

AEROSOL FORMATION IN THE VICINITY OF CARBON PARTICLE EVAPORATING UNDER THE ACTION OF LASER RADIATION

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Mathematical statement of the problem of evaporation of individual spherical high-melting particle in air accounting for recondensation of evaporated matter is described in this paper. An algorithm of numerical realization of the problem is presented. It was found that a cloud of submicron aerosol of high concentration forms in the vicinity of the particle. The results are discussed.

As of the present time, nonlinear effects appearing during combustion of carbon particles in the field of high-power laser radiation have been studied fairly well.¹⁻⁴ At intensities preceding the optical breakdown the nonlinear effects can also be related to evaporation of particle material^{5,6} with subsequent recondensation of the evaporated matter. The present paper deals with homogeneous condensation of supersaturated vapor in the vicinity of evaporating high-melting particle. The problem is specified by the presence of high temperature gradients that does not allow generalization of the results on recondensation obtained for liquid-droplet aerosol.⁷

MATHEMATICAL FORMULATION OF THE PROBLEM

Let us consider an individual carbon spherical particle of radius R_0 free-suspended in immovable air. The particle matter starts its evaporation heated with laser radiation of intensity I_0 to temperature higher than 3700 K. The analysis of characteristic times of the process revealed possible usage of quasistationary spherical-symmetric approximation of the problem of evaporation of an individual high-melting particle with regard to homogeneous recondensation of the evaporated matter.

For components of heterogeneous mixture we write the continuity (1)–(3) and heat balance (4) equations

$$\frac{1}{r^2} \frac{d}{dr} \left(-r^2 \rho_0 v y_1 + \rho_0 D(T) r^2 \frac{dy_1}{dr} \right) = \frac{dM}{dt} (r, y_1) \frac{1}{m_0}; \quad (1)$$

$$\frac{1}{r^2} \frac{d}{dr} \left(-r^2 \rho_0 v y_2 + \rho_0 D(T) r^2 \frac{dy_2}{dr} \right) = 0; \quad (2)$$

$$\frac{1}{r^2} \frac{d}{dr} \left(-r^2 \rho v c_{son}(r) \right) = -\frac{1}{m_0} \frac{dM}{dt} (r, y_1); \quad (3)$$

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \lambda_0(T) \frac{dT}{dr} + c_{p1} T (Q - q(r, y_1)) \right) + \rho(r) = 0. \quad (4)$$

The system of equations (1)–(4) is supplemented by boundary conditions

$$\left\{ \begin{aligned} 4\pi R_0^2 \rho_0 v y_1 - 4\pi R_0^2 \rho_0 D(T) \frac{dy_1}{dr} &= J_1, & (5) \\ 4\pi R_0^2 \rho_0 v y_2 - 4\pi R_0^2 \rho_0 D(T) \frac{dy_2}{dr} &= 0, & (6) \\ y_1(r = \infty) &= 0, & (7) \\ y_2(r = \infty) &= y_{2\infty}, & (8) \\ c_3(r = R_0) &= 0, & (9) \\ T(r = R_0) &= T_S, T(r = \infty) = T_0. & (10) \end{aligned} \right.$$

Here r is the running coordinate (the origin of coordinates coincides with the centre of evaporating particle); ρ_0 is the concentration of vapor-gas constituent of the heterogeneous mixture (in $1/m^3$); ρ_i is the concentration of the i th constituent of the heterogeneous mixture ($i=1$ is the carbon vapor C; $i=2$ is the air; and, $i=3$ is the condensed carbon); $y_i = \rho_i/\rho_0$ ($i=1, 2$) is the dimensionless concentration of vapor-gas mixture; $c_i = \rho_i/\rho$ is the dimensionless concentration of heterogeneous mixture constituents; ρ is the concentration of heterogeneous mixture; ρ_3 is the total number of condensed matter atoms per unit volume; m_0 is the mass of carbon atom; v is the velocity of Stefan flow ($v \ll c_{son}$, c_{son} is the sonic velocity); D is the coefficient of interdiffusion; dM/dt is the rate of vapor mass decrease per unit volume due to condensation; T is the temperature; λ_0 is the coefficient of thermal conductivity of vapor-gas mixture; c_{p1} is the carbon heat capacity on a single-atom basis; $Q = 4\pi r^2 \rho v$ is the total flow of mixture through a sphere of radius r ;

$q(r, y_1) = \frac{4\pi}{m_0} \int_{R_0}^r \frac{dM}{dt} (r, y_1) r^2 dr$ is the number of vapor

atoms condensed per unit time; and, $p(r)$ is the volume energy density per unit time due to heat release at phase transition and absorption of laser radiation energy by condensed aerosol. The particle surface temperature T_S is unambiguously determined by intensity of incident radiation and radius of evaporating particle. A flow of number of carbon atoms J_1 from the particle surface (a boundary condition (5)) is determined by kinetics of transport in the Knudsen layer. In the limiting case of solid medium under condition of low concentration of vapor $y_2/y_1 \gg 1$ the molecule flow J_1 is determined from the expression⁸

$$J_1 = 9.4 \pi R^2 (k_B T_S / 2\pi m_0)^{1/2} (\rho_1 - \rho_{sat}) Kn / \beta^2,$$

where k_B is the Boltzmann constant; ρ_{sat} is the concentration of saturated carbon vapors; Kn is the Knudsen number; $\beta = L/k_B T_S$ is the ratio of phase transition heat to the mean thermal energy of molecules.

The decrease of vapor phase is described by vapor condensation rate per unit time per unit volume

$$\frac{dM}{dt}(r, y_1) = \rho_c \int_{a_{cr}}^{\infty} 4\pi a^2 \frac{da}{dt}(r, y_1) f(t, r, a) da + \frac{4}{3} \pi a_{cr}^3 \rho_c I_{st}(r, y_1). \quad (11)$$

Here ρ_c is the carbon density; a is the radius of particle growing due to condensation; I_{st} is the stationary isothermal rate of nucleation⁹; and, f is the secondary size distribution function. In Ref. 11 the increase of the total mass of condensate is determined by formation of new stable nuclei of radius a_{cr} and condensation increase at rate $\frac{da}{dt}$ of nuclei previously formed. The radius of critical nucleus a_{cr} is calculated by the formula¹⁰

$$\frac{d \ln I_{st}}{d \ln s} = q_* + 1,$$

where q_* is the number of molecules in a nucleus of critical size, and $s = \rho_1/\rho_{sat}$ is the degree of supersaturation. This formula is valid for clusters of small size for which the Kelvin–Gibbs formula can fail. The dynamics of the secondary size distribution function is described by the kinetic equation¹¹

$$\frac{\partial f_0}{\partial t} + \dot{a} \frac{\partial f_0}{\partial a} + v \frac{\partial f_0}{\partial r} = \frac{I_{st}}{r} \delta(a - a_{cr}), \quad (12)$$

where $f_0 = f/\rho$, and $\delta(a - a_{cr})$ is the Dirac delta function.

The condition of independence of a growth rate \dot{a} on the radius in the free–molecular regime is used in Eq. (12). The boundary conditions are

$$\begin{cases} f_0(t = 0, r, a) = 0, \\ f_0(t, r = R_0, a) = 0, \\ f_0(t, r, a < a_{cr}) = 0. \end{cases} \quad (13)$$

It should be noted that Eq. (12) with the boundary conditions (13) describes homogeneous condensation of secondary particles of radius $a \geq a_{cr}$ (Ref. 11). Using the method of Laplace transform for function f_0 coupled with the principle of freezing \dot{a} , v , ρ , and I_{st} over the variable t we obtain the solution in the form

$$\begin{cases} f = \frac{I_{st}(\theta) r(r)}{\dot{a}(\theta) \rho(\theta)}, & \text{when } \int_{\theta}^x \frac{1}{v(r)} dr \leq t, \\ f = 0, & \text{when } \int_{\theta}^x \frac{1}{v(r)} dr > t. \end{cases} \quad (14)$$

Here θ is the coordinate where a stable nucleus of a_{cr} radius was formed. The value θ is the root of the equation

$$a = a_{cr} + \int_{\theta}^r \frac{\dot{a}(r)}{v(r)} dr.$$

It follows from the solution of kinetic equation (12) that the rate of nucleation plays the leading role in particle spectrum formation due to its strong dependence on thermodynamic parameters of the medium. The system of equations (1)–(4), (11), and (12) with boundary conditions (5)–(10), and (13) is closed with equations of ideal gas for constituents of vapor–gas

mixture, dynamics of radius of evaporating and growing particles, and can be solved using numerical methods.

METHOD OF SOLUTION

By introducing the substitution of variables $x^n = -R_0/r$ and $dz = \lambda_0 dT$ and differentiating Eqs. (1)–(4) we transform Eqs. (1) and (3) into the form

$$\frac{d^2 y_1}{d x^2} + \left[-\frac{1}{y_1} + \left(\frac{1-n}{x} + \frac{1}{\rho_0(x) D(x)} \frac{d(\rho_0 D)}{d x} - \frac{x^{n-1} Q n}{4\pi p D R_0} \right) \frac{1}{y_1} \right] \times \frac{d y_1}{d x} - \frac{4\pi R_0^3 n^2 d M / d t}{4\pi \rho_0 D R_0 m_0 y_1 x^{2n+2}} y_1 = -1, \quad (15)$$

$$c_3(x, y_1) = \frac{4\pi R_0 n}{m_0} \int_{-1}^x \left(\frac{d M}{d t}(x, y_1) / x^{3n+1} \right) dx, \quad (16)$$

$$\frac{d^2 z}{d x^2} + \left(-\frac{1}{z} + \frac{1-n}{x} \frac{dz}{dx} + \frac{c_{p1} n \eta_0(z) x^{n-1}}{4\pi R_0 z} \frac{dg(x, z)}{dx} + \frac{c_{p1} n x^{n-1}}{4\pi R_0 \lambda_0(z) z} \frac{dz}{dx} (g(x, z) - Q) \right) z = -1 - \frac{R_0^2 n^2 p(x)}{x^{2n+2}}, \quad (17)$$

where $\eta_0(z) = \int_0^z \frac{dz}{\lambda_0(z)}$.

The boundary conditions to the equations are

$$\begin{cases} y_1(x = -1) = y_{1s}, & (18) \\ y_1(x = 0) = 0, & (19) \\ c_3(x = -1) = 0, & (20) \\ z(x = -1) = z_s, & (21) \\ z(x = 0) = 0, & (22) \end{cases}$$

where $z_s = \int_{T_0}^{T_s} \lambda_0(T) dT$.

Equations (14) and (16) are interdependent therefore the concentration of vapor y_{1s} on the particle surface can be determined only by the iteration method. In this case the value y_{1s}^0 equal the vapor concentration on the particle surface without condensation must be taken as zero approximation. The value y_{1s}^0 is the root of transcendent equation²

$$-\ln(1 - y_{1s}^0) = (y_{sat}(T_s) - y_{1s}^0) R_0 (k_B T_s / 2\pi m_0)^{1/2} / D_{eff},$$

where $D_{eff} = 1 / \int_{R_0}^{\infty} \frac{T(r)}{T_s D(T) r^2} dr$.

The diffusion flow from the evaporating particle surface is determined from Eq. (10) with boundary conditions (18) and (19) is

$$j_d = -4\pi R_0^2 \rho_0 D(T_s) \frac{d y_1}{d r} / r = R_0.$$

Representing dy_1/dr in the form of difference derivative

$$\frac{d y_1}{d r} = \frac{y_1(R_0 + 2H) - y_1(R_0)}{2H} + O(H^2)$$

and using the boundary condition (5) for the evaporated matter flow we find the approximation for vapor concentration on the surface

$$y'_{1s} = (D \rho_0 y_1 (R_0 + 2H) / 2H + y_{sat} \alpha_s) / (\rho_0 v + \alpha_s + \rho_0 D / 2H), \quad (23)$$

where $\alpha_s = \xi \rho_0 (k_B T_s / 2 \pi m_0)$, $H = R_0 (1/x_1 - 1)$ is the step of the difference grid. When dividing an integration segment $[-1, 0]$ into 2^n portions $x_1 = -1 + 1/2^n$. The iteration approximation y'_{1s} is used as a boundary condition for Eq. (16). Using the obtained solution $y_1(x)$ based on Eq. (23) we construct the next approximation for boundary conditions (18) and (19). The iteration process is interrupted provided that $(y_{1s}^{i+1} - y_{1s}^i) / y_{1s}^i \leq 10^{-4}$. Equations (15) and (17) have a symbolic form

$$d^2 U / dx^2 + V(x, U) U = g(x)$$

and are solved by the Multhopp method.¹²

DISCUSSION ON RESULTS

As the numerical calculations showed, in the vicinity of evaporating particle there is a region of high supersaturation of vapor where the degree of supersaturation attains large values ($s \approx 2-3$) (Fig. 1).

The increase of supersaturation is caused by higher rate of carbon vapor atom inflow into the system as compared to the rate of vapor atom decrease due to condensation. In this region there occurs intense nucleation. The total area of secondary surface on which the vapor is condensed increases rapidly that results in the increase of vapor decrease rate and, as a result, decrease of degree of supersaturation. The vapor phase exhausts rapidly (Fig. 2). The base mass of vapor is condensed in a ring zone where the degree of supersaturation exceeds unity significantly (Fig. 1). Here an abrupt change of a portion of supercondensed matter equal to $f_c = \rho_3 / (\rho_3 + \rho_1)$ is observed (Fig. 3). In subsequent expansion of heterogeneous mixture the carbon vapor pressure slowly tends to the saturated vapor pressure (extension of this zone is 2-4 radius of particle). This is related to weak increase of secondary mass. After the equilibrium state is elapsed, the vapor becomes nonsaturated. The obtained specific dependences (Figs. 1, 2, and 3) are conditioned by sharp change (depending on distance) of saturated vapor pressure which decreases exponentially $\sim \exp(-L/k_B T(r))$ as well as the rate of nucleation.

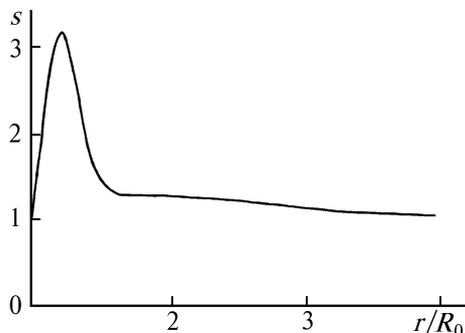


FIG. 1. The degree of supersaturation s of carbon vapor vs. the distance r/R_0 in the vicinity of particle of radius $R_0 = 100 \mu\text{m}$ under irradiance $I_0 = 2 \cdot 10^8 \text{ W/m}^2$.

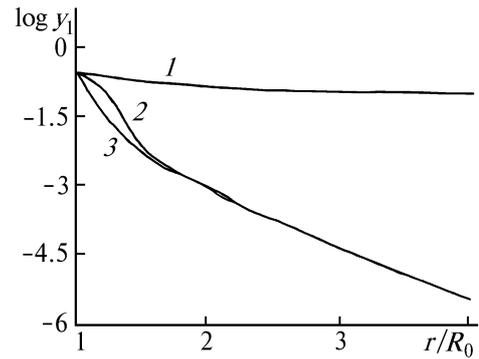


FIG. 2. The dimensionless concentration of carbon vapor in the vicinity of evaporating particle vs. the distance: 1) neglecting condensation of supersaturated carbon vapors; 2) taking account of supersaturation; and, 3) concentration of saturated carbon vapors. Here $R_0 = 100 \mu\text{m}$ and $I_0 = 2 \cdot 10^8 \text{ W/m}^2$.

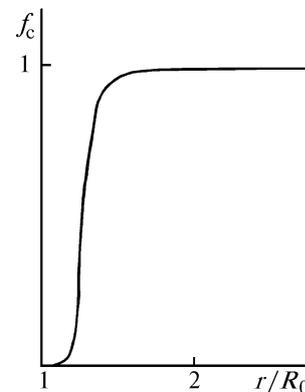


FIG. 3. A portion f_c of the supercondensed matter vs. the dimensionless distance. Here $R_0 = 100 \mu\text{m}$ and $I_0 = 2 \cdot 10^8 \text{ W/m}^2$.

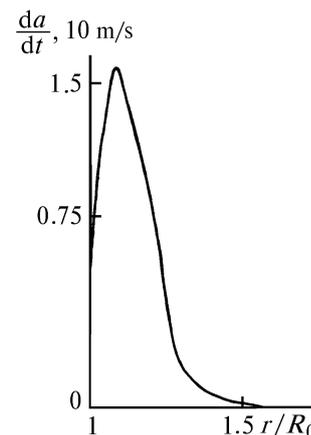


FIG. 4. The rate of change of the growing particle radius vs. the dimensionless distance. $R_0 = 100 \mu\text{m}$ and $I_0 = 2 \cdot 10^8 \text{ W/m}^2$.

The medium temperature in the vicinity of particle decreases as $\sim 1/r$. In the region of condensation the change of vapor-gas mixture temperature is $\Delta T \sim 1000 \text{ K}$ which corresponds to decrease of saturated vapor pressure by more

than an order of magnitude. As a result, the rate of secondary growth changes sharply (Fig. 4). The increase of the rate of secondary radius change is observed directly near the surface of evaporating particle at the distance $r \sim 1.08 R_0$. This fact is caused by increase of supersaturation degree. As the calculation indicated, a submicron aerosol is formed as a result of condensation that is related to short time of active growth of secondary particles:

$$t \sim \frac{dr}{v} = \frac{5 \cdot 10^{-5} \text{ m}}{10 \text{ m/s}} = 5 \cdot 10^{-6} \text{ s.}$$

During this time at the rate of increase $da/dt \sim 10^{-3} \text{ m/s}$ the particle radius attains $a \sim 5 \cdot 10^{-9} \text{ m}$. The radius of stable nucleus during this time is $\sim 10^{-9} \text{ m}$. The secondary size distribution function has a narrow spectrum since the rate of nucleation decreases with distance rapidly. Due to short time of active growth of particles the mean radius does not change practically with distance, and only a portion of modal-radius particles increases. Thus the numerical realization of the problem of particle evaporation in atmospheric air in a high-power optical field ascertained the following peculiarities:

1. Two- or three-fold vapor supersaturation is observed for large particles of $R_0 \sim 100 \mu\text{m}$ in the prebreakdown regime.

2. Active nucleation and active growth of concentrated particles occur in a narrow ring zone directly near the surface of primary particle.

3. A cloud of submicron aerosol forms in the vicinity of evaporating particle.

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