Pergamon

Int. J. Heat Mass Transfer. Vol. 41, No. 2, pp. 489–499, 1998

Printed in Great Britain
0017–9310/98 $19.00+0.00

PII: S0017–9310(97)00108–7

Heating of solid aerosol particles exposed to intense optical radiation

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(*Received* 8 *June* 1994)

Abstract—The absorbed energy release within solid spherical particles, due to laser radiation, can be substantially inhomogeneous. Under the influence of intense laser radiation heat transfer by conduction has no time for the equalization of temperature inside a particle, as a result its heating is also strongly inhomogeneous. Using the Mie theory, a theoretical investigation into the distribution of heat source function within the solid, absorbed particles is carried out. Based on a numerical solution of the heat conduction equation, taking into account the thermal dependencies of optical and thermophysical properties of particle material, the temperature distribution inside the particles is investigated in detail. © 1997 Elsevier Science Ltd.

1. INTRODUCTION

Considerable success in the development of laser irradiance sources and their increasing application for investigation of hydro- and atmospheres, optical location and communication, different physico-chemical and biological investigations and for various technical objectives, generated a need for an all-round study of the physics of interaction of laser radiation with various natural and artificial media. Various aspects of the problem of interaction of intense laser radiation with dispersed media evokes great interest. Among these, for example, are the propagation of intense radiation in a medium with particles, radiation-produced clearing of clouds, fogs, smoke and dust formations at the expense of radiation heating, evaporation and fragmentation of particles, forming them, optimization of technological processes based on the application of laser technologies (deposition of thin films, hardening and others).

Separate attempts to study the interaction of laser radiation with solid particles were made quite some time ago. As an example, one can mention ref. [1], where experimental results on the interaction of SiO_2, clay, NaCl, coal, slag, soot, charcoal and cement dust particles with CO_2-laser radiation ($I = 130$–210 W cm^{-2}) were discussed. Although the intensities of radiation in these experiments were not so high, it was clearly shown that combustion, melting, evaporation and jet motion of particles were possible under the action of radiation.

A large amount of investigations is available that are devoted to the interaction of soot and coal particles in air with laser radiation [2–4]. The range of the problems solved in this work is peculiar for being connected with chemical reactions which occur during the burning of particles. A discussion of these problems lies outside the scope of this paper. Investigations devoted to the optical breakdown of a medium under the action of laser radiation, as well as investigations of photophoretic and light jet movement of particles, do not enter into the scope of the present paper.

The objective of this investigation was to study, in detail, the absorption of the laser radiation, the distribution of the absorbed energy inside the particles, and the transformation of this energy into thermal energy with subsequent heating of the material to the phase transition temperature in the case of non-stationary regimes.

Based on the assumption of uniform and quasi-stationary heating, the authors of monograph [5] carried out a general theoretical consideration of evaporation of solid particles in diffusional and underexplosive gas kinetic regimes. They discussed the conditions under which not only the boiling temperature of the particle material, but also that of the start of violent spontaneous nucleation in the volume of a particle can be attained, i.e. the occurrence of the particle in the region of the metastable state of substance with its subsequent destruction. Some problems of the interaction of solid aerosol particles with intense laser radiation are also investigated in ref. [6].

However, the thermal mechanism of the destruction of particles can arise without the transition of the particle substance to a metastable state, but rather as a result of the cracking of particles on attainment of a certain magnitude of the temperature gradient at which the originating forces of mechanical stresses start to exceed the cohesion forces. In another case, the 'so-called' hot spots may arise within the particles under conditions of nonuniform release of heat inside its volume. At these hot spots the local melting of the

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NOMENCLATURE

C	heat capacity
E_r, E_θ, E_φ	electric field components at a given point inside a particle
E_0	electric field of incident laser beam
I	incident radiation intensity
m	complex refractive index of particle material
m_s	refractive index of surrounding medium
n	real part of complex refractive index
Q	power density of heat sources at a given point inside a particle
R	radius of homogeneous particle
R_1	radius of core
R_2	radius of the whole two-layered particle
r	radius coordinate
T	temperature
T_m	melting temperature of particle
t	time.

Greek symbols

α	coefficient of heat exchange between the particle and surrounding medium
ε	emissivity of a particle at its surface temperature
θ	angular coordinate
χ	imaginary part of the complex refractive index
λ_1	thermal conductivity
λ	radiation wavelength
σ	Stefan–Boltzmann constant
φ	angular coordinate
ρ	density.

Subscripts

m	melting
max	maximum
min	minimum
s	surrounding medium.

material begins with the formation of gas bubbles that execute convective motion. As several of these bubbles coalesce and reach the surface of the particle, a local explosion occurs with the ejection of the material or particle fragmentation. In either case, the degree of the nonuniformity of temperature distribution within the particle plays a decisive role.

Up to the present, the problem of inhomogeneous release of the laser radiation energy absorbed within the particle and especially the problem of inhomogeneous heating of particles by intense laser radiation were left untouched in the majority of such kinds of investigations. The estimations of heating, melting, evaporation (sublimation) of particles are made proceeding from the assumption of uniform evolution of heat in the particle volume and of uniform temperature distribution throughout the particle volume including the surface of the particle.

Just as any physical hypothesis, the above-mentioned assumptions about the character of temperature distribution within the particles exposed to intense laser radiation have their limits of applicability that in some cases are rather stringent.

In this paper, the results of a number of investigations are reported for inhomogeneous heating of particles. We elucidate the conditions in which the assumptions of uniform heating do not correspond to the processes occurring in particles. Therefore, we investigate, in detail, both the specific features of heat evolution and the regularities of the heating of particles under the action of intense laser radiation.

To show the necessity of such investigations, the results of some experiments are discussed below. These experiments cannot be explained with the help of the assumption of uniform heat evolution inside particles exposed to intense laser radiation.

Experimental investigation of the dynamics of laser heating of tungsten particles with $R = 35$–100 μm [7] revealed in particular that with radiation intensities $I = 5 \times 10^6$ W cm^{-2} the particle has no time to get to the boiling temperature, but breaks up into small pieces.

The interaction of intense ultraviolet laser radiation ($\lambda = 0.248$ μm) with small spherical particles of aluminum, calcium fluoride, glass, silicon carbide, tungsten, aluminum oxide and polystyrene/divinylbenzene (DVB) are considered experimentally in ref. [8]. The materials investigated generally ranged in size from 2.5 to 25 μm in radius with the exception of polystyrene/DVB spheres that had $R = 115$ μm. The pressure varied between 0.1 and 10^{-5} torr. The intensities of laser radiation were from 10^7 to 2×10^{11} W cm^{-2}.

The authors isolated a group of more absorbing materials (aluminum, silicon carbide, tungsten and polystyrene/DVB) and more transparent (calcium fluoride and glass) for a given wavelength. Interactions involving aluminum and calcium fluoride are given here as representatives of the two types. It is shown, experimentally, that under the action of high radiation intensities material is ejected from small aluminum particles in the direction of the laser pulse propagation. In contrast to aluminum interactions, transparent calcium fluoride particles ejected material in a much more symmetric fashion from the region of interaction. In the case of polystyrene/DVB the jetting of material occurs from the front surface of the particle opposite to the beam. There is no jetting in the shadow region of the particle. One possible explan-

nation of these dynamics is the size parameter of polystyrene/DVB particles that is one order of magnitude larger than that of any other material studied.

The results of experimental investigations of interactions of intense laser radiation ($\lambda = 10.6\ \mu m$, $I < 10^6$ W cm^{-2}) with separate spherical solid aerosol particles (NaCl, SiO_2, clay) are reported in ref. [9]. It is noted that in the range of particle radii $R = 5$–$25\ \mu m$ the heat evolution inside the particles is essentially nonuniform and in the region of the main maximum it is considerably larger than in the other regions of the particle volume. This causes the appearance of a gas phase in the 'hot spots'. For pulse CO_2-lasers, due to the small time of interaction of radiation with the material, the depth of temperature equalization throughout the SiO_2 particle volume does not exceed 10 μm for the time of a pulse. As a result, not the whole volume of the particle with $R > 20\ \mu m$ melts, and the ejection of the gas phase is accompanied by a partial jetting of the particle material.

The explosion and burning of large spherical aluminum particles with radii $R = 100$–$350\ \mu m$, under the action of a CO_2-laser operating in a continuous generation regime, are investigated experimentally in ref. [10]. It is found that at the intensities of the order of 400 W cm^{-2}, all of the particles start to burn. At the intensities from 10^3 to 4×10^3 W cm^{-2} larger particles are ignited, whereas smaller ones undergo intense fragmentation. At the intensities of $(6$–$8) \times 10^3$ W cm^{-2}, the particles explode at the time of ignition. The authors consider that the fragmentation of the particles is caused by their being heating up to the boiling aluminum temperature and, as a result, by the manifestation of the effects of explosive evaporation.

The large numbers of studies do not naturally exhaust all the works in which the interaction of intense laser radiation with solid material particles is studied experimentally or theoretically. Of the great number of such works we selected experiments in which the jetting of particle material is fixed, it is often nonsymmetric, occurs in the direction of an incident laser beam, or directed opposite to the beam. However, in all of the known cases the estimations and calculations of the temperature of particles or of their surface, as well as the rates of evaporation of particles are based on the assumption of the uniformity of inside heat evolution and central symmetric heating of particles.

2. STATE-OF-THE-ART

Based on the experience gained in the investigations of thermal explosion of water droplets [11], one can say straightaway that the boundaries of the validity of such an approximate consideration depends on the optical properties of the material at a given wavelength λ, its thermal diffusivity, particle radii and incident laser radiation intensity. It is possible to assume that in the case of slow heating, when the time of heating is much larger than the characteristic time $t_0 \sim R^2/(\lambda_1/c\rho)$, the nonuniformity of internal heat evolution can be neglected. However, when the intensity of radiation is sufficiently high and the melting temperature can be achieved for the times of order or smaller than t_0, in such cases temperature distribution inside a particle can be very nonuniform. It follows, from the above, that to receive the correct data on the heating of particles under the action of high intense laser radiation, it is necessary to have sufficient details and extensive information about the distribution of heat sources throughout the particle volumes, and also the correlation between the time of temperature relaxation and the time of the particle heating up to the temperature of phase transition or the appearance of the critical temperature gradient inside them.

This fact was taken into consideration in refs [12, 13], where investigations of the cracking of spherical ice particles under the action of the 10.6 μm radiation were made. It was assumed that the intensity of radiation was sufficiently high so that the temperature gradients that caused the appearance of destructive mechanical particle strains developed earlier than the melting temperature is reached at any point within the particle.

Calculations showed that heat evolution inside ice particles with the radius $R < 5\ \mu m$ was practically uniform, and that for larger particles the main part of the absorbed energy is transformed into heat in the illuminated region at the end of the main diameter of the particle. The main diameter is the particle diameter, which coincides with the propagation of incident radiation.

To obtain the temperature distribution inside a particle, we solve the following heat conduction equation in spherical coordinates with the origin at the particle centre subject to corresponding initial and boundary conditions:

$$C(T)\rho(T)\frac{\partial T(r,\theta,t)}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(\lambda_1(T)r^2\frac{\partial T}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\lambda_1(T)\sin\theta\frac{\partial T}{\partial\theta}\right) + Q(r,\theta,t,T) \quad (1)$$

where $0 < r < R$, $0 < \theta < \pi$. The temperature within the particle depends only on two coordinates r and θ, because the radiation is not polarized and the third coordinate φ is absent.

$$Q = \mathrm{IB}(4\pi n\chi/m_s\lambda) \quad (2)$$

$$B = (E_r^* E_r + E_\theta^* E_\theta + E_\varphi^* E_\varphi)E_0^{-2}. \quad (3)$$

The values of the electric field components inside the particle E_r, E_θ, E_φ are calculated from formulas (1.2.1)–(1.2.3) of ref. [11].

On the particle surface and at the center of the particle, the following conditions should be satisfied:

$$-\lambda_1(T)\left.\frac{\partial T}{\partial r}\right|_{r=R} = \alpha[T(R,\theta,t) - T_s] \quad (4)$$

$$|T(0,\theta,t)| < \infty \quad 0 \leqslant \theta \leqslant \pi \quad t > 0 \tag{5}$$

$$\left.\frac{\partial T}{\partial \theta}\right|_{\theta=0} = \left.\frac{\partial T}{\partial \theta}\right|_{\theta=\pi} = 0. \tag{6}$$

From additional premises specified in ref. [12], it follows that in the range of 210–273 K the limiting value of the local temperature gradient leading to the cracking of an ice sphere may be taken as $\Delta T = 10$ K.

The solution of the problem is possible only by numerical methods using a spatial–temporal grid and an iterative scheme. The specific method of the solution is described in refs [11–13]. The computations performed showed that maximum temperature drop is observed between the points in the illuminated and shadow hemispheres of the ice particle. For each size of particle there is some threshold value of the intensity and some threshold value of the absorbed energy at which a particle is destroyed not reaching melting.

It is noted that near the threshold intensities the amount of the absorbed energy before cracking is proportional to the volume of the particle, while at higher intensities the energy expenditures for the particle destruction are proportional to the cross-section of the particle.

The heating of aluminum oxide and carbon particles up to the melting temperature in vacuum under the action of intense laser radiation of different wavelengths was considered in refs [14, 15]. Instead of the boundary conditions (4) the following equation was used

$$-\lambda_1(T)\left.\frac{\partial T}{\partial r}\right|_{r=R} = \sigma\varepsilon T^4. \tag{7}$$

Calculations showed that just as in the case of water droplets, even with inhomogeneous heat evolution the heating of particles up to the melting temperature can be practically uniform if the intensity of irradiation is not very high and the time of reaching the melting temperature exceeds the characteristic relaxation time. At high intensities the time of attaining the melting temperature is small compared to the characteristic relaxation time and the heating is nonuniform. In carbon particles the melting begins on the illuminated surface, whereas in weakly absorbing aluminum oxide particles for radiation with $\lambda = 0.69$ μm the region of maximum temperatures appears in the shadow hemisphere of the particle.

The consideration of the heating of even small, but highly absorbing carbon particles at irradiation with $\lambda = 10.6$ μm [16] showed once again that in the case of highly absorbing particles it is very important to known the peculiarities of the distribution of absorbed energy within them for obtaining a correct solution of this problem. This is especially important for the study of interaction of intense laser radiation with metallic particles having, in a number of cases, high values of the real and imaginary parts of the complex refractive index ($m = n - i\chi$).

3. NUMERICAL RESULTS AND DISCUSSION

A characteristic feature of metallic particles exposed to the radiation at $\lambda = 10.6$ μm is the fact that due to high values of optical properties heat evolves in a very thin surface layer within which the absorbed energy decreases exponentially [17]. The main maximum of heat evolution is located on the illuminated surface of the particle at the point nearest to the source of radiation and the second maximum on the shadow surface. The appearance of the second surface heat evolution maximum on the shadow surface of highly absorbing particles has not yet found a simple explanation. Evidently, here occurs something that reminds the 'slide' of some portion of the energy that penetrated inside the skin-layer (due to the so-called surface waves).

It should be noted that with a decrease in the size of metallic particles the energy distribution in them becomes more uniform, especially when their size approaches the depth of radiation penetration $l_p = \lambda/(4\pi n\chi)$. However, in this case too the electric field strength on the surface of the particle and at its center differs 10-fold [18].

Up to recent times, the commonest opinion in the literature was that the highest maximum of heat evolution is always observed on the main diameter of the particle (the diameter parallel to the propagation of incident laser radiation). We determined that this is correct only when the real part of the refractive index of the particle material n exceeds 1 ($n > 1$).

Our investigations show that in particles with $n < 1$, for example, in aluminum oxide particles the distribution of heat sources for $\lambda = 10.6$ μm has a clearly expressed specific [19], namely, in a definite range of the size of particles the highest values of heat evolution are located not on the main diameter of the particle, but on the surface of a certain circular zone, with its position, described by the angular coordinate θ, being dependent on the size and the optical constants of the particle material (Fig. 1, Tables 1 and 2). As the radius of the particle increases, the maximum values of the energy density increase, while the location of the ring maximum first shifts to the side of larger angles and then to the opposite direction. At these maxima the energy density is substantially higher than on the main diameter of the particle. For larger particles a marked second maximum appears, which is located on the main diameter in the shadow hemisphere. Its height

Table 1. Maximum values of the quantity B and their angular coordinates for aluminum oxide particles of different radii

R (μm)	B_{max}	θ_{max} (deg)	R (μm)	B_{max}	θ_{max} (deg)
4	1.88	40	25	2.80	51
6	2.01	52	30	2.94	50
8	2.08	56	35	3.05	49
10	2.16	53	40	3.15	49
16	2.46	53	45	3.24	48
20	2.64	52	—	—	—

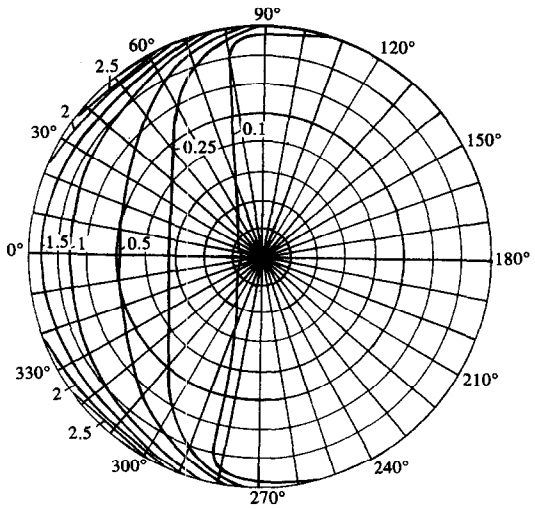

Fig. 1. Distribution of relative energy density over the section plane of a large circle of an aluminum oxide particle with $R = 25$ μm under the action of radiation with $\lambda = 10.6$ μm. The numbers at the curves are the values of B.

competes with the height of the maxima in the circular region depending on the relationship of the parameters n, χ, R and λ. The shift of the energy density maximum within weakly absorbing particles with $n < 1$ to the surface of the ring layer located at the angle θ can probably be explained by the existence of the so-called 'critical' range of scattering noted in refs [20, 21], in which the scattering of light by air bubbles in water was considered (the relative refractive index for visible light is $n = 3/4$).

To make it clear whether such peculiarity in the evolution of heat is preserved in highly absorbing particles with the real part of the refractive index $n < 1$, the present authors investigated [22] the distribution of the absorbed energy density of radiation with $\lambda = 0.53$ μm inside and on the surface of small ($0.03 < R < 2$ μm) silver and gold particles ($m_{Ag} = 0.053 - i3.1$ and $m_{Au} = 0.52 - i2.2$) [23].

Calculations showed that the distribution of absorbed energy inside the particles of intermediate regions ($0.08 < R < 0.0905$ μm for silver and $0.06 < R < 0.08$ μm for gold) is of greatest interest. As follows from ref. [22], as the particle size increases, the maximum of absorbed energy first shifts smoothly

Table 2. Maximum values of the quantity B and their angular coordinates for $\lambda = 10.6$ μm, $R = 10$ μm and variations of optical constants of particle material

	$n = 0.52$			$\chi = 0.063$	
χ	B_{max}	θ_{max} (deg)	n	B_{max}	θ_{max} (deg)
0.00	2.36	59	0.30	2.90	42
0.01	2.33	57	0.70	1.65	60
0.05	2.20	54	0.80	1.35	62
0.10	2.05	50	0.85	1.23	51
0.50	1.58	11	0.90	1.14	45
1.00	1.28	0	0.95	1.06	43

from the illuminated end of the main diameter to the surface of the circular zone with the angular coordinate $\theta \neq 0$ and then again returns to the main diameter of the particle, but now already in the shadow hemisphere. It seems that in large absorbing particles the effect of the 'critical' range of scattering is simply suppressed by high absorption. It is also interesting to note the fact that for the above-mentioned ranges of the sizes of particles, a peculiar diffraction picture in their neighboring field is observed. The highest maxima of this field, in which the energy density can be greater by an order of magnitude than that in the incident beam, are situated in the range of angles $\theta = 100 \div 140°$.

Such distribution of electrical energy density inside and near small particles with $n < 1$ seems also to be connected with the existence of the above-mentioned 'critical' range of scattering [20, 21], which for absorbing particles appears in a strictly definite range of sizes depending on specific values of n and χ of the particle material. The present authors were the first to discover this effect, which has not as yet been comprehensively investigated and explained. These investigations are being continued.

Now, let us consider in more detail the specific features of heating homogeneous and two-layered particles with high absorptivity under the action of intense laser radiation.

Investigations of the heating of homogeneous titanium and aluminum particles by highly intense laser radiation with $\lambda = 10.6$ μm was considered in detail in refs [24, 25]. As noted in ref. [17], in not very small metallic particles heat evolution, and, therefore, direct heating, is realized in a thin surface layer. The internal regions of particles are heated only at the expense of the thermal conductivity of the metal. Therefore, in the case of low intensities of radiation (slow heating regime) the temperature distribution can be practically uniform throughout the particle volume due to a long influence of radiation. Precisely this assumption is usually used by many authors for evaluating the effectiveness of heating metallic particles by laser radiation.

As already noted above, in the investigations of the present authors on the contrary fast heating regimes are discussed when the intensity of incident radiation is high enough for the temperature of phase transition (melting) to be reached at first only at separate 'hot' points of the particle volume.

For investigating the problem of laser heating of particles, which in a general case is formulated with the help of a system of equations (1)–(8), use is made, as already mentioned, of locally one-dimensional implicit scheme solved by the pivot method. Since the range of temperatures is sufficiently large, i.e. from room temperature $T = 293$ K to titanium melting temperature $T_m = 2000$ K and aluminum melting temperature $T_m = 933$ K, the temperature-dependent values of thermal conductivity, heat capacity and density of the above-mentioned metals were introduced

in accordance with the local values of temperature at each node of the grid on each temporal step with the help of approximating formulae. The values of the heat source functions were recalculated for the temperature-dependent values of n and χ with a certain optimum step over the volumetric average particle temperature. The conditions of the computations allowed us to obtain the error of the temperature values not exceeding 15–20%.

As a result of the investigations carried out, it is shown that the heating of particles along their main diameter has a substantially nonuniform character, with the inhomogeneities being higher, the higher the intensities of incident radiation and the larger the size of particles (at the same intensities). The time of attaining the first regions of melting inside particles with a fixed radius is approximately characterized by the dependence of the kind $t = aI^{-b}$. For small particles the time of their heating before the beginning of melting (t_m) increases with their radii. This effect is particularly evident in the range of sizes $1 < R < 7$ μm. For $R > 7$ μm the indicated dependence is expressed more weakly, whereas in the region $R > 15$ μm the function $t_m(R)$ is close to the asymptotic one [25].

For example, for titanium particles with different radii and with different incident radiation intensities it was shown in ref. [24] that it is possible, in principle to determine the regions of the values of R and I that correspond to homogeneous and inhomogeneous heating of particles before melting. Moreover, the approximate 10-fold excess of the time before the beginning of melting over the characteristic relaxation time is a criterion for defining the region of uniform heating.

Of particular interest are the results on the investigations of the heating of aluminum particles from the room temperature to the melting temperature $T = 933$ K by intense radiation with $\lambda = 0.53$ μm [26], since for this wavelength the real part of the refractive index $n < 1$, and the imaginary part is rather great ($m = 0.7 - i5.1$).

As expected, when heating particles by highly intense radiation, the maximum temperature is attained on the surface of the circular region; moreover, the higher the intensity, the closer the position of the maximum temperature to the maximum of heat evolution. With a decrease in the intensity, this temperature maximum smears and shifts to the side of smaller angles θ; it can even return to the illuminated end of the main diameter at intensities smaller than a certain value for each size of particle.

For small particles ($r < 0.3$ μm), the shift of the maximum temperature to the region $\theta = 0°$ occurs at higher incident radiation intensities. A clear idea about the temperature distribution on the particle surface and about the degree of the inhomogeneity of its heating is given by the dependencies presented in Fig. 2.

The distribution of temperature along the main diameter of such particles is qualitatively similar to

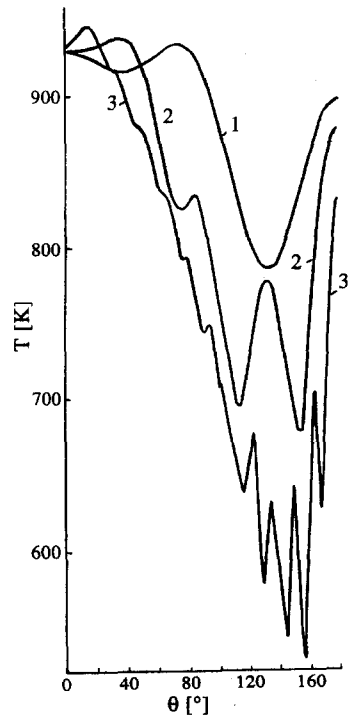

Fig. 2. Distribution of temperature over the aluminum particle surface under the action of radiation with $\lambda = 0.53$ μm, $I = 10^8$ W cm^{-2} up to the time of the beginning of melting at $R = 0.1$ (1), 0.3 (2), 1 (3) μm.

that obtained in refs [24, 25]. At not very high radiation intensities ($I < 5 \times 10^6$ W cm^{-2}) the dependence of the time before the beginning of melting has the character mentioned above ($t_m = aI^{-b}$). At higher intensities this regularity is violated, the time t_m decreases more and more rapidly with an increase in I.

As is known, the majority of metals and, consequently, metallic particles are oxidized when in air. Depending on specific physical conditions (temperature, humidity, etc.) and time, the thickness of the oxide layer formed can differ. For example, on aluminum particles at room temperature, the thickness of the oxidic film is 0.005–0.01 μm and at melting temperature it is of about 0.2 μm [27]. Since at the present time the powdery metals are finding an increasing use in the new technology (and often in active and corrosive media), it is of interest to investigate the regularities in heat evolution and heating of metallic particles with oxide films under the action of intense laser radiation of different wavelengths.

The computations of energy distribution inside such two-layered particles were first made in ref. [28]. They are based on the electromagnetic wave diffraction theory for a multilayered sphere [29, 30].

A highly characteristic situation with the absorbed energy distribution appears in the case of aluminum particles with an oxide film irradiated at $\lambda = 10.6$ μm. The optical properties of aluminum and aluminum oxide differ greatly ($m_{Al} = 30.6 - i64.8$ [31], $m_{Al_2O_3} =$

0.78 – i0.049 [32]). The appearance of an oxide film on the surface of aluminum particles leads to a noticeable increase in the effectiveness of energy absorption of such two-layered particles.

In the core of such a particle the heat evolution has the same character as in a homogeneous particle of aluminum with the maximum energy in the illuminated hemisphere, exponential decrease of energy in the thin surface layer and a lowered (compared to a homogeneous particle) heat evolution in the shadow hemisphere (Fig. 3). Just as in the case of homogeneous aluminum oxide particles, heat evolution inside the oxide layer is characterized by the presence of energy maximum shifted from the main diameter to the surface of the circular region with the angular coordinate depending on the particle size and the thickness of the layer. For example, for the same thickness of the layer $\Delta R = 0.1$ μm in the case of $R_2 = 3$ μm the heat evolution maximum lies in the region of the angles $\theta = 125$–$135°$ (in the shadow hemisphere), whereas for $R_2 = 10$ μm the heat evolution maximum shifts to the angular region $\theta \sim 75$–$80°$ (in the illuminated hemisphere, Fig. 3). The second in magnitude, but more narrow energy maximum that increases with an increase in temperature is observed in the shadow hemisphere in the region $\theta \sim 160$–$165°$. It should be noted that the distributions of absorbed energy over the core and oxide film surfaces differ substantially. Moreover, on increase in temperature the nonuniformity of heat evolution over the core surface

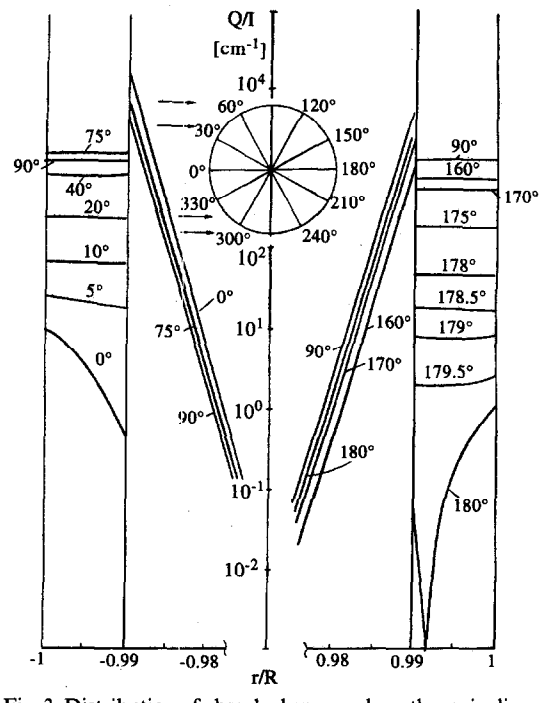

Fig. 3. Distribution of absorbed energy along the main diameter of aluminum with oxide shell particles at $R_1 = 9.9$ μm, $R_2 = 10$ μm, $\lambda = 10.6$ μm. The numbers at the curves are the values of the angle θ. Vertical lines are the boundaries between the core and the shell.

increases, whereas that over the layer surface somewhat decreases (as compared with the room temperature), in spite of the small relative thickness of the layer ($\Delta R/R \sim 1\%$).

The method of solving the problem of heating two-layered particles by light radiation was developed in ref. [33]. In the case of metallic particles with oxide film it is necessary to take into account the presence of absorption in the particle shell, as well as the change in the thermophysical properties of the core and the shell and their optical constants with an increase in temperature. According to the data of ref. [31], for aluminum we received approximate formulae of the kind $n_1 = 1.5 \times 10^5 T^{-1.5}$; $\chi_1 = 3.9 \times 10^4 T^{-1.125}$. For aluminum oxide such formulae were borrowed from ref. [32]: $n_2 = 0.78 + 1.47 \times 10^{-4} T$ and $\chi_2 = 1.94 \times 10^{-2} + 1.02 \times 10^{-4} T$. It is easy to see that with the change in the particle temperature from the room to the melting temperature ($T = 933$ K) the absorption index of aluminum decreases by almost a factor of 2.5, while the absorption index of the aluminum oxide increases by a factor of 2.3. As a result, for particles with $R_1 = 9.9$ μm, $R_2 = 10$ μm the quantity of energy absorbed in the core decreases by a factor of 1.3–1.4 and in the shell increases by a factor of about 1.5.

For the considered range of the size of particles ($R_2 = 1$–30 μm) their heating by highly intense radiation ($I > 10^6$ W cm^{-2}) is very nonuniform, with the nonuniformity increasing with an increase in I. For example, for particles with $R_1 = 9.9$ μm, $R_2 = 10$ μm the maximum temperature drop $\Delta T = T_{max} - T_{min}$ with $I = 1.5 \times 10^6$ W cm^{-2} is approximately 250 K, for $I = 10^7$ W cm^{-2} it is near 500 K and for $I = 10^8$ W cm^{-2} it approaches 900 K. The explanation is as follows. For high-intensity radiation the time for attaining the beginning of aluminum melting in a small part of the core volume of a particle is small and the heat conduction mechanism has no time to equalize the temperature throughout the particle volume.

The same occurs with an increase in the size of the particles. Thus, on exposure of particles with $R_2 = 1$, 3 and 10 μm (for the same oxide film thickness $\Delta R = 0.1$ μm) to radiation with $I = 5 \times 10^7$ W cm^{-2} the maximum temperature drop ΔT attains approximately 500, 650 and 750 K, respectively. This is due to the fact that in large particles the internal regions are less heated. Least heated is the region inside the particle, which is situated close to the main diameter near its center (Fig. 4). The maximum heating is attained on the external surface of the particle. The angular localization of the region of maximum heating depends on the particle size and shell thickness. In the case of small particles ($R_2 = 1$ μm) the region of the maximum heating lies on the circular layer surface along the direction perpendicular to the main diameter ($\theta = 90$–$270°$). With an increase in the size of particles to $R_2 = 3$ μm the region of maximum heating shifts to the shadow hemisphere ($\theta \sim 130°$). A further increase in the particle radii are accompanied by a shift

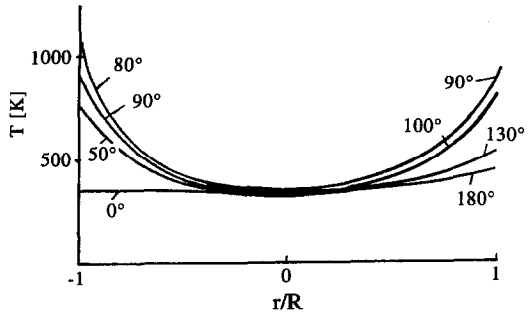

Fig. 4. Distribution of temperature along the main diameter of aluminum particles with oxide shells at $R_1 = 9.9$ μm, $R_2 = 10$ μm, $I = 5 \times 10^7$ W cm^{-2} up to the time of the beginning of melting of Al.

of the region of maximum heating to the illuminated hemisphere in the angular interval $\theta \sim 70$–$80°$ (Fig. 5).

Since the melting temperature of aluminum oxide $T_m = 2300$ K is considerably higher than that for the metal ($T_m = 933$ K), the first centers of melting appear in the region of the maximum heating on the surface of the core of two-layered particles. The time for the attainment of the beginning of the aluminum melting depends in a complicated manner on the core and the shell size and on the incident radiation intensity.

For particles with $R_2 > 5$ μm the distribution of absorbed energy within the core and shell changes inconsiderably. Therefore, for a constant shell thickness the time of the particle heating up to the beginning of melting increases due to a decrease in the fraction of the shell volume in the entire volume of particles of growing sizes.

At fixed dimensions of the core and shell the time up to the beginning of aluminum core melting is approximately inversely proportional to the incident radiation intensity.

As follows from the afore-mentioned, the heating of aluminum particles with oxide films differs substantially from the heating of homogeneous aluminum

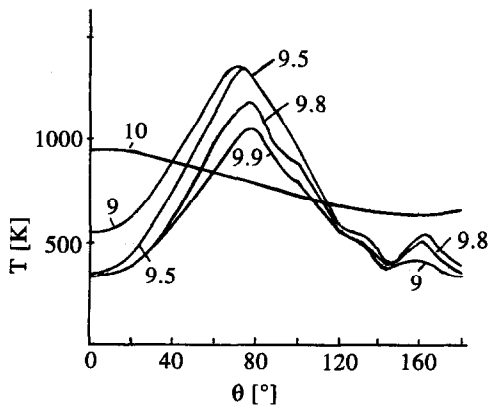

Fig. 5. Distribution of temperature on the surface of aluminum with oxide shell particles with $R_2 = 10$ μm at $I = 5 \times 10^7$ W cm^{-2} and at different core radii (shown near the curves) up to the moment of the beginning of aluminum melting.

particles in both the character of temperature distribution within the particles and the time of heating them up to the beginning of aluminum melting.

In nature [34, 35], as well as in different technological processes [36, 37] all kinds of hollow and two-layered particles are often encountered. The specific features of the absorption of radiation by hollow metallic and two-layered particles with metallic shells and also the optical fields within such particles are discussed in ref. [38]. To make investigation more specific, aluminum was selected as the metal, and the material of the type of polyethylene with $n_1 = 1.52$ and $\chi_1 = 0$ [39], was selected as the core of a two-layered particle. It is assumed that the particle is exposed to radiation with the wavelength $\lambda = 10.6$ μm.

The investigations showed that with an increase of the external radius R_2 the absorption of particles increases to a certain maximum. The value of this maximum depends on the thickness and optical properties of the shell. More intense absorption corresponds to thinner metallic shells. For example, for hollow particles with $R_2 = 3$ μm and $R_2 = 10$ μm, the decrease in the shell thickness by a factor of 10 leads to an increase in the radiation absorption efficiency factor by a factor of 25 and 10, respectively. For $R_1/R_2 < 0.995$ the radiation absorption by two-layered particles is still independent of the core material, but if $R_1/R_2 > 0.995$, the presence of the polymer core decreases the absorption of the two-layered particle as compared with the hollow particle in the radius range $R_2 < 2$ μm.

Such a change in the absorptive properties of hollow and two-layered particles on decrease in the metallic shell thickness seems to be connected with the following facts. Obviously, in the case of thick shells radiation is mainly reflected by the metal surface and is absorbed very little. In the case of thin shells, radiation begins to penetrate inside the particle and its greater part is absorbed in the shell due to multiple passage of beams in it. To confirm such an explanation, the energy distribution within hollow aluminum particles and two-layered metallized particles was considered. It was shown, in particular, that the absorption of the two-layered particles with $R_2 = 10$ μm is practically equal at the ratios $R_1/R_2 = 0.99$ and $R_1/R_2 = 0.995$, since the main part of the energy is evolved in a thin metal layer near the shell surface. In the case of two-layered particles with very thin shells ($R_1/R_2 < 0.999$) the energy density in the shell increases substantially, as a result of which the total absorption of such a particle increases (at a constant external radius R_2).

The distribution of heat sources within polyethylene particles covered with an aluminum shell is rather peculiar. First of all it should be noted that heat evolution differs from zero only in the bounds of metallic shells. The absorbed energy maximum is located on the illuminated surface of the particle (Fig. 6). Further from the surface, to the particle center, the quantity of the absorbed energy decreases both in the illumi-

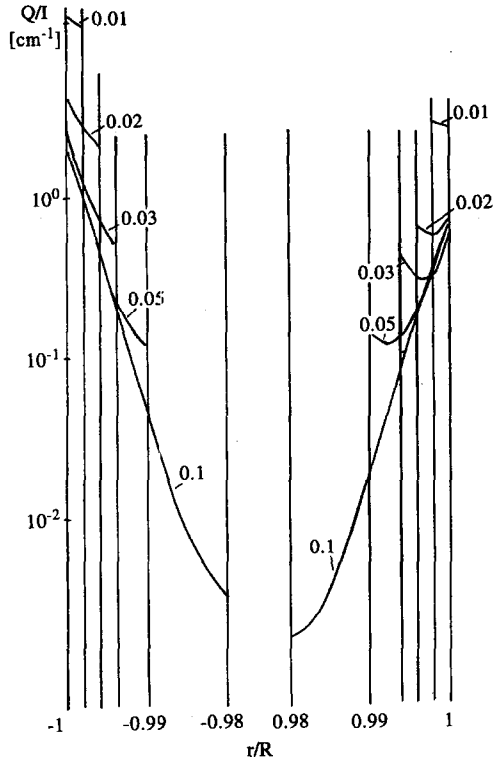

Fig. 6. Distribution of absorbed energy inside of the shell of metallized particles at $R_2 = 5$ μm. The numbers at the curves are shell thickness in μm. The vertical lines are the boundaries between the cores and shells.

nated and in the shadow part of the shell, but the character of the decrease differs essentially from the case of a homogeneous particle or a metallic core (Fig. 3). The maximum inhomogeneity in heat evolution is usually observed along the main particle diameter.

The specific value of heat evolution within an aluminum shell depends on its thickness. As the shell thickness decreases from $\Delta R = 0.05$ μm to $\Delta R = 0.02$ μm for $R_2 = 5$ μm the maximum value of the absorbed energy increases by a factor of 2.3. Beginning from a certain shell thickness the absorbed energy distribution inside it changes weakly after its further increase. This shell thickness depends on the value of the external particle radius. In the case of $R_2 = 1$ μm this value is equal to Δ0.05 μm; for $R_2 = 5$ μm the corresponding value is $\Delta R = 0.1$ μm

The heating of the above-mentioned two-layered particles by intense laser radiation is discussed within the range of the particle radii $1 > R_2 > 15$ μm for the shell thickness from 0.05 to 0.4 μm in the temperature range from the room to the melting temperature of polyethylene $T = 411$ K. It was taken into consideration that as polyethylene core does not practically absorb radiation with the wavelength $\lambda = 10.6$ μm, its heating takes place solely at the expense of heat transfer from the intensely absorbing metallic shell.

It is established that the region of maximum heating is located on the illuminated surface of the particle in the small angular range near $\theta = 0°$. Further from the particle surface to its center the heating temperature decreases and then increases again on approach to the shadow surface of the particle (the angular region near $\theta = 180°$). The nonuniformity of heating small particles with $R_2 < 5$ μm decreases noticeably on increase in the shell thickness. For particles of large radii ($R_2 = 5$ μm, Fig. 7) by the beginning of core melting the heating inhomogeneity depends weakly on the shell thickness and is approximately equal to $\Delta T = 110$–125 K for different values of ΔR. For particles with $R_2 > 10$ μm the quantity ΔT increases with the growth of the shell thickness at the expense of a weak change of the heating temperature at the center of the particle and noticeable increase of it on the surface with an increase of ΔR.

The heating time of the core of particles from room to melting temperature of polyethylene for the particles of middle sizes ($R_2 = 5$ μm) depends strongly on the shell thickness up to the value $\Delta R = 0.1$ μm, beginning from which the heat evolution in the shell depends weakly on its thickness. With a further increase of ΔR, the time of the attainment of T_m increases inconsiderably. As concerns the effect of the incident radiation intensity, then with an increase of I by an order of magnitude (from $I = 10^7$ W cm^{-2}) the

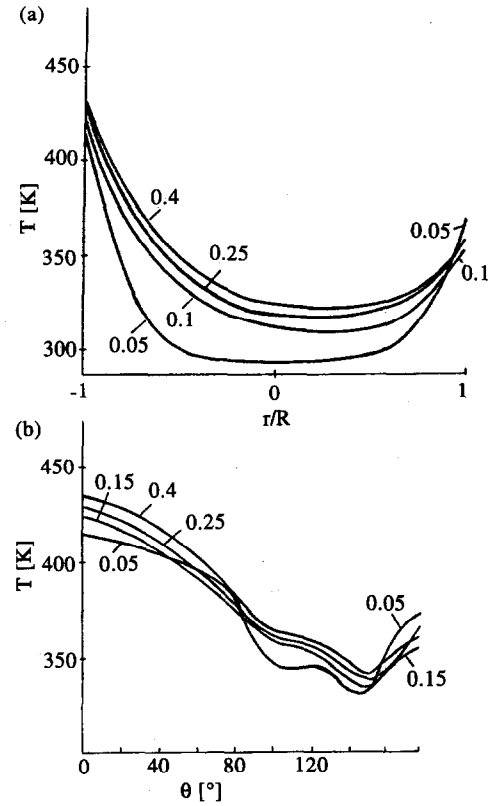

Fig. 7. Distribution of temperature inside metallized particles with different shell thickness up to the time of the beginning of melting: (a) along the main diameter ($\theta = 0$–$180°$), (b) on the particle surface at $R_2 = 5$ μm, $I = 5 \times 10^7$ W cm^{-2}. The numbers at the curves are the values of shell thickness in μm.

time of core heating up to T_m decreases by more than an order of magnitude.

Comparison of the behavior of curves in Figs 6 and 5 shows that the character of the heating of two kinds of two-layered particles under the action of intense laser radiation differs noticeably. The basic reason of this difference is the circumstance that the particle shells of aluminum oxide for a given wavelength are characterized by the real part of the refractive index $n < 1$.

It follows from the results of the investigations considered that under the action of highly intense laser radiation on solid absorbing particles their heating is usually strongly inhomogeneous. In this case the appearance of specific physical situations that manifest themselves in the thermal destruction of particles, partial (directed) ejection of their mass, noticeable decrease of threshold conditions of optical breakdown, formation of plasma and other nonlinear effects is possible. All of these effects are typical for the 'so-called' fast heating regimes when the nonuniformity of temperature distribution within the particles is preserved during the entire period of radiation action. These specific effects should be taken into account when studying the propagation of intense laser radiation in a medium with solid particles, photoablation of semiconducting materials, creation and deposition of thin films, industrial production of powder materials, laser machining of composites, metal matrix materials, and so on.

Unfortunately, up to the present time the boundary between the 'fast' and 'slow' regimes of the action of intense laser radiation on separate particles and systems of such particles remains uncertain. Only one work is known [40], in which an attempt is made to determine quantitatively this boundary for water aerosol droplets. The question of determining an analogous criterion for other disperse particles (metals, oxides, etc.) remains open. The authors consider the investigations described in this work as a necessary step on the way to determining such a criterion. At the same time, separate results of this work can surely be taken into account in a number of practical applications of the modern laser technology (some of them are enumerated above).

REFERENCES

1. Pogodaev, V. A., Action of the CO_2-laser radiation on solid particles. II *All-Union Symposium on Propagation of the Laser Radiation in the Atmosphere*, Tomsk (Abstracts), 1973, pp. 316–318.
2. Propagation of the high intense optical radiation in solid aerosol. Inter-institute collection, Barnaul, 1982.
3. Nonlinear interaction of intense laser radiation with solid aerosol. Inter-institute collection, Barnaul, 1988.
4. Bukatyi, V. I., Kronberg, T. K. and Shaiduk, A. M., The effect of nonlinear scattering on the dynamics of transparency of a burning aerosol. *Inzhenerno-Fizicheskiy Zhurnal*, 1988, **55**, 409–412.
5. Zuev, V. E., Kopytin, Yu. D. and Kuzikovskiy, A. V., *Nonlinear Optical Effects in Aerosol*. Izdatelstvo Nauka, Novosibirsk, 1980.
6. Zuev, V. E., Zemlyanov, A. A., Kopytin, Yu. D. and Koozikovskiy, A. V., *Intense Laser Radiation in the Atmospheric Aerosol*. Izdatelstvo Nauka, Novosibirsk, 1984.
7. Baranov, A. V., Klyukach, I. L. and Ubogov, S. A., *Laser Heating of Small Metallic Particles in Inert Gas. Interaction of the Radiation with Material*. Izdatelstvo Kuiybyshev Gosuniversitet, 1988.
8. Poulain, D. E., Alexander, D. R., Barton, J. P., Schaub, S. A. and Zhang, J., Interaction of intense ultraviolet laser radiation with solid aerosol. *Journal of Applied Physics*, 1990, **67**, 2283–2288.
9. Pogodaev, V. A., Solid aerosol particle in intense light field. Before breakup regime of the interaction. *Optika Atmosfery i Okeana*, 1992, **5**, 391–396.
10. Besprozvannykh, V. A., Ermakov, V. A. and Razdobreev, A. A., The explosion of metallic particles in the field of laser radiation. The combustion of heterogeneous and gaseous systems. *Proceedings of the 8th All-Union Symposium on Combustion and Explosion*, OIChF AN SSSR, Chernogolovka, 1986, pp. 58–62.
11. Prishivalko, A. P., *Optical and Heat Fields Inside Light Scattering Particles*. Izd. Nauka Tekhnika, Minsk, 1983.
12. Prishivalko, A. P., Semenov, L. P., Astafieva, L. G. and Leiko, S. T., The heating destruction of spherical ice particles under the action of radiation with wavelength $\lambda = 10.6$ μm. *Inzhenerno-Fizicheskiy Zhurnal*, 1988, **54**, 103–108.
13. Astafieva, L. G., Prishivalko, A. P., Semenov, L. P. and Leiko, S. T., The effect of the variation of temperature and optical constants of ice particles on their heating destruction by radiation with $\lambda = 10.6$ μm. *Optika Atmosfery*, 1988, **1**, 63–69.
14. Lednyova, G. P., Prishivalko, A. P., Leiko, S. T. and Katseva, I. R., The heating of solid particles by laser radiation in vacuum. Preprint of the Institute of Physics of the BSSR Academy of Sciences no. 491, Minsk, 1987.
15. Lednyeva, G. P. and Prishivalko, A. P., The heating of carbon particles by laser radiation in vacuum. *Izvestiya AN BSSR, Seriya Fiziko-Matematicheskikh Nauk*, 1989, pp. 60–64.
16. Dalzell, W. and Sarofim, A. F., Optical constants of soot and their application to heat-flux calculations. *Transactions of the ASME, Section C*, 1969, **91**, 100–104.
17. Astafieva, L. G., Prishivalko, A. P. and Leiko, S. T, About the movement of metallic particles in the field of intense optical radiation. *Fizika i Khimiya Obrobotki Materialov*, 1989, (4), 35–39.
18. Prishivalko, A. P. and Zarubo, I. V., Internal electrical fields in small metallic particles by laser radiation. *Izvestiya AN BSSR, Seriya Fiziko-Matermaticheskikh Nauk*, 1991, (5), 40–43.
19. Astafieva, L. G. and Prishivalko, A. P., The specific features of optical fields inside spherical particles with relative refractive index $n < 1$. *Optika i Spectroskopiya*, 1990, **69**, 398–401.
20. Marston, P. L., Critical angle scattering by a bubble: physical–optical approximation and observation. *Journal of the Optical Society of America*, 1979, **69**, 1205–1211.
21. Marston, P. L., Langley, D. C. and Kingsbury, D. L., Light scattering by bubbles in liquids: Mie theory, physical–optical approximation and experiments. *Applied Science Research*, 1982, **38**, 373–383.
22. Prishivalko, A. P. and Astafieva, L. G., About the distribution of laser beam energy in small metallic particles with $n < 1$. *Izvestiya AN SSSR, Metally*, 1991, (3), 192–195.
23. Zolotaryov, V. N., Morozov, V. N. and Smirnova, E. V., *Optical Constants of Natural and Technical Media*. Izd. Khimiya, Leningrad, 1984.
24. Astafieva, L. G., Prishivalko, A. P. and Leiko, S. T., The heating of metallic particles by laser radiation. *Fizika i Khimiya Obrabotki Materialov*, 1991, (2), 64–69.

25. Astafieva, L. G., Prishivalko, A. P. and Leiko, S. T., The heating of aluminum particles by radiation. *Teplofizika Vysokich Temperatur*, 1992, **30**, 579–583.
26. Astafieva, L. G. and Prishivalko, A. P., The heating of solid aluminum particles by intense radiation with $\lambda = 0.53$ μm. *Metallofizika*, 1991, **13**, 122–127.
27. Pochil, P. F., Belyayev, A. F., Frolov, Yu. V., Logachyov, S. V. and Korotkov, A. I., *Combustion of Powdered Metals in Active Media*. Izdatelstvo Nauka, Moscow, 1972.
28. Astafieva, L. G. and Prishivalko, A. P., Absorption of IR-radiation and optical fields inside metallic particles with oxide films. *Optika Atmosfery*, 1989, **9**, 1089–1092.
29. Toon, O. B. and Ackerman, T. P., Algorithms for the calculation of scattering by stratified spheres. *Applied Optics*, 1981, **20**, 3657–3664.
30. Bhandari, R., Scattering coefficients for a multilayered sphere: analytical expressions and algorithms. *Applied Optics*, 1985, **24**, 1960–1967.
31. Dreehsen, H. G., Hartwich, C., Schaefer, J. H. and Uhlenbusch, J., Measurement of the optical constants of Al above the melting point at $\lambda = 10.6$ μm. *Journal of Applied Physics*, 1984, **56**, 238–240.
32. Belov, N. N., Complex refractive index of aluminum oxide. *Kristallografiya*, 1989, **34**, 1185–1190.
33. Pastushenko, V. V., Prishivalko, A. P. and Leiko, S. T., Heating by electromagnetic radiation of absorbing particle covered of transparent shell. *Doklady AN BSSR*, 1979, **23**, 233–236.
34. Green, H. L. and Lane, W. R., *Particulate Clouds: Dust, Smokes and Mists*. Izdatelstvo Khimiya, Leningrad, 1972.
35. Cadle, R. D., *Solid Particles in Atmosphere and Space*. Izdatelstvo Mir, Moscow, 1969.
36. Kudinov, V. V., Puzanov, A. A. and Zambrzhitskiy, A. P., *Optics of Plasma Coatings*. Izdatelstvo Nauka, Moscow, 1981.
37. Kudinov, V. V. and Ivanov, V. M., *Deposition by Plasma of Refractory Coatings*. Izdatelstvo Nauka, Moscow, 1981.
38. Astafieva, L. G. and Prishivalko, A. P., The specific features of absorption of IR-radiation and of optical fields inside the hollow metallic and metallized particles. *Doklady AN BSSR*, 1990, **34**, 127–129.
39. Speranskaya, T. A. and Tarutina, Z. I., *Optical Properties of Polymers*. Izdatelstvo Khimiya, Leningrad, 1976.
40. Park, B. S. and Armstrong, R. L., Fast and slow heating regimes. *Applied Optics*, 1989, **28**, 3671–3680.