DUSTY AND SMOKY PLASMAS. SOME PROPERTIES AND APPLICATIONS

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Methods of application of low-temperature plasma with a condensed disperse phase, which dusty and smoky plasmas belong to, have been classified. Some properties of smoky plasma connected to the conditions of its production and resulted from its content, namely, a displacement of ionization equilibrium, long-range interaction, spatial nonhomogeneity of ionization equilibrium in the vicinity of charged smoke particles, electro-acoustical oscillations, and nucleation, have been discussed. An analysis of common and distinctive properties of dusty and smoky plasmas has been carried out.

Dusty and smoky plasmas are two kinds of low-temperature plasma which involves a condensed disperse phase. Determinative features of such plasma depend on the presence of fine particles in a liquid or solid state, particle characteristics, and interphase interaction at their surfaces. Among the peculiarities of plasma properties, one may note a displacement of ionization equilibrium in the gaseous phase containing particles with respect to that in identical gas plasma without them [1], the heterogeneity of the spatial distribution of the plasma ionization degree depending on the condensed phase parameters [2], oscillation processes in the condensed phase [3–5], formation of ordered structures and voids [6–8], variation of thermal and optical characteristics, etc.

Interphase interaction in plasma with a condensed disperse phase results also in varying the characteristics and properties of the condensed phase. Particles get certain charges; as a result, the plasma layer in their vicinity becomes perturbed, there appear the electrostatic interaction between particles and the forces of ionic pressure upon particle surfaces [9]. All that substantially affects the nucleation [10], condensation kinetics in plasma, and, consequently, the process of condensed phase formation and the size distribution function of the particles.

Practical application of plasma with a condensed disperse phase is caused by the specificity of its thermal and electrophysical characteristics and properties. Therefore, dusty and smoky plasmas have various fields of application, because, together with their common properties, they possess essential distinctions which are caused by the specificity of their production.

Dusty plasma is formed when dispersed particles are introduced artificially into gas-discharge plasma or into high-frequency plasma at low pressure [11]. It is also observed naturally, for example, in the interplanetary space, planetary rings, circumsolar dusty rings, comet tails, interstellar molecular clouds, phosphorescing clouds in the arctic tropo- and mesoshere, prethunderstorm clouds [12]. Its essential attribute is an insignificant role played by collisions in the gaseous phase and thermal electron emission from the surfaces of condensed particles. The former process is governed by a low, as a rule, pressure in the medium, and the latter one by a low temperature of particles and neutral components in the gaseous phase and by high values of the work function of electrons at the particles' surfaces. Therefore, ionization of gas atoms and molecules occurs due to an electric current or radiation, and plasma becomes nonequilibrium with significant differences in the electron, ion, and atom temperatures. The interphase charge exchange occurs through the absorption and emission of electrons by particles, with the particle charge being negative.

On the other hand, smoky plasma is formed when solid, gaseous, or liquid fuels, as well as synthetic fuel compositions doped with metal powders, are being burnt down [13]. Its determinative attribute is that smoke particles are formed immediately in plasma, have considerably smaller dimensions, and can be represented as metals, or metal oxides, or soot particles. In order to enhance the ionization degree of the gaseous phase, a fuel mixture is doped with readily ionized compounds of alkaline metal. Evaporation and dissociation of the dope produce an admixture of readily ionized atoms in the gaseous phase.

Smoky plasma, by essence, is a plasma made up of combustion products. It is formed as a flame, which is nonhomogeneous both in the distribution of temperature fields and in the distribution of component concentrations. The flame is divided conventionally

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into two characteristic altitude areas [14]. In the first one (it is the bottom part of the flame), intensive chemical reactions, that result in establishing the composition contents of the gaseous and condensed phases, take place. Here, the Boltzmann distribution of population of the atomic energy levels is violated, and the temperatures of the gaseous and condensed phases differ from each other by 20–40%.

In the second region of the flame, the composition of components comes to equilibrium, chemical reactions are completed, the temperature is close to the equilibrium one and can change within 2000–3000 K. In some areas, the conditions of local thermodynamic equilibrium are satisfied. It essentially influences the interphase interaction in plasma and its properties as a whole. Ionization of atoms in the gaseous phase is thermal, the concentration of free electrons reaches the value of about 10^{20} m^{-3}. Interphase interaction at the particle surfaces occurs through an exchange of electrons (sticking and thermionic emission), surface ionization of atoms, and recombination of ions [14]. The charges on the particle surfaces can be either positive or negative, depending on the particle and plasma parameters.

Thus, the properties of dusty and smoky plasmas differ substantially from each other. Interphase interaction through the exchange of energy, mass, and charges is typical of smoky plasma, which defines the kinetics of nonequilibrium processes, such as evaporation and condensation. On the other hand, it plays an essential role in attaining the quasi-equilibrium concentrations of plasma components and the charges of condensed particles under thermodynamic equilibrium conditions. Interphase interaction results in distributing the particles by size and charge, with the processes of formation of the dimensions and charges of particles being interconnected [10].

Smoke particles of metal oxides have a bimodal distribution [6]. For example, the plasma of combustion products of aluminum contains two fractions of aluminum oxide particles, 0.04—0.06 μm and more than a micron in dimension. If plasma is doped by Cs atoms, the electron concentration raises by almost two orders of magnitude up to a value of 10^{20} m^{-3}, and the particle dimension in the submicron size fraction decreases down to 0.01 μm. This evidences for the influence of the degree of plasma ionization on the average dimension of the particles that are formed during volume condensation. The electrostatic charge of the nucleus probably changes its free energy, which results in the variations of both the critical dimension of a charged nucleus [10] and the size distribution function.

Due to the surface processes of atom ionization and ion recombination and their proper account in the interphase interaction at the particle surfaces, the effect of a displacement of the ionization equilibrium in the gaseous phase of plasma with respect to the equilibrium in an identical gas plasma without particles was revealed [1]. Minimizing the free energy of dusty plasma, the folowing expression was obtained:

$$\mu_e + \mu_i - \mu_a = (\mu_a - \mu_i) n_p \frac{1}{e}\frac{\partial \rho}{\partial n_e} + e n_p \frac{\partial \varphi_s}{\partial n_e} \equiv \psi, \qquad (1)$$

where μ_e, μ_i, and μ_a are the chemical potentials of electrons, ions, and atoms, respectively; n_e and n_p are the concentrations of free electrons and smoke particles, respectively; ρ is the density of the space charge; φ_s is the potential of the particle surface; and ψ is the nonequilibrium parameter.

Solving Eq. (1) and substituting the expressions for the chemical potential in the ideal-gas approximation enable one to write down the condition of ionization equilibrium as the Saha equation with the effective value of the ionization potential $I_{\text{eff}} = I - \psi$. Hence, the nonequilibrium parameter characterizes the variation of the ionization potential of an isolated atom and, therefore, the displacement of ionization equilibrium in the gaseous phase owing to the presence of the space charge and the perturbation of plasma by the particle potential.

The researches of surface processes at the phase interface [15] and of electron and ion statistics in smoky plasma [16] showed that the nonequilibrium parameter can be expressed in terms of the plasma potential.

The physical meaning of *the plasma bulk potential* is defined as a work made by plasma when acquiring a unit bulk charge, provided that the charge of the opposite sign is accumulated on the particle surfaces. For a single charged smoke particle, we have

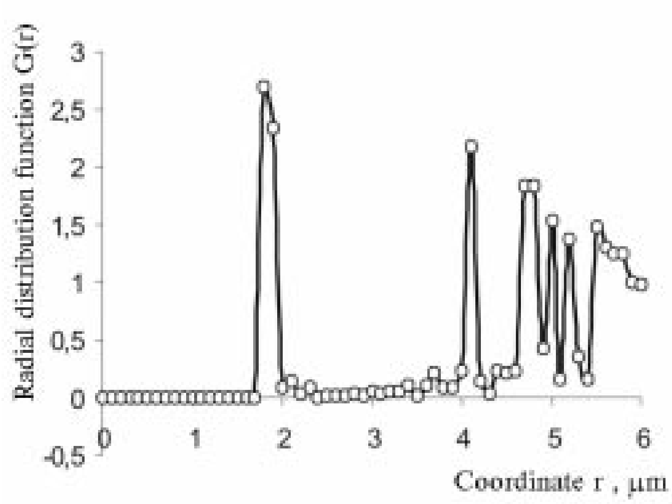
$$\varphi_{\text{pl}} = \frac{-4\pi\varepsilon_0}{eZ_p} \int\limits_{r_p}^{r_p+4D} \left(r \frac{d\varphi}{dr} \right)^2 dr - \langle \varphi \rangle, \qquad (2)$$

where Z_p is the charge of the particle in units of the electron charge, r_p is the radius of the particle, D is the Debye radius of screening, and

$$\langle \varphi \rangle = \frac{1}{V} \int_V \varphi(\mathbf{r}) dV$$

is the value of the bulk potential averaged over the volume.

The introduction of the notion of plasma bulk potential plays an important role, because it is the zero



Radial distribution function of submicron size particles

reading for the spatial distribution of the potential in plasma. Although the potential in electroneutral gas plasma can be reckoned from the zero value at infinity, in smoky plasma with particles different by charge signs and charge values, a macroscopical spatial nonhomogeneity in the degree of plasma ionization is observed. This results in the coordinate dependence of the plasma bulk potential and, therefore, of the zero reading.

Interaction of charged particles stimulates the formation of an ordered arrangement of particles in space. It was confirmed indirectly when the sampling of aluminum oxide particles from the flame onto a substrate was made [6, 17]. The processing of the same microphotos allowed us to construct the radial distribution function $G(r) = n_2(r)/n_1^2(r)$, where $n_1(r)$ and $n_2(r)$ are the unary and binary functions, respectively. When constructing the function $G(r)$, only the submicron-size particles were taken into account, so the dependence reflects a structurization of submicron-size particles in the field of larger ones. The availability of maxima indicates the existence of a long-range order interaction in the plasma under study, because the Debye screening length is about 0.1 μm, being much smaller than the average distance between particles (1.5 μm).

One of the reasons of *the long-range interaction* of charged particles at distances that exceed the effective range of Coulomb forces is the force of ionic pressure which arises owing to the non-uniform ionization of plasma near the particles [2]. Attaining an equilibrium at the particle–plasma interface depends on the gradient of the nonequilibrium parameter ψ, the presence of other particles, and the distances between them [16].

From the analysis of the Poisson equation, it follows that any function that is a solution of the Laplace equation $\Delta\varphi_0 = 0$ possesses all properties of the trivial solution. In the spherical coordinate system, this means that the plasma bulk potential can vary reciprocally to the coordinate. That is, the distribution of the plasma potential may look like

$$\varphi_{\rm pl}(r) = \varphi_{0s}\frac{r_p}{r} \quad (3)$$

around some particle with radius r_p that possesses the plasma bulk potential φ_{0s} at its surface. At the same time, plasma remains electroneutral everywhere where the measured value of the potential does not differ from that of $\varphi_{\rm pl}$:

$$n_e(\varphi_{\rm pl}) = n_{e0}\exp\frac{e\varphi_{\rm pl}(r)}{kT} = n_{i0}\exp\frac{-e\varphi_{\rm pl}(r)}{kT} = n_i(\varphi_{\rm pl}).$$

Since the nonequilibrium parameter ψ is linearly connected to the plasma bulk potential, its spatial dependence has to obey the same law. The value of this parameter at the surface of the particle depends only on the processes that take place here. Nevertheless, $\nabla\psi$ depends on the values of the nonequilibrium parameter at the surfaces of other particles that surround the selected one. The non-uniform distribution of $\nabla\psi$ in various directions from the particle surface towards other particles with various charges results in a non-uniform radial distribution of the plasma ionization degree and, consequently, the electron, ion, and atom concentrations. In accordance with that, the full momentum transferred to a particle of the condensed phase through collisions with plasma particles cannot be zero in some direction, which is determined by the radial asymmetry of $\nabla\psi$.

The force that acts upon the particle is determined only by the cumulative momentum, transmitted to the particle by ions and atoms due to collisions (the momentum transferred by electrons can be neglected, in view of their small mass). Calculating, by the conventional technique, the momentum transfer to the particle per unit of its surface, the surface density of force can presented in the form [18]

$$f = -\frac{e}{3\lambda}\frac{n_i}{N_a}\left(\nabla E + \frac{1}{kT}E^2 + \frac{1}{kT}E\nabla\psi_i\right), \quad (4)$$

where $\lambda = N_a^{-1/3}$ is the mean free path and $N_a = n_i + n_a$.

The value of the field strength E_s at the particle surface should be used in expression (4). For the examined distances between particles, this field is invoked by the particle itself and does not affect the interaction of particles with one another. Therefore, expression (4) is reduced to the formula

$$f(\varphi,\theta) = -\frac{en_{is}}{3\lambda N_a kT} E_s \nabla\psi_i(\varphi,\theta), \quad (5)$$

which determines the dependence of the force density on the φ and θ angles in the coordinate system of the selected particle.

Calculations provide the following values of $\nabla\psi$ along the axis that connects the centers of the micron- and submicron-size particles: $\nabla\psi_{s1} = 4 \times 10^5$ eV/m for the micron-size particle and $\nabla\psi_{s2} = -8 \times 10^7$ eV/m for the submicron-size one (the direction from the micron-size particle to the submicron-size one, i.e. from left to right, is assumed positive). In this case, there is a repulsive force between the particles. Submicron-size particles undergo the action of the same forces from other micron-size particles. Therefore, they are positioned at a half-distance between large particles, as is shown in the microphoto in work [17].

The collective interaction of condensed particles also manifests itself in *electro-acoustic oscillations*, which result from a charge fluctuation on the particle surface or when the particle is removed from its equilibrium position. The estimations of the natural frequency of oscillations gave the value of about 20 kHz [3]. For the experimental definition of the frequency spectrum of particle oscillations, a laminar diffusive flame with an aluminum powder containing particles with the average dimension equal to 4 μm was used [19].

The spectrum of low-frequency electro-acoustic oscillations in the smoky plasma of aluminum oxide occupies the frequency range from 10^4 to 2×10^5 Hz, with two characteristic frequencies with pronounced maxima at about 3×10^4 and 6×10^4 Hz [4]. The first maximum at 3×10^4 Hz has a large amplitude and rather a small dispersion. The halfwidth of the distribution function amounts to 7×10^3 Hz. The second maximum with a smaller amplitude has a larger halfwidth of 4×10^4 Hz.

Electrostatic properties of condensed particles influence the nucleation processes, in particular, they affect the critical dimensions of a nucleus. Taking the electrostatic energy of a charged nucleus into account in the expression for the free energy, the expression for the critical radius $R_{\rm cr}$ reads [10]

$$\frac{1}{R_{\rm cr0}} = \frac{1}{R_{\rm cr}} + \frac{\varepsilon_0\phi_s(\phi_s+\phi_{\rm pl})}{4\sigma R_{\rm cr}^2}, \quad R_{\rm cr0} = \frac{2\sigma V_l}{kT\ln(n/n_s)}, \quad (6)$$

where σ is the surface tension coefficient and $R_{\rm cr0}$ and $R_{\rm cr}$ are the critical radii of uncharged and charged nuclei, respectively.

There exists a range of the positive charge of a drop, where its critical radius decreases in comparison with that in the unperturbed state. One may assume that positively charged particles of dust are smaller than neutral and negative ones. When nuclei are formed in a medium that contains an excess number of electrons, e.g., due to an external source of β-rays, the critical radius of the charged nucleus increases with the increase of the electron concentration and the increase of the negative charge of the nucleus itself.

Now, let us consider *the basic directions and methods of application* of dusty and smoky plasmas.

1. Methods dealing with applying the smoky plasma properties and controlling its characteristics.

Thermal phenomena occurring in the plasma of combustion products serve as the sources of heat energy in furnace plants. In this case, smoke particles play a positive role in radiative heat transfer. However, slagging of the furnace walls complicates the process of burning and requires additional expenses. Thermal processes running at burning a fuel serve as the sources of momentum in rocket engines.

Electrophysical properties of smoky plasma are used in MHD- and EHD-generators, in MHD-pumps, for producing plasma with preset radiative and absorption characteristics. Here, particles can either promote the ionization processes, which depends on the character of interphase interactions, thus enhancing the degree of plasma ionization, or inhibit them.

The condensation of metal oxides in smoky plasma and the interrelation of this process with plasma properties created a unique opportunity for obtaining nano-size powders with preset characteristics [19]. Varying the conditions of the interphase interaction makes it possible to control not only the dimensions of particles, but their physical properties as well.

Oscillatory-wave processes in the condensed phase of plasma can be used for diagnostics. On the other hand, plasma with a condensed phase can be applied as a source of high-frequency acoustic waves.

2. Methods dealing with using smoky plasma that is formed in technological installations and devices.

Perspective seem the applications of plasma with a condensed disperse phase that is formed at the electroerosive processing of metal surfaces, electrospark doping, plasma-electrolytic polishing, and other methods of technological application of the electric-discharge phenomena in "metal–electrode" and "metal–electrolyte" systems [20]. To increase the efficiency of those processes, one must know the mechanisms of particle formation and the kinetics of their behavior in plasma upon the passage of large currents.

The plasma of an arc plasmotron, upon introducing powder-like materials, is used for the coating of solid surfaces to obtain their specific properties and protect them against corrosion.

It is of interest an opportunity to govern the process of formation of ultrathin films in electronic technology. If carbon materials are used, molecular complexes emerge in the form of fullerenes, which possess specific properties.

3. Methods dealing with applying the properties of dusty plasma that is observed in natural objects.

Dusty plasma is observed in the interplanetary space, planetary rings, circumsolar dusty rings, comet tails, as well as in interstellar molecular clouds. In the terrestrial atmosphere, typical features of dusty plasma are met in phosphorescing clouds in the arctic tropo- and mesoshere, in prethunderstorm clouds containing smoke-polluted air. Certainly, knowledge of properties of dusty plasma will allow the behavior of natural phenomena to be forecasted.

4. Methods of dusty plasma simulation for carrying out fundamental researches.

The artificial production of plasma in electric and high-frequency discharges made it possible to study its fundamental properties and characteristics [11]. An essential advantage in the simulation of physical phenomena was a discovery of plasma crystals [7, 8]. Laboratory dusty plasma is not only an exclusive physical model for studying the phase transitions and other properties of plasma crystals, but can also be used for the generation of oscillations and the creation of dusty formations with preset radiative and optical characteristics.

Thus, the unique properties and characteristics of dusty and smoky plasmas offer exclusive directions of their application.

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ПИЛОВА ТА ДИМОВА ПЛАЗМА. ДЕЯКІ ВЛАСТИВОСТІ ТА ЗАСТОСУВАННЯ

Г.С. Драган

Р е з ю м е

Проведено класифікацію методів застосування низькотемпературної плазми з конденсованою дисперсною фазою, різновидами якої є пилова і димова плазма. Описано деякі властивості димової плазми, пов'язані з умовами її одержання і складом, такі, як зміщення іонізаційної рівноваги, дальнодійна взаємодія, просторова неоднорідність іонізаційної равноваги навколо заряджених димових частинок. Проведено аналіз загальних і відмітних характеристик пилової і димової плазми.