HIGH-FREQUENCY ELECTRODE DISCHARGE AT REDUCED PRESSURE

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UDC 537.525.6

Results are presented for measured and calculated electron temperature and concentration, electric-field intensity, and electron collision frequency in a high frequency (hf) electrode discharge at pressures below atmospheric.

The high-frequency single electrode discharge at medium pressure is of great interest because of its expanding use in science and technology [1]. For a phenomenological description of the discharge and the processes occurring therein, a study of such parameters as charged-particle concentration, temperature and field distribution is of great importance.

At reduced pressure, ionization by electron collision plays a significant role [2]. The contribution of the electron component to the processes of excitation and dissociation, as well as to the mechanism for population of oscillatory–rotational levels is significant. The field distribution in the discharge plasma is an important parameter, if the field amplitude in the plasma is comparable to or exceeds the critical [3] value E_*:

$$E \geqslant E_* = [3mkTe^{2}\delta\,(\Omega^2 + \nu^2)]^{1/2} \tag{1}$$

Here T is the plasma temperature, Ω is the circular frequency of the hf field, ν is the effective collision frequency, and δ is the mean effective energy loss of an electron upon collision. In the zone of action of the field $E > E_*$ it is necessary to consider nonlinearity of the interaction processes and to introduce an approximation for the dependence of the transfer coefficients on the degree of "field" nonequilibrium of the plasma.

Methods for the study of discharge plasma properties at medium pressures are insufficiently developed. Spectroscopic methods [4] of measuring electron concentration n_e are inapplicable, since $n_e < 10^{14}$ cm^{-3}, and the intensity of the continuous spectrum is low. The probe method gives unreliable results because of the disturbing effect of the probe since the probe–plasma and plasma–ground capacitances are comparable in magnitude [5].

This study will present the results of spectroscopic, microwave, and calorimetric investigation of the parameters of an hf discharge in nitrogen in the pressure range 7.6–76 mm Hg.

A high-frequency discharge in nitrogen at pressures of 7.6–76 mm Hg is stable at power levels of several watts. The generator power in the experiments reached 16 kW, and the power introduced into the plasma was calorimetrically measured and reached 6 kW. The circular frequency of the field $\Omega \sim 2 \cdot 10^8$ Hz, i.e., the condition $\nu^2 \gg \Omega^2$ was fulfilled.

The discharge was created in a quartz chamber with a diameter of 90 mm and vertical length of 1000 mm, with a water-cooled copper–nickel electrode. Depending on the operating pressure, discharge length was from 60 to 90 cm, with diameter from 2 to 3.5 cm. Weak tangential torsion of the gas was used, and the linear plasma flow rate was small (~1 m/sec), so that flow turbulization could be neglected. The total power supplied to the plasma, its temperature T, the mean electron concentration $\langle n_e \rangle$, and the frequency of collision of electrons with neutral particles ν were measured.

Novosibirsk. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 4, pp. 13-17, July-August, 1973. Original article submitted May 15, 1972.

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The plasma oscillatory and rotational temperatures were measured with an ISP-51 spectrograph. In measurement of the oscillatory temperature T_v, the relative intensities of the maxima of the oscillatory bands of the second positive system of nitrogen with $\Delta v = 4.5$ were measured. The Frank–Condon factors were taken from [6]. To measure the rotational temperature T_r, photographic recordings of the spectrum were made by a camera with focal length $f = 270$ mm. Lines of the rotational structure of the 4278 Å band of the first negative system of nitrogen with $K = 10{-}23$ were measured. The gas temperature measured on the discharge axis changed from 2000 to 2500°K with increase in pressure. The oscillatory temperature underwent a corresponding change from 3800 to 2500°K. These T_v values agree with measurements in a glow discharge [7] and with values averaged over the radius for an electrodeless discharge of power 5 kW [8]. The T_v values in a high-frequency electrode discharge of [9] were higher.

The oscillatory temperatures in the ground state $x'\Sigma$ calculated according to [10] for 7.6 mm Hg coincided with T_r at this pressure. At 76 mm Hg, there is no such coincidence.

Microwave probing of the plasma was done by the directed-beam method in the wavelength range $\lambda = 1.1{-}1.5$ cm. A horn-lens method was used to focus the microwave beam to a spot of the order of a wavelength in dimension [11]. Preliminary experiments made it possible to select the plasmotron diameter such that absorption and scattering of the shf power on the walls could be neglected. The focal length f_c of the Teflon lenses was chosen with consideration of the condition that $f_c \gg \lambda, D$; $d^2/\lambda^2 \gg 1$ [11], where d is the lens diameter, and D is the diameter of the pinch. Probing was done at distances of 4, 16, and 28 cm from the electrode in a direction perpendicular to the discharge chamber axis.

Figure 1 shows the experimentally determined signal damping coefficients as functions of the working pressure in the discharge chamber at frequencies of 19 (1), 23 (2), 24 (3), 26 (4) GHz. Damping increases with increase in pressure. No sharp cutoff of the microwave signal was observed in the experiments. This may be explained by violation of the condition that $\nu/\omega \ll 0.01$ [12], deviations of experimental conditions from the requirements of geometrical optics [4], and by the fact that $\omega_0^2/\omega^2 > 0.01$, where ω_0 is the natural frequency of the plasma.

If the condition $\omega^2 \gg (c/D)^2$ is fulfilled, diffraction effects can be neglected in the first approximation [11]. Detailed consideration of the contribution of refraction, multiple reflection, and other types of losses to attenuation of a signal passing through the plasma is difficult [11]. Damping depends strongly on the radial concentration distribution, since

$$\langle n_e\rangle = 2\int_0^{D/2}\frac{n_e(r)}{D}\,dr$$

At pressures below 100 mm Hg the high-frequency single electrode discharge begins to lose its channelized quality (the conductive channel with a diameter of the order of 1 mm, specific for this type of discharge, begins to disappear), the diffusion mechanism of particle losses becomes significant, and the discharge is more homogeneous in a radial direction. The discharge model with a region Δ with sharp concentration gradients and $\Delta \ll D$ becomes admissible. In this case, the experimental damping data can be processed with the model of a plane layer for a cylindrical plasma column with sharp boundaries [11, 12]. Assuming that the reflected waves are incoherent [11], the reflection, transmission, and absorption of power

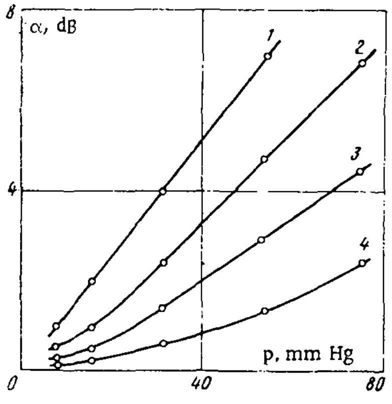

Fig. 1

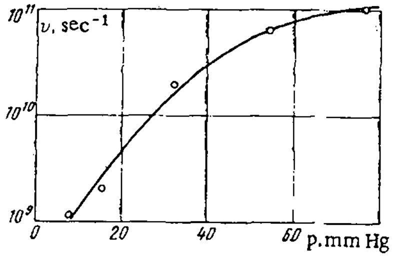

Fig. 2

can be calculated with the phase constant β and the damping constant α [11]. Calculations were conducted on a computer with an accuracy to 10^{-4} in each of the parameters. Expressions for α and β were taken in the form

$$\beta^2 = \frac{1}{2}\frac{\omega^2}{c^2}\left(1 - \frac{\omega_0^2}{\omega^2 + \nu^2}\right) + \frac{1}{2}\left[\left(1 - \frac{\omega_0^2}{\omega^2 + \nu^2}\right)^2 + \left(\frac{\omega_0^2}{\omega^2 + \nu^2}\frac{\nu}{\omega}\right)^2\right]^{1/2} \tag{2}$$

$$\alpha^2 = -\frac{1}{2}\frac{\omega^2}{c^2}\left(1 - \frac{\omega_0^2}{\omega^2 + \nu^2}\right) + \frac{1}{2}\left[\left(1 - \frac{\omega_0^2}{\omega^2 + \nu^2}\right)^2 + \left(\frac{\omega_0^2}{\omega^2 + \nu^2}\frac{\nu}{\omega}\right)^2\right]^{1/2} \tag{3}$$

since from preliminary estimates $\nu^2/\omega^2 > 0.01$ in high-pressure regions.

It follows from the calculations that in the pressure range 7.6-76 mm Hg, the mean electron concentration $\sim 10^{12}$ cm^{-3}, which agrees with the data of the literature on hf discharges at high pressure, for their approximations [9]. The data of [13] on n_e under analogous conditions appear to be low in value. This disagreement may be explained by the difference in power introduced into the plasma (a difference of almost one order).

Figure 2 shows the effective collision frequency ν as a function of pressure. For a Maxwellian distribution of electrons over velocity we have for ν

$$\nu = A\left(\frac{m}{2kT_e}\right)^{5/2} P_0 \int \sigma(v) \exp\left(-\frac{mv^2}{2kT_e}\right) v^5\, dv \tag{4}$$

where P_0 is the corrected pressure, $\sigma(v)$ is the collision section with momentum transfer. The electron–ion collision frequency ν_{ei} and the electron–electron collision frequency ν_{ee} are small in comparison with the electron–neutral particle collision frequency and make no significant contribution to ν. It follows from Eq. (4) that the nonlinearity of $\nu(P)$ (Fig. 2) indicates the complex relationship of T_e with pressure. Decrease in the growth of $\nu(P)$ in the high-pressure region is evidently related to the commencement of discharge channelization, i.e., to increase in the inhomogeneity $n_e(r)$.

The function $T_e(P)$ can be found from the energy balance equation

$$n_e T \nu_e(\varphi)(\varphi - 1) - T\nabla \varkappa \nabla\varphi - \sigma(\varphi) E^2 = 0 \tag{5}$$

Here $\varphi = T_e/T$, $\varkappa$ is the coefficient of thermal conductivity, $\sigma(\varphi)$ is the conductivity, determined for a Maxwell distribution by the relationship [15]

$$\sigma(\varphi) = \sigma_0 f(\varphi) \quad (\sigma_0 = 4\omega_0^2/3\pi^{3/2}\nu_0), \tag{6}$$

and E is the effective field in the plasma.

The ratio of energy loss by electron thermal conductivity [a portion of the second term in Eq. (5)] to energy loss by collision [the first term of Eq. (5)] is of the order of λ_*^2/D^2, where λ_* is the electron energy mean free path. Under the conditions here $\lambda_*^2/D^2 \ll 1$. Losses by thermal conductivity of the gas may also be neglected [9].

In evaluating $\sigma(\varphi)$ and E the dielectric permeability

$$\varepsilon = \varepsilon_0 - \frac{8\sqrt{2}\pi}{3}\frac{m^{1/2}T^{3/2}\omega_0 i}{n_e \Omega \nu}\int x \frac{\partial f}{\partial x} dx \tag{7}$$

for a Maxwellian distribution of electrons over velocity is calculated for the case realized by the experimental data $\nu/\Omega > 1$, $\omega_0^2/\varphi^{1/2}\Omega\nu_0 \gg \varepsilon_0$ [14].

It follows from the calculations that Im $(\varepsilon) \gg$ Re (ε) and Im $(\varepsilon) > 1$ over the entire pressure range studied. If, moreover, $D \ll L$, where L is the length of the discharge conductive zone, the discharge can be considered as a conductor with a decreasing field therein, in which dissipation of the hf power occurs basically through collision losses. The solution of the equation for propagation of the field in the direction of the z axis from the electrode, considering the low value of the Bessel function argument [15], leads to the conclusion that the damping coefficient changes

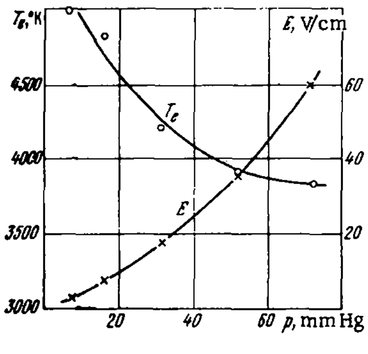

Fig. 3

only slightly over the length of the discharge. Losses by radiation in this case are also low [1], and the effective field in the discharge can be evaluated by calorimetric methods

$$Q \sim \int \sigma(\varphi) E^2 dV \tag{8}$$

The near electrode region with voltage drop ~400 V [2] is not considered.

From Eqs. (5) and (8) T_e and E may be determined. The values of δ for nitrogen in the range of gas temperatures 1000-5000°K and electron temperatures 1000-15000°K needed to calculate ν_e are given in [2, 16]. The functions $T_e(P)$ and E(P) are shown in Fig. 3. With increase in pressure a growth in E and decrease in T_e are observed. Over the entire pressure interval $E < E_*$. With increase in pressure the electron temperature approaches that of the gas, which increases with increased pressure; at P ~1 atm the hf plasma in nitrogen can be considered dynamically stable.

Note added in proof. From solving the system of equations of balance and electrodynamics describing an hf discharge, and also from recent spectroscopic measurements, it follows that T_e in the central portion of the discharge exceeds the values obtained in this study. This was to be expected, since the values of n_e and σ used for calculation of T_e based on Eqs. (5) and (8) are averaged values over the discharge radius, while a gas discharge plasma in reality is radially inhomogeneous in any of its parameters.

LITERATURE CITED

1. Notes on the Physics and Chemistry of Low Temperature Plasmas [in Russian], Nauka, Moscow (1971).
2. V. Farsky, and J. Janca, "Energetische Verhaltnisse in Plasma einpoliger Entl.," Beitrage Plasmaphys., 8, No. 2 (1968).
3. V. L. Ginzburg and A. V. Gurevich, "Nonlinear phenomena in a plasma located within a variable electromagnetic field," Usp. Fiz. Nauk, 70, No. 2 (1969).
4. Plasma Diagnostics [Russian translation], Mir, Moscow (1967).
5. O. Sager, "Frequency analysis of the probe current in an rf discharge," Proc. IEEE, 57, No. 2, 227-228 (1969).
6. R. N. Zare, E. O. Larsson, and R. A. Berg, "Frank—Condon factors for electronic band systems of molecular nitrogen," J. Molec. Spectroscop., 15, No. 2, 117-139 (1965).
7. S. S. Vasil'ev and O. N. Glagoleva, "Electron temperature as excitation temperature of electron-oscillatory levels of nitrogen molecules in discharges," Zh. Fiz. Khimii, 44, No. 8 (1970).
8. Z. G. Orlova, B. E. Zhestkov, and A. I. Omelik. "A spectral study of a high-frequency electrodeless nitrogen discharge plasma," in: Thermophysical Properties of Low-Temperature Plasma [in Russian], Nauka, Moscow (1970).
9. J. Janca, "Energy exchange in high-pressure discharge plasmas excited in diatomic gases," Czechoslov. J.Phys., B18, No. 7, 919-927 (1968).
10. M. Z. Novgorodov, V. N. Ochkin, and N. N. Sobolev, "Measurements of oscillatory temperature in CO_2 discharges," Zh. Tekh. Fiz., 40, No. 6 (1970).
11. V. E. Golant, Microwave Methods of Plasma Diagnostics [in Russian], Nauka, Moscow (1968).
12. H. Prinzler, "Zur Mikrowellendiagnostik stromstarker Entladungen mit Skineffekt," Beitr. Plasmaphys., 9, No. 5 (1969).
13. T. Bohatiel, "Torta sialte tipuri de descarcari de inalta frecventa la presiuni mai mici ca o atmosfera in aer, Ar, CO_2, H_2," Stud. Si cerc. fis., 22, No. 3 (1970).
14. F. G. Bass and Yu.G. Gurevich, "Nonlinear theory of electromagnetic wave propagation in a solid-state plasma in a gaseous discharge," Usp. Fiz. Nauk, 103, No. 3 (1971).
15. B. Z. Katsenelenbaum, High-Frequency Electrodynamics [in Russian], Nauka, Moscow (1966).
16. L. G. Cristhophorou and F. G. Carter, "Energy lost by slow electrons in collisions," Chem. Phys. Letter, 2, No. 8, 607-609 (1968).