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Filler Selection and Fabrication Methods for Highly Thermally Conductive Polymer Composites: A Review

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Abstract: In high-power electronic devices, the thermal conductivity of polymer composites is governed not simply by the intrinsically high thermal conductivity of the fillers, but by their ability to form continuous heat conduction pathways with few structural defects within the polymer matrix. This review examines the selection of carbon-based, ceramic, and metallic fillers, and discusses how hybrid filler systems, filler loading, and surface modification influence filler dispersion, interparticle distance, interfacial contact, and network formation. Vacuum-assisted filtration, template-assisted methods, electrospinning, field-assisted alignment, and three-dimensional printing are further considered in terms of their effects on filler arrangement and connectivity. Filler size, morphology, surface chemistry, and field responsiveness guide fabrication method selection. Fabrication then determines thermal network continuity. Future research needs to develop efficient thermal pathways at reduced filler content and porosity using stable, scalable processes.

Keywords: Thermal conductivity; Polymer; Thermal conduction networks; Fillers

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1. Introduction

Electronic devices continue to shrink as integration increases and power density rises. Heat accumulation in confined spaces undermines reliable operation throughout their service life [1]. Electronic devices require electrically insulating materials that dissipate heat without sacrificing flexibility or processability at low density. Polymers meet these requirements but generally have low intrinsic thermal conductivity. Adding thermally conductive fillers is therefore a common route to enhancing polymer thermal conductivity [2].

Thermally conductive fillers are commonly classified as carbon, ceramic, or metallic fillers. These filler types differ in intrinsic thermal conductivity and surface chemistry [3–7]. Fillers occur in particulate, fibrous, or platelet forms, with differences in size and geometric ratio. These characteristics affect their dispersion,

contact, orientation, and network formation within the polymer matrix [8–10]. Increasing filler loading reduces filler spacing and promotes contact. At high loadings, mixture viscosity rises, hindering processing and introducing defects that limit further gains in thermal conductivity [11–12]. Hybrid fillers with different sizes or morphologies improve network connectivity by filling the spaces between larger fillers and bridging adjacent particles. Improved connectivity depends on filler ratio and distribution [13–14]. Surface modification can improve filler dispersion and interfacial contact [15]. Suitable fabrication methods and processing conditions are determined by how filler type, size, morphology, loading, and surface chemistry affect filler dispersion, mixture viscosity, and filler orientation. Each method controls filler arrangement, connectivity, and orientation while requiring appropriate filler morphology and dispersion [16–18]. Common methods for preparing thermally conductive polymer composites include vacuum-assisted filtration, template-assisted methods, electrospinning, methods assisted by external fields, and three-dimensional printing [19–25]. Fabrication methods control filler distribution and connectivity to form heat conduction pathways with different orientations and continuity.

This review examines thermally conductive filler selection and how hybrid fillers, filler loading, and surface modification affect filler dispersion and contact in polymer composites. It also discusses how fabrication methods control filler network connectivity in polymer composites, providing guidance for filler selection and process design.

2. Selection and design of thermally conductive fillers

Filler selection requires compatibility with the polymer matrix and fabrication process rather than consideration of intrinsic thermal conductivity alone. Controlling filler formulation and surface chemistry improves dispersion and contact, promoting continuous heat conduction pathways.

2.1. Selection of fillers

Table 1 summarizes the thermal conductivities of different fillers. Carbon-based fillers provide high intrinsic thermal conductivity while remaining lightweight and chemically stable. In graphene and carbon nanotubes, strong sp^2 covalent bonds form ordered hexagonal structures that support rapid phonon transport along the graphene plane and nanotube axis [4]. Spinelli et al. incorporated multiwalled carbon nanotubes into epoxy resin. At a filler loading of only 0.5 wt%, the in-plane thermal conductivity of the composite increased by approximately 60% compared with that of neat epoxy [8]. Ceramic fillers conduct heat mainly through lattice vibrations and generally offer high electrical resistivity, thermal stability, and oxidation resistance. Al_2O_3 is inexpensive and chemically stable but has relatively low thermal conductivity. AlN has higher thermal conductivity but is susceptible to hydrolysis, while h BN combines high in-plane thermal conductivity with electrical insulation but is difficult to disperse uniformly in polymers. Xu et al. incorporated spherical Al_2O_3 into epoxy resin, achieving a thermal conductivity of 0.679 W m^{-1} K^{-1} at 50 vol%, approximately 270% higher than that of neat epoxy [9]. Fang et al. blended platelet BN with polyamide 6, increasing the thermal conductivity from 0.30 to 1.36 $W \cdot m^{-1} \cdot K^{-1}$ at 50 wt% BN [10]. Metallic fillers generally possess high intrinsic thermal conductivity because heat is transferred by both free electrons and lattice vibrations. Copper, aluminum, and iron readily form surface oxides that increase thermal resistance between fillers, whereas gold and silver resist oxidation but are expensive. Rösel et al. incorporated platelet copper into epoxy resin,

achieving an in-plane thermal conductivity of 7.9 $W \cdot m^{-1} \cdot K^{-1}$ at 50 vol%, approximately 1217% higher than that of neat epoxy [11]. Alhamidi et al. added spherical aluminum nanoparticles to a polybutylene terephthalate and polyethylene terephthalate matrix, reaching 0.59 $W \cdot m^{-1} \cdot K^{-1}$ at 25 vol%, about twice that of the unfilled matrix [12]. The differences among fillers in thermal conductivity, electrical properties, density, stability, and processability provide a basis for hybrid filler design to improve filler contact and heat conduction pathways.

Table 1. Thermal conductivity of various fillers

Filler	λ (W/mK)	Refs.	Filler	λ (W/mK)	Refs.	Filler	λ (W/mK)	Refs.
Au	345	[3]	Al_2O_3	20–29	[3]	AlN	200	[3]
Ag	450	[3]	ZnO	26	[3]	Diamond	2000	[3]
Cu	483	[3]	BeO	260	[6]	Graphite	100–400	[3]
Al	204	[3]	MgO	50	[7]	Graphene	5000	[4]
Fe	80	[5]	BN	400	[3]	CNT	3180–3500	[3]
SiC	80–120	[3]	Si_3N_4	180	[3]	Carbon Fibre	8–70	[3]

2.2. Hybrid filler design

Hybrid filler design combines fillers with different sizes or morphologies to fill spaces, bridge adjacent particles, and reduce platelet stacking [2]. Wang et al. combined platelet BN and AlN in epoxy resin [14]. At a total loading of 30 wt% and a BN to AlN mass ratio of 4:1, the thermal conductivity reached 1.278 $W \cdot m^{-1} \cdot K^{-1}$, 21.4% higher than that of the AlN-only composite. Choi et al. combined alumina microparticles with alumina nanowires in epoxy resin [15]. The composite containing 20 wt% microparticles and 10 wt% nanowires showed a thermal conductivity 109% higher than that containing 30 wt% microparticles alone because the nanowires connected adjacent particles and improved pathway continuity.

2.3. Filler loading

At low filler loadings, isolated fillers provide limited improvement because heat still passes mainly through the polymer matrix. Increasing filler loading reduces filler spacing and promotes contact, forming more continuous heat conduction pathways. Thermal conductivity may still increase gradually without a clear percolation threshold because interfacial thermal resistance, poor filler contact, pores, and structural defects cause phonon scattering [2]. Excessive filler loading increases mixture viscosity and may cause agglomeration, poor contact with the polymer matrix, and residual pores. Filler loading should therefore balance network connectivity, processability, and performance. High aspect ratio fillers, hybrid fillers, and preconstructed networks can improve filler contact at lower loadings.

2.4. Filler surface modification

Filler surface modification can be achieved through covalent bonding or noncovalent interactions. Covalent modification grafts functional groups through chemical bonds, whereas noncovalent modification relies on physical adsorption, hydrogen bonding, electrostatic interactions, or π-π interactions [2]. Surface modification can improve filler dispersion and strengthen contact with the polymer matrix and between adjacent fillers to promote heat transfer. Tang et al. grafted KH550 onto BN, increasing the thermal conductivity of the epoxy composite from 0.433 to 0.615 $W \cdot m^{-1} \cdot K^{-1}$ at a filler loading of 10 wt% [16]. Yuan et al. introduced hydroxyl groups onto BN nanosheets using H_2O_2 to improve their interaction with polyurethane, increasing the thermal

conductivity from 1.672 to 3.097 $W \cdot m^{-1} \cdot K^{-1}$ at a filler loading of 30 wt% [17]. He et al. modified spherical AlN to improve its contact with silicone rubber; at 75 wt%, the thermal conductivity increased from 0.95 to 1.14 $W \cdot m^{-1} \cdot K^{-1}$ [18]. Surface modification may increase thermal resistance or damage the filler structure. Oxidation introduces defects into the graphene lattice and increases phonon scattering [4]. Surface modification should therefore be controlled to improve filler dispersion and contact without introducing excessive thermal resistance or structural defects.

3. Fabrication methods for thermally conductive polymer composites

After selecting the filler type, morphology, loading, and surface state, appropriate fabrication methods are required to control filler dispersion, arrangement, and connectivity. Different fabrication methods impose different requirements on filler size, dispersion stability, mixture viscosity, and forming conditions, and produce thermally conductive filler pathways with different orientations and degrees of connectivity.

Vacuum-assisted filtration begins by uniformly dispersing thermally conductive fillers together with a polymer or polymer fibers in a liquid. During filtration, the fillers usually align parallel to the membrane, forming dense in-plane heat conduction pathways. This method is particularly effective in arranging and connecting platelet and fibrous fillers. These connected fillers form heat conduction pathways along the film plane. Dong et al. mixed rGO with an aramid nanofiber dispersion and prepared a composite film by vacuum-assisted filtration [19]. An in-plane thermal conductivity of 12.31 $W \cdot m^{-1} \cdot K^{-1}$ was achieved through parallel alignment of the rGO sheets, which formed continuous heat conduction pathways along the film plane. Vacuum-assisted filtration is suitable for preparing composite films with high filler loading and high in-plane thermal conductivity.

Template-assisted methods use removable templates to construct continuous filler networks for subsequent polymer infiltration. This approach enables continuous heat conduction pathways at low filler loadings. Wang et al. used ice templating to align AlN particles and Si_3N_4 nanowires [20]. At loadings of 65 wt% AlN and 1.5 wt% Si_3N_4 nanowires, the composite achieved a thermal conductivity of 1.64 $W \cdot m^{-1} \cdot K^{-1}$. Han et al. used NH_4HCO_3 particles as removable templates to construct a continuous BN network, followed by silicone rubber infiltration [21]. At a BN loading of 40.7 vol%, the thermal conductivity reached 3.89 $W \cdot m^{-1} \cdot K^{-1}$. Slurry concentration and freezing rate should be controlled to maintain network continuity.

Electrospinning uses a high electric field to form continuous polymer fibers containing thermally conductive fillers. The stretching of the jet can align elongated fillers along the fiber axis, producing heat conduction pathways along the deposited fibers. Zhang et al. electrospun thermoplastic polyurethane containing multiwalled carbon nanotubes and BN, followed by hot pressing [22]. At a total filler loading of 40 wt%, the membrane achieved an in-plane thermal conductivity of 7.28 $W \cdot m^{-1} \cdot K^{-1}$. Hot pressing reduced pores and increased contact between fibers.

Electric or magnetic fields can orient anisotropic fillers before polymer curing. Fillers with insufficient field response can be modified with dielectric or magnetic components to promote alignment. Jia et al. deposited Fe_3O_4 nanoparticles on BN nanosheets and aligned them through the thickness under a magnetic field [23]. At a filler loading of 40 wt%, the through-plane thermal conductivity reached 3.65 $W \cdot m^{-1} \cdot K^{-1}$. Xu et al. used the intrinsic diamagnetic anisotropy of carbon nanotubes to align them in a polymer [24]. At a loading of 10 wt%, the composite achieved a thermal conductivity of 1.10 $W \cdot m^{-1} \cdot K^{-1}$. Filler alignment is affected

by field strength, exposure time, mixture viscosity, and curing rate. High viscosity restricts filler rotation, whereas low viscosity may cause sedimentation.

Three-dimensional printing extrudes filler-containing polymer melts or slurries through a nozzle along a predefined path. Shear within the nozzle can orient platelet or fibrous fillers along the printing direction and form directional heat conduction pathways. Xiao et al. prepared BN and PDMS composites by direct ink writing [25]. At a BN loading of 40 wt% and a printing speed of 120 $mm \cdot s^{-1}$, the in-plane thermal conductivity reached 0.849 W·m-1·K-1, 87% higher than that of the compression-molded sample with the same filler loading. Mixture viscosity and printing direction should be controlled to ensure continuous extrusion, structural integrity, and continuous heat conduction pathways. Excessive viscosity may cause nozzle blockage, whereas insufficient viscosity may lead to structural collapse.

4. Conclusions and outlook

Filler type, size, morphology, loading, and surface chemistry determine filler dispersion, spacing, contact, and interaction with the polymer matrix. High thermal conductivity therefore depends not only on filler loading, but also on low interfacial thermal resistance and continuous filler networks. Filler design must be matched with appropriate fabrication methods for polymer composites to convert the intrinsic thermal conductivity of fillers into effective heat conduction throughout the composite. Future research should clarify how filler characteristics, network structure, pores, and processing conditions affect thermal conductivity. Continuous heat conduction pathways should be constructed at low filler loadings using reliable and scalable fabrication methods.

Disclosure statement

The author declares no conflict of interest.

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