Effect of Filler Loading, Geometry, Dispersion and Temperature on Thermal Conductivity of Polymer Nanocomposites

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Abstract

Using a unidirectional heat transfer apparatus, the roles of nanoparticle geometry, loading, dispersion and temperature on the thermal conductivity of polymer nanocomposites are investigated. The polymer nanocomposites (PNC) consist of epoxy matrices filled with silica nanopowder and carbon nanotubes, respectively, as well as poly (2-vinylpyridine) (P2VP) matrices loaded with silica nanoparticles. First, it is shown that thermal conductivity generally increases with nanofiller loading. These results are also reasonably described by the three phase Lewis-Nielsen or Halpin-Tsai analytical models. More importantly, it has been also demonstrated that the thermal conductivity of the polymer nanocomposites greatly depends on the dispersion state of the nanofillers. Furthermore, the effect of temperature on the thermal behavior of PNCs is briefly discussed. These results emphasize the important role of nanoparticles content and dispersion state on the thermal characteristics of polymer nanocomposites, which can be used to design composite materials with tunable thermal behavior.

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1. Introduction

Polymeric materials have been widely used for electronic packaging, high voltage transformers, cable terminations, and bushings or insulation for X-ray tubes [1–6]. Under these applications, the polymers are simultaneously exposed to mechanical and thermal loadings, and their thermal behavior plays a vital role in the overall performance of the entire system. In order to tune or improve the thermal conductivity of these macromolecules, nano sized inorganic particles have been incorporated into the polymer matrices[7–9]. Previous works suggested that the effective thermal conductivity of such composite materials is influenced by the thermal properties of the constituents [7,9,10], the thickness and characteristics of the interfacial layer [2,11,12], the filler's geometry/aspect ratio [13] and filler volume fraction [2–4,8,13–16]. However, to our knowledge, there is no systematic study to understand the combined effect of particle loading, geometry, dispersion state and temperature on the thermal conductivity of PNC.

In nanoparticle (NP) filled composites, the major issue is the clustering of the filler particles due to strong Van der Waals inter-NP forces. The extent of clustering or agglomeration has a direct dependence on the effective amount of filler and filler geometry. The tendency for agglomeration grows with the particle loading and/or the aspect ratio of the particle. Different mechanical and chemical techniques have been developed to overcome agglomeration over the years, but it still persists to some degree in highly loaded composite materials [17]. Recently, a polymer surface treatment using chemical solvent provided promising results in terms of dispersing the nanoparticles in polymers [17–22].

In this paper, we investigate the effect of nanofillers on the thermal conductivity of PNC, by accounting for the weight fraction, particle geometry and distribution of the fillers and the working temperature. Two types of fillers, higher aspect ratio carbon nanotubes and spherical silica, respectively, are used to characterize the role of particle geometry or aspect ratio. The effect of particle dispersion states is also investigated based on two different systems: carbon nanotube (CNT) samples with or without ultrasonication and silica-P2VP cast from different solvents. In addition, the effect of temperature on the thermal conductivity of the nanocomposites is also presented.

Analytical models are also used to model and understand these experimental findings. In the last few decades, different models have been proposed by implementing various analytical and semi-empirical techniques to estimate the mechanical and thermal property of composite materials. The series model, parallel model, the Lewis-Nielson (L-N), Maxwell Eucken-EMA (ME-EMA), Hamilton and Cheng-Vachon, and Halpin-Tsai models are a few proposed to determine the thermal conductivity of nanocomposites [2,3,11,23–25]. Most of these models take filler geometry, weight ratio of the filler, particle distribution, thermal conductivity of both the matrix and the filler, and interface thermal resistance (Kapitaz resistance) at the junction between the filler and the matrix into account. In this work, the three phase Lewis-Nielson and the modified Halpin-Tsai models are adopted due to their flexibility in allowing different material geometries, namely the particle, the matrix polymer and any interfacial material.

2. Materials and Methods

2.1 Materials

Three different types of PNCs, namely epoxy reinforced with silica particles (Type 1), epoxy reinforced with carbon nanotubes (CNTs) (Type 2), and silica-poly(2-vinylpyridine) [P2VP]

nanocomposites solvent cast from either methyl ethyl ketone (MEK) or pyridine solutions (Type 3) are considered. The weight fraction of the NPs was varied systematically in each class of PNC. The base matrix material used for Type 1 and Type 2 nanocomposites is Bisphenol-A epoxy (Buehler Epothin 2 Resin, Part-A) with compatible hardener, recommended by the manufacturer (Buehler Epothin 2 Hardener, Part-B). The nanoparticles used for Type 1 materials are spherical silica nanopowders with a primary particle size of 10 to 20 nm (BET) and 99.5 trace metal basis purchased from Sigma Aldrich. The nanoparticles used for Type 2 materials are tubular 95% pure CNTs, purchased from NanoLab Inc, with an outer diameter of 30 ± 15 nm and length of 5 to 20 microns. The silica NPs used in Type 3 samples were donated by Nissan Chemical Industries, which were incorporated into a poly (2-vinylpyridine) (P2VP) matrix ($M_w = 105$ Kg/mol, $M_w/M_n = 1.08$) purchased from polymer source Inc. The antioxidant Irganox 1010 (Ciba Specialty Chemicals, now BASF Switzerland) was used to avoid thermal degradation during annealing at 150°C for this last class of materials.

2.2 Nanocomposite preparation

For Type 1 nanocomposites, the as-received epoxy resin (Part-A) and silica nanopowder were weighed to obtain the desired NP weight fraction. The constituents were mixed mechanically in a glass beaker until the mixture turned to a uniform color. Then the mixture was ultrasonicated to obtain a uniform distribution of the filler in the matrix. The ultrasonication is made at 24% of the maximum amplitude with pulse mode of 2 sec ON and 1sec OFF for 30 mins. In order to maintain the temperature uniform during ultrasonication process the beaker was kept in an ice bath. The mixture was then kept in a vacuum chamber for about 10-30 mins and the air trapped during mechanical and ultrasonic mixing was sucked out. Afterwards, the right amount of hardener was added to the mixture (i.e. 1:2 ratio to the epoxy) and mixed thoroughly. The

mixture was brought back to and kept in the vacuum chamber again for a few minutes to remove any further trapped air. Following this the mixture was poured into silicon rubber molds that are coated with a release agent, and placed in a controlled hood to cure for 48 hrs. Finally, disc shaped samples with a diameter of 25.4 mm and a thickness of 1mm-1.5mm were prepared and the sample surfaces were polished with different grit size sand papers.

In Type 2, nanocomposite (CNT-epoxy) samples were also prepared following a similar procedure except for some changes in the ultrasonication parameters. CNT particles have a strong tendency to cluster together and usually a stronger shaking force is required to disperse them well. Ultrasonic waves are used to break the clustering bond, however, since CNTs have the ability to change their chemical form due to the sonic wave, the wave amplitude must be tuned, and the ultrasonication timing must be adjusted to keep the mixing temperature low. Considering these effects, the CNT-epoxy samples are ultrasonicated at 40 % of the maximum amplitude with pulse mode of 5sec ON and 10sec OFF for 1 hour. To probe the effect of particle distribution, samples were made following the same procedure but without using the ultrasonication.

The detailed fabrication process of Type 3 nanocomposites can be found elsewhere [18]- we briefly summarize the procedure here for completeness. First, P2VP and silica NPs with the appropriate weight ratio was mixed in either MEK or pyridine. The resulting solutions were vortex-shaken for 2 h and probe-ultrasonicated for 3 min. The solutions were then poured into a PTFE Petri dish, air dried in a fume hood for 5 days. To remove the residual solvent, the as-cast samples were then annealed at 150 °C for 24 h under vacuum.

2.3 Thermal conductivity characterization

The thermal conductivity of all samples was measured using a linear unidirectional heat transfer apparatus shown in Figure 1. The apparatus has six thermocouples to monitor the temperature flow through and across the cross section of the specimen. This device was set in an arrangement in which there is an electrical heat source at the top and a constantly flowing water sink at the bottom. The specimen was sandwiched between the two 25.4mm diameter brass rods. The top rod was connected with a heater while the bottom rod was connected to constantly running water. The sectioned schematic diagram of the experimental setup is shown in Figure 1, where T1-T6 represents the locations of the six thermocouples and T_H and T_C represents the top and lower surface of the specimen.

The heat source and the associated temperatures can be controlled by adjusting the input voltage. In this work, experiments were conducted at three different settings corresponding to 5V, 6.5V and 8V source voltages. During the test, once steady-state was achieved, the temperatures were measured using the thermocouples. The thermal conductivity of each specimen can be calculated from the temperature records and geometries, as given by Eq.1.

$$K_e = K_{Brass} \left[\frac{(\Delta X_{1-3}) * (T_1 - T_3)}{(\Delta X_{Specimen}) * (T_H - T_T)} \right] \dots \dots \dots (1)$$

Where K_{Brass} is the thermal conductivity of the brass conductors, $\Delta X_{Specimen}$ is the thickness of the test material; ΔX_{1-3} is the distance between the thermocouples T_1 and T_3 . T_H and T_C are temperature at the top and bottom surface of the specime, obtained by extrapolating temperature readings form T_1 - T_3 and T_4 - T_6 respectively.

3 Analytical Method

3.1 Three phase Lewis-Nielson (LN) Model

The three phase Lewis-Nielson (LN) model is adapted from the Halpin-Tsai (HT) mechanical model. It is a semi-empirical equation that considers the effect of particle size, shape of the particle (aspect ratio), weight ratio of the particle and the critical or maximum packing of particles [3]. It also accounts for the interface layer resistance by taking the layer as a third phase within the composite. The effective thermal conductivity of the composite can be given as:

$$K_e = K_m \left[\frac{(1 + \varepsilon * \eta * \varphi_{cp})}{(1 - \phi * \eta * \varphi_{cp})} \right] \dots \dots \dots (2)$$

Where, $\phi = 1 + \left[\frac{(1 - \varphi_m)}{(\varphi_m^2)} \right] \varphi_{cp}$, $\eta = \left[\frac{(K_i - K_m)}{(K_i + K_m * \varepsilon)} \right]$, ε is the aspect ratio, φ_m is the maximum packing factor and K_i is the combined thermal conductivity of the interface and particle respectively which is calculated as,

$$K_i = 1 / \left[\left[\frac{\varphi_p}{K_f(\varphi_p + V_i)} \right] + \left[\frac{\varphi_i}{K_m(\varphi_p + \varphi_i)} \right] \right] \dots \dots \dots (3)$$

K_m and K_f are the thermal conductivity of the matrix and the particle, respectively, φ_p is the volume fraction of the particles, and φ_i is the volume fraction of the interface layer which is obtained as shown from Eq.4:

$$\varphi_i = \varphi_p \left[\frac{(r_p + t)^3}{(r_p)^3} - 1 \right] \text{ and } \varphi_{cp} = \varphi_p * \delta \dots \dots \dots (4)$$

$\delta = \left(1 + \frac{t}{r_p} \right)^3$ and r_p is the radius of the particle, φ_{cp} is the combined volume fraction of

particle and interface layer. The effective thermal conductivity (K_e) obtained from this analytical model is shown in Fig. 2 and 3.

3.2 Halpin-Tsai (HT) Model

The Halpin-Tsai model is a semi-empirical model usually used to predict the elastic constants [2,23,24] especially for randomly oriented and high aspect fillers, such as for carbon nanotube reinforced composites. This model also considers different factors, such as the geometry of the filler, the properties of the filler and the matrix, volume fraction of the filler and filler distribution. In our study, the model was adopted and implemented to determine the effective thermal conductivity of nanocomposites made with CNT fillers. The model incorporates two parts as shown in Eq. 5, one for transverse and one for longitudinal thermal conductivities,

$$k_e = \frac{3}{8} \left[1 - 2 \left(\frac{l}{d} \right) * \left[\frac{\frac{k_f - d}{k_m - 4t}}{\frac{k_f + l}{k_m + 4t}} \right] \phi_f \right] * \left[1 - \left[\frac{\frac{k_f - d}{k_m - 4t}}{\frac{k_f - d}{k_m - 2t}} \right] \phi_f \right] * K_f + \frac{5}{8} \left[1 - 2 \left[\frac{\frac{k_f - d}{k_m - 4t}}{\frac{k_f + l}{k_m + 2t}} \right] \phi_f \right] * \left[1 - \left[\frac{\frac{k_f - d}{k_m - 4t}}{\frac{k_f + l}{k_m + 2t}} \right] \phi_f \right] * K_f \dots \dots \dots (5)$$

Where l , t and d are the length, thickness and diameter of the CNT filler, K_f and K_m are thermal conductivities of the filler and the matrix respectively and ϕ_f is the volume fraction of the filler CNT.

4 Results and Discussion

4.1 Effect of Particle Loading

Figure 2, presents the effective thermal conductivity of SiO₂-epoxy nanocomposites (type-1) as a function of particle content. As shown in the figure 2, the effective thermal conductivity (K_e) of the PNCs increases with an increase in the weight fraction of silica NPs. This should be attributed to the much higher thermal conductivity of silica materials (2 W/mK) compared to that of the epoxy matrix (0.15 W/mK). From Figure 2, it is also clear that the increment in K_e is not linear with the particle content, and qualitatively, it can be divided into three regions. In the first region at low particle loading, a rapid increase (~35% increment with inclusion of 2 wt % silica)

in the thermal conductivity was observed, where the nanoparticles were expected to be uniformly dispersed. In the second region, as the weight fraction of particles increases the slope of the increment in K_e is reduced (~14% increment from 2 wt % to 4 wt %), presumably due to particle clustering as more particles are added into the matrix. Going further (third region), when the filler content increased to larger than 6 wt %, the increment in K_e rose again. In the third region, higher particle packing is expected, and this packing brings a continuous particle to particle heat channel. As a result, the thermal barrier effect due to the local clustering is over dominated by the particle to particle channeling, and hence the thermal conductivity increases in region 3.

Figure 2 also provides the prediction of K_e as a function of particle loading by both the modified Halpin-Tsai and the Three phase Lewis-Nielsen models. Clearly, the Three phase Lewis-Nielsen model agrees much with the experimental results, with a maximum variation of 8%. Furthermore, the trend in the rate of increment in K_e for the three different regimes is quite well captured by this model. In contrast, the modified Halpin-Tsai model deviates appreciably from the experimental values, with the discrepancy increasing with the increase in the particle concentration. We believe that the failure of accounting for the interface effect in the Halpin-Tsai model should be the main reason for such a high discrepancy. Based on these results, we suggest that the Three phase Lewis-Nielsen model is reasonably appropriate for PNCs comprised of NPs with a spherical geometry or low aspect ratio.

The K_e of CNT-epoxy nanocomposites (Type-2) as a function of particle loading is shown in Figure 3. Similar to the silica filled system, the K_e of the CNTs filled PNCs increases monotonically with the amount of CNT incorporated. Due to the high thermal conductivity of the CNT fillers, the K_e of the composite materials increases dramatically with even a small amount of CNT added. For example, the K_e increases by ~32% with the addition of only 0.1 wt % CNT

into the plain epoxy and ~80% for the 0.4 wt % sample. At higher particle content, the rate of increment in K_e becomes smaller, which is the result of the particle agglomeration at high particle loadings.

Figure 3 also depicts the analytical prediction of K_e as a function of particle loading for CNT filled PNCs from the Lewis-Nielson and the modified Halpin-Tsai models, respectively. The Lewis-Nielson model shows an almost linear dependence on weight ratio in good agreement with experimental results at higher NP content. The modified Halpin-Tsai model, on the other hand, shows a much better prediction for experimental results across the whole range of compositions; the maximum deviation obtained from this model is about 5%. In general both models have shown appreciable approximations and agreement with the experimental result for CNT-epoxy composites, this indicates that the Lewis-Nielson model can also be effectively used for large aspect ratio fillers.

4.2 Effect of Particle Dispersion

Previous studies show that the materials properties specifically the mechanical behavior of PNCs depends strongly on the spatial organization of the added NPs. It is generally suggested that uniform particle dispersion is preferred to maximize the particle/polymer interfacial interaction and improve the mechanical properties of the nanocomposites. The effect of particle distribution on the effective thermal conductivity of the nanocomposites is investigated.

We have examined two different PNC systems: one is the CNTs reinforced epoxy and the other is composed of silica NPs and P2VP matrices. In the former case, the spatial distribution of CNTs is manipulated by the ultrasonication. As shown in Figure 4, prior to ultrasonication, the CNTs are strongly agglomerated in the matrices, with typical cluster sizes of ~microns (Figure 4a and 4b). In contrast, with ultrasonication, the large clusters are broken up and the dispersion

of CNTs is greatly improved; even individual particles can be found dispersed in the matrix (Figure 4c and 4d).

In another system, the silica NPs are added into a P2VP matrix, in which the distribution of silica NPs can be facilely controlled by the choice of the casting solvent. Our previous work [16] has shown that, in the films cast from MEK, a boundary layer of P2VP forms at the NP surface. The resulting “hairy” particles are sterically stabilized against agglomeration (Figure 5b). In contrast, in pyridine, P2VP does not adsorb onto the silica surface. Thus the bare silica particles are aggregated into fractal clusters due to polymer-induced depletion attraction (Figure 5a). We have also demonstrated that the aggregated morphology of pyridine cast samples cannot be completely annealed away due to the strong Van der Waals interaction between the particles.

With this capability we examined the effect of particle organization on the thermal behavior of the resulting composites. As shown in Figure 6, for the non-ultrasonicated samples, the effective thermal conductivity of the nanocomposites increased at NP loading at lower CNT content, but decreases as the weight fraction of the CNT increases. In contrast, the well-mixed ultrasonicated samples show a monotonic increase of thermal conductivity with loading. It is believed that the presence of agglomerated clusters causes the scattering of heat carrier phonons and reduces the flow of heat in a uniform direction, and as a result, the thermal conductivity reduces. In addition, the temperature discontinuity created by local high thermal conductivity of the agglomerated particle imposes non-uniform heat flow, which could potentially reduce the effective thermal conductivity of the nanocomposites.

For the other sets of samples, figure 7 compares the thermal conductivity of Type 3 PNCs cast from these two different solvents. First, as expected, for PNCs cast from both solvents, the effective thermal conductivity increases with the weight fraction of the silica nanofillers.

Interestingly, at low silica loadings (less than 40 wt %), the films cast in MEK generally transport heat more efficiently than those cast in pyridine. This can be reconciled by the fact that the average inter-particle distance is significantly smaller in the case of using MEK as the solvent where the NPs are well distributed. As a result, the phonons can propagate more easily than in the agglomerated case, making their transport in a more homogeneous way.

In contrast, when the particles are agglomerated, as the phonons prefer to go through the particle phase, the length of the polymer phase involved in phonon propagation becomes longer, leading to lower conductivity. Furthermore, at high silica content, e.g., 45 wt % and 50 wt %, the values of thermal conductivity for samples cast from different solvents nearly converge, probably because the particle phase percolates. In other words, the particle morphology evolves to be quite similar in these two cases [19], leading to no significant difference in thermal conductivity.

4.3 Effect of Temperature

Prior to understanding the effect of temperature on the thermal conductivity of the composites, we first examine the glass transition temperature (T_g) of these materials as a function of silica loading by using Differential Scanning Calorimetry (DSC). As shown in Figure 8, the T_g increases with the silica weight fraction, indicating favorable interaction between the polymer and the particle. Also note that the magnitude of T_g increase is small, i.e., less than 5 K, as observed in other works [1,26].

The effective thermal conductivity as a function of temperature for epoxy-Silica nanocomposites is shown in Figure 9. Note that each specimen has been tested at three different temperatures. At temperatures close to the glass transition, it is expected that the polymer chains are highly mobile. This should increase the thermal conductivity by allowing more phonon vibration and heat transportation. This effect can be seen in Figure 9, for pure epoxy and lower weight ratio

filler, where the effective thermal conductivity increases linearly with temperature. Clearly, as temperature increases, the resistance for the phonon propagation reduces, thus leading to faster heat transport.

As shown in Figure 9, at silica content less than 4 wt %, the thermal conductivity increases with the temperature, and almost remain constant for 4 wt %. Interestingly, at 6 wt %, the thermal conductivity first increases up to certain temperature and starts to drop. This indicates that there is a possibility that the presence of higher filler content within the polymer initiates phonon scattering at temperature close to the T_g .

5. Summary

The effective thermal conductivity of particle filled nanocomposites is examined both experimentally and numerically. The effects of particle loading, geometry and distribution, as well as the operating temperature on the effective thermal conductivity of nanocomposites are explored. From the current experimental and numerical results, the following points can be summarized:

- ◆ The thermal conductivity of the nanocomposites increases with nanofillers added, which depends strongly on the thermal properties of the fillers used.
- ◆ Dispersion of the NPs in the polymeric matrices is critical to the thermal conductivity of the resulting PNCs.
- ◆ The thermal conductivity of the nanocomposites with well dispersed particles increases with increasing the particles weight ratio. While, for non-uniform distribution of particles, the thermal conductivity decreases as weight ratio increases. This leads to the deduction that NP agglomeration in non-dispersed particles might have decreased the resistance for heat transport.

- ◆ The effective thermal conductivity of the nanocomposites with lower silica content, increases with testing temperature.

It was clearly demonstrated that for composites with spherical particles the Lewis –Neilson model is more appropriate, while both the Lewis-Neilson and the Halpin-Tsai models can be effectively used for nanocomposites with large aspect ratio particles.

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Figures

Figure 1: Schematic diagram of unidirectional/linear heat transfer device. T1-T8 represents the locations of the eight thermocouples, respectively.

Figure 2: The effective thermal conductivity (K_e) of SiO₂-epoxy nanocomposites (Type-1) as a function of silica weight fraction.

Figure 3: The effective thermal conductivity (K_e) of CNT-epoxy nanocomposites (Type-2) as a function of particle weight fraction.

Figure 4: TEM images of non-ultrasonicated (**a** and **b**) and ultrasonicated (**c** and **d**) naocomposites for 0.2 wt % CNT samples.

Figure 5: TEM images of P2VP matrices filled with 5 wt % silica NPs solvent cast from (a) pyridine (b) MEK. The samples had been thermally annealed for 24 h at 150°C under vacuum.

Figure 6: Variation of Effective Thermal Conductivity with weight ratio for Ultrasonicated and Non-ultrasonicated CNT composite at 30 °C.

Figure 7: Variation of Effective Thermal Conductivity with weight ratio for SiO₂-P2VP composite with solvent pyridine and MEK.

Figure 8: Glass transition temperature (Tg) of silica composite (Type-1) for different weight ratio.

Figure 9: Variation of Ke with Temperature (T) for different weight ratio of Silica composites.