



Three-Dimensional Boron Nitride Networks in Polymer Composites: Fabrication Strategies and Thermal-Management Applications

Xiaoyang Wang,^{1,2,8} Yue Lv,^{1,8} Tao Yang,^{3,*} Qingyu Yang,³ Jun Zhou,¹ Kai Yang,¹ Jinnan Ma,¹ Jiushuai Zhang,⁴ Jian Yang,⁵ Tao Zhou,⁶ Shiya Wu,^{7,*} and Xiaofei Chen^{1,*}

*Correspondence: yangtao@bit.edu.cn (T.Y.); swu@bit.edu.cn (S. W.); xfchen@bit.edu.cn (X. C.)

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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- BNNSs are promising insulating fillers for thermal management, yet random dispersion in polymers hinders continuous thermal pathways and limits composite thermal conductivity.
- Strategies to build 3D BN networks via external fields, templates and geometry design, clarifying structure–property relations for phonon transport and interfacial thermal resistance.
- Applications of 3D BN networks in electronics, phase-change storage and radiative cooling, and identifies future directions including responsive materials and data-driven optimization.



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¹School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China

²China Textile Academy, 3 Yanjingli Middle Street, Beijing 100025, China

³School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China

⁴Institute of Inorganic Chemistry, RWTH Aachen University, Landoltweg 1a 52074, Germany

⁵Department of Ophthalmology, The Second Hospital of Jilin University, Jilin 130041, China

⁶School of Biotechnology and Key Laboratory of Industrial Biotechnology of Ministry of Education, Jiangnan University, Wuxi 214122, China

⁷School of Cyberspace Science and Technology, Beijing Institute of Technology, Beijing 100081, China

⁸These authors contributed equally

*Correspondence: yangtao@bit.edu.cn (T.Y.); swu@bit.edu.cn (S.W.); xfchen@bit.edu.cn (X.C.)

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Boron nitride nanosheets (BNNs) combine high intrinsic thermal conductivity, electrical insulation, and chemical stability, making them attractive electrically insulating fillers for electronic thermal management applications. However, the random dispersion of BNNs within polymer matrices severely restricts the effective construction of thermally conductive pathways, resulting in limited thermal conductivity of the resultant composites. A central challenge is to construct continuous thermally conductive networks at relatively low BN loadings while reducing interfacial thermal resistance. To address this issue, this review first introduces two mainstream solutions: multi-dimensional filler hybridization and the construction of three-dimensional (3D) boron nitride (BN) networks. This review focuses on representative strategies for engineering BN-based thermal architectures, including external-field-driven alignment, template-assisted assembly, and geometry-guided structural design, with particular emphasis on the transition from directional alignment to multidirectionally interconnected 3D networks. It elucidates the structure–property relationships governing interfacial thermal resistance modulation and phonon transport enhancement. Furthermore, this review summarizes the recent advances in applications of 3D BN networks in electronic thermal management, phase-change energy storage, and radiative cooling, while also discussing key challenges related to scalability, durability, multifunctional integration, and adaptability to complex operating conditions. Finally, future research directions are discussed, highlighting smart-responsive thermal management materials, multiphysics-coupled design, and data-driven structural optimization, aiming to provide a theoretical foundation and technical guidance for the design and engineering application of next-generation high-performance polymeric thermal management materials.

INTRODUCTION

With the rapid development of high-power electronic devices toward miniaturization, high integration, and high-power density, the local heat flux density generated by chips and power modules has increased sharply.^{1,2} This trend imposes stringent requirements on the thermal transport performance, insulation reliability, and long-term stability of thermal interface materials (TIMs), packaging heat dissipation structures, and thermal management

systems.^{3,4} Particularly in fields such as 5G communication, artificial intelligence computing clusters, power semiconductors, and power batteries, excessively high hotspot temperatures have become one of the core bottlenecks restricting device performance, lifespan, and safety.^{5,6} Therefore, developing polymer-based thermal management materials that combine high thermal conductivity, electrical insulation, good mechanical flexibility, and processability constitutes an important research direction in the current electronics and energy sectors.^{7,8}

Most polymers possess intrinsically low thermal conductivities of approximately $0.1\text{--}0.5\text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ because their disordered molecular chains impede long-range vibrational energy transport. Incorporating high-thermal-conductivity fillers is therefore a widely used strategy, although randomly dispersed fillers introduce numerous polymer-rich gaps and filler–polymer interfaces.⁹ In this review, BNNs-based thermal architectures are classified into three categories. Orientation-enhanced anisotropic structures contain BNNs preferentially aligned along one direction and mainly enhance heat transport along the alignment direction. Laminated structures contain densely stacked BNNs and usually provide efficient in-plane heat spreading but limited through-thickness connectivity. Multidirectionally interconnected three-dimensional BN networks contain continuous BN-rich pathways extending across the material volume in multiple non-coplanar directions, thereby enabling heat transfer beyond a single alignment plane.^{10,11} This classification is used throughout the following sections to clarify the structural characteristics and application scope of different fabrication methods.

Among various nanofillers, the boron nitride nanosheets (BNNs) have attracted increasing attention in the thermal management field due to their unique two-dimensional (2D) structure, high in-plane thermal conductivity, excellent wide-bandgap insulating properties, and good chemical stability. Unlike carbon-based nanomaterials such as graphene and carbon nanotubes which are highly conductive, or MXenes which are prone to oxidation, BNNs exhibit distinct advantages due to their combined characteristics of high thermal conductivity, high insulation, and corrosion resistance. Furthermore, constructing 3D BN network skeletons not only significantly enhances the thermal conductivity of phase change materials and suppresses leakage but also extends their application to radiative cooling by leveraging their high infrared emissivity. Simultaneously, their oriented alignment within polymer

matrices enables the fabrication of electrically insulating films with high thermal conductivity for next-generation electronic devices. Consequently, BNNSs-based micro-nano composite technology is promoting a transition from single-function filler reinforcement to multi-physics coupling and multi-scale structural construction, providing a promising basis for developing a new generation of efficient and reliable thermal management systems. This paper systematically reviews the recent strategies for constructing 3D BN network skeletons and the recent research progress of corresponding composites in phase change thermal storage, radiative cooling, and advanced heat dissipation devices. It discusses interfacial heat conduction mechanisms and structure-property relationships and looks forward to future opportunities and challenges in this field. As summarized in Figure 1, external-field alignment, template-assisted assembly, and geometry-guided processing regulate BN orientation and connectivity in different ways, producing anisotropic, laminated, or multidirectionally interconnected architectures for application-specific thermal management.

THERMAL CONDUCTIVITY ENHANCEMENT MECHANISMS IN POLYMERIC MATERIALS

Heat transport in BN/polymer composites depends on the intrinsic thermal conductivity of BN, the orientation and continuity of the filler architecture, and the thermal resistance at filler–filler and filler–polymer interfaces. In randomly dispersed systems, heat repeatedly crosses polymer-rich gaps and poorly contacting interfaces. Directional alignment reduces resistance mainly along the preferred orientation, whereas the perpendicular direction may remain weakly connected. Laminated structures provide efficient in-plane transport but can retain substantial through-plane resistance. Multidirectionally interconnected 3D networks reduce polymer interruptions and provide continuous heat-transfer pathways in several spatial directions. Because amorphous polymers typically exhibit thermal conductivities of only 0.1–0.5 W·m⁻¹·K⁻¹, the network architecture becomes a central design variable after BN fillers are introduced.¹²

This theory effectively explains why the vast majority of high-thermal-conductivity materials are crystalline. As shown in Figure 2A, a highly ordered lattice structure enables thermal energy to rapidly propagate throughout the crystal via collective vibrational modes, known as phonons. Conversely, most polymer materials contain inherently amorphous regions where molecular chains are randomly arranged, causing atomic thermal vibrations to become disordered and severely impeding phonon transport (Figure 2B). Consequently, their intrinsic thermal conductivity typically ranges only between 0.1 and 0.5 W·m⁻¹·K⁻¹.¹³ In summary, there are significant structural differences between good and poor thermal conductors, such as metals versus polymers, which fundamentally arise from whether they possess continuous and ordered thermal transport pathways. Figure 2 therefore provides the mechanistic basis for the subsequent discussion by linking intrinsic heat transport in polymers with filler interconnection, interfacial thermal resistance, and the loading-dependent formation of thermally conductive pathways.

Enhancement of thermal conductivity in polymer matrices via thermally conductive fillers

The incorporation of functional fillers with high thermal conductivity—several orders of magnitude higher than that of the polymer matrix—into polymer materials can enhance the thermal conductivity of the composites to varying degrees depending on the filler loading. The underlying heat conduction mechanisms differ slightly, and currently accepted explanations include the thermal conduction pathway theory, the thermal percolation theory, and the thermal elasticity coefficient theory.^{16–18}

As the mainstream thermal conduction mechanism, the thermal conduction pathway theory posits that thermally conductive fillers gradually form interconnected heat-transfer pathways through contact with the polymer matrix. Heat transfer preferentially flows along high-thermal-conductivity paths.^{19,20} As illustrated in Figure 2C, at low filler loadings, filler particles are separated by the low-thermal-conductivity polymer matrix, exhibiting isolated island-like distributions without effective interconnection; consequently, no significant improvement in the composite's thermal conductivity is observed.^{21,22} However, when the filler loading reaches a critical value, particles begin to contact each other and form a continuous network (Figure 2D),

enabling efficient heat transfer through these pathways.²³ Research by Li confirmed this trend: the thermal conductivity of BNNSs/polydimethylsiloxane (PDMS) composites increased progressively with rising BNNSs loading, reaching 1.16 W·m⁻¹·K⁻¹ at a loading of 35 wt% (Figure 2E).¹⁵

Overall, enhancing the thermal conductivity of composites essentially hinges on whether the fillers can construct continuous thermal conduction pathways within the matrix. A marked increase in thermal conductivity may occur after a sufficiently connected filler network is established, although a universal thermal-percolation threshold is not observed in all polymer composites.

However, relying solely on increasing filler loading is not the optimal strategy. The interfacial thermal resistance between thermally conductive fillers and the polymer matrix can severely negate the benefits brought by highly conductive fillers. When the filler content is low and no thermal network is formed, the large interfacial thermal resistance between the fillers and the matrix significantly reduces the overall thermal conductivity of the composite.^{24–27} While increasing filler content to form networks can substantially boost thermal conductivity, directly incorporating high filler contents may compromise other critical properties of the polymer, such as mechanical, electrical, and optical performance.^{28,29} Consequently, current mainstream research focuses on efficiently constructing thermal conduction networks under relatively low filler loadings, thereby significantly improving the thermal conductivity of composites without sacrificing the pristine properties of the polymer.

Filler loading is reported as either weight fraction or volume fraction in the literature. Because the conversion between wt% and vol% depends on the densities and complete formulation of all components, these two metrics should not be directly compared across different polymer matrices and hybrid-filler systems. In this review, the original loading basis is retained and explicitly identified. Conversion is performed only when sufficient composition and density data are available; otherwise, the values are presented separately without direct quantitative ranking.

Construction of Thermal Conduction Networks and Interfacial Synergistic Optimization

The direct incorporation of thermally conductive fillers into a polymer matrix often fails to effectively enhance the thermal conductivity of composites, while high filler loadings inevitably compromise other intrinsic properties of the matrix. The core challenge lies in the efficient construction of inter-filler thermal conduction pathways. Currently, two mainstream strategies have emerged: the hybridization of multiple fillers with differences in morphology, size, or type and the directional construction of 3D spatial structures.

Secondary-Filler Bridging and Connectivity Enhancement in BN Networks. Introducing secondary thermally conductive fillers into BN/polymer composites represents a common strategy of multi-dimensional synergy. This strategy mainly improves thermal transport by constructing multidimensional hybrid networks, reducing interfacial thermal resistance, and optimizing filler packing density. Based on the morphology of the secondary phase fillers, they can be classified into zero-, one-, and two-dimensional fillers.³⁰

Secondary fillers contribute to BN-network formation by serving as junctions or bridges between neighboring BNNSs. Zero-dimensional particles fill local gaps and create point-contact junctions, one-dimensional fillers span spatially separated BNNSs and provide line-like interlayer bridges, and two-dimensional fillers enlarge overlapping contact areas between adjacent lamellae. These effects should be distinguished from a simple increase in filler packing density: local contacts improve short-range heat transfer, whereas a genuine 3D network requires these contacts to form continuous pathways across the material volume.

1. Zero-Dimensional Fillers

Common zero-dimensional (0D) thermally conductive fillers primarily include metal particles, diamond particles, and spherical alumina. Among these, the introduction of metal particles partially compromises the insulation performance of the material, while diamond particles, though avoiding this issue, are associated with relatively high costs. Alumina, an insulating ceramic filler, combines high thermal conductivity with low cost, making it a widely adopted filler in thermally conductive polymer composites, particu-

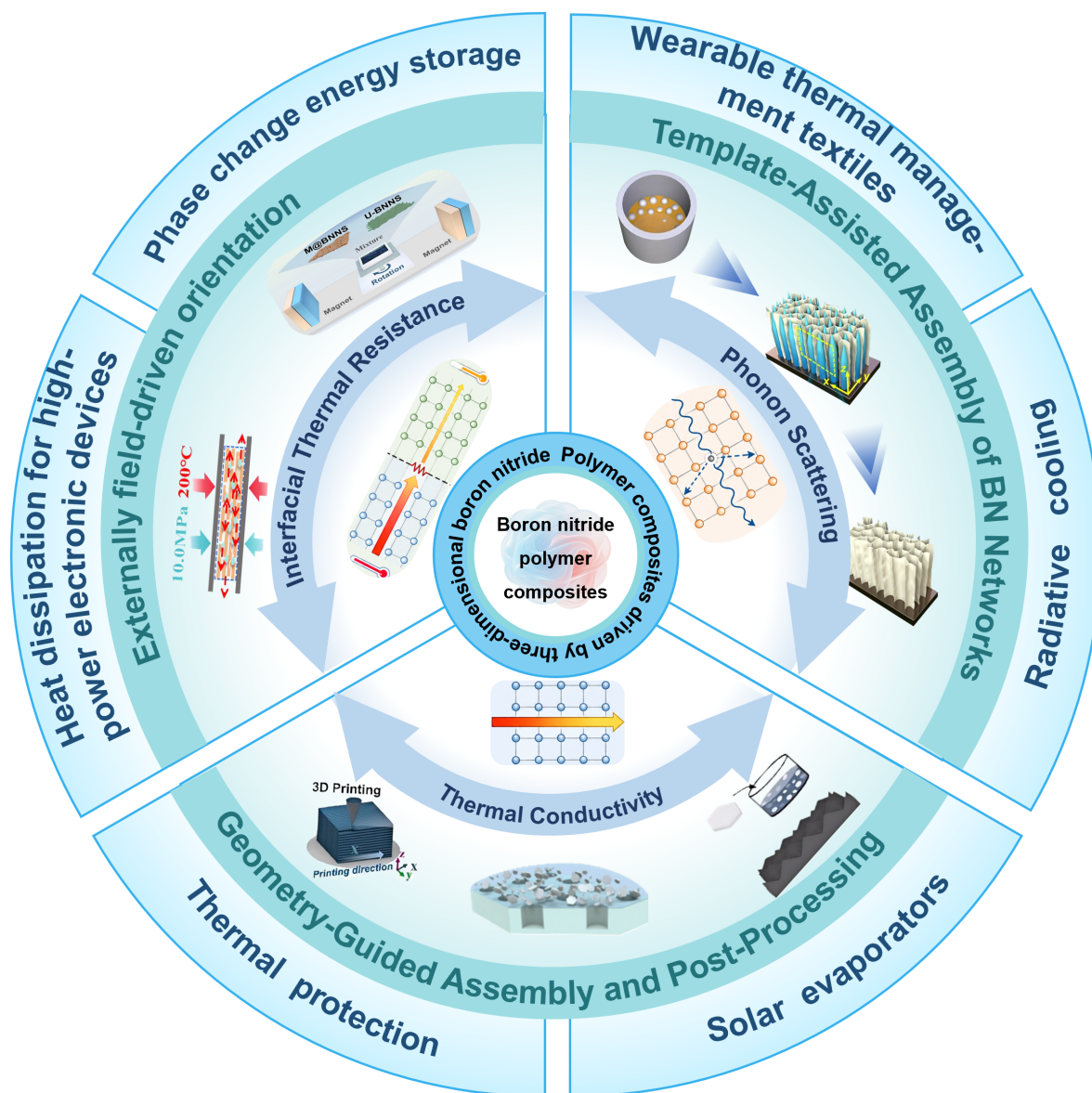


Figure 1. Schematic illustration of thermal conduction mechanisms, fabrication strategies, and target applications.

larly in its spherical form.^{31,32} Building on this foundation, Zhu et al. surface-functionalized Al_2O_3 with liquid metal (LM) and subsequently dispersed it among vertically compressed and aligned BNNs.³³ The resulting $\text{Al}_2\text{O}_3/\text{BN-LM}$ composite (80 wt% total hybrid fillers) exhibited a through-plane thermal conductivity of $7.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, an enhancement of 133% compared to the pure Al_2O_3 -based composite ($3.09 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) at the same filler loading.

Beyond these commonly used OD fillers, boron nitride nanocrystals (BNNs) have also been investigated as a class of OD thermally conductive fillers. Sharing the same lattice structure as BNNs, BNNs exhibit comparable phonon spectral features, leading to low phonon scattering at the interface. Moreover, the abundant hydroxyl and amino groups on their surfaces facilitate favorable interactions with the polymer matrix, thereby lowering the interfacial thermal resistance. With BNNs-decorated BNNs as the filler, the resulting flexible composite film delivered a through-plane thermal conductivity of $21.3 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and an in-plane thermal conductivity of $20.3 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 25 wt% loading.³⁴

In summary, OD fillers combined with BNNs—whether via heterogeneous bridging or homogeneous modification—reduce interfacial thermal resistance and enhance network connectivity, thus achieving efficient thermal conduction at low loadings. While OD fillers mitigate network discontinuity through point contacts that bridge BNNs gaps and minimize phonon scattering, trade-offs are required: metals compromise insulation, diamonds incur high costs, and conventional ceramics demand high loading. Thus, OD fillers

are best suited for flexible matrices like PDMS, particularly in flexible electronics and TIMs where low loading is critical to maintain compliance and processability. Accordingly, OD fillers primarily function as local junction nodes; they improve 3D connectivity only when these point contacts are distributed throughout an already interconnected BN framework.

2. One-Dimensional Fillers.

Compared to the point-contact limitation of OD fillers, one-dimensional (1D) fillers leverage their high aspect ratio and continuous linear structure to more readily bridge interconnected thermal conduction pathways within the matrix, thereby further enhancing the thermal conductivity and network stability of composites.

Carbon nanotubes (CNTs) represent the most prominent 1D filler, combining high thermal conductivity with lightweight properties. Han et al. anchored CNTs onto the surface of BNNs to construct hybrid fillers.³⁵ At a filler loading of 20 wt%, the resulting 3D BNNs-CNT/EP composite exhibited an in-plane thermal conductivity of $1.05 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, compared with $0.20 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the neat epoxy matrix, while the electrical insulation performance remained excellent. Based on this work, Fu et al. developed a flexible 3D porous skeleton using silanized cellulose nanofibers, and the thermal conductivity was further boosted upon the addition of BNNs and CNTs.³⁶

It should be noted, however, that the high electrical conductivity of CNTs may compromise the insulation performance of composites if introduced in excess. Boron nitride nanotubes (BNNs) could serve as an electrically insu-

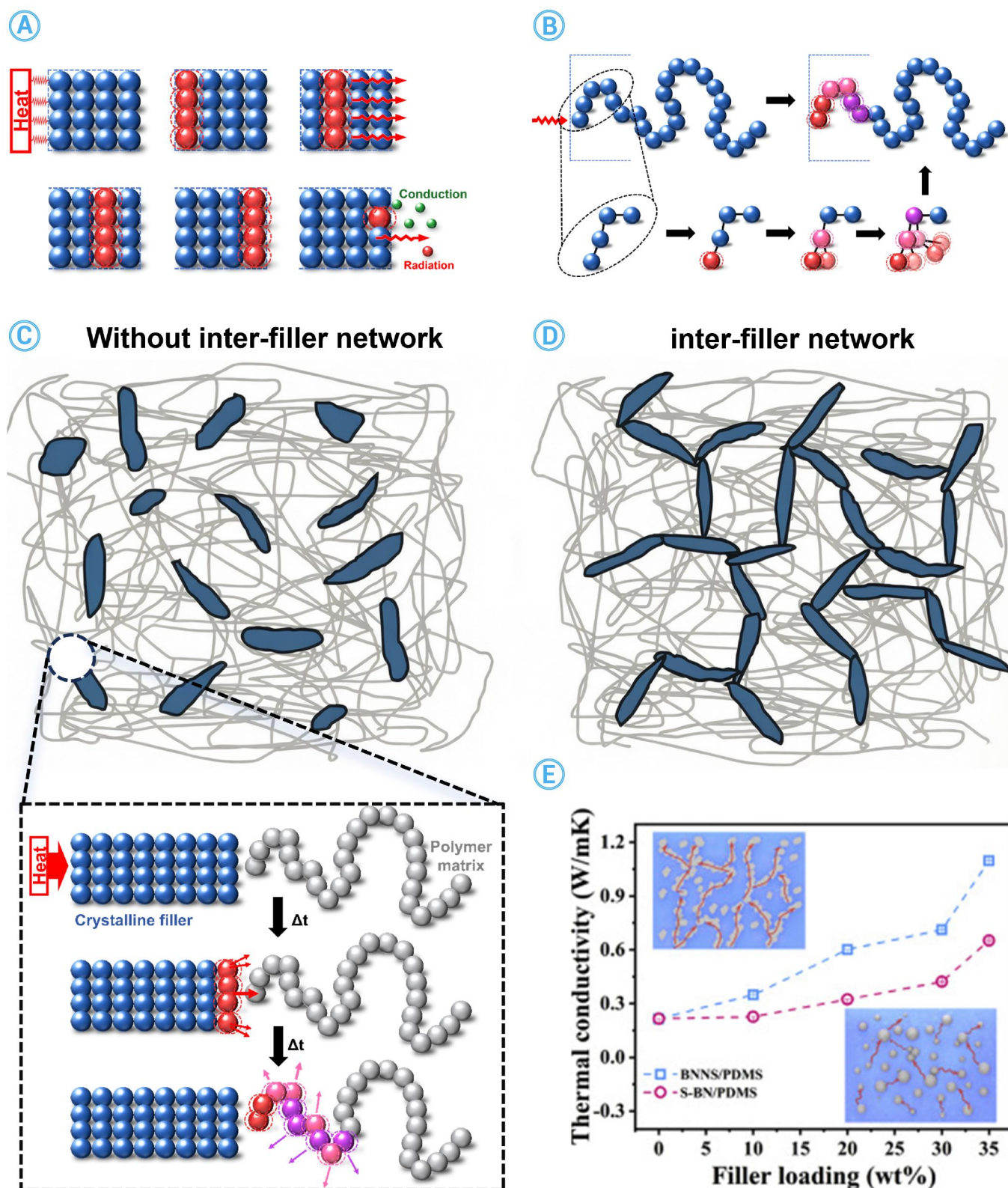


Figure 2. (A) Thermal conduction mechanism in crystalline materials; (B) Thermal conduction mechanism in amorphous polymers; Reproduced with permission from Ref. 12. Copyright 2016, Elsevier. (C) At low filler loading, no thermal conduction pathway is formed. Phonon conduction transfers between the fillers and the polymer matrix with high interfacial impedance, resulting in severe interfacial heat dissipation; (D) At higher filler loading, a thermal pathway is formed, improving the phonon transport efficiency; Reproduced with permission from Ref. 14. Copyright 2018, Elsevier. (E) The thermal conductivity of BN/PDMS composites increases with the increase in BN loading. Reproduced with permission from Ref. 15. Copyright 2016, Elsevier.

lating alternative.^{37,38} Mohanraman et al. reported that the introduction of just 1 wt% BNNTs into a composite containing 30 wt% BNNSs created 3D ther-

mal conduction pathways bridging the BNNSs layers, thereby elevating the thermal conductivity to $3.4 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ without compromising electrical insulating performance.³⁹ Nevertheless, the practical utility of BNNTs remains severely constrained by their high production costs.

In summary, 1D fillers leverage linear bridging to construct continuous 3D thermal networks at low percolation thresholds, thereby boosting phonon transport while avoiding the matrix degradation caused by high filler loadings. They address the network discontinuity and high contact resistance inherent in 0D systems through line-to-line bridging between BNNSs layers. However, trade-offs remain: excessive CNTs loading may compromise electrical insulation, whereas BNNTs are constrained by high production costs. Consequently, 1D fillers are well-suited for lightweight, high-performance matrices such as epoxy and phase-change materials in high-power electronic packaging and thermal management applications that demand a balanced combination of thermal conductivity, mechanical robustness, and electrical insulation. Owing to their high aspect ratios, 1D fillers are particularly effective as cross-plane or interlayer bridges and can convert isolated BNNSs domains into longer-range connected pathways.

3. Two-Dimensional Fillers.

Compared to 1D fillers, two-dimensional (2D) fillers replace line-to-line contacts with face-to-face contact, significantly enlarging the phonon transport cross-section. Graphene, BNNSs, and hexagonal boron nitride (h-BN) are currently the most common 2D thermally conductive fillers. Additionally, MXene,⁴⁰ as a novel 2D material, has garnered increasing attention from researchers due to its excellent in-plane thermal conductivity, with values reaching as high as $55.8 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for certain MXene compositions. Jiao et al. utilized electrostatic self-assembly combined with an ice-templating method to construct BN/MXene/silicone rubber (BN/MXene/SR) composites with a vertically oriented structure.⁴¹ At a filler loading of 20.75 wt%, the BN/MXene/SR composite achieved a thermal conductivity of $0.83 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, representing a 20.2% improvement over the BN-only composite ($0.69 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$).

Two-dimensional secondary fillers enlarge the overlapping contact area between adjacent BNNSs and can reduce contact resistance within lamellar networks. Their contribution to genuine 3D connectivity depends on whether the overlapping sheets also establish cross-plane junctions. Graphene and MXene may improve heat transfer and multifunctionality, although electrical conductivity, oxidation stability, and interfacial compatibility must be considered.

Secondary fillers of different morphologies can form point, line, and face contacts with BNNSs, with the contact area increasing sequentially.^{42–44} These additional contact sites reduce the reliance on direct contact between BN layers, enabling composites to cross the percolation threshold at relatively low filler loadings and achieve a nonlinear surge in thermal conductivity. It should be noted that 1D and 2D fillers exhibit anisotropy, and their thermal transport performance differs significantly between random dispersion and oriented alignment, which will be discussed in detail in subsequent sections.^{45–48}

4. Other Advanced Hybrid Fillers.

In addition, several novel hybrid filler configurations have been reported in recent studies,⁴⁹ such as core-shell architectures, interpenetrating networks, and sandwich-type structures.

Core-shell fillers generally consist of a highly thermally conductive core and a functional shell. This architecture can break the intrinsic in-plane orientation of BNNSs and modulate thermal conductivity anisotropy. BN is commonly employed as the shell material. In the work of Jiong et al., silane-coupling-agent-modified alumina was used as the core and hydroxylated BN as the shell.⁵⁰ At a total filler loading of 50 wt%, the 20 m-BN@80 m- Al_2O_3 film achieved a through-plane thermal conductivity of $2.3 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, corresponding to a 192% improvement over the m-BN film ($0.8 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) at the same filler loading, while the anisotropy index was reduced by 54%. In other studies, BNNSs can aggregate into thermally conductive cores through specific treatments. For instance, Li et al. fabricated core-shell heterostructured fillers with BN as the core and reduced graphene oxide (rGO) as the shell, achieving a composite thermal conductivity as high as $5.94 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.⁵¹

Interpenetrating structures introduce long-range bridging units into the conventional BN network. Unlike the 1D and 2D secondary fillers discussed

above, the bridging units required for interpenetrating structures are nanoribbons with extremely high aspect ratios, which establish cross-scale continuous thermally conductive networks through face-to-face contacts. Huang et al. synthesized boron nitride nanoribbons (BNNRs) with widths of 3–7 μm and lengths of 50–600 μm via chemical vapor deposition (CVD) and interpenetrated them into a BNNSs/melamine sponge (MS) skeleton.⁵² At a BN loading of 45 wt%, the composite exhibited an in-plane thermal conductivity of $3.72 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, compared with $0.17 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the neat matrix.

Sandwich structures are fabricated by laminating different filler layers, with each individual region containing only one type of filler. This configuration maximizes the utilization of the distinct thermal conduction characteristics of different fillers. In the study by Zhao et al., a covalent adaptive network (CAN) derived from vanillin was used as the matrix.⁵³ The outer layers consisted of BN/CAN to provide electrical insulation and thermal conduction, while the middle layer comprised CNTs/CAN for electromagnetic interference (EMI) shielding. Through high-temperature exchange reactions of imine bonds, the layers were welded into an integrated structure. The resulting material simultaneously exhibited a high thermal conductivity of $1.79 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, EMI shielding effectiveness exceeding 55 dB in the X-band, excellent electrical insulation on the order of $10^{-13} \text{ S}\cdot\text{m}^{-1}$, and a high modulus approaching 3800 MPa.

Compared to conventional hybridization strategies that rely solely on filler geometry, the main advantage of these novel connection modes lies in replacing simple physical stacking with interfacial chemical bonding or topological interlocking. Such strong coupling at the microscopic level not only reduces interfacial thermal resistance but also effectively overcomes the functional limitations of single-type fillers, enabling the integrated and synergistic enhancement of thermal conductivity, electromagnetic shielding, and mechanical performance.

Directional and Multidirectional Construction of BN Architectures.

Engineering directional and multidirectional BN architectures represents another effective route to improving thermal transport in polymer composites. Directional alignment shortens and regularizes heat-flow pathways along the preferred orientation but can leave transverse connectivity limited. In contrast, multidirectionally interconnected 3D networks extend BN-rich pathways across multiple non-coplanar directions and reduce polymer-rich interruptions. Essentially, directionally aligned BN networks exhibit faster phonon transport, lower interfacial thermal resistance, and higher thermal conductivity compared to randomly dispersed BN networks. Concurrently, the anisotropy of 2D BNNSs imparts anisotropic thermal conductivity to the aligned structure, making it suitable for applications requiring directional heat dissipation. Relevant research findings will be elaborated in detail below.

CONSTRUCTION STRATEGIES FOR BN THERMAL ARCHITECTURES

As previously discussed, engineering BNNS-based thermal architectures is an effective approach for improving the thermal conductivity of polymer composites. This chapter provides an overview of mainstream strategies for constructing directionally aligned, laminated, and multidirectionally interconnected BN architectures, as well as the latest advances involving secondary fillers to bridge these networks.

Externally field-driven orientation

As a typical 2D material, BNNSs possess atomic-level thickness and significant anisotropy. Under external force fields (shear, extrusion, or electric fields), BNNSs are prone to in-plane slippage or spatial rearrangement, enabling effective control over their crystallographic orientation.

Hot pressing is a commonly employed technique that induces preferential alignment of 2D BNNSs through a thermo-mechanical coupling field, offering advantages including operational simplicity, scalability, and high practicality.⁵⁹ Through hot-pressing-induced in-plane orientation of BNNSs, the composite exhibited an in-plane thermal conductivity of $3.85 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 40 wt% loading, which was 30 times higher than that of the neat epoxy resin ($0.127 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$).⁶⁰ The underlying principle of hot pressing relies on the synergistic effect of thermodynamics and rheology. However, the substantial viscosity of composites at high filler loadings imposes severe flow constraints, which significantly limit the construction of oriented structures by this method. Building upon this approach, the multilayer stacking hot-pressing technique employs a cyclic shear-stack-re-shear process: the

material is first hot-pressed into thin sheets, each serving as a single layer, and after multiple layers are prepared, they are stacked and subjected to a secondary hot-pressing step. Ji et al. utilized this technique to construct a highly vertically oriented hybrid network of BNNs and carbon fibers in a silicone rubber matrix, achieving a thermal conductivity of $9.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (Figure 3A).⁵⁴

In the fabrication of fibrous composites, conventional wet spinning also serves as an effective alignment technique. Yang et al. exploited the strong confinement effect and shear forces generated by sheath contraction to achieve a high degree of axial alignment of hydroxylated BNNs within the sheath precursor, yielding a core-sheath phase-change fiber with an axial thermal conductivity of $10.15 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (Figure 3B).⁵⁵

Magnetic field-assisted alignment technology enables the manipulation of magnetically responsive fillers in suspensions via an external magnetic field while simultaneously allowing composite fabrication. Since BNNs are inherently non-magnetic, they require modification with magnetic nanoparticles. Upon such modification, BNNs achieve a thermal conductivity of $12.1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a loading of 62.6 vol%.⁶¹ Nevertheless, this technique typically enhances thermal conductivity only along a single direction, and the through-plane thermal conductivity often falls below that of composites with randomly dispersed fillers.⁶² This limitation arises because the 2D fillers form continuous thermal pathways exclusively along the magnetic field direction, while insufficient interconnection in the perpendicular direction compromises overall thermal efficiency. Jiang et al. partially addressed this issue by incorporating non-magnetic hydrogen-bond-functionalized BN (U-BNNS) into magnetically modified BN fillers (M-BNNS).⁵⁶ At a total filler fraction of 35 wt%, the composite exhibited an in-plane thermal conductivity of $11.47 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, comparable to that of the single-oriented M-BNNS/WPU composite ($10.77 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$), while the through-plane thermal conductivity was enhanced by 194% to $2.88 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, far exceeding that of M-BNNS/WPU ($0.98 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) (Figure 3C). External strong electric fields can produce effects analogous to those of magnetic fields, with electrospinning being a representative example. Miao combined coaxial electrospinning with hot pressing to achieve an in-plane thermal conductivity of $6.99 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a filler loading of 10 wt%, compared with $0.47 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the neat PAN film, while the out-of-plane thermal conductivity reached $0.387 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (Figure 3D).⁵⁷

Beyond these established techniques, any controllable external force field capable of interacting with BNNs can, in principle, be exploited to construct oriented structures. For instance, Chen et al. employed a rotational shear force field that, at a shear rate of 8 rpm, induced the formation of shish-kebab structures in the polymer matrix and promoted the parallel alignment of BN (002) crystal planes along the shear direction (Figure 3E).⁵⁸ At a BN loading of 35 vol%, the in-plane thermal conductivity of the composite processed under rotational shear reached $1.26 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, representing a 3.3-fold increase over that of the composite processed without rotational shear, which exhibited an in-plane thermal conductivity of only $0.39 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

In summary, the prevailing strategy for constructing oriented filler structures relies on the application of external force fields—such as shear, magnetic, and electric fields—to drive directional alignment. Whether achieved via hot pressing, spinning, or electromagnetic induction, these approaches disrupt the random dispersion of fillers and enforce preferential heat-flow pathways. Nevertheless, such methods are often limited by unidirectional orientation characteristics or processing constraints at high filler loadings, rendering it challenging to achieve comprehensive 3D thermal percolation.

Template-assisted assembly of BN networks

The templating strategy employs various forms of templates to assemble thermally conductive fillers into 3D interconnected architectures with continuous heat-transfer networks. The ordered stacking of functional components within the composite can be achieved by controlling diverse material and processing parameters. Based on the types of templates utilized during assembly, templating methods can be classified into self-templating, sacrificial templating, foam templating, and ice templating, etc.⁶³

The ice-templating method is a directional assembly technique based on solid-liquid phase transition and physical confinement effects. Its underlying

principle relies on the repulsion exerted by growing ice crystals, which forces BNNs to undergo directional stacking at the freezing front. Subsequent freeze-drying removes the ice template, yielding a porous scaffold with an anisotropic structure. Using this technique to fabricate a 3D BN network, the composite achieved a through-plane thermal conductivity of $2.64 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a filler content of 21.88 vol%, substantially higher than the $0.21 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ of neat epoxy.⁶⁸ However, similar to magnetic-field-assisted alignment, conventional ice templating employs a single temperature gradient, resulting in unidirectional BNNs orientation. To address this limitation, Han et al. modified the traditional ice-templating approach by compressing the 3D scaffold after its construction (Figure 4A).⁶⁴ This compression caused the BN network to collapse into interlayer zigzag continuous phonon bridges, thereby optimizing the through-plane thermal conductivity. At a BNN content of 80 wt%, the film achieved an in-plane thermal conductivity of $26.16 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, representing a 48.9% improvement over that of the randomly gelled film, which exhibited an in-plane thermal conductivity of $17.57 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. Meanwhile, the through-plane thermal conductivity of the film reached $5.43 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, compared with $0.90 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the randomly gelled film. Alternatively, modulating the temperature gradient can achieve a similar effect. Zhao et al. employed a PDMS wedge to simultaneously establish horizontal and vertical temperature gradients in a BNNs/PU suspension, inducing the formation of a dual-oriented 3D network (Figure 4B).⁶⁵ At a BNNs loading of 80 vol%, this approach yielded high thermal conductivity in both directions, with in-plane and through-plane values of 39.0 and $11.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

The salt-templating method is an advanced synthetic technique based on a bottom-up strategy, utilizing inorganic metal salt crystals as structure-directing substrates to achieve controlled orientation and morphological confinement of 2D nanomaterials. Cui et al. employed sodium chloride as a removable sacrificial template to fabricate a BN-carbon nanofiber-polysulfone 3D thermally conductive scaffold (Figure 4C).⁶⁶ At a filler loading of 28.61 wt%, the epoxy composite with the as-prepared scaffold showed an isotropic thermal conductivity of $2.18 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. After carbonization at 1500°C , the thermal conductivity reached $2.85 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, versus $0.20 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for neat epoxy. Solvent-exchange-induced self-assembly is a dynamic self-assembly technique governed by solution thermodynamic regulation. This method achieves oriented assembly and rearrangement of 2D BNNs by precisely modulating the solubility parameters of the solvent environment, thereby disrupting the colloidal stability of the original dispersion system through entropy-driven or interfacial energy minimization principles. Chen et al. reconstructed hydrogen-bonding interfaces in an aqueous environment to composite hydroxylated BNNs with room-temperature self-healing glassy polyurethane (GPU), forming a biomimetic inverse nacre-like structure (Figure 4D).⁶⁷ At a filler loading of 40 wt%, this composite achieved a high in-plane thermal conductivity of $11.54 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

A key advantage of templating is its ability to regulate the spatial arrangement of BNNs through predefined confinement. Ice, salt, foam, and solvent-mediated templates can produce anisotropic or interconnected architectures with controllable pore structures. These methods provide a promising strategy for reducing the filler content required to form continuous heat-transfer pathways, although their practical performance depends strongly on scaffold integrity, porosity, and matrix infiltration.

Despite their structural advantages, template-assisted methods introduce several practical limitations. Residual porosity and incomplete polymer infiltration can interrupt filler contacts and increase the effective thermal resistance. Highly porous BN scaffolds may also be fragile during transfer, impregnation, compression, or long-term loading. Sacrificial-template removal, solvent exchange, and freeze-drying increase processing time, solvent consumption, and energy demand, while drying shrinkage can distort the designed architecture. In addition, small variations in freezing rate, template size, suspension stability, and infiltration conditions may cause batch-to-batch differences in pore morphology and thermal performance. Future studies should therefore report porosity, matrix-infiltration degree, shrinkage ratio, processing yield, mechanical integrity, and thermal-conductivity retention together with the initial thermal conductivity.

Geometry-guided assembly and post-processing

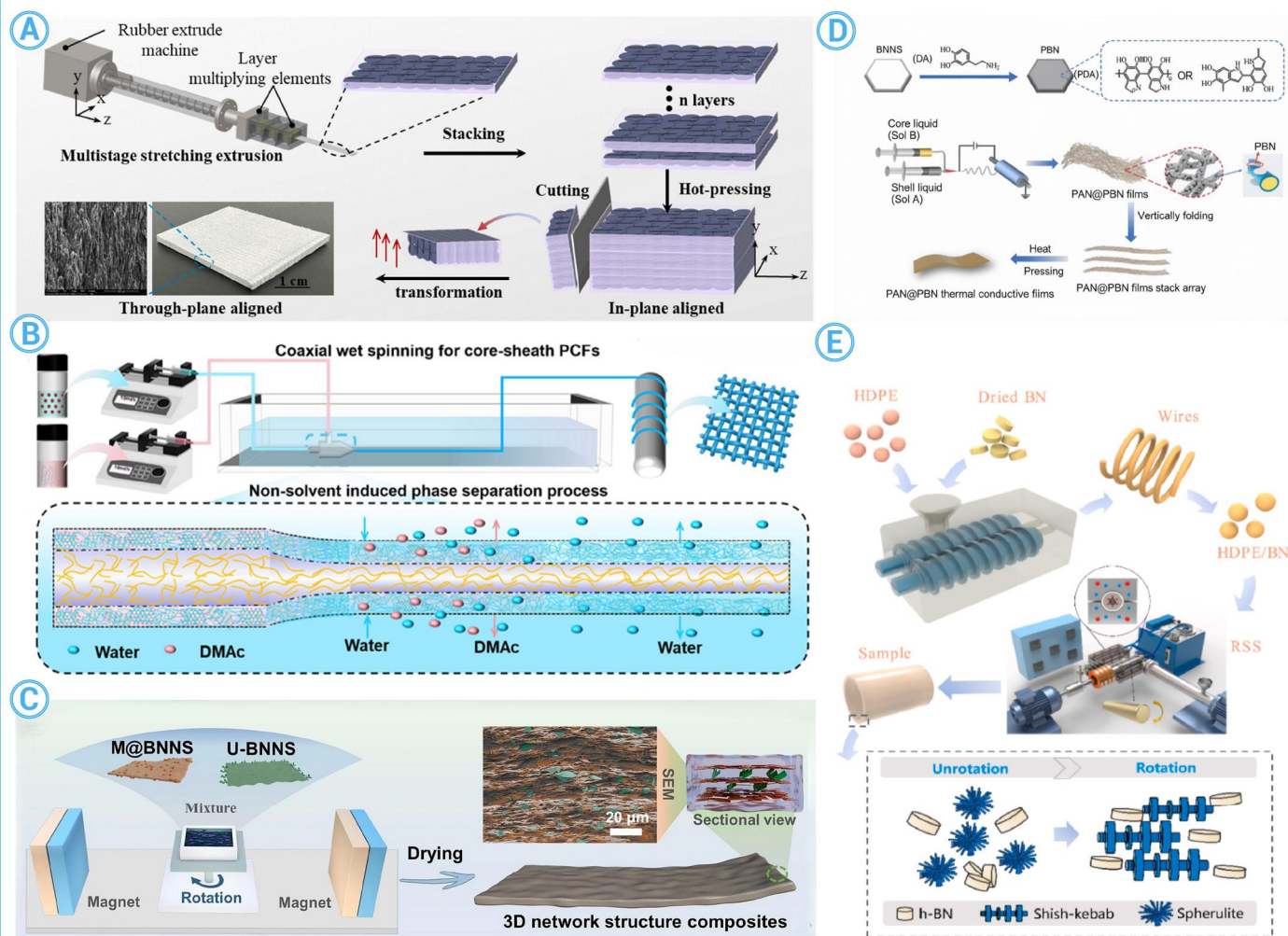


Figure 3. (A) Schematic illustration of the fabrication of silicone rubber composites with highly vertically aligned hexagonal BNNs/CF networks via multi-stage stretching extrusion, layer-by-layer stacking, and hot-pressing processes. Reproduced with permission from Ref. 54. Copyright 2023, Elsevier. (B) Schematic illustration of the core-sheath phase-change fibers containing axially aligned hydroxylated BNNs with high thermal conductivity, superior heat conduction performance, and ultrafast heat transfer via a coaxial wet-spinning process based on non-solvent induced phase separation (NIPS). Reproduced with permission from Ref. 55. Copyright 2025, Wiley. (C) Schematic diagram of the preparation of composites with 3D-bridge architecture based on magnetic field orientation. Reproduced with permission from Ref. 56. Copyright 2025, Springer. (D) Schematic illustration of the fabrication of dopamine-functionalized BNNs/polyacrylonitrile (PAN) core-shell nanofiber membranes via electrospinning combined with a hot-pressing process. Reproduced with permission from Ref. 57. Copyright 2025, Elsevier. (E) Schematic illustration of the fabrication of high density polyethylene (HDPE)/BN composites using a rotational shear system. Reproduced with permission from Ref. 58. Copyright 2023, Elsevier.

Beyond the conventional approaches of external force-field modulation and templating strategies, recent studies have proposed novel methodologies for constructing oriented networks, which are briefly outlined below.

The anisotropic drying shrinkage of gel-based matrices can provide an external driving force for BNNs orientation. Miao et al. exploited this effect to achieve dense packing of BNNs in sodium alginate (SA), attaining an in-plane thermal conductivity of $45.50 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a filler loading of 40 wt% (Figure 5A).⁶⁹

The high-aspect-ratio sheet-like morphology of BNNs renders them highly responsive to external physical fields. When BNNs are dispersed as individual sheets in a liquid polymer, the flow field response within confined spaces drives the sheets to rotate and align along specific directions. Specifically, Kim and co-workers designed two distinct microarray geometries—cylindrical⁷⁰ and tetrahedral⁷¹—in which the combined effects of flow-induced shear, inlet contraction, wall impingement, and the geometric confinement of the array templates collectively promoted the directional alignment of BNNs (Figure 5B & C).

Commercially available highly in-plane-oriented boron nitride films (BNFs) offer the advantages of high orientation and densely stacked lamellae. Taking advantage of this material, Niu et al. employed a mechanical cutting

approach to convert the in-plane orientation of BNFs into a through-plane orientation, achieving a through-plane thermal conductivity of $19.1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a loading of 37 vol% (Figure 5D).⁷²

These studies signify a broader transition in the field of TIMs, moving from a reliance on high filler loading toward a structure-oriented design philosophy. Researchers have expanded beyond simple filler-content optimization and increasingly employ physical-field induction, template-assisted assembly, and microstructural engineering to control BN orientation and connectivity within polymer matrices. This approach markedly curtails filler consumption while circumventing the inherent constraints of unidirectional heat conduction, thereby enabling the controlled synthesis of materials exhibiting high in-plane, through-plane, or isotropically enhanced thermal conductivity. These advancements collectively furnish both a theoretical underpinning and a basis for the development of next-generation lightweight and customizable thermal management materials tailored for high-performance, low-energy electronics.

THERMAL-MANAGEMENT APPLICATIONS OF BN-BASED THERMAL ARCHITECTURES

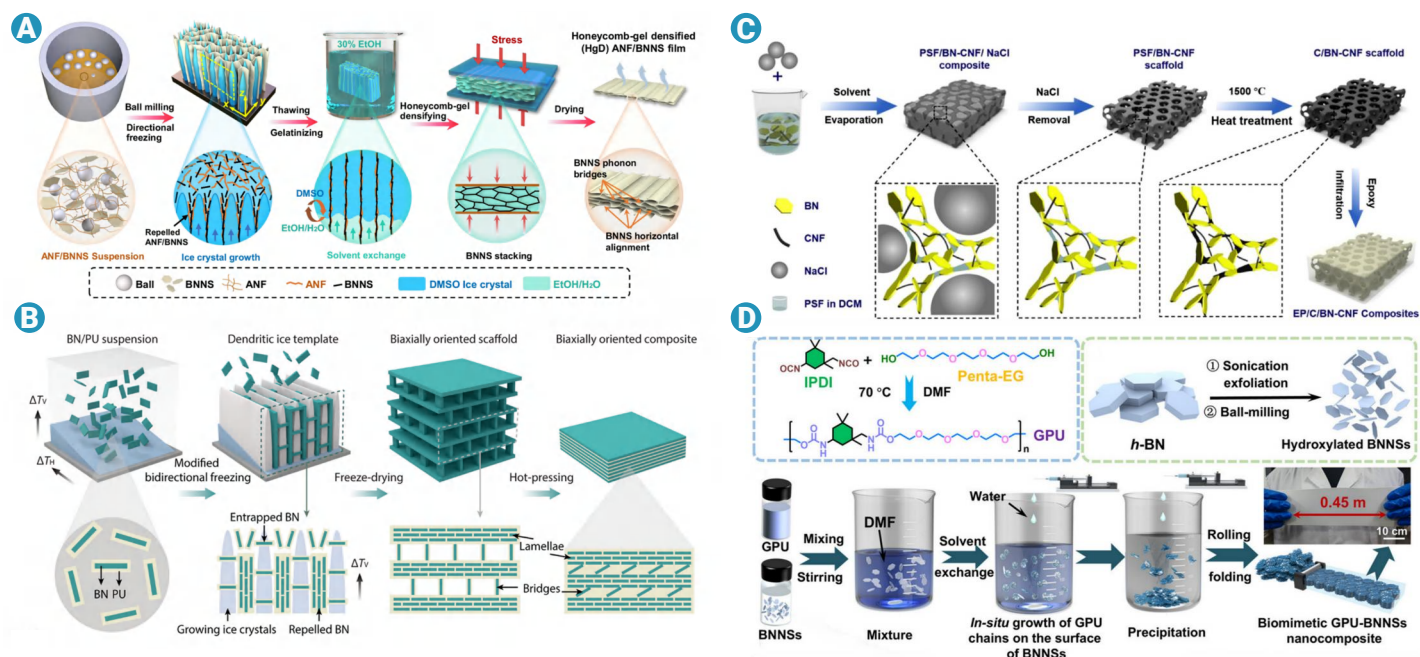


Figure 4. (A) Schematic illustration of the formation of aramid nanofibers (ANF)/BNNs films via a unidirectional freezing and thaw-gelation process. Reproduced with permission from Ref. 64. Copyright 2025, Springer Nature. (B) Schematic illustration of the fabrication of BN/polyurethane (PU) composites with a biaxially oriented network using a modified bidirectional freezing method. Reproduced with permission from Ref. 65. Copyright 2022, American Chemical Society. (C) Schematic illustration of the fabrication of epoxy resin (EP)/C/BNNs/cellulose nanofibrils (CNFs) composites via a salt-templating method. Reproduced with permission from Ref. 66. Copyright 2023, Elsevier. (D) Schematic illustration of the fabrication of GPU-BNNs nanocomposites via a solvent exchange-induced self-assembly strategy. Reproduced with permission from Ref. 67. Copyright 2025, Wiley.

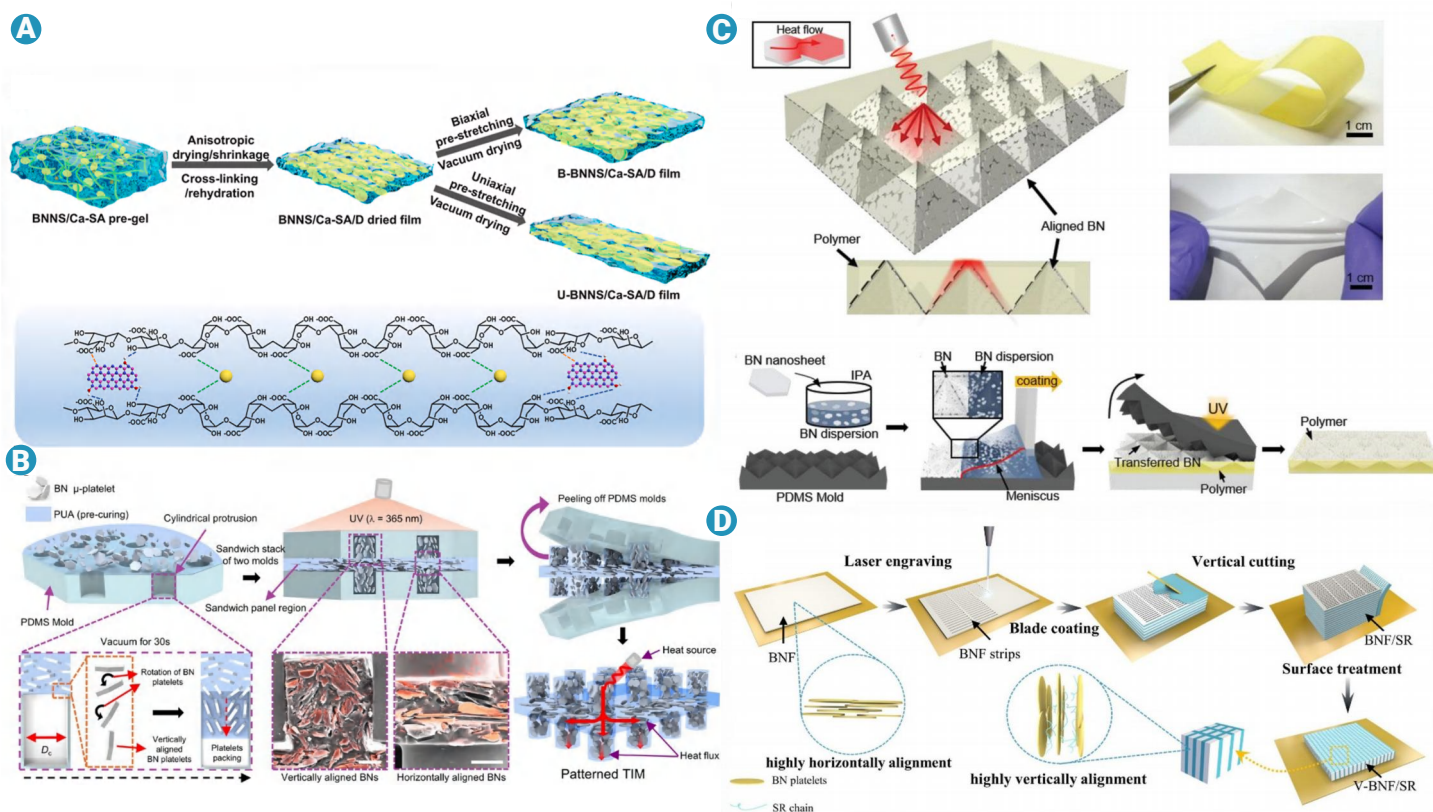


Figure 5. (A) Schematic illustration of the fabrication of a multilayer crosslinked oriented BNNs/Ca-SA/D film via stretch induction. Reproduced with permission from Ref. 69. Copyright 2026, Elsevier. (B) Assembly of BN flakes via micro-molding technology to further construct bi-directional thermally conductive composites. Reproduced with permission from Ref. 70. Copyright 2025, Wiley. (C) Assembly of continuous arrays of BNNs on a tetrahedral structure to construct 3D thermal pathways and further fabricate anisotropic thermally conductive composites. Reproduced with permission from Ref. 71. Copyright 2019, Wiley. (D) Schematic illustration of the fabrication of V-BNF/SR composites with a vertically aligned structure via a stacking-cutting method. Reproduced with permission from Ref. 72. Copyright 2024, Elsevier.

The construction of 3D BN networks not only enhances heat transport efficiency but also endows composites with designable anisotropy, structural stability, and interfacial synergy. Consequently, their applications have gradu-

ally expanded from traditional heat dissipation to diverse scenarios such as thermal energy storage, thermal protection, radiative cooling, and wearable thermal management.

Phase change energy storage

To address the bottlenecks of phase change materials (PCMs), including intrinsically low thermal conductivity, leakage tendency, and poor mechanical properties, constructing 3D thermally conductive skeletons and optimizing filler interfacial coupling have become core strategies for achieving efficient thermal management and structural-functional integration.⁷⁹

In the field of low-grade waste heat recovery, PCMs often face challenges such as low thermal conductivity, leakage, and insufficient mechanical strength.^{80,81} To this end, Tu et al. fabricated a flexible methyl vinyl silicone rubber (MVQ)/boron nitride (BN)/expanded graphite (EG) skeleton via a salt-templating method, followed by the incorporation of polyethylene glycol (PEG) to obtain a shape-stabilized phase-change composite.⁸² The resulting composite exhibited a thermal conductivity of $1.10 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and retained a compressive resilience of 92.26% after 100 compression cycles, demonstrating effective thermal transport together with mechanical recoverability. Building on this work, Fan et al. further enhanced the thermal conductivity of PCMs by employing a covalent hybridization strategy to construct BNNs/CNTs hybrid fillers.⁷³ At a filler loading of 15 wt%, a thermal conductivity of $5.92 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ was realized, successfully reducing the operating temperature of electronic devices by $10\text{--}30^\circ\text{C}$ (Figure 6A). These studies collectively provide high-performance solutions for passive thermal management in electronic equipment and energy storage systems.

Heat dissipation for high-power electronic devices

As the power density of electronic devices increases, the conflict between heat dissipation and electromagnetic compatibility (EMC) has become increasingly prominent.^{83,84} To address this challenge, researchers have constructed multi-dimensional filler networks, utilizing thermal-electrical performance decoupling and functional integration strategies to significantly enhance device operational reliability.⁸⁵

Li et al. constructed a continuous BN thermal conduction network within an SBS matrix.⁸⁶ At a BN loading of 40 wt%, the composite achieved a high thermal conductivity of $3.855 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. Under 2.45 GHz irradiation, the device temperature decreased by 44°C , effectively addressing the thermal accumulation issue in high-power systems and providing a scalable manufacturing solution for passive thermal management in electronic devices.⁸⁷ This strategy is equally applicable to precision optoelectronic devices. Lee et al. applied this approach to μLED implantable devices, utilizing BN layers as heat dissipation rails to suppress local overheating (Figure 6B).⁷⁴ This successfully inhibited light output attenuation and wavelength redshift, significantly enhancing illumination intensity and tissue penetration depth, thereby providing an approach for thermal management and precision therapy in high-power precision optoelectronic devices.

Thermal protection

In the field of power battery safety, thermal runaway propagation remains a critical hazard.^{88–90} Constructing multifunctional protection systems that integrate efficient heat dissipation with structural and electrochemical stability has become a key pathway to enhancing battery safety.

Zhang et al. constructed a synergistic thermally conductive network using ultra-high-molecular-weight polyethylene (UHMWPE) fiber fabrics and BN, enabling the composite to achieve high through-plane and in-plane thermal conductivities of 10.05 and $7.92 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively, while the puncture strength was significantly enhanced to 47.13 N (Figure 6C).⁷⁵ In ternary battery encapsulation applications, this strategy not only reduced the cycling temperature by over 10°C but also provided effective impact protection. Lithium metal batteries face another prominent thermal safety issue—the continuous growth of lithium dendrites may pierce the separator and cause short circuits.^{91,92} To address this, Wang et al. developed a trifunctional separator based on BN and poly(vinylidene fluoride)-hexafluoropropylene.⁹³ With an ultra-high in-plane thermal conductivity of $117.76 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ enabling uniform heat dissipation, the onset temperature of thermal runaway in pouch cells was elevated from 119.3°C to 196.7°C , while stable cycling of lithium symmetric cells was maintained for 1800 h, providing a critical separator design solution for high-safety lithium metal batteries. For lithium–sulfur batteries, Zhu et al. designed a super-aligned carbon nanotubes (SA-C)/super-aligned boron nitride @ carbon nanotubes (SA-BC) composite separator that integrates rapid heat dissipation, polysulfide anchoring, and dendrite suppression.⁹⁴ This separator enabled the batteries to maintain

excellent capacity retention after 300 cycles at a high temperature of 60°C and a high rate of 2C, with a per-cycle decay rate of only 0.026%, offering a novel design strategy for high-safety lithium–sulfur batteries.

Solar evaporators

Interfacial solar evaporation technology positions the photothermal conversion layer at the water–air interface and employs a localized heating strategy to substantially reduce heat capacity and thermal losses, thereby significantly enhancing evaporation efficiency. This technology imposes dual requirements on materials: on one hand, the materials must possess broadband strong light absorption and efficient photothermal conversion capability to ensure full capture of solar energy; on the other hand, they must achieve excellent thermal localization through the directional design of thermal conduction pathways, confining heat to the evaporation interface and preventing its dissipation into the bulk water, while cooperating with hydrophilic channels to guarantee continuous water supply.^{95,96} To meet these stringent demands, the construction of anisotropic structures that integrate both efficient thermally conductive networks and interfacial water regulation functions has become a key strategy for breaking through the bottlenecks of evaporation rate and long-term stability. Sun et al. utilized the shear force generated during 3D printing extrusion to induce the directional alignment of BNNs, combined with the broadband light absorption of Ti_3O_5 and the hydrophilic chitosan matrix, to simultaneously achieve dual anisotropy at both the material and geometric levels (Figure 6D).⁷⁶ The continuous phonon pathways constructed by BN, with an in-plane thermal conductivity of $2.73 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, enable rapid heat distribution across the evaporation interface, effectively preventing localized overheating.

Radiative cooling

Passive radiative cooling utilizes the atmospheric transparency window ($8\text{--}13 \mu\text{m}$) to dissipate heat to outer space without external electrical input during operation, providing a promising route for passive thermal management. However, the all-weather adaptability of this technology faces severe challenges. To address this issue, constructing a radiation-evaporation synergistic (RRE) mechanism is key to overcoming environmental dependency and achieving highly efficient passive cooling without external electrical input.^{97–100} Ran et al. used a polyvinyl alcohol hydrogel as the substrate, doping it with BNNs to increase solar reflectivity and mid-infrared emissivity to 0.9 and 0.93, respectively, while loading lithium bromide to optimize water management (Figure 6E).⁷⁷ This film achieved a sub-ambient cooling of 9.37°C under sunny conditions and maintained a cooling effect of 3.87°C even on cloudy days, successfully overcoming the weather-dependent limitations of single-mechanism technologies. When applied to photovoltaic modules, it reduced the surface temperature by 22.1°C and increased the output power by $20.3 \text{ W}\cdot\text{m}^{-2}$, providing a promising passive-cooling strategy for building energy efficiency and electronic device thermal management.

Wearable thermal management textiles

With the advancement of wearable electronics and the increasing frequency of extreme weather events, personal thermal-moisture comfort management has attracted increasing interest in smart textiles.¹⁰¹ Conventional fabrics are inherently limited in their ability to simultaneously achieve efficient thermal regulation and dynamic sweat transport, which commonly results in the concomitant sensation of overheating and stuffiness. Therefore, developing novel smart textiles that integrate both passive thermal management and moisture control functions is important.

Zhang et al. developed a Janus double-layer cooling textile (WCT): using biomass rush as a template to construct a BN thermal conduction network, combined with a wettability gradient to achieve rapid unidirectional sweat transport.⁷⁸ Compared to cotton fabrics, WCT reduced temperature by 1.5°C and 4.1°C under dry and wet conditions, respectively, while maintaining stable performance after 200 bending and washing cycles (Figure 6F). Liu et al. further proposed an adaptive thermoregulatory fabric (ATR), in which the outer layer (POM) achieves 94.2% solar reflectance and 96.2% infrared emissivity, while the inner layer—a BN/phase-change microsphere composite—provides a latent heat buffer of $149.4 \text{ J}\cdot\text{g}^{-1}$.¹⁰² Featuring asymmetric wettability for sweat transport within 3 s, this fabric demonstrated a measured daytime cooling of 6.6°C and nighttime warming of 1.7°C , effec-

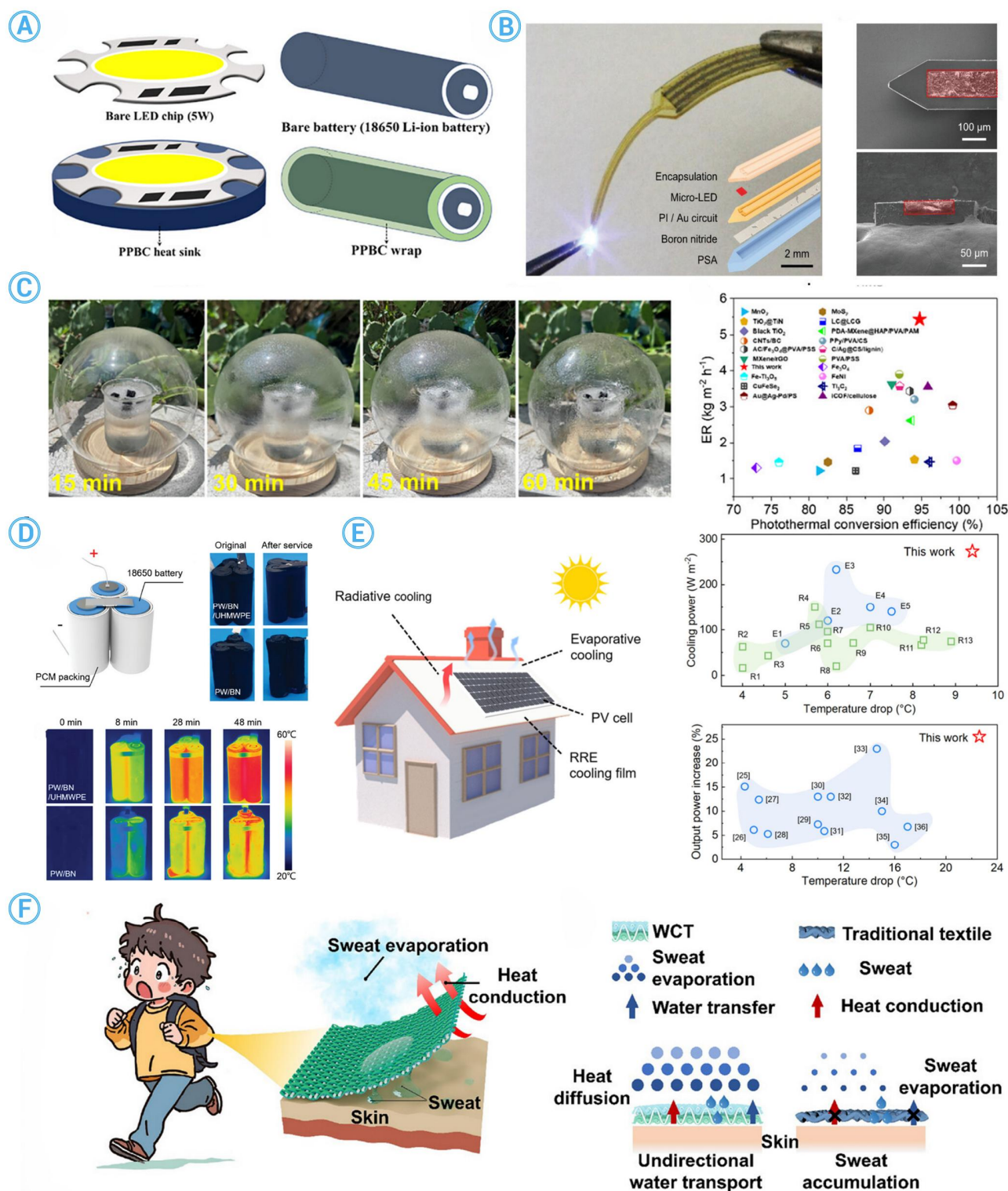


Figure 6. (A) Thermal Management Performance of Phase Change Composites with Covalently Interconnected BN/Carbon Nanotube Hybrid Networks in Commercial 18650 Batteries. Reproduced with permission from Ref. 73. Copyright 2026, Wiley. (B) Assembled BNNSSs-Based Heat Dissipation Directors for Neural Probes. Reproduced with permission from Ref. 74. Copyright 2023, Wiley. (C) Thermal Management and Mechanical Protection of Paraffin/BN Phase Change Composite Lithium-Ion Battery Systems Treated with a Polyethylene "Inner Layer". Reproduced with permission from Ref. 75. Copyright 2024, Wiley. (D) 3D Printing-Assisted Orientation of BN for High-Performance Solar Evaporators. Reproduced with permission from Ref. 76. Copyright 2026, Wiley. (E) Schematic Diagram of the Cooling Principle of Regenerable Radiative-Evaporative Cooling Films. Reproduced with permission from Ref. 77. Copyright 2026, Elsevier. (F) Wearable Textiles for Personal Thermal Management. Reproduced with permission from Ref. 78. Copyright 2026, Wiley.

tively resolving the overcooling issue associated with radiative cooling textiles at night.

RELIABILITY AND DATA-DRIVEN DESIGN

Long-term reliability under service conditions

High initial thermal conductivity does not necessarily guarantee stable

Table 1. Representative BN-based thermal architectures discussed in previous sections, along with their processing conditions, directional thermal conductivities, and target applications.

BN-based structures	Polymer matrix	Filler loading	Fabrication method	In-plane (k)	Through-plane (k)	Target application	Key feature / limitation	Architecture type	Ref.
$\text{Al}_2\text{O}_3/\text{BN-LM}$	PDMS	80 wt%	Mechanical grinding and hot pressing	–	7.2	TIMs	Vertical alignment, LM reduces ITR / High loading & cost	Orientation-enhanced anisotropic	33
BNNs/BNNs	SA/PVA	25 wt%	Ice-pressure-assisted assembly method	20.3	21.3	TIMs	BNNs self-modification, ice templating / Complex process	Multidirectionally interconnected 3D	34
BNNs/CNT	BPA epoxy resin	20 wt%	Curdan foaming and freeze-drying	1.05	–	TIMs	Curdan foaming, freeze-drying / Low TC	Multidirectionally interconnected 3D	35
BNNs/BNNTs	BPA epoxy resin	31 wt%	Vacuum-assisted SpeedMixer	2.6–3.4	–	TIMs	1D–2D synergy / Through-plane data missing	Multidirectionally interconnected 3D	39
BN/MXene	SR	20.75 wt%	Electrostatic self-assembly, ice-templating method, and hot pressing	–	0.83	TIMs, EMC	EMI absorption, ice templating / Low TC	Orientation-enhanced anisotropic	41
m-BN/m- Al_2O_3	PVA	50 wt%	Electrostatic self-assembly, solvent casting, and hot pressing	9.7	2.3	TIMs	Mechanical reinforcement / High anisotropy	Laminated	50
BNNs/BN NRs/MS	HDCN	45 wt%	Electrostatic L-b-L assembly, freeze-drying, and hot pressing	3.72	1.28	TIMs	Recyclable / Low through-plane TC	Multidirectionally interconnected 3D	52
BNNs/CF	SR	56.8 wt%	Multi-stage stretching extrusion (MSE), layer-by-layer stacking, and hot-pressing	–	9.5	TIMs	MSE extrusion / High loading	Orientation-enhanced anisotropic	54
BNNs-OH	CNF/PW @DMAC/LiCl	50 wt%	conventional wet spinning	10.15 (axial)	–	Phase-change fiber (PCF)	Phase-change, TC / Through-plane data missing	Orientation-enhanced anisotropic	55
M-BNNs/U-BNNs	WPU	35 wt%	Magnetic field-assisted alignment technology	11.47	2.88	TIMs	Magnetic alignment, flame retardant / Moderate TC	Multidirectionally interconnected 3D	56
PDA-BNNs	PAN	10 wt%	conventional wet spinning, Multilayer hot pressing	6.99	0.387	TIMs, flexible printed circuit boards (FPCBs)	Low dielectric constant / Very low through-plane TC	Laminated	57
BNNs	HDPE	35 vol%	Rotational shear system (RSS)	1.26	–	TIMs	Shish-kebab, high VST / Low absolute TC	Orientation-enhanced anisotropic	58
BNNs	ANF	80 wt%	ice-templating method, Compression densification	26.16	5.43	TIMs	Interlayer phonon bridges / High loading	Laminated	64
BNNs	PU	80 vol%	Bidirectional ice-templating method and hot pressing	39.0	11.5	TIMs	Bidirectional ice templating / Extremely high loading	Multidirectionally interconnected 3D	65
BN/CNF	Epoxy resin (E-51 grade)	28.61 wt%	salt-templating method	–	2.85	TIMs	Salt template, carbonization / High-temp treatment	Multidirectionally interconnected 3D	66
BNNs-OH	GPU	40 wt%	Solvent-exchange-induced self-assembly	11.54	–	Smart thermal interface materials (TIMs)	Self-healing, moisture-tolerant / Through-plane data missing	Laminated	67
BNNs	SA	40 wt%	Multiple crosslinking and stretch-induced orientation	45.50	–	TIMs, heat dissipation for wearable devices, and human body thermal management. TIMs for 3D integrated electronic devices	Highest in-plane TC / Extremely anisotropic	Laminated	69
BNNs μ -platelets	PUA	40 wt%	Micromolding and bidirectional orientation	7.4	6.9		Quasi-isotropic, flexible / Moderate TC	Multidirectionally interconnected 3D	70
BNNs	UV-curable acrylate polymer	16 wt%	Doctor blading, transfer printing, and planarization	11.05	1.15	TIMs	Stretchable, flexible / Low through-plane TC	Orientation-enhanced anisotropic	71
BNFs	SR	37 vol%	Stacking-and-cutting method	–	19.1	TIMs	Highest through-plane TC / Time-consuming process	Orientation-enhanced anisotropic	72

Note: Filler loadings are reported on the original wt% or vol% basis used in the cited publication. Values expressed on different bases should not be directly compared unless complete composition and density data are available.

*Thermal conductivity (TC), k ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$).

performance during long-term service. Thermal cycling, humidity, sustained compression, bending, washing, and repeated phase transitions can induce interfacial debonding, network cracking, pore evolution, and loss of filler–filler contact. These structural changes may gradually increase both bulk thermal resistance and contact thermal resistance. The dominant reliability requirement depends on the target application. Phase-change composites must resist leakage, volume changes, and repeated melting–solidification cycles; thermal interface materials must retain low contact resistance under compression and thermal cycling; wearable materials require bending, abrasion, and washing durability; and battery components must preserve structural integrity, thermal transport, and electrical insulation under elevated temperatures and electrochemical exposure.^{73,75,78,82,93,94}

Future studies should therefore report thermal-conductivity retention after standardized aging, thermal cycling, mechanical deformation, and humidity exposure. Contact resistance, dimensional stability, leakage ratio, mechanical-property retention, and electrical-insulation stability should be evaluated together with the initial thermal conductivity. Such reliability data is essential for distinguishing laboratory-scale conductivity enhancement from practically durable thermal-management performance.

Computational and data-driven design

Computational and data-driven methods can complement experimental design at several levels. Effective-medium and percolation models can estimate the effects of filler fraction and connectivity, while finite-element and molecular simulations can evaluate local heat-flow pathways, filler orientation, interfacial resistance, and network topology.^{17,28} Recent studies have already applied machine-learning models to predict the thermal conductivity of polymer composites using descriptors related to filler properties, polymer matrices, composition, and microstructural characteristics.^{103,104} Data-driven frameworks combining high-throughput numerical simulations, machine learning, optimization algorithms, and experiments have also been developed to determine filler–polymer and filler–filler interfacial thermal resistances in particulate-filled polymer composites.¹⁰⁵ More recently, Bayesian optimization has been employed to optimize filler formulations in thermally conductive polymer composites, demonstrating the potential of data-driven methods to accelerate composition screening and experimental design.¹⁰⁶ These approaches provide a route to screen filler dimensions, loading, interface treatment, and processing parameters before experimental fabrication and to identify nonlinear relationships among composition, architecture, and directional thermal conductivity.⁹⁰

The reliability of these approaches currently remains limited by small datasets and inconsistent reporting. Many published studies omit the heat-flow direction, specimen thickness, porosity, orientation factor, polymer baseline, or complete filler formulation. A useful database should therefore include matrix chemistry, BN type and dimensions, secondary fillers, wt% or vol%, porosity, orientation, interface treatment, processing route, in-plane and through-plane thermal conductivity, mechanical properties, and reliability data. Standardized descriptors and uncertainty reporting would enable more credible prediction and inverse design of BN-network architectures.

CONCLUSION

This review systematically summarizes the fabrication strategies and application progress of polymer composites based on 3D BN networks. It first identifies the intrinsically low thermal conductivity of conventional polymers as a critical bottleneck restricting their thermal management performance. The current mainstream solution is the incorporation of highly thermally conductive fillers; however, low filler loadings fail to establish effective thermal pathways, while high filler loadings significantly degrade processability and mechanical properties. To address this dilemma, two main strategies have emerged: one is the hybridization of multi-dimensional fillers (0D/1D/2D) to construct additional synergistic thermally conductive networks through point, line, and plane contacts, thereby reducing interfacial thermal resistance; the other is the construction of 3D BN networks, which enable the controlled fabrication of continuous thermal conduction channels at low filler loadings, significantly reduce polymer-rich interruptions and improve heat-transfer continuity, while potentially limiting the filler content required for a target thermal performance.

For application-oriented material selection, through-plane thermally conductive networks are preferred when heat must be transferred from a localized source to an external heat sink, as in thermal interface materials, chip packaging, and thermally conductive battery separators. In-plane networks are more suitable for lateral heat spreading in heat spreaders, wearable textiles, radiative-cooling films, and solar evaporators. Multidirectionally interconnected 3D networks are advantageous for phase-change composites, battery packs, and devices containing distributed or spatially varying heat sources. The optimal architecture should therefore be selected according to the heat-flow boundary condition, required mechanical compliance, and service environment rather than by maximizing a single thermal-conductivity value. This review summarizes three representative routes for engineering BN-based thermal architectures—external-field-driven alignment, template-assisted assembly, and geometry-guided structural design—and discusses how these strategies generate directionally aligned, laminated, or multidirectionally interconnected networks for different thermal-management scenarios. Finally, it points out that the complexity of fabrication processes and the difficulty of scaling up remain core bottlenecks, while the long-term interfacial stability between fillers and the polymer matrix requires further verification. Future efforts should be directed toward the development of low-cost, high-throughput fabrication technologies, the development of standardized, direction-resolved datasets and validated computational models to support data-driven screening and inverse design of BN-network architectures, and the strengthening of research on service behavior under multi-physics coupling conditions, thereby promoting the further engineering application of 3D BN network composites in new energy, 5G communication, aerospace, and other fields.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

No primary research results, software or code have been included, and no new data were generated or analyzed as part of this review.