

THERMAL CONDUCTIVITY OF TWO-PHASE SYSTEM
ZnO-GRAPHITE AND MgO-GRAPHITE

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Alexandria University, Egypt.Abstract:

Thermal conductivity of a two-phase (solid-solid) powder system was measured using a semi-absolute static method. Measurements were made on disk form samples having fixed porosity. Zinc oxide (ZnO) and magnesium oxide (MgO) powders were chosen to be the continuous solid phase. Each was mixed with different concentrations of graphite (the discontinuous phase). The experimental results have been compared with the available theoretical models for estimating either an effective thermal conductivity of such a system or lower and upper bounds of this property.

Introduction

The thermal conductivity is a very important property for applications that are involved with heat transfer. However, it is one of the most difficult transport coefficient to be measured with an acceptable degree of accuracy. In addition, the thermal conductivity has a high degree of difficulty to understand on standard physical basis as simply electronic and lattice contributions. This is especially at high temperature and for multiphase systems where so many factors are affecting heat conduction mechanism.

The aim of this work is to report about an experimental study on the thermal conductivity of solid-solid mixture system in which particles of one solid material are embedded in another continuous solid material. Experimental efforts were concentrated on two-phase porous materials which consist of a solid phase in the form of small particles and a saturating fluid phase filling the pore spaces. On the other hand, comparatively very little data was reported on solid-solid mixture system. The most recent study(1), performed in this laboratory, dealt with measuring thermal conductivity of MgO-graphite mixture with sodium silicate as a bonding matrix.

Various theoretical calculations on estimating the effective thermal conductivity K_E of two-phase systems have been reported in the literature(2-8). A lack of a general theoretical expression to determine K_E , which holds for all types of two-phase systems, has led to attempts to find bounds on K_E . The oldest serious attempt was due to Hashin and Shtrikman(9) followed by so many workers(10-16). We are

going to limit out comparison of the experimental results with only four theoretical expressions of K_E and two sets of bounds. These are collected as follows:

1. Maxwell(2), pioneered in the study of the thermal conductivity of two-phase mixtures, derived an expression to predict conductivity on the basis of potential theory:

$$K_E = K_c \left[\frac{K_d + 2K_c + 2\beta \Delta K}{K_d + 2K_c - \beta \Delta K} \right] \quad (1)$$

2. Russell (3) obtained the following formula:

$$K_E = K_c = \left[\frac{1 - \beta^{2/3} + K_d \beta^{2/3} / K_c}{(1 - \beta^{2/3} + \beta) + K_d (\beta^{2/3} - \beta) / K_c} \right] \quad (2)$$

3. Cheng and Vachon(7), modifying and extending Tsao's theoretical technique(6), predicted the thermal conductivity of heterogeneous solid mixtures as:

$$(K_E)^{-1} = \frac{1}{\sqrt{C \Delta K (K_c + B \Delta K)}} \ln \frac{\sqrt{K_c + B \Delta K} + (B/2) \sqrt{C \Delta K}}{\sqrt{K_c + B \Delta K} - (B/2) \sqrt{C \Delta K}} + (1-B)/K_c \quad (3)$$

where

$$B = \sqrt{3\beta/2} \quad \text{and} \quad C = 4\sqrt{2/3\beta}.$$

4. Chaurasia et al (8) considered separately the parallel and the series distribution to the heat flow direction as the first offers maximum value of K_E while the second distribution contributes to the minimum value. Then they used a weighted geometric mean of K_E (max.) and K_E (min.) and expressed K_E as:

$$K_E = (dK_d + bK_c)^n \left(\frac{K_f K_c}{cK_c + aK_d} \right)^{1-n} \quad (4)$$

where a, b, c and d are model elements:

$$a + b + c + d = 1, \quad c + d = \beta, \quad b + d = n$$

and where n is a weighting factor that gives the fraction of the sample having alternate zones of phases parallel to the direction of heat flow while the (1-n)th fraction of the sample is oriented perpendicular to the heat flow direction. The element d can be evaluated using $d = \beta^{1.3}$ and the element b is obtained through the expressions

$$b = (1/\Psi) \left(\frac{1-\beta}{1+\beta} \right)^3$$

where Ψ is the sphericity of the particles which was taken to be 0.82.

5. Devera and Strieder (14) used a random sphere model and applied variational techniques to obtain upper (K_U) and lower (K_L) bounds on K_E for a two-phase solid mixture of conductivities K_d and K_c with weight fractions β and $(1-\beta)$ respectively:

$$K_U \leq [\beta K_d + (1-\beta)K_c - m_o \ln \beta] (1 - m_o \ln \beta)^{-1}, \quad (5)$$

where

$$m_o = \frac{\beta \Delta K}{3K_c + 2\beta \Delta K},$$

and

$$(K_L)^{-1} \leq \left[\frac{\beta}{K_d} + \frac{1-\beta}{K_c} + \frac{2w_o \ln \beta}{K_c} \right] (1 + 2w_o \ln \beta)^{-1}, \quad (6)$$

where

$$w_o = \left(\beta \frac{\Delta K}{K_d K_c} \right) \left(\beta \frac{\Delta K}{K_d K_c} + \frac{3}{K_c} \right)^{-1}$$

6. Recently Pande et al(16) were able to optimise the weighted geometric mean of resistors considered for determining of two-phase systems and obtained the following set of bounds:

$$K_U = \frac{1}{(e^b - 1)} (\Delta K e^{b\beta} + K_c e^b - K_d), \quad (7)$$

and

$$K_L = \frac{1}{(e^a - 1)} (\Delta K e^{a\beta} + K_c e^a - K_d), \quad (8)$$

where

$$a = \frac{\Delta K}{(K_c K_d)^{1/2}} \quad \text{and} \quad b = \frac{K_d^2 - K_c^2}{K_d^2 + K_c^2}$$

In the above expressions (1-8), subscript c stands for the continuous phase and d for the discontinuous phase, β is the weight fraction of the discontinuous phase and ΔK represents the difference between the two conductivities ($K_d - K_c$).

Experimental Procedures

First, the powder of each phase was purified from all loosely adhering particles and from any magnetic materials. In order to achieve an approximate uniformity of particle size, each powder was passed through a standard sieve (mesh no. 100). Then, the powder was dried in an electric furnace at temperature of about 120°C for four hours. This temperature was chosen to be much lower than the oxidizing temperature of graphite (17). In order to form mixtures of different concentrations, the two powders (ZnO or MgO and graphite) of different weight proportions were thoroughly mixed together. Samples were prepared in the form of disks of fixed porosity using an Amstler Universal cupping machine with fixed pressure of 15 tons for all samples. The thermal conductivity was then measured for each sample using an apparatus whose description and theory can be found elsewhere (18,19).

Results and Discussions

The experimental thermal conductivities of both systems were obtained at room temperature. The data is plotted as a function of graphite concentration and are presented in Figs. (1) and (2) for Zinc oxide-graphite mixture system and in Figs (3) and (4) for magnesium oxide-graphite mixture system. Also are plotted, for comparison, the calculated effective thermal conductivity K_E according to the four expressions eqs. (1-4) in Figs. (1) and (3). In Figs. (2) and (4) are given the calculated upper and lower bounds of the thermal conductivity using eqs. (5-8).

a- Zn-Gr. mixture system

As can be seen from Fig. (1), the experimental data of ZnO-Gr. system agree reasonably well in terms of behaviour in the whole range of β with K_E calculated from Maxwell's and Russel's expressions. In terms of magnitude, the maximum deviation is about 10% for $\beta < 0.5$ and the deviation decreases as β increases. The calculated K_E using eqs. (3,4) deviate from K_{exp} , both in magnitude and behaviour. This can be understood in terms of the convergence of K_E calculated using expressions (3,4) as β approaches zero and unity. For Cheng and Vachon (7) expression: K_E tends to K_C as β approaches zero while it diverges quickly as β tends to unity. This may be due restricting the distribution of the discontinuous phase in the mixture to a parabolic one which is unrealistic. Looking carefully to Chaurasia et al (8) theoretical model, one finds that the model dements a , b , c , and d , and the weighting factor n are completely dependent on the volume fraction β and are entirely independent on the system. This means that these elements have fixed values for a certain β for any two-phase system irrespective of particle distribution and orientation, and of the constituent thermal conductivities K_C and K_D . There is also a divergence of K as β approaches zero or unity.

One can notice from Fig. (2) that the experimental data nearly coincides with the lower bound K_L of the theoretical model due to Devera & Strieder (14).

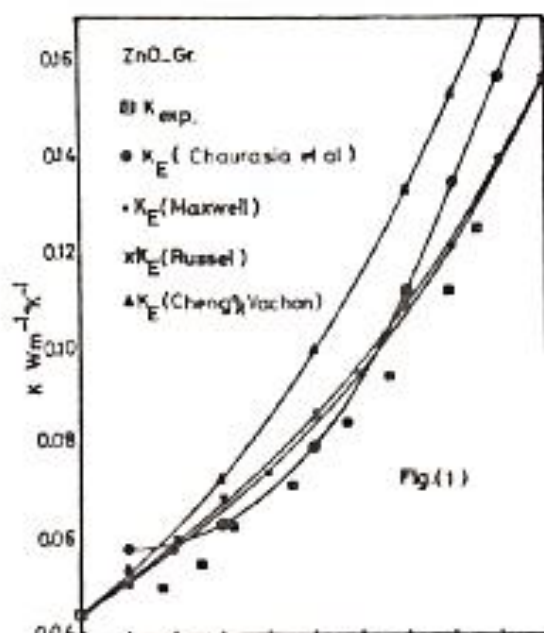


Fig. (1)

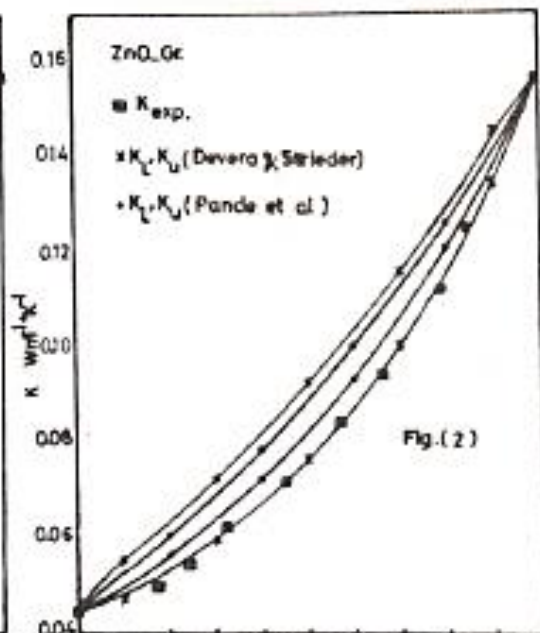


Fig. (2)

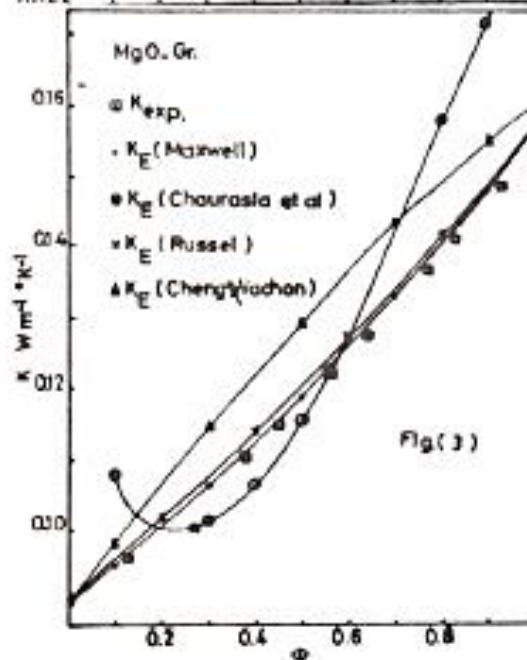


Fig. (3)

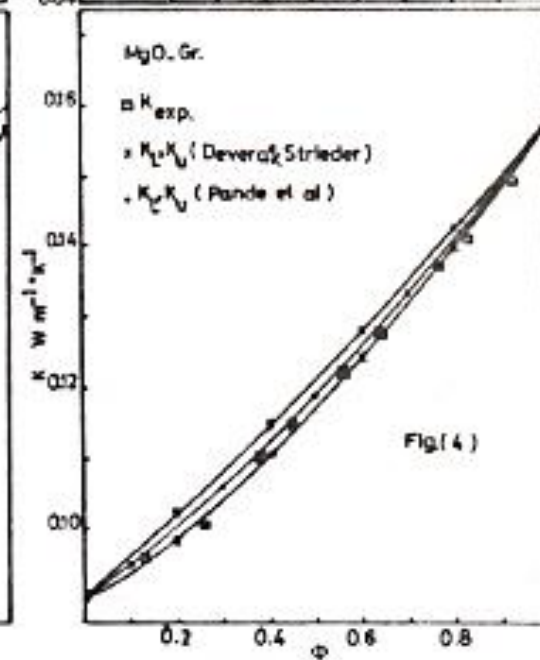


Fig. (4)

Fig. (1): Experimental thermal conductivity (K_{exp}) versus weight fraction of graphite (β) for ZnO-Gr. system together with calculated effective thermal conductivity (K_E) using eqs. (1-4).

Fig. (2): K_{exp} vs. β for ZnO-Gr. system as compared to two sets of bounds (K_L, K_U) using eqs. (5-8).

Fig. (3): K_{exp} vs. β for MgO-Gr. system together with K_E calculated using eqs. (1-4).

Fig. (4): K_{exp} vs. β for MgO-Gr. system as compared to two sets of bounds (K_L, K_U) eqs. (5-8).

It is not surprising that K_{exp} agree well with K_L rather than to be in between the bounds. This is because the sample is not completely packed and so the existence of even very few air pockets in the sample would lower the measured conductivity. The other model due to Pande et al (16) gives a smooth set of bounds similar in behaviour to K_{exp} , but its K_L is higher than K_{exp} , at most by 10%. This last model gives K_L and K_U completely enclosed within the first set with a relatively less area covered by bounds. Pande et al (16) claimed that a theoretical model which gives the least area of bounds is the most realistic one.

b- MgO-Gr. mixture system:

It is clear from Fig. (3) that K_{exp} of this system behaves with respect to the calculated K_C in a similar way to the ZnO-Gr. system. It is also to be noted that K_G (of Gr.) is only about 1.74 K_C (of MgO) so that one set of bounds (pande et al (16) gives nearly identical K_L and K_U while the corresponding valuesw of the other set of bounds (Davera and Strieder (14) are close to each other and consequently this set has a very small area of bounds. The experimental data agree quite well with both sets of bounds as can be seen from Fig. (4).

Comparing our experimental results to those reported on MgO-Gr. system by Ammar and Mahmoud (1), one finds that the oscillations of K_{exp} observed in their results, for $0.30 < \beta < 0.60$, may be due to the effect of the bonding matrix (sodium silicate) on the conductivity but out of this range of β , K_{exp} is nearly similar in behaviour to the present results. The effect of bonding the particles in the mixture by a glassy liquid on the effective thermal conductivity still requires further experimentation and more theoretical study.

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