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Mini Review

Engineered Nanocomposites for Critical Thermal Management Needs – Opportunities and Challenges

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Abstract

Hexagonal boron nitride (h-BN) and graphite are isoelectronic, so are their nanosheets, and they are both highly thermally conductive. However, h-BN derived nanosheets (BNNs) are electronically insulating, thus ideally suited for polymeric nanocomposites of high thermal conductivity but no electrical conductivity for a variety of critical thermal management needs. Highlighted here are advances in the development of polymer/BNNs composites and devices, excellent opportunities for further improvements, and also technical and other challenges, especially with the growing applications of 3D printing techniques.

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In the era of rapid advancement in transformative technologies, including especially artificial intelligence (AI), big data, advanced semiconductor chips, and the growing popularity of electric vehicles (EVs), there are enormous demands for and consumptions of energies, which are accompanied by the heat generation as a troubling or even in some cases prohibitive byproduct. Thus, in equally high demand are enabling thermal management (TM) technologies for the effective heat transport and dissipation, which are dependent on materials of high thermal conductivity (TC), including metals such as aluminum and copper and some carbon-based materials. For example, various combinations of aluminum and graphite are used for cooling the batteries in many EVs. However, graphite and even aluminum could become fuels at high temperatures, and their high electrical conductivity (EC) might disqualify their uses as thermal interface materials (TIMs) in chips and other devices. In this regard, isoelectronic with graphite and also of a similar layered structure is hexagonal boron nitride (h-BN), nicknamed white graphite, whose decoupled thermal and electric transport properties (high TC but no EC) are uniquely suited for TM applications. Indeed, h-BN nanosheets (BNNs, Figure 1) and their polymeric composites are being actively investigated, with some major advances already achieved [1-25].

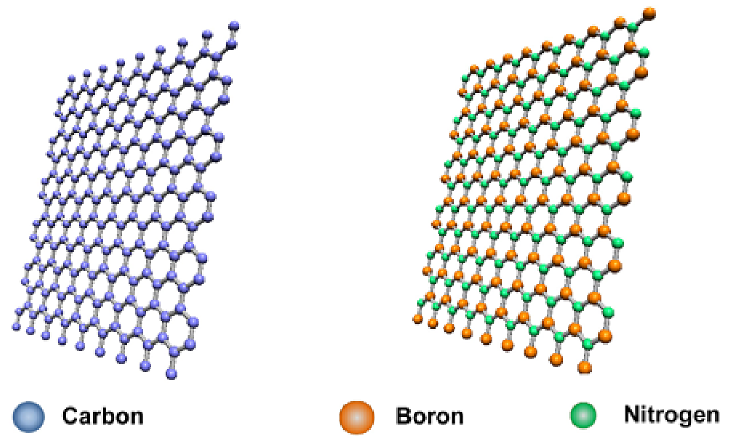

Figure 1: Single-layer graphene (left) versus hexagonal boron nitride (h-BN) nanosheet (right). Their comparison in nanosheets of multiple layers is similar, except for the stronger inter-layer interactions in h-BN.

Few-layer BNNs are mostly produced from the exfoliation of h-BN, for which the vigorous sonication of h-BN in isopropanol (IPA) as a preferred solvent has been popular [24-26]. Further improvements in the exfoliation efficiency and in the quality of the resulting BNNs, characterized by their larger aspect ratios and smooth surfaces, could be achieved by adding aqueous ammonia to IPA for the sonication [25, 27]. However, the inter-layer force is significantly stronger in h-BN than in graphite, making the exfoliation of h-BN more difficult and the resulting BNNs not as well defined nanosheets as their carbon counterparts. Beyond the commonly employed vigorous sonication, other processing tools and/or conditions might be explored for the right level of shear forces to produce BNNs of better qualities and in higher yields.

BNNs as planar nanofillers are anisotropic in thermal transport, with much higher in-plane TC than cross-plane TC. Therefore, more popular have been their uses in polymeric nanocomposite films designed for high in-plane TCs. Wet casting has been a convenient method for the fabrication of polymer/BNNs composite films, in which the filler BNNs are driven to align in the film plane, thus the observed high in-plane TC values [20, 28, 29]. For example, the dispersion of BNNs into polyethylene (PE) for PE/BNNs composite films resulted in monotonically increasing in-plane TCs with higher BNNs loadings (Figure 2) [29]. The crosslinking in the composite films could apparently enhance the TC performance significantly (Figure 2) [29]. A more effective strategy for further in-plane alignment of BNNs in composite films, such as those of poly (vinyl alcohol) (PVA) as the matrix polymer, is to apply the needed shear force with mechanical stretching of the films, resulting in substantially higher in-plane TCs (Figure 3) [20, 30]. As might be expected, the enlarged in-plane TCs associated with the more effective in-plane alignment of BNNs in the composite films are at the expenses of cross-plane TCs in the same films, generally with the cross-plane/in-plane TC ratios lowered to less than 10%, which could serve as an indicator of how well the filler BNNs in composite films are in-plane aligned. Among other shear force induced alignment methods for polymer/BNNs composite films of enhanced in-plane TCs is mechanical extrusion of the films [14, 31].

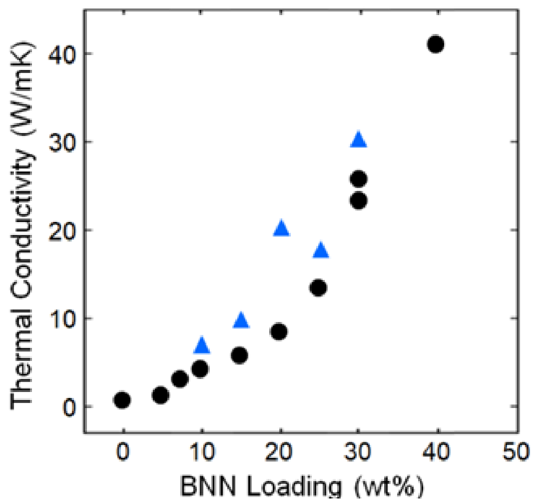

Figure 2: TC values of the PE/BNNs (circle), and crosslinked PE/BNNs (triangle) composite films at different loadings of BNNs. (From reference 29).

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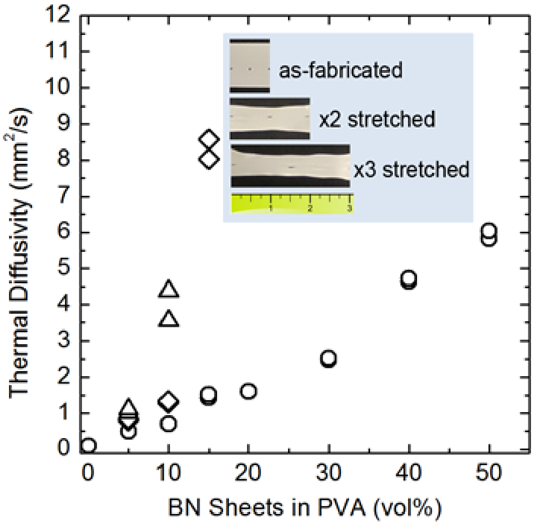

Figure 3: Observed thermal diffusivity values at different loadings of BNNs in PVA/BNNs nanocomposite films as fabricated (circle) and mechanically stretched (×2: diamond; and ×3: triangle). Shown in the inset are photos of as-fabricated and stretched films with 10% loading of BNNs. (From reference 20).

3D printing has emerged to represent an effective and versatile fabrication method for nanocomposites and related devices. Naturally it has been employed for the preparation of polymer/BNNs composite films for in-plane thermal transport purposes. Some of the more popular printing methods are limited to or more suitable for composite films of relatively low loadings of BNNs, offering advantages over simple wet casting in terms of more versatile and controllable fabrications, though in terms of in-plane TC performances the potential for major improvements seems limited. There are also other printing methods capable of preparing films with high loadings of BNNs, somewhat analogous to a more controlled extrusion method. Nevertheless, 3D printing should in principle be majorly advantageous for the fabrication of composite films and structures in which the filler BNNs are cross-plane aligned for high cross-plane thermal transport [32-35], because such composite configurations are pretty much beyond the capability of conventional wet casting. Illustrated in Figure 4 is a representative 3D printing fabrication of films (3.2 mm in thickness) in which the filler BNNs are cross-plane aligned in the embedded rod structures, with the observed TCs increasing linearly with loadings of BNNs (Figure 4) [32]. However, the TC of only 1.5 W/mK at >20 wt/% loading of BNNs leaves a lot of room for improvements.

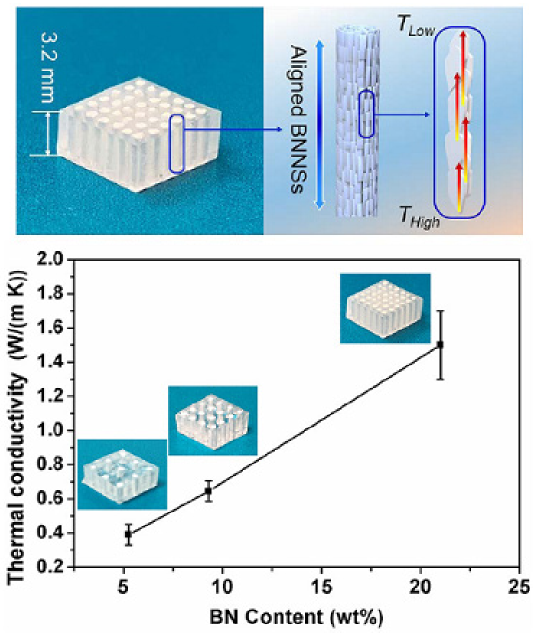

Figure 4: Upper: Thick film of polydimethylsiloxane (PDMS) embedded with the through-film-plane aligned BN rods. Lower: The observed dependence of the through-film-plane thermal conductivity on the BN loading. (From reference 32).

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In fact, the need for major performance improvements represents a general challenge for 3D printing of polymer/BNNs composites designed for TM applications. For such a challenge, specifically developed and implemented printing strategies and protocols are in demand for addressing the critical issues associated with the challenge. The aimed improvements include:

(i) To make the printed structures more dense via reducing voids and porosity and enhancing the interfacial bonding between the polymer matrix and filler BNNs. For example, the voids and structural discontinuities could be minimized by improving the filler dispersion, optimizing the viscosity of feedstocks, adjusting various printing parameters (layer height, hatching space, printing speed, temperature, etc.), and applying suitable post-processing techniques such as annealing or infiltration to densify the printed parts.

(ii) To push for the uniform dispersion of BNNs within the polymer matrix during the feedstock preparation and the subsequent printing process. Strategies for the uniform dispersion include the use of BNNs that are surface functionalized with molecules fully compatible with the matrix polymer, the selection of suitable solvents or dispersing agents, and the energetic shear mixing.

(iii) To reduce and minimize the anisotropy and discontinuities arising from the layer-by-layer deposition in the printing.

(iv) To reach high filler loading, for which the associated high viscosity in the feedstock might impede the homogeneous flow and deposition during printing, thereby reducing processability and uniformity. Several mitigation strategies could be applied by balancing the filler content with rheological properties (leveraging non-Newtonian shear-thinning flow behavior in the fabrication process), using hybrid filler systems, adjusting printing temperature, and adding dispersing agents or rheology modifiers to maintain processability while preserving the desired mechanical or functional properties.

(v) To enable the fabrication of the composites in special form factors, especially ultra-thin films in which BNNs are cross-film-plane aligned for thermal transport needs in some high-end electronic devices, as most commercially available systems are not designed for such fine structures that require high precision. Nevertheless, there are promising developments in the relevant research field that are applicable to the printing of the ultra-thin films, including strategies for the precise control over deposition methods, the vat photopolymerization process with two-photon excitation, micro-extrusion, inkjet printing, and electrohydrodynamic printing.

In summary and conclusion, polymer/BNNs composites in various form factors engineered for high in-plane or cross-plane TCs but electrically insulating have shown great promises. Further rapid and broad developments of these materials and their derived devices for much improved performances, driven by the critical thermal management needs in some of the most exciting current and emerging technologies, may be envisaged.

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Conflict of Interest

No Conflict of Interest.

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