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1. Abstract

To study the increase in the thermal conductivity of polymers embedded with conductive fillers, two fillers, BN particles and Al wool fibers, are blended with two different polymers in various volume percentage fractions. As such, the effect of filler type and volume fraction is observed on the thermal conductivity enhancement of the polymer composite. The thermal conductivity is shown to rise most dramatically with Al wool but reaches its greatest magnitude using BN particles. For these material combinations, the increase in volume fraction of conductive filler is observed to be more significant than polymer type in the enhancement of thermal conductivity.

2. Introduction

In the design of reliable products in the computer industry, heat transfer plays a critical role and tends to limit the technical advances made in electronics. As electronic components become faster, they generate more heat in smaller confines of space causing significant design constraints unless adequate thermal solutions are perceived and is the motivation for this study in polymer composite thermal conductivities.

At the Engineering Design Research Center (EDRC), the process known as Shape Deposition Manufacturing (SDM), allows several materials to be deposited in layers to ultimately form a part. Applied to portable, wearable computers, which need to be nigged, small, and power-efficient, SDM can be used to embed the heat-generating electronics inside a thermally conductive polymer composite substrate. This has been shown to be very effective as a thermal design for both transient and steady-state results (Egan and Amon, 1996). It has been shown that adding conductive fillers to polymers can yield polymer composites whose thermal conductivities are much higher than those of the polymer (Bujard, 1988). In this study, a simple experimental method using the principle of inverse engineering is used to approximate the conductivity of several polymer composites. Two polymers and two fillers demonstrate the importance of type and volume percentage of filler on the thermal conductivity of the composite.

3. **Material** Combinations

The two polymers are Epon 815 Resin and Epi-Cure 3290 Curing Agent from Shell, Inc. and LUC-4180 Urethane Elastomer from Ad-Tech. The former was proposed by Advanced Ceramics for its mixing characteristics with Tillers, while the later is known for its excellent machinability and is currently being used in the Frogman, a wearable computer designed and manufactured at Carnegie Mellon.

Two fillers are selected based for their extreme differences. The first is Polartherm 670, previously HCJ-48, from Advanced Ceramics. It is a BN powder having a mean particle diameter of 225 |im. The second is Al wool and consists of a fine mesh of very thin hair-like strands of aluminum. Thus, the first is a thermally conductive, electrically insulative ceramic particle, whereas the second conducts both thermally and electrically and consists of continuous fibers. As will be seen, these two fillers behave very differently when mixed with their respective polymers.

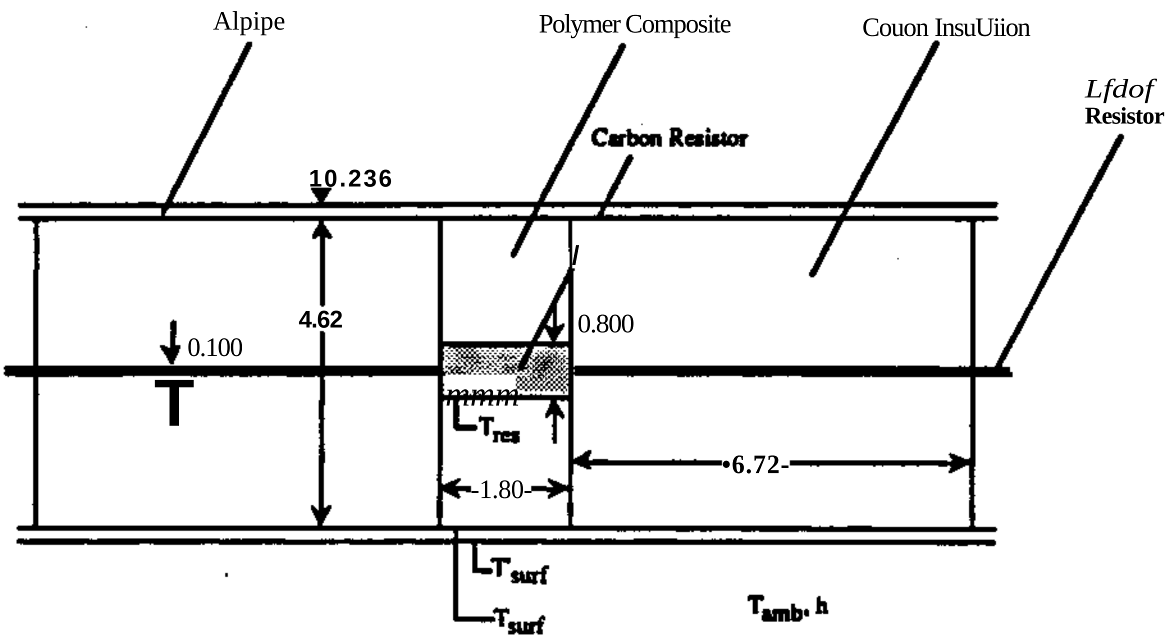
4. **Experimental** Apparatus

To approximate the thermal conductivity of polymer composites, the mixture is poured into a cylindrical tube of aluminum with a carbon resistor embedded in the center producing 1.5 W of heat. With proper insulation, such an experimental specimen can approximate one dimensional radial heat transfer. By comparing experimental data with numerical simulation results, the conductivity of the polymer composite may be approximated by predicting the radial heat flow through the composite. Table 1 shows the material properties used both in the analytical treatment of the heat transfer problem as well as in the numerical simulations. Property values are taken from *Fundamentals of Heat Transfer,* Incropera and DeWitt. A schematic of the experimental specimen is shown in Figure 1.

Table 1 Material Properties of Experimental Specimen

Material	k (W/m K)	o (ks/m^3)	C_B (J/k* K)
6061 Al	170	2640	903
Polymer Composite	7<n	?<2)	?<*>
Cotton Insulation	7<*>	80	1300
Carbon Resistor	1.6	...	...
Tin Lead	66	—	...

Note: (1) 0.1 < k...,, < 4.0 Wfai K
(2) 1000 < p_{eomp} < 1500 kg/m3
(3) Cp = 800 J/kg K (approx.)
(4) k... = 0.06 W/m K (approx.)



(all dimensions in cm)

Fig. 1 Experimental specimen used to approximate thermal conductivity of polymer composites.

5. Methodology for Thermal Conductivity Characterization

To approximate the thermal conductivity of the polymer composite, it is initially assumed that all heat generated by the resistor flows radially outward and is dissipated from the Al tube through natural convection and radiation exchange. Using this assumption, the thermal conductivity of the polymer is first approximated using the following equation:

$$k_{comp} = \frac{\frac{1}{4/r} q_r \left(\frac{M}{R_{res}} \quad \frac{I}{R_{tube}} \right)}{\Delta T_{res} - \Delta T_{surf}} \qquad (1)$$

where k_{comp} = thermal conductivity of polymer composite
q_r = portion of heat transferred radially through polymer composite to Al tube
R_{ret} = radius of carbon resistor
R_{tube} = radius of tube
ΔT_{surf} = temperature rise above ambient at surface of Al tube
ΔT_{m} = temperature rise above ambient at surface of resistor

Since AT_{furf} and AT^ are experimentally determined, only q_r needs to be known. But this would lead to applying the assumption that the heat flow is truly one dimensional. Because some heat does escape into the insulation, numerical simulations are conducted until the temperatures obtained from the simulations converge to those of the experimental data. The spectral element method is the numerical technique underlying the simulations and is a hybrid of two techniques: the finite clement method and the spectral technique.

The numerical simulations predict the steady-state temperature Held for a conduction-only domain where h and T_{amb} satisfy the boundary conditions. The heat transfer coefficient, h, specifics the energy release from the Al tube due to both natural convection and radiation. Using experimental data such as AT_{surf} and q_{tot}, h is determined through an energy balance.

To conduct the numerical simulations, k_{cot} must also be ascertained. The conductivity of cotton is taken from Incropcra and DcWitt and is assumed to be 0.06 W/m K. A sensitivity analysis of this assumption is performed to estimate the effect of k_{cot} on thermal phenomena of the specimens. Using a previous experimental result with a polymer of known thermal conductivity, k_{col} was varied from 0.065 to 0.300 W/m K in the numerical simulations with the result that AT_{re}, changed by less than 8%. This shows that the temperature field is not too sensitive to the thermal conductivity of the insulator as long as k_{int} is less than k_{corop} by a factor of 3 or more.

Using k_{eoa}, from (1) as the initial estimate and with all other variables known, kco.p i^s varied in the numerical simulations until the temperature field agrees with the experimental results.

6. Assumptions

Because of the simplicity of the experimental apparatus, certain assumptions need to be made and are listed below.

1. Negligible heat flow through insulated ends but some heat flow into insulation
2. Al tube is isothermal both in radial and axial directions
3. Negligible contact resistance at the thermocouple interfaces
4. Power generation is steady and exactly 1.5 W
5. Thermal conductivity of cotton insulation is 0.06 W/m K for all specimens
6. Carbon resistor is placed accurately in center of specimen
7. All voids between filler particles is occupied by the polymer

7. Types of Experiments and Experimental Data

In all, 10 experiments are conducted. Two experiments determine the thermal conductivity of the two polymers, Epon 8IS Resin with Epi-Curc 3290 Curing Agent (Epon) and LUC-4180 Urethane Elastomer (LUC). With Epon, BN is used as the conductive filler and is varied in volume percentages of 10, 20 and 30 percent. Two conductive fillers are used with LUC, BN and Al wool. The BN is varied in volume percentages of 20 and 30 percent, while the Al wool is varied in volume percentages of 0.78, 1.S5, and 2.33 percent. The experimental data is presented in Table 2.

Table 2 Experimental Data of Epon and LUC Polymer Composite Specimens

Polymer	Volume Percentage of Conductive Filler	AT^CC)	AT^. (°C)
Epon	(none)	4.87	57.05
"	10% BN	4.76	42.23
"	20% BN	5.19	22.48
"	30% BN	5.36	20.19
LUC	(none)	5.16	69.89
"	20% BN	5.72	35.09
"	30% BN	6.15	27.45
"	0.78% Al wool	5.97	51.99
"	1.55% Al wool	5.90	53.84
"	2.33% Al wool	5.11	35.36

Using an energy balance, the average value of h can be determined from the following equation.

$$q_{tot} = h\ A_{surf}\ \Delta T \wedge \quad (2)$$

The heat transfer coefficient for each experiment is shown in Table 3.

Table 3 **Heat** Transfer Coefficients Along Al Tube

Experiment	h (W/m^2 K)
Epon with 0% BN	12.63
Epon with 10% BN	12.93
Epon with 20% BN	11.86
Epon with 30% BN	11.48
LUC with 0% BN	11.92
LUC with 20% BN	10.76
LUC with 30% BN	10.00
LUC with 0.78% Al wool	10.31
LUC with 1,55% Al wool	10.42
LUC with 2.33% Al wool	12.04

8. **Results from** Numerical Simulations

An iterative method approximates the thermal conductivity of each composite using numerical simulations to match experimental temperature results. The initial guess for k_{comp} is given by (1). With all other variables held constant, k_{comp} is manipulated until AT_{ret} and AT_{iurf} closely agree with the experimental data. The numerical simulation results are presented in Table 4.

Table 4 Results Showing Iterative Refinement of $k_{co \cdot F}$

Polymer Composite	k (W/m K)	AT«,I,. CO	AT«J.. CO
Epon with no filler	.30	62.99	4.77
ATJ_ = 57.05 °C	.35	56.12	4.77
AT.J.,, = 4.87 «C	.32	60.04	4.77
Epon with 10% BN	0.44	47.03	4.68
AT J^ = 42.23 *C	0.50	42.60	4.69
AT«rfU = 4.76°C	0.51	41.95	4.69
Epon with 20% BN	1.03	24.79	5.11
AT J^ = 22.48 *C	1.10	23.61	5.11
AT.J., = 5.19 °C	1.20	22.15	5.11
Epon with 30% BN	1.25	21.67	5.27
ATJ^ = 20.19 °C	1.40	20.00	5.27
$AT_{wrf}L_{ip}$ = 5.36-C	1.36	20.41	5.28
LUC with no filler	0.24	74.40	5.01
AT J^ = 69.89 °C	0.30	63.26	5.03
AT.J^ = 5.16 °C	0.27	68.34	5.02
LUC with 20% BN	0.63	36.47	5.57
ATJ^ = 35.09 •C	0.70	33.67	5.58
AT^^ = 5.72 °C	0.66	35.20	5.57
LUC with 30% BN	0.90	28.29	5.98
ATJ^, = 27.45 °C	1.00	26.21	5.98
AT^^, = 6.15 °C	0.93	27.62	5.98
LUC with 0.78% Al wool	0.37	54.79	5.77
ATJ^, = 51.99 °C	0.45	47.31	5.78
AT.J..P = 5.97 »C	0.40	51.69	5.77
LUC with 1.55% Al wool	0.39	52.62	5.71
ATJ«^ = 53.84 °C	0.35	57.05	5.71
AT,^ = 5.90 °C	0.38	53.65	5.71
LUC with 2.33% Al wool	0.61	36.82	5.02
AT J^ = 35.36 °C	0.64	35.49	5.02
AT.J_ = 5.11«C			

Table S shows the best approximation of the thermal conductivity of the polymer composites using this iterative technique as well as the level of conductivity enhancement.

Table 5 Thermal Conductivity Approximations of the Polymer Composites

Polymer Composite	kc-p (WAn K)	k_{comp}/k_{poly}
Epon with no filler	0.34	1.00
Epon with 10% BN	0.51	1.50
Epon with 20% BN	1.18	3.47
Epon with 30% BN	1.38	4.06
LUC with no Tiller	0.26	1.00
LUC with 20% BN	0.66	2.54
LUC with 30% BN	0.93	3.58
LUC with 0.78% Al wool	0.40	1.54
LUC with 1.55% Al wool	0.38	1.46
LUC with 2.33% Al wool	0.64	2.46

A graphical representation of this compilation is shown in Figure 2 below.

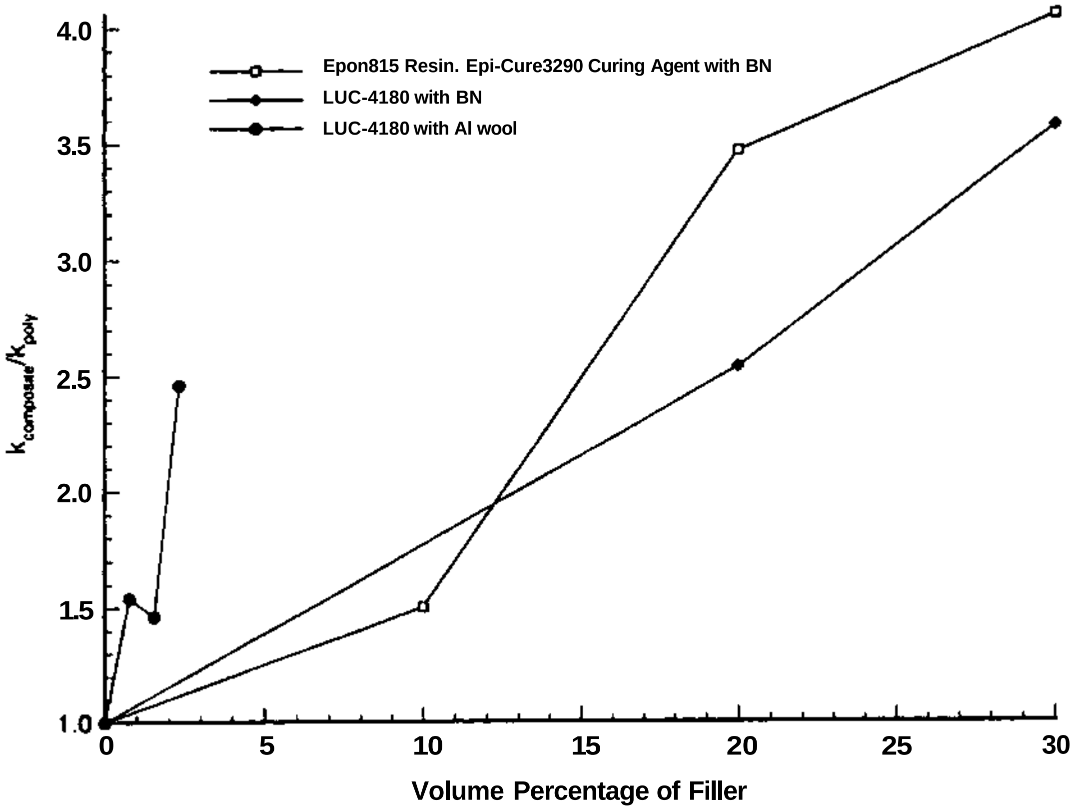


Fig. 2 Thermal conductivity enhancement due to addition of conductive fillers

Figure 2 shows the dramatic increase in thermal conductivity with increased volume fraction of conductive fillers. This trend has been extensively supported in the literature (Bigg, 1986). Although it appears that the conductivity of the Al wool sample increases at a faster rate than the others, it must be realized that the 2.33 volume percentage packing is actually quite dense, and higher packing ratios may be impractical. However, it does show that the Al wool allows the heat to propagate through a continuous weave of strands, creating heat flow paths through the polymer, thus dramatically increasing the thermal conductivity of the composite.

An interesting note is that a rough estimate of the thermal conductivity of a polymer composite may be made by simply knowing the thermal conductivity and volume fractions of the constituent components. To find the possible range of conductivities expected from the mixture of conductive filler and polymer, a thermal resistance approach is used. The upper and lower limits of the conductivity of the composite depend on the configuration of the materials with respect to the direction of the heat flow. In the case for the upper limit of the conductivity, the filler particles conglomerate in a direction parallel to the heat flow allowing the heat to bypass the polymer. In the other extreme, the filler particles form a layer across the composite perpendicular to the heat flow.

The parallel resistance network provides the upper limit of the conductivity of the composite where most of the heat passes through the material of higher conductivity. For a composite of two materials a and b, its formulation takes the form: kcoap = v_gk, + $v_b k_b$, where v is the volume fraction of the constituent materials and k, the conductivity. The volume fraction is used as a basis for measuring the relative width of the each layer of the individual materials.

The series resistance network, on the other hand, places a lower bound on the conductivity. Since the heat must flow through both materials, the less conductive material has its largest possible effect on the overall conductivity in this configuration, causing the lower limit of the conductivity of the composite to be ascertained. The series formulation takes the form of:
keep = k.kb/Cv.kb + v_bk.).

To test whether the series or parallel configuration is more dominant, a linear combination of the form: k_{comp} = a k_{tcriM} + (J $k_{pgr \cdot lu}$, is assumed to hold for all possible values of k_{eOBp}. As a result, the coefficients indicate the nature of the configuration, yielding an implication of the aggregate particle dispersion in each specimen. Figure 3 shows the series weighting factor for each experiment. Evidently, the filler behaves as if it were in the series configuration. This result, however, may indicate that significant contact resistances or voids exist between the polymer and the filler since visual observation suggests a high degree of parallel distribution in the Epon specimens, for example.

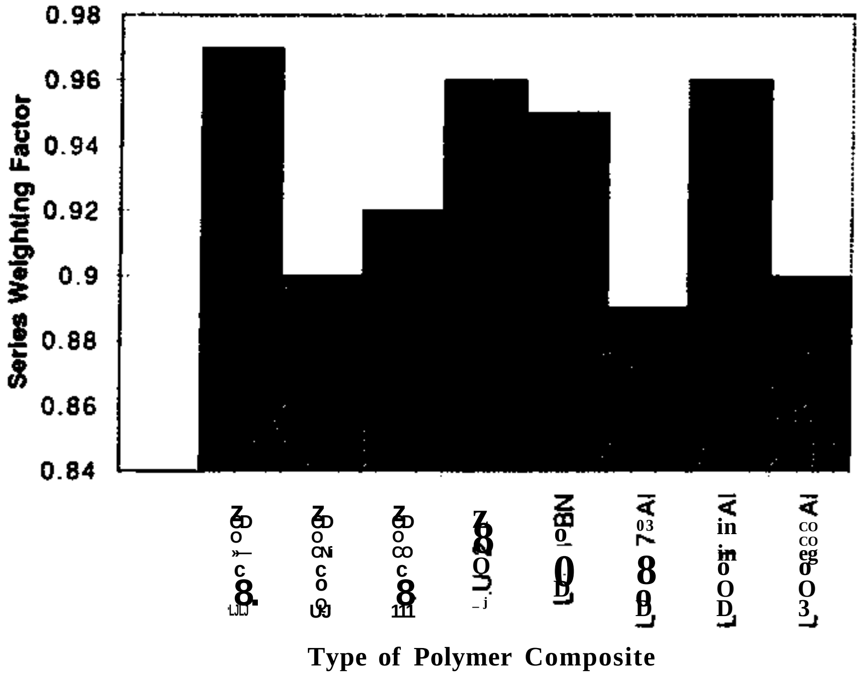


Fig. 3 Particle Dispersion Characteristics of **the Polymer** Composite Specimens

9. Conclusions

It is interesting that regardless of the polymer, filler, or volume percentage of filler, the scries approximation is very close to the predicted value of thermal conductivity. As volume percentage increases, the particle dispersion seems to become more parallel, causing an even greater increase in the thermal conductivity than the increase in volume percentage would by itself predict.

For application purposes, the LUC-4180 is the superior polymer, not necessarily for its thermal properties but because it is far less of health hazard than the Epon 815 Resin and hardens more quickly, allowing a more uniform dispersion of the BN to develop due to a smaller amount of paniculate settling.

Although the Al wool has excellent thermal characteristics, it is electrically conductive as well, a negative property for embedded electronics applications. The BN is not electrically conductive and can more easily be blended into the polymer. It is, therefore, a very good conductive filler, and its enhancement effect may be increased by creating continuous strands parallel to the heat flow.

In this study, it has been attempted to account for every possible source of error due to the simplicity of the experimental apparatus. Therefore, a high degree of emphasis is placed on the assumptions. First, the assumption that no heat escapes from the outer ends of the aluminum cylinder seems to be accurate due to the close correlation between $AT_{,ttrf}l._{,p}$ and $AT_{ilirf}l.._{f..}$ Second, the approach of starting the iterative procedure with an analytical approximation and then refining with two dimensional numerical simulations does not require the assumption of one dimensional radial heat flow, increasing the accuracy of the thermal conductivity characterization.

Some of the largest sources of error is the assumption that the resistor is exactly in the center of the Al cylinder, the actual composition of the polymer composite samples, and the assumption regarding the conductivity of the cotton insulation. Although, these errors, if combined, may be significant, it is thought that the accuracy of the experiments is accurate enough to characterize the thermal conductivities with reasonable certainty.

10. Acknowledgments

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