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# IMPORTANCE & APPLICATIONS OF NANOTECHNOLOGY

# Polymers and Nanocomposites with High Thermal Conductivity

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## Abstract

Polymers are primarily insulating materials and are used in a vast array of applications because of their low cost, availability, and ease of manufacture. However, polymers have very low thermal conductivity, which is why they are used as insulators. Thermal conductivity is nevertheless a crucial parameter in new technologies and applications such as aerospace and mechanical instrumentation. Many smart advanced materials in electronic appliances tend to give off heat and increase the temperature, which degrades performance. High temperatures can at times affect the mechanical stability of polymeric parts and their operation.

Different approaches have been put forward to enhance the heat conductivity of polymers; most involve the incorporation of thermally conductive nanoparticles. This chapter reviews recent experimental and theoretical approaches to increasing the thermal conductivity of polymeric composites. Crucially, improved thermal conductivity requires conductive channels to dispel heat; thus, the percolation threshold must be reached.

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## Improving thermal conductivity using non-metallic fillers

To improve the thermal conductivity of non-conductor materials such as polymers, researchers have focused on either incorporating several types of fillers or modifying the structural aspects of the material. Better thermal conductivity depends on the fillers, their nature, their structure, and other features. Numerous types of thermally conductive nano-fillers (mostly crystalline) have been explored in recent years and have led to more or less comparable results for carbon-based fillers (graphite, carbon nanotubes, carbon black, graphene), metallic fillers (Ag, Cu, Al, TiO<sub>2</sub>, AlN), ceramic fillers (BN, Si, ZrB<sub>2</sub>), vegetal fibers, and others. Studies that have used various filler combinations to achieve a synergetic effect have yielded similar improvement rates [1,2].

Most findings confirm that when the composite structure has a physical percolation threshold, no noteworthy enhancement in thermal conductivity can be achieved. Thermal conductivity differs from electrical conductivity in several keyways as a simple consequence of their fundamental mechanisms. More studies are needed to determine how the dispersion state influences the thermal conductivity of the resulting material [3]. The mechanism of thermal conductance at the filler/matrix interface is one crucial issue to focus on in future work to better understand heat transfer in composite materials. The aspect ratio is also an important parameter to consider, as shown schematically in Figure 1 for epoxy composites. Zhou et al. [4] measured the percolation threshold for micro-SiC/epoxy composites at approximately 55%wt. At this loading, the thermal conductivity increased dramatically, but this high filler content affected the mechanical properties of the composites.

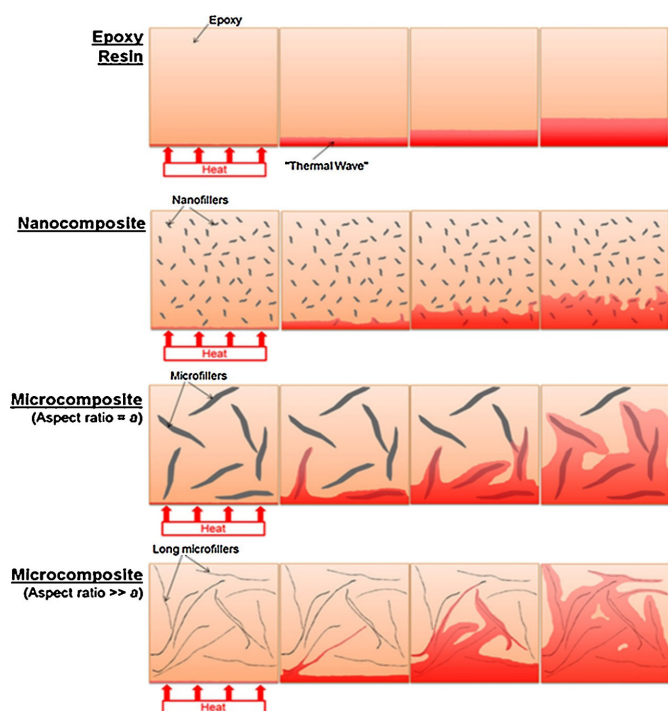
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**Table 1:** Thermal properties of various materials [5].

Material	Thermal Conductivity (W/m K)	CTE (10 <sup>-6</sup> /°C)	Density (g/cm <sup>3</sup> )
Aluminum	247	23	2.7
Gold	315	14	19.32
Copper	398	17	8.9
Lead	30	39	11
Molybdenum	142	4.9	10.22
Tungsten	155	4.5	19.3
Invar	10	1.6	8.05
Kovar	17	5.1	8.36
Diamond	2000	0.9	3.51
Beryllium oxide	260	6	3
Aluminum nitride	320	4.5	3.3
Silicon carbide	270	3.7	3.3

Chung et al. [5] reviewed materials in terms of their thermal conductivity, including metals, carbons, ceramics and composites, and showed that effective heat transfer requires good contact between the different surfaces involved. The Coefficient of Thermal Expansion (CTE) should also be taken into consideration, as shown in Table 1.



**Figure 1:** Heat transfer mechanism in composites, as a function of size and aspect ratios [1].

Han et al. [6] reviewed the thermal conductivity of carbon nanotubes and their polymer nanocomposites. Some of these thermal conductivities are listed in Table 1 and Table 2. Different nanoparticles can be used to improve the thermal conductivity of polymers. For example, HDPE filled with 7vol% nanometer size expanded graphite has a thermal conductivity of 1.59W/mK, twice that of microcomposites (0.78W/mK) at the same volume content [7]. 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 2.50W/mK, respectively [8]. Carbon nanofiber is also reported to improve the thermal conductivity of polymer composites [9,10]. 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. This is because CNT couples very high thermal conductivity with an outstanding aspect ratio, thus forming a percolating network at very low loadings.

Shtein et al. [11] studied the effect of the particle size of thermally conductive graphene-polymer composites. They reported the dispersion of commercially available graphene nanoplatelets in a polymer matrix, which formed a composite with an ultrahigh thermal conductivity of 12.4 W/m K (vs 0.2 W/m K for neat polymer). The percolation threshold was  $\sim \phi > 0.17$ . Grosiord et al. [12] suggested a toolbox for dispersing carbon nanotubes into polymers to obtain conductive nanocomposites. Three dispersion techniques were reviewed: the direct mixing of the CNT and the polymer, modification of either the polymer matrix or the CNT walls to improve the wetting of the filler with the matrix material, and the addition of a third component. The basic concept was the generation of a stable colloidal system containing both a suspension of CNTs stabilized by surfactant molecules in water, and polymer latex. After removal of the water, the resulting powder is processed into the desired shape. The third technique was reported as a promising route to produce conductive nanocomposites with low percolation thresholds as well as good conductivity levels. The resulting composite was shown to be highly flexible with respect to the choice of the polymer matrix, and can be applied to any polymer synthesized by emulsion polymerization, or made into a polymer latex form artificially; for example, CNT/polystyrene latex [13,14].

**Table 2:** Thermal conductivities of some key polymers [6].

Material	Thermal Conductivity at 25°C (W/mK)
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

Fukushima et al. [15] reported on the thermal conductivity of exfoliated graphite nanocomposites. Metal fillers are more affordable but have the disadvantage of high density. The use of lighter weight carbon-based compounds as conductive fillers has been widely investigated, including carbon blacks, carbon fibers, graphite, carbon fibers, and carbon nanotubes. CNTs exhibit a longitudinal thermal conductivity of 2800–6000 W/mK 2800–6000 W/mK for a single nanotube at room temperature [6].

Carbon blacks are inexpensive and commonly used as fillers to increase electrical conductivity, but often have relatively low thermal conductivity. Carbon fibers and carbon nanotubes have excellent thermal conductivity, but their cost is still too high for many applications. For example, the conductivity of carbon fiber-based composites are usually in the range of 2 W/mK to 20 W/mK. Graphite (crystalline) can be as high as 3000 W/mK, and thus may serve as a promising candidate. However, like all nanocomposites, good dispersion is crucial. Not only should the nanoparticles be well dispersed, the percolation threshold must be met in order to exhibit thermal conductivity [16].

### Improving thermal conductivity using metallic fillers

Mamunya et al. [17] examined the electrical and thermal conductivity of polymers filled with metal powders. The authors studied the electrical and thermal conductivity of systems based on Epoxy Resin (ER) and Poly (Vinyl Chloride) (PVC) filled with metal powders of copper and nickel with different particle shapes. The thermal and electrical conductivities strongly depended on the filler concentration. For example, when a polymer matrix with a conductivity  $\sigma_p$  was filled with a dispersed filler with a conductivity of  $\sigma_f$  the composite acquired a conductivity value of  $\sigma$ . When the volume filler fraction  $\phi$  reached a critical value  $\phi_c$  (the so-called percolation threshold), an Infinite Conductive cluster (IC) formed and the composite became conductive [16].

Mamunya et al. [17] compared experimental data with the following model:

$$\log \lambda = \log \lambda_p + (\log \lambda_f - \log \lambda_p) \left( \frac{\phi}{F} \right)^N$$

where F is the filling limit,  $\phi$  is the percolation volume of conductive sites, and  $\lambda, \lambda_p, \lambda_f$  are the overall, polymer and filler thermal conductivities, respectively. The N parameter related to the topological peculiarities of the systems was filled with dispersed filler. The parameters characterizing the thermal conductivity of the composites are listed in Table 3.

**Table 3:** The parameters characterizing the thermal conductivity of the composites [17].

Composite	$\log \lambda_p$ (Wt/mK)	$\log \lambda_f$ (Wt/mK)	$\log \lambda_f$ (Wt/mK)	N
ER-Cu	-0.64	0.55	2.59	1.0
PVC-Cu	-0.78	0.46	2.59	1.0
ER-Ni	-0.64	0.58	1.95	1.0
PVC-Ni	-0.78	0.09	1.95	1.3

Weidenfeller et al. [18] reported on thermal conductivity, thermal diffusivity, and the specific heat capacity of particle-filled polypropylene. Through the addition of metal and oxide particles to plastics, the thermal transport properties, heat capacity, and density of the polymers were varied systematically. Composite samples of Polypropylene (PP) with various fillers in

different fractions (up to 50 vol%) were prepared by an injection molding process. The thermal conductivity of the polypropylene increased from 0.27 up to 2.5 W/(mK) with 30 vol% talc or with 35 vol% Fe<sub>3</sub>O<sub>4</sub> in the polypropylene matrix.

Wong et al. [19] investigated the thermal conductivity, elastic modulus, and Coefficient of Thermal Expansion (CTE) of polymer composites filled with ceramic or metallic particles. As expected, the thermal conductivity of alumina and SCAN filled composites was much higher than those filled with silica. At a volume loading of 50%, the thermal conductivity of Silica-Coated Aluminum Nitride (SCAN)-filled composites was 10 times the intrinsic thermal conductivity of the epoxy resin. The aluminum-filled epoxy exhibited roughly 5 times the intrinsic thermal conductivity of the epoxy resin with a load of 50%.

### Improving thermal conductivity via coating

Almost all the scientific work on coatings has focused on Thermal Barrier Coatings (TBC); i.e., thermal insulations [20]. Understanding the heat flux mechanism makes it possible to better select materials and treatments. A phase diagram of the selected materials or alloys is a key component in the design of conductive layers. Cao et al. [21] studied the properties of ceramic for thermal applications. The properties are listed in Table 4.

**Table 4:** Properties of selected ceramic materials [21]

Materials	Properties
ZrO <sub>2</sub>	$T_m = 2973K$ $D_{th} = 0.43 \times 10^{-6} m^2s^{-1}$ (1273K) $\lambda = 2.17 Wm^{-1}K^{-1}$ (1273K) $E = 21 GPa$ (1373K) $\alpha = 15.3 \times 10^{-6} K^{-1}$ (1273K) $\nu = 0.25$
3YSZ	$T_m = 2973K$ $D_{th} = 0.58 \times 10^{-6} m^2s^{-1}$ (1273K) $\lambda = 2.12 Wm^{-1}K^{-1}$ (1273K) $C_p = 0.64 Jg^{-1}K^{-1}$ (1273K) $\alpha = 11.5 \times 10^{-6} K^{-1}$ (293–1273K)
8YSZ (plasma-sprayed)	$E = 40 GPa$ (293K) $\alpha = 10.7 \times 10^{-6} K^{-1}$ (293–1273K) $\nu = 0.22$
18YSZ	$\alpha = 10.53 \times 10^{-6} K^{-1}$ (1273K)
5 wt.% CaO + ZrO <sub>2</sub>	$T_{softening} = 2558K$ $E = 149.3 GPa$ (293K) $\alpha = 9.91 \times 10^{-6} K^{-1}$ (1273K) $\nu = 0.28$
Mullite	$T_m = 2123K$ $\lambda = 3.3 Wm^{-1}K^{-1}$ (1400K) $E = 30 GPa$ (293K) $\alpha = 5.3 \times 10^{-6} K^{-1}$ (293–1273K) $\nu = 0.25$
Al <sub>2</sub> O <sub>3</sub>	$T_m = 2323K$ $D_{th} = 0.47 \times 10^{-6} m^2s^{-1}$ (1273K) $\lambda = 5.8 Wm^{-1}K^{-1}$ (1400K) $E = 30 GPa$ (293K) $\alpha = 9.6 \times 10^{-6} K^{-1}$ (1273K) $\nu = 0.26$

Al <sub>2</sub> O <sub>3</sub> (TGO)	$E = 360 \text{ GPa (293K)}$ $\alpha = 8 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1273K)}$ $\nu = 0.22$
Al <sub>2</sub> O <sub>3</sub> + TiO <sub>2</sub>	$D_{th} = 0.65 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ (1273K)}$ $\alpha = 5.56 \times 10^{-6} \text{ K}^{-1} \text{ (1073K)}$
CeO <sub>2</sub>	$T_m = 2873\text{K}$ $D_{th} = 0.86 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ (1273K)}$ $C_p = 0.47 \text{ Jg}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\lambda = 2.77 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1273K)}$ $E = 172 \text{ GPa (293K)}$ $\alpha = 13 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1500K)}$ $\nu = 0.27 – 0.31$
La <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub>	$T_m = 2573\text{K}$ $D_{th} = 0.54 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ (1273K)}$ $C_p = 0.49 \text{ Jg}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\lambda = 1.56 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1273K)}$ $E = 175 \text{ GPa (293K)}$ $\alpha = 9.1 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1273K)}$
BaZrO <sub>3</sub>	$T_m = 2963\text{K}$ $D_{th} = 1.25 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ (1273K)}$ $C_p = 0.45 \text{ Jg}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\lambda = 3.42 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1273K)}$ $E = 181 \text{ GPa (293K)}$ $\alpha = 8.1 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1273K)}$
TiO <sub>2</sub>	$T_m = 2098\text{K}$ $D_{th} = 0.52 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ (1073K)}$ $\lambda = 3.3 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1400K)}$ $E = 283 \text{ GPa (293K)}$ $\alpha = 9.4 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1500K)}$ $\nu = 0.28$
Garnet (Y <sub>3</sub> Al <sub>5</sub> O <sub>12</sub> )	$T_m = 2243\text{K}$ $\lambda = 3.0 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\alpha = 9.1 \times 10^{-6} \text{ K}^{-1}$
Lanthanum aluminate (LaMgAl <sub>11</sub> O <sub>19</sub> )	$C_p = 0.86 \text{ Jg}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\lambda = 1.7 \text{ Wm}^{-1}\text{K}^{-1} \text{ (1273K)}$ $\alpha = 10.1 \times 10^{-6} \text{ K}^{-1} \text{ (298 – 1473K)}$
LaPO <sub>4</sub>	$T_m = 2343\text{K}$ $\lambda = 1.8 \text{ Wm}^{-1}\text{K}^{-1} \text{ (973K)}$ $E = 133 \text{ GPa (293K)}$ $\alpha = 10.5 \times 10^{-6} \text{ K}^{-1} \text{ (1273K)}$ $\nu = 0.28 \text{ (293K)}$
NiCoCrAlY (bond coat of TBC)	$E = 86 \text{ GPa (293K)}$ $\alpha = 17.5 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1273K)}$ $\nu = 0.3$
IN737 superalloy (Substrate of TBC)	$E = 197 \text{ GPa (293K)}$ $\alpha = 16 \times 10^{-6} \text{ K}^{-1} \text{ (293 – 1273K)}$ $\nu = 0.3$

Symbols in Table 4 have the following meanings:  $D_{th}$ , thermal diffusivity;  $E$ , Young's modulus;  $\alpha$ , thermal expansion coefficient;  $\lambda$ , thermal conductivity;  $C_p$ , heat capacity;  $\nu$ , Poisson's number;  $T_m$ , melting point; TGO, thermally grown oxide on bond coat.

Hsiao et al. [22] patented a method for forming a protective and thermally conductive layer on a work piece. The thickness of the protective layer ranged from 160 to 500  $\mu\text{m}$ . Different alloys were used as the conductive layer including Al/B<sub>4</sub>C, Co-Cr-Al-Y/Al<sub>2</sub>O<sub>3</sub>, Al-Cu-Mo-W, and Cr<sub>3</sub>C<sub>2</sub>-Ni-Cr/SiC-Ni. The heat

conductive layer was applied by a high velocity oxy-fuel (HVOF) spray-coating technique. In a thermal conductivity test, when Silicon Carbide (SiC) was added to Cr<sub>3</sub>C<sub>2</sub>, the thermal conductivity of the coating layer increased from  $\sim 46 \text{ W/mK}$  to  $\sim 71 \text{ W/mK}$ .

Okada et al. [23] patented a method for the production of a conductive sheet. Most conventional heat conductive sheets are prepared from a blend of silicone rubber, Silica (SiO<sub>2</sub>), boron nitride, magnesium oxide, etc. However, they used a conductive resin layer containing binder resin and a heat conductive filler dispersed in the binder. These binder resins were generally composed of two resins such as silicone gel, urethane resin, a synthetic rubber type resin, and a thermoplastic resin of an acrylic type. Different particulate fillers such as silicon carbide, boron nitride, aluminum oxide, aluminum nitride, and mixtures of these particles were tested. Generally, the film thickness was within the range of 1 to 25  $\mu\text{m}$ . The heat conductive sheet was reported to acquire a high heat conductivity of at least  $2.0\text{--}2.6 \text{ W/mK}$ .

Kostamo et al. [24] received a patent for the Atomic Layer Deposition (ALD) of a heat conductive coating. Their method involves depositing on the surface of the substrate at least one thin continuous layer of a first material by ALD, where the first material has a lower heat conductivity than the substrate. The first and/or the second material can be made of material chosen from a group comprising aluminum, magnesium, hafnium, titanium, tantalum, and zirconium. This method may further involve depositing at least one thin continuous layer of a second material by ALD on at least one layer of the first material.

#### 1-D and 2-D composites comprising thermal conductivity

Hill et al. [25] studied the thermal conductivity of platelet-filled polymer composites. Platelet-shaped powders of similar size, shape, and aspect ratio were chosen as fillers for an epoxy composite. The chosen fillers included BN, SiC, TiB<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub>.

Each filler was added to an epoxy molding. The thermal conductivity of the epoxy composites prepared from each of the platelet fillers was measured over a wide range of filler loadings. The findings showed that the increase in the thermal conductivity of the composites filled with platelet-shaped ceramic fillers was practically independent of the intrinsic filler thermal conductivity. Soft filler platelets, such as BN, were found to deform under stress, making it possible to achieve higher density and greater surface contact between particles than observed for the harder, more rigid fillers. The higher density and improved contact appeared to result in the formation of conductive clusters of particles in the composite with improved capability to transfer heat. This deformation and surface contact clearly explained the improved thermal performance of BN-filled composites over that of composites filled with rigid particles, such as TiB<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and SiC, all of which are approximately the same shape as the BN. Hill et al. also suggested a theoretical model, but with some restrictions on the experimental data.

Kurabayashi et al. [26] reported the anisotropic thermal properties of solid polymers. It is well-known that amorphous phases in a polymer, as well as crystalline phases, exhibit significant thermal conduction anisotropy when the polymer chains are partially aligned with each other. This is because the thermal energy transports more efficiently along the polymer chain, which consists of strong carbon-carbon covalent bonds, than perpendicular to the polymer chain, where the thermal energy is carried by the weak van der Waals interaction of molecules.

This behavior is observed in any kind of polymer material, whether crystalline or amorphous.

Shen et al. [27] studied the thermal conductivity of polyethylene fibers. Bulk polymers typically have thermal conductivities on the order of  $0.1 \text{ Wm}^{-1}\text{K}^{-1}$  [28]. However, aligned polymer chains exhibit high thermal conductivity. Commercially oriented polyethylene fibers, with diameters ranging from 10 to 25  $\mu\text{m}$ , have been found to have an enhanced thermal conductivity of  $30\text{-}40 \text{ Wm}^{-1}\text{K}^{-1}$  at around room temperature [29,30]. Shen et al. [27] developed a new technique for fabrication on nanowires. The thermal conductivity of these nanofibers was as high as  $104 \text{ Wm}^{-1}\text{K}^{-1}$ , which is greater than many metals. The fibers were created from a polyethylene gel prepared from Ultra-High Molecular Weight Polyethylene (UHMWPE). The fibers were drawn mechanically at a controlled speed.

Zhou et al. [31] studied thermally conductive composites prepared from a mixture of silicon nitride ( $\text{Si}_3\text{N}_4$ ) particles with a Ultra-High Molecular Weight Polyethylene (UHMWPE)/Linear-Low Density Polyethylene (LLDPE) blend. By adding 0-20 %  $\text{Si}_3\text{N}_4$  filler particles, the composite's thermal conductivity increased from 0.2 to  $\sim 1.0 \text{ Wm}^{-1}\text{K}^{-1}$ . The composite thermal conductivity was further enhanced to  $1.8 \text{ Wm}^{-1}\text{K}^{-1}$  by decreasing the  $\text{Si}_3\text{N}_4$  particle size from 35, 3 and 0.2  $\mu\text{m}$ , and using a coupling agent for the composites with higher filler content.

Models for the calculation of thermal conductivity as a function of filler concentrations

Most of the notation below draws on Nielsen et al. [32], who explored the thermal conductivity of particulate-filled polymers as the basic model for thermal conductivity:

$$\frac{\kappa}{\kappa_1} = \frac{1 + AB\phi_2}{1 - B\psi\phi_2}$$

$$A = \kappa_E - 1$$

$$B = \frac{\frac{\kappa_2}{\kappa_1} - 1}{\frac{\kappa_2}{\kappa_1} + A}$$

$$\psi = 1 + \left( \frac{1 - \phi_m}{\phi_m^2} \right) \phi_2$$

where  $\kappa$  is the thermal conductivity of the composite, and  $\kappa_1$  and  $\kappa_2$  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,  $\psi$  is a function related to the maximum packing fraction  $\phi_m$ , of the filler, and  $\phi_2$  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 of 0.5. The maximum packing fraction  $\phi_m$ , is the ratio of the density of the filler material to the maximum density of the bulk powder.

Zimmer et al. [3] studied the effects of aluminum nanoparticles and carbon nanotubes, with different concentrations, on through-thickness conductivity. The nanocomposites were prepared using epoxy and compared to nanoparticle/fiber-reinforced composites.

Liang et al. [33,34] suggested a theoretical model for heat transfer in polymer composites filled with inorganic hollow micro-spheres. The composites were ternary systems (polymer, glass, and air). This heat transfer mainly involved three mechanisms: (1) thermal conduction between solid and gas; (2) thermal radiation between the hollow micro-sphere surfaces; and (3) natural thermal convection of the gas in the micro-hollow spheres. The following theoretical model was suggested:

$$k_{\text{eff}} = \left[ \frac{1}{k_p} \left( 1 - \frac{6\phi_f}{\pi} \right)^{1/3} + 2 \left( k_p \left( \frac{4\pi}{3\phi_f} \right)^{1/3} + \pi \left( \frac{2\phi_f}{9\pi} \right)^{1/3} \right) \times \left( k_g \left( \frac{\rho_s - \rho_a}{\rho_g - \rho_a} \right) + k_a \left( \frac{\rho_g - \rho_s}{\rho_g - \rho_a} \right) - k_p \right) \right]^{-1}$$

where  $k_{\text{eff}}$  is the effective thermal conductivity,  $\phi_f$  is the volume fraction of filler, and  $\rho_g, \rho_a$ , and  $\rho_s$  are the effective densities of the sphere shell, gas, and micro-sphere, respectively. They showed that the theoretical estimations were similar to the finite element simulations of the effective thermal conductivity of hollow glass bead-filled polypropylene composites at lower concentration of the particles ( $\phi_f \leq 20\%$ ).

Agari et al. [35-37] studied the thermal conductivity of filled polymers and suggested a conduction model, which was supported by experimental data. The model was based on several conduction models of composites. Later work has considered random dispersed particles. In filled composites, thermal conductivity is the highest when conductive blocks are formed, i.e., particles are gathered and arranged parallel to the flux direction. Thermal conductivity is the lowest when these particle blocks are arranged in series in the direction of the heat flux. These parallel and series conductions are expressed by the following equations, respectively:

*Parallel conduction:*

$$(1) \lambda = V \cdot \lambda_2 + (1 - V) \cdot \lambda_1$$

*Series conduction :*

$$(2) \frac{1}{\lambda} = \frac{V}{\lambda_2} + \frac{(1 - V)}{\lambda_1}$$

where  $\lambda$  = the thermal conductivity of the composite,  $\lambda_1$  = the thermal conductivity of polymer,  $\lambda_2$  = the thermal conductivity of the particles, and V = the volume content of the particles. Agari et al. assumed that a combined equation should be used:

$$(3) \log \lambda = V \cdot C_2 \cdot \log \lambda_2 + (1 - V) \cdot \log (C_1 \cdot \lambda_1)$$

where  $C_2$  is the ease factor in forming conductive chains of particles and is smaller than 1 and larger than 0. In the preparation of a composite, particles can affect crystallinity and the crystal size of polymer and change the thermal conductivity of the polymer. This effect is taken into consideration and  $\lambda$ , is replaced by  $C_1\lambda_1$ . Some values of  $C_1$  and  $C_2$  are listed in Table 5.

**Table 5:** Values of  $C_1$  and  $C_2$  [36].

Materials	$C_1$	$C_2$
Polyethylene filled with graphite	0.898	0.882
Polyethylene filled with copper	1.073	0.888
Polyethylene filled with $Al_2O_3$	0.859	0.902
Polyamide filled with graphite	1.017	0.922
Polystyrene filled with graphite	1.024	0.892

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