

Thermal Conductivity of a Polymer Composite

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SYNOPSIS

A prediction equation for thermal conductivity of polymer composites reported in our previous papers has been revised in terms of two view points: (1) estimation of thermal conductivity of a composite using an idea of reduced thermal conductivity; and (2) the effect of ease in forming conductive filler chains on thermal conductivity is related to the *CVF* value in electric conductivity of the composite. The new equation was confirmed to be adaptable to thermal conductivities of varieties of polymer composite systems filled with spherical or irregular fillers. The equation was also considered to explain thermal conductivity of polymer composites filled with fibers. Further, it was found that thermal conductivities of fiber composites can be estimated by introducing a factor of the *CVF* value or aspect ratio (*L/D*) into the new equation. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

Many reports have been published on the measurement of thermal conductivity¹⁻¹⁹ and several thermal conductive models^{1,14,17,20-26} have been proposed for polymer composites. In previous papers, we compared some of these equations: Maxwell-Eucken's,²¹ Bruggeman's,²² Nielsen's,²⁶ Cheng and Vochon's,²⁵ and ours,¹ of which the last one showed the closest approximation to experimental data over a wide range of filler content. The equation, however, is still insufficient, since it includes a coefficient related to ease in forming conductive filler chains that is unexpectedly affected considerably by increasing filler thermal conductivity.

In this study, we improve the equation and propose a revision that overcomes the above defect. Its adaptability is then discussed using experimental data on over 40 types of polymer composites appearing in the literature,¹⁻¹⁹ including those of the present authors.^{1,3-6}

IMPROVEMENT OF PREDICTION EQUATION FOR THERMAL CONDUCTIVITY OF POLYMER COMPOSITES

We proposed eq. (1) for predicting thermal conductivity of a polymer composite in the previous report¹:

$$\log \lambda = V \cdot C_2 \cdot \log \lambda_2 + (1 - V) \cdot \log(C_1 \cdot \lambda_1) \quad (1)$$

where λ is the thermal conductivity of a composite; λ_1 , the thermal conductivity of a polymer; λ_2 , the thermal conductivity of fillers; V , the volume content of fillers; C_1 , the coefficient of the effect on crystallinity and crystal size of a polymer; and C_2 , the coefficient of ease in forming conductive chains of fillers.

However, this equation is insufficient, because C_2 increases significantly with increasing of λ_2 . Thus, we improve it basically as follows:

There are many types of composite systems in which polymer materials contain fillers. In those systems, thermal conductivity of the composite is the highest in the event that all fillers are gathered to form a conductive block and another block of polymer is arranged in parallel in the direction of

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thermal flux (parallel conduction) and is the lowest in the case where these blocks are arranged in a series in the direction of thermal flux (series conduction). Thermal conductivities of composites in the parallel and series conduction can be estimated, respectively, by the following equations:

Parallel conduction:

$$\lambda = V \cdot \lambda_2 + (1 - V) \cdot \lambda_1 \quad (2)$$

Series conduction:

$$1/\lambda = V/\lambda_2 + (1 - V)/\lambda_1 \quad (3)$$

We assume a generalization of eqs. (2) and (3) to the following eq. (4), where, if $n = 1$ or -1 , eq. (4) corresponds to eq. (2) or (3), respectively. n takes a value between 1 and -1 for each composite, depending on the geometry of filler and polymer in a composite:

$$\lambda^n = V \cdot (\lambda_2)^n + (1 - V) \cdot (\lambda_1)^n \quad (4)$$

To simplify the calculation by a scalar quantity, thermal conductivities of the composite and fillers were divided by that of the polymer. Then, λ/λ_1 and λ_2/λ_1 were defined as "reduced thermal conductivities" of the composite and the filler, respectively, and eq. (4) is rearranged to eq. (5):

$$(\lambda/\lambda_1)^n = V \cdot (\lambda_2/\lambda_1)^n + (1 - V) \cdot 1^n \quad (5)$$

However, eq. (3) was derived on the assumption that each polymer and filler made each block, and any difference in the geometrical factor (n) of the composite, polymer, or fillers was not considered. There are many types of composites with various compositions, where fillers are divided and dispersed in the polymer matrix, while the polymer matrix is the continuous phase with many holes. Since the ease in forming conductive filler chains is different depending on every dispersion system, the geometrical factors of fillers and polymer matrix are affected by its changing. Therefore, the index (n) of each element (composite, fillers, or polymer) is replaced by an index (n , $C_f \cdot n$ or $C_{pg} \cdot n$ respectively), and eq. (5) is converted into eq. (6):

$$(\lambda/\lambda_1)^n = V \cdot (\lambda_2/\lambda_1)^{C_f \cdot n} + (1 - V) \cdot 1^{C_{pg} \cdot n} \quad (6)$$

where $C_f (> 0)$ is a factor related to ease in forming conductive chains of fillers, while $1^{C_{pg} \cdot n} = 1$.

It has been reported²⁷ that, in the preparation of a composite, fillers can affect crystallinity and the crystal size of the polymer, resulting in changing the thermal conductivity. This effect is taken into consideration and λ_1 is replaced by $C_p \cdot \lambda_1$. Therefore, eq. (6) is converted into

$$\begin{aligned} [\lambda/(C_p \cdot \lambda_1)]^n \\ = V \cdot [\lambda_2/(C_p \cdot \lambda_1)]^{(C_f \cdot n)} + 1 - V \end{aligned} \quad (7)$$

If the dispersion state can be made uniform, n can be assumed to be in the neighborhood of 0, and λ^n may be approximated by $1 + n \cdot \log \lambda$.

Therefore, eq. (7) can be converted into the following equation:

$$\log[\lambda/(C_p \cdot \lambda_1)] = V \cdot C_f \cdot \log[\lambda_2/(C_p \cdot \lambda_1)] \quad (8)$$

Further, eq. (8) can be rearranged to

$$\log \lambda = A \cdot V + B \quad (9)$$

$$A = C_f \cdot \log[\lambda_2/(C_p \cdot \lambda_1)]$$

$$B = \log(C_p \cdot \lambda_1)$$

This equation means that logarithms of the thermal conductivities of composites are linearly related to the volume contents of fillers, as expressed by eq. (1), although the C_f value in eq. (9) is different from the C_2 value in eq. (1).

Critical volume fraction (CVF)²⁸ in electric conductivity is defined as the content of fillers at the inflection point of the S-shaped curve obtained by plotting logarithms of electric conductivities of polymer composite filled with conductive fillers, against the volume content of fillers. It is known^{28,29} that CVF decreases with increasing ease in forming conductive chains of fillers. Thus, it is considered that C_f increases with decreasing CVF , $0 < CVF < 1$ and $C_f > 0$. To satisfy these conditions, C_f is assumed to be represented as in eq. (10):

$$C_f = \log(1/CVF)$$

Consequently, by substituting eq. (10), eq. (8) can be rearranged to eq. (11):

$$\begin{aligned} \log[\lambda/(C_p \cdot \lambda_1)] \\ = V \cdot \log(1/CVF) \cdot \log[\lambda_2/(C_p \cdot \lambda_1)] \end{aligned} \quad (11)$$

Table I C_p and C_f , Obtained from Our Experimental Data

No.	Polymer	Fillers	λ_1^a	λ_2^b	C_p	C_f	CVF^c	Ref.
1	PE	Graphite	6.96	0.500	0.898	0.850	0.165	1
2	PE	Copper	6.96	0.946	1.073	0.857	0.146	1
3	PE	Al ₂ O ₃	6.96	0.079	0.859	0.865	—	1
4	PA	Graphite	4.60	0.500	1.017	0.905	0.131	1
5	PS	Graphite	3.33	0.500	1.024	0.874	0.155	1
6	PE	Al ₂ O ₃	6.8	0.080	1.03	0.718	—	3
7	PE	SiO ₂	6.8	0.023	1.00	0.722	—	3
8	PE	SiO ₂	6.8	0.013	1.00	0.838	—	3
9	PS	SiO ₂	3.9	0.023	1.06	0.887	—	3
10	PS	SiO ₂	3.9	0.013	1.06	0.987	—	3
11	PS ^d	PE	3.83	8.09×10^{-4}	0.970	0.823	—	6

^a Thermal conductivity of polymer ($\times 10^{-4}$ cal/s cm °C).

^b Thermal conductivity of fillers (cal/s cm °C).

^c Critical volume fraction (CVF) in electric conductivity of the composite.

^d Containing 2 wt % of SEBS block copolymer.

APPLICATION OF THE NEW MODEL TO EXPERIMENTAL DATA

1. Our Data on Thermal Conductivities of Polymer Composites

We found^{1,3,6} that logarithms of thermal conductivities of several types of polymer composites are linearly related to volume contents of fillers, over a wide range from low to superhigh content. This

means experimental confirmation of eq. (1). Therefore, since the above relation is maintained in all types of the dispersion systems, it is considered that eq. (9) is also adaptable for composites. Coefficients C_p and C_f were estimated by eq. (9), as indicated in Table I. All C_p were also approximately 1. C_f was plotted against logarithms of reduced thermal conductivity of fillers [$\log(\lambda_2/\lambda_1)$], as shown in Figure 1. All C_f were found around a constant value (= 0.85), which approximately corresponds to the

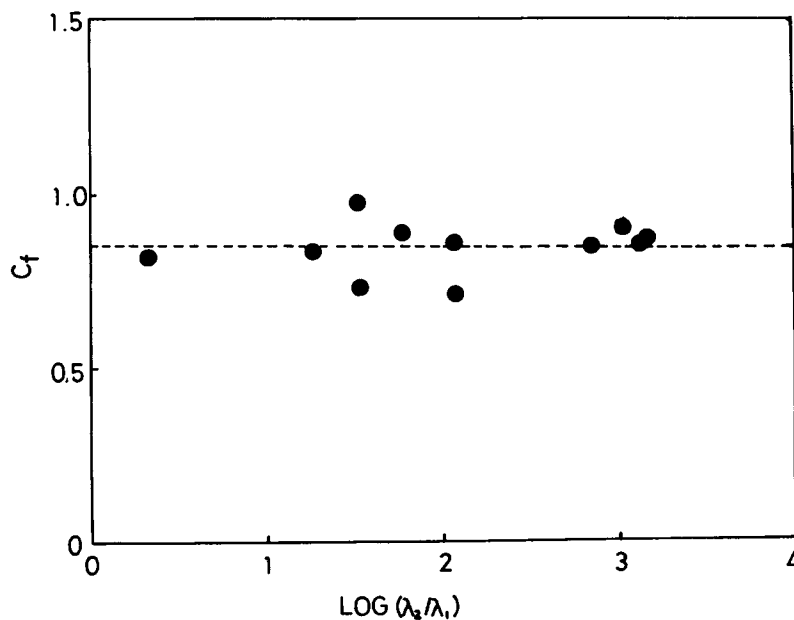


Figure 1 C_f , obtained from our experimental data on thermal conductivities of composites, against $\log(\lambda_2/\lambda_1)$.

logarithms of $1/CVF$ in these composite systems (no. 1–no. 5; 0.783–0.880). Therefore, it was confirmed that eq. (11) could explain the thermal conductivities of polymer composites.

2. On the Effects of Types of Dispersion for Formulating Polymer Composites

It was reported⁴ by us that thermal conductivities of composites are strongly affected by types of dispersion systems (melt mixture, roll-milled mixture, solution mixture, and powder mixture), although eq. (1) was adaptable to the thermal conductivity of composites independent of the dispersion systems. Thus, it meant that logarithms of thermal conductivities are linearly related with volume content of filler in any type of the dispersion systems and that eq. (9) is also adaptable to thermal conductivity of a composite of any type of dispersion system.

C_p and C_f in eq. (9) for polyethylene filled with graphite are shown in Table II. C_p varies slightly. Thus, it seems that crystallinity and the crystal size of the polymer are affected by fillers for this system. C_f 's are plotted against $\log(1/CVF)$, as shown in Figure 2. All C_f 's are approximately on a line, whose slope and intercept are 1 and 0, respectively. This result proves the relation of eq. (10). Therefore, eq. (11) was confirmed to be adaptable to thermal conductivities independent of dispersion systems.

3. Other Reported Data of Thermal Conductivities of Polymer Composites

Many other reports^{7–17} have been published on thermal conductivities of polymer composites filled with spherical- or irregular-shaped fillers. We evaluated the linear relationship between logarithms of thermal conductivities of these polymer composites

Table II C_p and C_f , Obtained from Our Experimental Data for Polyethylene Filled with Graphite

No.	λ_1^a	λ_2^b	C_p	C_f	CVF^c	Ref.
12	6.96	0.5	0.888	0.863	0.15	4
13	6.96	0.5	0.980	1.030	0.10	4
14	6.96	0.5	0.967	1.097	0.10	4
15	6.96	0.5	1.007	1.304	0.06	4

^a Thermal conductivity of polymer ($\times 10^{-4}$ cal/s cm °C).

^b Thermal conductivity of fillers (cal/s cm °C).

^c Critical volume fraction (CVF) in electric conductivity of the composite.

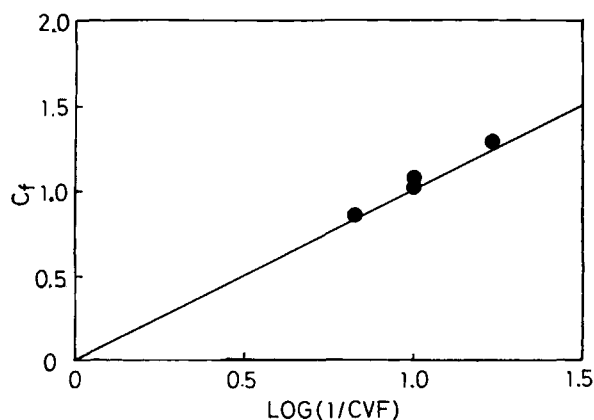


Figure 2 C_f , obtained from our experimental data on thermal conductivities for several dispersion systems, against $\log(1/CVF)$.

and the volume content of fillers, shown in the literature, by the method of least squares. Logarithms of thermal conductivity of those composites lie on a line against volume content of fillers^{7,8,17} as shown for typical examples in Figure 3. The correlation coefficients for linearities were all approximately 1, as shown in Table III. Thus, eq. (9) was considered

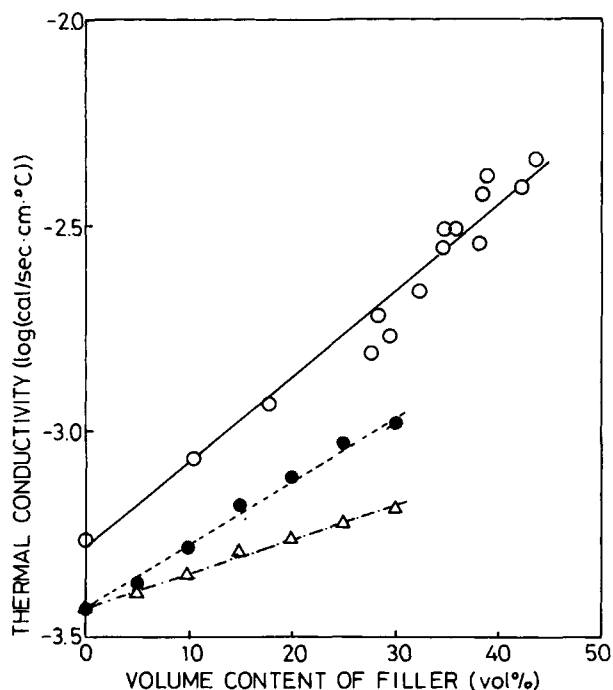


Figure 3 Logarithms of thermal conductivity in typical of composites: (O) poly(methyl methacrylate) filled with copper; (●) polystyrene filled with Al_2O_3 ; (Δ) polystyrene filled with glass.

generally adaptable to the thermal conductivities of these polymer composites.

C_p and C_f were estimated by eq. (9), as shown in Table IV. C_p stays almost constant, suggesting that the crystallinity and crystal size of polymers were not affected by fillers for these composites.

C_f varied depending on the variation of composite systems. This C_f variation must be caused by the difference of the slopes in eq. (9). Thus, Figure 4 shows the slopes (A) in eq. (9), plotted against $\log(\lambda_2/\lambda_1)$. Most of values of A deviated widely from the solid line, which was estimated by eq. (11) in the assumption that $CVF = 0.142$ (i.e., $C_f = 0.85$ for our dispersion systems cited above). This must have occurred because these dispersion systems, differing from ours, have values of A approximately between dotted lines for $CVF = 0.1$ and 0.4 .

There have been many reports²⁹ on electric conductivities of polymer composites with various values of CVF (0.1–0.4). We found³⁰ that the values of CVF differ depending on the dispersion types. It was reported that CVF varied widely with differences in surface energy of polymers, even if prepared by the same type of dispersion method. Thus, it was considered that the difference in dispersion systems for several types of composites changed the value of

A to result in varying of C_f . Therefore, eq. (11) can be considered to be adaptable, in general, to thermal conductivities of polymer composites.

4. The Effect of Aspect Ratio on Thermal Conductivities of Polymer Composites

It has been known^{17,20,23} that thermal conductivities of polymer composites are affected by filler shape and improved by increasing the aspect ratio (L/D) of fillers. In general, most of thermal conduction models have been organized by assuming a shape for fillers. For example, the Maxwell–Eucken model assumes that a spherical block, where all the fillers gather, exists at the center of a cubic block of the composite. On the other hand, eq. (11) did not define directly any effect of filler shapes. However, filler shape is expected to affect the ease of forming conductive filler chains, changing C_f in eq. (11). Thus, we considered introducing the effect of filler shape into C_f in the equation.

Our previous report⁵ proved that logarithms of thermal conductivities of composites filled with several types of carbon fibers were linearly related to the volume contents of fibers. Here, it was confirmed that fiber length did not decrease by breakdown

Table III Adoptability of Eq. (9) to Experimental Data Appearing in the Literature on Several Types of Composites

16	Epoxy resin	Silver	5.83	1.003	Spherical	0.995	8, 9, 11
17	Epoxy resin	Copper	5.83	0.915	Spherical	0.996	8, 9, 10
18	PMMA	Copper	5.40	0.871	$0.8 \times 0.8 \times 0.45$	0.978	17
19	Epoxy resin	Aluminum	3.63	0.525	Spherical	0.998	8, 12
20	Silicon rubber	Aluminum	4.51	0.525	Spherical	1.000	8, 13
21	Epoxy resin	Aluminum	5.40	0.525	Irregular	0.991	8, 15
22	Epoxy resin	SiO ₂	5.35	0.202	Irregular	0.998	8, 16
23	Epoxy resin	Tin	5.83	0.147	Irregular	0.997	8, 9
24	PE	MgO	8.01	0.131	Irregular	0.995	7, 8
25	PS	MgO	3.70	0.131	Irregular	0.990	7, 8
26	PE	Al ₂ O ₃	8.01	0.074	Irregular	0.995	7, 8
27	PS	Al ₂ O ₃	3.70	0.074	Irregular	0.995	7, 8
28	Epoxy resin	Al ₂ O ₃	5.25	0.073	Irregular	0.998	19
29	PMMA	Al ₂ O ₃	5.40	0.085	Spherical	0.991	17
30	Epoxy resin	St. steel ^c	5.83	0.039	Spherical	0.999	8, 9
31	PE	CaO	8.01	0.036	Irregular	0.999	7, 8
32	PS	CaO	3.70	0.036	Irregular	0.991	7, 8
33	PMMA	TiO ₂	4.93	0.017	Irregular	0.993	18
34	Epoxy resin	SiO ₂	5.25	0.015	Irregular	0.999	19
35	Epoxy resin	Glass	5.25	0.0021	Spherical	0.997	19
36	PE	Glass	8.01	0.0024	Irregular	0.998	7, 8
37	PS	Glass	3.70	0.0024	Irregular	0.998	7, 8

^a Thermal conductivity of polymer ($\times 10^{-4}$ cal/s cm °C).

^b Thermal conductivity of fillers (cal/s cm °C).

^c Stainless steel.

Table IV C_p and C_f , Obtained from Literature Data on Several Types of Composites

No.	Polymer	Fillers	$\log(\lambda_2/\lambda_1)^a$	A^b	C_p	C_f
16	Epoxy resin	Silver	3.24	1.220	1.002	0.379
17	Epoxy resin	Copper	3.20	1.400	0.933	0.434
18	PMMA	Copper	3.21	2.170	0.906	0.668
19	Epoxy resin	Aluminum	3.16	1.310	0.972	0.412
20	Silicon rubber	Aluminum	3.07	1.230	1.002	0.401
21	Epoxy resin	Aluminum	2.99	1.590	0.938	0.528
22	Epoxy resin	SiO ₂	2.58	1.410	0.977	0.543
23	Epoxy resin	Tin	2.40	0.964	1.041	0.404
24	PE	MgO	2.21	1.530	1.016	0.692
25	PS	MgO	2.55	1.480	1.029	0.582
26	PE	Al ₂ O ₃	1.97	1.520	1.007	0.772
27	PS	Al ₂ O ₃	2.30	1.880	1.014	0.819
28	Epoxy resin	Al ₂ O ₃	2.14	2.010	0.968	0.933
29	PMMA	Al ₂ O ₃	2.20	1.710	1.121	0.798
30	Epoxy resin	St. steel ^c	1.83	1.680	0.997	0.997
31	PE	CaO	1.65	1.400	0.980	0.842
32	PS	CaO	1.99	1.450	1.014	0.737
33	PMMA	TiO ₂	1.54	1.630	0.981	1.053
34	Epoxy resin	SiO ₂	1.46	1.230	0.988	0.840
35	Epoxy resin	Glass	0.60	0.638	1.004	1.062
36	PE	Glass	0.48	0.592	1.000	1.242
37	PS	Glass	0.81	0.825	1.007	1.019

^a λ_1 : thermal conductivity of polymer, λ_2 : thermal conductivity of fillers.

^b Value of slope (A) in eq. (9).

^c Stainless steel.

during preparation of specimens. Thus, eq. (9) was considered to be adaptable to thermal conductivities of such composites.

Since direct measurement of thermal conductivity of carbon fiber was difficult, the conductivity hitherto reported has a broad range from 1.2 to 4.8×10^{-2}

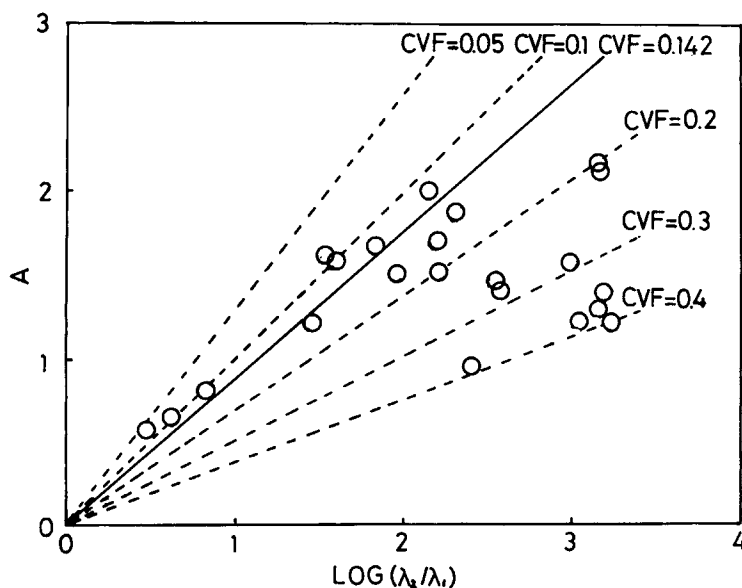


Figure 4 Slopes (A) in eq. (9) from literature data on thermal conductivities of composites, against $\log(\lambda_2/\lambda_1)$.

Table V C_p and C_f , Obtained from Our Experimental Data on Polyethylene Filled with Carbon Fiber

No.	L_n/D^a	L_w/D^b	λ_1^c	λ_2^d	C_p	C_f	CVF^e	Ref.
38	1.0	1.0	6.8	1.2-4.8	0.974	0.521	0.301	5
39	4.9	6.0	6.8	1.2-4.8	1.063	0.696	0.152	5
40	13.4	21.6	6.8	1.2-4.8	1.066	0.919	0.046	5
41	21.8	45.3	6.8	1.2-4.8	0.976	1.070	0.016	5

^a Aspect ratio of number-average length to diameter of a fiber.

^b Aspect ratio of weight-average length to diameter of a fiber.

^c Thermal conductivity of polymer ($\times 10^{-4}$ cal/s cm $^{\circ}$ C).

^d Thermal conductivity of fillers ($\times 10^{-2}$ cal/s cm $^{\circ}$ C).

^e Critical volume fraction (CVF) for electric conductivity of the composite.

cal/s cm $^{\circ}$ C.³¹ Here, it is possible to calculate the value by using eq. (11) for thermal conductivity of composite no. 38 ($L/D = 1$). Thus, the thermal conductivity of the carbon fiber was estimated as 3.8×10^{-2} cal/s cm $^{\circ}$ C, by substituting the CVF ($= 0.301$) and C_p ($= 0.974$) for composite no. 38 into eq. (11). This value falls within the range of the above reported data. Further, using this value (3.8×10^{-2} cal/s cm $^{\circ}$ C), several C_p and C_f values for other L/D 's (nos. 39-41) were estimated by eq. (9), as shown in Table V. C_p 's were approximately 1, and it was considered that crystallinity and the crystal size of polymers are almost unaffected by the L/D of fillers. In contrast, C_f increased considerably with increasing fiber length. This occurs because the increase in fiber length favors ease of forming conductive fiber chains.

C_f was plotted against $\log(1/CVF)$ for several types of carbon fiber composites, as shown in Figure

5. All data do not lie on the dotted line, whose slope and intercept are 1 and 0, respectively, but approximately on the solid line, whose slope is approximately 0.5. This occurred because the aspect ratio of fibers increases electric conductivity more effectively than it increases the thermal conductivity of a composite. Thus, for thermal conductivity of a composite, the effect of decreasing CVF by increasing the aspect ratio should be lower than by improving the ease of forming conductive filler chains, i.e., the dispersion state. Then, eq. (11) can be modified to eq. (12) for fiber composites:

$$\begin{aligned} \log(\lambda/(C_p \cdot \lambda_1)) &= V \cdot C_f \cdot \log[\lambda_2/(C_p \cdot \lambda_1)] \\ C_f &= C \cdot \log(1/CVF) + (1 - C) \cdot C_{f1} \\ C_{f1} &= \log(1/CVF_1), \quad C = 0.5 \end{aligned} \quad (12)$$

where CVF_1 and C_{f1} are defined as CVF and C_f , respectively, for $L/D = 1$.

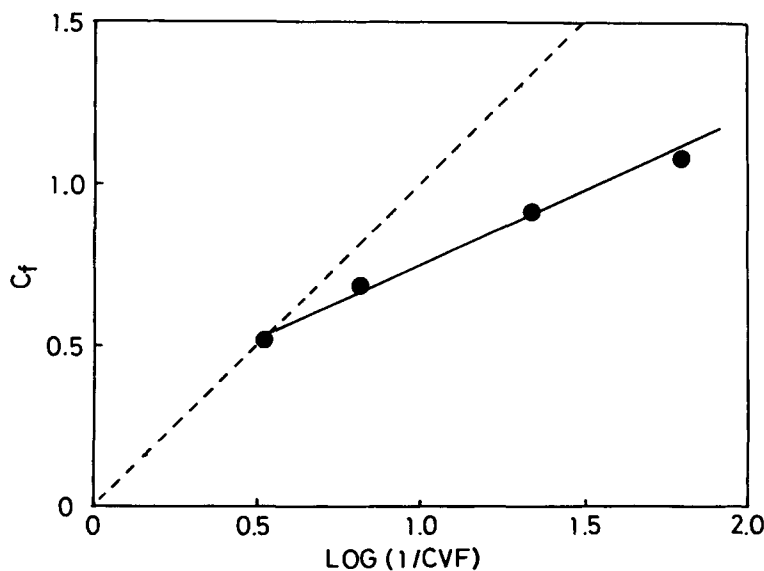


Figure 5 C_f , obtained from our experimental data on thermal conductivities of fiber composites, against $\log(1/CVF)$.

Table VI C_p and C_f for Experimental Data Appearing in the Literature on Several Types of Fiber Composites

No.	Polymer	Fillers	λ_1^a	λ_2^b	Aspect Ratio ^c	Correlation Coefficient	C_p	C_f	Ref.
42	PMMA	Copper	5.4	0.871	5.7	0.986	1.034	0.860	17
43	PP	Aluminum	6.21	0.525	12.5	0.983	0.944	1.080	8

^a Thermal conductivity of polymer ($\times 10^{-4}$ cal/s cm °C).

^b Thermal conductivity of fillers (cal/s cm °C).

^c Aspect ratio (L/D) of a fiber.

For other data^{8,17} on polymer composites filled with fibers having two differing aspect ratios, we evaluated the linearities between the logarithms of thermal conductivity and the volume content of fibers by the method of least squares. As shown in Table VI, the correlation coefficients of linearity were all approximately 1. Thus, eq. (9) was confirmed to be adaptable to the thermal conductivities of polymer composite systems filled with differently shaped fillers, i.e., fibers.

C_p and C_f for the above data were also estimated by eq. (9) and are shown in Table VI. C_p 's were approximately 1; hence, the thermal conductivities of these polymers were not affected by fillers. C_f in-

creased significantly with increasing aspect ratio of fillers as with the carbon fiber composite. The deviation of C_f in the other data^{8,17} from that in the carbon fiber composites might occur not only because differences in dispersion systems, but also because of the breakdown of fibers during the preparation of specimens.

Further, C_f 's were plotted against $\log(L_n/D)$ and $\log(L_w/D)$ for multiple data on thermal conductivities of the fiber composites (Fig. 6). C_f 's derived from the experimental data of other are close to our data plotted against $\log(L_n/D)$. Thus, $\log(L_n/D)$ was considered more suitable for representing $\log(L/D)$ than was $\log(L_w/D)$. This result is in contrast to the case of electric conductivity of composites.³² It probably occurred because the effect of longer fibers in filled composites on thermal conductivity was weaker than that on electric conductivity of composites.

Figure 7 shows $\log(C_f - C_{f1})$ plotted against $\log[\log(L/D)]$. All data points for each case of L_n/D or L_w/D lie on a straight line and can be represented as eq. (13):

$$\log(C_f - C_{f1}) = E \cdot \log[\log(L/D)] + F \quad (13)$$

where $E = 1.72$ and $F = -0.482$ for L_n/D , while $E = 1.52$ and $F = -0.591$ for L_w/D .

Thus, eq. (11) can be converted into eq. (14) by substituting eq. (13) into eq. (11):

$$\begin{aligned} \log[\lambda/(C_p \cdot \lambda_1)] &= V \cdot C_f \cdot \log[\lambda_2/(C_p \cdot \lambda_1)] \\ C_f &= 10^F \cdot \log(L/D)^E + C_{f1} \end{aligned} \quad (14)$$

Therefore, thermal conductivities of fiber composites are thought to be estimated by substituting C_{f1} (C_f for $L/D = 1$) and CVF values or the aspect ratio (L/D) into eq. (12) or (14), respectively.

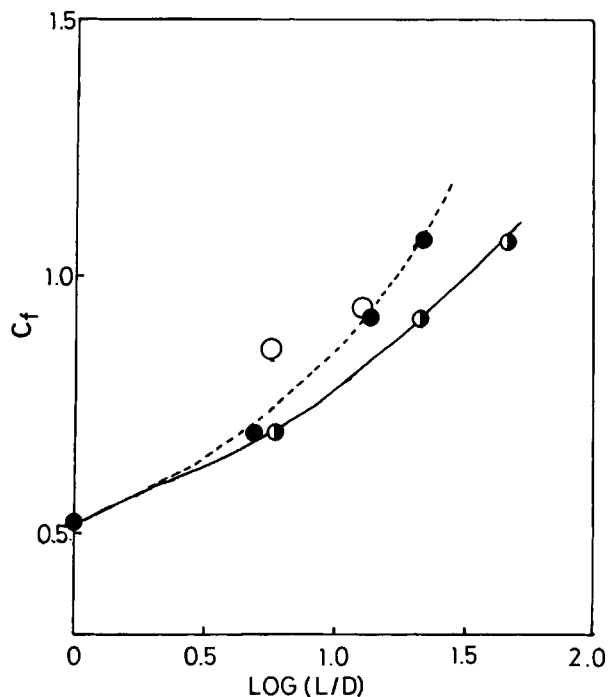


Figure 6 C_f against $\log(L/D)$: (●) our data against $\log(L_n/D)$; (⊙) our data against $\log(L_w/D)$; (○) literature data.

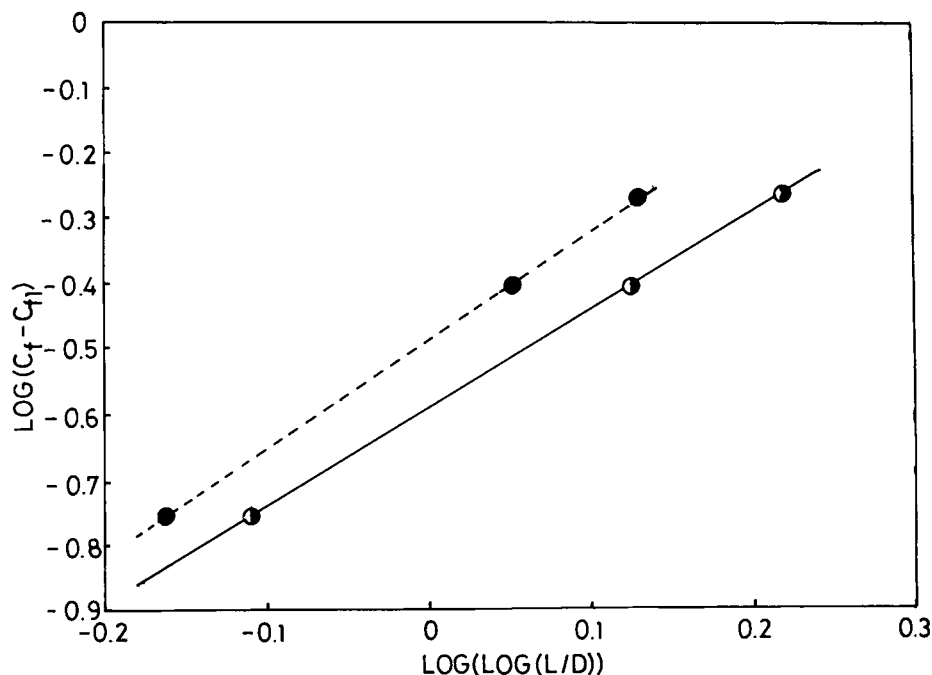


Figure 7 $\log(C_f - C_{f1})$ against $\log[\log(L/D)]$: (●) our data against $\log(L_n/D)$; (○) our data against $\log(L_w/D)$.

CONCLUSION

An equation previously proposed by the present authors for predicting thermal conductivity of polymer composites had a deficiency in that the coefficient of ease in forming conductive filler chains increases largely with increasing thermal conductivity of fillers. In this study, we improved it basically to a new prediction equation, which was derived based on two viewpoints: One is to estimate thermal conductivity of composites, using an idea of reduced thermal conductivity. Another is to relate the effect of the ease in forming conductive filler chains on thermal conductivity, with the *CVF* value for electric conductivity of the composite.

The new equation was confirmed to be adaptable to thermal conductivities of over 36 types of polymer composite systems filled with spherical- or irregular-shaped fillers, which were reported by us and others. Then, it was found that the difference of dispersion state of fillers strongly affects the thermal conductivity of the composite, resulting in varying C_f in the new equation.

Further, the new equation was confirmed to be adaptable to thermal conductivity of polymer composites filled with fibers, which were reported by us and others, by introducing the factors of C_{f1} (C_f for $L/D = 1$) and either the *CVF* value or L/D .

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