A numerical investigation of the electrical characteristics of the electrode boundary layer of a slightly ionized plasma of molecular gases

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The hydrodynamic problem of determining the electrical characteristics of the electrode region in a slightly ionized plasma in chemical equilibrium was formulated in [1] and subsequently analyzed more than once. The present article is devoted to a numerical solution of this problem. We note that, besides the independent interest, such a solution is also of interest for estimating the degree of accuracy of various approximate approaches.

The problem under consideration is a boundary-value problem for a system of nonlinear, ordinary differential equations; for the conditions of practical interest this system contains two small parameters to the leading derivatives, while in the case of a relatively low electrode temperature it also contains a third small parameter in the exponent. Certain difficulties arise in the direct numerical solution of problems of this type, and therefore one or another simplifying assumptions were made in [2-4], devoted to the numerical solution of this problem. For example, in [2, 3] the electrode layer is subdivided into a space-charge layer and a quasineutral region, and the solution of the problem is sought separately in each region with subsequent joining. In this case the ionization of neutral atoms and the recombination of charged particles in the space-charge layer are ignored, which prevents a correct description of the behavior of the volt-ampere characteristic curves of the electrode region of molecular-gas plasma for high densities of current to the electrode [5]. Some important terms of the system of determining equations were not taken into account in [4], and in [2-4] the problem was solved by the shooting method.

An efficient iteration algorithm based on the trial-and-error method is developed in the present article to obtain a direct numerical solution of the problem under consideration. Calculation results are given for the case of a plasma of combustion products with a potassium admixture and a wide range of electrode temperatures, and a detailed comparison is made with the results of calculations by the analytical equations of [5], obtained by the method of joined asymptotic expansions, and with experimental data.

I. Statement of the Problem

Let us consider a multicomponent, slightly ionized plasma of molecular gases containing neutral components, positive singly charged ions of atoms of one of the neutral components
(an easily ionized admixture), and electrons. The plasma borders on a plane, perfectly catalytic, and is nonemitting electrode. The ionization of admixture atoms can occur in collisions both with electrons and with molecules of one of the neutral components; electrons or molecules of this neutral component can take part as the third body in ion–electron recombination. It is assumed that the plasma pressure and the molar concentrations of the neutral components are constant, and convective transfer of charged particles is ignored. The plasma temperature is assumed to be a given function of the y coordinate (the y axis is directed normal to the electrode).

In dimensionless variables we have the following nonlinear boundary-value problem [1, 5]:

\( I_i = -a(z_i - \theta^{-1} \lambda_i E); \)  
(1.1)

\( I_e = -a(z_e + \theta^{-1} \lambda_e E); \)  
(1.2)

\( \chi I_i = \frac{2b}{1 + \beta} (1 + cz_i)(r - z_i \lambda_e); \)  
(1.3)

\( I_e = \beta I_i - j; \)  
(1.4)

\( e\theta E' = z_i - z_e; \)  
(1.5)

\( \eta = 0, z_i = z_e = 0, \eta \to \infty, z_i \to 1, z_e \to 1, \)  
(1.6)

\( \eta = \frac{y}{L}, z_i = \frac{x_i}{x_i \infty}, I_i = \frac{I_i}{x_i \infty \lambda_{i \infty}}, E = \frac{e\lambda_i E}{kT \infty} (j = i, e), \)

\( a = \frac{nD_i}{x_n \infty D_{i \infty}}, \theta = \frac{T}{T \infty}, b = \frac{k_{\lambda i} x_i}{k_{\lambda i}} \frac{u}{x_i}, c = \frac{k_{\lambda e} x_i}{k_{\lambda e} \infty}, \)

\( r = \frac{x_i}{x_i \infty} = \theta^{\frac{1}{2}} e^{-x}, x_i = \frac{n_{i \infty}}{x_i}, t = m \frac{1 - \theta}{\theta}, \chi = \frac{d_{i \infty}}{2L}, \)

\( \beta = \frac{D_i}{D_e}, j = \frac{I_i \infty}{eD_{i \infty} n_{i \infty}}, e = \frac{k_{\lambda i}^2}{L^2}, m = \frac{I}{2kT \infty}, \)

\( d_i = \left[ \frac{4D_i}{k_{\lambda i} n_{i \infty} n_{i \infty} (1 + \beta)} \right]^{1/2}, \)

\( h = \left( \frac{kT}{4\pi n_{i \infty} e^2} \right)^{1/2}. \)

Here \( J_i, J_e, x_i, \) and \( x_e \) are the number densities of the diffusional fluxes and the molar concentrations of ions and electrons; \( n \) and \( T, \) total particle concentration and the plasma temperature; \( e \) is the electron charge; \( E, \) electric field strength; \( k, \) Boltzmann constant; \( k_{\lambda i} \) and \( k_{\lambda e}, \) recombination rate constants for reactions with the participation of molecules of a neutral component and electrons as the third body, respectively; \( D_i \) and \( D_e, \) coefficients of diffusion of ions and electrons (their ratio \( \beta \) is assumed to be constant); \( x_i, \) molar concentration of the neutral component, the molecules of which take part in ionization and recombination reactions; \( j^0, \) density of electric current to the electrode (a given value); \( n_{i \infty}, \) quasineutral concentration of charged particles in local chemical equilibrium; \( I, \) ionization energy of the admixture atoms; \( L, \) characteristic scale of variation of plasma temperature; a prime denotes differentiation with respect to \( \eta; \) the index \( \infty \) is assigned to values of the respective quantities in the undisturbed plasma.

2. Algorithm for Numerical Solution

The inequality \( \epsilon \ll 1 \) holds for conditions of practical interest. In this case the Poisson equation (1.5) is unsuitable for determining the electric field \( E, \) since there is a small parameter in front of the only term containing the electric field in this equation [6]. Therefore, we convert the system of differential equations (1.1)–(1.5) to a form more suitable for numerical solution [7],

\[ [e\theta E' + (\beta - 1) z_i] = -eEE' + \frac{1 + \beta}{a} z_i E - \frac{\beta}{a} j; \]

\[ z_i^2 - \frac{a}{\beta} z_i E = \frac{2b}{x(1 + \beta)} (1 + e(z_i - e\theta E'))(z_i^2 - e\theta E' z_i - r^2). \]

The boundary conditions take the form
To obtain the exact solution of this problem we use an iteration process, at each step of which the following linearized equations are solved jointly:

\[
\begin{align*}
\varepsilon_0 \partial E' + (\beta - 1) z_i' &= - \frac{\beta}{a} j + \varepsilon_0 E' - \frac{4 + \beta}{6} z_i E - \varepsilon_0 E' + \frac{4 + \beta}{6} z_i E - \varepsilon_0 E + \frac{4 + \beta}{6} \frac{E_0}{E} z_i, \\
\left[ \frac{a}{6} z_i E + az_i - \frac{a}{6} z_i E - \frac{a}{6} \frac{E_0}{E} z_i \right]' &= \frac{20}{\zeta (1 + \beta)} [1 + c(\varepsilon_0 - \varepsilon_0 E') + (r + z_i - \varepsilon_0 E') z_i - (r + z_i) r].
\end{align*}
\]

Quantities marked by a tilde are taken from the preceding iteration.

The linearized equations are solved by matrix trial and error (we used a generalization of scalar trial and error [8] to the case of a system of equations) using a two-point difference scheme of fourth-order accuracy [8]. To obtain a solution to within \(10^{-6}\) requires 10-30 iterations.

We note that in the outer part of the electrode boundary layer (in chemical equilibrium) the solution is described by simple analytical expressions. For economy of computer time, therefore, the interval of numerical integration can be shortened and integration can be carried out from the wall to the region in chemical equilibrium. In this case the second boundary condition (2.1) can be replaced by the following condition:

\[
\eta = \Lambda, \quad z_i = r, \quad E = \frac{\beta_0}{(1 + \beta)aj} j + \frac{\beta - 1}{\beta + 1} \frac{\theta_0}{r}.
\]

The quantity \(\Lambda\) is chosen large enough so that the distributions of ion concentration and electric field obtained from the numerical solution join sufficiently smoothly with the distributions in chemical equilibrium. Under this condition the solution proves to be independent of the concrete choice of the quantity \(\Lambda\).

In the region of the boundary layer of space charge (the Debye layer) the step of the difference grid must be small enough to assure the desired accuracy.

After solving the formulated problem one can find the electrode voltage drop and the electron density distribution corresponding to a given value of \(j\): 

\[
\psi_0 = \int_0^\infty (E - E_0) \, d\eta = \int_0^\Lambda E d\eta - \frac{\beta}{1 + \beta} j \Lambda + \frac{1}{1 + \beta} \int_\Lambda^\infty [(\beta - 1) \frac{\theta_0}{r} + \beta j (\frac{\theta_0}{ar} - 1)] \, d\eta,
\]

\[
z_e = z_i - \varepsilon_0 E'.
\]

The calculation of each volt–ampere characteristic curve starts with \(j = 0\), at which the choice of the initial approximation is not important; for all subsequent values of \(j\) the solution obtained for the preceding value is used as the initial approximation.

As an example of the application of the formulated method, in Figs. 1–3 we present the results of calculations for the case of a plasma of combustion products with a potassium admixture for a temperature distribution of plasma in the boundary layer described by the model equation

\[
\theta = 1 + (\theta_0 - 1)e^{-\eta},
\]

where \(\theta_0\) is the dimensionless temperature of the electrode (a given parameter). The quantities \(k_{r1}\) and \(k_{re}\) were taken from [9], and to calculate the diffusion coefficients we used the equations

\[
D_i = 0.082 \left( \frac{p}{1K} \right)^{2} \frac{1 Pa cm^2}{p \cdot sec}, \quad D_e = 48 \left( \frac{p}{1K} \right)^{2} \frac{1 Pa cm^2}{p \cdot sec},
\]

constructed from the numerical data of [10, 11], where \(p\) is the plasma pressure.

It was assumed that \(T_0 = 2700^\circ K, p = 1 \ atm, L = 1\ cm, \) the molar concentration of potassium atoms is 1%, and the molar concentration of water molecules (these molecules take part
in ionization and recombination reactions) is \( x_1 = 18\% \), and then \( \varepsilon = 10^{-9} \), \( \chi = 10^{-5} \), \( \beta = 1/220 \), \( c = 0.7 \), and \( m = 9.3 \).

The cathode branches of the volt-ampere characteristic curves \( \psi_w(j) \) (curve 1) and the thickness of the Debye layer as a function of the current density, \( \eta_D(j) \) (curve 2; for determinacy, the thickness of the Debye layer is understood as the value of the \( \eta \) coordinate at which the difference between the functions \( z_i \) and \( z_e \) becomes 10\%), are presented in Figs. 1-3 for the cases of \( \theta_w = 1, 0.6, \) and 0.2, respectively.

In all cases the volt-ampere characteristic curve demonstrates ion saturation. In other words, when the current density reaches a certain critical value the inclination of the volt-ampere characteristic curve to the current axis starts to increase.

With a decrease in the wall temperature from \( \theta_w = 1 \) to \( \theta_w = 0.2 \) the qualitative form of the functions \( \psi_w(j) \) and \( \eta_D(j) \) changes. Whereas a monotonic increase in the inclination of the volt-ampere characteristic curves to the current axis is characteristic of the cases of \( \theta_w = 1 \) and 0.6, in the case of \( \theta_w = 0.2 \) the volt-ampere characteristic curve has a bend. For \( \theta_w = 1 \) the dependence of the thickness of the Debye layer on the current density is close to linear in the saturation section, for \( \theta_w = 0.6 \) the rise in \( \eta_D \) with an increase in \( j \) goes considerably more slowly, and for \( \theta_w = 0.2 \) there is a sharp increase in \( \eta_D \) at the start of the saturation section and an ever slower rise with a further increase in \( j \).

The relative errors of the asymptotic equations for the functions \( \psi_w(j) \) and \( \eta_D(j) \) are also given in Figs. 1-3.

The following parametric equations were obtained in [5] to describe the volt-ampere characteristic curves of the electrode boundary layer:

\[
\psi_w = -2\varepsilon^{-1/2}\chi^{-1/2} \int_0^{\eta_D} \left[ \int_0^\eta a^{-1} \left( \int_b \eta_D \right) d\eta \right]^{1/2} d\eta.
\]

The expected region of applicability of these equations is confined to values of the parameter \( \eta_D \) which are not too small. The errors in the representations of the functions \( \psi_w(j) \) and \( \eta_D(j) \) given by these equations are plotted with the lines 3 and 4, respectively, in Figs. 1-3.

The following equations were also obtained in [5] for the case of a hot electrode:

\[
\psi_w(j) = \frac{\beta}{1 - \beta} \left[ \frac{-\theta}{ar} - 1 \right] d\eta - e^{-\varepsilon^{1/2}\chi^{1/2}} \left( \frac{2a}{b} \right)^{1/2} 2\pi \Phi \left( \frac{\sqrt{1/4 + 2\varepsilon \gamma}}{4\varepsilon} \right),
\]

\[
\eta_D(j) = -\frac{\sqrt{1/4 + 2\varepsilon \gamma}}{2b\theta^{3/2}},
\]

\[
\Phi(u) = \left( \frac{1}{2} \right) (u^2 + u)^{1/2} - \frac{1}{8} \ln \left( 2(u^2 + u)^{1/2} + 2u + 1 \right),
\]

\[
\gamma = r \left[ \frac{br(8 + 3\varepsilon \gamma)}{8a} \right]^{1/3}.
\]
The expected region of applicability of these equations is limited by the inequality 
\[ j < -2a_w \gamma \chi^{-1/2}. \] The errors of these equations are represented by the lines 5 and 6 in Fig. 1.

From Figs. 1-3 it follows, in particular, that over most of the section of ion saturation the error of the respective asymptotic equations does not exceed 10%.

3. Comparison with Experimental Data

In accordance with the above, to calculate the volt-ampere characteristic curves, in addition to information about the transfer and kinetic coefficients of the plasma, one must assign the profiles of temperature and concentration of admixture atoms in the boundary layer, as well as the values of the pressure and the concentration of water molecules. Unfortunately, among the numerous reports now published and devoted to the experimental investigation of a diffuse discharge, we were unable to find reports in which these quantities were measured along with the discharge characteristics. The most detailed data are given in [12, 13], and they were taken for interpretation in the present article.

The measurements of [12, 13] were made in a plasma stream of combustion products at atmospheric pressure. The distribution of plasma temperature in the outer part of the boundary layer (in the region of \( T \gtrsim 2000\,\text{K} \); we note that this distribution proved close to a power-law distribution with an exponent of \( 1/10 \)) and the conductivity in the core of the stream were measured along with the discharge characteristics.

In making the calculations discussed below, the kinetic and transfer coefficients of the plasma were calculated just as in Sec. 2, the pressure was taken as \( 10^5 \,\text{Pa} \), and the molar concentration of water molecules as 36%. For the temperature profile in the entire boundary layer we used the equation

\[
\frac{1}{T} = \frac{1}{T_w} + \left( \frac{1}{T_w} - \frac{1}{T_\infty} \right) \left( \frac{y}{\delta} \right)^{1/10},
\]

where \( T_\infty = 2600\,\text{K} \); \( T_w = 420 \) and \( 520\,\text{K} \); \( \delta = 0.42 \,\text{cm} \) for \( T_w = 420\,\text{K} \) and \( \delta = 0.36 \,\text{cm} \) for \( T_w = 520\,\text{K} \).

Finally, the molar concentration of the impurity (potassium atoms) was taken as constant in the boundary layer and determined from the experimentally measured conductivity \( \sigma \) of the stream core:

\[
x_n = 2.7 \times 10^{-5} \left( \frac{\sigma}{\text{S/m}} \right)^2. 
\]

The calculated thickness \( y_D \) of the Debye layer at the electrode, which is at a floating potential, as a function of \( \sigma \) is presented in Fig. 4 (curves 1 and 2). The quantity \( y_D \) was defined as the distance from the electrode surface at which the charge separation is 10% (curve 1) or 1% (curve 2). The experimental data of [12], obtained by the plasma-condenser method, are plotted with points. The agreement between theoretical and experimental data is satisfactory.

In Figs. 5 and 6 we present theoretical volt-ampere characteristic curves (lines) and experimental points \( \psi^* = (kT_w/e)\psi_w \) for \( T_w = 420 \) and \( 520\,\text{K} \), respectively. Curve 1 and the
light points correspond to $\sigma = 1 \text{ S/m}$, curve 2 and the crosses to $\sigma = 3 \text{ S/m}$, and curve 3 and the dark points to $\sigma = 5 \text{ S/m}$ in Fig. 5 and $\sigma = 5.8 \text{ S/m}$ in Fig. 6. The general form of the theoretical and experimental characteristic curves is the same [for example, it is interesting to note that both the theoretical and the experimental values of $\psi_0^0$ for the case of $T_w = 420^\circ\text{K}$ (see Fig. 5) exceed by several times the corresponding values for the case of $T_w = 520^\circ\text{K}$ (see Fig. 6)], although quantitatively the theoretical voltage drops exceed the experimental ones. This disagreement increases with an increase in $\psi_0^0$.

One of the possible causes of this disagreement is that the values of the kinetic coefficients and the plasma temperature distribution adopted for the calculation do not correspond to the experimental conditions. As an example, in Fig. 6 we present theoretical curves for the case of $\sigma = 5.8 \text{ S/m}$, in the calculation of which we successively increased the values of the function $k_{1x}(y)$ by an order of magnitude (curve 4), increased the content of potassium atoms by five times (curve 5), and reduced the exponent in the temperature distribution law to $1/12$ (curve 6). It is seen that these curves are in considerably better quantitative agreement with the experimental characteristic curve.

For a final clarification of the causes of this disagreement, new experiments must be run satisfying the requirements listed in the present section.

**LITERATURE CITED**

THEORY OF ANISOTROPIC FERROMAGNETIC COLLOIDS

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Introduction. In present theoretical descriptions of the flow of a ferrofluid (a stabilized surface-active colloid consisting of ferromagnetic particles in a fluid carrier) various approaches and approximations have been used [1-12]. One of the first hydrodynamic models of a ferrofluid [1] assumed that the magnetization relaxation time $\tau$ was small in comparison to the characteristic macroscopic times of the problem; in this case one can put $\tau = 0$, and consider the magnetization of the medium as given by an equilibrium equation of state.

Unlike this approach, in [2-4], models were developed where the relaxation time $\tau$ is finite, so that the change in magnetization is given by a relaxation equation, which together with the other equations of the theory describe the magnetization dynamics of magnetically isotropic ferrofluids. From the microscopic point of view, magnetic isotropy of the ferrofluid implies that the magnetization is frozen in the particles or the generalized magnetic anisotropy constant $K$ of the particles is infinite. This means that the orientational change of the magnetization in these models is determined entirely by the rotation of the particles (Brownian relaxation [5] with relaxation time $\tau_2$).

In the case where the thermal energy $kT$ is comparable to the (finite) magnetic anisotropy energy of a particle $kV$, where $V$ is the volume of a particle and $k$ is the Boltzmann constant, the latter energy is not large enough to keep the magnetic moment frozen inside the particle against the thermal fluctuations. The orientational change of the magnetization in the general case where the magnetic moments are partially frozen will be determined both by diffusion of the moment with respect to the particle (Néel relaxation with relaxation time $\tau_1$ [5]), and by the Brownian rotation of the particles, which for hydrodynamical flow and for $\tau_1 \ll \tau_2$ will determine the macroscopic magnetic anisotropy of the ferrofluid.

The first treatment of the effect of partial freezing of the magnetic moments on the effective viscosity of the ferrosuspension was considered in [10]. However, this treatment, based on kinetic ideas, did not yield a definite formula for the effective viscosity, which would contain the previously known formula for the rigid magnetic dipole model, or a general macroscopic equation of motion.

A series of papers [8, 11, 12] are of interest, in which a general macroscopic equation was formulated taking into account the finiteness of the magnetic anisotropy energy of the particles by introducing into the theory a macroscopic vector parameter $\mathbf{A}$ defined as the internal magnetic anisotropy field. In the equilibrium state this parameter is taken to be proportional to the magnetization $\mathbf{A} = \alpha m$, where the coefficient $\alpha$ is defined as a parameter giving the degree of freezing of the magnetic moments with respect to the particles.

Calculations of the dependence of the viscosity on the field strength $\xi$ in a state of partial equilibrium using these equations [8] yields remarkable agreement between theory and experiment for a suspension of magnetite in kerosene. However, this agreement should not be taken as unequivocal support for the above interpretation of the parameter $\alpha$ (or for the assumption that the deviation of the experimental viscosity dependence from the rigid dipole