

KINETICS OF MOLECULES, RADICALS AND CHARGED PARTICLES IN A STATIONARY PLASMA-BEAM DISCHARGE

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ABSTRACT

The work gives the results of solving a self-consistent problem permitting to calculate macroscopic properties of electrons, molecules and also relative concentrations of radicals and charged particles being formed in a plasma-beam discharge depending on the parameters of the discharge. Experimental data of the formation of hydrocarbon radicals in a discharge are presented also in this paper.

1. KINETICS OF ELECTRONS AND MOLECULES

As shown in /1/ the kinetic equation for electrons in a plasma-beam discharge at a high density of neutral particles has the form:

$$\frac{\partial f}{\partial t} = \frac{W}{n_e} D_e(f) - St(f) - D_z(f) \quad (1)$$

where $f(\epsilon)$ is the electron energy distribution function, W - is the energy volume density of Langmuir oscillations; n_e - is the electron concentration.

The first right part member of eq-n (1) describes the electron heating in the Langmuir oscillations field, the second shows the interaction of electrons with particles of weakly ionized gas, the third - the loss of electrons. Since the member showing the interaction of electrons with gas particles depends on their quantum state, then along with eq-n (1) it is necessary to solve a system of eq-ns describing the quantum states distribution function of the particles. Since many chemical reactions proceed with the participation of oscillatorily excited molecules, we shall first of all be interested in the kinetics of oscillatory levels. In the case of two-atomic molecules it is described by the following system of eq-ns /2/

$$\frac{\partial F}{\partial t} = Q(F, F) - P(F) + n_e \Psi(F, f) - D(F) \quad (2)$$

where $F(V)$ is the oscillatory levels distribution function of molecules. The first right-hand member of (2) describes a one-quantum exchange $V-V'$, the second - one-quantum processes $V-T$ - relaxation; the third - the process of electron interaction with molecule oscillatory levels, the fourth - the gas flow through the discharge zone. It may be shown that in the stationary case the character of the processes taking place in the discharge zone is defined by the following parameters:

$W/\rho_0, \rho_0 \tau_0, \rho_0 L$ where ρ_0 is the neutral gas pressure, τ_0 is the characteristic time the working gas spends in the discharge zone, L is the characteristic discharge dimension that defines the rate of ambipolar diffusion. It should be noted that eq-n (1) turns out to be principally nonlinear with respect to the distribution function f , as the electron heating rate is proportional to $1/n_e$, and n_e is defined by the electron number balance equation.

The system eq-n (1,2) has been solved with the help of a specially developed multistep iteration method. The calculations are performed for molecular hydrogen.

Fig.1 shows the dependence of the ionization degree $\alpha = n_e/n_0$ the mean electron energy \bar{U} and the value $W/n_e \tau_0$ on the parameter $\rho_0 \tau_0$ at $W/\rho_0 = 10^{-7}$ J/cm³. Torr and $\rho_0 L = 10$ Torr.cm.

Fig.2 gives the oscillatory levels distribution function of molecules in the saturation regime for the values $W/\rho_0 = 10^{-8}, 10^{-7}$ J/cm³.Torr. The dependence of the oscillatory energy ϵ_v and the loading efficiency of the oscillatory levels η^v on the parameter $\rho_0 \tau_0$ at $W/\rho_0 = 10^{-7}$ J/cm³.Torr and $\rho_0 L = 10$ Torr.cm is given in Fig.3.

Fig.4 shows the dependence of the relative energy losses η^M (here: V^- - is the oscillatory excitation, DI^- - is the dissociations, EL - are the elastic losses, EI are the electron-ion collisions, EE is the electron excitation, I - ionization, DE - diffusion, $VT-V-T$ relaxation, W - dissociation from the upper oscillatory level) from $\rho_0 \tau_0$ at $W/\rho_0 = 10^{-7}$ J/cm³.Torr, $\rho_0 L = 10$ Torr.cm.

2. KINETICS OF RADICALS AND CHARGED PARTICLES

To investigate the effectiveness of the dissociation and ionization processes in a stationary plasma-beam discharge, the present work also considers the reactions employing heavy particles M : ions (H^+, H_2^+, H_3^+), inexcited molecules and radicals - hydrogen atoms in the ground and a metastable state $H(2s)$. The system of equations of heavy particles balance was solved together with Boltzman's kinetic eq-n (1) and the quasineutrality eq-n for charged particles. It was supposed that the life-time of charged particles is defined by the rate of the ambipolar diffusion, and the life-time of radicals, neutral molecules and metastable atoms-by their presence time in the working zone of the discharge. As a result of a self-consistent calculation besides the distribution function of the electron and their various macroscopic properties, the degrees of gas ionization and dissociation were calculated and also the concentration of ions and metastable atoms in dependence of the given parameters:

Fig.5,6 give the dependences of the degree of ionization (X_i) and dissociation (X_D) on the gas pressure ρ_0 for different values of W (J/cm³), calculated at $\tau = 10^{-4}$ (sec),

and Fig. 7, 8 give the dependences of χ_i and χ_D on the time τ (sec) calculated at $p_0 = 3 \cdot 10^{-2}$ Torr.

3. EXPERIMENTAL RESULTS

The experimental research of hydrocarbon radical formation was carried out on the installation described in /4/. Chemically pure methane was used as a work gas. The analysis of the reaction products was carried out by the mass-spectrometric method. The table (Fig. 9) presents the results of mass-spectrometric measurements. The first and the fourth lines show the data of the methane mass-spectre without any discharge at the pressure of $6 \cdot 10^{-2}$ Torr and $2 \cdot 10^{-2}$ Torr correspondingly. At the pressure inside the chamber $\sim 6 \cdot 10^{-2}$ Torr and the beam power of the order of 1 kw the formation of CH_3 mostly takes place, and at the pressure $\sim 2 \cdot 10^{-2}$ Torr only peaks C_2H_2 , CH_2 , H_2 are observed.

4. CONCLUSION

The experimental and conclusion results show that by changing the plasma-beam discharge parameters such as pressure and electron beam power, it is possible to purposefully govern the kinetics of plasma-chemical reactions. The solution of the self-consistent problem permits in the frame of a theoretical model to calculate the optimal parameters of a plasma-beam discharge necessary for obtaining the maximum effectiveness of the specified product outcome.

References

- (1) A.A. Ivanov, T.K. Soboleva, P.N. Yushmanov, Fizika plazmy (russ.), 3, 152, (1977)
- (2) J.W. Rich, J. Appl. Phys. 42, 2719 (1971).
- (3) R. Winkler, P. Michel, S. Pfan, V.V. Starykh, J.W. Wilhelm, SPIG-80, Dubrovnik, 1980, Contr. Paper, p. 168.
- (4) A.M. Alekseev et al., 4th Int. Symp. on Plasma Chemistry, Zurich, 1979, p. 427-432.

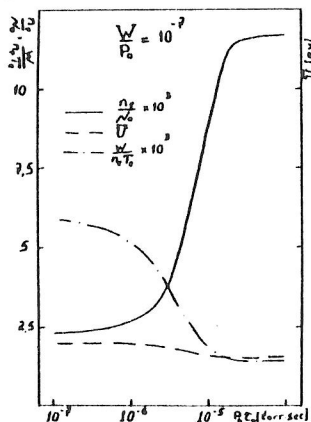


Fig. 1

Pres.	C_2H_2	C_2H_4	C_2H_6	CH_3	CH_4	CH_3	CH_2	CH	C	H_2	J_n	U_n
$p_{ch} p_m$	29	28	26	17	16	15	14	13	12	2	A	κB
1 $6 \cdot 10^2$ $9 \cdot 10^4$	29	3	0	28	140	84	17	9	3	0	0	0
2 $6 \cdot 10^2$ $7 \cdot 10^4$	8	8	16	5	46	45	9	5	2	29	0,7	1
3 $6 \cdot 10^2$ $5 \cdot 10^4$	8	8	14	4	37	36	7	4	2	19	1	1,25
4 $2 \cdot 10^2$ $4 \cdot 10^4$	15	10	0	15	120	105	15	5	2	0	0	0
5 $1 \cdot 10^2$ $3 \cdot 10^4$	0	61	0	0	0	0	18	0	0	74	0,7	1,25

Fig. 9

