# ELECTRON-BEAM SUSTAINED GLOW DISCHARGE IN A N<sub>2</sub>+CO GAS MIXTURE AT CRYOGENIC TEMPERATURE

V.V.Azharonok, <u>I.I.Filatova</u>, N.I.Chubrik, V.D.Shimanovich, V.A.Gurashvili\*, V.N.Kuzmin\*, N.G.Turkin\*, A.A.Vaselenok\*

Institute of Molecular and Atomic Physics of Belarus Academy of Sciences, F.Scaryna Avenue 70, Minsk, 220072, Belarus \*Troitsk Institute of Innovative and Thermonuclear Investigations, Troitsk, Moscow Region, 142052, Russia

### Abstract

A quasi-continuum electron-beam sustained glow discharge in a flow of  $N_2$ +CO gas mixture at cryogenic temperature was studied by the emission spectroscopy methods. Effective values of electron-ion recombination and rate of electron adhesion to electronegative molecules (Fe(CO)<sub>5</sub>, Ni(CO)<sub>4</sub>, H<sub>2</sub>O) present in the discharge have been determined in dependence of reduced electric field strength E/N.

#### Introduction

A non-self-sustained glow discharge in a flow of molecular gases ionized by a high power electron beam is widely used to excite active medium of plasmachemical reactors and powerful CO-lasers due to the high degree of the vibrational excitation of molecules in the discharge plasma. Degree of vibrational non- equilibrium of the discharge plasma depends in many respects on plasma's neutral component temperature  $T_g$  as well as on free electron loss processes as a result of electron-ion recombination and electron adhesion to electronegative molecules. An influence of impurities of O<sub>2</sub>, H<sub>2</sub>O, C<sub>3</sub>F<sub>6</sub> molecules on plasma conductivity in CO-laser was studied at T<sub>g</sub>=100 K [1]. Electron adhesion processes to Fe(CO)<sub>5</sub>, Ni(CO)<sub>4</sub> molecules was considered at T<sub>g</sub>=300 K [2].

The present work is devoted to investigations of gas temperature  $T_g$  fields in a non-self-sustained glow discharge in a flow of N<sub>2</sub>:CO= 10:1 mixture at cryogenic temperature and to studies of the processes determining discharge plasma vibrational non-equilibrium.

# Experiment

The experimental apparatus have been described in detail in our previous publication [3]. The discharge was operated in a discharge chamber with an equipotential netted anode and a segmented cathode disposed at an angle of  $6^{\circ}$  to each other. A distance between electrodes L at the chamber entrance was equal to 5 mm. The gas mixture was pre-ionized by electron beam with energy  $E_e$  of 100 keV. Values of total pressure and speed of the mixture pumped through the discharge chamber were equal to 60 Torr and 100 m/s, correspondingly. Gas

temperature  $T_g$  of the mixture at the chamber's entrance was maintained at 100 K. The discharge duration was varied in the range of 0.2-20 s.

Emission spectra were registered in the range of  $\Delta\lambda$ =200-700 nm with spectral resolutions of 0.1, 2.4 nm and a spatial resolution of ~0.1 mm. To measure gas temperature, a method was used [3] of registering the unresolved rotation structure of an electronic-vibrational bands of diatomic molecules. As pirometric bands, those of the 2+ system of N<sub>2</sub> and 1- system of N<sub>2</sub><sup>+</sup> were used.

Effective values of electron-ion recombination  $\beta_{ei}$  and rate of electron adhesion  $v_e$  have been determined in the range of reduced electric field strength variation  $E/N=(0.8 - 1.2) \cdot 10^{-16} \text{ V cm}^2$  using the balance equation

$$q_e = v_e \cdot n_e + \beta_e \cdot n_+ \cdot n_e$$

(under condition of  $n_{-} << n_{e}$ ), where  $q_{e}$  is the measured value of the rate of working medium ionization by the electron beam;  $n_{-}$ ,  $n_{+}$  and  $n_{e}$  are the concentrations of negative ions, positive ions and electrons, correspondingly.

#### **Results and discussion**

The N<sub>2</sub>+CO plasma spectrum in the investigated wavelength range  $\Delta\lambda$  includes mainly molecular bands of N<sub>2</sub> second positive, N<sub>2</sub><sup>+</sup> first negative, CN violet, C<sub>2</sub> Svan systems, NO  $\beta$ -system and FeI, NiI atomic lines. The most likely mechanisms of NO, CN and C<sub>2</sub> molecule formation in plasma are the interactions between vibrationally excited N<sub>2</sub>(X,V"), CO(X,V") molecules and products of their dissociation leading to degradation of N<sub>2</sub>+CO mixture. Presence of FeI and NiI atoms in plasma testifies that in the discharge the decomposing takes place of Fe(CO)<sub>5</sub> and Ni(CO)<sub>4</sub> molecules which are usually formed in metal balloons at CO pressure of approximately 100 atm.

Axial distributions of radiation plasma intensity and gas temperature  $T_g$  are ingomogeneous (fig. 1, 2). The temperature  $T_g$  values are equal to 670 K near the

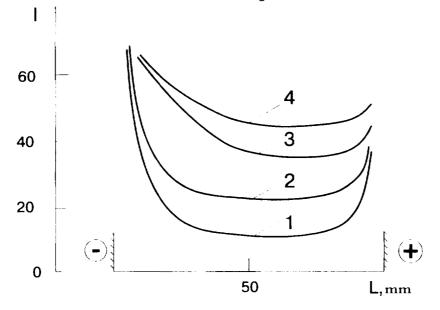


Fig. 1. The emission intensity of the non-self-sustained discharge  $N_2$ +CO plasma along the electrode gap: molecular bands of  $N_2$  (1);  $N_2^+$  (2);  $C_2$  (3), CN (4)

cathode, 500 K near the anode and 120 K in the center of the electrode gap (fig.2). In the vicinity of electrodes the discharge energy goes mainly to excitation of electronic states of  $N_2$ ,  $N_2^+$ , CN,  $C_2$  molecules (fig.1).

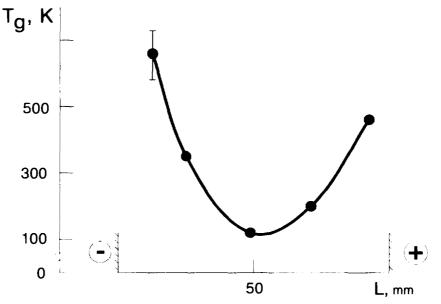


Fig. 2. The N<sub>2</sub>+CO plasma's neutral component temperature  $T_g$  distribution along the electrode gap

The molecules can be deactivated by quenching at intermolecular collisions followed by essential heat releasing. In the central zone of the electrode gap the main portion of the discharge energy is utilized to excite vibrational levels of N<sub>2</sub> and CO molecules ground state  $(X^{1}\Sigma)$ . Therefore, the radiation intensity of N<sub>2</sub>, N<sub>2</sub><sup>+</sup> molecules is lower in the center of the electrode gap rather then near electrodes. By contrast with it the radiation intensity of CN and C<sub>2</sub> molecules does not practically change along L. This is due to high efficiency of quasi-resonant energy exchange between vibrationally excited N<sub>2</sub>(X,V">9), CO(X,V">10) molecules and C<sub>2</sub>(X, V"=0), CN(X, V"=0) molecules being in the ground electronic state [4, 5]:

$$CO(X,V"\geq 10) + C_2 \rightarrow CO(X,V") + C_2(d^3\pi),$$
  

$$CO(X,V"\geq 10) + CN \rightarrow CO(X,V") + CN(B^2\Sigma),$$
  

$$N_2(X,V"\geq 12) + CN \rightarrow N_2(X,V") + CN(B^2\Sigma).$$

Evaluations of energy defects between N<sub>2</sub>(X,V") and C<sub>2</sub>( $d^3\pi$ , V') have allowed us to establish that the following mechanism of quasi-resonant excitation of C<sub>2</sub>( $d^3\pi$ ) molecules is possible in the discharge in the N<sub>2</sub>+CO mixture:

$$N_2(X,V''\geq 9) + C_2 \rightarrow N_2(X,V'') + C_2(d^3\pi).$$

Inhomogenety of molecular bands radiation intensity and gas temperature  $T_g$  gradient along the electrode gap can lead to rise of the discharge instability and to an exchange of environment optical properties in the direction of electric current. In the case of discharge duration increasing (20 s) a widening of the near-anode zone to the discharge center has been observed. Simultaneously the

stratification of the discharge along the electrode gap near the anode took place. There were three characteristic zones of plasma radiation intensity with dimensions of ~5 mm each. The discharge stratification was accompanied by modulation of electric current and voltage with a frequency of ~0.25 Hz. The most probable cause of the discharge stratification is the local reduction of the plasma conductivity resulting from processes of losing free electrons due to electron adhesion to electronegative molecules of impurities (Fe(CO)<sub>5</sub>, Ni(CO)<sub>4</sub>, H<sub>2</sub>O) present in discharge and molecules of NO formed in plasma.

The table presents the  $\beta_{ei}$  and  $\nu_e$  values measured at  $T_g = 120$  K in the N<sub>2</sub>+CO mixture previously purified from the impurities down to the level of P\*/P~1 ppm, where P\* is a partial pressure of the impurities.

P*/P, ppm	<u> </u>	1		>50
$E/N \cdot 10^{-16}, V \cdot cm^2$	0.6	0.7	0.8	0.8
$v_{e} \cdot 10^{4}, s^{-1}$	1.5	1.3	1.2	300
$\beta_{ei} \cdot 10^{-8}, \text{ cm}^2 \cdot \text{s}^{-1}$	0.6	0.7	0.8	60

For comparison the data of free electron lose rates in non-purified ( $P^*/P > 50$  ppm) cryogenic mixture are given. At  $T_g=120$  K the value of  $\beta_{ei}$  is the order of magnitude higher than that at  $T_g=300$  K [2]. Purification of the mixture (from Fe(CO)<sub>5</sub>, Ni(CO)<sub>4</sub> impurities) makes electron adhesion rate two orders of magnitude lower and allows specific energy contribution into discharge to be W/G=300-350 J/g at electron beam current density of  $j_e=(7-12)$   $\mu$ A/cm<sup>2</sup>. To achieve the same values of W/G for non-purified mixture the value of  $j_e=2$  mA/cm<sup>2</sup> is required.

## Conclusion

Processes of quasi-resonant energy exchange between CO and  $N_2$  molecules and CN,  $C_2$  radicals formed in the electron-beam sustained glow discharge in a flow of CO+N<sub>2</sub> mixture at cryogenic temperature play a vital role in the reduction of the vibronic excitation degree of the mixture.

Presence of electronegative molecules  $Fe(CO)_5$ ,  $Ni(CO)_4$  in the discharge plasma and the gradient of the plasma's neutral component temperature ( $\Delta T_g \sim 550$  K) along the electrode gap lead to the decrease of plasma conductivity and to the discharge stratification in the near-anode zone where the discharge sustained electron beam enters the discharge chamber.

- [1] Shashkov V.M., Shchekotov E.Yu.: Rep. of AS USSR, 306 (1989) 1397 (in Russian).
- [2] Center R.E.: J. Appl. Phys., 44 (1973) 3538.
- [3] Azharonok V.V., Gurashvili V.A., Kusmin V.N. at all.: Plasma Phys. Rep., **19** (1993) 469. [4] Grigor'yants G.M., Dymshits B.M., Ionih Yu.Z.: Optics and Spectroscopy, **65** (1988) 766
- (in Russian).
- [5] Taieb G., Leday F.: Canad. J. Phys., 48 (1970) 1956.