



## Recent progress on thermal conductive and electrical insulating polymer composites

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### ABSTRACT

Thermal conductive and electrical insulating polymer composites have gained more and more attention due to their vital importance for various applications, especially electronic packing, thermal management, etc. Herein, recent progress in this field are reviewed to provide guidelines for researchers. Filler variety, filler network morphology, filler size, the use of hybrid filler, the nano-structure of filler, the selective distribution of filler and thermal conductivity of polymer matrix are considered as important issues influencing the final thermal and electrical properties. This paper represents a short review to systematically understand the enhancement in thermal conductivity and electrical resistivity. Then, future perspectives for thermal conductive and electrical insulating polymer composites are briefly discussed.

### 1. Introduction

Heat transferring is a widely concerned issue that affects the performance, reliability and lifetime of electronic devices. In the past decades, microelectronics are widely developed which demands those products to be more miniaturized, lighter, more efficient and multi-functional. The miniaturization, densification, high frequency and power inevitably lead to frequently aggregated heat and overheated devices. The investigation and application of materials with both high thermal conductivity and excellent electrical insulation are urgently needed.

Considering their electrical insulation, lightweight, low-cost and good processing ability, polymers have been widely studied for electronics packaging [1,2]. However, unlike metal and inorganic materials, the thermal conductivity of polymers is quite low, usually in the range of 0.1~0.5 W/(mK) [3]. This greatly limits the application of polymers for heat transfer devices. Polymer composites with ultrahigh thermal conductive fillers have gained more and more attention and considered to be the next generation heat transfer devices in electronics. Typical thermal conductive fillers, including aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) [4,5], aluminum nitride (AlN) [6], boron nitride (BN) [7–9], graphene [10] and carbon nanotube [3,11], are usually introduced into polymers by solution mixing or melt processing. The range of thermal conductivity can be controlled by conductive filler content or filler morphology. Generally, the addition of ceramic fillers such as BN does not change the electrical insulation properties of polymer matrix, while the addition of carbon fillers with ultrahigh electrical conductivity

might largely damage the electrical insulation properties.

In this short review, conductive fillers are discussed systematically to determine the effect on both thermal conductivity and electrical insulation properties of polymer composites. The varieties of fillers, morphologies, size, the use of hybrid fillers, the nano-structure of fillers, the selective distribution of fillers and the thermal conductivity of polymer matrix are considered as the main issues influencing the thermal and electrical properties. Recent research progress are reviewed to systematically understand the enhancement in thermal conductivity and electrical resistivity. Then, future perspectives for thermal conductive and electrical insulating polymer composites are discussed at last.

### 2. Thermal conductive mechanism of polymer composites

#### 2.1. The mechanism of heat transferring in solid

Microscopically speaking, heat transferring is a process that conducted by micro-particles such as molecules or atoms vibration and transferring within adjacent ones. In solid, there are two kinds of conduction mechanism: free electrons conduction and phonons conduction. Free electron conduction mainly exists in metals. While polymers, which are mostly saturated and non-free electrons, can only rely on the lattice vibration as well as phonons conduction to transfer heat. Thus, increasing the crystallinity of polymers is an efficient approach to enhance the thermal conductivity, but it is rather difficult to realize it during application due to the range of crystallinity could be achieved is

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rather small.

## 2.2. The thermal conductive mechanism of polymer composites

It is regarded that the thermal conductive mechanism of intrinsic polymers is rather easy to understand, but as for thermal conductive polymer composites, the mechanism has not been clearly discussed yet. Three main explanations are introduced here: the thermal conductive pathway (network) mechanism [12]; the thermal conductive percolation mechanism [13] and the thermoelastic coefficient mechanism [14]. Generally, the pathway mechanism and the percolation mechanism are accepted and mostly used by researchers. The pathway mechanism indicates that conductive fillers are isolated and surrounded by polymer matrix at low content. With increasing filler content, thermal conductive network is established by fillers connecting with each other. Hence, heat transfers through the conductive network of fillers, which largely enhances the thermal conductivity of polymer composites. While the percolation mechanism is applicable to fillers with ultrahigh thermal conductivity, such as graphene and carbon nanotubes (CNTs). Researchers indicate that percolation phenomenon is difficult to be observed in most composites. However, the thermal conductivity can be lifted dramatically when the content of graphene or CNTs reaches the percolation value, which is mostly responsible to the percolation behavior of conductive fillers. Therefore, the choice of conductive fillers and the aggregative state determine the thermal conductivity of polymer composites to a great extent, and that will be discussed at large in this review.

## 3. Novel design and recent progress on achieving thermal conductive and electrical insulating polymer composites

### 3.1. Present problems on achieving high thermal conductive (TC) and electrical insulating (EI) polymer composites

Polymer composites with excellent TC and EI properties have been widely studied in recent years, but problem still exists and demands urgent solutions. Especially, on one hand, high thermal conductivity is usually obtained by adding a large amount of fillers, which inevitably results in the destruction of mechanical properties of polymer composites; on the other hand, ultrahigh TC fillers like graphene and CNTs will bring about additional electrical conductivity increasing simultaneously, which is not required for TC and EI polymer composites.

### 3.2. Thermal conductivity of bulk polymers

The thermal conductivity of polymer matrix has a crucial effect on the overall thermal conductivity of polymer composites. However, bulk polymers are usually known as thermal insulators, with thermal conductivity of about 0.1–0.5 W/mK. Generally speaking, polymers with rigid aromatic backbones display higher thermal conductivity, such as polyacetylene, poly(p-phenylene benzobisthiazole), poly(p-phenylene), polythiophene [15]. It is because that the  $\pi$ - $\pi$  conjugated structure in rigid backbones benefits the transferring of phonon. In addition, the orientation of molecular chains is also investigated recently to enhance the thermal conductivity of polymer matrix. Shearing, mechanical drawing and spinning are developed to induce the orientation of PE molecular chains [16–18]. And it is found that the thermal conductivity parallel to the orientation direction increased significantly comparing to neat PE, while the thermal conductivity perpendicular to the orientation direction changed slightly.

### 3.3. Materials used for TC and EI polymer composites

#### 3.3.1. TC fillers

Due to the rather low thermal conductivity of polymer matrix, TC fillers with high thermal conductivity are introduced into polymer

**Table 1**  
Thermal conductivity of various fillers at room temperature.

	Thermal conductive fillers	Thermal conductivity (W/mK)
Metallic fillers	Copper(Cu)	483
	Silver (Ag)	450
	Gold (Au)	345
	Aluminum (Al)	204
Ceramic fillers	Silicon (Si)	200
	Aluminum oxide (Al <sub>2</sub> O <sub>3</sub> )	20–29
	Silicon dioxide (SiO <sub>2</sub> )	1–10
	Zinc oxide (ZnO)	26
	Boron Nitride (BN)	250–300
	Aluminum nitride (AlN)	200
	Silicon Nitride (Si <sub>3</sub> N <sub>4</sub> )	180
Silicon carbide (SiC)	80–120	
Carbon fillers	Carbon black	6–174
	Diamond	2000
	Graphite	100–400
	Carbon Fibre	8–70
	Graphene	4840–5300
	MWCNT	3180
	SWCNT	3500

composites. Ordinarily, high filler loading (> 30 vol%) is required to obtain significant enhancement in thermal conductivity. Table 1 shows the thermal conductivity of typical TC fillers. The addition of metallic fillers might result in both increase in thermal conductivity and electrical conductivity. Besides, the increase of composites density and the poor oxidation resistance will limit the application in many situations. Ceramic fillers such as boron nitride(BN), silicon carbide(SiC) [19,20] with excellent thermal conductivity and electrical resistivity, are ideal TC and EI fillers. The particle size, particle morphology, surface modification and filler distribution are systematically studied to determine their effect on thermal conductivity of polymer composites. Carbon fillers have both ultrahigh thermal conductivity and electrical conductivity. The intrinsic thermal conductivity of CNTs is about 3000 W/mK in axial direction [21], 10 times higher than most ceramic fillers. However, the introduction of carbon fillers results in rapid increase on electrical conductivity simultaneously. Special surface treatments are often required to avoid the damage of electrical insulation property.

#### 3.3.2. Single ceramic filler system

The advantage of composites with one kind of ceramic filler is that it will maintain its electrical resistance no matter how much loading of fillers is used. In fact, to gain such ceramic/polymer composites with largely enhanced thermal conductivity, the loadings is typically up to 30 vol%, even as high as 70 vol% [4,6], which cause great sacrifice in mechanical properties and processing ability. Chao Jiang et al. [6] fabricated AlN/Hydantion epoxy composites and the flexural strength decreases sharply with 58 vol% AlN loadings. Moreover, high filler loading will introduce more interface between fillers and polymer matrix, results in rapid increased interfacial thermal resistance. It is believed that morphology tuning of ceramic fillers is necessary to overcome above shortcomings. Yong Hu et al. [4] prepared Al<sub>2</sub>O<sub>3</sub>/epoxy composites by using a novel processing technique consisting of gel-casting, sintering and vacuum infiltration methods. They found that comparing to homo-dispersed Al<sub>2</sub>O<sub>3</sub> or simple physical contacted Al<sub>2</sub>O<sub>3</sub>, the sintered Al<sub>2</sub>O<sub>3</sub> were chemical bonded with neighboring particles, so that it provides more TC pathway and less interfacial thermal resistance. Chemical bonding also strengthens the mechanical properties of composites. As a result, high Al<sub>2</sub>O<sub>3</sub> loadings up to 70 vol% were achieved and the flexural strength and thermal conductivity of the Al<sub>2</sub>O<sub>3</sub>/epoxy composites reached as high as 305 MPa and 13.46 W/mK, respectively, comparing to 121 MPa and 3.60 W/mK at Al<sub>2</sub>O<sub>3</sub> loadings of 55 vol% with no chemical bonding.

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