MECHANICAL AND THERMAL PROPERTIES OF METAL-PARTICULATE FILLED POLYMERS

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The effect of metallic fillers on thermal conductivity and mechanical properties of High Density Polyethylene is investigated experimentally and compared with the existing theoretical and empirical models. Thermal conductivities of samples produced by the addition of various volumetric percentages of Al,Cu and Pb particles in HDPE matrix are measured with hot-wire technique and these experimental values are compared with the calculated ones from Maxwell and Lewis & Nielsen models.Concerning the mechanical properties of metal filled polymers, a brief literature survey is made; tensile test results of HDPE filled with various volume fractions of Al powder are given.

INTRODUCTION

Polymers are often processed with the addition of various kinds of fillers and additives, in order to obtain final products of desired physical properties. Some of the reasons for the use of such fillers are:
 a) To improve thermal conductivity and diffusivity,
 b) To have a certain degree of electrical conductivity and magnetic permeability,
 c) To stiffen the matrix and make it more rigid,
 d) To reduce the internal stresses in injection molded parts,
 e) To reduce creep,
 f) To get a better appearance of the final product.

To make an adequate use of filled polymers the variation of physical properties with the kind and percentage of filler materials must be known.

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THERMAL CONDUCTIVITY OF METAL FILLED POLYMERS

Theoretical Models

Numerous theoretical and empirical models predicting thermal conductivities of mixtures of polymers and fillers are found in the literature (1).

Using potential theory Maxwell(1) obtained a simple relationship for the conductivity of randomly distributed and non-interacting homogeneous spheres in a homogeneous continuous medium:

$$k_c = k_p \frac{k_m + 2.k_p + 2.\phi.(k_m - k_p)}{k_m + 2.k_p - \phi.(k_m - k_p)} \qquad (1)$$

in which, k_c, k_p, k_m are respectively thermal conductivities of the composite (metal particle filled polymer), continuous phase (polymer), and discrete phase (metal particles), ϕ is the volume fraction of filler (discrete phase). This model predicts fairly well thermal conductivities at low filler concentrations.

The semi-theoretical model developed by Lewis and Nielsen(2) include the effect of the shape of the particles and the orientation or type of packing for a two-phase system:

$$k_c = k_p . \left(\frac{1 + A.B.\phi}{1 - B.\phi.\beta} \right) \qquad (2)$$

$$B = \frac{k_m / k_p - 1}{k_m / k_p + A} \quad \text{and} \quad \beta = 1 + \left(\frac{1 - \phi_m}{\phi_m^2} \right).\phi \qquad (3)$$

"A" is the shape factor and "ϕ_m" is a constant corresponding to the type of packing. In the case of randomly packed spherical particles A=1.5 and ϕ_m=0.637.

Experimental Results and Discussion

Aluminium, copper and lead particles in the form of fine powder are mixed with high density polyethylene powder at various volumetric concentrations. The particle sizes are respectively 3 microns for Aluminium powder, 8-12 microns for copper powder, 4 microns for lead powder. Rectangular shaped samples of 100mm length, 60mm width and 15mm thickness are prepared by placing the metal-polymer powder mixture in a die and melting it at 180°C under pressure. After cooling and solidifi-

cation,the thermal conductivity of samples are measured by hot-wire measuring technique, at room temperature, with an accuracy of ±5% and reproducibility of ±2%. For each specimen,the thermal conductivity is measured five times and the mean values are listed in Tables 1,2 and plotted in figure 1. The experimetal values of thermal conductivity are compared to calculated values from Maxwell and Lewis & Nielsen models. At low metal filler concentrations, the models predict fairly well the thermal conductivity of the composite,whereas at higher concentrations, the models underestimate the thermal conductivity.

TABLE 1 - Thermal Conductivities of Al-HDPE Composites.

% Al Volumetric	Experimental Results k(W/m.K)	Maxwell Model k(W/m.K)	Lewis & Nielsen Model k(W/m.K)
0	0.568	0.568	0.568
1	0.605	0.585	0.577
1.58	0.586	0.595	0.583
2.36	0.637	0.608	0.590
2.47	0.843	0.611	0.592
3.50	0.858	0.629	0.602
4.25	0.898	0.643	0.610
4.9	0.905	0.655	0.617
5.71	0.995	0.670	0.625
6.42	0.968	0.683	0.633
7.12	1.194	0.697	0.642
7.80	1.261	0.710	0.649
8.50	1.169	0.725	0.658

TABLE 2 - Thermal Conductivities of Cu-HDPE Composites.

% Cu Volumetric	Experimental Results k(W/m.K)	Maxwell Model k(W/m.K)	Lewis & Nielsen Model k(W/m.K)
0	0.477	0.477	0.477
1.92	0.539	0.505	0.491
3.75	0.540	0.533	0.504
5.52	0.572	0.560	0.516
7.23	0.645	0.588	0.528
8.72	0.716	0.613	0.539
10.47	0.725	0.644	0.551

MECHANICAL PROPERTIES OF METAL FILLED POLYMERS

Although there is no good general theory about the stress-strain behavior of filled systems , it is known from observations that generally fillers cause a large decrease in elongation to break and also fillers often decrease the tensile strength of a material. The simple model developed by Nielsen (2) explains in a semiquantitative manner many of the stress-strain properties of filled systems.For the case of perfect adhesion and for any kind of a stress-strain curve, the model predicts that the elongation to break of a system filled with particles of approximately spherical shape is:

$$\frac{\varepsilon_B(Filled)}{\varepsilon_B(Unfilled)} = 1 - \phi^{1/3} \tag{4}$$

ϕ is the volume fraction of filler. Especially at low concentrations of filler, when there is good adhesion, the filler causes a drastic decrease in elongation to break. The addition of fillers to polymers can also greatly reduce creep.

The specific volume of filled material is not always a linear additive function of the specific volumes of the components,but depends also on the nature of the components and the fabrication technique.In the extreme case voids are incorporated into the system during fabrication and the composite material becomes porous; in this case the tensile strength can be approximated by the empirical equation as a function of porosity (3):

$$\sigma_B = \sigma_{B0} \cdot e^{-aP} \tag{5}$$

σ_B is the tensile strength of the composite, σ_{B0} is the tensile strength when the material is nonporous (has no holes or voids), "a" is a constant, "p" is the porosity defined as the volume fraction of holes or void space in the material.

During tensile tests, because of large stresses and strains, voids are created during the stretching of the specimen. This phenomenon which is due either to poor interfacial adhesion or to the breaking up of aggregates of low strength, is called dewetting (4). After dewetting takes place, tensile strength and modulus decrease greatly.

The mechanical properties of filled systems depend upon the size of the particles. The largest effect is

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usually on the tensile strength. As the particle size decreases, tensile strength increases. Also , as the particle size decreases, the modulus and yield strength increase, while the elongations at yield and at break decrease. The tensile strength and other properties are approximately a linear function of the reciprocal of the particle size.

Experimental Results and Discussion

Standard specimens for tensile tests are prepared by first mixing High Density Polyethylene powder with various volume fractions of aluminium powder, then placing the mixed powder in a die and allowing to melt at 180°C under pressure; after solidification the standard specimen is obtained. The average grain size of aluminium particles is 3 microns.

Tensile tests were performed at room temperature (20°C),the strain rate being 5mm/min.Results of tensile strength versus volume fraction of Al are given in Fig.2. From this figure, it may be noticed that there is a first sharp decrease in tensile strength at low volume fractions of Al filler, then a gradual decrease as the volume fraction of filler increases. Upon exami- nation of the ruptured specimen after the tensile tests, it was evident that dewetting occured which contribu- ted to the big decrease in tensile strength.

SYMBOLS USED

k = thermal conductivity (W/m.K)

$\emptyset$ = volume fraction of filler material (%)

σ_B = tensile strength of the composite (MN/m^2)

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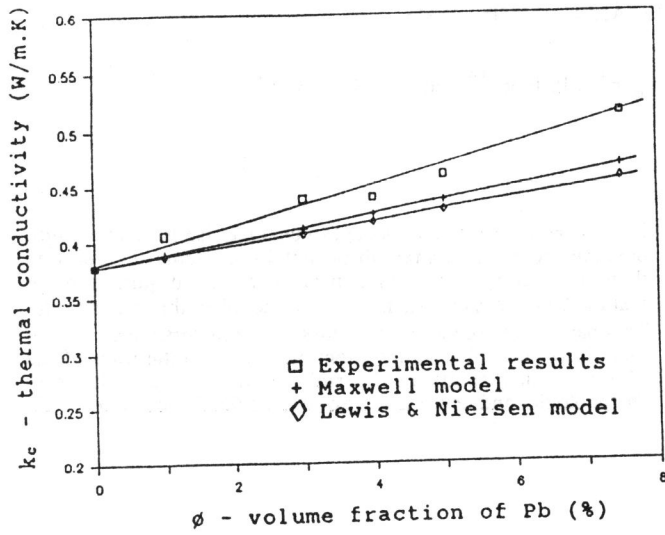
Figure 1. Thermal conductivity of HDPE-Pb composite
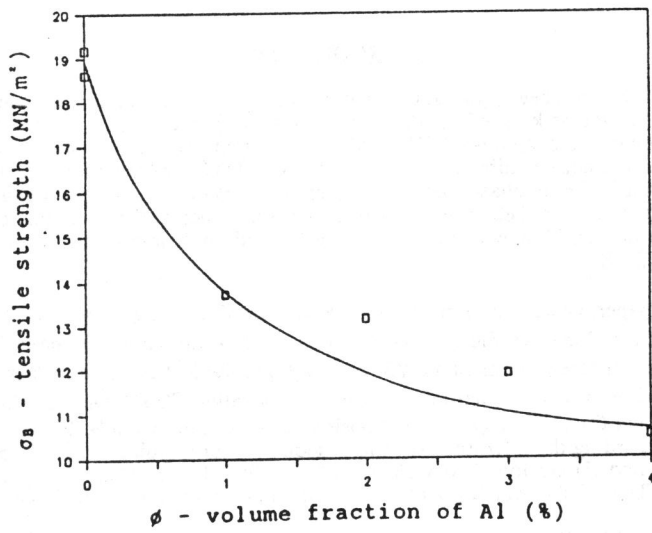plotted against volume fraction of Pb

Figure 2. Tensile strength of HDPE-Al composite plotted
against volume fraction of Al

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