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# A modified differential approximation for thermal radiation of semitransparent nonisothermal particles: application to optical diagnostics of plasma spraying

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## Abstract

A modified differential approximation proposed recently by the author is formulated for nonisothermal particles. This approximation called  $MDP_0$  (modified  $DP_0$ ) is much simpler than the radiation transfer equation (RTE). Comparison with the RTE solution confirms an acceptable accuracy of  $MDP_0$ . An improved model of particle heating in plasma spraying is proposed. This model, which includes  $MDP_0$  equations, takes into account the radiation–conduction interaction inside the particle. Calculations for metal oxide particles in a typical plasma jet show that the particle color temperature, which is usually determined in monitoring the plasma spraying, is very sensitive to the absorption index of the particle substance. This effect should be taken into account in experimental evaluations of the bulk temperature of particles. © 2002 Elsevier Science Ltd. All rights reserved.

*Keywords:* Thermal radiation; Particles; Optical diagnostics

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## 1. Introduction

In some practical applications, one deals with intensive heating or cooling of semitransparent particles. One example is the problem of vapor explosion due to thermal interaction of molten uranium oxide droplets with ambient water. Another one is the problem of oxide particles heating in plasma spraying. In both cases, the temperature difference between the center and the surface of the particle cannot be ignored and one needs it in calculations of thermal radiation from nonisothermal particles.

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### Nomenclature

$B$	Plank's function
$C_1, C_2$	coefficients in MDP <sub>0</sub> equation
$C_D$	drag coefficient
$D$	radiation diffusion coefficient
$I_0^-, I_0^+, g_0$	functions of radius in MDP <sub>0</sub>
$L$	latent heat of melting
$P$	heat generation rate
$r$	radial coordinate, $0 \leq r \leq r_p$
$R$	Fresnel's reflection coefficient
$x$	diffraction parameter
$y$	axial coordinate (along the jet axis).

### Greek symbols

$\mu = \cos \theta$ ,	$\theta$ is measured from the radius-vector direction,
$\alpha$	absorption coefficient
$\mu_c$	cosine of Brewster's angle
$\eta$	dynamic viscosity

### Subscripts and superscripts

c	color
e	external
g	gas or plasma
m	melting
p	particle
s	particle surface
$\lambda$	spectral dependent
*	peculiar value

A rigorous statement of the problem must take into account effects of interference as is done in the Mie theory. For solving this problem, one can use the general algorithm for a stratified sphere developed in paper [1]. At the same time, in the case of large particles, for which the temperature difference is more important, one can try to use the geometrical optics approximation and the radiation transfer theory. The applicability of geometrical optics approximation has been examined in papers [2–9]. A comparison with the Mie theory calculations, presented in recent papers [7–9], showed that geometrical optics approach is usually rather accurate, even for not very large particles (size parameter  $x \geq 20$ ), both for the particle emissivity value and for the heat generation profile. At the same time, the calculations based on the geometrical optics and RTE are still too complicated. To simplify the calculations, in recent papers [7–9], the modified differential approximation has been proposed.

In this paper, the equations of MDP<sub>0</sub>-approximation for the general case of nonisothermal particle are derived and the accuracy of this approximation is evaluated by comparison with RTE solution both for isothermal and nonisothermal particles. In the second part of the paper, MDP<sub>0</sub> is employed to develop a combined heat transfer model of particle heating in plasma spraying. Calculations for metal oxide particles in a typical plasma jet show a considerable difference between usually measured color temperature of the particle and their bulk temperature, which is the most interesting value in monitoring the plasma spraying.

## 2. Differential approximation for radiation transfer

The radiation transfer equation for a spherical volume of absorbing medium with the absorption coefficient  $\alpha_\lambda(r)$ , temperature profile  $T_p(r)$  and constant index of refraction  $n$  is as follows [10,11]:

$$\mu \frac{\partial I_\lambda}{\partial r} + \frac{1 - \mu^2}{r} \frac{\partial I_\lambda}{\partial \mu} + \alpha_\lambda I_\lambda = 2\pi n^2 \alpha_\lambda B_\lambda(T_p). \tag{1}$$

Here,  $I_\lambda(r, \mu)$  is the radiation intensity at point  $r$  in the direction  $-1 \leq \mu = \cos \theta \leq 1$  integrated over the azimuth. The boundary conditions (symmetry at  $r = 0$  and Snell’s law at  $r = r_p$ ) are

$$I_\lambda(0, -\mu) = I_\lambda(0, \mu), \quad I_\lambda(r_p, -\mu) = R(\mu)I_\lambda(r_p, \mu), \tag{2}$$

where  $0 \leq \mu \leq 1$ ,  $R(n, \mu)$  is Fresnel’s reflection coefficient for unpolarized radiation [12]

$$R = (R_{\parallel} + R_{\perp})/2, \quad R_{\parallel} = \left\{ \frac{\mu - n\sqrt{1 - n^2(1 - \mu^2)}}{\mu + n\sqrt{1 - n^2(1 - \mu^2)}} \right\}^2, \tag{3}$$

$$R_{\perp} = \left\{ \frac{n\mu - \sqrt{1 - n^2(1 - \mu^2)}}{n\mu + \sqrt{1 - n^2(1 - \mu^2)}} \right\}^2, \quad \mu > \mu_c,$$

$$R_{\parallel} = R_{\perp} = 1, \quad \mu \leq \mu_c = \sqrt{1 - 1/n^2}.$$

We assume here that the index of absorption is small in comparison with the medium index of refraction:  $\kappa \ll n$ .

The angle-averaged heat generation rate for a spherical layer of unit thickness is equivalent to the divergence of the integral radiation flux:

$$P(r) = \frac{1}{r^2} \frac{d}{dr} \left( r^2 \int_0^\infty q_\lambda d\lambda \right) = \int_0^\infty \alpha_\lambda \left[ 4\pi n^2 B_\lambda(T) - \int_{-1}^1 I_\lambda(r, \mu) d\mu \right] d\lambda. \tag{4}$$

The spectral radiation intensity on the outer surface of the volume and the corresponding spectral radiation flux are defined as follows:

$$I_\lambda^e(a, \mu_c) = [1 - R(\mu)]I_\lambda(a, \mu), \quad \mu = \sqrt{1 - (1 - \mu_c^2)/n^2}, \quad 0 \leq \mu_c \leq 1, \tag{5}$$

$$q_\lambda^e(a) = \int_0^1 I_\lambda^e(a, \mu_c) \mu_c d\mu_c = \int_{\mu_c}^1 [1 - R(\mu)]I_\lambda(a, \mu) \mu d\mu.$$

In the two-flux (or DP<sub>0</sub>) approximation, the radiation intensity is presented in the form

$$I_\lambda(r, \mu) = \begin{cases} I_0^-(r), & -1 \leq \mu < 0, \\ I_0^+(r), & 0 < \mu \leq 1. \end{cases} \tag{6}$$

An analysis of angular dependencies of the radiation intensity showed that expression (6) is appropriate only in the range  $r \leq r_* = r_p/n$  and the following approximation of the function  $I_\lambda(r, \mu)$  is reasonable for the spherical layer  $r_* < r \leq r_p$  [7–9]:

$$I_\lambda(r, \mu) = \begin{cases} I_0^-(r), & -1 \leq \mu < -\mu_*, \\ 2\pi n^2 B_\lambda(T_p), & -\mu_* < \mu < \mu_*, \quad \mu_*(r) = \sqrt{1 - \left(\frac{r_*}{r}\right)^2}, \\ I_0^+(r), & \mu_* < \mu \leq 1. \end{cases} \tag{7}$$

The change of the angular dependence from (6) to (7) at  $r=r_*$  is explained by the internal reflection of the radiation emitted by elementary volumes placed at  $r > r_*$ . Note that Eqs. (7) transform to (6) if we put  $\mu_* = 0$  when  $r \leq r_*$ . Integrating radiation transfer Eq. (1) over  $\mu$  separately on intervals  $-1 \leq \mu < \mu_*$  and  $\mu_* < \mu \leq 1$ , and introducing the function  $g_0 = I_0^- + I_0^+$ , we obtain the following boundary-value problem in the modified DP<sub>0</sub>-approximation (MDP<sub>0</sub>):

$$\begin{aligned} &-\frac{1}{r^2} \frac{d}{dr} \left( r^2 D_\lambda \frac{dg_0}{dr} \right) - (C_1 - 1) \frac{2D_\lambda}{r} \frac{dg_0}{dr} + C_2 \alpha_\lambda [g_0 - 4\pi n^2 B_\lambda(T)] = 0, \\ &C_1 = \begin{cases} 1, & r \leq r_* \\ (1 - \mu_*)/(2\mu_*), & r > r_* \end{cases} \quad C_2 = \begin{cases} 1, & r \leq r_* \\ (1 + \mu_*)^{-2}, & r > r_* \end{cases} \quad D_\lambda = 1/(4\alpha_\lambda), \\ &\left. \frac{dg_0}{dr} \right|_{r=0} = 0, \quad \frac{n}{n^2 + 1} g_0(r_p) + D_\lambda (1 + \mu_c) \left. \frac{dg_0}{dr} \right|_{r_p} = 0. \end{aligned} \tag{8}$$

For simplicity, the normal reflectivity  $R(1)$  is used in the boundary condition. The spectral radiation flux on the particle surface and the heat generation rate inside the particle are calculated as follows:

$$q_\lambda^e = \frac{g_0(r_p)}{n(n^2 + 1)}, \quad P = \int_0^\infty P_\lambda d\lambda, \quad P_\lambda = \begin{cases} w, & r \leq r_* \\ (1 - \mu_*)w, & r > r_* \end{cases} \quad w = \alpha_\lambda [4\pi n^2 B_\lambda(T) - g_0]. \tag{9}$$

The mathematical problem statement in MDP<sub>0</sub>-approximation is much simpler than RTE. It is sufficient to note that the computational time on the same finite-difference mesh decreases in two orders of magnitude when the MDP<sub>0</sub> is used. At the same time, preliminary evaluations of the accuracy of MDP<sub>0</sub> in the case of isothermal particles [7–9] have shown that the error of this approximation is not too large. Some additional results for isothermal particle with  $n = 2$  are presented in Fig. 1. The dimensionless heat generation rate is defined as

$$\bar{P}(\bar{r}) = P(\bar{r}) / \left[ 3 \int_0^1 P(\bar{r}) \bar{r}^2 d\bar{r} \right], \quad \bar{r} = r/r_p. \tag{10}$$

The optical thickness is  $\tau_0 = \alpha_\lambda r_p$ . One can see that MDP<sub>0</sub> gives qualitatively correct radial profiles of the heat generation rate and the error in particle emissivity in the range of moderate optical thickness

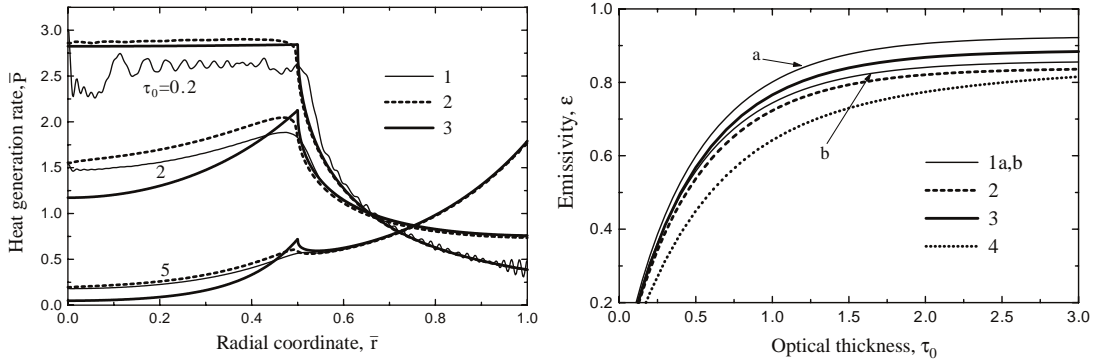


Fig. 1. Heat generation profiles in isothermal particle and the particle emissivity: (1) Mie theory ((1)  $x = 100$ , 1(a) 50, 1(b), 300), (2) numerical solution of RTE, (3) MDP<sub>0</sub>, (4) DP<sub>0</sub>.

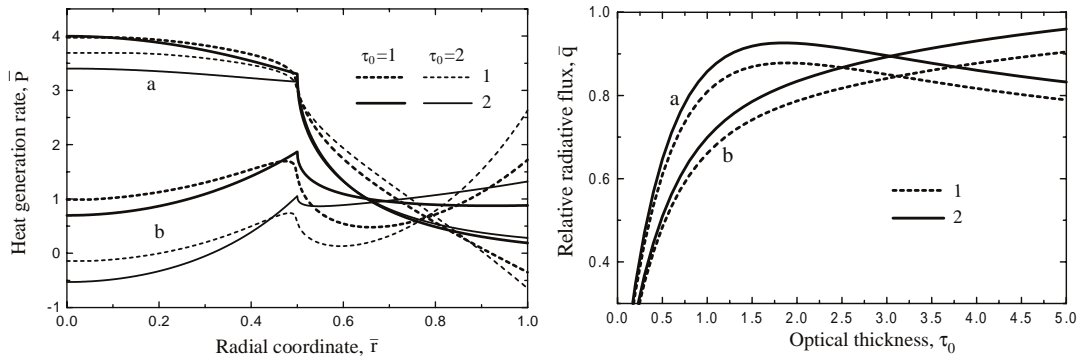


Fig. 2. Heat generation profiles and relative radiation flux for nonisothermal particles: (1) numerical solution of RTE, (2) MDP<sub>0</sub>-approximation, (a)  $\Delta T_p = -500$  K, (b)  $\Delta T_p = 500$  K.

is considerably less than that of DP<sub>0</sub>-approximation. To evaluate the error of MDP<sub>0</sub> consider also a nonisothermal particle with the temperature profile

$$T_p = T_0 + \Delta T_p \bar{r}^2, \quad \Delta T_p = T_s - T_0. \tag{11}$$

The bulk temperature of the particle

$$\bar{T}_p = \int_0^1 T_p(\bar{r}) \bar{r}^2 d\bar{r} = T_0 + 0.6 \Delta T_p. \tag{12}$$

Assuming  $\alpha_\lambda = \text{const}$ , we use the value of optical thickness  $\tau_0$  as a parameter of the problem. Some numerical results for particles with  $n = 2$ ,  $\bar{T}_p = 3000$  K,  $\Delta T_p = \pm 500$  K are presented in Fig. 2, where the relative radiation flux is defined as  $\bar{q} = q/\sigma T_p^4$ . It is obvious that the accuracy of MDP<sub>0</sub> remains rather high in the case of nonisothermal particles in a wide range of the optical thickness. Having in mind, the known results on the accuracy of diffusion-like approximations [13,14], one should not expect a considerable increase in the error of MDP<sub>0</sub> for other temperature profiles. In the next section of the paper, the application of MDP<sub>0</sub>-approximation to the problem of optical diagnostics of the bulk temperature of particles in plasma spraying is considered.

### 3. Particle motion and heating in plasma jet

Numerical simulation of behavior of particles in a plasma jet is necessary for understanding the process of plasma spraying used for coating deposition and for advance material forming. It is especially important because of developing experimental techniques and intensive experimental investigations of plasma spraying [15–17]. Intensive heating in the jet leads to considerable temperature difference inside the particle. This effect has been first analyzed in earlier papers [18,19]. At the same time, the present-day theoretical models of particle heating do not take into account the particle semi-transparency. As a result, the evaluations of the thermal radiation effect on the particle temperature are not quite correct for metal oxides (Al<sub>2</sub>O<sub>3</sub>, Cr<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, ZrO<sub>2</sub>), which are widely used in plasma spraying. Below the more general model is proposed and some numerical results for a typical plasma jet are given.

#### 3.1. Theoretical model

Particle concentration in a plasma spray jet is usually small. Therefore, particles do not interact with each other and their influence on plasma flow parameters is not large [15,17]. From the other hand, the optical thickness of the jet is small and thermal radiation effects may be easily taken into account in the heat balance for a single particle. The resulting equations for a particle moving along the jet axis are as follows:

$$u_p \frac{du_p}{dy} = \frac{3}{8r_p} C_D \frac{\rho_g}{\rho_p} (u_g - u_p) |u_g - u_p|, \quad u_p(0) = u_{p0}, \tag{13}$$

$$\rho_p c_p u_p \frac{\partial T_p}{\partial y} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 k_p \frac{\partial T_p}{\partial r} \right) - P, \quad T_p(0, r) = T_{p0}(r),$$

$$\left. \frac{\partial T_p}{\partial r} \right|_{r=0} = 0, \quad \left. k_p \frac{\partial T_p}{\partial r} \right|_{r=r_p} = k_g \frac{Nu}{2r_p} [T_g - T_p(y, r_0)]. \tag{14}$$

The latent heat of melting may be treated as an equivalent increasing the specific heat capacity in the narrow temperature interval near the melting temperature. Possible evaporation of particle is not considered in the paper. The value of heat generation rate  $P$  on the right hand side of (14) is determined by the use of MDP<sub>0</sub>-approximation.

The drag coefficient and the Nusselt number are calculated by using the following relations [20]:

$$C_D = 24/(Re_p + 4.33\varphi) + \frac{4.5 + 0.38\psi}{1 + \psi} \exp\left(-\frac{0.5M_p}{\sqrt{Re_p}}\right) + 0.6\varphi \left[1 - \exp\left(-\frac{M_p}{Re_p}\right)\right], \tag{15}$$

$$\varphi = M_p \sqrt{\gamma/2}, \quad \psi = 0.03Re_p + 0.48\sqrt{Re_p}, \quad Re_p = 2\rho_g |u_g - u_p| r_p / \eta_g,$$

$$Nu = \frac{\zeta}{1 + 3.42\zeta M_p / (Re_p Pr)}, \quad \zeta = 2 + 0.459Re_p^{0.55} Pr^{0.33}. \tag{16}$$

For plasma jet conditions (especially for evaporating particles), Eqs. (15) and (16) need further improvement [15,21].

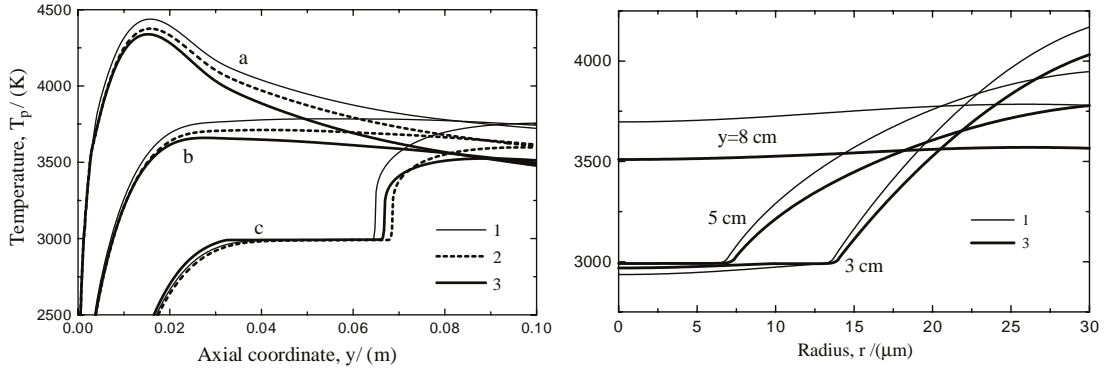


Fig. 3. Effect of thermal radiation on particle temperature: (1) without radiation, (2,3) with radiation ((2)  $\alpha_i = 10^4 \text{ m}^{-1}$ , (3)  $10^5 \text{ m}^{-1}$ ); (a) surface, (b) average temperature  $\bar{T}_p$ , (c) center.

### 3.2. Numerical results

Consider a typical argon plasma jet and take the following approximations of the plasma velocity and temperature variation along the jet axis [15]:

$$u_g = u_1 / [1 + (u_1/u_2 - 1)\bar{y}^2], \quad T_g = T_1 / [1 + (T_1/T_2 - 1)\bar{y}^2], \quad \bar{y} = y/y_0, \quad (17)$$

where  $y_0 = 0.1 \text{ m}$ ,  $u_1 = 600 \text{ m/s}$ ,  $u_2 = 150 \text{ m/s}$ ,  $T_1 = 12000 \text{ K}$ ,  $T_2 = 2000 \text{ K}$ . Thermal properties of plasma from [17] are approximated by functions

$$c_g = \begin{cases} 500, & \tilde{T} \leq 8 \\ 500 + 280(\tilde{T} - 8)^2, & \tilde{T} > 8. \end{cases} \quad k_g = 0.1 + 0.013(\tilde{T} - 2)^2, \quad \eta_g \cdot 10^5 = 10 + 2.5(\tilde{T} - 2), \quad \tilde{T} = T_g/1000. \quad (18)$$

Here, all physical values are expressed in SI units.

Calculations are performed for zirconium oxide particles of radius  $r_p = 30 \mu\text{m}$  with the following physical parameters:  $\rho_p = 5150 \text{ kg/m}^3$ ,  $c_p = 600 \text{ J/(kg K)}$ ,  $k_p = 1.5 \text{ W/(m K)}$ ,  $L = 730 \text{ kJ/kg}$ . The value of  $n = 2$  is used [22] and two variants of the constant absorption coefficient are considered:  $\alpha_i = 10^4$  and  $10^5 \text{ m}^{-1}$  [23]. Numerical data on the particle temperature variation along the jet are presented in Fig. 3. One can see the great temperature difference in the particle during the melting. The radiation decreases the particle temperature and the temperature difference inside the particle but this effect is usually less than 300 K. The time of melting may be increased or decreased due to radiation. The sign of this effect depends on the optical properties of the particle substance.

## 4. Color temperature of semitransparent particles

The particle temperature is usually determined experimentally from the ratio of the thermal radiation detected at two closely related wavelength (two-color pyrometry) [16]. As a result, the so-called color temperature is obtained. In the case of a semitransparent particle, the color temperature

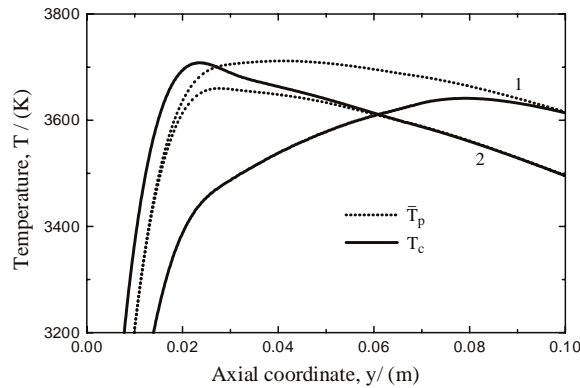


Fig. 4. Comparison of the color temperature  $T_c$  with the bulk temperature  $\bar{T}_p$  of oxide particle in the plasma spraying jet: (1)  $\alpha_\lambda = 10^4 \text{ m}^{-1}$ , (2)  $\alpha_\lambda = 10^5 \text{ m}^{-1}$ .

$T_c$  is intermediate between the surface temperature  $T_s$  and the center temperature  $T_0$  of the particle. Having in mind that one usually interested in the bulk (volume averaged) temperature  $\bar{T}_p$ , we consider the relationship between this value and the color temperature. Numerical results presented in Fig. 4 show that the color temperature may be less or greater than the bulk one. In the case of small optical thickness of the particle ( $\alpha_\lambda = 10^4 \text{ m}^{-1}$ ,  $\tau_0 = 0.3$ ) we have  $T_c > \bar{T}_p$ ; in the opposite case of comparably large optical thickness ( $\alpha_\lambda = 10^5 \text{ m}^{-1}$ ,  $\tau_0 = 3$ ) we have  $T_c < \bar{T}_p$ . The difference  $|T_c - \bar{T}_p|$  in the region of intensive heating of the particle is about 200–300 K and then decreases with decreasing the temperature difference inside the particle.

A comparison of the calculated surface temperature for  $\text{Al}_2\text{O}_3$  particles ( $r_p = 23.5 \text{ }\mu\text{m}$ ) with experimental data was given by Fiszdon [19]. It has been found that experimental (most likely, color) temperature was less than the surface one. The data of [19] show that this discrepancy decreases with the distance from the jet origin from 400 K at  $y = 8 \text{ cm}$  to 200 K at  $y = 18 \text{ cm}$ . Unfortunately, there is no complete set of parameters of that plasma jet to repeat the calculations of [19]. Nevertheless, in all likelihood, the observed difference between experimental temperature and calculated surface temperature may be explained by semi-transparency of aluminum oxide particles.

It is important to take into account the temperature dependence of oxide absorption coefficient. A considerable increasing of  $\alpha_\lambda$  with temperature (as usually observed for  $\text{Al}_2\text{O}_3$  [13]) may lead to more high color temperature of particle during the heating.

## 5. Conclusion

- The problem of thermal radiation from large nonisothermal spherical particles of semitransparent material is considered. The new differential approximation ( $\text{MDP}_0$ ) proposed recently by the author is compared against the exact numerical solution of the radiation transfer equation. The acceptable accuracy of  $\text{MDP}_0$  in a wide range of the problem parameters is shown.
- An improved model of particle heating in plasma spraying is proposed. This model, which includes  $\text{MDP}_0$  equations, takes into account the radiation–conduction interaction inside the particle. Effect of radiation is illustrated by numerical example for a typical plasma jet.



- Plasma spraying calculations show that color temperature of metal oxide particles may be less or greater than their bulk temperature depending on the spectral absorption coefficient of particle substance. The typical difference of about 200–300 K between color and bulk temperatures of oxide particles should be taken into account in optical diagnostics of plasma spraying process.

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