Thermal Conductivity of Particulate-Filled Polymers

Sundstrom and Lee[1] recently published in this journal data on the thermal conductivity of polystyrene and polyethylene containing several types of fillers. Their data tended to follow the theoretical equation of Bruggeman[2] at low concentrations of filler and the theoretical equation of Cheng and Vachon[3] at higher concentrations. I wish to point out that the data over the entire concentration range can be fitted better by the Kerner equation[4] or the Halpin-Tsai equations[5,6] as modified by Lewis and Nielsen.[7-9]

The modified equations for thermal conductivity (or electrical conductivity or elastic moduli also) are:

$$\frac{k}{k_1} = \frac{1 + AB\phi_2}{1 - B\psi\phi_2} \tag{1}$$

$$A = k_E - 1; \; B = \frac{k_2/k_1 - 1}{k_2/k_1 + A} \tag{2}$$

$$\psi \doteq 1 + [(1 - \phi_m)/\phi_m^2]\phi_2. \tag{3}$$

In these equations, k is the thermal conductivity of the composite, k_1 and k_2 are the thermal conductivities of the polymer and filler, respectively, A is a constant related to the generalized Einstein coefficient k_E, B is a constant related to the relative conductivity of the components, ψ is a function related to the maximum packing fraction ϕ_m of the filler, and ϕ_2 is the volume fraction of the filler. The Einstein coefficient k_E has been published for a number of suspensions; for instance, $k_E = 2.5$ for rigid spheres suspended in a medium with a Poisson ratio[9] of 0.5. The maximum packing fraction ϕ_m is the ratio of the density of the filler material to the maximum density of the bulk powder.

Data on four systems covering the extremes studied by Sundstrom and Lee are shown in Figures 1 and 2 along with the predictions of eqs. (1) to (3). The value of A for glass spheres is 1.5 ($k_E = 2.5$). The exact value of A for MgO powder is not known, but a value of $A = 3$ fits the data well and is about what would be expected for irregularly shaped particles. Unagglomerated cubic particles (rock salt) have been shown to have a value[10] of $A \doteq 2$. More irregularly shaped particles, especially if somewhat agglomerated, should have a value of A greater than 2. The exact value of the packing fraction ϕ_m also is not known, but a value of 0.64 was assumed. This is the ϕ_m value for random close packing of spheres, and many other powders have a similar value. The theoretical curves, as determined from eqs. (1) to (3), agree quite well in general

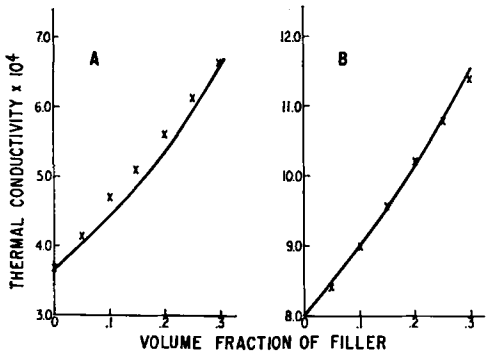

Fig. 1. Thermal conductivity (cal/sec cm °C). Solid lines are theoretical; (X) experimental points of Sundstrom and Lee; (A) glass beads in polystyrene; (B) glass beads in polyethylene.

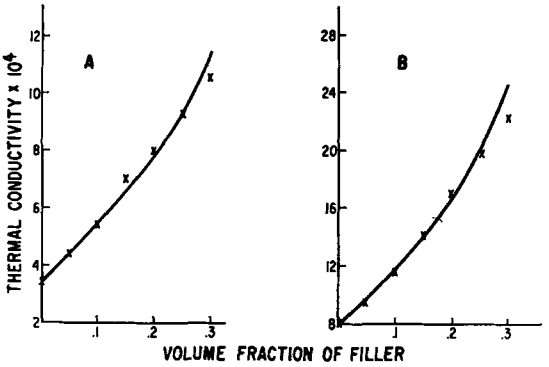

Fig. 2. Thermal conductivity (cal/sec cm °C). Solid lines are theoretical; (X) experimental points of Sundstrom and Lee; (A) MgO in polystyrene; (B) MgO in polyethylene.

with the experimental results. The fit throughout the entire concentration range is generally better than that shown in the paper of Sundstrom and Lee where other theoretical curves were tested. Since the actual experimental values of the Einstein coefficient and the packing fractions were not available, "reasonable" values were assumed. No attempt was made to obtain better fit by optimizing the values for k_E and ϕ_m. The important point, however, is that the equations proposed here generally fit the experimental data better than other equations. Although Sundstrom and Lee[1] give no data on metal-filled polymers, the equations do fit such data very well as shown in reference 9.

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