USE OF SHORT-WAVELENGTH TECHNIQUE IN SMM AND MM RANGE FOR DIAGNOSTICS OF NONEQUILIBRIUM PLASMACHEMICAL UHF DISCHARGE AT MODERATE PRESSURE IN CO_2

A.V.Golubev, V.G.Zatsepin and E.A.Tishchenko,
Institute for Physical Problems, Moscow, USSR

A.K.Vakar and E.G.Krasheninnikov,
Institute of Atomic Energy, Moscow, USSR

INTRODUCTION

Dissociation of carbon dioxide up to CO and O_2 with the use of CO for producing of H_2 /1/ is one of the most important plasmachemical processes. Energetic efficiency of this process in the case of slightly ionized plasma essentially depends on the mechanism of dissociation of CO_2; the maximum energetic efficiency in the nonequilibrium plasma ($T_e > T_v > T_0$, where T_e, T_v, T_0 - electron, vibrational and translational gas temperatures, correspondingly), is achieved by dissociation of CO_2 via vibrational excitation of molecules in the ground electron state /2/ . The efficiency of this mechanism depends on thedegree of the plasma ionization, determining the pumping rate of the vibrational degrees of freedom of molecules and parameter E_{eff}/N (E_{eff} is effective electric field in plasma, N - gas density), determining the energetic balanse of electronic energy in plasma. This data can be received by probing the plasma with the transverse electromagnetic waves.

EXPERIMENTAL

This work is devoted to investigation of electron component parameters of stationary UHF plasma discharge at moderate pressures (p=50 ÷ 250 torr) in CO_2.Fig.1 presents the scheme of reactor chamber. We have used the UHF source (c.w. magnetron) which excites the TE_{10}-mode in a rectangular waveguide (90 x 45 mm) (frequency of oscillations - 2,4 GHz ; radiated power 2,5 kWt). Discharge was created in a quartz tube (8) (28 mm i.d.) transversing the waveguide (1) perpendicularly to its wide walls. Gas entered the reactor through the aperture (5), that provides the tangential movement of stream around the reactor axis to stabilize the discharge. For side-on plasma probing rectangular holes (20 x 40 mm) were cut in the narrow walls of waveguide. They were closed with one-dimentional wire grids (2) (period 0,2 mm) to prevent the UHF field leakage. Experiments were conducted at gas pressures p= 50 ÷ 250 torr and specific plasma energy $q_v = W_p/Q = 3 \div 5$ J/cm^3(here W_p is the power absorbed in plasma, Q is the expenditure of CO_2).

Diagnostic installation (Fig.2) consists of two identical homodyne Mach-Zehnder interferometers with a backward-wave tube (BWT) as a source. One of them formed by polarizers P_2, P_4 and mirrors M_1, M_2 (called below as N1) was used for plasma probing; the other (number 2) consisting of polarizers P_2, P_3 and mirrors M_1, M_3 served as BWT frequency discriminator of the automatic frequency control (a.f.c.) system. Quasy-optical beam of radiation formed by lens L_1 and polarised by P_1 in a horisontal plane entered the interferometer through the polarizer P_2. One-dimentional grids P_2- P_5 were used to split the beams so that plasma was probed with a horisontally polarised wave, focused by lenses (7) (see Fig.1). This wave easily penetrates through the shield grids of reactor chamber. The polarizers P_6 and P_7 used as mixers of the ortogonally polarised waves superposed with P_4, P_3 and detectors D_1, D_2 (n-InSb at 4,2 K) are placed at the outputs of interferometers. The wavelength of the BWT radiation was modulated by sawtooth law with a frequency of 30 kHz. Amplitude of modulation is chosen so that wave phase shift is transfered onto the first harmonic of modulation frequency. The signal of first harmonic was selectively amplified and used for phase-amplitude analysis with phase-voltage converter and linear detector. Phase and amplitude of the beam transmitted through plasma were recorded as functions of discharge displacement across the probing beam. The width of the beam was defined by relative apertures of lenses L_2 and L_3; it was about 3,5 mm at λ =1,7 mm (λ is the wavelength of probing radiation).

PHASE STABILIZATION

To analyse the phase stability of measuring interferometer in the absence of plasma we consider the installation as a chain of converters, characterised by the definite transfere coefficients. BWT is a converter of collector voltage U into radiation frequency ω . Interferometer N1 with a detector D_1 transform the frequency ω into the phase shift of the beat signal first harmonic $\varphi_1 = \ell_1\omega/c$ in respect with the modulating signal, where ℓ_1 =250 cm is the difference between the optical lengthes of channels, C is light speed. Phase instability

$$\Delta\varphi_1 = \frac{\ell_1 \Delta\omega}{c} + \frac{\omega \Delta\ell_1}{c} \qquad (1)$$

is determined by the frequency drift $\Delta\omega$ of BWT radiation, caused by d.c. supply voltage U fluctuations, and interferometer base instability $\Delta\ell_1$ due to the external conditions changes (atmospheric pressure, humidity etc), resulting in fluctuations of air permittivity. Frequency of BWT radiation is stabilized by means of feedback circuit, including reference interferometer N2 with the base ℓ_2 , detector D_2, phase-voltage converter (with the transmittance K_{φ_2}) and coupling unit (with gain K_c), forming the feedback signal to controle BWT collector voltage (Fig.2). Frequency instability is equal to

$$\Delta\omega = K_\omega \Delta U = \frac{K_\omega \Delta U_{0\,eqv}}{1+K} - \frac{K_\omega K_{\varphi_2} K_c \frac{\omega}{c} \Delta\ell_2}{1+K}, \qquad (2)$$

where $\Delta\ell_2$ is the reference interferometer base instability due to the external conditions changes, $\Delta U_{0\,eqv}$ are all internal a.f.c. system instabilities, $\mathcal{K}_\omega$ - differential transmittance of BWT, $\mathcal{K}=\mathcal{K}_\omega \mathcal{K}_{\varphi_2} \mathcal{K}_c \ell_2/c$ - the feedback loop gain. Substituting $\Delta\omega$ into (1), we obtain for $\mathcal{K}=400 \gg 1$:

$$\Delta\varphi_1 = \frac{\ell_1}{\ell_2}\,\frac{\Delta U_{0\,eqv}}{\mathcal{K}_c \mathcal{K}_{\varphi_2}} + \frac{\omega \ell_1}{c}\left(\frac{\Delta\ell_1}{\ell_1} - \frac{\Delta\ell_2}{\ell_2}\right). \qquad (3)$$

Phase instability is determined by the differential drift of interferometers N1 and N2. Thus, if $\ell_1 \approx \ell_2$ then the interferometer drifts are partially compensated because the external condition changes equally influence both interferometers.

Influence of voltage instabilities $\Delta U_{0\,eqv}$ can be substantially eliminated by means of a balance method, i.e. by measuring the interferometer phase shift difference $\varphi_1 - \varphi_2$ (Fig.1, switch in position 2). In this case phase drift $\Delta(\varphi_1-\varphi_2) = \Delta\varphi_1 - \Delta\varphi_2$ where $\Delta\varphi_2 = \Delta U_{0\,eqv}/\mathcal{K}_c \mathcal{K}_{\varphi_2}$ - the reference interferometer phase instability, which is equal to

$$\Delta\varphi_1 - \Delta\varphi_2 = \frac{\ell_1 - \ell_2}{\ell_2}\,\frac{\Delta U_{0\,eqv}}{\mathcal{K}_c \mathcal{K}_{\varphi_2}} + \frac{\omega \ell_1}{c}\left(\frac{\Delta\ell_1}{\ell_1} - \frac{\Delta\ell_2}{\ell_2}\right). \qquad (4)$$

The phase influence of $\Delta U_{0\,eqv}$ is vanishing for $\ell_1 \approx \ell_2$. The use of the balance method of phase measurement combined with an automatic BWT radiation frequency control increased the phase sensitivity of the interferometer to 10^{-2} radn that permits to measure plasma electron densities in the range of $n \cdot 2a = 10^{10} - 10^{14}$ cm^{-2}.

RESULTS

Fig.3 presents the experimental curves of phase and modulus of microwave beam transmittivity for one of the plasma regimes. Phase shift in plasma is determined as a difference between curves 2 and 1 (curves 1 correspond to the quartz tube without plasma). The plasma transmittivity is obtained by division of respective amplitude functions. Characteristic diameter of plasma discharge $2a$ was taken as a distance between the phase profile points with $\Delta\varphi = \Delta\varphi(0)/2$, where $\Delta\varphi(0)$ is a phase shift at the plasma diameter. The pressure dependences of n and ν_{eff} on plasma axis under constant specific energy q_v are shown in Fig.4.

At the constant specific energy $q_v = 3{,}5$ J/cm^3 plasma diameter substantially depends on gas pressure. At pressures p=50 ÷ 100 torr the discharge has the diffused features ($2a \simeq 18 \div 14$ mm). For p $\gg$ 150 torr plasma diameter sharply diminishes to 5 ÷ 6 mm. This transition is accompanied by substantial increase of n (see Fig.4) and slower change of ν_{eff} , that causes the growth of plasma conductivity. In the whole range of the investigated plasma parameters ν_{eff} was greater than Ω (magnetron frequency $\Omega = 1{,}5 \cdot 10^{10}\,s^{-1}$). Thus, the real part σ_r of plasma complex conductivity

$$\sigma = \sigma_r + i\sigma_i = \frac{ne^2}{m}\,\frac{\nu_{eff}}{\Omega^2 + \nu_{eff}^2} - i\,\frac{ne^2}{m}\,\frac{\Omega}{\Omega^2 + \nu_{eff}^2}$$

is greater than $|\sigma_i|$ and UHF energy absorption is due to active losses; the specific power equals to

$$W_1 = \sigma_r E_{eff}^2 = \frac{ne^2}{m} \frac{E_{eff}}{\nu_{eff}} . \qquad (5)$$

Eqn.(5) provides determination of E_{eff} by the measured values of n, ν_{eff} and W_1; the translational temperature of plasma discharge /3/ at given gas pressure was used to evaluate parameter E_{eff}/N.

At the specific energy q_v =3,5 J/cm^3 the maximum energetic efficiency of CO_2 decomposition ($\eta \simeq 80\%$) was obtained for p=120 torr /3/ when E_{eff}/N =$(2 \div 3)\cdot 10^{-16}$ V·cm^2 and n/N = = $(4 \div 5)\cdot 10^{-6}$. Under this conditions the main part of UHF power is spent on the excitation of antisymmetric vibrational mode of CO_2 molecule /4/ . Degree of ionization n/N =$(4-5)\cdot 10^{-6}$ exceeds the theoretical minimum value n/N =$3\cdot 10^{-6}$, which corresponds to effective dissociation of CO_2 via the vibrational excitation of molecules in the ground electron state /2/.

Thus, the measurements of plasma electron component parameters confirms the essential participation of vibrational excitation in the process of CO_2 dissociation at the plasma regimes being investigated.

REFERENCES

(1) I.G.Belousov, V.A.Legasov, V.D.Rusanov,"Voprosy atomnoy nauki i tehniki"(IAE, Moscow, 1977), 2(3), p.158-163.

(2) V.D.Rusanov, A.A.Fridman, G.V.Sholin, JTP(Journ.of Tech. Phys., in Russian), 49, 2169 (1979).

(3) Yu.P.Butylkin, V.K.Zivotov, E.G.Krasheninnikov, JTP, 51, 925, (1981).

(4) I.V.Kochetov, V.B.Pevgov, L.S.Polak et al., "Plasmohimicheskie protsessy", edited by L.S.Polak (Moscow, "Nauka", 1979), p.4-43.

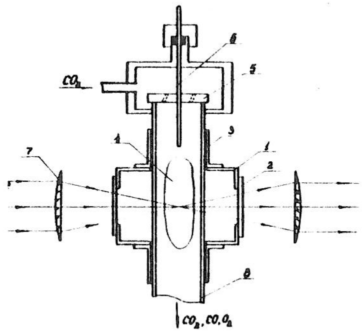

Fig.1 Plasmachemical reactor

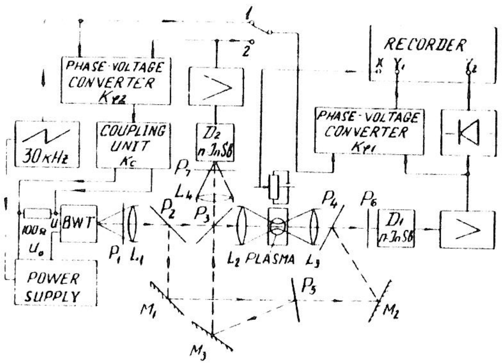

Fig.2 Scheme of interferometer. P_1-P_7 - one-dimentional grids (period 0,06 mm); L_1-L_4 polyethylen lenses (diameter 54 mm, focal distanse 140 mm); M_1-M_3 mirrors. Dashed lines-microwave radiation, solid lines - electrical circuits.

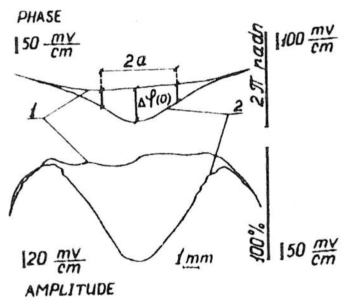

Fig.3. Experimental phase and modulus of trasmission coefficient (p=150 torr; q_v = 3,5 J/cm^3).

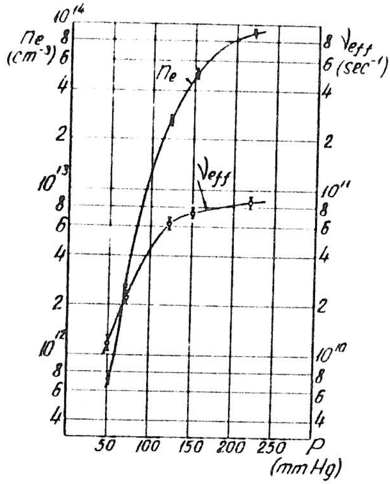

Fig.4. Dependence of electron density (n) and effective collision frequency (ν_{eff}) at the discharge axis on the pressure for constant q_{ν} .