

Thermal Conductivities of Powder-Filled Epoxy Resins

F. LIN,* G. S. BHATIA, and J. D. FORD†

Department of Chemical Engineering, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1

SYNOPSIS

Effective thermal conductivities of powder-filled epoxy composites were experimentally obtained using a transient method. Fillers used were aluminum and cupric oxide. Comparisons of these data with published correlations indicate that Agari's model can give better fits. Nielsen's model may also give a good prediction if the shape of particles can be determined and values of parameters A and ϕ_m are available. An alternative method of dealing with parameter C_2 in Agari's model is suggested. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

With increasing demand for plastics or resins in different branches of engineering, the addition of other materials—metals, graphite, oxides, glass, etc.—has been widely applied. The resulting composites combine the advantages of plastics or resins with those of the additives, which could provide improvements in the physical properties. In particular, cupric oxide-filled plastics can be used in microwave heating applications. For such heterogeneous systems, then, more information on the physical properties is required. The determination of effective thermal conductivity, for example, has led to numerous experimental and theoretical studies. In this work, the effective thermal conductivities of epoxy resins containing aluminum or cupric oxide powder with various filler concentrations were obtained by a transient method, based on direct contact condensation of steam on a sphere. The results for aluminum-epoxy composites were compared with data from the literature, and both sets of data compared with predictions from several well-known models.

Models in the Literature

For a two-component composite, the simplest alternatives would be with the materials arranged in

either parallel or series with respect to heat flow, which gives the upper or lower bounds of effective thermal conductivity. Considering that the actual value of conductivity must be located somewhere between the two extremes, Woodside and Messmer¹ suggested the use of a weighted geometric mean of the thermal conductivities of the two components. However, Parrott and Stuckes² indicated that this model was mathematically unsound and tended to overestimate the conductivity of the system.

The well-known Maxwell-Eucken³ equation was frequently used because of its simplicity:

$$k_c = k_m \left(\frac{k_f + 2k_m + 2\phi(k_f - k_m)}{k_f + 2k_m - \phi(k_f - k_m)} \right) \quad (1)$$

where k_c , k_m , and k_f are thermal conductivities of composite, matrix, and filler, respectively, and ϕ is the volume fraction of filler. Based on different assumptions, Bruggeman⁴ developed an implicit correlation:

$$1 - \phi = \left(\frac{k_f - k_c}{k_f - k_m} \right) \left(\frac{k_m}{k_c} \right)^{1/3} \quad (2)$$

An empirical parameter of sphericity was introduced by Hamilton and Crosser,⁵ by which the shape of filler particles was taken into account. When particles are spherical, their formula is identical with eq. (1).

Cheng and Vachon⁶ extended Tsao's⁷ model assuming a parabolic distribution of the filler:

$$\frac{1}{k_c} = \frac{1}{MN} \ln \frac{2N + BM}{2N - BM} + \frac{1 - B}{k_m} \quad (3)$$

* Visiting Scholar, East China University of Chemical Technology, Shanghai, People's Republic of China.

† To whom correspondence should be addressed.

where

$$B = \left(\frac{3}{2}\phi\right)^{1/2}, \quad C = 4\left(\frac{2}{3}\phi\right)^{1/2},$$

$$M = [C(k_f - k_m)]^{1/2},$$

$$N = [k_m + B(k_f - k_m)]^{1/2}, \quad \text{for } k_f > k_m.$$

From experimental results, Sundstrom and Lee⁸ concluded that the Bruggeman equation gave better prediction up to 10% of filler by volume, whereas the Cheng-Vachon equation was more accurate in the range of 15–25% by volume. Nielsen⁹ indicated that their data could be fitted by the Lewis and Nielsen¹⁰ equation, in which effects of the shape of particles and type of packing were included:

$$k_c = k_m \left(\frac{1 + AB\phi}{1 - B\psi\phi} \right) \quad (4)$$

where

$$B = \frac{k_f/k_m - 1}{k_f/k_m + A}, \quad \psi = 1 + \left(\frac{1 - \phi_m}{\phi_m^2} \right) \phi$$

where A is a constant related to the generalized Einstein coefficient k_E , and ϕ_m , the maximum packing fraction of the filler. Some values of A and ϕ_m were given.¹¹

Oleynikova and Dushchenko¹² showed that several formulae available in the literature were different expressions of the Maxwell-Eucken correlation. They recommended eqs. (1) and (2) for calculation of thermal conductivities of two-component systems containing less than 12% by volume of filler. Progelhof et al.¹³ reviewed numerous correlations in the literature and concluded that the Lewis and Nielsen correlation fitted the data best for the range of fillers tested.

Based on the generalization of models for parallel and series conduction in composites, Agari and Uno¹⁴ developed a new model:

$$\log k_c = \phi C_2 \log k_f + (1 - \phi) \log(C_1 k_m) \quad (5)$$

Two new parameters were introduced, which could be obtained by regression from experimental data. C_1 is a measure of the effect of particles on the secondary structure of the polymer and C_2 measures the ease of particles to form conductive chains. Later, they¹⁵ modified it with an aspect ratio to take the shape of particles into account. Generally speaking, this semiempirical model seems to fit the experimental data well. However, adequate experi-

ments need to be completed before it can be fully established.

EXPERIMENTAL

Background

The effective thermal conductivity of a composite, k_c , is defined as the product of effective thermal diffusivity, α_c , density, ρ_c , and heat capacity, C_{pc} :

$$k_c = \alpha_c \rho_c C_{pc} \quad (6)$$

In this study, a transient method is used to determine the effective thermal diffusivity of a composite, and then the effective thermal conductivity can be readily obtained by eq. (6).

The well-known problem of heat conduction into a sphere initially at a uniform temperature, when suddenly placed in a higher constant temperature field, has been thoroughly analyzed. In the present case, a sphere is heated by being exposed to saturated steam. The condensing steam provides a very high heat transfer coefficient as the boundary condition for the governing equation, by which the Biot number can be considered to tend to infinity. The study of Lin and Ford¹⁶ showed that for a lead sphere of diameter 5.12 cm, with a condensing coefficient of 11.2 kW/m² K, the external heat transfer resistance due to the thin condensate film was very small, as assumed.

In this study, $k_{\text{epoxy}} \ll k_{\text{lead}}$ (less than 1% of k_{lead}) and the resistances within the spheres are much greater than that of the lead sphere. Therefore, the assumption of negligible resistance of the condensate film is reasonable. The temperatures at the center of the spheres are measured as a function of time. The solution then becomes

$$\theta = 2 \sum_{n=1}^{\infty} (-1)^{n+1} \exp[-(n\pi)^2 Fo] \quad (7)$$

where

$$\theta = \frac{T_s - T}{T_s - T_i}, \quad Fo = \frac{\alpha t}{r_0^2}$$

and T_s is the temperature of steam; T_i , the initial temperature of the sphere; t , the time; and r_0 , the radius of the sphere. After an initial time interval (e.g., $Fo = 0.12$), θ can be obtained from the leading term of the series, neglecting higher terms:

$$\theta = 2 \exp[-\pi^2 Fo] \quad (8a)$$

or in logarithmic form:

$$\ln \theta = \ln 2 - \pi^2 Fo = \ln 2 - \frac{\pi^2 \alpha}{r_0^2} t \quad (8b)$$

The linear relation between $\ln \theta$ and t is the basis of the measurements in this study. Five or six points ($\theta < 0.3$) from the linear portion of the curves are enough to obtain the slope ($-\pi^2 \alpha / r_0^2$) by means of least squares.

Sample Preparation

The spheres were made by injecting an epoxy-powder mixture into an aluminum mold and curing at an elevated temperature. The mold consisted of two smooth-faced aluminum blocks with matching hemispherical portions (2.6 cm in diameter) machined out at the center of the smooth-faced side. Joining the two blocks together formed a hollow sphere in the center of the mold. A small hole was drilled from the top of the blocks to allow for injection of the mixture into the mold. A pin of diameter 0.16 cm and length 1.30 cm protruding radially was placed at the hemispherical wall of one block so that the molded sphere would have a preformed hole exactly at its center.

The mold was cleaned and smoothed using an emery cloth, and the inside faces sprayed with Teflon. The two components of epoxy (resin and curing agent manufactured by Conap Inc. and supplied by Amethane Inc.) were placed in separate containers, heated at 70°C for 5 min to reduce their viscosities, and weighed into another container according to a 5 : 1 ratio of curing agent to resin. The cupric oxide powder (average particle diameter 3 μm) was weighed in and the three components mixed thoroughly. The mixture was kept at 70°C for 15 min with occasional mixing and poured into a 20 cm^3 hypodermic syringe, by which the mixture was injected into the clamped mold. After the mold was placed in an oven at 70°C for 5 h, the cured sample was removed and any rough edges on the sphere filed away using an emery cloth.

The procedure for preparing epoxy-aluminum spheres was similar to that described above except that the system was partially cured outside the mold. After heating the three-component mixture in a boiling bath with continuous stirring for 5 min, its viscosity increased rapidly. After injection, the mold opening was blocked with a small cork and the mold turned every 30 s for 10 min. The mold was then placed in an oven for 2 h at 110°C to complete the curing. This was done to prevent the heavier alu-

minum powder (average particle diameter 7 μm) from settling during the curing, allowing a more uniform distribution of the powder in the sphere. The above procedures were worked out after sectioning spheres to check for uniform particle distribution. The measurements of diameter, density, and heat capacity of the prepared spheres can be found in the work of Bhatia.¹⁷

Measurements

The arrangement of experimental apparatus and procedure can be found in the study of Lin and Ford.¹⁶ The temperature probe was a fast response (time constant 0.5 s) chromel-alumel thermocouple (type K) in a flexible stainless-steel sheath of outside diameter 0.159 cm. The thermocouple probe was forced into the hole in the sphere until a tight fit was obtained. The temperature readings from the thermocouple were recorded on a Sargent-Welch chart recorder (Model SRG) as temperature vs. time. Check runs with an insulating sheath on the thermocouple confirmed the accuracy of the above procedure.

RESULTS AND DISCUSSION

Typical curves of temperature vs. time and $\ln \theta$ vs. time for samples of pure epoxy, aluminum-epoxy, and cupric oxide-epoxy spheres are shown in Figures 1 and 2.

Epoxy

Two samples of pure epoxy (without filler) were cast. The average density and heat capacity were obtained as 1130 kg/m^3 and 1.83 $\text{kJ}/\text{kg K}$. The thermal diffusivity of epoxy of $1.07 \times 10^{-7} \text{ m}^2/\text{s}$ from the regression produces a thermal conductivity of 0.221 $\text{W}/\text{m K}$. Table I shows the comparison of measured thermal conductivity of epoxy resin with those in the literature. It can be noticed that despite the variation in the properties of epoxy resin as well as its composition, the present result of thermal conductivity falls into the right range. This also confirms that the measuring technique used can produce reliable results.

Epoxy with Aluminum Powder

Since the mixture was prepared by weight, the conversion of mass fraction to volume fraction of filler in the mixture was calculated using the following equation:

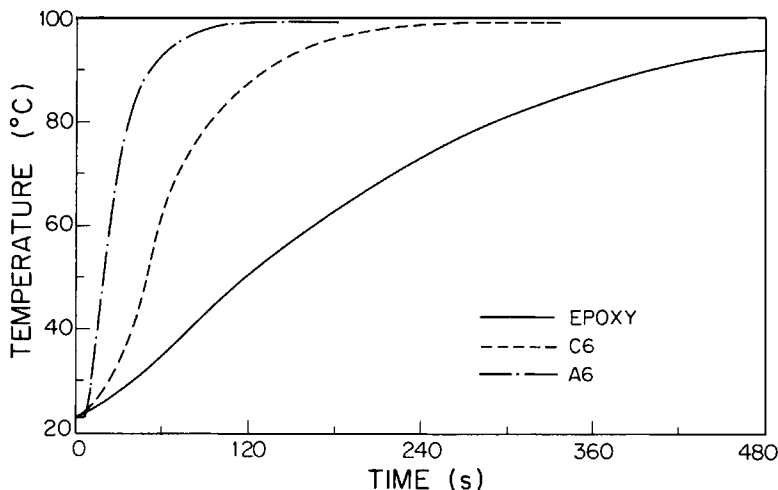


Figure 1 Heating curves of epoxy, CuO-epoxy (C6), and Al-epoxy (A6) spheres.

$$\phi = \frac{\rho_c}{\rho_p} X \tag{9}$$

where ρ_p is the density of powder, and X , the mass fraction of powder in the composite.

Table II demonstrates the effect of the addition of aluminum powder on the thermal diffusivity and conductivity. It can be seen that the effect is small at low filler concentrations but increases rapidly as the concentration of Al powder reached higher levels. For example, the thermal conductivity of the mixture jumps from 120 to over 800% of that for pure epoxy, whereas the filler concentration increases from 4.5 to 44.5%. These results provide an opportunity to compare them with some published works (Kline¹⁸ and Bigg²²), shown in Figure 3, in which

thermal conductivities are normalized as factors of the conductivity of the matrix to allow for comparison between them. The results of Kline displayed as a function of mass fraction were also transformed to volume fraction. It can be noticed that the results from the present study are located between those two sets, but much closer to those of Kline, especially at higher filler concentrations. That the results of Bigg are much higher than those of the other two sets can be attributed to the high ratio of length to diameter of the particles (12.5), these particles being more likely to "bridge" to conduct heat flow. Kline used fine aluminum powder (average diameter 15 μm), supposedly spherical. In the present study, the aluminum powder used has an aspect ratio about 3.

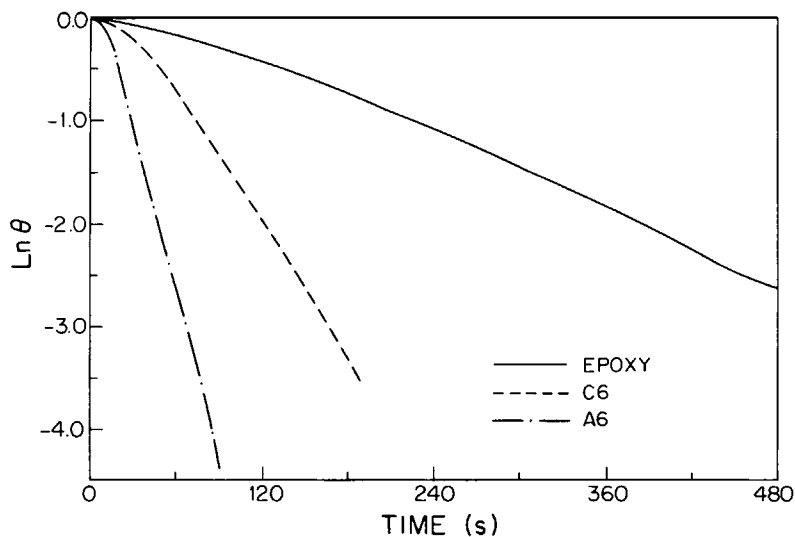


Figure 2 Ln θ vs. time for epoxy, CuO-epoxy (C6), and Al-epoxy (A6) spheres.

Table I Comparison of Thermal Conductivity of Epoxy Resins

Source	Kline ¹⁸	Garrett and Rosenburg ¹⁹	Kusy and Corneliussen ²⁰	Hansen and Tomkiewicz ²¹	Present
k (W/m K)	0.188 (60°C)	0.22 (27°C)	0.225	0.264	0.221 (60°C)
$\rho \times 10^{-3}$ (kg/m ³)	1.19	1.21	—	—	1.13
Composition	Epon 828 with <i>m</i> -phenylenediamine in a ratio of 85 : 15	Araldite MY740 with HY906 and DY602 in a ratio of 100 : 80 : 2	—	—	AD1000A with RN1000 in a ratio of 100 : 20

Therefore, the results shown in the figure can be expected.

Epoxy with Cupric Oxide Powder

From Table III, it can be seen that the addition of cupric oxide powder increases the thermal diffusivity and conductivity of the composite substantially. When the composite contains 28% CuO powder by volume, the thermal conductivity increases 3.6 times. Samples of C2 and C3 were used to test the reproducibility of the sample preparation and experimental technique. The thermal conductivities of these two samples, with 0.3% difference of cupric oxide by volume, agree with each other within 1%. Hence, the reproducibility of experiments is confirmed.

It is interesting to notice that at comparable filler concentrations the effect of the two powders are of the same order of magnitude. For example, k_c of A4 and C6 are 0.877 and 0.788 W/m K, whereas their filler concentrations are 29.6 and 28.0% by volume, respectively. These results confirm what was found by Cullen et al.,²³ who concluded that once the thermal conductivity of the filler becomes much larger than that of the matrix (over 50 times), a further

increase in the thermal conductivity of the filler at moderate filler concentrations has little effect on that of the composite. They compared the effect of copper and carbon steel on plastics and found a difference of only 5–6% between the composite conductivities, although the conductivity of copper is over five times that of carbon steel. In this study, the conductivity of aluminum (236 W/m K) is 25 times that of cupric oxide (9.21 W/m K), which is over 40 times higher than that of epoxy resins (0.221 W/m K).

Comparisons

Comparisons of experimental data with those models for Al-epoxy composites are shown in Figure 4. The average deviations between experimental data and predictions from the models are 18.7, 18.2, 5.9, and 4.5% for Bruggeman, Cheng, Nielsen, and Agari, respectively. For the first two models, experimental results are close to the predictions at low filler concentrations, whereas the predictions deviate significantly from experimental results at higher filler concentrations. The deviations of Bruggeman's and Cheng's models are comparable in the experimental range. Cheng's overestimates at low filler concen-

Table II Effect of Aluminum Powder Concentration

Sample No.	X (%)	ϕ (%)	$r_0 \times 10^2$ (m)	$\rho_c \times 10^{-3}$ (kg/m ³)	C_{pc} (kJ/kg K)	$\alpha_c \times 10^7$ (m ² /s)	k_c (W/m K)	k_c/k_m
Epoxy	0.0	0.0	1.27	1.13	1.83	1.07	0.221	1.00
A1	10.1	4.5	1.29	1.20	1.68	1.34	0.270	1.22
A2	24.0	10.9	1.29	1.23	1.73	1.62	0.345	1.56
A3	38.5	20.4	1.29	1.43	1.57	2.38	0.534	2.42
A4	50.0	29.6	1.29	1.60	1.51	3.63	0.877	3.97
A5	62.5	39.3	1.30	1.70	1.25	5.79	1.230	5.57
A6	69.5	44.5	1.30	1.73	1.21	8.66	1.813	8.20

$$\rho_{Al} = 2700 \text{ kg/m}^3, k_{Al} = 236 \text{ W/m K.}$$

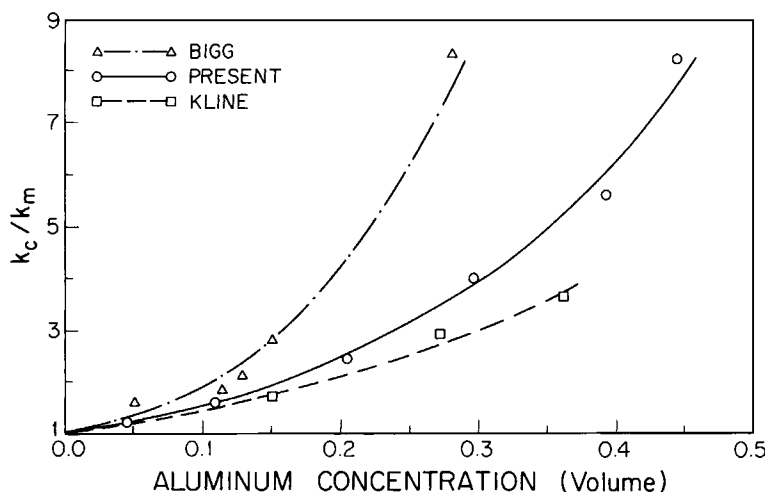


Figure 3 Comparison between literature and present results for Al-plastic systems.

trations and gives better predictions than those of Bruggeman's in the range of 10–30%. They crossover at about 35%.

Since Nielsen's model is semiempirical, parameters of A and ϕ_m need to be suitably chosen. Nielsen⁹ suggested the values of $A = 3$ and $\phi_m = 0.64$ for irregularly shaped particles. The values of ϕ_m for random loose packing of spheres and randomly oriented rods are 0.60 and 0.52. In this study, $\phi_m = 0.56$ gives better predictions. From Al powder photographs, some particles appear roughly spherical and others have a rodlike shape. Agari's model is also semiempirical, and the regression provides the best fit with parameters of $C_1 = 0.974$ and $C_2 = 0.567$ for Al-epoxy composites.

Comparisons for CuO-epoxy composites are shown in Figure 5 and have profiles similar to those in Figure 4. The average deviations are 11.8, 15.7, 10.5, and 2.6% for the four models. Cheng's and Bruggeman's crossover is at 30% of filler by volume. Parameters of $A = 3$ and $\phi_m = 0.6$ are used for Niel-

sen's equation, but it cannot fit the experimental data as well as those for the Al-epoxy composite. CuO powder photographs show a totally irregularly shaped particle, which suggests that these parameters need to be adjusted, but the values of parameters are not available. Parameters in Agari's model are obtained as $C_1 = 0.930$ and $C_2 = 1.447$.

According to Agari's arguments, both C_1 and C_2 should be in the range zero to unity. From this study, values of C_1 are close to unity and this means that particles of filler have little effect on the secondary structure of epoxy resins. The value of C_2 for Al-epoxy falls in the designated range, whereas that for CuO-epoxy, however, exceeds this restriction. Agari et al.²⁴ attributed their results of C_2 (greater than 1) not only to the ease in forming conductive chains of particles, but also to the thermal contact resistance between polymer and particles. They²⁵ claimed that the conductivity of a composite was significantly affected by the dispersion state. Values of C_2 (from -1.88 to 2.32) were obtained from four dif-

Table III Effect of Cupric Oxide Powder Concentration

Sample No.	X (%)	ϕ (%)	$r_0 \times 10^2$ (m)	$\rho_c \times 10^{-3}$ (kg/m ³)	C_{pc} (kJ/kg K)	$\alpha_c \times 10^7$ (m ² /s)	k_c (W/m K)	k_c/k_m
Epoxy	0.0	0.0	1.27	1.13	1.83	1.07	0.221	1.00
C1	8.5	1.6	1.28	1.21	1.60	1.15	0.223	1.01
C2	24.5	5.3	1.28	1.38	1.57	1.17	0.254	1.15
C3	26.0	5.6	1.28	1.38	1.55	1.18	0.252	1.14
C4	35.1	8.3	1.29	1.51	1.50	1.34	0.304	1.38
C5	50.0	17.0	1.28	2.18	1.05	2.10	0.481	2.18
C6	69.0	28.0	1.29	2.60	0.95	3.19	0.788	3.57

$$\rho_{CuO} = 6400 \text{ kg/m}^3; k_{CuO} = 9.21 \text{ W/m K.}$$

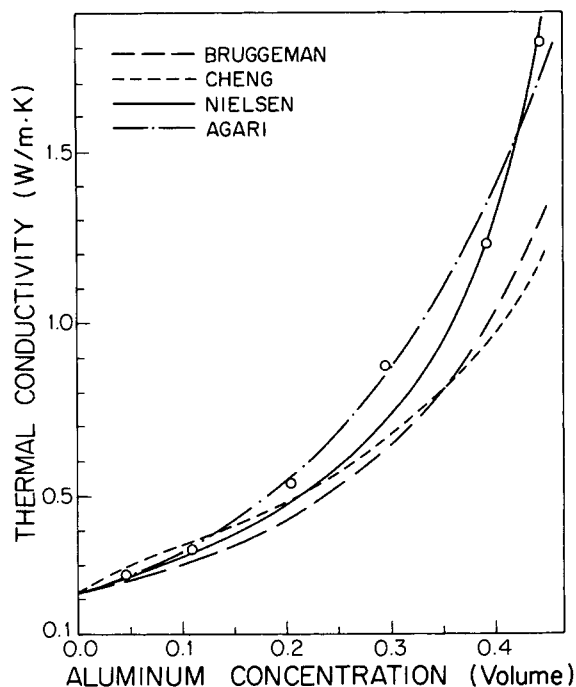


Figure 4 Comparison of experimental results with correlations for Al-epoxy composites.

ferent dispersion states. Critical volume fraction of particles and magnification of conductivity of particles were thought responsible for the variation of C_2 . All these make the explanation dubious.

From the least-squares regression, it was found that the parameter C_2 varied with the unit used. For example, the values of C_2 were 0.567, -1.76, and 5.13 with respect to the units of W/m K, W/cm K, and cal/s cm K for the thermal conductivity of the Al-epoxy composite. This raised the question of why C_2 varied with the unit. Equation (5) can be rearranged as

$$k_c = \left[\frac{k_f^{C_2}}{C_1 k_m} \right]^\phi (C_1 k_m) \quad (5a)$$

It can be noticed that the dimensions of two sides in eq. (5a) are nonhomogeneous unless C_2 equals unity. Equation (5) can be also written as

$$\log k_c = a + b\phi \quad (5b)$$

where $a = \log(C_1 k_m)$ and $b = C_2 \log k_f - \log(C_1 k_m)$. Therefore, $\log k_c$ is a linear function of ϕ with slope b and intercept a . Set $k' = nk$, where $n (> 0)$ is the unit conversion factor. Then, $\log k'_c$ has a slope of b' and an intercept of a' instead. From the regression, it can be obtained that $a' = a + \log n$ and $b' = b$.

The conclusion is that $C'_1 = C_1$ and $C'_2 = (m + C_2)/(m + 1)$, where $m = \log n/\log k_f$. C_2 is obviously related to the unit used for thermal conductivity. Therefore, it seems to us that the significance of C_2 designated by Agari is unsuitable.

Kusy and Corneliussen²⁰ reviewed Taylor's²⁶ study in which a densely packed powder of Al exhibited only 1/1000 the conductivity of a piece of bar stock and that of Cu showed only 1/400 that of the wrought material. Therefore, they proposed a metallic conduction coefficient, λ , in the parallel model:

$$k_c = \phi(\lambda k_f) + (1 - \phi)k_m \quad (10)$$

where λ could be a polynomial of volume fraction of filler, ϕ . They claimed that this model gave quite good predictions for the experiments of PVC/Ni and PVC/Cu mixtures in the range of $0 \leq \phi \leq 0.15$. This model is also tested with good fits in this study but the curves are not shown in Figures 4 and 5. The average deviations are 4.0 and 6.4% for CuO-epoxy and Al-epoxy composites, respectively.

Based on the above, we suggest an alternative:

$$\log k_c = \phi \log(C_2 k_f) + (1 - \phi) \log(C_1 k_m) \quad (10)$$

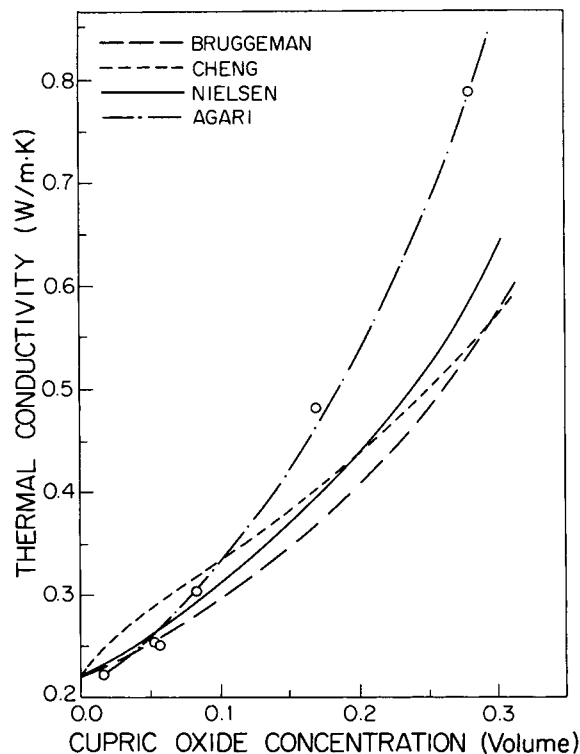


Figure 5 Comparison of experimental results with correlations for CuO-epoxy composites.

C_1 and C_2 can be also obtained by the same procedure with the same deviations as for eq. (5). Here, C_2 does not vary with the unit used. The values of C_2 are 0.094 and 2.70 for Al-epoxy and CuO-epoxy composites, respectively. C_2 may still be a measure of the ease of particles to form conductive chains, but is also a measure of the contact between particles and the matrix.

CONCLUSIONS

In this study, thermal conductivities of epoxy composite were obtained via measurement of thermal diffusivities by a transient method. Epoxy served as the composite matrix and its thermal conductivity was experimentally determined as 0.221 W/m K, comparing favorably with published data.

Two sets of effective thermal conductivities vs. volume fraction of filler were obtained. The set for CuO-epoxy composites (up to $\phi = 0.28$) was not found in the literature. The comparison of the set for the Al-epoxy composite with those of Kline and Bigg demonstrates the effect of the shape factor of particles on the thermal conductivity of the composite.

Experimental results are compared with five models. Bruggeman's gives better predictions up to about 10% of filler by volume while Cheng's overestimates. In the range of $0.1 < \phi < 0.2$, Cheng's predictions are better than those of Bruggeman's. Beyond that, both models produce significant deviations. Nielsen's model may be used up to a high-volume fraction of fillers if the shape of particles can be determined and values of parameters (A and ϕ_m) are available. Both Kusy's and Agari's models can fit experimental results well by regression. Even though Agari's model produces the smallest deviation, the significance of parameter C_2 is still not clear since it varies with the unit used. An alternative formulation containing C_2 is proposed.

REFERENCES

1. W. Woodside and J. H. Messmer, *J. Appl. Phys.*, **32**, 1688 (1961).
2. J. E. Parrott and A. D. Stuckes, *Thermal Conductivity of Solids*, Pion, London, 1975, p. 131.
3. A. Eucken, *Forsch. Gebiete Ingenieurw. B3 Forschungsheft*, **353**, 16 (1932).
4. D. A. G. Bruggeman, *Ann. Phys.*, **24**, 636 (1935).
5. R. L. Hamilton and O. K. Crosser, *Ind. Eng. Chem. Fund.*, **1**, 187 (1962).
6. S. C. Cheng and R. I. Vachon, *Int. J. Heat Mass Transfer*, **12**, 249 (1969).
7. G. T.-N. Tsao, *Ind. Eng. Chem.*, **53**, 395 (1961).
8. D. W. Sundstrom and Y.-D. Lee, *J. Appl. Polym. Sci.*, **16**, 3159 (1972).
9. L. E. Nielsen, *J. Appl. Polym. Sci.*, **17**, 3819 (1973).
10. T. B. Lewis and L. E. Nielsen, *J. Appl. Polym. Sci.*, **14**, 1449 (1970).
11. L. E. Nielsen, *Ind. Eng. Chem. Fund.*, **13**, 17 (1974).
12. N. M. Oleynikova and V. P. Dushchenko, *Heat Transfer-Soviet Rev.*, **8**, 133 (1976).
13. R. C. Progelhof, J. L. Throne, and R. R. Ruetsch, *Polym. Eng. Sci.*, **16**, 615 (1976).
14. Y. Agari and T. Uno, *J. Appl. Polym. Sci.*, **32**, 5705 (1986).
15. Y. Agari, A. Ueda, and S. Nagai, *J. Appl. Polym. Sci.*, **43**, 1117 (1991).
16. F. Lin and J. D. Ford, *Can. J. Chem. Eng.*, **62**, 125 (1984).
17. G. S. Bhatia, M.A.Sc. Thesis, University of Waterloo, 1990.
18. D. E. Kline, *J. Polym. Sci.*, **50**, 441 (1961).
19. K. W. Garrett and H. M. Rosenberg, *J. Phys. D Appl. Phys.*, **7**, 1247 (1974).
20. R. P. Kusy and R. D. Corneliusen, *Polym. Eng. Sci.*, **15**, 107 (1975).
21. D. Hansen and R. Tomkiewicz, *Polym. Eng. Sci.*, **15**, 353 (1975).
22. D. M. Bigg, *Polym. Eng. Sci.*, **19**, 1188 (1979).
23. D. L. Cullen, M. S. Zawojski, and A. I. Holbrook, *Plast. Eng.*, **44**, 37 (1988).
24. Y. Agari, A. Ueda, M. Tanaka, and S. Nagai, *J. Appl. Polym. Sci.*, **40**, 929 (1990).
25. Y. Agari, A. Ueda, and S. Nagai, *J. Appl. Polym. Sci.*, **42**, 1665 (1991).
26. Taylor, Aluminum Company Research Laboratories, experimental data in J. Delmonte's *Metal-Filled Plastics*, Reinhold, New York, 1961, p. 68.

Received August 7, 1992

Accepted January 14, 1993