RF}$ and E is the tension of an electrical component of a RF field. It is shown, that factor of excitation of a N2 ($C^{3}\Pi_{u}$) state is decreased more than on the order, while E/ω is increased from 10⁻⁷ up to 10⁻⁵ V•s/cm; at the same time the constant of ($B^{2}\Sigma^{+}_{u}$) state excitation of a N₂⁺ changes poorly.

We have estimated the meanings of E/ω for the RFCD. It was $E/\omega \sim 10^{-6}$ and $\sim 10^{-5}$ V··s/cm for f_{RF} =5.28 and 80 MHz correspondently. In this connection it is possible to consider, that the observable redistributions of the radiation intensity in the bands (2+) of N₂ and (1-) of N₂⁺ with variation of frequency are connected to the different excitation efficiency of N₂ ($C^{3}\Pi_{u}$) and N₂+($B^{2}\Sigma^{+}_{u}$) states. The occurrence of a luminescence in bands (2+) N₂ in the discharge at f_{RF} =5.28 MHz in mixtures without CO₂ (N₂+He or N₂ at P > 10 Torr) could be connected to the absence of suppression of both metastable states (2^{1,3}S) of HeI and and vibration levels of nitrogen in the $X^{1}\Sigma g^{+}$, v' ground electronic state by CO₂ molecules in these mixtures. As it well known these states participate actively in formation of a high-energy wing of the $f_e(\varepsilon_e)$ function of electrons distribution on energy in RFCD.

3.1. Width of shields in a RFCD in CO₂+N₂+He gas mixtures

The existence of some sharp areas near the electrodes, which differs from other regions of a discharge both on a luminescence intensity, and on plasma components is one of the most interesting feature of an α -phase RFCD. There is a great uncompensated ionic charge here (such layers of a positive spatial charge are named *shields*) [1, 4]. For the plasma-chemical reactors it is very promising region, where a lot of ionic processes are very intensive. On the contrary for gas lasers these regions are non-working and could be as narrow as it possible - the lasing could

exist only in a region between two boundary shields. But in both cases it is necessary to know the real shield's width for any plasma device optimization.

We have observed (Fig. 1) that the I(L) distributions are a little bit asymmetrical - the widths of the shields near the grounded (ΔL_{\perp}) and powered (ΔL_{RF}) electrodes are differed. The relation between ΔL_{RF} and ΔL_{\perp} also varies: $\Delta L_{RF} < \Delta L_{\perp}$ is realized at frequency f_{RF} = 5,28 MHz and $\Delta L_{RF} \approx \Delta L_{\perp}$ - at f_{RF} = 81 MHz. In a discharge in pure nitrogen a pressure raising up to 5 Torr is accompanied by growth of intensity of a luminescence in both zones irrespective of frequency f_{RF} . In a range 5 < P < 40 Torr intensity I(L) poorly depends on pressure, and at P > 40 Torr (f_{RF} = 5,28 MHz) it sharply descends. The sizes of both zones practically do not vary with growth of P. The increasing of a CO₂ content in a mixture N₂ + CO₂ from P_{CO2} = 0.5 up to 2 Torr (f_{RF} = 81 MHz, P_{N2} = 5 Torr) is accompanied by a drop of a discharge luminescence intensity in whole and diminution of ΔL_{RF} and ΔL_{\perp} widths (Fig.2 b). In a N₂ + He mixture (f_{RF} = 5,28 MHz, P_{N2} = 5 Torr) while the partial pressure of helium raises from 1 to 10 Torr the intensity in ΔL_{RF} and ΔL_{\perp} zones essentially increases, but their widths practically do not vary.

With growth of a current an intensity of plasma emission and widths of zones ΔL_{RF} and ΔL_{\perp} are increased for researched operating duties of a discharge. The registered modifications I(L), ΔL_{RF} and ΔL_{\perp} as a function of pressure of nitrogen, magnitude of a discharge current, partial content of CO₂ and helium could be connected to singularities of kinetics of level population and ionization of the molecules, present in a discharge, so as radiative and collisional deactivation of their exited electronic, vibrational and rotational states [7, 8].

The most unexpected feature of these plots is the observation that the spatial structure of a discharge essentially depends on frequency of RF field, but the width of the shields is almost independent on it. It sufficiently differs from the theoretical predictions. According to the elementary theory of RFCD [1] both the width of positive ions shields and its position from the electrodes are almost equal to the amplitude Δ_m of the electron oscillation at the RF field

$$\Delta_m = e E_0 / m v \omega, \qquad (1)$$

where E_{θ} is strength of a RF field, *e* and *m* are charge and mass of electrons, *v* is an effective number of electron collision.

As one could see from (1), the growth of gas pressure should reduce in magnification of collisions number in volume, and therefore - in a diminution of mobility of electrons; as a corollary, the calculated width of the shields owes varies in inverse proportion to pressure of gas. According to our measurements such dependence is not so strong and unambiguous (see, for example mentioned above dependencies $\Delta L_{RF}(P)$ and $\Delta L_{\perp}(P)$).

At the same time from the expression (1) it follows that the width of the shields has to monotonously grow with a diminution of a driving frequency, as it is

shown at Fig. 3 b by solid line. In a good accordance with this predictions the shield width association with the gas pressure in our experiments is quite close to reciprocal proportionality (fig. 3 a), but even in this case the dependence on frequency of excitation is extremely weak - in the certain contradiction with the simplest theory. This discrepancy is more evident from the experimental data shown at Fig. 3 b. We have observed that with rising of frequency the width of a luminescence maxima could even increased and the drop of intensity in central zone becomes less expressed. So as a rule the shield widths are constant or slowly increased with $\boldsymbol{\omega}$ instead of inversely proportional diminishing with $\boldsymbol{\omega}$, as a mention theory predicts.

The reason of so essential divergence of experimental outcomes with predictions of the theory is not yet clear. Partially it could be connected with incompetence of a model [9], especially in the case of relative low frequency (in this case a discharge condition are closer to the alternating current, then to the RF case; besides the existing theory does not take into account a lot of specific effects in molecular gases such as an electron attachment for mixtures containing

electronegative gases and so on). It is also necessary to define more precisely some details of experiment; in a nearest future we assume to investigate such dependencies in other gases (in particular - in pure inert gases) and in DT with the gap size variation.

Earlier similar regularities were not observed; there are 2 reasons of this. According to predictions of the theory [9] at rather low frequencies $\boldsymbol{\omega}$ the maxima of a luminescence intensity for ions N₂⁺ should be superimposed practically at any pressure and spatial distribution of intensity in DT with narrow gaps was not even investigated in any article. Besides in majority of works it was investigated only the spatial distribution of integrated plasma intensity, which could mask the information about shields' width as the intensity distribution for a neutral components of plasma luminescence is much smoother than in a band 391,4 nm.

All mentioned spatial nonuniformities could play an important role in the RFD pumped molecular lasers designing. In particular, the width of the shields, where lasing is impossible, put a natural limit on the transversal dimensions of laser active elements L - it could be at least

$$L > 2 \Delta_m. \tag{2}$$

According to the expression (1) it means that at rather low RF frequencies of a field the theoretically estimated transversal sizes of an active element have to be rather great (for instance for $\omega = 5,28$ MHz the boundary value of L for various gases at pressure of several Torr and strengths of a field of the order 10 V/ cm is usual not less then 2 - 3 cm). Most of modern CO₂ lasers are designed under these conditions (1). It reduced in a very low gain (a gain coefficient in a gas discharge lasers is proportional to 1/L) and in a diminishing of the optimal gas pressure which both could limited the output power and efficiency of such lasers.

With the help of obtained outcomes it is possible to solve a lot of designing problems and to realize a good laser conditions with essential smaller transversal sizes, that will allow to develop lasers with much higher gain, output power and efficiency under the relative low-frequency pumping. On our sight [10, 11] it opens new prospects for the development of novel high power gas lasers for various material processing and other applications.

3.2. Neutral component heating at RFCD plasma

It is known, that thermal stability of RFCD and efficiency of laser pumping with such discharge in many respects are determined by losses of input electrical energy on heating of active media which are described by the kinetical temperature T_g of the gas. For CO₂-lasers the optimum level of inversion of the active media is usually reached at $T_g < 600$ K [12, 13]. Unfortunately this quantity (that is widely used in a numerical modeling of RF pumped CO₂-lasers [26]) has not received in most experiments and could only estimate. As a rule a spatial distribution of the gas temperature in RFCD is unknown. The absence of data on T_g (*l*) distributions does not allow estimating gradients of the gain and distortion of laser beam structure caused by the thermal heterogeneity of the active media density.

In the present article the gas temperature distribution in various spatial zones of the planar RFCD in CO₂-N₂-He laser gas mixtures was experimentally determined for the first time. The gas kinetic temperature T_g has been deduced from the half-width $\lambda_{1/2}$ of rotationally unresolved spectral bands V - V' = 0-3, 0-4, 1-5, 0-5 of the second positive (2+) system of N₂. The algorithm of gas temperature T_g definition is based on comparison of measured $\lambda_{1/2}$ with its meanings $\lambda_{1/2}^*$, calculated from band contours of the appropriate modeling spectra, for various rotary temperatures T_{rot} in view of a real hardware function of the monochromator and registration setup [14, 15]. The dependence of $\lambda_{1/2}^*$ on T_{rot} for a band V' - V' = 0-5 of (2 +) system of nitrogen is given in fig. 4a. A halfwidth $\lambda_{1/2}^*$ is linearly increased with growth of the T_{rot} .

It is shown that the obtained from spectroscopic measurements rotational temperature T_{rot} values reflect the true gas kinetic temperature T_g . The distribution of molecules number on rotational levels j' of the N₂ ($C^3\Pi$) state is close to the Boltzman function

$$N_{j'} \sim \exp[-F_{j'}/T_{rot}]. \tag{3}$$

It is confirmed by the data in fig. 4b, where the measured relative quantity $\ln N_j$ are given for various spatial zones of an interelectrode gap in depending on energy $F_{j'} \sim j'(j'+1)$ of rotational levels. The absence of breaks in these distributions testifies to temperature uniformity of radiating volume in a direction of supervision. The opportunity of an identification T_{rot} with T_g follows from a parity $\tau_C/\tau_{RT} >> 5$

between the life τ_c time of $C^3\Pi$ state of N₂ (in view of its quenching at intermolecular interaction) and time τ_{RT} of its rotational (*R*-*T*) relaxation [14]. Estimated error of measurements ($\Delta T_g/T_g$) did not exceed 5 %.

The dependences $T_g(l)$ for RFCD at f_{RF} =5.28 and 81 MHz in pure nitrogen and $N_2/CO_2/He$ mixtures are given in fig. 5a,b. The received distributions T_g (l) are essentially non-uniform along the interelectrode gap. The structure of $T_g(l)$ depends on a discharge current, mixture composition and pressure of working gas, but poorly depends on the changes of excitation frequency. In a RFCD in pure nitrogen at P <15 Torr and reduced currents the distribution of temperature $T_g(l)$ along the gap is close to parabolic, with a maximum on distance $l \approx 0.4L$ from the RF-electrode. This temperature maximum increases with pressure of nitrogen growing from 1 up to 15 Torr (Fig. 5b). The magnification of current and pressure of nitrogen up to 20 Torr is accompanied by a growth of T_g in zones closed to electrodes (Fig. 5a). In a center of DC thus temperature does not vary, but the form of distributions $T_g(l)$ changes from the parabolic to two-humped with maxima of temperature located on the distances l $\approx 0.23L$ and $l \approx 0.17L$ from RF electrode and $l \approx 0.33L$ and $l \approx 0.4L$ from grounded electrode for $f_{\rm RF}$ =5.28 and 81 MHz accordingly. As a rule such transition to twohumped distribution of T_g (l) was accompanied with appreciable growth of temperature in a vicinity of electrodes (up to 600 K) (fig. 5a,b). In the central zone of the interelectrode gap T_g values change poorly (less, then ~200K). The temperature $\Delta T_g / \Delta l$ gradient on the distance from the shields region near the powered electrode, were the temperature reaches a maximal value, to the midgap is about 700 K/cm (f_{RF} =5.28 MHz at P_{N2} = 20 Torr, *i* =1.08 A).

In N₂ + CO₂ + He mixtures at P < 20 Torr a T_g (*l*) profile is similar to obtained for RFCD in pure N₂ at low currents and P < 20 Torr (Fig. 5c). The magnification of CO_2 partial pressure leads to T_g growing. In a triple mixture at P > 40 Torr and high current (up to 1 A) a value of the temperature near RF-electrode essentially increases, but in a center of DC it practically does not vary. The T(l) distribution in mixtures N₂: CO₂: He = 20 : 1: 20 (P = 41Torr) at f_{RF} = 5,28 MHz is characterized by a higher temperature in the center of DC and smaller one near RF-electrode on a comparison with the discharge in pure N_2 at P = 20 Torr. It can be observed in fig. 5a,c that the $\Delta T_g/\Delta l$ gradient in discharge in N₂/CO₂/He mixtures smaller than that in pure nitrogen discharge: $\Delta T_g / \Delta l \approx 350$ K/cm ($f_{\rm RF}$ = 5.28 MHz, N₂/CO₂/He =20/1/20, P = 41 Torr, i = 2.2 A) and $\Delta T_g / \Delta l \approx 180$ K/cm ($f_{\rm RF} = 81$ MHz, N₂/CO₂/He = 10/2/5, P = 17 Torr, i = 0.7 A). As a rule it is connected with the high-temperature zone expansion in a direction of the gap center. The meanings of temperature here depend on a ratio of a molecular component in a mixture. With increase of the CO₂ contents in a mixture $N_2/CO_2/He$ the gas temperature is growing (for instance, for mixtures N₂: CO₂: He =10 :2 :5 a changing P_{CO2} from 0.5 up to 3 Torr increases the T_g near the RF-electrode on 400 K). In $N_2/CO_2/He$ mixture with high partial He pressure $(N_2/CO_2/He = 20/1/20, P=41 \text{ Torr}, f_{RF}=5.28 \text{ MHz}) T_g(l)$ profiles are shown more higher T_g values in the center of the electrode gap and more lover ones near the

powered electrode on comparison with the discharge in pure nitrogen at P=20 Torr (Fig. 5a).

For all investigated regimes of discharge the asymmetry in distributions $T_g(l)$ takes place: as a rule T_g is maximal at the upper (RF) electrode. It could be caused both by the heterogeneity of energy input in the different parts of discharge [16], and by occurrence of convective thermal flows in gas connected to presence of gradients of temperature and gas density [17]. For the benefit of last in particular, the character of change of the $T_g(l)$ contour testifies: it depends on pressure of gas in the discharge chamber. At small pressure (< 5 Torr) a free molecular flow dominates, with the removal of heat from the volume due to diffusion of the excited molecules to walls with subsequent deactivation. In this case structure $T_g(l)$ is practically parabolic with a maximum at the DT center. At pressure more than 5 Torr the processes of ascendant thermal flows and free convection of heat begin to play an essential role.. It results in a displacement of a maximum temperature to the upper electrode. The further increase of pressure can result in an establishment of complex structure of $T_g(l)$, determined by alternation of ascendant and descendant flows in gas volume; it bringing to occurrence of specific cellular structure of temperature fields with a plenty of small maxima. But in general it also leads to increase of gas temperature in a vicinity of the top electrode [17].

The results of the present researches of gas kinetic temperature fields are used to account the spatial distribution of input electrical energy losses into the gas heating in plasma of RFCD (f_{RF} =40 MHz) in nitrogen and air at medium pressures [18]. The meanings of temperature, received by us, are close to the data of theoretical accounts T_g [19, 20] for the central zone of the gap. However the shape of the T_g (*l*) distributions in [19, 20]in all cases is close to a "parabolic", which differs from the experimental data. Such discrepancies are connected to features of physical model used in [19, 20].

The most probable channels of an enclosed electrical energy dissipation are considered on the base of calculation of an electrons energy balance in a gas discharge in N₂ + CO₂ + He [18, 19] with adding of obtained temperature distributions T_g (*l*) in a RFCD plasmas. So in pure nitrogen discharge or mixture N₂ + CO₂ + He at currents *i* < 0.5 A ($f_{RF} = 81$ MHz) and *i* < 1.8 A ($f_{RF} = 5.28$ MHz) the T_g temperature peaks near the electrodes does not registered (Fig. 5). In this conditions of the discharge operation the processes of *V*-*T* – relaxation of N₂($X^{1}\Sigma$, *V* ") molecules bring the main input into the gas heating. At the time an efficiency of the electrodes at higher currents shows a presence of a gas heating source in plasma, which is not compensated by heat conduction to the discharge chamber walls. Such a source, both with the deactivation of N₂ vibrational levels, may be the self-quenching of metastable $A^{3}\Sigma$ states of N₂:

$$2 \operatorname{N}_{2}(A^{3}\Sigma) \to \operatorname{N}_{2}(B^{3}\Pi) + \operatorname{N}_{2}(X^{1}\Sigma) + \Delta E$$
(4)

with the extraction of an energy ΔE in heat [21].

The heating of RFCD plasma in N₂ and mixtures N₂ + CO₂ + He in central zone of DT is determined by electrons elastic impacts with molecules, vibrotranslational relaxation of molecules and deactivation of vibration levels of N₂ (X, V"). It also reduces in lower values T_g in a center of DT on a comparison with near electrodes zones of a discharge. The drop of the temperature near the RF-electrode in the N₂/CO₂/He discharge with high partial helium pressure on a comparison with discharge in pure N₂ at *i*>1.8 A (Fig. 5b) is due to effective quenching of the N₂ $A^3\Sigma$ state by CO₂ and He molecules [22, 7]. Furthermore helium takes heat away to the discharge chamber walls. At the same time an observed raise of T_g in the gap center at adding of CO₂ and He to nitrogen is stimulated by the processes of elastic scattering of electrons on CO₂, He molecules and *V*-*T*-relaxation of CO₂ molecules.

Thus, in a mixture $N_2 + CO_2 + He$ the basic contribution to the heating of a plasma's neutral component in a RFCD in the center of the DT is brought by the processes of *V*-*T* relaxation of vibrationally excited N_2 and CO_2 molecules, but near the electrodes the significant role in the heating, alongside with *V*-*T*-relaxation of these molecules is played the processes of metastable states deactivation (mainly - $A^3\Sigma$ states of a molecule N_2).

The data of T_g measurements near the electrodes are in a good agreement with the results of our measurements of temperature of the electrodes, executed with the help of the thermocouple. The last testifies that the heating of electrodes is defined basically by the thermal flow of the plasma's neutral component to the electrode surface.

3.3. Influence of water vapor on RFCD plasma parameters

In obtained RFCD spectra both in pure nitrogen and its mixtures with He we usually have observed the intensive bands of OH (transition $A^2\Sigma-X^2\Pi$), as well H_a and H_β lines of hydrogen. Obviously, it is connected with the uncontrollable impurities of water in container with nitrogen used in our experiments. As a rule in case of N₂+He mixtures the band intensity of hydroxide molecules is much higher, than in the discharge in pure nitrogen. It is characteristic, that if we add a few Torr of water in an initial mixture, an OH-band intensity could appreciably diminished. It is necessary to note, that the influence of water vapor on CO₂ laser operation is well known [12, 23], but the role of H₂O dissociation in molecular plasma has not discussed. It is known only, that in many cases the hydroxide molecules formed during this dissociation are characterized by strongly nonequilibrium functions of distribution on internal degrees of freedom [24].

It is shown in [24], in conditions similar to our experiment, the dissociation from the basic state X^1A_1 of water molecule and the excitation of electronic states of OH (in particular $A^2\Sigma$) takes place due to direct electronic impact:

$$H_2O(X^1A_1) + e \rightarrow OH(X^2\Pi) + H + e$$
(5)

with the subsequent excitation

(6)
$$OH(X^2\Pi) + e \rightarrow OH(A^2\Sigma) + e.$$

It could be a result of dissociation reactions of molecules H_2O through the excited electronic states B^1A_1 :

(8)

$$H_2O(X^1A_1)+e \rightarrow H_2O(B^1A_1)+e,$$
 $H_2O(B^1A_1)\rightarrow OH(A^2\Sigma)+H.$

Both mechanisms are connected to the direct electronic impact. However in the processes mentioned above electrons of various energies E_e take part. A threshold meaning of E_e for processes of the first group is ~4 eV, of the second ~ 10 eV. As a result of processes of the first group is the occurrence in RFCD of "cold" OH(A² Σ) molecules with rotational temperature T_{rot} , close to T_g . The second group of processes is characterized by formation of ensemble of "hot" OH(A² Σ) molecules with $T_{rot} \sim 10\ 000\ K$. A thermalization of this high-energy ensemble can leads to increase of T_g and, as a result, to the reduction of gain in an active media.

Necessary condition for processes (7) - (8) is the existence in the discharge a group of electrons with high energy ($E_e > 10 \text{ eV}$). As it is shown in our paper [25], the surplus (in comparison with the direct current discharge) of such electrons could occur in RFCD with frequencies of excitation 150 -200 MHz, or in mixtures containing He (as a result of super elastic collisions of electrons with metastable atoms HeI ($2^{1,3}$ S) and processes of associative ionization of He):

$$\operatorname{HeI}(2^{3}S) + \operatorname{HeI}(2^{3}S) \to \operatorname{He}_{2}^{+} + e.$$
(9)

In this case deactivation of vibrationally excited (v', v'') nitrogen molecules in the ground electronic state $X^{1}\Sigma_{g}^{+}$ is accelerated at collisions of the second sort mixtures:

$$N_{2}(X^{1}\Sigma_{g}^{+}, v') + e(E_{e}^{'}) \rightarrow N_{2}(X^{1}\Sigma_{g}^{+}, v'') + e(E_{e}^{''}), \ E_{e}^{''} > E_{e}^{'}.$$
(10)

So the occurrence in the discharge of the electrons with $E_e > 10$ eV stimulates the processes (8) - (10). As a result we observe a radiation intensity increase in OH bands in comparison with the discharge in water vapor with nitrogen, where the

basic channels of OH ($A^2\Sigma$) formation are, on our sight, the processes (5), (6). For both case the distributions of intensities in OH bands is different as well: in H₂O+N₂ mixtures the radiation in a band has maxima, basically, in lines from rotationally levels j'≤10, while in mixes H₂O+N₂+He the significant share of radiation is connected with a site of a spectrum appropriate to transitions with 10< j' ≤30. As a rule the last is a characteristic attribute of presence in the discharge of "hot" OH ($A^2\Sigma$) molecules.

The observed changes in an integrated intensity of radiation in bands of OH in function of the CO₂ contents in mixtures can be interpreted as delay of processes (7), (8) in this case because of suppression of HeI metastable states by CO₂ molecules, and as decrease of the efficiency of process (10) in mixtures N₂+CO₂ in result of V-V'-exchange between molecules N₂ ($X^{1}\Sigma_{g}^{+}$, v') and CO₂ ($X^{1}\Sigma_{g}^{+}$, v'').

It is necessary to note, that under certain conditions of RFCD in gas mixtures with water vapor the dominant mechanism of H_2O dissociation can be the exothermic reaction with participation of the vibrationally-excited H_2O^* molecules [26]:

$$H_2O + e \to H_2O^* + e, \tag{11}$$

$$H_2 O^* \rightarrow H_2 + 1/2 O_2 - \Delta E, \qquad (12)$$

where $\Delta E \sim 2.6$ eV is a power threshold of the given reaction. In this case, as it was shown in [26], there are good conditions for cooling of molecular plasma. Temperature of electrons T_e could be reduced up to meaning as low as 0.2 eV, which is much lower than the temperature $T_e \sim 1.5$ -2 eV that is optimal for CO₂ lasers operation conditions.

Taking into account the important role of water and OH molecules in all plasma processes the given researches will be continued. Simultaneously at realization of experiments it is necessary to pay attention to necessity of elimination even of the smallest impurity of water vapor in initial gases.

3. Conclusion

First, it should be outlined, that the classical emission spectroscopy technique in combination with modern matrix detection schemes gives the possibilities of quantitative analysis of space structure of macroscopic plasma parameters even at elevated pressures. The studies of molecular spectra for different gas pressures and excitation frequencies can be attracted to understanding of space-temporal plasma dynamics in compact discharges structures.

Some of the observed features of RFCD could play an important role in the planning of applications of such discharge. Thus to the various purposes such as chemical reactors and laser devices the behavior of spatial structure of plasma is

rather essential. In the first case the most essential is to find the conditions, when the layers in RFCD are the narrowest and are close located to a surface of electrodes. In the second case, for creation of powerful compact gas lasers with transversal RF pumping, the interest represents the observed effect of the weak dependence of shields width of excitation frequency. It follows, that at smaller pumping frequencies it is possible to use considerably large volumes of gas. The same time, the opportunity to work at small width of shields means, that the large laser signal amplification is achieved also. Such combination provide more flexible compromise in between of a number of problems in such lasers designing and allows to expect for creation of lasers with the output power up to several kW at frequencies of excitation from 1 up to 10 MHz and to realize a good laser conditions with essential smaller transversal sizes, that will allow to develop lasers with much higher efficiency and output beam quality. On our sight some new prospects for the development of such lasers with low-frequency pumping could be realized by the engineering of the lasers with closed cycle of gas flow For development of such devices there can be very useful results of the chapter 3.3 connected to influence of the small additives of water vapor.

The same time some of our observations make the questions from physical point of view. Two of them seems be of the most importance. The first is the low dependency of volume charge sheets on electric field frequency in contradiction with standard RF discharges theory. The next one is the existence of the central deeps in the gas temperatures stationary space profiles. The only physical sense of such result is to propose the strong heat consuming processes in the bulk of the plasma. Such results have not been observed formerly for the DC discharges. These observations has to be studied in more details.

This work was supported in part by the grant of the Foundation for Fundamental Research of the Belarus (project $\Phi 02P-082$) and by the grant RBRF 02-02-81008 of Russia.

5. REFERENCES

- 1. Yu.Raizer, M.Shneider, H.Jatsenko. *The high-frequency capacitive discharge*. (Moscow, "Science.", 1995 in Rus.)
- 2. V.V.Azharonok, I.I. Filatova, L.N. Orlov, e.a., Proc. SPIE.-2000.- V. 4165.- p. 90-100.
- 3. V. V. Azharonok, I. I. Filatova, e.a. Proc. 18th Int. Symp. on the Physics of Ionized Gases, Kotor (1996) 428-431
- V. V. Azharonok, I. I. Filatova, V. D. Shimanovich, L.N.Orlov, JAS, <u>68</u>, N5, 2001, p. 634-638
- 5. W. T. Armstrong, R. A. Roussel-Dupre, R. Karl et al., Proceedings ICPIG-18, Swansea, 1987, p.850
- 6. O. Ivanov, S. Lirin., Plasma physics, <u>18</u>, 1992, p. 124-127(in Russian).

- 7. D.I. Slovetskij. *Mechanisms of chemical responses in non-equilibrium plasma*.(Moscow "Science", 1980 in Rus.)
- 8. N.A. Jatsenko, J. of Techn. Physics, <u>51</u>, 1981, p. 1195 (in Russian).
- 9. Yu. P.Raiser. *Physics of Gas Discharge*. (Moscow, "Nauka", 1987 in Rus.)
- 10. L.N. Orlov, V. V. Azharonok , J.Appl.Spectr., <u>66</u>, №5, 1999, p..715-718 (in Russian).
- 11. L.N. Orlov, V. V. Azharonok, Abstracts of the XIV ICPC, 1-8.08.1999, Prague (Czech).- p. 39.
- 12. I.I. Wedenov. *Physics of high-flow-rate CO₂ lasers* (Moscow, 1982 in Rus.)
- 13. K. M. Abramski, A. D. Golley, H. J. Baker, D. R. Hall, IEEE J. Quant. Electronics, <u>52</u>, 1996, p. 340- 345.
- 14. V Nevdakh., L.Orlov, J. Techn. Phys. (Poland), <u>40</u>, No.3, 1999, p. 221-224.
- 15. V. V. Azharonok, V.V. Gurashvili, V.N. Kuzmin e.a., Plasma physics, <u>19</u>, 1993, p. 903-909 (in Russian).
- 16. S. Stamou, D. Mataras, D. Rapakoulias, J. Phys. D: Appl. Phys., <u>31</u>, 1998, p. 2513-2520.
- 17. O.G. Martynenko, Ju.A. Sokovishin. *Free-convective heat transfer* (Minsk, "Science & Technics", 1982, in Rus.).
- 18. A. Kovalev, L. Rahimov, N. Suetin, V. Feoktistov, Proc. III Conf. on Gas Discharge Physics, Kiev, 1986, p. 107.
- 19. A. A. Kuznetsov, M. Z. Novgorodov, V. N. Ochkin, e.a., Proc. SPIE, <u>4165</u>, 2000, p. 140-149
- 20. B. I. Ilukhin, Yu. B. Udalov, I.V.Kochetov e.a., Appl. Physics, <u>B 62</u>, 1996 p. 113-127.
- 21. A. Berdyshev, A. Vicharev e.a. High Temp. Physics, <u>26</u>, 1988, p. 661-666 (in Russian).
- 22. J.W. Dreyer, D. Perner, C.R. Roy. J. Chem. Phys., 61 (1972) 3164.
- 23. D.Hall, C.Hill. In: *Handbook of Molecular Lasers*/ ed.by P.Cheo, M.Dekker, 1987.
- 24. V. N. Ochkin, S.J.Savinov, N.N.Sobolev., Proceedings of FIAN, 157, 1985., p. 6-85 (in Russian).
- 25. L.Orlov, S.al-Havat.// Publ.Observ. Astron. Belgrade, 1996, №53 p.137-139.
- 26. K.Smith, R.M. Thompson. *Computer Modelling of Gas Laser*.(New York: Plenum, 1978).



Fig. 1. The basic circuit of measuring installation: vacuum chamber (1), discharge chamber (2), camera lens (3), monochromator (4), CCD-camera (5), computer and monitor (6).



Fig. 2. Intensity distribution of the N₂⁺ band emission at $\lambda = 391.4$ nm of RFCD along interelectrode gap L: a – pure nitrogen, i = 0.7 A; b – gas mixtures, i = 0.7 A.



Fig. 4. Test graphs for the rotational temperature measurements



Fig.5a. Temperature profile $T_g(l)$ in a discharge gap at f_{RF} =5.28 MHz (pure N₂ at i = 0.42A (\Box) and 1.08 A (\bullet); mixture N₂/CO₂/He = 20/1/20 at i = 1.84 A (\Box), 2.2 A (\bullet)).



Fig.5.b. Temperature profile $T_g(l)$ in a discharge gap at $f_{RF} = 81$ MHz (pure N₂ at i = 0.42 A (\Box) and 1.08 A (\bullet); mixture N₂/CO₂/He=20/1/20 at i = 1.84 A (\Box), 2.2 A (\bullet)).



Fig. 5.c. Temperature profile $T_g(l)$ in a discharge gap at $f_{RF} = 81$ MHz for triple mixture.