High Temperature. Vol. 42, No. 5, 2004, pp. 776–784. Translated from Teplofizika Vysokikh Temperatur, Vol. 42, No. 5, 2004, pp. 772–779.

HEAT AND MASS TRANSFER AND PHYSICAL GASDYNAMICS

Approximate Models of Radiation Scattering in Hollow-Microsphere Ceramics

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Received December 2, 2003

Abstract—Theoretical models are suggested, which explain the strong scattering of visible and infrared radiation in hollow-microsphere ceramics and are based on the assumption of predominant scattering of radiation in the contact zones of neighboring microspheres. The approximation of the results of calculations by the Mie theory is used in one of the models to estimate the transport coefficient of attenuation; the other model, which produces approximately the same results, involves the use of solution for radiation scattering by random-oriented plates. The validity of the suggested theoretical description and derived approximate formulas is confirmed by comparing the calculated values of the spectral coefficient of radiation diffusion to the experimental data for two types of heat-insulating ceramics made of hollow spherical particles of alumina.

INTRODUCTION

The theoretical description of the optical properties of dispersed materials in the visible and infrared spectral regions presents one of the most important problems of the current theory of heat transfer, because it defines in many respects the possibility of computational analysis of the characteristics of modern high-porosity heat-insulating materials. A review of early publications on this subject, made by Vortmeyer [1], contains descriptions of both discrete and continual models. The latter include various options of a model of radiative thermal conductivity which differ from one another by the semiempirical formulas for the coefficient of radiative thermal conductivity of dispersed material. In addition to providing information about subsequent publications on the subject, more recent review papers [2, 3] for the first time made a distinction between the region of independent scattering of radiation by individual particles and the region of marked interference effects in the near and far fields united by the common term “dependent scattering”. In the case of independent scattering, the interaction of radiation with particles of dispersed materials is the same as in the absence of neighboring particles. In so doing, the properties of the material may be calculated, for example, using the Mie theory for individual particles or fibers [4, 5]. This situation is observed, for example, for numerous high-porosity fibrous heat-insulating materials [5–7]. The theoretical description of the optical properties of materials with close-packed particles is, on the contrary, very difficult to make. By now, only some semiempirical models have been developed, which relate to disperse systems of special type. A number of papers were devoted to close-packed spherical particles [8–11]; approximate solutions are available for foamy structures [12, 13] and porous ceramics [14, 15]. The current status of investigations in this field is discussed in the review by Baillis and Sacadura [16].

The recent paper of Moiseev *et al.* [17] contains experimental data on the optical properties of an interesting new high-temperature heat-insulating material of caked hollow microspheres. The information about the structure of the material given in [17] is fairly complete and may be used both for estimating the error of the classical model of independent scattering and for constructing a theoretical model which takes into account the additional scattering of radiation in a close-packed dispersed system.

It is not only with high-temperature applications that interest in materials containing ceramic or glass microspheres is associated. In view of this, note the paper by German and Grinchuk [18] which deals with the heat-shielding properties of a composite coating of hollow microspheres and a binder. Such coatings are used for additional heat shielding of buildings. The theoretical model of heat transfer in such semitransparent coatings suggested in [18] includes, as an essential element, the description of thermal radiation transfer in a medium containing microspheres 10 to 50 μm in diameter, the distances between which are commensurable with the dimensions of the microspheres. The impact of the effects associated with the high concentration of microspheres is estimated using the solution [19] of the problem on scattering of a spherical wave whose source is located at a close range to a particle.

In this paper, I deal with simpler estimates relating to a particular case of thin-walled hollow microspheres whose diameter significantly exceeds the radiation wavelength.

OPTICAL PROPERTIES OF SEMITRANSPARENT HOLLOW PARTICLES

The absorption and scattering of radiation by isolated hollow spherical particles may be calculated using the well-known solution which represents a generalization of the classical Mie theory [4, 5, 20]. In the semi-transparency region, the absorption is relatively low and weakly affects the scattering and attenuation of radiation by particles. In addition, the low absorption is very sensitive to the presence of minor impurities of absorbing substances. Therefore, I will restrict myself to determining only the transport factor of attenuation efficiency Q_{tr} which is required for the calculation of the spectral coefficient of radiation diffusion D_λ in a dispersed system of particles being treated.

The value of Q_{tr} depends on the following dimensionless parameters: the particle diffraction parameter $x = 2\pi r/\lambda$ (r is the outer radius of particle and λ is the radiation wavelength), the complex refractive index of the particle matter $m = n - i\kappa$ (n is the refractive index and κ is the index of absorption), and the relative thickness of the particle wall $\Delta = \delta/\lambda$. As was mentioned above, at $\kappa \ll 1$, the dependence on the absorption index κ may be ignored assuming that $\kappa = 0$. At the same time, the spectral dependence of the refractive index must be taken into account. In the spectral range of $0.5 < \lambda < 3.5$ μm, which is of most interest from the standpoint of engineering applications of high-temperature ceramics, the refractive index of the majority of metal oxides varies in the range from 1.5 to 2. Figure 1 gives the calculated dependences $Q_{tr}(\Delta)$ for these values of the refractive index and different fixed values of the particle diffraction parameter. The preassigned high values of the diffraction parameter correspond to the range of practical interest, because the particle radius usually exceeds 20 μm [17]. In preassigning the range of variation of the parameter Δ, it was taken into account that the particle wall thickness did not exceed 5 μm and that $\delta/r < 0.1$ [17]. To make the physical picture complete, the calculation data for very small values of Δ are given as well. At the same time, the region of $\Delta > 1.5$, in which the $Q_{tr}(\Delta)$ dependence is obvious, is not shown in Fig. 1. The calculations reveal that the curves for different values of x differ insignificantly from one another. This result implies the degeneracy of the general solution and transition to the region of anomalous diffraction at $x \gg 1$ [21]. Note the large-scale oscillations of the $Q_{tr}(\Delta)$ curves caused by the interference of radiation passing through the particle. However, these oscillations are of no consequence as regards real polydisperse systems which contain particles with walls of different thickness. It is interesting that the oscillations of $Q_{tr}(\Delta)$ are approximately symmetric relative to some constant value of $\bar{Q}_{tr}$ which increases with the refractive index. The results of additional calculations, partly given in Fig. 2, showed the dependence $\bar{Q}_{tr}(n)$ to be close to linear and to be adequately approximated by the simple formula

$$\bar{Q}_{tr} = 0.36(n-1). \quad (1)$$

We will consider the physical meaning of the limiting dependence $Q_{tr}(\Delta)$ for large thin-walled spherical particles. Because the value of Q_{tr} does not depend on the particle radius, it is natural to assume that different elements of such a particle scatter radiation independently of one another. In this case, a particle may be represented as a combination of a large number of isolated small flat plates.

The coefficient of reflection of nonpolarized radiation for each one of such plates may be determined using the known solution [22]

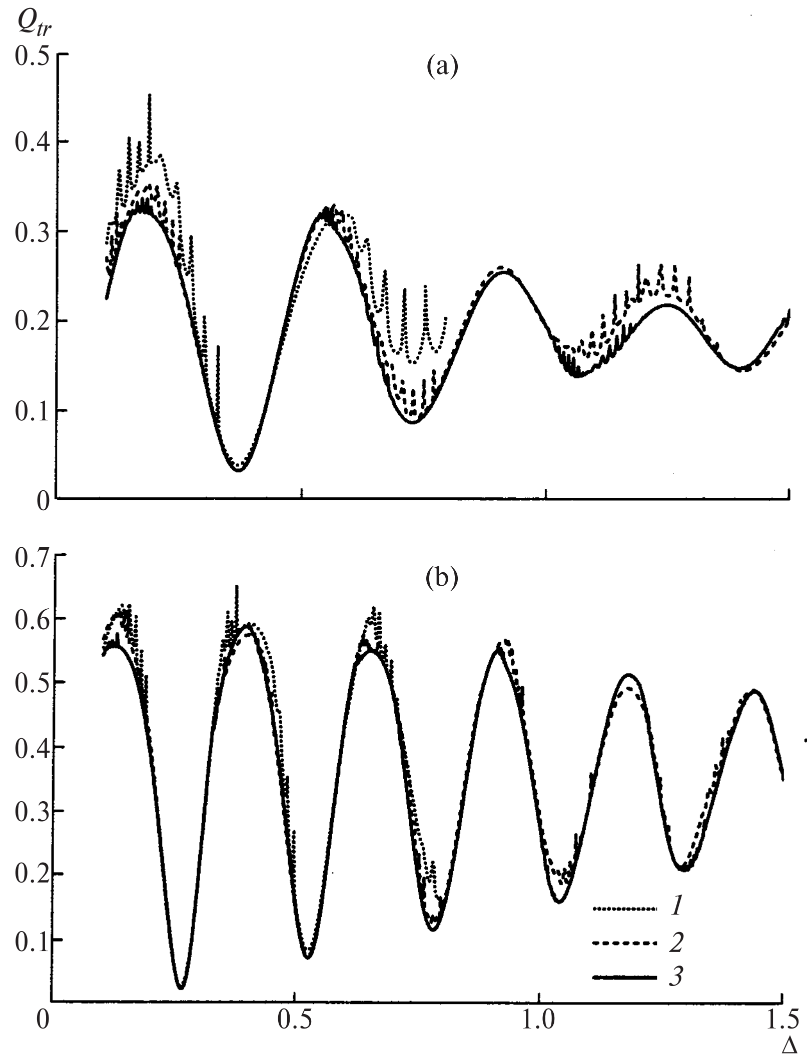

Fig. 1. The transport factor of attenuation efficiency for large hollow spherical particles of weakly absorbing matter with the refractive index n = (a) 1.5 and (b) 2 as a function of the dimensionless thickness of particle walls Δ: (*1*) x = 50, (*2*) 100, (*3*) 200.

$$R = (R_{\|} + R_{\perp})/2,$$

$$R_{\|} = \frac{2\rho_{\|}^2 - 2\rho_{\|}^2\cos(2\beta)}{1 + \rho_{\|}^4 - 2\rho_{\|}^2\cos(2\beta)},$$

$$R_{\perp} = \frac{2\rho_{\perp}^2 - 2\rho_{\perp}^2\cos(2\beta)}{1 + \rho_{\perp}^4 - 2\rho_{\perp}^2\cos(2\beta)}, \quad (2)$$

$$\rho_{\|} = \frac{\cos\theta - n\cos\theta'}{\cos\theta + n\cos\theta'}, \quad \rho_{\perp} = \frac{n\cos\theta - \cos\theta'}{n\cos\theta + \cos\theta'},$$

$$\beta = 2\pi\Delta\cos\theta', \quad \sin\theta/\sin\theta' = n,$$

where θ is the angle between the direction of incident radiation and the normal to the plate surface. Because the angle between the directions of incident and reflected radiation is $\pi - 2\theta$, the transport factor of efficiency attenuation is calculated by the obvious relation

$$Q_{tr}^{(1)}(\theta) = R(\theta)[1 + \cos(2\theta)]. \quad (3)$$

If the plates which make up a hollow spherical particle are placed in the same volume but are randomly oriented in space, the transport factor of attenuation efficiency of such a dispersed system may be calculated by the formula

$$Q_{tr} = 4\int_0^{\pi/2} Q_{tr}^{(1)}(\theta)\cos\theta d\theta. \quad (4)$$

If we take into account the distribution of plates with respect to their orientations in forming the shell of a hollow particle, a more complex relation is derived instead of formula (4),

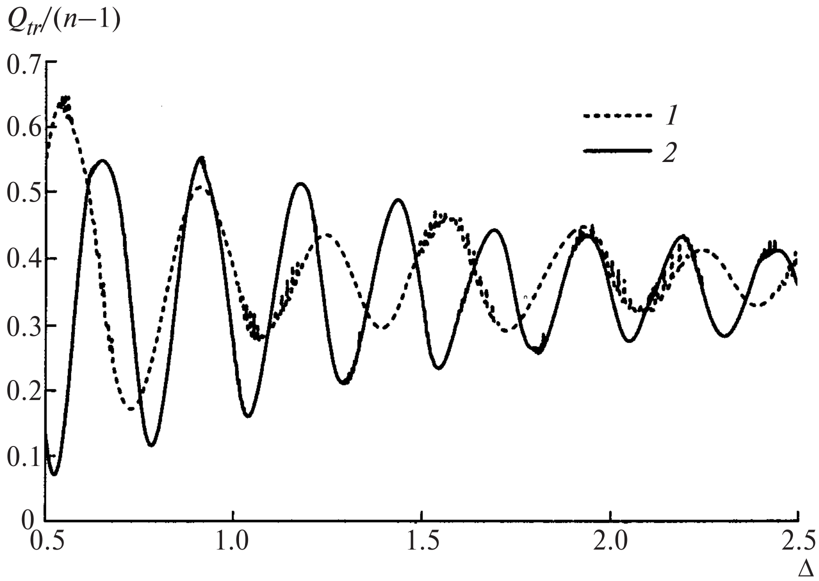

Fig. 2. Estimation of the mean values of the transport factor of attenuation efficiency for large hollow spherical particles. Calculation by the Mie theory for $x = 200$: (*1*) $n = 1.5$, (*2*) $n = 2$.

$$Q_{tr} = 2\int_0^{\pi/2} [2 - R(\theta)]Q_{tr}^{(1)}(\theta)\cos\theta\sin\theta d\theta. \quad (5)$$

Also taken into account in this case is the insignificant shading of the hemisphere located on the side opposite to incident radiation.

Figure 3 gives comparison of the results of calculations by the Mie theory for a large hollow spherical particle to the results of calculations by Eqs. (4) and (5) for a combination of small plates. One can see that Eq. (5) describes well the scattering of radiation by a spherical particle. This supports the assumption that different elements of a large hollow particle scatter radiation independently of one another. At the same time, the randomly oriented plates attenuate the radiation to a much greater extent; in this case, the following approximate relation may be recommended instead of Eq. (1) for the average value of the transport coefficient of attenuation efficiency:

$$\bar{Q}_{tr} = 1.3(n - 1). \quad (6)$$

OPTICAL PROPERTIES OF A RAREFIED POLYDISPERSE SYSTEM

If we ignore the effects associated with the close packing of hollow spherical particles in a material, we can readily calculate the transport coefficient of attenuation Σ_{tr} for a polydisperse system of such particles. We ignore the weak dependence of Q_{tr} on r and take into account only the distribution over the particle wall thickness $F(\delta)$ to derive, at $\delta << r$ [5],

$$\Sigma_{tr} = \frac{1-p}{4}\int_{\delta_1}^{\delta_2} Q_{tr}F(\delta)d\delta \Big/ \int_{\delta_1}^{\delta_2} \delta F(\delta)d\delta. \quad (7)$$

Here, $p = 1 - \rho/\rho_s$ is the porosity of the dispersed system (ρ is the density of the dispersed system and ρ_s is the density of particle matter). In the case of uniform distribution in the interval (δ_1, δ_2), the function $F(\delta) = 1/(\delta_2 - \delta_1)$, and we have, instead of Eq. (7),

$$\Sigma_{tr} = \frac{1-p}{2}\frac{\int_{\delta_1}^{\delta_2} Q_{tr}d\delta}{\delta_2^2 - \delta_1^2}, \quad (8)$$

or, assuming that $\int_{\delta_1}^{\delta_2} Q_{tr}d\delta = \bar{Q}_{tr}(\delta_2 - \delta_1)$,

$$\Sigma_{tr} = \frac{(1-p)\bar{Q}_{tr}}{4\bar{\delta}}, \quad (9)$$

where $\bar{\delta} = (\delta_1 + \delta_2)/2$. We use the approximate formula (1) and write Eq. (9), in the form

$$\Sigma_{tr} = 0.09\frac{(1-p)(n-1)}{\bar{\delta}}. \quad (10)$$

The spectral coefficient of radiation diffusion is defined as [5]

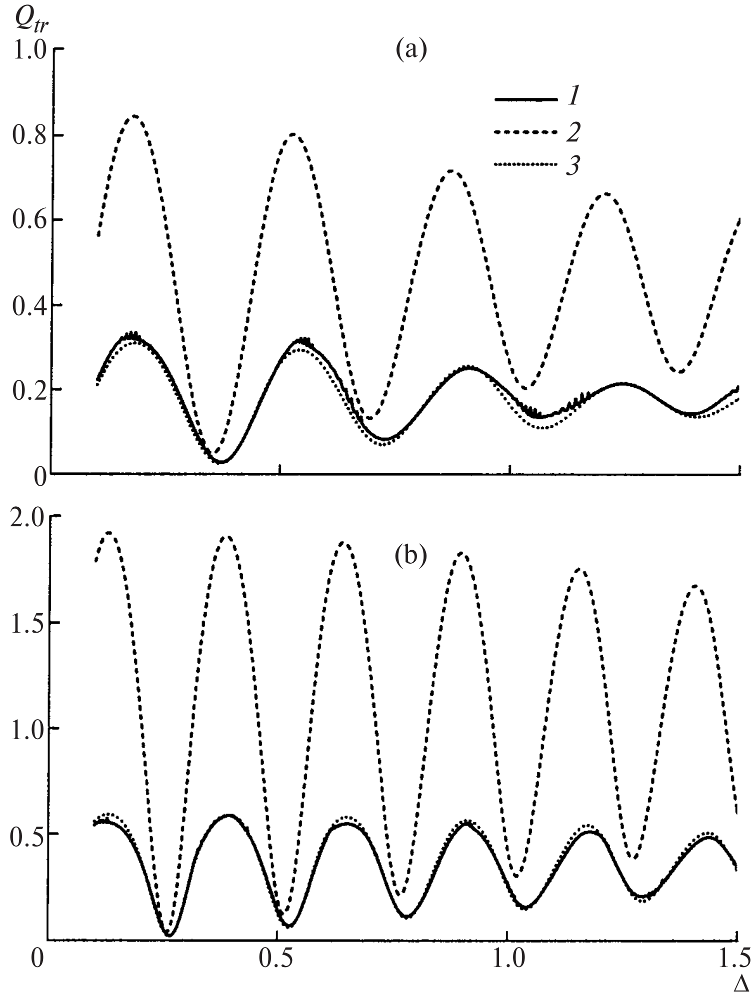

Fig. 3. Comparison of the attenuation of radiation by a large hollow particle and by a set of flat plates of weakly absorbing matter with the refractive index n = (a) 1.5 and (b) 2: (*1*) calculation by the Mie theory for x = 200, (*2*) random-oriented plates, (*3*) plates on a spherical surface.

$$D_\lambda = \frac{1}{3\Sigma_{tr}}. \quad (11)$$

We substitute expression (10) into Eq. (11) to derive

$$D_\lambda \approx \frac{3.7\bar{\delta}}{(1-p)(n-1)}. \quad (12)$$

One can see that the radiation diffusion coefficient is directly proportional to the mean thickness of the particle walls and inversely proportional to the concentration of particles in the dispersed system. Equation (12) is valid for rarefied dispersed systems and, therefore, does not include the distance between particles.

MODELING OF THE OPTICAL PROPERTIES OF A DENSE DISPERSE SYSTEM

The mean interparticle clearance in a material made up by close-packed hollow spherical particles is much less than the particle diameter. Under these conditions one cannot ignore the effect of the close packing of particles on the scattering of radiation. On the contrary, the interference between elements of a single hollow particle spaced apart may be ignored compared to the interaction between elements of neighboring particles. The latter statement is based on the results of foregoing theoretical analysis which revealed that individual elements of a large thin-walled spherical particle scatter radiation independently of one another.

Different methods are possible of describing the scattering of radiation in contact zones of neighboring particles. For example, a qualitatively correct solution may be derived by treating a rarefied dispersed system of hollow spherical particles modeling randomly oriented clearances. It is natural to assume that such particles have the same wall thickness as real spherical

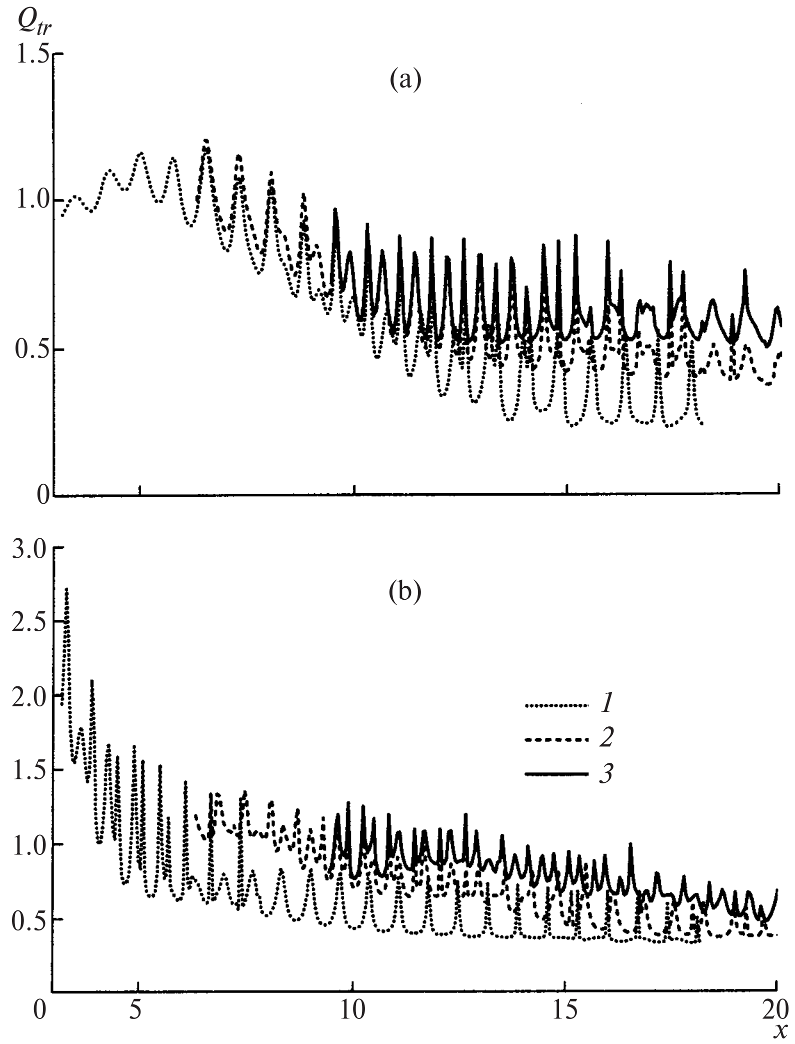

Fig. 4. The transport factor of attenuation efficiency for non-absorbing hollow particles in the Mie scattering region as a function of the diffraction parameter x: (a) $n = 1.5$, (b) $n = 2$; (*1*) $\Delta = 0.5$, (*2*) $\Delta = 1$, (*3*) $\Delta = 2$.

particles, and their mass concentration coincides with the density of the material.

Obviously, the transport coefficient of attenuation of such a model dispersed system may still be calculated by formula (9); however, in determining the value of $\bar{Q}_{tr}$, one must take into account that the new particles are much finer, and the approximate formula (1) is not suitable in their case.

We will again turn to the calculations by the Mie theory, but for the case of finer hollow particles. The results of such calculations are given in Fig. 4. One can see that, with the diffraction parameter of $3 < x < 20$, the transport coefficient of attenuation efficiency assumes much higher values than those in the large particle limit ($x > 50$). In so doing, the value of $\bar{Q}_{tr}$ continues to approximately linearly increase with the refractive index. The dispersed composition of new particles which model the clearances may hardly be determined single-valuedly. For a quantitative estimation of the equivalent mean value of $\bar{Q}_{tr}$, it is natural to assume that the maximal contribution is made by particles with the diffraction parameter $x \approx 5$ to 10, which scatter the radiation more intensively. The following approximate expression may be written for such particles:

$$\bar{Q}_{tr} = 1.5(n-1). \tag{13}$$

The substitution of Eq. (13) into (9) and (11) gives

$$\Sigma_{tr} \approx 0.38\frac{(1-p)(n-1)}{\delta}, \tag{14}$$

$$D_{\lambda} \approx \frac{0.9\bar{\delta}}{(1-p)(n-1)}. \tag{15}$$

As was to be expected, Eq. (15) produces much lower values of the spectral coefficient of radiation diffusion than Eq. (12).

An obvious disadvantage of the model discussed above consists in that it ignores the complex shape of the clearances which are described by a set of hollow spherical particles. In so doing, the increase in scattering compared to a rarefied dispersed system is attributed to the interaction between closely-spaced elements of the particle walls.

We will treat an alternative model in which emphasis is laid on the complex shape of the clearances, while the scattering by isolated elements of the particle walls is, on the contrary, taken to be independent. In view of this, we should recall the foregoing result obtained for a dispersed system of thin plates. It turned out that random-oriented plates scattered the radiation much more actively than in the case of orientation they would have if they were located on the surface of a sphere. The scattering is enhanced only because of the orientation of the plates relative to the direction of lighting rather than due to the interaction between the plates in the case of their high concentration.

In the new model, the differently oriented walls of complex-shaped clearances are treated as a combination of random-oriented plates of the same wall thickness. In this case, Eq. (6) must be used for the mean transport factor of efficiency, and we derive, instead of Eqs. (14) and (15),

$$\Sigma_{tr} \approx 0.32\frac{(1-p)(n-1)}{\delta}, \qquad (16)$$

$$D_\lambda \approx \frac{\bar{\delta}}{(1-p)(n-1)}. \qquad (17)$$

Equations (16) and (17) almost coincide with the analogous relations (14) and (15), except for the insignificant difference in the coefficients.

COMPARISON WITH EXPERIMENTAL DATA

The results of calculations by the suggested theoretical models may be compared to the experimental data of [17] on the spectral coefficient of radiation diffusion for two types of heat-insulating ceramics made of hollow spherical particles of alumina. The KMB-0.44 ceramics investigated in [17] (ρ = 440 kg/m^3, p = 0.89) consisted of larger particles (r_{32} = 65 to 70 μm) with an average wall thickness $\bar{\delta}$ = 3.16 μm,

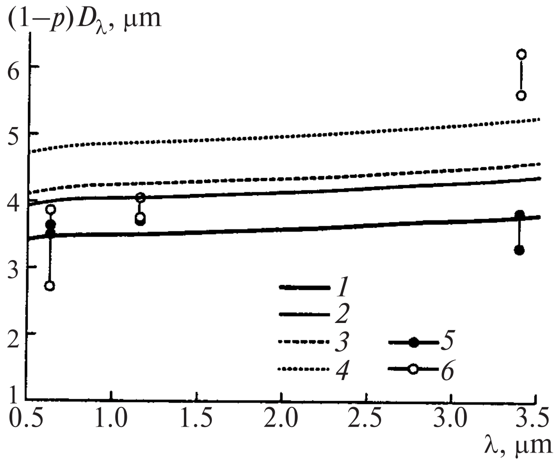

Fig. 5. Comparison of the calculated values of the spectral coefficient of radiation diffusion (curves *1* and *2*, model of spherical particles; curves *3* and *4*, model of random-oriented plates) to the experimental data of [17] (vertical segments *5* and *6*): (*1*, *3*, *5*) KMB-0.44 ceramics, (*2*, *4*, *6*) KMB-1.1 ceramics.

and the KMB-1.1 ceramics (ρ = 1100 kg/m^3, p = 0.73) consisted of particles with r_{32} = 55 μm and $\bar{\delta}$ = = 3.63 μm. The experimentally obtained values of D_λ for these two ceramics significantly differ from each other. Turning to relations (15) and (17), we can assume that this is largely due to the different porosities of the ceramics. Indeed, the values of $(1-p)D_\lambda$ for two ceramics given in Fig. 5 are close to each other.

Also plotted in Fig. 5 are calculated dependences obtained using formulas (15) and (17) in view of the spectral dependence of the refractive index of alumina according to the data of Malitson [23]. One can see that both theoretical models adequately describe the experimental data of [17]. The maximal divergence between theory and experiment is observed for the KMB-1.1 ceramics in the long-wave region. This result may be attributed to the impurities in the ceramics observed in [17], which cause an increase in the effective index of absorption; as a result, the attenuation efficiency factor decreases [5]. Another possible reason for the decrease in the attenuation efficiency factor and for the respective increase in the coefficient of radiation diffusion for denser ceramics in the long-wave range consists in the decrease in the relative size of clearances between the microspheres, when a significant part of clearances becomes less than the radiation wavelength. Note further that it is in the long-wave spectral region that the shell thickness distribution of particles $F(\delta)$ affects the properties of a dispersed system most strongly; Moiseev *et al.* [17] give only the ranges of variation of δ for isolated fractions of particles.

Note that Eq. (12) for a rarefied dispersed system gives approximately four times the values of the radiation diffusion coefficient than relations (15) and (17) in the suggested models.

Apparently, a rigorous physical model of the optical properties of the material being treated must include both the electromagnetic interaction of closely-spaced walls of neighboring particles and the complex shape of the interparticle clearances (complex conditions of lighting of particle elements). The first one of the suggested models, in which the clearances are represented as a combination of relatively fine hollow spherical particles, lays emphasis on the effect of dependent scattering of closely-spaced particle walls. The second model, on the contrary, ignores the effect of close packing and gives a maximal estimate of the effect of orientation of particle elements. Nevertheless, the resultant relations differ little from one another. According to both theoretical models, the spectral coefficient of radiation diffusion D_λ is inversely proportional to $(1 - p)$, the difference between the values of $(1 - p)D_\lambda$ for the ceramics being treated is attributed to the difference in the mean thickness of the particle walls, and the increase in D_λ in the long-wave region is associated with the decrease in the refractive index of alumina.

We will now treat the evolution of the physical pattern of radiation scattering (attenuation) by large hollow spherical particles (which do not absorb radiation) under conditions of hypothetical variation of the concentration of particles in a dispersed system. While the interparticle clearance significantly exceeds their own dimensions, the radiation scattered by neighboring particles reaches the particle being treated in the form of plane waves, which corresponds to the formulation of the scattering problem in the Mie theory. In so doing, a significant part of the particle surface turns out to be lit at glancing angles; as a result, the transport factor of attenuation efficiency Q_{tr} turns out to be relatively low (see curves *1* in Fig. 2). When the particle concentration increases, the clearance between neighboring particles becomes commensurable with their own dimensions, and the lighting conditions change. The pattern approaches that in the case when the elements of the particle surface are random-oriented with respect to incident radiation. Therefore, Q_{tr} increases (from curves *1* towards curves *2* in Fig. 3) even under conditions of independent scattering of radiation by particles. The maximal estimation of the respective variation of the radiation diffusion coefficient D_λ is given by the model of random-oriented plates. When the particles continue to come closer together until they are close-packed in the dispersed system, the dependent scattering of radiation by elements of neighboring particles starts playing a significant part. A qualitative estimate of the associated additional decrease in D_λ may be obtained using the model in which the clearances between particles are represented as relatively fine hollow spherical particles scattering the radiation independently of one another.

CONCLUSIONS

Two approximate theoretical models have been suggested, which explain the strong scattering of visible and infrared radiation in ceramics consisting of hollow microspheres. Models based on the assumption of predominant scattering of radiation in the contact zones of neighboring microspheres differ by the physical interpretation of the increase in scattering. The transport coefficient of attenuation and the spectral coefficient of radiation diffusion are estimated using either an approximation of the results of calculations by the Mie theory or a similar approximation of analytical solution for the scattering of radiation by random-oriented plates.

The results of comparison of the calculated values of the spectral coefficient of radiation diffusion to experimental data for two types of heat-insulating ceramics of hollow spherical particles of alumina confirmed the validity of both suggested models (they produce close results) both for qualitative analysis and for theoretical estimation of the optical properties of materials of the treated type.

The evolution of the physical pattern of scattering of radiation by large hollow spherical particles as a result of variation of the particle concentration in a dispersed system has been discussed. An increase in the particle concentration is first accompanied by the variation of the conditions of lighting of individual elements of the surface of each particle (this is accounted for in the model of random-oriented plates), after which the dependent scattering of radiation by elements of neighboring particles comes to play an important part.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research (grant no. 04-02-16014).

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