## NONSTEADY TRANSFER IN DISPERSE AND HETEROGENEOUS MEDIA

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The Laplace transform is used to obtain a unique equation for describing essentially nonsteady heat or mass transfer in a heterogeneous medium. The equation is used to analyze the heating of a half-space through a plane boundary.

A detailed survey of the methods used to describe nonsteady heat transfer in dense granular media can be found in [1]. The possibilities of the methods are discussed in connection with experimental data on the heating of different types of layers from a flat wall. The methods take two approaches, each of which is largely phenomenological. The first approach involves taking the solutions of the system of equations of heterogeneous transfer with a time-independent interphase heat-transfer coefficient and extending them to the region of heat-transfer time scales in which this system is invalid [1]. The second approach is based on the use of an ordinary parabolic heat-conduction equation [2] with allowance for the additional thermal resistance near the wall. This leads to the formulation of a boundary condition of the third kind for the wall.

Although useful empirical formulas for the heat flux from the wall can be obtained by appropriate selection of the coefficients in the equations for interphase heat flow or the boundary condition, neither method adequately describes the physics of the transport process. In fact, as was shown in [3], the above system can be used only when the transience of the flow is slight. The best evidence of the conditional nature of studies that have employed the second approach is that physically substantiated attempts to refine the thermal resistance (see [4-6], for example) have only worsened the agreement between the theoretical and experimental results. Generalization of methods of the steady-state theory to essentially nonsteady transport processes requires direct analysis of the exchange of the continuous phase with individual elements of the discrete phase, as was proposed in [3]. Calculations of this type were performed in [7, 8] (as well as [9]) for the rows of particles closest to the wall.

To simplify the problem, we propose to ignore the nonuniformity of the temperature distribution over the particle surface and to assume it to be equal to the mean temperature of the continuous phase at the point corresponding to the center of the particle. The validity of these assumptions was discussed in [3]; the assumption of uniformity of temperature at scales on the order of the dimensions of a particle is a necessary condition for the applicability of continuum methods in the description of heat transfer in heterogeneous media. Some ramifications of further generalizations are discussed below.

The equation of convective heat transfer in the continuous phase is written in the standard form

$$\varepsilon d_1 c_1 \left( \frac{\partial}{\partial t} + \mathbf{u} \nabla \right) T_1 = \lambda_* \Delta T_1 - nq. \tag{1}$$

To calculate the heat flux q from the continuous phase to a single particle in the approximation being examined, we have the usual problem of the heat conduction inside a particle with a boundary condition of the first kind. If the particles are spherical, then the solution of the problem, with the initial condition  $\tau(0, r|\mathbf{x}) = T_1(0, \mathbf{x})$  has the form [10]

$$\tau(t, r \mid \mathbf{x}) = T_1^{\circ}(\mathbf{x}) + \frac{2\pi\kappa_2}{ar} \sum_{j=1}^{\infty} (-1)^{j-1} \sin \frac{j\pi r}{a} \int_0^t \exp\left[-\frac{\kappa_2 j^2 \pi^2}{a^2} (t-t')\right] \left[T_1(t', \mathbf{x}) - T_1^{\circ}(\mathbf{x})\right] dt',$$

$$T_1^{\circ}(\mathbf{x}) = T_1(0, \mathbf{x}),$$
(2)

where the argument is a parameter in this case. Applying the Laplace transform to (2) (the

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transformed quantities will be denoted by an asterisk above the quantity), we obtain

$$\tau^* = \frac{T_1^\circ}{p} + \frac{a}{r} \left( T_1^* - \frac{T_1^\circ}{p} \right) \frac{\operatorname{sh} \alpha r'}{\operatorname{sh} \alpha \pi}, \quad r' = \frac{\pi r}{a}, \quad \alpha = \frac{a}{\pi} \sqrt{\frac{p}{\varkappa_2}}.$$
 (3)

We use this result to calculate the transform of the heat flux to a particle:

$$q^* = 4\pi a^2 \lambda_2 \frac{d\tau^*}{dr} \Big|_{r=a} = -4\pi a \lambda_2 \left[ 1 - a \sqrt{\frac{p}{\varkappa_2}} \operatorname{cth}\left(a \sqrt{\frac{p}{\varkappa_2}}\right) \right] \left(T_1^* - \frac{T_1^*}{p}\right). \tag{4}$$

Considering that for a layer of identical spherical particles  $n = 3(1 - \epsilon)/(4\pi a^3)$  and inserting (4) into Eq. (1) after Laplace transformation of the latter, we arrive at the equation

$$\varepsilon d_1 c_1 \left( p T_1^* - T_1^\circ + \mathbf{u} \nabla T_1^* \right) = \lambda_* \Delta T_1^* + \frac{3\lambda_2}{a^2} \left( 1 - \varepsilon \right) \left[ 1 - a \sqrt{\frac{p}{\varkappa_2}} \operatorname{cth} \left( a \sqrt{\frac{p}{\varkappa_2}} \right) \right] \left( T_1^* - \frac{T_1^\circ}{p} \right).$$
(5)

Given the appropriate boundary conditions, the solution of this equation yields the transform of the mean temperature of the continuous phase. The transform of the mean temperature of the disperse phase can be obtained by averaging (3) over the volume of the particle:

$$T_2^* = \frac{T_1^*}{p} + 3\left(T_1^* - \frac{T_1^*}{p}\right) \left[\frac{1}{a} \sqrt{\frac{\varkappa_2}{p}} \operatorname{cth}\left(a \sqrt{\frac{p}{\varkappa_2}}\right) - \frac{\varkappa_2}{a^2 p}\right].$$
(6)

Thus, the description of nonsteady heat transfer has been reduced, first, to the solution of the corresponding boundary-value problem for linear Eq. (5) and, second, to calculation of the originals of the resulting transforms. The first problem is routine, while the second can be solved either by asymptotic methods or by established methods of numerically inverting Laplace transforms [11]. It should be noted that  $T_2(t, x)$  can always be found from the known  $T_1(t, x)$  and can be determined directly by averaging (2) without inverting (6).

For times t  $\gg a^2/\kappa_2$ , the term in the square brackets in (5) can be represented in the form of an expansion in powers of p. Limiting ourselves to the first two terms of expansion, we have

$$1-a\sqrt{\frac{p}{\varkappa_2}} \operatorname{cth}\left(a\sqrt{\frac{p}{\varkappa_2}}\right) \approx -\frac{1}{3}\frac{a^2}{\varkappa_2}p + \frac{1}{45}\frac{a^4}{\varkappa_2^2}p^2.$$

If we use this in (5) and we perform the inverse Laplace transformation, we obtain the elliptic equation

$$\left[\varepsilon d_1 c_1 + (1-\varepsilon) d_2 c_2\right] \frac{\partial T_1}{\partial t} + \varepsilon d_1 c_1 \left(\mathbf{u} \nabla\right) T_1 = \lambda_* \Delta T_1 + \frac{(1-\varepsilon) a^2 d_2 c_2}{15 \varkappa_2} \frac{\partial^2 T_1}{\partial t^2}$$

with the auxiliary condition  $\partial T_1/\partial t = 0$ , t = 0. It should be noted that this equation coincides exactly with that formulated in [3].

Let us examine the features of nonsteady heat transfer, using as an example the unidimensional problem of the heating of a granular layer with stationary phases from a flat wall when the initial temperature of the phases is zero. In this case, Eq. (5) can be written in the form

$$d^{2}T_{1}^{*}/d\xi^{2} = [p' - k(1 - \sqrt{p'} \operatorname{cth} \sqrt{p'})]T_{1}^{*}, \qquad (7)$$

where we have introduced the dimensionless distance  $\xi$ , the time (Fourier number) Fo, and the parameter k:

$$\xi = \sqrt{\frac{\epsilon \lambda_1 \kappa_2}{\lambda_* \kappa_1}} \frac{x}{a}, \quad \text{Fo} = \frac{\kappa_2 t}{a^2}, \quad k = \frac{3(1-\epsilon) d_2 c_2}{\epsilon d_1 c_1}, \tag{8}$$

while p' is the Laplace transform parameter, corresponding to the time Fo (the primes with these quantities will henceforth be omitted).







Fig. 1. Profiles of the dimensionless temperatures of the phases for the first  $T_i/T_w$  (a) and second  $T_i/AQ_w$  (b) boundary-value problems with different log Fo (numbers next to curves); the top and bottom solid curves pertain to the continuous and disperse phases, respectively, while the dashed lines correspond to the solution of the parabolic equation.

Fig. 2. Relaxation of the temperatures of the continuous (1) and disperse (2) phases for the first boundary-value problem at  $\xi = 0.05$ , k = 10 (a) and k = 100 (b); the dashed curves represent the solution of the parabolic equation.

We assign a boundary condition of the third kind on the wall  $\xi = 0$ ; after Laplace transformation, this condition is written in the form:

$$\lambda_* \frac{dT_1^*}{d\xi} \bigg|_{\xi=0} = \frac{1}{R} \left( T_1^* - T_w^* \right).$$
(9)

Solving (7) with condition (9) and the condition that  $T_1^*$  vanish at  $\xi \rightarrow \infty$  (which determines the choice of the origin for the temperature coordinate), we have

$$T_{1}^{*} = C(p) \exp \{-\xi [p + k(\sqrt{p} \operatorname{cth} \sqrt{p} - 1)]^{1/2}\},\$$

$$C(p) = \frac{T_{w}^{*}}{1 + k' [p + k(\sqrt{p} \operatorname{cth} \sqrt{p} - 1)]^{1/2}},\$$

$$k' = b\sqrt{\epsilon \lambda_{1} \varkappa_{2}/\lambda_{*} \varkappa_{1}},$$
(10)

where b is a coefficient with accounts for the irregularity of the arrangement of the particles near the wall. It is introduced into the expression  $R = ba/\lambda_*$  for contact thermal resistance [2].



Fig. 3. Dependence of Nu on Fo for the first boundary-value problem (a) and dependence of the dimensionless wall temperature  $T_w/AQ_w$  on Fo (b) for the second boundary-value problem: 1-8) k = 0, 10, 32, 100, 317, 1000, 3163, 10,000.

Introducidng the Nusselt number (with constant  $T_w$ , i.e.,  $T_w^* = T_w/p$ ):

$$Nu = \frac{AQ_w}{T_w}, \quad Q_w = -\lambda_* \frac{\partial T_1}{\partial x} \bigg|_{x=0}, \quad A = \frac{a}{\lambda_*} \sqrt{\frac{\kappa_1 \lambda_*}{\epsilon \kappa_2 \lambda_1}}, \quad (11)$$

we obtain the following from (10) for its transform

$$Nu^{*} = \frac{1}{\sqrt{p}} \left[ \left( 1 + k \frac{\sqrt{p} \operatorname{cth} \sqrt{p} - 1}{p} \right)^{-1/2} + k' \sqrt{p} \right]^{-1}.$$
 (12)

At  $p \ll 1$  (Fo  $\gg 1$ ), we have Nu\*  $\approx (1 + k/3)^{1/2} p^{-1/2}$ , while at  $p \gg 1$  (Fo  $\ll 1$ ), we have Nu\*  $\approx (k'p)^{-1}$ .

When a boundary condition of the first kind is used for  $T_1^*$  on the wall, the previous expression from (10) is valid and formulas for C(p) and Nu\* are obtained from (10) and (12) with k' = 0, i.e.

$$C(p) = T_w^*, \quad \mathrm{Nu}^* = \frac{1}{\sqrt{p}} \left[ 1 + k \frac{\sqrt{p} \operatorname{cth} \sqrt{p} - 1}{p} \right]^{1/2}.$$
 (13)

The asymptote Nu\* at p  $\ll$  l coincides with the asymptote for the third boundary-value problem, while at p  $\gg$  l we have Nu\*  $\approx$  p  $^{-1/2}.$ 

When a boundary condition of the second kind is assigned on the wall, the quantity  $T_1^*$  is again expressed by the formula in (10) but

$$C(p) = T_{w}^{*} = \frac{AQ_{w}^{*}}{\left[p + k\left(V\overline{p}\operatorname{cth} V\overline{p} - 1\right)\right]^{1/2}}.$$
(14)

At  $p \ll 1$  in the case  $Q_w * = Q_w/p$ , we have  $T_w * \approx AQ_w(1 + k/3)^{-1/2}p^{-3/2}$ , while at  $p \gg 1$  we have  $T_w * \approx AQ_w p^{-3/2}$ . All of the above-examined asymptotes correspond to the solutions of problems for an ordinary parabolic equation, but with different physical parameters.

To find the originals of the resulting solutions, we resorted to numerical inversion of the Laplace transform on the basis of a fourth-order interpolational method [11]. This is presently the most accurate available method. Such an inversion is exact for functions of the form  $p^{-8}P_n(1/p)$ , where  $P_n$  represents a polynomial of up to and including degree seven.

Let us examine the character of the temperature fields in a granular mass with boundary conditions of the first and second kinds. Figure 1a shows profiles of the dimensionless temperatures of the phases  $T_1/T_W$  and  $T_2/T_W$  for the first boundary-value problem at different moments of dimensionless time Fo. Also shown are profiles of mean temperature which follow from the solution of the corresponding problem for a parabolic Fourier equation with the thermophysical parameters characteristic of the granular medium as a whole. Similar information for the second boundary-value problem is shown in Fig. 1b. The curves in Fig. 1 re-





flect the dynamics of heating of the disperse phase and dispersion medium at different distances from the boundary of the body. For sufficiently low values of Fo, the solutions of the parabolic equation appreciably underestimate the temperature of the dispersion medium and appreciably overestimate the temperature of the disperse phase; this result is evidently connected with the fact that the one-temperature formulation of the problem does not account for the finite rate of interphase heat transfer. It is clear that this difference is greater, the greater the parameter k. The curves in Fig. 2 provide information on the relaxation of the phase temperatures to the "parabolic" temperature at different k.

It is of considerable practical interest to examine the dynamics of the change in the heat flow to a granular mass with a boundary condition of the first kind and a wall temperature satisfying a boundary condition of the second kind. Figure 3a shows the dependences of Nu on Fo with constant  $T_w$  and different k changing due to variation of the properties of the particles as the properties of the continuous phase remain constant. At Fo  $\rightarrow$  0, all of these relations lie on a single straight line in logarithmic coordinates, corresponding to Nu =  $(\pi Fo)^{-1/2}$ . When Fo  $\rightarrow \infty$ , they become parallel lines, with Nu =  $[(1 + k/3)/(\pi Fo)]^{1/2}$ . For intermediate values of the Fourier number, there is a region in which the change in Nu with Fo is greatly slowed and, roughly speaking, Nu ~ Fo<sup>-1/4</sup>. The existence of such a region, expanding with an increase in k, has been confirmed by numerous experiments conducted for different types of granular layers [1, 2] and was substantiated theoretically in [3].

Figure 3b shows similar curves illustrating the change in wall temperature with the prescription of a constant heat flow to a granular body. This case is also characterized by a region Fo in which the increase in temperature with Fo is relatively slow. Again, the region expands with an increase in the parameter k.

Figure 4 shows experimental data on the heating of granular layers of glass or slag spheres filled with air, helium, carbon dioxide, or freon. The data was taken from Fig. 2 in [1]. Figure 4a also shows theoretical curves obtained by inverting Eq. (13), which is valid for the first boundary-value problem. With  $k = 10^3$ , Fig. 4b shows the relations Nu(Fo) for the third boundary-value problem. These relations follow from (12) and correspond to different values of the parameter k'. Also shown are the anologous curves for the parabolic heat-conduction equation for a granular layer as a homogeneous heat-conducting medium.

Figure 4 leads to the general conclusion that by allowing for relaxation phenomena connected with interphase heat transfer within the framework of the two-temperature model of disperse medium examined here, we can obtain agreement between experimental and theoretical results at considerably lower values of k' (and thus, lower values of contact resistance) than by means of the traditionally used one-temperature model. However, as for other wellknown experimental results of the same type (see [12-14], for example) the accuracy of the experimental data (Fig. 4) is on the whole high enough so as to permit an unambiguous conclusion regarding the relative significance of the effects connected, first, with relaxation phenomena and, second, with the influence of the contact resistance at the wall on heat flow. To clearly separate these contributions, it will evidently be necessary to conduct special experiments in which the nonuniformity of the layer which develops near the wall due to its impermeability to particles is completely excluded. In principle, such experiments could be conducted by using solid composite materials with a dense-packed disperse phase and phases having different thermophysical properties.

The developed method of reducing problems of heat conduction in disperse media to the solution of a single equation for the Laplace transform of the mean temperature field of the continuous phase is easily generalized to more complicated situations, such as when heat conduction is significant outside the particle as well as inside or when the temperature field is nonuniform. In accordance with the method proposed in [15], if we consider a disperse medium with stationary phases surrounded by a spherical particle of the discrete phase and we regard this medium as a quasiuniform continuum with thermophysical properties dependent on the distance to the particle surface, we have the following problem:

$$p\left[d_{1}c_{1}\varepsilon\left(r\right) + d_{2}c_{2}\left(1 - \varepsilon\left(r\right)\right)\right]\varphi^{*} = \lambda_{*}\nabla\left[f\left(r \mid \varkappa\right)\nabla\varphi^{*}\right] - n\left(r\right)q^{*}, r \ge a,$$

$$p\tau^{*} - T_{1}^{0} = \varkappa_{2}\Delta\tau^{*}, \quad 0 \leqslant r < a; \quad \tau^{*} < \infty; \quad r = 0;$$

$$T_{1}^{*} + \varphi^{*} = \tau^{*}, \ \lambda_{*}n\left[\nabla T_{1}^{*} + f\left(a \mid \varkappa\right)\nabla\varphi^{*}\right] = \lambda_{2}n\nabla\tau^{*}, \quad r = a;$$

$$\varphi^{*} \rightarrow 0, \quad r \rightarrow \infty; \quad \varkappa = \lambda_{2}/\lambda_{1}.$$
(15)

The solution of this problem determines not only the distribution of the transform  $\tau^*$  of temperature inside the particle, but also the deviation  $\Phi^*$  of the temperature transform in its neighborhood from the quantity  $T_1^*$ , which may depend arbitrarily on  $\mathbf{x} + \mathbf{r}$ . Here, is the radius vector of the center of the particle (it is usually sufficient to consider only the first two terms of the Taylor expansion of  $\tau^*$  in powers of r). The form of the functions  $\varepsilon(\mathbf{r})$  (and, thus,  $n(\mathbf{r})$ ) and  $f(\mathbf{r}/\kappa)$  are determined by the packing of the granular system: different models for random packing were examined in [16]. If contact conductivity over the particle skeleton is absent, then  $\lambda_* f(a|\kappa) = \lambda_1$ . Equation (3) corresponds to the solution of problem (15) in the case when  $\Phi^*$  nearly vanishes, while the dependence of  $T_1^*$  on the coordinates is ignored.

In principle, the theory can be generalized to heterogeneous media of a different structure by using a semiempirical method to isolate representative elements of both phases and evaluate nonsteady interphase heat transfer by analyzing heat conduction within and in the neighborhood and such elements. The literature contains examples of similar modeling of heterogeneous media as sets of alternate layers or interacting blocks of more complex geometry consisting of uniform phases.

In conclusion, we note that, in a mathematical sense, formulation of problems on diffusive-convective mass transfer in granular systems is fully analogous to the problem examined above. Its further generalization can be connected with the presence of sources or sinks of a diffusive impurity created by sorption-desorption or chemical reactions. This point is very important for describing and analyzing processes in granular and porous beds of catalysts. Nonsteady phenomena are important in a practical sense, and these phenomena can be studied on the basis the model developed above. Also important to study are processes involving the propagation of moisture in cloddy soil, the filling of nonuniform porous media by a fluid, the establishment of steady-state filtration, and many other relaxation phenomena in filtration flows.

#### NOTATION

A, parameter introduced in (11); *a*, particle radius; b, coefficient accounting for irregularity of particle packing near the wall; c, heat capacity; d, density; Fo, dimensionless time (Fourier number); k, k', dimensionless parameters determined in (8) and (10); Nu, Nusselt number; n, numerical concentration of particles; p, Laplace transform parameter;  $Q_w$ , dimensional heat flux to the granular layer from the wall; q, heat flux to a particle; R, contact thermal resistance; r, radial coordinate in the system connected with the particle; T<sub>i</sub>, mean temperatures of the phases; T<sub>w</sub>, wall temperature; t, time; u, rate of convective transport; x, space coordinate;  $\alpha$ , parameter in (3);  $\varepsilon$ , porosity;  $\kappa_i$ , thermal diffusivity of the materials of the phases;  $\kappa$ , parameter in (15);  $\lambda$ , thermal conductivity;  $\lambda_x$ , effective thermal conductivity;  $\xi$ , dimensionless coordinate;  $\tau$ , temperature inside particle. Indices: 1 and 2 pertain to the continuous and discrete phases, respectively; an asterisk above a quantity denotes its Laplace transform.

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# NON-NEWTONIAN PROPERTIES OF EMULSIONS IN SOLUTIONS OF SURFACE-ACTIVE

# AGENTS

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The adsorption of surface-active agents (surfactants) on channels changes the effective viscosity of an emulsion and gives it non-Newtonian properties.

Establishing the form of rheological equations of state of disperse systems is one of the most important problems in the physical mechanics of mixtures. This problem is far from being resolved even for the simplest systems - suspensions of rigid particles or Newtonian drops in a Newtonian fluid. The situation is even more complicated if physicochemical processes which alter the structure of the flow near the particle are taking place on the surface of a particle or drop. Such phenomena can have a significant effect on the behavior of the mixture as a whole. Meanwhile, the result of this effect is impossible to predict by means of a phenomenological modeling of continuum equations.

Here, we study the rheological properties of emulsions whose drops might adsorb an impurity contained in the dispersion medium. It was shown in [1] that the capillary effects which occur in this case impart non-Newtonian properties to the emulsion even when the disperse phase and the dispersion medium are Newtonian fluids. However, it was assumed in [1] that sorption-desorption processes take place at an infinitely high rate. Below, we consider the finiteness of these processes. At the same time, we correct the errors allowed in [1]. As in [1], we examine limitingly dilute mixtures in which we can ignore any particle interaction. The surface tension of the drops is assumed to be strong enough to ensure that they are spherical in form during the flow process.

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