

$i, j, k$  = any cartesian coordinate

### Superscript

— = arithmetic average

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# THERMAL CONDUCTIVITY OF HETEROGENEOUS TWO-COMPONENT SYSTEMS

R. L. HAMILTON, *Celanese Corp. of America, Summit, N. J.*  
O. K. CROSSER, *University of Oklahoma, Norman, Okla.*

The influence of included particle shape, composition, and pure component conductivity upon the thermal conductivity of heterogeneous two-component mixtures consisting of a continuous and a discontinuous phase was studied. Thermal conductivities were measured for mixtures of balsa wood and aluminum particles as several shapes in rubber at selected compositions. Thermal conductivities for widely different heterogeneous two-component systems can be calculated accurately using an equation in which the empirical shape factor  $n$  depends on thermal conductivities of the phases and the included particle shape. The influence of particle shape on  $n$  can be accounted for using the sphericity of the particle.

**A**LARGE AMOUNT OF RESEARCH has been concerned with the transfer of heat through heterogeneous systems. These systems take many forms: packed beds and composite insulations (17), foamed metals or glasses (6, 13), emulsions (10), mixtures of solids in gases (12) or liquids (17), gels (9), alloys (1), ceramics (15), and many others. These studies usually seek to determine the thermal conductivity of such a system or to measure the additional contribution to the total heat transfer offered by convection or radiation. In either case, it is important to establish accurately the amount of conduction heat transfer which occurs. The thermal conductivity of any system describes the heat transferred only by the conduction process. It was the purpose of this research to develop procedures for calculating or correlating the thermal conductivity of two-component heterogeneous mixtures as a function of the conductivity of the pure materials, the composition of the mixture, and the manner in which the pure materials were distributed throughout the mixture.

### Theoretical

Most of the studies on conductivity of heterogeneous mixtures have discussed electrical conductivity; Brown (3) and de Vries (17) give several references. Even though these discussions apply equally well to thermal conductivity (9, 17, 18), a new description of the theory seems desirable. The usual model of a heterogeneous system consists of a continuous phase (phase 1) with a discontinuous phase (phase 2) dispersed

within the continuous phase as particles of various shapes in either regular or irregular array. On this basis, the definition for the mixture thermal conductivity given by Hamilton (8) is:

$$K = \frac{K_1 V_1 (dT/dx)_1 + K_2 V_2 (dT/dx)_2}{V_1 (dT/dx)_1 + V_2 (dT/dx)_2} \quad (1)$$

where  $(dT/dx)_1$  and  $(dT/dx)_2$  are the over-all average temperature gradients in the two phases. The advantage of this definition lies in the fact that an average gradient ratio can be determined from the theoretical work of Maxwell (14) and Fricke (7) to be of the form:

$$\frac{(dT/dx)_2}{(dT/dx)_1} = \frac{nK_1}{K_2 + (n-1)K_1} \quad (2)$$

where  $n$  depends on the shape of the dispersed particles and upon the ratio of the conductivities of the two phases. From solutions of Laplace's equation in spherical coordinates (14), Equation 2 becomes:

$$\frac{(dT/dx)_2}{(dT/dx)_1} = \frac{3K_1}{K_2 + 2K_1} \quad (3)$$

for spherical particles. Maxwell's equation can be obtained by substituting Equation 3 into Equation 1. Thus, when the particles of the dispersed phase are spheres, the theoretical result shows that  $n$  depends neither on the size of the particles nor upon the relative size of the conductivities of the two phases. This suggests that other particle shapes should be

referred to the case of spheres. Equation 2 can be used with  $n$  as an empirical constant which must be determined experimentally for mixtures containing particles of arbitrary shapes. Substituting Equation 2 into Equation 1 gives:

$$K = K_1 \left[ \frac{K_2 + (n-1)K_1 - (n-1)V_2(K_1 - K_2)}{K_2 + (n-1)K_1 + V_2(K_1 - K_2)} \right] \quad (4)$$

Fricke (7) examined theoretically the conductivity resulting when the particles are ellipsoids. In terms of his "shape factor" which he calls  $X$ ,  $n$  is  $1 + X$ . Fricke's results show that  $n$  is a function of  $K_1$ ,  $K_2$ , and the oblateness or prolateness of the ellipsoids and indicate that  $n$  will not depend strongly on particle shape unless the conductivity of the discontinuous phase particles is at least a factor of 100 larger than the conductivity of the continuous phase. At this high conductivity ratio, nonspherical shapes (all other circumstances being the same) will increase the conductivity above that for spheres.

Solutions in closed form are not feasible for other than ellipsoidal particles (with spheres a special case) because of the difficulty in solving Laplace's equation for arbitrary boundary conditions and geometry. Equation 4 is used, and the object of this work is to determine, from previous as well as present experiments, the factors which affect  $n$  and, hence, to determine the applicability of Equation 4.

### Previous Experimental Results

Data on mixtures whose components have nearly equal conductivities usually agree with Equation 4 for  $n = 3$  even when the particles are not spherical (9, 10, 17). In these mixtures the possibility of convection or radiation was not eliminated, however, and it was necessary in this study to examine mixtures in which conduction only would occur.

Data have also been presented on mixtures whose components have widely different conductivities. An important example is the measurements of Johnson (9) on mixtures of

copper spheres in gelatin and on drop-shaped aluminum particles in gelatin. The results on copper spheres in gelatin agree well with Equation 4 using an  $n$  of 3, even though the conductivity ratio was over 1000 and compositions ranged from 5 to 20 vol. %. However, Johnson's results for drop-shaped aluminum particles suspended in the same gelatin gave higher values of thermal conductivity than were obtained with the copper spheres. These data show a significant shape effect, because the conductivity of copper is about twice that of aluminum (229/118). The thermal conductivity of graphite flakes (roughly disks) in silicone oil (16) also shows a shape effect. Unfortunately, investigators did not always give sufficient information about the specific shape of the particles used in their mixtures, so that little quantitative information about this effect can be obtained from their data.

This examination of previous experimental and theoretical work indicates that Equation 4 applies to mixtures whose component conductivities are nearly equal and to mixtures whose included particles are spheres, independent of the conductivity ratio. The effect of sphere size has not been investigated experimentally, but the theoretical results (Equation 3) indicate there is no effect. Also, for nonspherical shapes there is a significant increase in mixture conductivity, provided the conductivity of the components differs by a factor of about 100 or more. There was not, however, enough detailed experimental work on which to base a general correlation.

This experimental work was, therefore, directed toward the determination of  $n$  for those shapes for which theoretical results are not feasible and toward the determinations of the influence of particle size for systems of high conductivity ratio. The experiments used mixtures designed so that only conduction would contribute to heat transfer and so that the effect of large variations in shape could be investigated.

### Experimental

**Apparatus.** The apparatus used in these determinations consisted of an electrically heated sphere which was surrounded by a spherical shell of the mixture (Figure 1). Measurements of the steady state temperature difference across this shell were used with the thickness of the spherical shell to obtain values of thermal conductivity. The inside diameter of the test shell was 1.50 inches and the outside diameter was 2.10 inches. The heater consisted of a glass-insulated, 24-gage Constantan wire with a resistance of 0.40 ohm imbedded in a sphere of solder. The electric current through this resistor was measured by a General Electric Type AP-11 A-C ammeter of 0.25% accuracy class. This current was controlled by a Powerstat (Superior Electric Co.) and regulated by a Sola (Sola Electric Co.) a.c. regulator.

Copper-Constantan thermocouples were used to measure temperatures. The hot junctions of two of the thermocouples were imbedded in the solder which formed the heater, and those of two other thermocouples were imbedded in the outside surface of the rubber mixture. The cold junctions of all thermocouples were kept in a water bath at the temperature of the room. The voltages of these thermocouples were measured ( $\pm 2$  mv.) with a Leeds and Northrup Model 8662 potentiometer.

**Procedure.** The mixture shell was prepared by stirring a weighed amount of particles into a measured volume of Silastic (Dow-Corning Corp.) RTV-502 rubber and then adding 3 drops of catalyst which accelerated the vulcanization process. This catalyzed mixture was then poured into the mold which consisted of an inside, celluloid sphere, 1.50 inches in diameter, and an outside, hollow latex sphere. After the

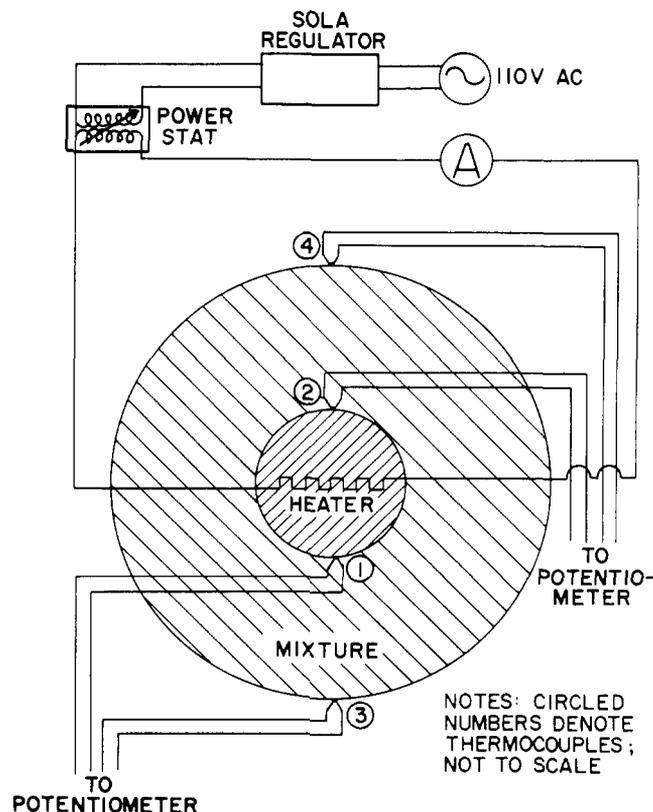


Figure 1. Conductivity cell

vulcanization was complete—usually about 10 minutes—the outside of the mold was removed and the shell was cut along a great circle to remove the celluloid sphere so the heater could be placed inside the shell. A small amount of freshly catalyzed rubber was spread over the surface of the heater to be sure the heater surface would be in good thermal contact with the inner surface of the shell. After enclosing the heater in the shell, the two thermocouple junctions were imbedded in the outer surface. The cell was then suspended inside a quiet air bath and the heater was turned on. After thermocouple readings became constant for a period of 2 hours, the inside and outside thermocouple voltages and the heater current were recorded. These data, the heater resistance, and the inside and outside radii of the test shell established the thermal conductivity.

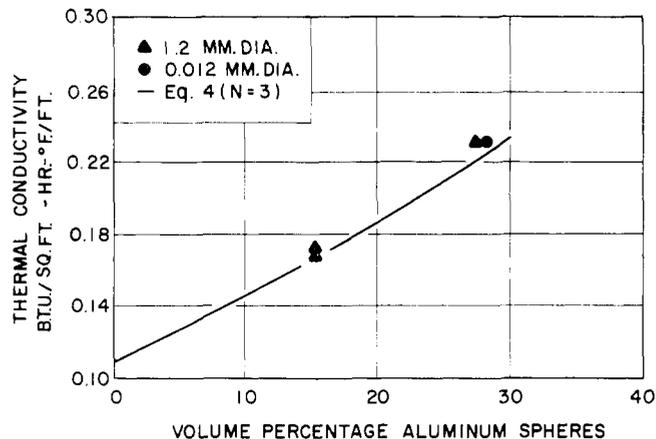
The first conductivity measurement was made on pure rubber. The measured value was  $0.109 \pm 0.002$  compared with  $0.127$  B.t.u./sq. ft.-hour-° F./ft. as given (4) by the manufacturer. This difference was probably due to differences in the vulcanization process, the amounts of catalyst added, and other factors. For the purposes of these experiments and calculations, a value of  $0.109$  was used for the rubber conductivity. During these preliminary measurements, the outside thermocouple junctions were moved from place to place around the outside surface of the spherical shell. This showed that the outside temperature varied less than  $0.1^\circ$  F. over the outer surface of the shell. X-ray photographs of the mixtures showed (8) that the particles were distributed uniformly through the mixture with no indication of preferential orientation. Additional tests showed that the temperature difference across the shell was directly proportional to the square of the heater current, as it should be if heat was being transferred only by conduction with no radiation or convection.

## Results

Table I gives measured values of thermal conductivity for mixtures of Silastic rubber and variously shaped particles of aluminum and balsa at selected volume compositions at  $95^\circ$  F. The results for the variously shaped aluminum particles clearly show an effect of shape. At the same volume composition, the conductivity of a mixture which contains cylinders, with a length to diameter ratio of 10 to 1, is nearly 1.5 times the conductivity of a mixture which contains spheres. However, the results obtained using nonspherical balsa wood

**Table I. Thermal Conductivity of Mixtures of Rubber and Particles of Aluminum or Balsa (Discontinuous Phase)**

Material	Shape	Particle Dimensions, Mm.	Volume % Particles	Measured Conductivity of Mixture, B.t.u./Sq. Ft.-Hr.-° F./Ft.
Aluminum	Spheres	1.2 diam.	27.0	0.234
Aluminum	Spheres	1.2 diam.	27.0	0.235
Aluminum	Spheres	1.2 diam.	15.5	0.173
Aluminum	Spheres	1.2 diam.	15.5	0.168
Aluminum	Spheres	0.012 diam.	27.5	0.238
Aluminum	Cylinders	5 × 1	15.5	2.04
Aluminum	Parallelepipeds	1.6 × 1.6 × 0.4	15.5	0.216
Aluminum	Cylinders	2.7 × 0.27	15.5	0.236
Balsa	Disks	0.8 × 7.2	25.0	0.087
Balsa	Disks	0.8 × 7.2	25.0	0.086
Balsa	Cubes	0.8 × 0.8	25.0	0.084
Balsa	Disks	0.8 × 7.2	14.0	0.096



**Figure 2. Comparison of Equation 4 with data on rubber-aluminum sphere mixtures**

particles show no shape effect. In all cases, the conductivity increases with increasing amounts of the higher conductivity phase.

The first four rows of Table I show the reproducibility of the determination to be about 2% for the more sensitive cases, which are those involving aluminum particles. The conductivities and densities of aluminum, balsa, and copper were taken from Forsythe (5), and Johnson's (9) values for gelatin were used in the calculations.

## Discussion

The comparison between the measurements for mixtures of aluminum spheres in rubber with Equation 4 using an  $n$  of 3 is shown in Figure 2. The agreement is satisfactory, and one must conclude that Equation 4 is appropriate for spherical inclusions regardless of the ratio of the conductivities of the two phases and regardless of the size of the sphere, at least in the size range used in these experiments.

The results for aluminum particles show a substantial shape effect. This effect can be accounted for by introducing  $n$  into Equation 4 as a function of particle shape. The sphericity function has been used to characterize shape for particles falling through fluids (2). The sphericity is defined as the ratio of the surface area of a sphere, with a volume equal to that of the particle, to the surface area of the particle. All the data obtained in this study can be correlated with Equation 4, provided one sets:

$$n = 3/\Psi \quad (5)$$

where  $\Psi$  is the sphericity. Figure 3 compares Equation 4 using this value for  $n$ , and the agreement is quite satisfactory.

According to the theoretical discussion, the effect of shape should not be important when the ratio of conductivities of the two phases is below about 100. The values obtained for mixtures of rubber and balsa wood agree completely with this idea, as shown in Table II, which compares measured values with those calculated from Equation 4 with  $n = 3$ .

Thus, Equation 4 agrees completely with the experimental results on widely different mixtures containing particles of different shapes.

An attempt was made to compare this general correlation with previous, applicable experimental results. Neither Johnson nor Sibbitt gave quantitative information about the shapes of the particles they used. Johnson gave a length to diameter ratio of 5 or more, and if the sphericity of these

**Table II. Comparison of Equation 4 with Data on Balsa Wood-Rubber Mixtures**

Volume Fraction Balsa	Sphericity of Balsa Particles	Measured Conductivity, B.t.u./Sq. Ft.-Hr.-° F./Ft.	Calcd. Conductivity (Eq. 4, $n = 3$ ), <sup>a</sup> B.t.u./Sq. Ft.-Hr.-° F./Ft.
0.25	0.5	0.087	0.086
0.25	0.5	0.086	0.086
0.25	0.8	0.084	0.086
0.14	0.5	0.096	0.095

<sup>a</sup>  $K_2 = 0.0242$ .

particles was 0.5, his results would agree with Equation 4 ( $n = 3/\Psi$ ) because  $n = 6$  for his data. Observation of graphite flakes, similar to those used by Sibbitt, showed the sphericity to be about 0.5. Because  $n$  must also be 6 for these data,  $n = 3/\Psi$ . On the basis of these comparisons, the data on these mixtures all are correctly predicted by Equation 4 with  $n = 3/\Psi$ .

It was naturally of interest to compare this functional value of  $n$  with the functional value from Fricke's theory. His results for  $n$  were computed in terms of sphericity. These computations showed that:

$$n = 3/\Psi^2 \tag{6}$$

for prolate ellipsoids, and:

$$n = 3/\Psi^{1.5} \tag{7}$$

for oblate ellipsoids. These do not compare favorably with  $n = 3/\Psi$ . Several factors were examined to explain the difference between Equation 5 and Equations 6 and 7.

Preliminary experiments have shown the effect of particle orientation to be large. However, in view of the x-ray examination of the mixtures and the care taken to ensure random orientation, this explanation was discarded. It was suggested that sphericity might not be the correct function with which to describe particle shapes, and other shape functions were tried with no better agreement between Fricke's results and all the data. Johnson made the same comparison on the basis of length to diameter ratio and found essentially these differences. With the lack of any data specifically contrary to the correlation presented here, this last explanation has been accepted, and it is suggested that Equation 5 be used to determine  $n$  as a function of particle shape. The preceding discussion referred to those systems for which the conductivity of the discontinuous phase was much larger than the conductivity of the continuous phase. Francl (6) and Marathe (13) have presented data for the inverse case, where the conductivity of the discontinuous phase is very much lower than that of the continuous phase. Francl correlated his data for porous metals with:

$$K = K_1 (1 - V_2) \tag{8}$$

where  $K$  is the conductivity of the composite,  $K_1$  is the conductivity of the pure metal, and  $V_2$  is "void" fraction or volume fraction of air in the powder. Marathe (13) found the same behavior for compressed, powdered copper. The general correlation should reduce to Equation 7 for these mixtures. Because  $K_1$  is much greater than  $K_2$  and because  $n$  is not less than 3,  $(n - 1)K_1$  will be much greater than  $K_2$ . Therefore, Equation 4 reduces to:

$$K = K_1 \left[ \frac{(1 - V_2)}{1 + \frac{V_2}{n - 1}} \right] \tag{9}$$

Over any practical range of porosity, Equation 9 agrees numerically with 8. The maximum difference is about 25% at such large porosities ( $V_2 = 0.5$ ) that no data are available with which to test the difference between Equations 8 and 9. Over the range of compositions which Francl studied, a valid test requires an experimental precision of at least 3%. His results were not presented in a form which permits a choice between Equations 8 and 9, and Francl's equation can be considered a special, simplified case of the correlation.

The variability of the thermal conductivity as a function of the particle (void) shape is contained in  $n$ . Equation 8 corresponds to an infinite value for  $n$ , and Equation 9 corresponds to a finite value for  $n$ . Except for very high porosities, only precise measurements can distinguish these values and, therefore, mixtures of this kind will show little dependence on the shape of void regions.

Thus, both cases possible for two-components systems containing phases of very different conductivity are correlated by Equation 4. If the continuous phase has the higher conductivity, there is little shape effect. If the discontinuous phase has the higher conductivity, there is a strong shape effect which can be accounted for using Equation 4 with  $n$  in terms of sphericity.

**Limitations**

In many practical cases, a number of complicating circumstances can appear. There can be serious question about which is the continuous phase [see Figure 4b of (7)]. This is a vital point, as this analysis shows, because the conductivity of a mixture composed of materials with rather different conductivities varies enormously depending upon which phase is continuous. (Foamed metals show clearly that the predominating phase need not be the continuous phase.) Additional compounds can be formed at the interfaces between the two phases. The particles can be so close together, even touching, so that the shape of the particles may be doubtful (7). The particles may not all have the same shape, and they may not be randomly oriented. All these complications will have a large effect on the mixture conductivity in cases where the phases have widely different conductivity. Other transfer mechanisms, particularly radiation transfer, will influence measured values of conductivity for heterogeneous materials. Additional phases might even be present.

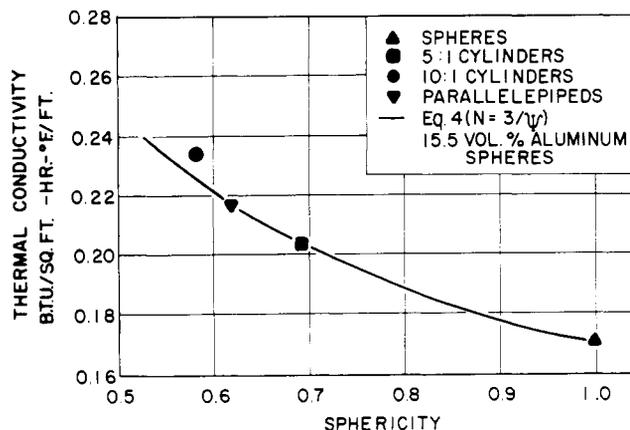


Figure 3. Comparison of Equation 4 with data as a function of particle sphericity

In spite of all these difficulties, the results obtained in this study can be of important help. Each of the complications mentioned can be approached on a more successful and rational basis if one assumes that Equation 4 correctly denotes the purely conductive heat transfer for the heterogeneous array. If a sample exhibits a strongly different behavior, then it would be best to assume that some other compound or mechanism or geometry makes a major contribution to the thermal transfer and to search for that contribution. It is believed that this view is entirely justified by the results obtained, and that more sagacious attacks upon these other difficulties will be found from it.

### Conclusions

The thermal conductivities of widely different types of two-component heterogeneous mixtures can be correlated using Equation 4. For mixtures in which the conductivity of the discontinuous phase is the larger by more than a factor of 100, Equation 5 should be used to obtain  $n$ , accounting for the influence of particle shape. For other mixtures the value of  $n$  in Equation 4 may be taken as 3.

### Nomenclature

- $K$  = thermal conductivity of the mixture, B.t.u. sq. ft.-hr.-° F./ft.  
 $K_1$  = thermal conductivity of the continuous phase, B.t.u. sq. ft.-hr.-° F./ft.  
 $K_2$  = thermal conductivity of discontinuous phase, B.t.u. sq. ft.-hr.-° F./ft.  
 $V_1$  = volume fraction of continuous phase, dimensionless  
 $V_2$  = volume fraction of discontinuous phase, dimensionless  
 $X$  = "shape factor" for ellipsoids in Fricke's equation, dimensionless

- $n$  = empirical shape factor, dimensionless  
 $\Psi$  = sphericity, dimensionless  
 $T$  = temperature, ° F.

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# MULTICOMPONENT VISCOSITIES OF GASEOUS MIXTURES AT HIGH TEMPERATURES

S. C. SAXENA<sup>1</sup> AND T. K. S. NARAYANAN *Chemical Engineering Division, Atomic Energy Establishment, Trombay, Bombay, India*

Binary diffusion and pure viscosity data are used to compute binary viscosities according to an expression derived from rigorous kinetic theory for monatomic gases. The rigorous expression is based on the assumption of central forces. An approximate but simpler formula proposed earlier by Wilke is also considered. A scheme is proposed for computing multicomponent viscosities from the known pure and related binary mixture viscosities at two compositions, and is tested for several ternary mixtures. This procedure permits calculation of the multicomponent viscosities for mixtures permuting out of the nine binary systems considered here with much less computation than for the rigorous method. The constants required for such a calculation are tabulated.

HIGH temperature viscosity data for gaseous mixtures are extremely useful to design engineers in problems involving fluid flow. Unfortunately, there are very few experimental data of this nature (10, 13). The only alternative, therefore, has been to calculate the multicomponent viscosity from the

<sup>1</sup> Present address, University of Rajasthan, Jaipur, India.

theoretical expression in conjunction with information about the intermolecular forces derived from some other source. According to the rigorous kinetic theory of Chapman and Enskog, the coefficient of viscosity of a  $\nu$ -component mixture,  $\eta_{\text{mix}}$ , is given by Equation 1. The development is based on the assumption of central forces.