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Thermal Conductivity of Nanoparticles Filled Polymers

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1. Introduction

Thermal conductivity of polymers is an important thermal property for both polymer applications and processing. Polymers typically have intrinsic thermal conductivity much lower than those for metals or ceramic materials, and therefore are good thermal insulators. Further enhancement of this thermal insulating quality can be achieved by foaming polymers. In other applications which require higher thermal conductivity, such as in electronic packaging and encapsulations, satellite devices, and in areas where good heat dissipation, low thermal expansion and light weight are needed, polymers reinforced with fillers, organic or inorganic, are becoming more and more common in producing advanced polymer composites for these applications (Hodgin & Estes, 1999; Tavman, 2004; Lee & Eun, 2004; Liu & Mather, 2004; Ishida & Heights, 1999; Frank & Phillip, 2002; Hermansen, 2001; Ishida, 2000). Most polymeric materials are processed and fabricated at elevated temperatures, often above their melting temperatures. This process may be long and expensive because of the low thermal conductivity of polymers. Subsequently, the cooling process or annealing may also be controlled by heat transport properties of polymers, which eventually affect the physical properties of the materials. One example is crystalline polymers, for which the structural and morphological features may be significantly changed with the speed of cooling. Careful consideration in designing polymer processing is vital to achieve desired properties.

For one-dimensional and rectilinear heat flow, the steady-state heat transfer in polymeric materials can be described by the Fourier's law of heat conduction:

$$q = -k\frac{dt}{dx} \tag{1}$$

where q is the heat flux (i.e., the heat transfer rate per unit area normal to the direction of flow), x is the thickness of the material, dT/dx is the temperature gradient per unit length, and the proportionality constant k is known as the thermal conductivity. The units for thermal conductivity k are expressed as W/(m K) in SI units, Btu in./(ft² h °F) in English units, and cal/(cm s ℃) in cgs units. The corresponding units for heat flux are expressed as W/(m²), Btu/(ft² h), and cal/(cm² s), respectively.

Heat transfer involves the transport of energy from one place to another by energy carriers. In a gas phase, gas molecules carry energy either by random molecular motion (diffusion) or by

an overall drift of the molecules in a certain direction (advection). In liquids, energy can be transported by diffusion and advection of molecules. In solids, phonons, electrons, or photons transport energy. Phonons, quantized modes of vibration occurring in a rigid crystal lattice, are the primary mechanism of heat conduction in most polymers since free movement of electrons is not possible (Majumdar, 1998). In view of theoretical prediction, the Debye equation is usually used to calculate the thermal conductivity of polymers (Han & Fina, 2010).

$$\lambda = \frac{C_p v l}{3} \tag{2}$$

where C_p is the specific heat capacity per unit volume; v is the average phonon velocity; and l is the phonon mean free path.

For amorphous polymers, l is an extremely small constant (i.e. a few angstroms) due to phonon scattering from numerous defects, leading to a very low thermal conductivity of polymers (Agari et al., 1997). Table 1 displays the thermal conductivities of some polymers (T'Joen et al., 2009), (Hu et al., 2007) and (Speight, 2005).

Material	Thermal Conductivity at 25°C (W/m K)
Low density polyethylene (LDPE)	0.30
High density polyethylene (HDPE)	0.44
Polypropylene (PP)	0.11
Polystyrene (PS)	0.14
Polymethylmethacrylate (PMMA)	0.21
Nylon-6 (PA6)	0.25
Nylon-6.6 (PA66)	0.26
Poly(ethylene terephthalate) (PET)	0.15
Poly(butylene terephthalate) (PBT)	0.29
Polycarbonate (PC)	0.20
Poly(acrylonitrile-butadiene-styrene) copolymer (ABS)	0.33
Polyetheretherketone (PEEK)	0.25
Polyphenylene sulfide (PPS)	0.30
Polysulfone (PSU)	0.22
Polyphenylsulfone (PPSU)	0.35
Polyvinyl chloride (PVC)	0.19
Polyvinylidene difluoride (PVDF)	0.19
Polytetrafluoroethylene (PTFE)	0.27
Poly(ethylene vinyl acetate) (EVA)	0.34
Polyimide, Thermoplastic (PI)	0.11
Poly(dimethylsiloxane) (PDMS)	0.25
Epoxy resin	0.19

Table 1. Thermal conductivities of some polymers (T'Joen et al., 2009), (Hu et al., 2007) and (Speight, 2005).

3. Thermal conductivity – measurement and modeling

3.1 Methods for thermal conductivity measurements

Several methods, as reviewed elsewhere (Tritt & Weston, 2004) and (Rides et al., 2009), have been proposed and used for measurement of the thermal conductivity of polymers and composites. Classical steady-state methods measure the temperature difference across the specimens in response to an applied heating power, either as an absolute value or by comparison with a reference material put in series or in parallel to the sample to be measured. However, these methods are often time consuming and require relatively bulky specimens.

Several non steady-state methods have also been developed, including hot wire and hot plate methods, temperature wave method and laser flash techniques (Nunes dos Santos, 2007). Among these, laser-flash thermal diffusivity measurement is widely used, being a relatively fast method, using small specimens (Nunes dos Santos, 2007), (Nunes dos Santos, 2005) and (Gaal et al., 2004). In this method, the sample surface is irradiated with a very short laser pulse and the temperature rise is measured on the opposite side of the specimen, permitting calculation of the thermal diffusivity of the material, after proper mathematical elaboration. The thermal conductivity k is then calculated according to Eq. (3):

$$k=aC_p\rho \tag{3}$$

where a, C_p and ρ are the thermal diffusivity, heat capacity and density, respectively.

Differential scanning calorimetry (DSC) methods may also be used, applying an oscillary (Marcus & Blaine, 1994) or step temperature profile (Merzlyakov & Schick, 2001) and analyzing the dynamic response.

Significant experimental error may be involved in thermal conductivity measurements, due to difficulties in controlling the test conditions, such as the thermal contact resistance with the sample, leading to accuracy of thermal conductivity measurements typically in the range of 5–10%. In indirect methods, such as those calculating the thermal conductivity from the thermal diffusivity, experimental errors on density and heat capacity values will also contribute to the experimental error in the thermal conductivity.

3.2 Modeling of thermal conductivity in composites

Several different models developed to predict the thermal conductivity of traditional polymer composites are reviewed elsewhere (Bigg, 1995), (Zhou et al., 2007), (Zeng et al., 2009) and (Wang et al., 2008). The fundamentals are recalled in this section.

The two basic models representing the upper bound and the lower bound for thermal conductivity of composites are the rule of mixture and the so-called series model, respectively. In the rule of mixture model, also referred to as the parallel model, each phase is assumed to contribute independently to the overall conductivity, proportionally to its volume fraction (Eq. (4)):

$$k_c=k_p\Phi_p+k_m\Phi_m \tag{4}$$

where k_c, k_p, k_m are the thermal conductivity of the composite, particle, matrix, respectively, and Φ_p, Φ_m volume fractions of particles and matrix, respectively. The parallel model maximizes the contribution of the conductive phase and implicitly assumes perfect contact between particles in a fully percolating network. This model has some relevance to the case of continuous fiber composites in the direction parallel to fibers, but generally results in very large overestimation for other types of composites.

On the other hand, the basic series model assumes no contact between particles and thus the contribution of particles is confined to the region of matrix embedding the particle. The conductivity of composites accordingly with the series model is predicted by Eq. (5):

$$k_c = \frac{1}{(\phi_m + k_m) + (\phi_p + k_p)} \tag{5}$$

Most of the experimental results were found to fall in between the two models. However, the lower bound model is usually closer to the experimental data compared to the rule of mixture (Ebadi-Dehaghani et al., 2011; Bigg, 1995), which brought to a number of different models derived from the basic series model, generally introducing some more complex weighted averages on thermal conductivities and volume fractions of particles and matrix. These so-called second-order models including equations by Hashin and Shtrikman, Hamilton and Crosser, Hatta and Taya, Agari, Cheng and Vachon as well as by Nielsen (Bigg, 1995), (Zhou et al., 2007) and (Okamoto & Ishida 1999), appear to reasonably fit most of the experimental data for composites based on isotropic particles as well as short fibers and flakes with limited aspect ratio, up to loadings of about 30% in volume.

In the case of the geometric mean model, the effective thermal conductivity of the composite is given by:

$$K_c = K_m^{\phi_m} + K_f^{\phi_f} \tag{6}$$

Lewis and Nielsen modified the Halpin-Tsai equation (Nielsen et al., 1994) to include the effect of the shape of the particles and the orientation or type of packing for a two-phase system.

$$K_c = K_m \left[\frac{1 + AB\phi_f}{1 - B\psi\phi_f}\right] \tag{7}$$

Where

$$B = \frac{\frac{K_f}{K_m} - 1}{\frac{K_f}{K_m} - A} \quad \psi = 1 + \left(\frac{1 - \phi_{max}}{{\phi_{max}}^2}\right)\phi_f \tag{8}$$

The values of A and Φ_{max} were given for many geometric shapes and orientations (Weidenfeller et al., 2004).

This model appears to reasonably fit most of the experimental data for composites based on isotropic particles as well as short fibers and flakes with limited aspect ratio, up to loading of about 30% in volume. For higher loadings, the Nielsen's model appear to best fit the rapid increase of thermal conductivity above 30 vol.%, thanks for the introduction of the maximum packing factor into the fitting equation, despite the evaluation of maximum packing factor in

real composites may present difficulties due to particle size distribution and particle dispersion in the matrix. However, the basic assumption of separated particles in the effective medium approach is not valid in principle for highly filled composites, where contacts are likely to occur, possibly leading to thermally conductive paths (Tavman, 1996).

Maxwell, using potential theory, obtained an exact solution for the conductivity of randomly distributed and non-interacting homogeneous spheres in a homogeneous medium:

$$K_c = K_m \left[\frac{K_f + 2K_m + 2\phi_f (K_f - K_m)}{K_f + 2K_m - \phi_f (K_f - K_m)} \right] \tag{9}$$

Other theoretical models have attempted to explain the thermal conductivity of two-phased composites. Some of these models, such as those by Bruggeman, Botcher, De Loor, and Ce Wen Nan et al., equations 6 to 9 respectively, have been used for prediction of thermal conductivity of carbon nanotube composites (Bruggeman, 1935; Böttcher, 1952; deLoor, 1956; Nan et al., 2004).

$$K_c = \frac{K_m}{(1-\phi_f)^3} \tag{10}$$

$$K_c = \frac{K_m}{(1-\phi_f)} \tag{11}$$

$$K_c = \frac{K_m (1+\phi_f)}{1-2\phi_f} \tag{12}$$

$$K_c = \frac{K_m \left[3+\phi_f \left(\frac{K_f}{K_m} \right) \right]}{3-2\phi_f} \tag{13}$$

In order to take into account fluctuations in thermal conductivity in the composites, Zhi et al. (Zhi et al., 2009) proposed the concept of heat-transfer passages, to model the conduction in regions where interparticle distance is low, applying the series model to "packed-belt" of conductive particles.

Even though these macroscopic approaches may be of interest from the engineering point of view, they deliver little or no information about the physical background of the observed behavior. As an example, very limited interpretation is given to the rapidly increasing conductivity with filler content above a certain filler loading (typically above 30 vol.%), or why the experimental results are so far away from the upper bound conductivity, even for highly percolated systems.

Attempts to model thermal conductivity taking into account the interfacial thermal resistance between conductive particles and matrix have been reported by several research groups (Nan et al., 1997), (Every et al., 1992), (Dunn & Taya, 1993), (Lipton & Vernescu, 1996) and (Torquato & Rintoul, 1995) and applied particles with different geometries and topologies, including aligned continuous fibers, laminated flat plates, spheres, as well as disoriented ellipsoidal particles. In general, these models provided an improved fit with experimental data for ceramic based composites than models not accounting for interface thermal resistance. These approaches generally assume conductive particles to be isolated in the matrix and take into account the thermal resistance in heat transfer between conductive

particle and matrix, also known as Kapitza resistance, from the name of the discoverer of the temperature discontinuity at the metal–liquid interface. A very simple proof of thermal interfacial resistance is the fact that a thermal conductivity lower than the reference matrix was experimentally found with some composites containing particles with thermal conductivity higher than the matrix (Nan et al., 1997) and (Every et al., 1992). This phenomenon is explained by the very low efficiency of heat transfer between particles and matrix, so that the higher thermal conductivity of the filler cannot be taken into advantage and the composite behaves like a hollow material, thus reducing its conductivity compared to the dense reference matrix. Evaluation of the effective thermal conductivity of composite polymers by considering the filler size distribution law was investigated by Holotescu et al (Holotescu et al., 2009).

They presented an empirical model for the effective thermal conductivity (ETC) of a polymer composite that includes dependency on the filler size distribution—chosen as the Rosin-Rammler distribution. The ETC is determined based on certain hypotheses that connect the behavior of a real composite material A, to that of a model composite material B, filled with mono-dimensional filler. The application of these hypotheses to the Maxwell model for ETC is presented. The validation of the new model and its characteristic equation was carried out using experimental data from the reference. The comparison showed that by using the size distribution law a very good fit between the equation of the new model (the size distribution model for the ETC) and the reference experimental results is obtained, even for high volume fractions, up to about 50%.

4. Crystallinity and temperature dependence

Polymer crystallinity strongly affects their thermal conductivity, which roughly varies from 0.2 W/m K for amorphous polymers such as polymethylmethacrylate (PMMA) or polystyrene (PS), to 0.5 W/m K for highly crystalline polymers as high-density polyethylene (HDPE) (Hu et al., 2007). The thermal conductivity of semi-crystalline polymers is reported to increase with crystallinity. As an example, the thermal conductivity of polytetrafluoroethylene (PTFE) was found to increase linearly with crystallinity at 232 °C (Price & Jarratt, 2002).

However, there is a large scatter in the reported experimental data of thermal conductivity of crystalline polymers, even including some contradictory results. It should be noticed that the thermal conductivities of polymers depend on many factors, such as chemical constituents, bond strength, structure type, side group molecular weight, molecular density distribution, type and strength of defects or structural faults, size of intermediate range order, processing conditions and temperature. Furthermore, due to the phonon scattering at the interface between the amorphous and crystalline phase and complex factors on crystallinity of polymer, the prediction of the thermal conductivity vs. crystallinity presents a significant degree of complexity (Han & Fina, 2010).

Semicrystalline and amorphous polymers also vary considerably in the temperature dependence of the thermal conductivity. At low temperature, semicrystalline polymers display a temperature dependence similar to that obtained from highly imperfect crystals, having a maximum in the temperature range near 100 K, shifting to lower temperatures and higher thermal conductivities as the crystallinity increases (Greig & Hardy, 1981) and (Yano

& Yamaoka, 1995), while amorphous polymers display temperature dependence similar to that obtained for inorganic glasses with no maximum, but a significant plateau region at low temperature range (Reese, 1969). The thermal conductivity of an amorphous polymer increases with increasing temperature to the glass transition temperature (T_g), while it decreases above T_g (Zhong et al., 2001) and (Dashora & Gupta, 1996). The study of the thermal conductivity of some amorphous and partially crystalline polymers (PE, PS, PTFE and epoxy resin) as a function of temperature in a common-use range (273–373 K) indicates that the conductivity of amorphous polymers increases with temperature and that the conductivity is significantly higher in crystalline than amorphous regions (Kline, 1961).

From the general overview given in the preceding, it appears that very limited thermal conductivity is usually characteristic of polymers. On the other hand, there are many reasons to increase thermal conductivity of polymer-based materials in various industrial applications including circuit boards in power electronics, heat exchangers, electronics appliances and machinery. This justifies the recent significant research efforts on thermally conductive composite materials to overcome the limitations of traditional polymers.

5. Fillers for thermally conductive composites

Many applications would benefit from the use of polymers with enhanced thermal conductivity. For example, when used as heat sinks in electric or electronic systems, composites with a thermal conductivity approximately from 1 to 30 W/m K are required (King et al., 1999). The thermal conductivity of polymers has been traditionally enhanced by the addition of thermally conductive fillers, including graphite, carbon black, carbon fibers, ceramic or metal particles (see Table 2) (Pierson, 1993), (Wypych, 2000), (Fischer, 2006),

Material	**Thermal Conductivity at 25 °C (W/m K)**
Graphite	100∼400 (on plane)
Carbon black	6∼174
Carbon Nanotubes	2000∼6000
Diamond	2000
PAN-based Carbon Fibre	8∼70 (along the axis)
Pitch-based Carbon Fibre	530∼1100 (along the axis)
Copper	483
Silver	450
Gold	345
Aluminum	204
Nickel	158
Boron Nitride	250∼300
Aluminum nitride	200
Beryllium oxide	260
Aluminum oxide	20∼29

Table 2. Thermal conductivities of some thermally conductive fillers (Pierson, 1993), (Wypych, 2000), (Fischer, 2006), (Wolff & Wang, 1993) and (Kelly, 1981).

(Wolff & Wang, 1993) and (Kelly, 1981). It is worth noticing that significant scatter of data are typically reported for thermal conductivity of fillers. This is caused by several factors, including filler purity, crystallinity, particle size and measurement method. It is also important to point out that some materials, typically fibers and layers, are highly anisotropic and can show much higher conductivity along a main axis or on a plane, compared to perpendicular direction.

High filler loadings (>30 vol.%) are typically necessary to achieve the appropriate level of thermal conductivity in thermally conductive polymer composites, which represents a significant processing challenge. Indeed, the processing requirements, such as possibility to be extruded and injection molded, often limit the amount of fillers in the formulation and, consequently, the thermal conductivity performance (King et al., 2008). Moreover, high inorganic filler loading dramatically alters the polymer mechanical behavior and density. For these reasons, obtaining composites having thermal conductivities higher than 4 W/m K and usual polymer processability is very challenging at present (Han & Fina, 2010).

5.1 Carbon-based fillers

Carbon-based fillers appear to be the best promising fillers, coupling high thermal conductivity and lightweight. Graphite, carbon fiber and carbon black are well-known traditional carbon-based fillers. Graphite is usually recognized as the best conductive filler because of its good thermal conductivity, low cost and fair dispersability in polymer matrix (Causin et al., 2006) and (Tu & Ye, 2009). Single graphene sheets constituting graphite show intrinsically high thermal conductivity of about 800 W/m K (Liu et al., 2008) or higher (theoretically estimated to be as high as 5300 W/m K (Veca et al., 2009) and (Stankovich et al., 2006)), this determining the high thermal conductivity of graphite, usually reported in the range from 100 to 400 W/m K. Expanded graphite (EG), an exfoliated form of graphite with layers of 20–100 nm thickness, has also been used in polymer composites (Ganguli et al., 2008), for which the thermal conductivity depends on the exfoliation degree (Park et al., 2008), its dispersion in matrix (Mu & Feng, 2007) and the aspect ratio of the EG (Kalaitzidou et al., 2007).

Thermal conductivity of exfoliated graphite nanocomposites was investigated by Fukushima et al. (Fukushima et al., 2006). Since the late 1990's, research has been reported where intercalated, expanded, and/or exfoliated graphite nanoflakes could also be used as reinforcements in polymer systems. The key point to utilizing graphite as a platelet nanoreinforcement is in the ability to exfoliate graphite using Graphite Intercalated Compounds (GICs). Natural graphite is still abundant and its cost is quite low compared to the other nano–size carbon materials, the cost of producing graphite nanoplatelets is expected to be ~$5/lb. This is significantly less expensive than single wall nanotubes (SWNT) (>$45000/lb) or vapor grown carbon fiber (VGCF) ($40–50/lb), yet the mechanical, electrical, and thermal properties of crystalline graphite flakes are comparable to those of SWNT and VGCF. The use of exfoliated graphite flakes (xGnP) opens up many new applications where electromagnetic shielding, high thermal conductivity, gas barrier resistance or low flammability are required. A special thermal treatment was developed to exfoliate graphite flakes for the production of nylon and high density polypropylene nanocomposites. X-ray diffraction (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were used to assess the degree of exfoliation of the

graphite platelets and the morphology of the nanocomposites. The thermal conductivity of these composites was investigated by three different methods, namely, by DSC, modified hot wire, and halogen flash lamp methods. The addition of small amounts of exfoliated graphite flakes showed a marked improvement in thermal and electrical conductivity of the composites.

Carbon fiber, typically vapor grown carbon fiber (VGCF), is another important carbon-based filler. Polymer/VGCF composites have been reviewed by Tibbetts et al. (Tibbetts et al., 2007). Since VGCF is composed of an annular geometry parallel to the fiber axis, thermal conductive properties along the fiber axis are very different from the transverse direction (estimated up to 2000 W/m K in the axial direction vs. 10–110 W/m K in the transverse direction (Chen & Ting, 2002) and (Zhang et al, 2000)), directly affecting the thermal conductivity of aligned composites (Mohammed & Uttandaraman, 2009) and (Kuriger et al., 2002).

Carbon black particles are aggregates of graphite microcrystals and characteristic of their particle size (10–500 nm) and surface area (25–150 m^2/g) (Pierson, 1993). Carbon black is reported to contribute to electrical conductivity rather than thermal conductivity (Wong et al., 2001), (Abdel-Aal et al., 2008) and (King et al., 2006).

5.2 Metallic fillers

The filling of a polymer with metallic particles may result in both increase of thermal conductivity and electrical conductivity in the composites. However, a density increase is also obtained when adding significant metal loadings to the polymer matrix, thus limiting applications when lightweight is required. Metallic particles used for thermal conductivity improvement include powders of aluminum, silver, copper and nickel. Boudenne et al studied the thermal behavior of polypropylene filled with copper particles (Kumlutaş et al., 2003). In this work thermal conductivity, diffusivity, effusivity and specific heat of polypropylene matrix filled with copper particles of two different sizes were investigated. A parallel study of the evolution of the electrical conductivity was also carried out. The highest heat transport ability was observed for the composites filled with the smaller particles. The Agari's model provides a good estimation of the thermal conductivity of composites for all filler concentrations. Polymers modified with the inclusion of metallic particles include polyethylene (Kumlutaş et al., 2003), polypropylene (Boudenne et al., 2005), polyamide (Tekce et al., 2007), polyvinylchloride and epoxy resins (Mamunya et al., 2002), showing thermal conductivity performance depending on the thermal conductivity of the metallic fillers, the particle shape and size, the volume fraction and spatial arrangement in the polymer matrix. Thermal conductivity of metal powder-polymer feedstock for powder injection moulding was studied by Kowalski et al. (Kowalski et al., 1999).

Thermal conductivity of a powder injection molding feedstock (mixture of metal powders and polymers) in solid and molten states has been measured by using the laser flash method. The filler material was 316L stainless steel powder and its content in the mixture amounted 60% by volume. An attempt has been made to employ two most promising existing mathematical models (theoretical Maxwell- and semi-theoretical Lewis & Nielsen model) to calculate the thermal conductivity of the mixture (see section 1.3.2). Comparison of the experimental and calculated results has revealed that the Lewis & Nielsen model

predicts better than Maxwell model the thermal conductivity of the feedstock. As the difference between the calculated (Maxwell model) and the measured results amounts to 15–85%, it is suggested that it can only be used for preliminary assessment of the thermal conductivity of so highly filled composite material. If accurate thermal conductivity data are required (as in case of numerical simulation of the powder injection moulding process), measurement of this property has to be performed if meaningful simulation results are to be expected.

5.3 Ceramic fillers

Ceramic powder reinforced polymer materials have been used extensively as electronic materials. Being aware of the high electrical conductivity of metallic particles, several ceramic materials such as aluminum nitride (AlN), boron nitride (BN), silicon carbide (SiC) and beryllium oxide (BeO) gained more attention as thermally conductive fillers due to their high thermal conductivity and electrical resistivity (Nu et al., 2008) and (Ishida & Rimdusit, 1998). Thermal conductivities of composites with ceramic filler are influenced by filler packing density(Ohashi et al., 2005), particle size and size distribution (Yu et al., 2002) and (Mu et al., 2007), surface treatment (Gu et al., 2009) and mixing methods (Zhou et al., 2007). Models and theories for predicting the thermal conductivity of polymer composites were discussed. Effective Medium Theory (EMT), Agari model and Nielsen model respectively are introduced and are applied as predictions for the thermal conductivity of ceramic particle filled polymer composites. Thermal conductivity of experimentally prepared Si_3N_4/epoxy composite and some data cited from the literature are discussed using the above theories. Feasibility of the three methods as a prediction in the whole volume fraction region of the filler from 0 to 1 was evaluated for a comparison. As a conclusion: both EMT and Nielsen model can give a well prediction for the thermal conductivity at a low volume fraction of the filler; Agari model give a better prediction in the whole range, but with larger error percentage (He et al., 2007).

6. Nanocomposites for thermal conductivity

Polymer nanocomposites are commonly defined as the combination of a polymer matrix and additives that have at least one dimension in the nanometer range. The additives can be one-dimensional (examples include nanotubes and fibres), two-dimensional (which include layered minerals like clay), or three-dimensional (including spherical particles). Over the past decade, polymer nanocomposites have attracted considerable interests in both academia and industry, owing to their outstanding mechanical properties like elastic stiffness and strength with only a small amount of the nanoadditives. This is caused by the large surface area to volume ratio of nanoadditives when compared to the micro- and macro-additives. Other superior properties of polymer nano-composites include barrier resistance, flame retardancy, scratch/wear resistance, as well as optical, magnetic, thermal conductivity and electrical properties. Polymer based nanocomposites can be obtained by the addition of nanoscale particles which are classified into three categories depending on their dimensions: nanoparticles, nanotubes and nanolayers. The interest in using nanoscaled fillers in polymer matrices is the potential for unique properties deriving from the nanoscopic dimensions and inherent extreme aspect ratios of the nanofillers (Mai et al., 2006). Kumar et al. (Kumar et al., 2009) summarized six interrelated characteristics of

nanocomposites over conventional micro-composites: (1) low-percolation threshold (about 0.1–2 vol.%), (2) particle–particle correlation (orientation and position) arising at low-volume fractions (less than 0.001), (3) large number density of particles per particle volume (10^6 to 10^8 particles/μm^3), (4) extensive interfacial area per volume of particles (10^3 to 10^4 m^2/ml), (5) short distances between particles (10–50 nm at 1–8 vol.%) and (6) comparable size scales among the rigid nanoparticles inclusion, distance between particles, and the relaxation volume of polymer chains (Han & Fina, 2010).

Different nanoparticles have been used to improve thermal conductivity of polymers. As a few examples, HDPE filled with 7 vol.% nanometer size expanded graphite has a thermal conductivity of 1.59 W/m K, twice that of microcomposites (0.78 W/m K) at the same volume content (Ye et al., 2006). Poly(vinyl butyral) (PVB), PS, PMMA and poly(ethylene vinyl alcohol) (PEVA) based nanocomposites with 24 wt.% boron nitride nanotubes (BNNT) have thermal conductivities of 1.80, 3.61, 3.16 and 2.50 W/m K, respectively (Zhi et al., 2009). Carbon nanofiber was also reported to improve the thermal conductivity of polymer composites (Sui et al., 2008) and (Elgafy & Lafdi, 2005). However, the most widely used and studied nanoparticles for thermal conductivity are certainly carbon nanotubes (either single wall-SWCNT or multiwall-MWCNT), which have attracted growing research interest. Indeed, CNT couples very high thermal conductivity with outstanding aspect ratio, thus forming percolating network at very low loadings.

Droval and co-workers (Droval et al., 2006) investigated the effect of boron nitride (BN), talc ($Mg_3Si_4O_{10}$ $(OH)_2$), aluminum nitride (AlN) and aluminum oxide (Al_2O_3) particles, and their impact on thermal properties. Lewis and Nielson, Cheng and Vachon, Agari and Uno models were used to predict the evolution of thermal conductivity with filler content and were found to describe correctly thermal conductivity. Only BN shows a real exponential increase of conductivity over 20% v/v filler. Consequently, in best conditions introducing 30% v/v of BN allows the thermal conductivity to be multiplied by six.

A technology has been developed for making carbon-ceramic composite refractories by combining carbon fibers as reinforcing component with a mixture matrix, which allows one to make refractory components of various sizes and geometry, including thin-walled large constructions (Chernenko et al., 2009). The heat resistance of these composite refractories increases with the bulk silicization during ceramic production on a carbon-carbon substrate. The degree of silicization is determined by the volume of the open microporosity of transport type, which is formed by pyrolysis of a polymer coke-forming matrix in the initial carbon plastic. The transport micropores are produced by a modification of the phenol-formaldehyde resin additive treatment, which does not give rise to coke on pyrolysis. As a result, the content of open pores in the carbon framework attains 55%, which enables one to make a silicized composite refractory of density up to 2.7 g/cm^3 with a compressive strength of 250 – 300 MPa, bending strength 120–140, and tensile strength 60–80 MPa, elastic modulus 120–140 GPa, linear expansion coefficient 3.5×10^{-6} – 4.5×10^{-6} K^{-1}, and thermal conductivity 6 – 8 W/(m K). These refractories are widely used in various branches of industry. Thermal conductivity of particle filled polyethylene composite materials was investigated by Kumlutas et al. (Kumlutas et al., 2003). In this study, the effective thermal conductivity of aluminum filled high-density polyethylene composites is investigated numerically as a function of filler concentration. The obtained values are compared with experimental results and the existing theoretical and empirical models. The thermal

conductivity is measured by a modified hot-wire technique. For numerical study, the effective thermal conductivity of particle-filled composite was calculated numerically using the micro structural images of them. By identifying each pixel with a finite difference equation and accompanying appropriate image processing, the effective thermal conductivity of composite material is determined numerically. As a result of this study, numerical results, experimental values and all the models are close to each other at low particle content. For particle content greater than 10%, the effective thermal conductivity is exponentially formed. All the models fail to predict thermal conductivity in this region. But, numerical results give satisfactory values in the whole range of aluminum particle content.

6.1 Nanocomposites using inorganic fillers

Thermally conductive polymer nanocomposites based on polypropylene has been studied (Vakili et al., 2011; Ebadi-Dehaghani et al., 2011). In this study three nanocomposite containing 5 to 15 wt% of ZnO and CaCO3 nanoparticles prepared by extrusion were used. The thermal conductivity (TC) of compression moulded polypropylene (PP) and PP filled nanoparticles was studied using thermal conductivity analyser (TCA). The effect of nanoparticle content and crystallinity on thermal conductivity was investigated using conventional methods like SEM, XRD and DSC. The incorporation of nanoparticles improved crystallinity and thermal conductivity simultaneously. The experimental TC values of PP nanocomposites with different level of nanoparticles concentration showed a linear increase with an increase in crystallinity.

6.1.1 Differential Scanning Calorimetry (DSC)

DSC measurements were investigated by conventional differential scanning calorimeter Labsys TG (Setaram Instumentation, Caluire, France). A pellet of extruded sample, with a weight of 8-10 mg, was placed into an alumina pan in the presence of air as the furnace atmosphere. Measurements were performed from ambient temperature up to 200°C with heating rate of 10°C/min. The DSC results for pure PP and nanocomposites, The Tm (peak temperature of melting) and ΔHm (enthalpy of melting), are listed in Table 3.

The degree of crystallinity of a specimen can be calculated from the melting heat of crystallization according to the following equation:

$$X_c = \frac{\Delta H_m}{\Delta H_0(1 - W_f)} \times 100 \tag{14}$$

Where w_f is the weight fraction of nanofiller and ΔH_0=207.1 Jg^{-1} is the melting heat of 100% crystalline PP (Bai et al., 1999).

The DSC results indicated that the addition of both nanoparticles to the PP caused only a marginal effect on melting temperature (T_m) and no correlation of the results with the filler concentration could be established. The calculated degree of crystallinity of the PP phase increased with increasing content of both nanoparticles, indicating that the nanofillers nucleated the crystallization process. (Frormann et al., 2008) This implies that the existence of nanoparticles facilitates the crystallization of PP and this effect becomes more evident with higher nanoparticle content (Zhao & Li, 2006). Similar results for PP/CaCO3

nanocomposites, PP/carbon nanotube composites and PP/nanoclay composites (Han & Fina , 2011; Frormann et al., 2008; Vakili et al., 2011) have been reported. However there are some contradicting results in the literature (Zhao & Li, 2006).

	Neat PP	PP+5wt% nanofiller	PP+10wt% nanofiller	PP+15wt% nanofiller
T_m (°C)	167.8	168.5	168.8	167.0
ΔH_m (Jg^{-1})	78.0	91.2	108.9	107.7
X_c (%)	37.7	44.0	52.6	52.0

a)

	Neat PP	PP+5wt% nanofiller	PP+10wt% nanofiller	PP+15wt% nanofiller
T_m (°C)	167.8	168.6	168.1	168.4
ΔH_m (Jg^{-1})	78.0	81.4	85.8	104.6
X_c (%)	37.7	39.3	41.4	50.5

b)

Table 3. Crystallization parameters of neat PP and nanocomposites. a) PP/ZnO nanocomposite. b) PP/$CaCO_3$ nanocomposite.

6.1.2 Thermal conductivity measurement

Thermal conductivity was measured using a TCA Thermal Conductivity Analyser (TCA-200LT-A, Netzsch, Selb, Germany) with the guarded heat flow meter method. Each compression molded sample (30cm×30cm sheets with 10 mm thickness) was placed between two heated surface controlled at different temperatures with a flow of heat from the hot to the cold surface. When thermal equilibrium was attained thermal conductivity data were taken within an accuracy of 3%. Fig. 1 compares the effect of the nanoparticles' content on the thermal conductivity of the nanocomposites. As seen, the value of thermal conductivity increased with an increase in the nanoparticle concentration up to 64% and 82% for $CaCO_3$ and ZnO respectively. The increase in TC for ZnO nanoparticles is more than $CaCO_3$ due to the nature of nanofiller and also crystallinity degree regarding to DSC results (Table 3). These increases in TC for both nanoparticles are higher than that of reported values for CNF in a PP matrix (Frormann et al., 2008).

The values obtained from the experimental study of PP nanocomposites were compared with several TC models (Figs 2.a and 2.b). As seen the experimental results were found to fall in between the Series and Parallel models. However, the lower bound model (series) is usually closer to the experimental data.

Maxwell, Lewis & Nielson, Bruggeman, Bottcher and De Loor models predict fairly well thermal conductivity values up to 10 wt% for PP/ZnO nanocomposites (Fig. 2.a). In the concentration of 15 wt% no model could predict well the TC values and all of the mentioned models underestimated the TC values of nanocomposite whereas in the case 5 wt% all models overestimated the TC values.

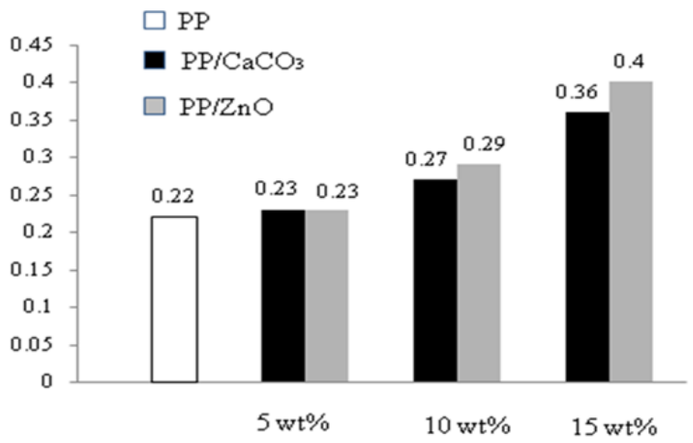

Fig. 1. Comparing the effect of nanoparticles on the TC of PP.

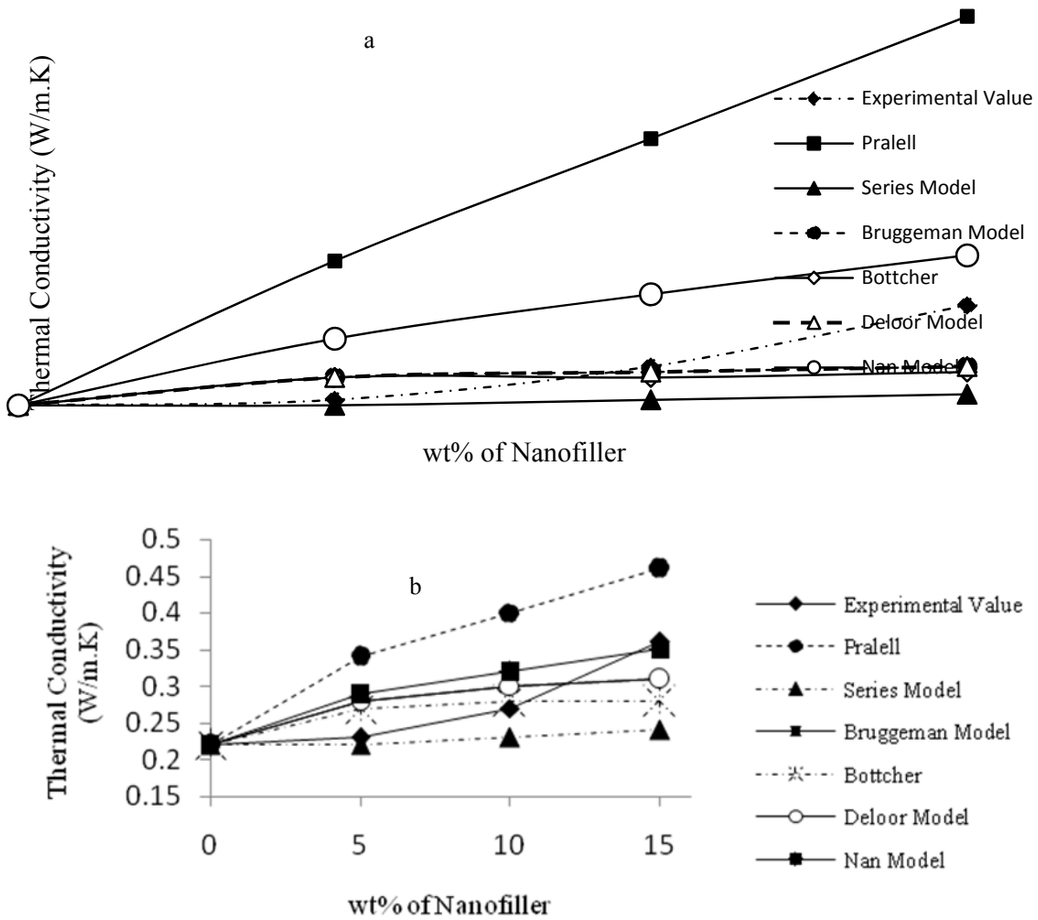

Fig. 2. Comparing the experimental TC values vs nanoparticle content with theoretical models. a) PP/ZnO nanocomposite b) PP/$CaCO_3$ nanocomposite.

This fact can be attributed to the intrinsic thermal conductivity of both nanoparticles and their large surface area which even at lower loadings of nanofillers they are still effective to transfer heat through the samples (Frormann et al., 2008). At a higher volume fraction, this effect becomes stronger. Fig. 2.b the values obtained from the experimental study for PP/$CaCO_3$ nanocomposites are compared with a number of TC models. As seen the Ce Wen Nan model predicts fairly well the thermal conductivity values up to 15 wt%. For the concentration of 10 wt% all the models predict the TC values well. In the case of 15 wt% other models underestimated the TC values of nanocomposites except for the Ce Wen Nan model, whereas for 5 wt% all models overestimated the TC value. The predicted TC values by the models depend on the nature of nanofiller and their relative concentrations (Weidenfeller et al., 2004; Frormann et al., 2008).

The TC improvement in PP/ZnO nanocomposite is greater than that of PP/calcium carbonate nanocomposites. This fact can be attributed to intrinsic thermal conductivity of the ZnO nanoparticles. Several models have been used for prediction of TC in the nanocomposites (see section 3.2). In the PP/ZnO nanocomposites TC values correlated well with the values predicted by Series, Maxwell, Lewis & Nielson, Bruggeman and De Loor models up to 10 wt%.

7. Conclusions

As electronic devices tend to become slimmer and more integrated, heat management become a central task for device design and application. Similar issues are faced in several other applications, including electric motors and generators, heat exchangers in power generation, automotive, etc. Metallic materials are widely used as heat dissipation materials, but there have been many attempts to replace the metallic materials with highly thermally conductive polymer based composites due to their lightweight, corrosion resistance, easy processing and lower manufacturing cost.

Thermally conductive polymer based composites are tentatively prepared by the incorporation of thermally conductive fillers. The outstanding thermal conductivity of mentioned fillers makes them a promising candidates to obtain highly thermally conductive polymer based composites.

PP nanocomposites were prepared by melt extrusion in a twin screw extruder. The introduction of nanoparticles resulted in an increase in crystallinity. Scanning electron microscopy (SEM) indicated a good dispersion of the nanofillers within the PP matrix that might enhance the thermal conductivity of the nanocomposites even at lower nanofiller loadings owing to enhanced filler-matrix interaction. The thermal conductivity of PP/ZnO nanocomposites had an increase of 82% at 15 wt% concentration comparing to that of pure PP, while for PP/$CaCO_3$ nanocomposite with same level of nanoparticle content it was 64%, so it is concluded that ZnO nanoparticles had more intrinsic potential to improve thermal conductivity of PP comparing to $CaCO_3$ nanoparticles regarding to its nature and crystallinity.

The thermal conductivity was increased from K=0.22 W/mK for pure PP by 64% for the sample with 15 wt% of $CaCO_3$ nanoparticles. These results for both nanocomposites (PP/ZnO and PP/$CaCO_3$) are higher than the values which reported for CNF in a PP matrix

Frormann L, Iqbal A, Abdullah S.A. 2008. The measured values were also compared with various models in the investigated range of nanofiller concentration. The Series, Maxwell, Lewis & Nielson, Bruggeman, Bottcher, De Loor and Ce Wen Nan models predicted fairly well the thermal conductivity values for the samples containing more than 5 wt% of nanoparticles. The experimental TC values of PP nanocomposites showed a linear increase with an increase in concentration and crystallinity.

8. Abbreviations

ABS poly(acrylonitrile-butadiene-styrene) copolymer
AlN aluminum nitride
BeO beryllium oxide
BN boron nitride
BNNT boron nitride nanotubes
CNT carbon nanotube
C_p heat capacity
DSC differential scanning calorimetry
DWCNT double-walled carbon nanotube
EG expanded graphite
EPDM ethylene propylene diene rubber
EVA poly(ethylene vinyl acetate)
GNP graphite nanoplatelet
HDPE high density polyethylene
K thermal conductivity (in some figures taken from literature referred as K_e = effective thermal conductivity)
k_c thermal conductivity of composite
k_m thermal conductivity of matrix
k_p thermal conductivity of particle
l phonon mean free path
L length parameter
LDPE low density polyethylene
MWCNT multi-walled carbon nanotube
PA6 polyamide 6
PA66 polyamide 6-6
PBT poly(butylene terephthalate)
PC polycarbonate
PDMS poly(dimethylsiloxane)
PE polyethylene
PEEK polyetheretherketone
PET poly(ethylene terephthalate)
PEVA poly(ethylene vinyl alcohol)
PI polyimide
PMDA pyromellitic dianhydride
PMMA polymethylmethacrylate
PP polypropylene
PPS polyphenylene sulfide
PPSU polyphenylsulfone

PS polystyrene
PSU polysulfone
PTFE polytetrafluoroethylene
PU polyurethane
PVB poly(vinyl butyral)
PVC polyvinyl chloride
PVDF polyvinylidene difluoride
R_k interfacial resistance
SiC silicon carbide
SWCNT single-walled carbon nanotube
T_g glass transition temperature
v average phonon velocity
VGCF vapor grown carbon fiber
A thermal diffusivity
ρ density of the material
Φ_m volume fractions of matrix
Φ_p volume fractions of particles
w_f weight fraction of particles

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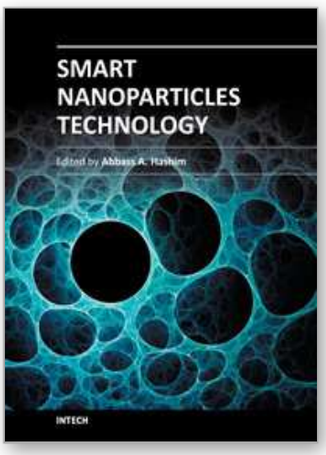

Smart Nanoparticles Technology

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In the last few years, Nanoparticles and their applications dramatically diverted science in the direction of brand new philosophy. The properties of many conventional materials changed when formed from nanoparticles. Nanoparticles have a greater surface area per weight than larger particles which causes them to be more reactive and effective than other molecules. In this book, we (InTech publisher, editor and authors) have invested a lot of effort to include 25 most advanced technology chapters. The book is organised into three well-heeled parts. We would like to invite all Nanotechnology scientists to read and share the knowledge and contents of this book.

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