CONTRIBUTION OF CHARGE TRANSFER PROCESSES TO HEAT TRANSFER IN THE TREATMENT OF METALLIC PARTICLES IN A LOW-DENSITY PLASMA JET

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It is shown that the effectiveness of the plasma treatment of dispersed metallic particles is due to ionization of the gas, the participation of electrons in transfer processes, and electrostatic charging of the particles.

The thermal action of a low-temperature plasma on dispersed particles lies at the basis of a number of technical applications [1]. Many of the studies which have been devoted to modeling these processes have investigated phenomena which determine the specific features of plasma treatment – phenomena such as heat transfer in the presence of large temperature gradients [2, 3], rarefaction of the gaseous medium [4], high flow velocities [2, 5], etc. One of the most important effects is connected with ionization of the gas and the presence of charges – electrons and ions – in the flow together with neutral molecules. Different approaches [6-12] have been taken to account for the features of plasma treatment due to the presence of a space charge. The authors of [10-12] made an analogy between the behavior in a plasma of objects such as spherical particle, an isolated electrostatic probe, and a manmade satellite in the ionosphere. The laws governing the interaction of these bodies with the plasma are connected with the accumulation of charges on them and the creation of local electric fields which in turn influence the motion of the electrons and ions. The methods of molecular-kinetic theory [13] have proven to be effective for describing the interaction of bodies with low-density ionized gases. These methods have been successfully employed in problems of probe diagnostics [14] and astronautics [15]. However, these investigations have focused mainly on charge distribution and transfer rather than thermal effects, and it is these effects that are of the primary interest in the plasma treatment of materials.

Formidable mathematical difficulties are encountered in attempts to solve the kinetic problem in general form, and an analytical description of the effect of a plasma on particles is possible only in certain limiting cases. Here, we analyze the role of charge transfer processes in heat transfer by using the example of the treatment of metallic particles in a low-density ($\mathrm{Kn} = \ell/P_p \gg 1$) subsonic ($S = V/(2kT_{h\infty}/m_h)^{1/2} < 1$) low-temperature plasma jet accompanied by strong ($\xi_D = R_p/r_D \gg 1$) and weak ($\xi_D < 1$) Debye shielding. The restrictions imposed allow us to ignore distortions of the spherically-symmetric electrostatic field near a metallic particle due to "shadow" effects. These distortions are smoothed out by the random thermal motion of the electrons and ions. Also, since their collision cross sections with the particles in strongly- and weakly-shielded plasmas depend only on the surface potential, when we determined the flows of mass, momentum, and energy in the free-molecular regime we used familiar Maxwell distributions for velocity in the undisturbed region of a plasma jet moving relative to a particle with the velocity $V = V_g - V_p$. For the same reason, we also used Maxwell distributions for molecules, electrons, and ions scattered by the surface of the particle.

Due to the substantial difference in the mean thermal velocities of electrons and ions ($\bar{v}_e/\bar{v}_i \sim (m_i/m_e)^{1/2} \gg 1$), a metallic particle in a plasma obtains an excess negative potential $\varphi_p = \varphi_f$. The magnitude of this potential is determined from the condition of equality of the flows of electrons and ions collected by the particle $\Gamma_e^-(\varphi_f) = \Gamma_i^-(\varphi_f)$.

Standard calculations (see [11], for example) make it possible to obtain expressions for the flows of charge and heat transmitted to a particle by molecules, electrons, and ions:

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$$I_j^- = 4\pi R_p^2 N_{j\infty} (kT_{j\infty}/2\pi m_j)^{1/2}\, j_j^-,$$
$$Q_j = Q_j^- - Q_j^+ = 4\pi R_p^2 P_{j\infty} (2kT_{j\infty}/\pi m_j)^{1/2} q_j. \tag{1}$$

The dimensionless fluxes j_j^- and q_j in (1) for a negatively-charged particle in a low-density subsonic jet are represented as follows:

$$j_e^- = \exp(-y_f), \quad j_i^- = 1 + \tau y_f + \frac{1}{3} S^2 (1 - \tau y_f),$$
$$q_a = 1 + S^2 - \left(1 + \frac{1}{3} S^2\right)\tau_s, \quad q_e = \left(1 + \frac{1}{2} y_f + \frac{1}{2} w_e\right) \exp(-y_f), \tag{2}$$
$$q_i = 1 + \frac{1}{2}\tau y_f + S^2 \left(1 + \frac{1}{6}\tau y_f\right) +$$
$$+ \left[1 + \tau y_f + \frac{1}{3} S^2 (1 - \tau y_f)\right] \left(\frac{1}{2} w_i - \tau_s\right).$$

Here $y_f = -e\varphi_f/kT_{e\infty}$, $\tau_s = T_s/T_{h\infty}$, while the parameter τ takes the value $\tau = 0$ at $\xi_D >> 1$ and $\tau = T_{e\infty}/T_{i\infty}$ at $\xi_D < 1$, $w_j = W_j/kT_{j\infty}$, where $W_a = 0$, $W_e = \Phi_e$, $W_i = E_i - \Phi_e - e\varphi_f$. In the relations presented above, it was considered that along with kinetic energy, the recombination of electrons and the neutralization of ions on the particle surface results in the transfer of the energy associated with their charge state – the work function Φ_e and the effective ionization energy at the surface $E_i' = E_i - \Phi_e$, respectively. Emission of electrons by the particle is not taken into account here. For metallic particles, thermionic processes become important at temperatures greater than melting point only when the concentration of charge carriers in the plasma is very low. The latter prevails either when the plasma is of very low density or when the degree of ionization η is very low (and the role of plasma electrons and ions in heat transfer can thus be ignored). Simple estimates based on the use of th Richardson formula make it possible to determine the conditions under which thermoelectrons begin to make a significant contribution to the total charge flow. For example, for metallic particles with the temperature T_s = 2500-3000 K in an argon plasma at normal pressure $P_g = 10^5$ Pa, this occurs at $T_g \lesssim 8000$ K ($\eta \approx 6 \cdot 10^{-4}$). For a plasma with the temperature T_g = 10,000 K, it occurs at $P_g \lesssim 10^2$ Pa ($\eta \approx 0.25$). Features of the behavior of emitting particles in a plasma were examined in [6, 7, 16-18].

The expressions for the fluxes contain small squared corrections for velocity $\sim S_j^2$. Thus, in (2) we have omitted the terms which include the velocity ratio for the electrons $S_e \ll S_h \equiv S$. The equilibrium (floating) potential of the particle depends slightly on the velocity of the jet. In a subsonic (S = 0-1) argon plasma, for example, y_f = 5.6-5.3 at $\xi_D \gg 1$ and y_f = 4.0-4.2 at $\xi_D < 1$. An important feature connected with the features of plasma treatment due to ionization is the fact that the energy of electrostatic interaction between electrons and ions on the one hand and charged particles on the other hand is considerably greater than the mean energy of their thermal motion ($y_f > 1$).

At high particle temperatures, it is also necessary to consider vaporization and radiative losses of the particle. The corresponding mass and energy fluxes are written in the form

$$I_v^+ = 4\pi R_p^2 N_{vs} (kT_s/2\pi m_v)^{1/2},$$
$$Q_v^+ = 4\pi R_p^2 P_{vs} (2kT_s/\pi m_v)^{1/2} \left(1 + \frac{1}{2} w_v\right), \quad Q_r^+ = 4\pi R_p^2 \varepsilon_T \sigma T_s^4. \tag{3}$$

Here $w_v = L_v/kT_s$, $N_{vs} = P_{vs}/kT_s$, P_{vs} is the vapor pressure above the curved surface of the particle determined from the Clausius–Clapeyron condition with allowance for the Kelvin correction; ε_T is the absorptivity of a metallic particle.

The force acting on a metallic particle in a plasma flow is connected with the transfer of momentum by the heavy particles in the plasma – molecules [13] and ions [11, 15]. the contribution of electrons to the resistance force is negligible. The total drag $F = F_a + F_i$ is made up of the components due to the surface collision (F_{sh}) and reflection (F_{rh})

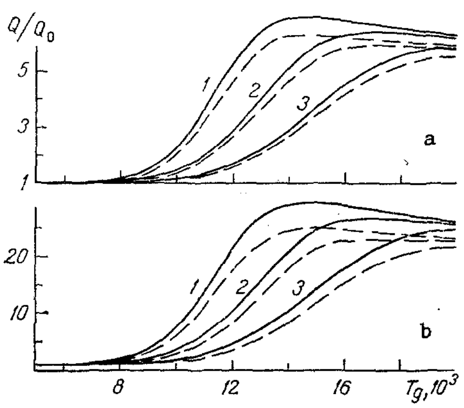

Fig. 1. Ratio of the heat fluxes Q/Q_0 on a metal particle (T_p = 1000 K) with and without allowance for ionization in relation to the temperature T_g of a low-density argon plasma jet (a) $\xi_D \gg 1$, b) $\xi_D < 1$; 1) P_g = 0.001 MPa; 2) 0.01; 3) 0.1; solid lines – $V = V_g - V_p = 0$; dashed lines – V = 1000 m/sec. T_g, K.

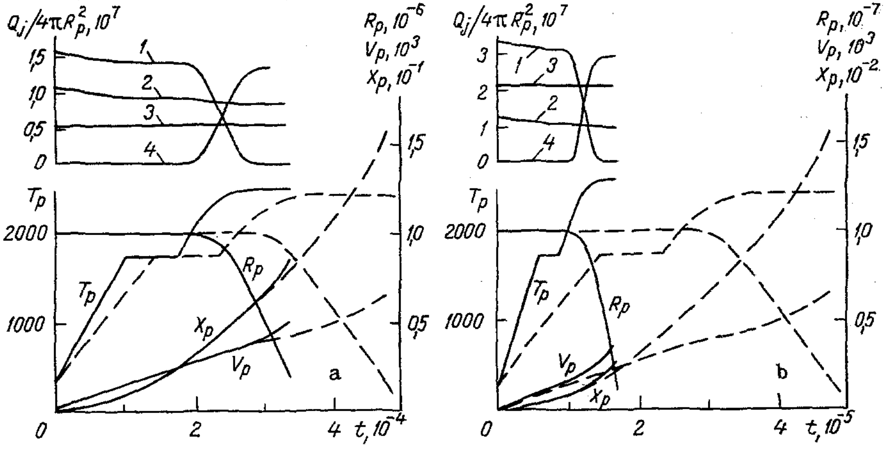

Fig. 2. Heating, melting, and vaporization of particles of nickel in a low-density argon plasma jet with P_g = 0.01 MPa, I_g = 10,000 K, V_g = 1000 m/sec (a: R_{p0} = 1 μm; b: 0.1): 1) Q, 2) Q_a; 3) $Q_e + Q_i$; 4) Q_v^+; solid lines: calculation with allowance for ionization; dashed lines: without allowance. $Q_j/4\pi R_p^2$, W/m²; R_p, X_p, m; V_p, m/sec; t, sec.

of molecules and ions and to Coulomb interaction (F_{Ci}) between a charged particle and ions moving along curved paths and not colliding directly with the particle. The components of the drag force can be represented in the form

$$F_{kh} = \frac{16}{3}\pi^{1/2}R_p^2 P_{h\infty} S\psi_{kh}, \quad k = s,\ r,\ C,\ h = a,\ i. \tag{4}$$

At S < 1, the dimensionless functions ψ_{kh} are determined by the relations:

$$\psi_{sa} = 1, \quad \psi_{ra} = \frac{\pi}{8}\tau_s^{1/2}, \quad \psi_{si} = 1 + \frac{1}{2}\tau y_f,$$
$$\psi_{ri} = \frac{1}{4}\tau_s^{1/2}\left[\frac{2\pi^{1/2}}{3}(\tau y_f)^{3/2} - \frac{\pi}{2}\tau y_f + \pi^{1/2}(\tau y_f)^{1/2} + \right.$$
$$\left. + \frac{\pi}{2}\left(1 + \frac{1}{2}\tau\right) + \dots\right], \tag{5}$$

$$\psi_{Ci} = \frac{1}{2}(\tau y_f)^2 L_C \exp(-\xi_D^2 \tau y_f),$$

where

$$L_C = \ln\{[(e\varphi_f/m_i\bar{v}_i^2)^2 + (r_D/R_p)^2]^{1/2}/(1 - e\varphi_f/m_i\bar{v}_i^2)\}; \quad \bar{v}_i = (8kT_{i\infty}/\pi m_i)^{1/2}.$$

The effect of charge transfer processes on the effectiveness of plasma treatment can be evaluated using the simple example of the treatment of thermally thin (Bi ≪ 1, $T_p \simeq T_s$) metal particles in a low-density isothermal ($T_{j\infty} = T_g$ = const) low-temperature plasma jet. The equations of motion, heating, and vaporization of a particle are written as follows

$$\frac{4}{3}\pi R_p^3 \rho_p \frac{aV_p}{dt} = F \equiv F_a + F_i,$$

$$\frac{4}{3}\pi R_p^3 \rho_p C_p \frac{dT_p}{dt} = Q \equiv Q_a + Q_e + Q_i - Q_v^+ - Q_r^+, \tag{6}$$

$$4\pi R_p^2 \rho_p \frac{dR_p}{dt} = -m_v I_v^+$$

with the initial conditions $V_p = 0$, $T_p = T_{p0}$, $R_p = R_{p0}$ at $t = 0$. The optical thickness and dust content of the jet are assumed to be low, so we do not consider the radiant flux of the plasma, the mutual effect of the particles, or the disturbance of ionization equilibrium due to the presence of a readily-ionized component (metal vapors) in the plasma jet.

The electron and ion flows make a substantial contribution to the overall heat balance even in the case of low degrees of ionization. Figure 1 shows the ratio of the fluxes incident on a metal particle Q/Q_0 calculated with and without allowance for ionization, respectively, when the plasma is regarded as a molecular gas with the same temperature T_g and pressure P_g. The high efficiency of heat transfer in the collision of electrons and ions with particles is due to the fact that the energies released on the surface W_e and W_i are considerably greater than the mean energy associated with thermal motion. The role of plasma phenomena with weak Debye shielding is particularly large for small particles, when the effective cross section for collisions between ions and charged particles is linearly connected with the potential of the particle and is considerably greater than the geometric cross section. For this reason, in a weakly-shielded plasma, there is an increase in the drag on the particle compared to the case of a molecular gas.

Figure 2 shows results of calculations of the time dependences of the heat flux Q_j, temperature T_p, radius R_p, velocity V_p, and coordinate X_p of nickel particles in a rarefied argon jet (in the example being examined, $\eta \simeq 0.027$, $r_D \simeq 0.15$ μm, $\ell_a \simeq 24$ μm, $\ell_i \simeq 11$ μm, $\ell_c \simeq 1300$ μm). The calculations were performed with and without allowance for plasma effects. The dynamics of the plasma treatment is determined by the rates of heat transfer to and from the particles and the total heat fluxes in each of these directions. Despite the low degree of ionization ($\eta \sim 3\%$), electrons and ions account for a significant fraction of the energy transmitted to the particles. Also, the rate of heating of the particles in the initial section of the jet is markedly higher in the plasma than in the molecular gas. Heat removal is intensified with an increase in particle temperature, while the heating rate is slowed. Calculations show that the energy losses associated with radiation are significant only for particles of refractory metals with a high boiling point (such as tungsten) and are not important in the given case. The laws governing the behavior of the particles in the flow are connected with their vaporization at temperatures above the melting point, vaporization becoming the main channel for heat loss in this case. Due to the decrease in radius, a particle in the plasma jet undergoes rapid acceleration. Intensive vaporization of the particle occurs in the quasisteady regime with a nearly constant temperature T_v. Thus, all of the energy transferred from the plasma to the particle is expended on phase transformation. An important feature of th eprocess is the substantial difference between the particle vaporization temperature T_v found from the condition $Q(T_v) = 0$ ($T_v = 2480$ K at $R_{p0} = 1$ μm and 2570 K at $R_{p0} = 0.1$ μm) and the boiling point of the material T_b (at $P_g = 0.01$ MPa, $T_b = 2664$ K for Ni). Failure to allow for ionization leads to smaller theoretical values of T_v and – due to the heavy exponential dependence of vaporization rate on temperature – to significantly overstated treatment times. These differences, due to plasma effects, are largest for submicron metallic particles with $R_p < r_D$.

Thus, heat and mass exchange between particles and a plasma flow are appreciably influenced by charge transfer processes and electrostatic charging of the particles. The high efficiency of plasma treatment of particles is due to the participation of electrons and ions in transer processes, the contribution of the energy of the charge state to heat transfer, and the fact that the geometric cross section of the electrons and ions differs considerably from the cross section for their collisions with the charged particles.

NOTATION

Bi, Biot number; C, heat capacity; e, electron charge; E_i, ionization energy; F, drag; I, flux of plasma particles, vapor flow; k, Boltzmann constant; $Kn = \ell/R_p$, Knudsen number; ℓ, mean free path; L_v, heat of vaporization referred to one molecule; m, mass; N, theoretical concentration; P, pressure; Q, heat flux; $r_D = (kT_{e\infty}/8\pi e^2 N_{e\infty})^{1/2}$, Debye shielding length; R_p, particle radius; $S_j = V/(2kT_{j\infty}/m_j)^{1/2}$, velocity ratio; t, time; T, temperature; $\bar{v}$, mean thermal velocity of plasma particles; V, velocity of jet, particle; X_p, coordinate of a particle in the jet; $y_f = -e\varphi_f/kT_{e\infty}$, dimensionless potential of a particle; $\eta = N_{i\infty}/(N_{\alpha\infty} + N_{i\infty})$, degree of ionization; $\xi_D = R_p/r_D$, Debye shielding parameter; ρ, density; σ, Stefan–Boltzman constant; φ, potential; Φ_e, work function. Indices: α, molecules; e, electrons; i, ions, g, plasma, gas; h, heavy particle of the plasma (molecules and ions); v, vapor; b, boiling; r, radiation, reflection; p, particle; s, surface; ∞, value away from a particle in an undisturbed region of the plasma; +(−), in the direction away from (toward) the particle.

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