#### HEAT TRANSFER IN GRANULAR SYSTEMS

## WITH A RANDOM STRUCTURE

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Heat transfer through a granular system with a random structure in the state of free filling is discussed. A model of random structure is proposed and a method for calculating the effective thermal conductivity of a granular system is presented.

It was shown in [1] that there is need for a new model of granular systems satisfying the following requirements: the model must be stable, isotropic, reflect the shape of the particles over the whole range of variation of porosity, and for large values of the porosity consist of relatively large conglomerates of contacting particles. The last requirement is imposed to try to bring the model closer to the structure actually observed in a highly porous granular material.

A granular system is made up of a "carcass" consisting of a random but relatively dense stacking of grains (first order structure) and larger voids penetrating the carcass and forming with it a structure with interpenetrating components (second order structure). It was shown in [1, 2] that the effective thermal conductivity of the whole granular system can be found from the relation

$$\frac{\lambda}{\lambda_c} = C_2^2 + v_{22}(1 - C_2^2) + \frac{2v_{22}C_2(1 - C_2)}{C_2v_{22} + (1 - C_2)}, \quad v_{22} = \frac{\lambda_{22}}{\lambda_c}.$$
(1)

The parameter  $C_2$  is related to the volume concentration  $m_{22}$  of the gaseous component in the second order structure by the equation

$$m_{22} = 2C_2^3 - 3C_2^2 + 1, \tag{2}$$

which is solved in [2]. If the total porosity of the system is  $m_2$  and the porosity of the carcass is  $m_{21}$ , then [1, 2]

$$m_{22} = (m_2 - m_{21}) (1 - m_{21})^{-1}.$$
(3)

Thus the effective thermal conductivity of the whole system can be calculated by Eqs. (1)-(3) for known values of the thermal conductivity of the carcass  $\lambda_c$ , the thermal conductivity of the large voids  $\lambda_{22}$ , and the porosity of the carcass  $m_{21}$ .

The model of the carcass used in [1, 2] to determine these parameters assumed spherical grains with tetragonal packing; i.e. a carcass with a constant, and for spheres of the same size, smallest porosity  $m_{21} = 0.26$ .

It is assumed that the particles are solid and therefore any value of the porosity of a granular system  $m_2 > 0.26$  is reflected in the model by the appearance of second order structure (a network of large voids). With an increase in porosity the fraction occupied by the carcass is decreased, and the spatial network of voids in the second order structure fills all the remaining volume of the system (Fig. 1a).

On the basis of the above, and using certain additional assumptions, expressions were derived for the thermal conductivities  $\lambda_c$  and  $\lambda_{22}$ , and thus the system of equations (1)-(3) was closed. Values of the effective thermal conductivity of granular systems calculated by this method were in satisfactory agreement

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with numerous experimental data. In spite of this there remained a certain dissatisfaction with the very artificial character of the model of a carcass in which the porosity and the nature of the particle stacking are rigidly fixed. We present below another model of the structure of the carcass taking account of the restrictions indicated which as far as possible approximates the actual structure of the carcass. With certain reservations this model can describe the effective thermal conductivity not only of the carcass but of the whole granular system.

Model of a Random Granular System. The analytical determination of the geometrical parameters of the model of a granular system discussed above [1] is based on the conversion from a random to an ordered arrangement of grains in the system. Papers have appeared in which the geometrical parameters of a model similar to that described above [1, 2] were determined directly for a random structure without converting to an ordered model. One of the most interesting descriptions, in our opinion, is given by Kunii and Smith [3] and M. G. Kaganer [4] who determine the geometrical parameters of the model by using the dependence of the average number of contacts N per grain (coordination number) on the porosity of the system  $m_2$ .

Analytical and experimental attempts to determine the form of this relation have had a limited success. Eremeev [5] established a more general form of this relation analytically (Fig. 2)

$$N = \frac{m_{21} + 3 + \sqrt{m_{21}^2 - 10m_{21} + 9}}{2m_{21}} \,. \tag{4}$$

By using (4) the number of contacts between particles can be found for various values of  $m_{21}$  and the form of a model of a granular system can be described. Figure 1b, c, and d show plane cross sections of the structure of granular systems with various porosities.

A change in N corresponds to a change in the nature of the stacking of the particles, and this makes the model more flexible than the one considered above [1] with ordered stacking. This is the most attractive feature of the method. However, two fundamental difficulties arise in the realization of this idea. The first is related to the establishment of the form of the relation  $N = N(m_{21})$ , and the second to the justification of the transition from the whole granular system to a certain element with average geometrical parameters having a thermal conductivity equal to that of the system. It is shown in [2] that the basic idea proposed in [3, 4] was not adequately developed and the authors were forced to recommend calculating the thermal conductivity by semiempirical relations.

We have attempted to describe the heat-transfer process in a random granular structure by using Eq. (4).

Heat Transfer in a Granular System. Suppose the granular system consists of rounded particles having a thermal conductivity higher than that of the material in the pores. Most of the heat flux in such systems occurs through the regions surrounding the contacts. A single contact region is several times smaller than the diameter of a particle [6].

We take advantage of this feature of heat transfer in a granular system and divide the heat flux into individual tubes of flow such that the axis of a tube passes through a particle from contact points at entrance and exit (Fig. 3a).

We assume that the thermal conductivity of each tube is equal to the effective thermal conductivity of the whole granular system (assumption 1). This assumption stems from the supposition that a tube of flow is much longer than the transverse dimensions of the particles; i.e., each tube of flow passes through a large number of particles of nonelongated form randomly filled the volume of the granular system.

We separate a tube into the elements i-1, i, i + 1, ... (Fig. 3a); each element is contained between two planes perpendicular to the heat flux; the c-c plane passes through a contact point and the b-b plane divides the particle in half. The lateral surface of a tube is formed by adiabatic walls.

The thermal resistance of a tube is equal to the sum of the thermal resistances of its elements. These elements can be divided into two types. In the first type there are no through pores of the granular system; elements of the second type contain such pores.

Figure 4 shows a special case of granular stacking — ordered cubical stacking of spheres. It is easy to see that each sphere is in contact with six other spheres at the points K, L, M, O, P, and E; the contacts O, M, P, and E occur for through pores, and the cross sectional area of the through pores is



Fig. 1. Plane sections of the structure of granular systems with various porosities. a, c, d actual structures; b) idealized chain structure. a) m = 0.9; b) 0.9; c) 0.7; d) 0.4.

Fig. 2. Relation of the coordination number N to the porosity  $m_{21}$ . The dashed lines represent calculations with Eq. (4). The coordination numbers for various methods of stacking of spheres are: 1) tetragonal,  $m_2 = 0.26$ ; 2) hexagonal,  $m_2 = 0.4$ ; 3) cubical body-centered,  $m_2 = 0.32$ ; 4) cubical,  $m_2 = 0.47$ ; 5) diamond stacking structure.

shown in Fig. 4a and b as shaded areas. Both types of elements of a tube of flow are shown in Fig. 3b and c. We note that through pores are present only in those tube elements of the second type for which the a-a plane (Fig. 3c) intersects the b-b plane within the limits of the tube.

We consider first heat transfer in tube elements of the first type: the lateral boundaries of such elements are generally curved and heat transfer in them is difficult to describe mathematically. We make assumption 2: the thermal resistance of a curved element is equal to the thermal resistance of the "rectified" element in which the lateral adiabatic surfaces in the particle are parallel to the direction of the total heat flux; in this case the cross sectional area of an element is maintained. Consequently the change in



Fig. 3. Tubes of flow in a granular system; a) tube of flow; b, c) first and second types of elements comprising a tube of flow.



Fig. 4. Determination of the area of through pores: a) through pores in cubical stacking of spheres; b) equivalent volume spheres replacing polyhedra; c) model element of random structure of a granular system with average parameters.

direction of flow lines in the rectified element of a tube of flow occurs only at the a-a plane near the contact region (Fig. 3b).

Real particles can be angular in shape (ground materials) and curved with various radii of curvature. For simplicity we assume (assumption 3) that the regions near contacts in each element of a tube are formed by spheres with an average radius r.

The area  $S_3$  of the spherical surface per contact is equal to the ratio of the total surface of a particle  $S = 4\pi r^2$  to the coordination number N; i.e.,  $S_3 = 4\pi r^2/N$ , and we take the cross section of a tube of flow as a cricle of radius  $r_3$  (Fig. 4c). To find the relation between  $r_3$  and N we use the equations [7]

$$r_3^2 = h_c (2r - h_c), \quad S_3 = 2\pi r h_c,$$

where h<sub>c</sub> is the height of the spherical segment.

After some simple transformations we find that

$$r_{3} = r_{3}/r = 2\sqrt{N - 1/N}.$$
 (5)

The relation  $y_3 = y_3(m_{21})$  can be determined by using Eq. (4). It can be shown that elements of tubes of the first type have identical thermal resistances equal to the thermal resistance of the central element shown in Fig. 4c.

As pointed out above, contact points in elements of the second type are closed to the plane of separation b—b, and the heat flux depends critically on the volume fraction of the through pores. The number of elements of the first and second type in a random granular system are clearly not very different. We take the next step in the schematization of the process by assuming that the effective thermal conductivity of a tube of flow is equal to the thermal conductivity of a certain combination element with average parameters (assumption 4). The element with average parameters is an element of the first type with a prescribed average fraction of through pores per element of the second type.

To determine the average fraction of through pores we construct tangent planes about each particle at its points of contact with neighboring particles. The set of planes forms a spatial polyhedron. It is easy to see that the average porosity of such polyhedra is the same as that of the whole system. In view of the arbitrary shape of the particles we replace the polyhedra by a system of concentric spheres having the same volume (assumption 5) and the same porosity, as shown in Fig. 4b.

The ratio  $R_4/r$ , which is required later, can be found by using Fig. 4b. By definition the volume of the outer sphere  $V_0$  and that of the particle V are related to the porosity  $m_{21}$  by the expression  $(V_0 - V)$   $/V = m_{21}$ , from which

$$R_4/r = (1 - m_{21})^{-\frac{1}{3}}.$$
 (6)

We make a quantitative estimate of the fraction of through pores by comparing the relative areas of the pores. In other words we set up the ratio of the area of the through pores per particle to the average cross sectional area of a particle. We make a further simplification of the model (assumption 6) by representing the through pores by a cylinder with an annular base surrounding the central element of the tube (Fig. 4c). In this case we assume that the relative area of the base of the cylinder is equal to the relative area of the through pores. We write the last condition in the form

$$\pi (R_4^2 - r^2)/\pi r^2 = \pi (r_4^2 - r_3^2)/\pi r_3^2$$
,

which together with Eqs. (4) and (5) leads to

$$y_4 = y_3 \left(1 - m_{21}\right)^{-\frac{1}{3}}.$$
 (7)

Equations (4), (5), and (7) completely characterize the geometrical parameters of an average element (Fig. 4c) which must reflect the characteristics of the whole granular system. Equations (4)-(7) are derived for ideally smooth rigid particles. Figure 4c shows schematically the distortion of the shape of a particle in the contact zone. It is represented by a nominal contact spot of radius  $r_2$  and an actual contact spot of radius  $r_1$ . We do not take account of the dimensions of the contact spot in view of its small size. For freely poured grains  $y_2 = r_2/r = 1-3 \cdot 10^{-3}$ ,  $y_1 = r_1/r_1 = (10^{-3}-10^{-1})y_2$ . Methods of estimating the dimensions of the nominal and actual contact spots are discussed in [1, 2].

The model considered is isotropic and stable, but for large values of the porosity the particles in the granular system are stretched into thin chains as shown in Fig. 1b. We recall that in actual granular systems large conglomerates of particles are formed for large porosities (Fig. 1a). Therefore the range of applicability of the present model is restricted by the following limits of variation of the porosity;  $0 \le m_2 \le 0.4$ . For a porosity  $m_2 > 0.4$  through pores of the second order structure appear in the system. The thermal conductivity of an element with average parameters (Fig. 4c) is equal to the effective thermal conductivity of the whole system if the five basic assumptions made above do not distort the actual process too much.

Let us consider heat transfer through an element with average parameters (Fig. 4c) having isotherms as bases and adiabatics as lateral surfaces. The thermal resistance of the element can be calculated by breaking it down either by isothermal planes perpendicular to the heat flux or by adiabatic surfaces parallel to the heat flux. The breaking down by isothermal planes underestimates the thermal resistance, and breaking it down by adiabatic surfaces overestimates it [2]. A combination of these methods is used in [2]: in this case an additional isotherm is introduced into the model. In the element under consideration with average parameters this isotherm coincides with the plane shaded in Fig. 4c. Using the combination described an expression can be found for the thermal conductivity of the carcass  $\lambda_c$ :

$$\lambda_{\mathbf{c}} = \frac{\lambda_{1}}{y_{4}^{2}} \left\langle \frac{y_{1}^{2}}{0.5h_{\mathbf{r}} + (1 - 0.5h_{\mathbf{r}})\Phi} + \left\{ \frac{D}{y_{3}^{2}} + \left[ \frac{A}{1 - 0.5h_{\mathbf{r}} - B + 0.5h_{\mathbf{r}}/v_{\mathrm{mz}}} + \frac{2v_{\mathrm{sz}}}{1 - v_{\mathrm{sz}}} \left( D - F + \omega \ln \frac{\omega - D}{\omega - F} \right) \right]^{-1} \right\}^{-1} + v_{\mathrm{sp}}E \right\rangle,$$
(8)

where

$$A = y_2^2 - y_1^2; \quad E = y_4^2 - y_3^2; \quad D = \sqrt{1 - y_3^2}; \quad F = \sqrt{1 - y_2^2};$$
  
$$y_1 = \frac{r_1}{r}; \quad y_3 = \frac{r_3}{r}; \quad w = \frac{1}{1 - v_g} (1 - v_g D + B/Hd); \quad \Phi \simeq 0.017 + 0.4y_1$$

Equation (8) can be appreciably simplified in a number of practically important cases.

For example, for freely poured grains under a pressure of a gas filler H > 100 torr with particle sizes d > 0.1 mm the effect of microroughness can be neglected and the grains can be assumed smooth and rigid  $h_r$ ,  $y_1$ ,  $y_2 = 0$ . In this case Eq. (8) takes the form

$$\lambda_{\rm c} \simeq \frac{\lambda_{\rm I}}{y_{\rm 4}^2} \left\{ \frac{D}{y_{\rm 3}^2} + \left[ \frac{2\nu_{\rm g}}{1 - \nu_{\rm g}} \left( D - 1 + w \ln \frac{w - D}{w - 1} \right) \right]^{-1} \right\}^{-1} + \frac{\lambda_{\rm sp} E}{y_{\rm 4}^2} \,. \tag{9}$$

If the thermal conductivity of the material in the pores is appreciably lower than the thermal conductivity of the particles  $\nu_i \ll 1$ , Eq. (8) simplifies to

$$\lambda_{c} \simeq \frac{\lambda_{1}}{y_{4}^{2}} \left\{ \frac{D}{y_{3}^{2}} + \left[ 2v_{g} \left( D - 1 + \ln \frac{1 - D}{v_{g}} \right) \right]^{-1} \right\}^{-1}.$$
 (10)

We estimate the expected difference between the calculated and experimental values of the effective thermal conductivity of granular systems.





The theoretical and experimental values of the effective thermal conductivity can differ because of the schematization of the geometrical parameters of the model, the use of approximate mathematical methods, and the uncertainty or spread in the handbook values of the thermal conductivity of the grains, the coefficient of accommodation etc., or information on the material under study as the average particle size, the microroughness, the pressure of the gas filler etc.

The reliability of the method for determining the geometrical parameters of the model is to a certain extent supported by the fact that the geometrical parameters of the model calculated for ordered and random stacking of spheres turned out to differ by no more than 10%. An approximate mathematical apparatus is used to describe the heat-transfer process because a more rigorous description involves marked mathematical difficulties and becomes too complicated to receive wide acceptance in engineering calculations. However, the problem of estimating the error of approximate methods and developing simpler but sufficiently accurate methods for the mathematical description of heat transfer is still urgent and will remain so.

Preliminary calculations show that the uncertainty (spread) of the input data leads to a difference between the calculated and experimental values of  $\lambda$  of 10-15%.

To test the validity of the new model of a granular system with a random structure the values of the effective thermal conductivity calculated by using Eqs. (1)-(8) were compared with experimental data cited in the literature for eleven different granular systems (80 experimental points). The particles were of steel, quartz, zirconium, magnesium oxide, zirconium oxide, diphenyls, graphite, and sand. The thermal conductivity of the particles varied from 0.2 to 50 W/m deg K. The material in the pores was: air, carbon dioxide, hydrogen, helium, argon, nitrogen, methane, water, ethyl alcohol, benzene, and oil. The thermal conductivity of the material in the pores varied from 0 to 0.6 W/m deg K. The pressure of the gas filler varied from  $10^{-6}$  to  $10^{5}$  torr. The dimensions of the particles varied from  $10^{-3}$  to 10 mm. The porosity of the granular systems varied from 0.3 to 0.96. The temperature range was  $100-2600^{\circ}$ K.

Figure 5 compares the calculated and experimental data on the thermal conductivity of various granular materials with a gas filler at atmospheric pressure. Under these conditions the effect of the contact parameters can be neglected and we can take  $h_r$ ,  $y_1$ ,  $y_2 = 0$  with an error of less than 10%. It is clear that the calculated results agree with experiment over the whole range of variation of the defining parameters. A histogram of the difference between the calculated and experimental values follows closely the form of the normal distribution curve with no systematic deviations. The mean-square deviation of the calculated from experiment for 80 points does not exceed 15%. The difference between the calculated and experimental values increases as the pressure of the filler gas is lowered (H < 10 torr) because of the uncertainty in our values of the dimensions of the actual and nominal contact spots, which practically determine the value of the effective thermal conductivity of the filling under these conditions.

In conclusion we compare the model of granular systems with the ordered stacking of particles in the carcass [2] and the model considered above with a random structure of the carcass.

The two models give practically the same results for the thermal conductivity of granular systems. The calculations do not differ in complexity, and the informational possibilities of the models are identical. In the new model the problem of determining the thermal conductivity of the carcass is solved more exactly; this is, the limitation imposed on the porosity of the carcass ( $m_{21} = 0.26$ ) is removed, which of course does not give any clear preferences. Nevertheless we consider the present investigation expedient, giving rise to a logical internal development of research in this field, since its results enable us to draw conclusions on the suitability of the proposed model and the method for calculating the effective thermal conductivity of granular materials in engineering practice, and also on the absence of basic preference for taking account of the random nature of the structure of actual granular materials.

### NOTATION

$\lambda_1$ and $\lambda_g$	are the thermal conductivities of the particles and the gas filler, W/m • deg K;
$\lambda_{mol}$ and $\lambda_{rad}$	are the molecular and radiative components of the thermal conduc- tivity of the material in the pores, W/m · deg K;
$\mathbf{r}_1$ , $\mathbf{y}_1$ and $\mathbf{r}_2$ , $\mathbf{y}_2$	are the absolute (m) and relative radii of the actual and nominal contact spots;
$\mathbf{r}_3$ , $\mathbf{y}_3$ and $\mathbf{r}_4$ , $\mathbf{y}_4$	are the absolute (m) and relative radii of an element with average parameters;
$h_r$ and $h_r$	are the average absolute (m) and relative depths of the microrough- nesses;
H,	is the pressure in the gas filter, mm Hg;
d = 2r	is the diameter of a particle, m;
$\gamma = c_p/c_v$	is the ratio of the specific heat at constant pressure to that at con- stant volume;
<i>a</i>	is the accommodation coefficient of gas molecules at the surface of particles;
$\lambda_0$	is the molecular mean free path at atmospheric pressure $H_0$ , m;
Pr	is the Prandtl number.

### LITERATURE CITED

- 1. G. N. Dul'nev, Yu. P. Zarichnyak, and B. L. Muratova, Inzh.-Fiz. Zh., 16, No. 6 (1969).
- 2. Yu. P. Zarichnyak, Thermal Conductivity of Granular and Loosely-Bound Materials, Author's Abstract of Candidate's Dissertation [in Russian] LITMO, Leningrad (1970).
- 3. D. Kunii and J. M. Smith, A. I. Ch. E. Journal, 6, No. 1 (1960).
- 4. M. G. Kaganer, "Thermal Insulation in Low-Temperature Technology [in Russian], Mashinostroenie (1966).
- 5. M. A. Eremeev, "On the average number of contacts in granular systems," in: Energy and Material Transfer Processes at Low Temperatures in Vacuo [in Russian], Minsk (1973).
- 6. V. S. Nikitin, Investigation of the Effective Electrical Conductivity of Fillings of a Dispersed Material at Low Temperatures, Author's Abstract of Candidate's Dissertation [in Russian], ITMO AN BSSR, Minsk (1969).

7. I. N. Bronshtein and K. A. Semendyaev, Mathematics Handbook [in Russian], Moscow (1957).