A PREDICTION OF THE BOUNDS ON THE EFFECTIVE THERMAL CONDUCTIVITY OF GRANULAR MATERIALS

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Abstract-Two models are developed to describe the effective thermal conductivity of randomly packed granular systems based on a one dimensional Ohm's Law method. These models are shown to represent upper and lower bounds on the effective conductivity of all normally distributed stochastic mixtures. An empirical factor has been obtained to account for three dimensional thermal effects. Comparisons to experimental data indicate that the modified correlation is generally accurate to within $\pm 20\%$ over a wide range of constituent materials.

NOMENCLATURE

- area of heat transfer; $A_{\rm S}$
- thermal conductivity of continuous phase; k_c
- k_d , thermal conductivity of discontinuous phase;
- k_{en} effective thermal conductivity assuming uniform heat flux;
- effective thermal conductivity assuming k_e , parallel isotherms;
- thermal resistance; R_{\rm}
- time; \bar{t} .
- solid fraction of granular material; $\bar{\varepsilon}$.
- mode of solid fraction distribution; μ ,
- σ . standard deviation of solid fraction distribution.

1. INTRODUCTION

THE PROBLEM of determining the effective thermal conductivity of randomly packed granular materials is one which frequently occurs in engineering practice. As a consequence there has been a considerable effort in the past to develop suitable analytical models. In general these models have proved satisfactory provided that the constituent conductivities are of similar magnitude. For heterogeneous systems which do not fulfil this condition it has been found that available analytical models produce considerable error [l].

The difficulty in developing an adequate model does not arise from any ignorance of the fundamental laws, rather the problem arises in complications in their application. A detailed solution of the conduction problem would require a knowledge of the shape, size, location and conductivity of each particle in the system together with the interaction between particles. Such knowledge is difficult to represent for randomly packed systems. To overcome these difficulties, investigators have tended to make a series of simplifying assumptions. The two basic approaches may generally be classified as follows :

1. Fourier's Law models-These models utilize an idealized geometry for which the temperature field may be solved. Knowing the temperature field the effective thermal conductivity may be solved using the Fourier-Biot Law. The idealized geometry will differ significantly from the geometry of randomly packed structures. The formulations so developed are extrapolated to randomly packed systems and differ primarily in the extrapolation technique.

2. Ohm's' 'Law models-Simplified repetitive geometries are assumed to be representative of the randomly packed system. The problem is further simplified by assuming one dimensional heat transfer. An equivalent electrical network is then developed and the thermal conductivity of the system is obtained from the thermal-electrical analogy.

2. OHM'S LAW MODELS

The Fourier's Law models received considerable attention during the early period of model development for heterogeneous systems. More recently attention has centered more toward the Ohm's Law models. Actually there exist only a very limited number of configurations for which closed form solutions exist for the Fourier equation. Once these solutions were exhausted researchers were forced toward development of the Ohm's Law models in hopes of developing a more general solution.

A simple Ohm's Law model is shown in Fig. l(a). Here a heterogeneous material with solid volume fraction $\tilde{\varepsilon}$, is represented as a series of idealized cubic particles arranged in a square array. The uniform spacing between particles is maintained such that the idealized system retains the proper volume fractions. A typical unit cell is shown in the figure. It is assumed that the effective thermal conductivity may be determined by considering the equivalent electrical resistances in parallel and in series and by applying Ohm's Law. In taking such an approach it is necessary to assume one dimensional heat transfer. Two options occur here: It may be assumed that the heat flux is uniform in the direction of heat transfer or that the isotherms are straight and parallel. These two assumptions lead respectively to the equivalent electrical networks shown in Figs. l(b) and (c).

It should be noted that the two networks result in dif-

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FIG. 1. Cubic lattice representation of granular material.

ferent effective conductivities. Both are approximations to the exact solution and differ because of the simplifying assumptions. It is important to note that both solutions form bounds to the real solution to the proposed model. This is more clearly seen when one considers a nodal point representation of the temperature field in the proposed system. If the resistances normal to the direction of heat flow are assumed to be very large, the heat flow will remain uniform in the direction of the overall temperature gradient. Conversely, if the lateral resistances are taken as zero the potential will be equal in each lateral plane. Therefore, the assumption of linear and parallel heat flux lines is equivalent i to the assumption of infinite lateral resistances; the assumption of parallel isotherms is equivalent to that of zero lateral resistance. The actual resistance will, of course, fall somewhere between zero and infinity so that these two cases form the bounding conditions. Whether the two solutions represent the bounding conditions for the physical system depends upon the suitability of the proposed model. It can therefore be (c) (c) (d) stated that the limitations placed on available Ohm's Law models may be attributed to two principle causes: FIG. 2. Equivalent geometries for uniform heat flux.

- 1. Unrealistic Geometrical Assumptions Most common Ohm's Law models utilize highly idealized The channels behave as resistances in parallel. The particle shapes (usually parallelopipeds) and un- overall resistance is then given as: realistic particle arrays (usually cubic). Both assumptions deviate significantly from the physical situation found in randomly packed granular materials frequently found in nature.
- 2. Unrealistic Heat Flow Assumptions-The assumption of one dimensional heat flow, which is almost universally to be found in Ohm's Law models, is not justified for systems with constituents of widely differing thermal conductivities.

Any model development which might serve to eliminate or reduce differences between the analytical model and actual granular systems should also serve to extend the range of applicability of the proposed correlation.

3. THEORETICAL DEVELOPMENT

Recognizing the two principal causes of failure for If the area of the channels is allowed to approach zero existing correlations, it has been decided to approach as the number of channels approaches infinity the

them systematically, solving first the geometrical relationships. In this case fully stochastic arrangements will be used.

Consider a typical unit cell of the heterogeneous system shown in Fig. 2(a). Divide the unit cell into uniform sized channels by passing both vertical and horizontal planes through the element. These planes are to be oriented parallel to the direction of heat flux and are to be equi-spaced. If the channels arc sufficiently small compared to the dimensions of the solid particles, they will appear as consisting of sections of the continuous and discontinuous phases placed in series. Assuming a uniform heat flux in each channel, $K_{\rm g}(1-\epsilon^{\frac{2}{3}})$ the order of the series resistances does not influence lar material, the overall resistance. Consequently, the two components may be separated as shown in Fig. 2(b). The resistance of the channel is then given by

$$
R_i = \frac{\varepsilon_i}{k_d \Delta A} + \frac{(1 - \varepsilon_i)}{k_c \Delta A}.
$$
 (1)

$$
\frac{1}{R_e} = \frac{1}{R_1} + \frac{1}{R_2} + \dots + \frac{1}{R_n}.
$$
 (2)

The order of these channels and their shape may be altered so long as the individual channel resistances remain unchanged. They may then be distorted to a unit width by changing the vertical dimension while retaining the unit length. They are then arranged in order of decreasing solid fraction. This geometry is shown in Fig. 2(c). The effective thermal conductivity for the specified element is

$$
k_{e_0} = \sum_{i=1} \frac{k_c k_d \Delta A_i}{k_c \varepsilon_i + k_d (1 - \varepsilon_i)}
$$
(3)

as the number of channels approaches infinity the

summation in equation (3) may be replaced by an integral

$$
k_{e_0} = \int_A \frac{k_c k_d \, \mathrm{d}A}{k_c \, \varepsilon + k_d (1 - \varepsilon)} \tag{4}
$$

$$
k_{e_0} = \int_0^1 \int_0^1 \frac{k_c k_d}{k_d + \varepsilon (k_c - k_d)} dx dy
$$
 (5)

$$
= \int_0^1 \frac{k_c k_d}{k_d + \varepsilon (k_c - k_d)} dx
$$
 (6)

since the integrand is constant in the horizontal direction.The corresponding geometry is shown in Fig. 2(d).

B. Parallel *isotherms*

Consider a unit cube of the heterogeneous system shown in Fig. 3(a). The system is to be divided into a series of fine lamina oriented normal to the direction of heat flow as shown. These elements are chosen to be sufficiently thin that the cross sectional area of the solid particles are essentially constant throughout its width. Assuming parallel isotherms within the unit cube, the discontinuous (solid) and continuous phases will act as resistances in parallel within each lamina.

FIG. 3. Equivalent geometries for linear isotherms.

The order of the parallel resistances may be changed arbitrarily without affecting the overall resistance of the parallel circuit. Thus the resistances of the solid elements may be grouped together in each lamina as shown in Fig. 3(b). A simple calculation then shows that the equivalent resistance is given by the equation:

$$
ke_{x} = \frac{1}{\sum_{m=1}^{L} \frac{\Delta x}{k_c + (k_d - k_c)\epsilon_m}}.
$$
 (7)

Where the solid fraction in the mth lamina is given by ε_m and the width of each lamina is Δx .

Note the laminae act as resistances in series. Again the order of the resistances does not affect the overall thermal resistance so that the elements may be rearranged in order of decreasing solid fraction as shown in Fig. 3. If the width of the laminae is allowed to approach zero then the effective thermal conductivity for Fig. 3(d) becomes :

$$
ke_{\infty} = \frac{1}{\int_0^1 \frac{dx}{k_c + (k_d - k_c)\varepsilon}}.
$$
 (8)

C. Evaluation of the solid fraction, ε

Both equations for zero and infinite lateral conductivity, (6) and (S), require a knowledge of the functional relation between the solid area fraction and position to solve the integral. The technique for obtaining such a relation was developed by Tsao [2]. Consider the arbitrary material distribution shown in Fig. 2(c). The vertical position of each lamina is determined by its solid fraction. The portion of the elements below a particular element is then equal to the portion of elements having larger solid fractions.

$$
x = P(\varepsilon_i > \varepsilon) \tag{9}
$$

$$
x = \int_{\varepsilon}^{1} f(\phi) d\phi.
$$
 (10)

Differentiating this equation,

$$
dx = -f(\varepsilon) d\varepsilon. \tag{11}
$$

This relationship may be substituted into equations (6) and (8) for the thermal conductivity with uniform heat flux and parallel isotherms. The limits of integration must be changed accordingly.

$$
k_{e_0} = \int_1^0 \frac{k_e k_d f(\varepsilon)}{k_d + \varepsilon (k_c - k_d)} d\varepsilon \tag{12}
$$

$$
\frac{1}{k_{e_{\epsilon}}} = \int_{1}^{0} \frac{f(\epsilon)}{k_{c} + \epsilon(k_{d} - k_{c})} d\epsilon.
$$
 (13)

Equations (12) and (13) are entirely general in that no assumptions have yet been made regarding the particle shape or size distribution. The effects of these parameters on the solid area fraction were studied by Debbas and Rumph [3] and Haughey and Beveridge [4]. These sources found experimentally that the distribution of the solid area fraction is Gaussian for most packings. A notable exception occurs after prolonged vibration of a sample. This packing produces large regions of ordered distribution and strong anisotropic effects. Similarly large particle size variations tend to allow sifting of smaller particles into the lower regions of a given sample. This produces a definite bulk porosity gradient in the vertical direction. In such cases the radial distribution remains normal. Neglecting all such non-normal distributions the frequency distribution may be taken as Gaussian

$$
f(t) = \frac{e^{-\frac{1}{2}t^2}}{\int_a^b e^{-\frac{1}{2}t^2} dt}.
$$
 (14)

The integral in the denominator serves to normalize the truncated Gaussian distribution. By replacing the standardized random variable equation (14) may be written in the form :

$$
f(\varepsilon) = \frac{+e^{-\frac{1}{2} \left(\frac{\varepsilon-\mu}{\sigma}\right)^2}}{\frac{\sigma \sqrt{\pi}}{\sqrt{2}} \left[\text{erf}\left(\frac{1-\mu}{\sigma \sqrt{2}}\right) - \text{erf}\left(\frac{0-\mu}{\sigma \sqrt{2}}\right) \right]},
$$
 (15)

The mode of the distribution, μ , and the standard deviation of the solid area fraction, σ , remain to be evaluated. The mode may be defined implicitly in terms of the standard deviation from the basic geometry of the mixture. The total solid volume is equal to the sum of the elemental solid volumes.

$$
\bar{\mathbf{z}} = \int_0^1 \varepsilon \, \mathrm{d}\mathbf{x}.\tag{16}
$$

Using the results of equation (11) this expression may be written:

$$
\bar{\varepsilon} = \int_{1}^{\infty} -\varepsilon f(\varepsilon) \, \mathrm{d}x. \tag{17}
$$

Determination of an exact expression for the standard deviation is difficult. Strange [5] reports that the standard deviation is given by the relation.

$$
\sigma = \frac{\text{constant}}{\sqrt{M}}\tag{18}
$$

where M is a measure of the sample size.

For the correlation to be meaningful there must exist a minimum size sample such that for all larger samples the effective thermal conductivity is constant. This is in fact the case as shown in Fig. 4. Here σ is varied in the equations for the effective thermal conductivity

FIG. 4. Effect of standard deviation in area fraction upon effective conductivity.

using uniform heat flux and parallel isotherms respectively. It may also be noted that the calculational results bracket the range of experimental data. This is, of course, the expected trend since the assumption of uniform heat flux and parallel isotherms correspond to those of zero and infinite lateral conductivity respectively.

4. **DEVELOPMENT OF AK EFFECTIVE CONDUCTIVITY CORRELATION**

Equations (7) and (8) have been obtained to predict the effective thermal conductivity oftwo phase mixtures as a function of the constituent conductivities and the volume fractions. The distribution of the two phases is found by taking σ as any sufficiently large number, σ > 100, and solving equations (15) and (17) implicitly for the corresponding value of μ . The resulting thermal conductivity expressions are considered to be geometrically realistic for most random packings of spherical or semispherical particles but are based alternately on the assumptions of zero or infinite lateral conductivity.

Since the lateral conductivity of the mixture will fall between these limits the two equations form a set of bounding limits for the physical case. Numerous authors have proposed correlations which effectively imply that one of these two assumptions is sufficiently close to the physical system that it may be used in obtaining an effective conductivity expression. In spite of arguments presented by proponents of both methods, it does not appear possible to select, *a priori*, a correlation which is more consistent with the physical system. The preferred method would then be to compare both correlations to experimental results and to make a selection based on the demonstrated results.

Such a comparison has been made and the results are shown graphically in Figs. $5-7$. These figures represent randomly packed granular systems with porosities of 0.31. 0.43 and 0.58 respectively. The

FIG. 5. Comparison of bounding conductivities with experimental data for a porosity of 0.31.

selected values represent a range of packing fractions so that they may be considered as representative of all random packings. The non-dimensionalized effective conductivity is given as a function of the constituent conductivity ratios, k_c/k_d . The experimental values are taken from data listed in Table 1. As shown in the figures virtually all data falls between the two limiting curves. For a constituent conductivity ratio, $k_d/k_c < 10$ either bounding equation gives reasonably accurate results. For $k_d/k_c > 100$ both correlations deviate significantly from the experimental data. This indicates the reason why previous Ohm's Law models have generally not been successful at higher constituent conductivity ratios. No doubt some of the discrepancy has been countered by frequently assuming a somewhat distorted array. Uniform heat flux models have been developed such that an inordinate amount of the higher conductivity material is arranged in series with itself.

	Conductivity, (kcal/m h K) \times 100							
Case	Fluid phase	Solid phase	Experiment	Uniform heat flux	Parallel isotherms	(k_d/k_c)	$1-\tilde{\varepsilon}$	Ref.
$\mathbf{1}$	Air	Calcite	21.4	12.1	64.8	128.6	0.493	[6]
2	2.41 Air	310 Steel	22.4	16.1	261.1	684.6	0.489	$[7]$
3	2.41 Helium	1650 Steel	75.5	60.5	341.2	138.1	0.489	$[7]$
4	11.95 Glycerin	1650 Steel	246.0	170.6	456.4	36.3	0.489	$[7]$
5	45.4 Water	1650 Steel	272.0	187.4	471.9	32.0	0.489	$[7]$
6	51.6 CO ₂	1650 Basalt	1.015	0.0047	2.26	134.2	0.470	$^{[8]}$
7	0.080 EtOH	k0,8 Calcite	63.6	49.9	101.0	19.7	0.465	$[9]$
$\,8\,$	15.7 Air	310 Calcite	25.0		65.7			
	2.41	310		11.9		128.6	0.458	[6]
9	Air 2.13	Calcite 310	17.5	10.6	65.7	145.5	0.454	$[9]$
10	EtOH 15.7	Calcite 310	63.5	48.7	10.3	19.7	0.454	$[9]$
11	Water 50.5	Calcite 310	127.0	103.8	154.3	6.1	0.453	$[10]$
12	Air 2.12	Calcite 310	19.0	10.7	64.7	146.2	0.451	$[10]$
13	Air	Calcite	17.2	10.6	65.6	145.5	0.451	$[10]$
14	2.13 Air	310 Lead	30.4	16.1	455.0	1260.7	0.450	[6]
15	2.34 Water	2950 Calcite	118.0	104.4	154.3	6.1	0.447	$[10]$
16	50.8 Air	310 Quartz	36.5	14.0	170.1	398.7	0.440	$[9]$
17	2.38 Air	950 Lead	23.9	15.4	475.7	1260.7	0.439	[6]
18	2.34 Water	2950 Silica	216.0	154.3	356,0	17.9	0.439	$[7]$
19	54.5 Air	973.9 Quartz	26.8	12.7	176.3	420.0	0.438	$[11]$
20	2.25 Air	945 Coal	11.8	6.2	13.6	16.0	0.437	$[11]$
21	2.25 Hydrogen	36 Coal	25.3	21.8	27,7	2.2	0.437	$[11]$
22	16.6 Air	36 Silica	21.89	13.0	181.7	421.9353	0.437	$[7]$
23	2.308 Air	973.9 Steel	34.1	15.8	604.7	1645.2996	0.435	
	2.34	3850						[6]
24	EtOH 29.34	Silica 973.9	144.7	98.4	302,0	33.198	0.434	$^{[7]}$
25	Air 2.34	Lead 2950	34.4	15.4	475.7	1260.6841	0.433	[6]
26	Water 54.5	Silica 973.9	244.9	154.3	356.1	17.8689	0.431	$[7]$
27	IC8 12.29	Glass 93.96	35.14	26.5	44.6	7.6485	0.431	$^{[7]}$
28	Oil 15.4	Lead 2410	81.5	73.1	533.3	156.4935	0.430	$[10]$
29	Water	Silica	224.9	154.3	356.1	17.8689	0.430	$[7]$
30	54.5 Water	973.9 Silica	217.4	154.3	356.1	17.8689	0.430	$[7]$
31	54.5 Water	973.9 Silica	218.9	154.3	356.1	17.8689	0.430	$[7]$
32	54.5 Water	973.9 Silica	218.9	154.3	356.1	17.8689	0.430	$^{[7]}$
33	54.5 H ₂	973.9 SiC	91.0	65.0	371.3	104.3771	0.429	$[12]$
	14.85	1550						

Table 1. Comparison of bounding conductivities to experimental conductivities

Table 1 Continued

Conductivity, (keal in h K) \times 100

96.8

Thus the calculated equations are raised above the lower bounding curve. Similarly for parallel isotherm models an excessive amount of the higher conductivity material can be placed in parallel with itself and in series with the low conductivity phase. This distortion tends to result in a lower calculated conductivity than that of the upper bound.

The approach to be recommended here is to avoid all distortions of the granular system geometry. Recognizing that some error is introduced by the one dimensional heat transfer assumptions a correction factor is to be applied to one of the bounding equations. Such acorrection factor must at the present time be obtained empirically. Selecting the upper bound equation an iso-

FIG. 6. Comparison of bounding conductivities with experi-
mental data for a porosity of 0.43.
mental data for a porosity of 0.58. mental data for a porosity of 0.58.

thermal distortion factor, \bar{F}_{∞} , may be introduced such that:

$$
\overline{F}_{\alpha} = \frac{\overline{k}}{\overline{k}_{e\alpha}}.
$$
 (19)

It is assumed that the isothermal distortion factor may be expressed as a polynomial function of the constituent conductivity ratio k_d/k_c and the solid fraction $\bar{\varepsilon}$. Using data from Table 1 values of \bar{F}_{∞} have been correlated using a least squares technique. The resulting correlation was found to be:

$$
\ln F_{\infty} = -0.1439 - 0.72359 \ln(k_d/k_c) + 0.020114 [\ln(k_d/k_c)]^2 + 3.0260 \tilde{\epsilon}.
$$
 (20)

FIG. 8. Comparison of experimental and calculated results.

FIG. 9. Comparison of experimental results with calculated conductivity for the Schumann and Voss equation.

A comparison between predicted and calculated data from Table 1 is shown in Fig. 8. It should be noted that the correlation will fit virtually all of the data points within $\pm 30\%$. The average error is calculated to be 21% . This range of accuracy appears quite good in consideration of the variety of sources from which data was selected and the wide range of particle sizes and shapes included. Moreover it was necessary to assume certain constituent conductivities in that their exact composition was not always given by researchers.

FIG. 10. Comparison ofexperimental results with calculated conductivity for the Meredith and Tobias equation.

FIG. 11. Comparison ofexperimental results with calculated conductivity for the Bruggeman equation.

For comparative purposes four well known correlations developed by Schumann and Voss $|11|$, Meredith and Tobias [18], Bruggeman [19] and Lord Rayleigh [20] have been evaluated. Calculational results are illustrated in Figs. 9-12 respectively. The average error for these correlations is calculated to be 24.9, 31.6, 26.9 and 33.3% respectively. The new correlation is seen to reduce this average error significantly. The authors attribute this improvement to the assumed geometrical form.

FIG. 12. Comparison of experimental results with calculated conductivity for the Lord Rayleigh equation.

In conclusion, two bounding equations to the effective thermal conductivity of granular materials have been developed. These equations indicate a limit to the usefulness of one dimensional models and hence to the Ohm's Law approach in evaluating such systems without empirical correlation. The model which has been proposed for evaluating the thermal conductivity of granular systems offers the unique advantage of being based on a geometrically realistic geometry. While distortion of the heat flux lines and isotherms can not yet be adequately handled by analytical development they are considered by means of an empirical curve fit. Thus both of the major factors affecting errors in simplified models have been avoided in this development.

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ESTIMATION DES LIAISONS POUR LE CALCUL DE LA CONDUCTIVITE THERMIQUE EFFECTIVE DES MATERIAUX GRANULEUX

Résumé – Pour décrire la conductivité thermique effective de systèmes de grains agglomérés au hasard, on développe deux modèles basés sur la méthode de la loi d'Ohm monodimensionnelle. Ces modèles représentent les liaisons supérieures et inférieures pour tous les mélanges stochastiques normalement distribués. On obtient un facteur empirique pour tenir compte des effets thermiques tridimensionnels. Des comparaisons avec les résultats expérimentaux indiquent que la corrélation modifiée est généralement correcte à $\pm 20\%$ pour une large gamme de matériaux constitutifs.

BESTIMMUNG DER GRENZWERTE DER EFFEKTIVEN WARMELEITFAHIGKEIT GRANULIERTER MATERIALIEN

Zusammenfassung-Zur Beschreibung der effektiven Wärmeleitfähigkeit von Granulatsystemen willkürlicher Packung werden zwei auf dem eindimensionalen Ohmschen Gesetz basierende Modelle entwickelt. Diese Modelle geben die obere und untere Grenze der effektiven Wärmeleitfähigkeit für eine normal verteilte stochastische Mischung wieder. Mit Hilfe eines empirischen Faktors werden dreidimensionale Wärmeleiteffekte erfaßt. Ein Vergleich mit Meßwerten zeigt, daß die modifizierte Beziehung für eine Vielzahl von Materialien uauf $\pm 20\%$ genau stimmt.

РАСЧЕТ ГРАНИЦ ЭФФЕКТИВНОЙ УДЕЛЬНОЙ ТЕПЛОПРОВОДНОСТИ ЗЕРНИСТЫХ МАТЕРИАЛОВ

Аннотация — Разработаны две модели для описания эффективной удельной теплопроводности дисперсных систем с беспорядочной укладкой частиц на основании одномерного метода закона Ома. Показано, как эти модели определяют верхнюю и нижнюю границы эффективной удельной теплопроводности всех стохастических смесей с нормальным распределением. Получен эмпирический коэффициент для учета трехмерных тепловых эффектов. Сравнение с экспериментальными данными показывает, что модифицированная корреляция имеет точность около +20% для широкого диапазона компонентов смеси.