THERMAL CONDUCTIVITY OF CONSOLIDATED SYSTEMS

G. N. Dul'nev, M. A. Eremeev, and Yu. P. Zarichnyak

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A structural model and method for calculating the effective thermal conductivity of consolidated granular materials are proposed.

Formulation of the Problem. A considerable number of natural and artificial materials of varying origin are known which can be converted by the term "consolidated materials." Among them are sintered, cemented, flocculated, welded, etc. materials. We consider those consolidated materials which are obtained by pressing or sintering of granular systems that are initially in the state of a free charge.

Visual study of such materials has shown that the greater part of their structure is a spatial skeleton of adhering particles. In this case, the pores formed by the skeleton are considerably larger than the pores between the particles (Fig. 1a). Using the rules for transition from a random system to ordered models (Fig. 1b) and from the latter to an elementary cell (Fig. 1c), the subsequent study of heat transport will be carried out within the framework of the elementary cell [1].

We denote by V_T, V_1, V_2, and $V = V_1 + V_2 + V_T$ the volumes of the solid particles, of the pores between the particles in the skeleton, of the large pores in the structure, and the total volume of the system (see Fig. 1c); we introduce the notation m_{2k} for the porosity of the skeleton (first-order structure), m_{22} for the porosity of the second-order structure, and for total porosity of the system, m_2:

$$m_2 = (V_1 + V_2)/V, \quad m_{22} = V_2/V, \quad m_{2к} = V_1/(1 - m_{22})\ V, \tag{1}$$

where $(1 - m_{22})V$ is the volume of the skeleton. It is easy to show that the following relation,

$$m_{22} = 1 - \frac{1 - m_2}{1 - m_{2к}} \tag{2}$$

exists between these parameters.

The effective thermal conductivity λ of a structure with interpenetrating components is related to the effective thermal conductivities λ_k of the skeleton and λ_2 of the components in the large pores by a system of equations [1] which takes the form

$$\frac{\lambda}{\lambda_к} = C_2^2 + \nu_2 (1 - C_2)^2 + \frac{2\nu_2 C_2 (1 - C_2)}{\nu_2 C_2 + (1 - C_2)}, \quad \nu_2 = \frac{\lambda_2}{\lambda_к},$$
$$m_{22} = 2C_2^3 - 3C_2^2 + 1. \tag{3}$$

using new notation.

Thus further analysis is connected with the determination of the porosity m_{22} of the second-order structure and of the values of the coefficients of thermal conductivity λ_2 and λ_k. All these parameters depend on the nature of the deformation of the system and on the state of the free charge of the grains before the formation of the consolidated material.

Analysis of Skeleton Deformation. We consider geometric deformation of the charge during sintering and pressing. The fact that the skeleton sketched in Fig. 1a is not completely broken down is evidence of

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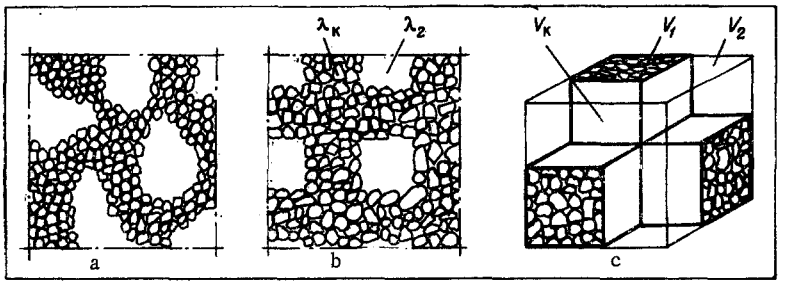

Fig. 1. Transition to an ordered model of granular structure: a) actual structure; b) ordered structure; c) elementary cell.

its stability. In our opinion, the stability can be explained in the following manner: in the initial stages of sintering and pressing the skeleton is partially broken down, and the particles being deformed agglomerate with the agglomeration of the particles, in turn, strengthening the skeleton. In addition, highly porous materials are often prepared by introducing a slightly compressible filler (rubber, for example) into the large pores, which also strengthens the skeleton. We therefore assume that reduction of charge porosity during pressing and sintering occurs both because of spreading of the skeleton (with consequent reduction of the porosity m_{22}) and because of deformation of the particles in the skeleton.

We introduce the notation m_2^0, m_{2k}^0, and m_{22}^0 corresponding to the total porosity, skeleton porosity, and porosity of the second-order structure in the original state of the free charge (before pressing). It is obvious that a relation similar to Eq. (2) exists between these parameters, i.e.,

$$m_{22}^0 = 1 - \frac{1 - m_2^0}{1 - m_{2к}^0}. \tag{4}$$

Analysis of a large amount of data for granular systems allows one to assume the most probable value of the porosity of the skeleton in the state of a free charge with particles from 0.01 to 1 mm in size is $m_{2k}^0 \simeq 0.4$.* We consider heat transfer in the skeleton and find an expression for its effective thermal conductivity λ_k.

There are two possible solutions for this problem: represent the arrangement of the particles within the skeleton in the form of some kind of ordered structure or assume the particles are arranged randomly within the skeleton. The first way, which is geometrically more visualizable, has been realized [2]. It is of interest to select the second way, having used the results of [3] where it was shown that the effective thermal conductivity of a skeleton with randomly arranged granules is equal to the effective thermal conductivity of its volume element, a two-dimensional representation of which is shown in Fig. 2a. It consists of a solid component (portion of a spherical granule) in the form of a cylinder of radius r_3 and height r with a spherical base of radius r and a gaseous or liquid component filling the pore in the form of a cylinder of radius r_4. The relative dimensions of the volume element, $y_3 = r_3/r$ and $y_4^0 = r_4^0/r$, are uniquely related by the expressions in [3] to the porosity m_{2k}^0 and the coordination number N, which is the average number of contacts per particle. Using the new notation for the parameters, we rewrite these expressions:

$$N = (m_{2к}^0 + 3 + \sqrt{m_{2к}^0 - 10m_{2к}^0 + 9})/2m_{2к}^0, \tag{5}$$

$$y = 2\sqrt{N-1}/N, \qquad y_4^0 = y_3(1 - m_{2к}^0)^{-\frac{1}{3}}. \tag{6}$$

In particular, Eqs. (5) and (6) show that for $m_{2k}^0 = 0.4$, the parameters $N = 7.09$, $y_4^0 = 0.825$, and $y_3 = 0.696$.

In the model we represent the deformation of the granular system during pressing or sintering. For simplicity, we make the following assumptions.

1. The coordination number N for the skeleton depends only on the porosity in the initial state and does not change during deformation (N = const).

*It is assumed the original granular system has not been subjected to special treatment such as vibratory shaking, for example.

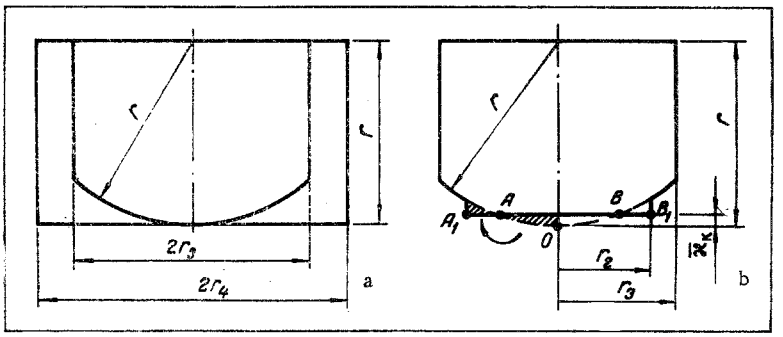

Fig. 2. Model of granular system (a) and particle deformation (b).

2. Linear deformation of a particle in the contact regions (reduction of radius during compression) is the same in all directions and is $\bar{\varkappa}_k$ (Fig. 2b). For the deformed system, the relative radius of the model decreases in comparison with the original value y_4^0,

$$y_4 = r_4/r = y_4^0(1 - \varkappa_k), \quad \varkappa_k = \bar{\varkappa}_k/r. \tag{7}$$

We find a relation between the relative particle deformation $\varkappa_k$ and the relative radius of the actual point of contact $y_2 = r_2/r$, where r_2 is the radius of contact. To do this, we assume that during deformation the particle material is displaced in the region around the contact point in the manner shown in Fig. 2b. Such a scheme is close to reality for plastic deformation of nonbrittle particles. In fact, because of the small size of the original contact points, the specific pressures at the contacts far exceed the creep limit for granular systems even for small pressing forces.

For simplicity, we further assume that the area around the contact is a cylinder of radius r_2 (Fig. 2b). In deformation of particles by an amount $\bar{\varkappa}_k$, the volume of the spherical segment AOB is displaced in the area around the contact as shown by the arrow in Fig. 2b. Furthermore, one must satisfy the condition requiring equality between the volume of a cylinder with a base radius r_2 and a height $\bar{h}_c = r - \sqrt{r^2 - r_2^2}$ and the volume of a spherical segment with the same base radius and height $\bar{h}_c + \bar{\varkappa}_k$

$$V_c = \pi r_2^2 \bar{h}_c = \pi r^3 y_2^2 h_c, \quad V_s = \frac{1}{3}\pi r^3 (h_c + \varkappa_k)(3 - h_c - \varkappa_k), \quad h_c = \bar{h}_c/r. \tag{8}$$

Equating V_c and V_s and making the transformations, we obtain a relation between the relative radius y_2 of the contact point and the relative particle deformation $\varkappa_k$:

$$y_2 = \left\{1 - \left[1 - \frac{3}{4}(1 + \varkappa_k) + \sqrt{\frac{9}{16}(1 + \varkappa_k) - 3\varkappa_k}\right]^2\right\}^{1/2}. \tag{9}$$

It is obvious that the condition $y_{2max} = y_3$ must be satisfied in the case of maximum deformation of the skeleton, i.e., its conversion into a monolith ($\varkappa_k = \varkappa_{kmax}$). A relation between the maximum deformation $\varkappa_{kmax}$ and the initial porosity of the skeleton can be obtained by studying unit volume of the skeleton i.e., a cube of volume V ($1 \times 1 \times 1 = 1$). In this case, the volume occupied by the solid component is $V_1 = V(1 - m_{2k}^0) = (1 - \varkappa_k)^3$, whence

$$\varkappa_{kmax} = 1 - \sqrt[3]{1 - m_{2k}}. \tag{10}$$

However, in the case of maximum deformation, a calculation based on Eq. (9) yields $y_{2max} > y_3$, which cannot actually be. In our opinion, the reason for this discrepancy is that the contact point was assumed circular in the derivation of Eq. (9); in fact, the contact point will have a circular shape only up to certain values $\varkappa_k < \varkappa_{kmax}$ ($\varkappa_k < 10^{-2}$) and with maximum deformation the boundaries of the areas around the contact points must merge with the contact point having a shape that is almost polygonal. In our view, therefore, calculations based on Eq. (9) should give values of y_2 approximating the actual values only for small deformations. To calculate y_2 over the entire range of deformations, $0 \le \varkappa_k \le \varkappa_{kmax}$, we consider it advisable to approximate Eq. (9) in such a way that it satisfies the additional condition $y_{2max} = y_3$. We represent the relation $y_2 = y(\varkappa_k)$ in the form $y_2 = A\sqrt{\varkappa_k}$, and then

$$y_{2max} = y_3 = A\sqrt{\varkappa_{kmax}}. \tag{11}$$

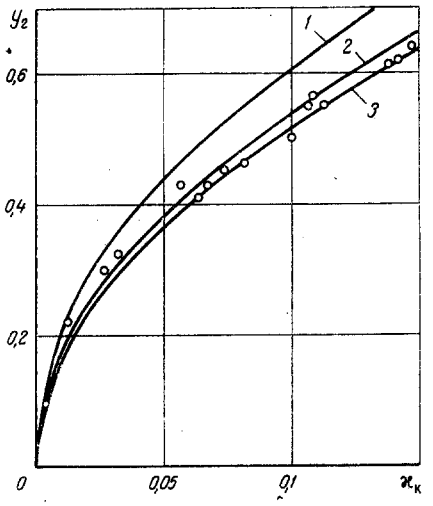

Fig. 3. Dependence of relative radius y_2 of particle contact point on relative compression $\varkappa_k$: 1) calculated from Eq. (9); 2, 3) calculated from Eq. (14) for $m^0_{2k} = 0.4$ and 0.47; points are experimental data ($m^0_{2k} = 0.47$).

Replacing y_3 and $\varkappa_{kmax}$ in Eq. (11) by their values from Eqs. (6) and (10), we obtain an expression for A:

$$A = \frac{2\sqrt{N-1}}{N\sqrt{1-\sqrt[3]{1-m^0_{2к}}}}. \tag{12}$$

Then substituting A from Eq. (12) into Eq. (11), we find y_2:

$$y_2 = \frac{2}{N}\left(\frac{\varkappa_к (N-1)}{1-\sqrt[3]{1-m^0_{2к}}}\right). \tag{13}$$

Equation (13) satisfies the conformity condition $y_{2max} = y_3$. For the overwhelming majority of actual granular systems with a charge porosity $m^0_2 \geq 0.4$, the porosity of the charge skeleton $m^0_{2k} \simeq 0.4$ and the coordination number N = 7.09. In this case, Eq. (13) simplifies to

$$y_2 \simeq 1.755\sqrt{\varkappa_к}. \tag{14}$$

Figure 3 presents calculated relations $y_2 = y(\varkappa_k)$ for $m^0_2 = 0.4$ and 0.47, which were obtained from Eqs. (9) and (14), as well as the results of our measurements of contact points for deformation of a plasticine sphere from six sides (faces of a cube). It is clear from Fig. 3 that the theoretical and experimental relations $y_2 = y(\varkappa_k)$ are in good agreement over the entire range of $\varkappa_k$ when $m^0_2 = 0.47$.

Model of Deformation Process. We introduce the concept of the total relative linear deformation $\varkappa$ of a system, which is equal to the ratio between the changed dimension (after deformation) and the original dimension (Fig. 4), and we find a relation between $\varkappa$ and m_2 for deformation of the system. To do this, we consider a unit volume V_0 (a cube 1 × 1 × 1) in the as yet undeformed system where the fractional volume of the solid component with volume V_1 is $V_1/V_0 = 1-m^0_2$, and the linear deformation of the system is numerically equal to the relative deformation $\varkappa$. After deformation, the volume of the system becomes $V = (1-\varkappa)^3$ and the ratio of the volume V_1 of the solid component to this volume is $1-m_2$, i.e.,

$$V_1/V = \frac{1-m^0_2}{(1-\varkappa)^3} = 1-m_2, \quad \text{or} \quad \varkappa = 1-\sqrt[3]{\frac{1-m^0_2}{1-m_2}}. \tag{15}$$

For maximum deformation ($m_2 = 0$), we have

$$\varkappa_{max} = 1-\sqrt[3]{1-m^0_2}. \tag{16}$$

We assume that the relative deformation $\varkappa_k$ of the particles in the skeleton and the relative deformation $\varkappa - \varkappa_k$ of the skeleton itself (its spread) occur simultaneously so that they are proportional to the deformation $\varkappa$ (Fig. 4). Then the deformation of particles in the skeleton is

$$\varkappa_к = \varkappa\frac{\varkappa_{kmax}}{\varkappa_{max}} = \frac{1-\sqrt[3]{1-m^0_{2к}}}{1-\sqrt[3]{1-m^0_2}}\left(1-\sqrt[3]{\frac{1-m^0_2}{1-m_2}}\right). \tag{17}$$

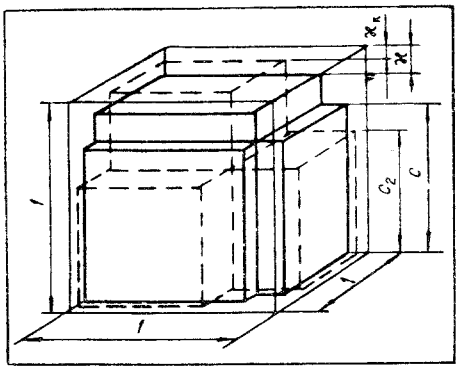

Fig. 4

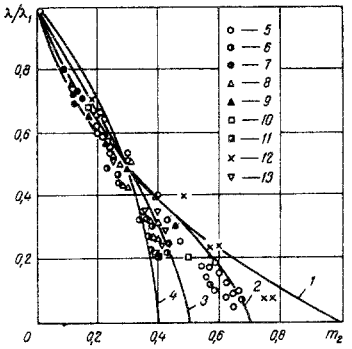

Fig. 5

Fig. 4. Model deformation because of deformation of particles and skeleton.

Fig. 5. Comparison of theoretical and experimental data in the temperature range 20 to 500°C: 1-4) calculated by the proposed method for $m_2^0 = 0.4$, 0.5, 0.7, and 1.0; 5, 6, 7) sintered nickel [6], LITMO (S. Buravoi) and MÉI (V. Alekseev); 8, 10, 11) sintered iron, copper, bronze OF 8.5-0.3, LITMO data (S. Buravoi); 9) sintered iron [4]; 12) graphite [6]; 13) bronze [5].

One of the basic assumptions is contained in this proportionality condition. We recall that the skeleton deformation $\varkappa - \varkappa_k$ was produced by its partial spread, i.e., the particles in it are rearranged and the cross section of the skeleton becomes thicker. Therefore the porosity of the second-order structure (between skeletal blocks) decreases during deformation of the system.

We find a relation between the variation of the porosity m_{22} of the second-order structure and the corresponding deformation $\varkappa_k$ of the particles and $\varkappa$ of the system as a whole. To do this, it is necessary to replace m_2 and m_{2k} in Eq. (2) by the corresponding values of m_2^0 and m_{2k}^0. It can be shown that the relations between m_2 and m_2^0 and between m_{2k} and m_{2k}^0 are determined by expressions similar to Eqs. (10) and (16), i.e.,

$$m_2 = 1 - \frac{1 - m_2^0}{(1 - \varkappa)^3}, \quad m_{2\text{к}} = 1 - \frac{1 - m_{2\text{к}}^0}{(1 - \varkappa_{\text{к}})^3}. \tag{18}$$

Substituting m_2 and m_{2k} from Eq. (18) into Eq. (2), we obtain

$$m_{22} = 1 - \frac{1 - m_2^0}{1 - m_{2\text{к}}^0} \left(\frac{1 - \varkappa_{\text{к}}}{1 - \varkappa} \right)^3. \tag{19}$$

Thermal Conductivity of Skeleton. We determine the effective thermal conductivity of the skeleton of the system (first-order structure). For this, we use the expression from [2] for the calculation of λ for granular systems where the heat flow through the pores between the particles can be neglected ($\lambda_2 = 0$) and the relative height of microroughnesses in the contact region is zero (there are no microroughnesses in consolidated systems):

$$\lambda_{\text{к}} \simeq \lambda_1 (y_2/y_4)^2 \Phi^{-1}. \tag{20}$$

The function Φ takes into account spreading out of the flow from the contact point over the cross section of a particle and, in first approximation, can be assumed to be

$$\Phi \simeq y_2/y_3. \tag{21}$$

Equation (20) then takes the form

$$\lambda_{\text{к}} = \lambda_1 y_2 y_3 y_4^{-2}. \tag{22}$$

Thermal Conductivity of the Consolidated System. Using Eq. (3), one can calculate the effective thermal conductivity of a consolidated granular material as a whole. Usually the thermal conductivities of pores and grains in consolidated materials are considerably different, $\lambda_2/\lambda_1 \ll 10^{-2}$, and the size of an actual contact point ($y_1 = y_2 = r_2/r \geq 10^{-2}$) is considerably greater than the size of the contact point in a free

charge ($y_1, y_2 < 10^{-3}$). In this case, the main heat flow occurs along the skeleton of consolidated particles and Eq. (3) is significantly simplified to

$$\lambda/\lambda_к \simeq C_2^2 . \tag{23}$$

As shown by our analysis, however, the use of the approximate expression (23) leads to some underestimate for calculated values of $\lambda/\lambda_к$ since it was assumed in the derivation of Eqs. (3) and (23) that the lines of heat flow were parallel to the overall heat flow. In fact, the lines of flow, being curved, spread apart at expansion points (nodes) of the skeleton and converge at points of contraction. We take this situation into account.

Where there is considerable difference between the thermal conductivities of the skeleton and of the component in the pores of the second-order structure ($\nu_2 < 0.3$ or $\nu_2 > 3$), one should take into account curvature of the lines of flow in sections filled by the different components. A rigorous consideration of this situation entails mathematical difficulties. If one considers that subdivision of the elementary cell by adiabatic planes [3] yields some underestimate for calculated values of the thermal conductivity of the second-order structure, and subdivision by isothermal planes perpendicular to the heat flow yields an overestimate of the value of the thermal conductivity [7]

$$\frac{\lambda}{\lambda_1} = \left[\frac{1 - C_2}{C_2^2 + \nu_2 (1 - C_2^2)} + \frac{C_2}{C_2 (2 - C_2) + \nu_2 (1 - C_2)^2} \right]^{-1}, \tag{24}$$

one can use as an approximation their arithmetic mean, i.e.,

$$\lambda = 0.5 [\lambda_{(3)} + \lambda_{(24)}]. \tag{25}$$

The approximate expression (25) gives values which differ from the true values (results of a numerical solution) by less than 5% for arbitrary values of ν_2. It is simpler to use Eq. (3) in the region $0.3 < \nu_2 < 3$.

Thus, if we know the coefficients of thermal conductivity λ_1 and λ_2 for the components, the system porosity m_2, and the initial porosities m_2^0 of the charge and $m_{2к}^0$ of the skeleton, successive application of Eqs. (5), (6), (17), (13), (22), (19), (3), (24), and (25) makes possible the calculation of the effective thermal conductivity of a consolidated material.

The relation $\lambda/\lambda_1 = f(m_2)$ for consolidated granular systems with various initial porosities m_2^0 (from 0.7 to 0.4) is shown in Fig. 5 as calculated by the proposed method. Plotted in the same figure are known experimental values of λ/λ_1 for cermets produced by hot pressing of powdered nickel, iron, copper, bronze, and graphite. It is clear from the figure that experimental values of λ/λ_1 fall in the region bounded by calculated relations for initial porosity range 0.7–0.4. Our observations show that the overwhelming majority of metal powders have an initial porosity in the range specified, i.e., from 0.7 to 0.4. We therefore consider the agreement between calculation and experiment demonstrated in Fig. 5 to be satisfactory.

In addition, by looking at the behavior of the calculated relations $\lambda/\lambda_1 = f(m_2)$ for various initial porosities m_2^0, one can conclude that the thermal conductivity of the components and their concentrations in a consolidated granular system do not determine its effective thermal conductivity uniquely. To calculate the effective thermal conductivity, it is necessary to know an additional parameter, the porosity of the granular material in the free-charge state.

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