



Review

Thermal conductivity of transparent and flexible polymers containing fillers: A literature review



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ABSTRACT

Advances in micro and nanotechnology have motivated a number of recent studies on transparent and flexible polymers (TFP). In this study, a literature review on a thermal conductivity (TC) of these polymers containing fillers is first presented. Three major types of TFPs considered are poly-dimethyl siloxane (PDMS), polyethylene terephthalate (PET), and polyimides (PI). It was found from the literature review that carbon-based fillers, particularly multi-walled carbon nanotubes, and ceramic fillers have been considered as the most promising candidate for the TC enhancement of PDMS and PI, respectively. However, since very limited number of studies considered the TC of PET composites, it becomes a motivation for further investigations. This study provides better understandings in the research trends of polymer sciences, particularly the TFPs.

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1. Introduction

1.1. Thermal conductivity of pure polymers

Thermal conductivity (TC or k) is an important property in material science, research, electronics, building insulation, and related fields, particularly where high operating temperature is required. It characterizes the ability of a material to conduct heat flow. Recently, TC has been studied extensively for both polymer applications and processing. However, polymers typically have

intrinsic TC that is much lower than those of metals or ceramic materials, making them good thermal insulators. The TCs of some well-known polymers listed in Table 1 indicate that the TC of pure polymers is commonly very low and less than 0.5 W/(m·K).

The TC of a material is evaluated primarily by Fourier's law in steady-state thermal conduction. However, for polymer composite materials, the Debye kinetic model is usually used for theoretical prediction [4].

$$k = \frac{C_p v \lambda}{3}$$

where C_p is the specific heat capacity per unit volume, v is the average velocity of phonon, and λ is the phonon mean free path (i.e.

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Table 1
TCs of some well-known polymers at room temperature [1–3].

Polymer name	k [W/(m·K)]
High density polyethylene (HDPE)	0.44
Polyphenylsulfone (PPSU)	0.35
Poly(ethylene vinyl acetate) (EVA)	0.34
Poly(acrylonitrile–butadiene–styrene) copolymer (ABS)	0.33
Low density polyethylene (LDPE)	0.30
Polyphenylene sulfide (PPS)	0.30
Poly(butylene terephthalate) (PBT)	0.29
Polytetrafluoroethylene (PTFE)	0.27
Nylon-6,6 (PA66)	0.26
Nylon-6 (PA6)	0.25
Polyetheretherketone (PEEK)	0.25
Polytetrafluoroethylene (PTFE)	0.25
Poly(dimethylsiloxane) (PDMS)	0.25
Polysulfone (PSU)	0.22
Polymethylmethacrylate (PMMA)	0.21
Polycarbonate (PC)	0.20
Polyvinyl chloride (PVC)	0.19
Polyvinylidene difluoride (PVDF)	0.19
Epoxy resin	0.19
Poly(ethylene terephthalate) (PET)	0.15
Polystyrene (PS)	0.14
Polypropylene (PP)	0.12
Polyethylene (PE)	0.11
Polyimide, Thermoplastic (PI)	0.11

average distance that a phonon travels between two collisions). According to the quantum mechanics, waves are particles. Therefore, the elastic waves (or sound waves) arisen from lattice vibration of immobile positive ions and free electrons (electron gas) in a solid are particles too, and these particles are called “phonons”. In this regard, a solid can be considered as a dilute gas of phonons. For an isotropic solid structure, the TC can be determined by Debye kinetic model mentioned above, and the phonon mean free path in solid form of polymers can be considered as it in gas matters. Lopes and Felisberti [5] indicated that λ is a main parameter affecting the TC of composite materials. Furthermore, for amorphous polymers, λ is an extremely small constant (i.e. a few angstroms) due to phonon scattering from numerous defects. That is why the TC of polymers is very low, as mentioned previously.

The TC of various polymers can be found in a polymer handbook written by Yang [6] for more detail. Measurement techniques were provided by Tritt for determining the TC of a bulk material, such as the radial flow method, laser-flash diffusivity, and pulse-power method [7]. The membrane method, bridge method, and photothermal emission method for thin-film materials have also been

Table 2
TCs of highly conductive fillers at room temperature [10–13].

Group	Filler	k [W/(m·K)]
Carbon-based fillers	Carbon nanotubes (CNT)	2000–6000
	Diamond	2000
	Pitch-based carbon fiber	530–1100 (along the axis)
	Graphite	100–400 (on plane)
	Carbon black (CB)	6–174
	PAN-based carbon fiber	8–70 (along the axis)
Metallic fillers	Copper (Cu)	483
	Silver (Ag)	450
	Gold (Au)	345
	Aluminum (Al)	204
	Nickel (Ni)	158
Ceramic fillers	Boron nitride (BN)	250–300
	Beryllium oxide (BeO)	260
	Aluminum nitride (AlN)	200
	Aluminum oxide (Al ₂ O ₃)	30

considered. These approaches are very useful for understanding the physical properties and thermal behaviors of polymers, particularly the TC concerned in this study.

1.2. Thermal conductivity of highly conductive fillers

Advanced polymer composites reinforced with organic or inorganic fillers are becoming more and more common for applications that require higher TC, such as electronic packaging and encapsulations, satellite devices, and areas where good heat dissipation, low thermal expansion, and light weight are needed [8,9]. The TC of polymers has traditionally been enhanced by the addition of highly conductive fillers, including graphite, carbon black, carbon fibers, and ceramic or metal particles. Table 2 [10–13] shows that the TC of fillers is much higher than that of the pure polymers shown in Table 1. For example, the TC of planar graphite reported is very high, up to 3000 W/(m·K) obtained in [14].

It is evident that polymer composites filled with thermally conductive fillers have many advantages due to their easy processability, low cost, and durability against corrosion. In addition, Tavman [15] provided the following main reasons for using fillers: (1) improving thermal conductivity and diffusivity; (2) obtaining a certain degree of electrical conductivity, magnetic permeability; (3) stiffening the matrix and making it more rigid; (4) reducing internal stresses in injection molded parts; (5) reducing creep; (6) reducing the cost of the material; (7) improving the appearance of the final product.

Fillers can be classified into three common groups: carbon-based fillers, metallic fillers, and ceramic fillers, as shown in Table 2 [4]. Carbon-based fillers are known as ideal thermally conductive fillers for polymer composites because they have a higher TC, excellent corrosion resistance, and a lower thermal expansion coefficient than metals. King et al. [16] studied TC variation by adding three different types of carbon (carbon black, synthetic graphite particles, and carbon fiber) to Vectra A950RX Liquid Crystal Polymer. They indicated that for single filler systems, synthetic graphite caused the largest increase in TC. For combinations of fillers, the composites containing carbon black and synthetic graphite caused the largest increase in composite TC. In addition, many studies demonstrated that the TC of polymer composites increases with the filler content. Such fillers include plasticized carbon black [17], vapor-grown carbon nanofiber (VGCF) [18], and multi-walled carbon nanotubes (MWNTs) [19]. Polymer composites filled with VGCF are estimated to have a very high TC of 1260 W/(m·K) [20], and tenfold improvement in TC compared to pure epoxy [21].

The TC of polymers can be enhanced using metallic fillers, such as aluminum powder [22,23], copper powder, or other particles [24,25]. Tekce et al. [26] indicated that the TC of copper-filled polyamide composites depends on the TC of the filler particles, their shape and size, volume fraction (VF), and spatial arrangement in the polymer matrix. Ishida and Rimdusit [27] achieved a TC of 32.5 W/(m·K) with a boron nitride-filled polybenzoxazine at a maximum filler loading of 78.5% by volume (vol%), or 88% by weight (wt%). The effects of boron nitride (BN) content, the particle size of HDPE, and the temperature on the TC of the composites were investigated by Zhou et al. [28]. The results indicated that the TC is higher for the larger size HDPE than for the smaller one. The TC increases with increasing filler content. In addition, the combined use of BN particles and alumina short fiber results in higher TC of the composites compared to when only BN particles are used.

By adding spherical aluminum nitride (AlN) into polymers, Ohashi et al. [29] achieved an excellent TC of 8.2 W/(m·K). Yu et al. [30] also indicated that the TC of composites filled with AlN reinforcement is higher with larger particle size. The TC is five times that of

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