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Review of Modification Ways for Thermal Interface Materials

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Abstract. The rapid increase in power density in modern electronic devices has elevated thermal management to a critical technological challenge. Thermal interface materials (TIMs) are essential for efficient heat dissipation, and their performance is highly dependent on the incorporation and optimization of functional fillers. Numerous modification strategies have been developed, but there remains a lack of systematic guidance for selecting and integrating these approaches based on specific application requirements. This review categorizes TIM enhancement strategies into three primary approaches: filler selection, surface modification, and structural design. Filler selection is analyzed with respect to material type, morphology, and thermal conductivity, highlighting their suitability for different operational scenarios. Surface modification techniques are examined for their ability to reduce interfacial thermal resistance and improve filler-matrix compatibility. Structural design strategies, including three-dimensional networks and hierarchical architectures, are discussed for their role in overcoming intrinsic performance limitations. Integrating these strategies, a three-step decision framework is proposed, linking material choice, interface engineering, and structural optimization in a coherent workflow. This framework provides a practical guide for the rational design of high-performance TIMs and offers insights into tailoring materials to meet the thermal management demands of diverse electronic applications. The review aims to bridge theoretical understanding and practical implementation, supporting more efficient and reliable electronic device operation.

Keywords: Thermal Interface Materials, Modification Strategies, Filler Selection, Structural Design

1. Introduction

As the power density of electronic devices continues to increase, conventional thermal interface materials (TIMs) are increasingly unable to meet thermal management requirements due to their intrinsic low thermal conductivity. Enhancing TIM performance requires a synergistic integration of three complementary strategies: filler selection, surface modification, and structural design. Current research primarily emphasizes the incorporation of high-thermal-conductivity fillers to form composite TIMs. This approach aligns with the principles of framework materials, which involve precise design and multiscale control of both the composition and architecture of the composite to optimize heat transfer pathways. This paper systematically examines the three key strategies—filler selection, surface modification, and structural design—highlighting how their coordinated

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application can significantly improve the thermal performance of TIMs for advanced electronic applications.

2. Modification methods: examples and suggestions

2.1. Filler change—the basic parts for building good heat conduction

Filler modification typically involves physically or chemically altering the inherent properties, structure, or phase of the filler to address its natural limitations in application. Selecting the appropriate shape, size, and dimensional structure from among various filler types is a critical step in optimizing performance. Commonly used fillers include metals, ceramics, and carbon-based materials, each offering distinct advantages and challenges in thermal interface applications [1].

2.1.1. Metal fillers

Metal fillers follow the electron heat conduction mechanism, so they have good points like high thermal conductivity and thermal stability [1]. Normal metal filler choices are all about picking from metals like gold, silver, copper, and they are usually put in as powder [1]. But because the metal powder's size makes the effect somewhat limited, we need to change to more flexible shapes, and this change points right to the study of metal nanowires.

Metal nanowires (NWs) are a one-dimensional nanostructure material. Because they have a high length-to-diameter ratio, they can more easily make a complete heat conduction loop inside the polymer than powder fillers, since you can put more in and they have more surface area. Also, metal nanowires, because of the high ratio, help make a continuous heat conduction path in the polymer, so it lowers the percolation threshold. For example, copper nanowires CuNWs because of the high ratio, can make the composite thermal conductivity go up about 20% at a very low amount (0.15 vol%however), showing it can make very good heat conduction networks [2]. On the other hand, because 1D metal NWs have a high ratio, they can have line contact or even face contact with each other, and this bigger contact area will greatly lower the thermal resistance coming from heat transfer, which is better than the point contact of nanoparticles.

But the high surface area from the NWs structure isn't only a good thing; for example, there is a big interface area between the NWs and the polymer, and if the interface bonding isn't good, it will block heat transfer, and this block might cancel out or be worse than the good results.

2.1.2. Carbon-based fillers

Carbon-based fillers have attracted considerable attention due to their superior thermal conductivity and availability in multiple dimensional forms [3]. Two-dimensional graphene exhibits an in-plane thermal conductivity theoretically reaching ~5300 $W \cdot m^{-1} \cdot K^{-1}$, while one-dimensional carbon nanotubes (CNTs) possess axial thermal conductivities between 3000–6000 $W \cdot m^{-1} \cdot K^{-1}$, significantly exceeding those of conventional metallic copper (~400 $W \cdot m^{-1} \cdot K^{-1}$) and ceramic fillers such as boron nitride (~400 $W \cdot m^{-1} \cdot K^{-1}$) [3]. However, this high performance introduces challenges. First, the intrinsic electrical conductivity of carbon fillers can create insulation issues in certain applications. Second, the large surface area from zero-dimensional carbon black to two-dimensional graphene promotes aggregation and increases interfacial thermal resistance. Consequently, the combination of surface modification and structural design is critical to retain high thermal conductivity while ensuring proper dispersion and functional performance [3].

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Carbon-based materials exist in various dimensional forms, each with distinct advantages. Zero-dimensional carbon black is inexpensive but prone to aggregation, limiting thermal conductivity enhancement. One-dimensional CNTs possess high aspect ratios, enabling network formation, mechanical reinforcement, and electrical conductivity. Two-dimensional graphene and graphene nanoplatelets (GNPs) provide sheet-like structures that facilitate efficient planar heat conduction and can form high-performance thermal conduction layers. Three-dimensional carbon sponges offer pre-formed skeletal networks that serve as structural frameworks. Additionally, all carbon-based materials exhibit low density and high specific strength, making them suitable for lightweight thermal management applications. Three-dimensional carbon sponges constructed pre-formed skeletal networks that serve as structural frameworks [4].

Despite these advantages, key challenges remain. High electrical conductivity can compromise insulation, while strong van der Waals interactions lead to aggregation and elevated interfacial thermal resistance. These issues limit the practical performance of carbon-based TIMs and must be addressed through appropriate surface modification and structural design strategies to optimize thermal conductivity and overall functionality.

2.2. Surface modification

For TIMs, the main goal of surface modification is to lower phonon scattering, to reduce the total thermal resistance between the filler and the matrix, and between the TIM and the interface. This is very important for making the TIM conduct heat better in the end.

The value of surface modification is that it directly deals with the main problem of high interface thermal resistance. For example, by treating boron nitride nanosheets with a silane coupling agent, we can put functional groups on their surface. This not only makes them spread out better through the steric hindrance effect, but also can form strong chemical bonds with the polymer matrix. This greatly reduces the scattering of phonons at the interface and directly makes the TIM's overall thermal conductivity higher [5].

2.3. Structural design

Structural design is about actively building continuous and efficient heat flow paths inside the composite material. Its biggest advantage is that it can greatly lower the amount of filler needed to make the heat network work (the percolation threshold), so you can get high thermal conductivity with a low filler amount. Designing at this level is also very good for solving the problem of making an efficient heat network with little filler, from a big picture. As shown in [6,7], they utilized the ice template method to construct surface-modified boron nitride nanosheets into a three-dimensional vertical network. This not only makes the fillers connect well, but its pore structure also helps keep the TIM flexible. This idea of 'change first, build framework later' is a very good example of making both thermal conductivity and mechanical properties better together.

3. The connection between methods and how to choose or use them together

From looking at the three big modification methods above, we can see they are not just side-by-side. There is actually a step-by-step order, except that when the requirements are not high, you can just modify from one of the levels. Designing a TIM for a specific situation should follow a certain logical order. I. Figure out the basic needs after knowing the application scene. II. Decide how to use the modification methods together, or choose between them, in that scene.

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1. Choosing fillers based on the scene.

All designs start by clearly defining the application scene. If you want the best thermal conductivity, you should choose carbon materials or specific ceramics that have the highest own conductivity. If you need high flexibility, then you should look at one- or two-dimensional fillers (like nanowires, nanosheets) that can form a network with a low filler amount. If cost is the main thing, then cost-effective micron-sized ceramics become the practical choice. The decisions here set the highest possible performance and the base for the whole material system.

2. Surface modification – it's needed everywhere.

After choosing the filler, surface modification is a must-do second step. No matter which filler you pick, the natural interface compatibility between the filler and the polymer matrix is a key problem that affects thermal conductivity. So, using chemical ways for surface modifications to make them spread better, stick to the matrix strongly, and effectively lower the interface thermal resistance, is almost always necessary.

3. Structural design – use it when you need it.

When the first two steps still can't meet the performance needs, then Structural design is turned on as a performance "booster". For scenes with the highest thermal limits, building a 3D thermal network is the best way. For scenes that need both good thermal conductivity and flexibility, mixing fillers of different dimensions in a controlled way is a better choice. And in very cost-sensitive uses, this step can often be skipped.

4. Conclusion

The future development of thermal interface materials (TIMs) is increasingly moving toward integrating multiple modification strategies. Rather than optimizing individual methods in isolation, research should focus on the synergistic application of filler selection, surface modification, and structural design. By coordinating these approaches across different scales and perspectives, significant improvements in the thermal conductivity and overall performance of composite TIMs can be achieved. This shift emphasizes designing composite materials with consideration of their interactions at the microscale, mesoscale, and macroscale, rather than solely enhancing single aspects.

A key direction for future research is the combination of multiple functionalities within TIMs. Advanced composites should not only provide efficient heat conduction but also integrate properties such as electrical insulation, mechanical flexibility, and electromagnetic interference shielding. Achieving multifunctionality requires careful design of both the fillers and the matrix, ensuring that these additional properties do not compromise thermal performance. The development of such multifunctional TIMs can expand their applicability across electronics, energy systems, and aerospace industries.

Another focus is precise interface engineering and long-term reliability. In-situ characterization techniques combined with computational simulations can elucidate phonon transport mechanisms at the atomic and interfacial levels, allowing targeted control of thermal resistance. Simultaneously, the durability of TIMs under thermal cycling, aging, and mechanical stress must be evaluated, alongside the adoption of low-cost and environmentally sustainable fabrication methods. Progress in these areas will facilitate the transition of TIMs from laboratory research to widespread industrial applications.

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