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HEATING OF NANOPARTICLES IN LOW-PRESSURE PLASMA JETS

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The heating of nanoparticles in a low-pressure plasma jet was studied with the help of computer simulation. Modeling of the expansion of a plasma jet with a dispersed phase, which was a mixture of nanoparticles of two sizes, was carried out within the framework of a multi-fluid axisymmetric hydrodynamic model. As a result of the calculations, the spatial distributions of the plasma parameters at different times after the plasma jet injection were obtained. The simulation results show that the temperature of nanoparticles in the plasma jet depends not only on their size, but also on the percentage composition of the mixture of dust particles. The reason for this is the influence of the size of nanoparticles on the spatial distribution of ion concentration, which play a decisive role in the heating of dust particles due to recombination on their surfaces.

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INTRODUCTION

Chemically active low-temperature plasmas have been known to be efficient sources of nanoparticle formation for quite a while. Nanoparticles are applied in various industries because of their specific properties and high surface/volume ratios. In particular, an important application of nanoparticles is the creation of thin nanostructured films on substrates. Recently, a new process, which uses a plasma torch operating at low pressure has been developed with the aim of depositing uniform thin layers on large surfaces [1, 2]. In this plasma spraying process plasma jets are used as a heat sources to melt and accelerate the injected nanoparticles which subsequently impinge and solidify on a substrate. To create nanostructured films with the required properties, it is necessary to be able to control the parameters of nanoparticles in the flow, in particular their energy, temperature and charge.

Nanoparticles produced in low-temperature plasmas often have crystalline structure, which suggests high temperatures during synthesis [3], a puzzling aspect considering that the gas temperature in low-pressure plasmas is often close to room temperature.

Simulation of the expansion of plasma jets in rarefied gas was performed in [4], where it was shown that dust particles in plasma jets can be heated to a high temperature, that is much higher than the plasma temperature. This heating is associated with several mechanisms, namely the recombination of ions on dust particles, the bombardment of ions and electrons on the surface of dust particles. In addition, dust particles in the stream are cooled due to collisions with neutral atoms. The balance of these processes determines the temperature of dust particles, which depends on their size.

In this paper we will analyze the energy exchange of dust particles with plasma, if the plasma jet contains dust particles of two different sizes.

1. MODEL AND SIMULATION METHOD

In this paper, the expansion of an axially symmetric plasma jet with nanoparticles into a rarefied neutral gas is studied. It is assumed that the plasma velocity V_0 and its density ρ_0 are constant at the inlet during the plasma jet expansion. The plasma considered in this article consists of electrons, neutral argon atoms, singly ionized argon ions and nanoparticles that differ in size: radius of small particles is $r_{d1} = 5$ nm and radius of large particles is $r_{d2} = 5$ nm. In the model ions, electrons and neutral atoms have the same drift velocity $\vec{w} = (u, v)$ due to effective momentum exchange, and dust particles have drift velocity $\overrightarrow{w_d} = (u_d, v_d)$. Here u, u_d are radial velocity components and v, $v_d$are axial velocity components.

The ions temperature equals to neutral atoms temperature T, but electrons temperature T_e can differ from them.

The continuity equation for heavy plasma component (ions and neutral atoms) is equal to

$$\frac{\partial n}{\partial t} + div\left(n\vec{w}\right) = 0. \tag{1}$$

In equation (1) n is sum of ion density n_i and neutral atom density n_a.

The continuity equation for ions is equal to

$$\frac{\partial n_i}{\partial t} + div\left(n_i\vec{w}\right) = -\sum_{\alpha=1}^{2} I_{i\alpha} n_{d\alpha} / e, \tag{2}$$

where n_e is electron concentration, $n_{d\alpha}$ is concentration of dust particles of type α. The right hand side in (2) describes the ion destruction due to recombination at the interaction with dust particles. The ion current on dust particle of type $\alpha I_{i\alpha}$ is described by OLM theory [5].

The continuity equations for dust particles is equal to

$$\frac{\partial n_{d\alpha}}{\partial t} + div\left(n_{d\alpha}\vec{w}_d\right) = 0\ . \tag{3}$$

The momentum equations for heavy plasma particles (ions and atoms) are given by

$$\frac{\partial(nu)}{\partial t}+div(nu\vec{w})=-\frac{1}{m_i}\frac{\partial P}{\partial r}-\frac{\sum_{\alpha=1}^{2}n_{d\alpha}f_{r\alpha}}{m_i}+\frac{e}{m_i}n_iE_r, \quad (4)$$

$$\frac{\partial(nv)}{\partial t}+div(nv\vec{w})=-\frac{1}{m_i}\frac{\partial P}{\partial z}-\frac{\sum_{\alpha=1}^{2}n_{d\alpha}f_{z\alpha}}{m_i}+\frac{e}{m_i}n_iE_z, \quad (5)$$

where $P=nkT$ is the plasma pressure, E_r and E_z are radial and axial electric field components, m_i is ion mass (in our case the neutral atom mass is equal to the ion mass). In equations (4), (5) $\vec{f}_\alpha$ is force of the aerodynamic interaction between plasma and dust particle of type α ($f_{r\alpha}$ and $f_{z\alpha}$ its components along axis r and z). It consists of a friction force between dust particles and neutral particles $\vec{f}_{dn}^{\alpha}$, as well as between the ions and the dust particles $\vec{f}_{di}^{\alpha}$.

According [5] neutral drag force can be approximate as

$$\vec{f}_{dn}^{\alpha}=\frac{8}{3}\sqrt{2\pi}r_{d\alpha}^{2}n_a mV_{Tn}(\vec{w}-\vec{w}_d), \quad (6)$$

where $V_{Tn}=(w^2+8kT/\pi m)^{1/2}$ is total atom speed (a combination of directed and thermal speeds). The ion drag force can be expressed as [5]

$$\vec{f}_{di}^{\alpha}=n_i mV_{Tn}\sigma_{col}^{\alpha}\vec{w}+n_i mV_{Tn}\sigma_{coul}^{\alpha}\vec{w}. \quad (7)$$

In (6), (7) σ_{col}^{α} (σ_{coul}^{α}) is the momentum collision cross section corresponding to the collection of ions by direct ion impacts (electrostatic Coulomb collisions).

The momentum equations for dust particles are given by

$$\frac{\partial(\rho_d u_d)}{\partial t}+div(\rho_d u_d\vec{w}_d)=\sum_{\alpha=1}^{2}n_{d\alpha}f_{r\alpha}+\sum_{\alpha=1}^{2}q_{d\alpha}n_{d\alpha}E_r, \quad (8)$$

$$\frac{\partial(\rho_d v_d)}{\partial t}+div(\rho_d v_d\vec{w}_d)=\sum_{\alpha=1}^{2}n_{d\alpha}f_{z\alpha}+\sum_{\alpha=1}^{2}q_{d\alpha}n_{d\alpha}E_z. \quad (9)$$

Equations for internal energies ions and atoms ε, electrons ε_e and dust particles $\varepsilon_{d\alpha}$ are given by

$$\frac{\partial(\rho\varepsilon)}{\partial t}+div(\rho\varepsilon\vec{w})+P\,div\,\vec{w}=Q_{ei}+Q_{en}-n_dQ, \quad (10)$$

$$\frac{\partial(\rho\varepsilon_e)}{\partial t}+div(\rho\varepsilon_e\vec{w})+P_e\,div\,\vec{w}+div\,q_e=$$
$$-Q_{ei}-Q_{en}-\sum_{\alpha=1}^{2}n_{d\alpha}Q_{ed}^{\alpha}, \quad (11)$$

$$\frac{\partial(\rho_{d\alpha}\varepsilon_{d\alpha})}{\partial t}+div(\rho_{d\alpha}\varepsilon_{d\alpha}\vec{w}_d)=n_{d\alpha}\left(Q_{ad}^{\alpha}+Q_{id}^{\alpha}+Q_{ed}^{\alpha}\right). \quad (12)$$

In (12) $\rho_{d\alpha}=n_{d\alpha}m_\alpha$ is the density of dust particles of type α, in (10) P, P_e are partial pressures of the heavy plasma component and electrons, in (8), (9) ρ_d is total density of the mixture of dust components. In equations (10)-(12) $Q_{ad}^{\alpha}, Q_{ed}^{\alpha}, Q_{id}^{\alpha}$ are the energy exchanges between a dust particleof type α and neutral atoms, electrons and ions [6, 7], Q_{ei} is the energy exchange between electrons and ions, Q_{en} is the energy exchange between electrons and neutrals. The heat flux of electrons in (11) is given by $\vec{q}_e=-\chi(T_e)\nabla T_e$, where $\chi(T_e)$ is the coefficient of electron thermal conductivity.

The equation of conservation of momentum for electrons if we neglect their inertia, as well as the force of friction of the electrons with atoms, ions and dust particles has the form

$$en_e\vec{E}=-\nabla P_e. \quad (13)$$

Relation (13) expresses the electric field $\vec{E}$.

The system of equations (1)-(13) is solved numerically by the method of large particles [8].

2. RESULTS AND DISCUSSION

The numerical simulations of the plasma jet expansion were carried out with the following parameters at the inlet: the plasma flow velocity $v_0=40$ m/c, the radius of the inlet $R_0=0.01$ m, the ratio of the total density of the dust component to the density of the plasma $\rho_d/\rho_0=0.1$, plasma pressure $P_0=40$ Torr. The degree of plasma ionization was $\alpha=5\cdot10^{-5}$. Calculations were performed at different ratios of densities small and large dust particles ρ_{d1}/ρ_{d2} in the plasma jet.

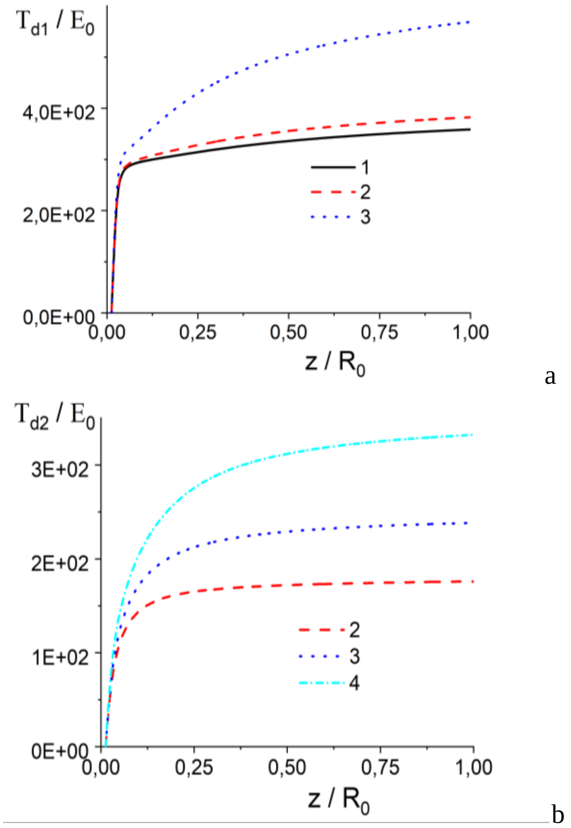
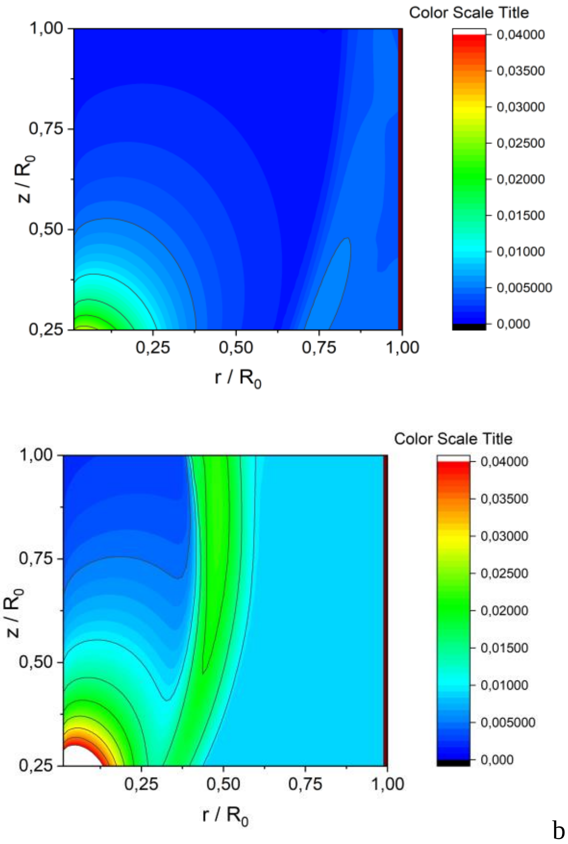


Fig. 1. Spatial temperature distributions of small dust particles (a) and big dust particles (b) on jet axis for different compositions of the mixture of dust components (curves 1 – $\rho_{d1}=\rho_d$, $\rho_{d2}=0$; 2 – $\rho_{d1}=0.8\rho_d$, $\rho_{d2}=0.2\rho_d$; 3 – $\rho_{d1}=0.2\rho_d$, $\rho_{d2}=0.8\rho_d$; 4 – $\rho_{d1}=0$, $\rho_{d2}=\rho_d$)

Fig. 1 shows spatial temperature distributions of small dust particles (a) and large dust particles (b) on the jet axis. Here the temperature is normalized to energy $E_0=m_iv_0^2$.

As it was shown in [4], the temperature of dust particles in a plasma jet at low pressures is determined by the balance of energy flows to its surface, the main ones of which are caused by collisions of ions, hot electrons and atoms with dust particles. When ions collide with dust particles, their recombination occurs, and the ionization energy is transferred to the surface of the dust particle. When atoms collide with dust, the dust cools down as a result of energy exchange, since the temperature of the plasma is usually lower than the surface temperature of the dust particles. Therefore, significant heating of nanoparticles is observed near the inlet, where the concentration of ions and electrons is high. When the coordinate increases, the process of heating the dust slows down, which is associated with a rapid decrease in the concentration of ions and electrons during the expansion of the plasma jet.



a

b

Fig. 2. Spatial distributions of the plasma density in the case when the plasma jet contains particles with a radius r_d=5 nm (a) and r_d=50 nm (b)

Fig. 1 also shows that the temperature of small dust particles increases when large particles are added to the mixture of dust particles. On the contrary, the temperature of large dust particles decreases when small particles are added to the mixture.

To illustrate this statement, consider the spatial distributions of the plasma density in different calculation modes that differ in the percentage composition of the dust mixture.

Fig. 2 shows the spatial distributions of the plasma density for cases when the plasma jet contains particles of the same size. It can be seen that if small nanoparticles are present in the jet (see Fig. 2,a), it expands in the radial direction much more than in the case when nanoparticles of a larger size are present in it (see Fig. 2,b).

This result is also confirmed by the radial distributions of the plasma density, which are shown in Fig. 3 for different compositions of the mixture of nanoparticles at a distance $z = 0.05\ m$ from the inlet.

This figure also indicates that the plasma density in the axial region of the jet increases as the fraction of large nanoparticles in the mixture increases, provided the mass of the dust component is the same.

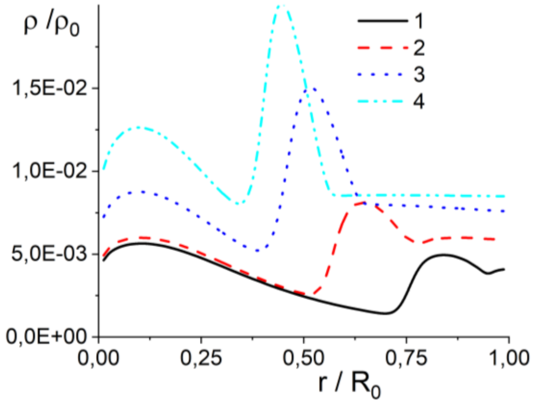


Fig. 3. Radial distributions of plasmadensity at $z = 0.05$ m for the same mixture composition of dust component as in Fig. 1

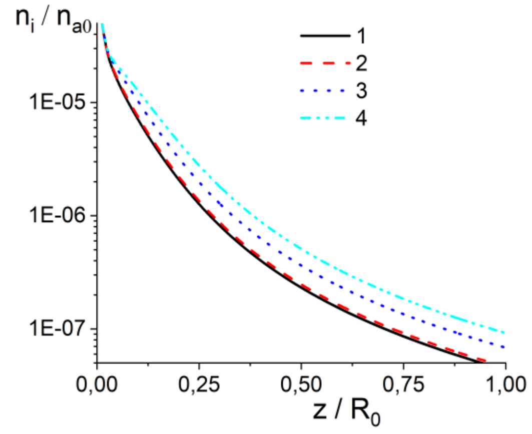


Fig. 4. Spatial distributions of ion density on the jet axis (curves 1-4 corresponds cases indicated in Fig. 1)

Nanoparticles have the same effect on ion density. This can be seen from Fig. 4, which shows the axial distributions of the ions density for the mixture compositions indicated in Fig. 1. Here n_{a0} is neutral atoms concentration on the inlet.

Thus, with an increase in the proportion of large dustparticles, the concentration of ions in the central part of the plasma jet increases.As a result, the flow of ions to the dust particles increases, and therefore the energy flow on their surface due to recombination increases.

CONCLUSIONS

The expansion of a low-pressure plasma jet containing nanoparticles of various sizes and the heating of nanoparticles in it were studied. The main results can be summarized as follows:

When the proportion of large dust particles in the dust mixture increases, the concentration of ions in the axial region of the plasma jet increases.

An increase in the concentration of ions leads to an increase in the temperature of nanoparticles due to an increase in the number of ion recombination processes when they collide with dust particles.

As a consequence of the influence of the size of dust particles on the parameters of the plasma jet, with an increase in the proportion of large particles in the dust mixture, the temperature of small particles increases, and the temperature of large particles decreases.

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НАГРІВАННЯ НАНОЧАСТИНОК У ПЛАЗМОВИХ СТРУМЕНЯХ НИЗЬКОГО ТИСКУ

О.Ю. Кравченко, І.С. Марущак

За допомогою комп'ютерного моделювання досліджено нагрівання наночастинок у струмені плазми низького тиску. Моделювання розширення плазмового струменя з дисперсною фазою, яка являла собою суміш наночастинок двох розмірів, виконано в рамках багаторідинної осесиметричної гідродинамічної моделі. В результаті розрахунків отримано просторові розподіли параметрів плазми в різні моменти часу після інжекції плазмового струменя. Результати моделювання показують, що температура наночастинок у плазмовому струмені залежить не тільки від їх розміру, але й від процентного складу суміші частинок пилу. Причиною цього є вплив розміру наночастинок на просторовий розподіл концентрації іонів, які відіграють вирішальну роль у нагріванні частинок пилу внаслідок рекомбінації на їх поверхні.