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Uniaxially Stretched Polyethylene/Boron Nitride Nanocomposite Films with Metal-like Thermal Conductivity

Ri-Chao Zhang^{1,2*}, Zirui Huang¹, Zhihuan Huang¹, Meiling Zhong¹, Dongmian Zang¹, Ai Lu³, Yuanfei Lin⁴, Bronagh Millar², Graham Garett², Gary Menary², Dehui Ji¹, Lixian Song⁵, Quanchao Zhang¹, and Jiali Zhang¹, Dan Sun^{2*}

¹*School of Materials Science and Engineering, East China Jiaotong University, Nanchang 330013, China;*

²*School of Mechanical and Aerospace Engineering, Queen's University Belfast, Belfast BT9 5AH, UK;*

³*Institute of Chemical Materials, China Academy of Engineering Physics, Mianyang 621900, Sichuan, China;*

⁴*South China Advanced Institute for Soft Matter Science and Technology, South China University of Technology, Guangzhou 510640, China*

⁵*College of Materials Science and Engineering, Southwest University of Science and Technology, Mianyang 621002, Sichuan, People's Republic of China*

* Correspondence: d.sun@qub.ac.uk; zrcws@zju.edu.cn;

Abstract: Achieving ultra-high thermal conductivity while maintaining the electrical insulation of polymers is highly desirable for many applications such as thermal

management and packaging. In this work, polyethylene / boron nitride nanoplatelets (PE/BNNP) nanocomposite film was produced through melt processing followed by uniaxial stretching. Microstructural analysis reveals that the stretched composite film features a co-continuous network structure which consists of oriented lamellae bridged by both stretched polymer chains and aligned BNNPs along the stretching direction. The resulting film exhibit a metal-like thermal conductivity as high as $106 \text{ W m}^{-1} \text{ K}^{-1}$ and it is believed that the unique network structure has enabled efficient phonon transfer across the film, resulting in superior thermal transporting performance. This work shines a light on the design and scalable manufacturing of high performance functional polymer-based composites for future thermal management applications.

Keywords: polyethylene; boron nitride nanoplatelets; nanocomposites; thermal conductivity

1. Introduction

The rapid development in electronic device integration and miniaturization requires high performance thermally conductive materials for effective thermal management, as this is essential for prolonged device life and reliability [1-3]. For certain industries such as the aerospace sector, lightweight, durable and thermally conductive materials with electrical insulation are highly desirable [4]. Polymers are widely deployed in modern technologies ranging from soft robotics [5], organic electronics [6], to wearable computing devices [7] and artificial skin [8], owing to their unique properties such as low cost, lightweight, durability, flexibility, corrosion resistance and easy/scalable processing [9]. Unfortunately, their low thermal conductivity ($\kappa = 0.1 \text{ W m}^{-1} \text{ K}^{-1}$) usually prevents them from many advanced functional applications [10]. Technically, thermally conductive polymers can be

manufactured by incorporating high loadings (e.g., 50 w.t.%) of thermally conductive fillers [11-13], such as boron nitride (BN) [14-16], into the polymer matrix to form a composite. Unfortunately, the high filler content can significantly increase the polymer rheology, creating difficulty in the volume production of such composites into final products through common processing techniques such as melt-processing and extrusion. In addition, very high filler content usually leads to particle agglomeration, which can damage the integrity of the material and hence compromise the mechanical properties (such as ductility) of the resulting composites. Despite the recent research in this area, the thermal conductivity of such composites is still far from ideal. For polymers and polymer based composites, achieving a thermal conductivity approaching that of the metallic material still represents a significant challenge.

Polyethylene (PE) is one of the semi-crystalline thermoplastic polymers which finds wide applications in the industry [17]. Single chain PE can be treated as a quasi-one-dimensional (1-D) crystal with very high thermal conductivity [18] along the chain direction, based on the non-ergodic characteristics of 1-D conductors [19]. Recent atomic simulation results [20] show that the thermal conductivity of a single PE chain in the axial direction can be as high as $1400 \text{ W m}^{-1} \text{ K}^{-1}$ and that the thermal conductivity of a single crystal lamella can reach $237 \text{ W m}^{-1} \text{ K}^{-1}$. Unfortunately, the thermally conductive single crystal lamellae within bulk PE are isolated by randomly distributed entangled chains (amorphous phase) and the overall thermal conductivity of the bulk polymer is greatly compromised ($0.2\sim 0.5 \text{ W m}^{-1} \text{ K}^{-1}$) [21] due to the phonon scattering at the interfaces between lamella and the amorphous phase. Despite such limitation, the thermal

characteristics of PE can be modified by manipulating their microstructures and/or crystallinity through externally applied forces [22-29]. For instance, PE films with improved thermal conductivity ($65 \text{ W m}^{-1} \text{ K}^{-1}$) have been obtained under a uni-axial stretching ratio of 200 [26]. PE nanofibers with considerably high thermal conductivity ($\sim 104 \text{ W m}^{-1} \text{ K}^{-1}$) have been fabricated through a two-stage hot-drawn method [23]. Despite the promising results, the process-structure-property relationship of the above mentioned material systems as well as the associated heat transfer mechanisms within such material systems remain poorly understood. In addition, the extremely thin films or nanofibers produced at very high stretching ratios in the early reports may limit their applications in practical volume manufacturing.

In this work, we produced PE/BNNP nanocomposite films with up to 15 w.t.% BNNP ($\kappa = 300 \text{ W m}^{-1} \text{ K}^{-1}$ [30]). Uniform BNNP dispersion in PE matrix has been achieved through combined melt polymer processing and uni-axial stretching techniques. Under optimal conditions ($\phi_{\text{BNNP}} = 15 \text{ w.t.\%}$ and a stretching ratio of 5), the nanocomposite film can achieve a metal-like thermal conductivity ($\kappa = 106 \text{ W m}^{-1} \text{ K}^{-1}$) that outperforms that of many pure metals (e.g., stainless steel, $\sim 14 \text{ W m}^{-1} \text{ K}^{-1}$) [32]. This is the first report where PE/BNNP nanocomposites is engineered through primary and secondary polymer processing to achieve a unique microstructure and ultra-high thermal conductivity. The work demonstrates the strong potential of our approach for the production of cost-effective high performance thermally conductive polymer composites through scalable manufacturing process.

2. Experimental

2.1 Materials

High density polyethylene pellets (HDPE HTA-108, average MW 123,400, 0.961 g/cm³) were supplied by ExxonMobil. h-boron nitride nanoplatelets (BNNP, 99%, 2.25 g/cm³) with diameter of 0.1-3 μm and thickness of $\sim 100\text{ nm}$ were purchased from Sigma-Aldrich, UK.

2.2 Nanocomposite processing

As is shown in Figure 1, nanocomposites with $\phi_{\text{BNNP}} = 5\text{ w.t.\%}$, $10\text{ w.t.\%}$ and $15\text{ w.t.\%}$, respectively, were prepared by blending ground PE powders with BN powders at a speed of 2000 rpm/min using a PRISM Pilot 3 high speed mixer. The premixed blends were then melt-mixed in a Collin ZK 25 twin-screw extruder and pelletized. The bespoke screw configuration profile of the extruder ensures an excellent dispersion of nanofillers within the polymer matrix even at high nanofiller loading up to 20 w.t.% [31]. The pellets were then extruded into a 1 mm thick, 100 mm wide sheet using a single-screw extruder with a finishing calendar/chill roll unit. Afterwards, the extruded PE sheets were cut into 76 mm $\times$ 76 mm squares for uniaxial stretching using an in-house biaxial stretcher. PE sheet (76 mm $\times$ 76 mm $\times$ 1 mm) was first heated to 160 °C and hold for 4 min to erase any materials processing history. The sample was then cooled down to 110 °C at a cooling rate of 80 °C/min, followed by uniaxial stretching at different stretching ratios ($\lambda = 1, 2, 3, 4, 5$) in air. Further details regarding material processing can be found in the Supplementary Information.

Figure 1. Preparation of PE/BNNP composite film (a) raw materials, (b) raw powers were mixed using high speed mixture, (c) mixed powers were extruded / pelletized using a single screw extruder, (d) as-extruded PE/BNNP firm made from pellets, (e) PE/BNNP nanocomposite film undergoing uniaxial stretching.

2.3 Characterization

Transmission electron microscopy (TEM, JEOL JEM-2100F) was used to observe the BNNP structure. Microtomed specimen with thickness less than 100 nm was prepared by a Reichert-Jung Ultracut E. ulstra-Microtome using a 30° angle diamond knife at room temperature. Field-emission scanning electron microscopy (SEM, Nova NanoSEM450, FEI, USA) was used to investigate the microstructure of the polymer and nanocomposite samples. Specimens were etched at ambient temperature for 48h. The etching reagent is potassium permanganate (0.5%) in a mixture of concentrated sulfuric acid (98%) and concentrated nitric acid (65%). After washing and drying completely, the etched surface was gold sputtered and observed at an accelerating voltage of 20 kV. Thermal diffusivities (α) and specific heat capacity (C_p) were measured using a laser-flash conductometer (LFA 467, NETZSCH, Germany) based on xenon flash lamp source at room temperature under ambient conditions. The thermal conductivity was determined by comparison with a pyroceramic reference sample. The composite samples were 25.4 mm in diameter. At least three samples were tested for each group and average value was

taken. κ was calculated by $\kappa = \alpha \times \rho \times C_p$, where ρ is the density of materials. Two-dimensional wide-angle (2D-WAXD) X-ray scattering measurements were carried out using Hefei National Synchrotron Radiation Laborator (NSRL) X-ray Diffractometer. A 2D CCD X-ray detector (MARUSA) (1024×1024 pixels in an area of $150 \times 150 \mu\text{m}^2$) was employed to collect the signal pattern (X-ray wavelength: $1.54 \text{ \AA}$). More details regarding sample characterization can be found in Supporting Information.

3. Results and discussions

Figure 2a shows typical TEM image of BNNP fillers which feature irregular plate shape. BNNP in microtomed PE/BNNP nanocomposite film (Figure 2b) shows that the nanoparticle retains its typical multi-layer stacked structure when embedded in the polymer.

Figure 2 TEM images of (a) typical BNNPs and (b) BNNP embedded in PE.

To better understand the morphological evolution of polymer and nanocomposites under the stretching process, SEM was used to observe the microstructure of the PE and PE/BNNP nanocomposites. For unstretched PE, it can be seen that the etching process can effectively remove the top amorphous layer from the as-extruded polymer (Figure 3

(a)), revealing the randomly arranged lamellar structure of PE crystals (Figure 3 (b)).

Figure 3. Top view SEM images of (a) unetched as-extruded PE and etched (b) as-extruded PE, (c) stretched PE film, (d) stretched PE film at higher magnification, inset shows close-up image of PE lamellae (indicated by blue arrows) along the stretched polymer chain (indicated by green arrow) (d) stretched PE/BNNP nanocomposite film, (e) stretched PE/BNNP nanocomposite film at higher magnification, (g) and (h) cross-sectional view SEM images of stretched PE/BNNP nanocomposites (Yellow arrows depict stretching direction, $\lambda = 5$ for all stretched samples).

After the stretching process ($\lambda = 5$), the lamellae within the neat PE appeared were more orderly aligned, see Figure 3(c) and (d). Figure 3(d) inset shows that the PE lamellae (indicated by blue arrows) grew epitaxially on the stretched chains (indicated by green arrows) upon stretching and they were perpendicular to the chain. Figure 3 (e) shows that for stretched PE/BNNP nanocomposite film, the PE lamellae showed similar aligned arrangement, with well dispersed BNNPs. No sign of BNNP percolation network was observed. Close up image in Figure 3(f) shows that apart from the regularly aligned PE lamellae, some lamellae were also found to be epitaxially adsorbed along the BNNP edges and/or partially covered the BNNP surface. In order to further confirm the BNNP distribution within the stretched nanocomposite film, the microstructure of the composite film was also imaged from its cross-sectional surface (see Figure 3 (g) and (h)). As expected, BNNPs were well dispersed and aligned along the stretching direction. The fact that BNNPs were isolated from each other further confirmed the thermal transport within the composite is not through BNNP percolation network.

Figure 4 shows a schematic illustrating the re-alignment of PE lamellae, stretched chains

and the BNNP as a result of the uniaxial stretching.

Figure 4. Schematics showing (a) pristine PE with randomly distributed lamellae and PE chains (b) Uniaxially stretched PE with aligned lamellae and PE single chains (c) Uniaxially stretched PE/BNNP nanocomposite with aligned lamellae, PE single chains and BNNPs.

In-plane thermal conductivities of all sample films were measured using the laser flash method following established protocol detailed elsewhere [33-35]. The technique captures the temperature rise at the back surface of the specimen produced by a short energy pulse on the top surface. Figure 5 (a)-(d) summarizes the thermal conductivity results of neat PE films and the nanocomposite films with varying BNNP concentrations ($\phi_{BNNP} = 5$ w.t.%, 10 w.t.% and 15 w.t.%, respectively), at different λ . As shown in Figure 5 (a), the κ of neat PE films increased with increasing λ and reached a maxima of ~ 2.41 $\text{W m}^{-1} \text{K}^{-1}$ at $\lambda = 5$ (approx. 7.5 times greater than that of the unstretched PE (~ 0.32 $\text{W m}^{-1} \text{K}^{-1}$)). Note that within the neat polymer, both PE lamellae and the stretched chains can contribute to the thermal conduction. Since the λ we used (maximum $\lambda = 5$) was moderate, the lamellae orientation may not be perfect, and the stretched molecular chains may not be long enough to connect all lamellae to form an highly effective thermal conduction path, and hence the enhancement of κ in stretched neat PE was very limited.

Figure 5 (a) Thermal conductivity of neat PE films as a function of Λ . (b) Thermal conductivity of unstretched nanocomposite films as a function of ϕ_{BNNP} . (c) Thermal conductivity of PE/BNNP nanocomposite films as a function of ϕ_{BNNP} at various Λ . (d) Experimental data and curves fitting for thermoreflectance signal as a function of time ($\phi_{\text{BNNP}} = 15$ w.t.% at $\Lambda = 3, 4$ and 5 , respectively). (e) Thermal conductivity of nanocomposite films ($\phi_{\text{BNNP}} = 15$ w.t.%) as a function of Λ . (f) Enhancement of κ for nanocomposite films stretched at $\Lambda = 5$ as a function of ϕ_{BNNP} .

When BNNPs were introduced, their large aspect ratio allowed them to “bridge” the neighboring lamellae within the unstretched composites, thus facilitating the heat transfer. As expected, κ of the nanocomposite films increased with increasing ϕ_{BNNP} and a maximum κ of $1.29 \text{ W m}^{-1} \text{ K}^{-1}$ was achieved at $\phi_{\text{BNNP}} = 15$ w.t.%, see Figure 5 (b). With uniaxial stretching, the κ of nanocomposite films can be further enhanced, see Figure 5 (c). Notably, under $\Lambda = 5$, the κ for nanocomposite increased dramatically from $3.1 \text{ W m}^{-1} \text{ K}^{-1}$ to $106.2 \text{ W m}^{-1} \text{ K}^{-1}$ at $\phi_{\text{BNNP}} = 15$ w.t.%. Such a dramatic boost of κ may be

related to the presence of multiple thermal conduction pathways within the composite film due to (1) greater number of stretched PE chains, (2) more orderly lamellae alignment [36-38] and (3) increased BNNP “bridges”. The results indicate that both ϕ_{BNNP} and Δ complement each other in enhancing the nanocomposite thermal conductivity. Figure 5 (d) shows the thermorefectance signal evolution as a function of time, from which one can obtain the samples in-plane thermal diffusivity under varying Δ ($\phi_{BNNP} = 15$ w.t.%). Figure 5 (e) shows the κ value of the nanocomposite films for $\phi_{BNNP} = 15$ w.t.% under various Δ . The κ increased with Δ exponentially, suggesting possibility for further conductivity enhancement beyond $\Delta = 5$. The enhancement of κ in Figure 5 (f) can be calculated according to Equation 1.

$$\Delta = (\kappa_c - \kappa_{pe} / \kappa_{pe}) \times 100\% \quad \text{Eq (1)}$$

where κ_{pe} represents the thermal conductivity of unstretched neat PE, κ_c represents the thermal conductivity of nanocomposite films.

The enhancement of κ (Δ) increases with increasing ϕ_{BNNP} with a dramatic increase at $\phi_{BNNP} = 10$ w.t.%, indicating the formation of an effective thermal transport network within stretched nanocomposite film at this ϕ_{BNNP} .

To further confirm the impact of uniaxial stretching process on the PE lamellae alignment, the degree of orientation and crystallinity of PE lamellar was studied by a two-dimensional wide-angle synchrotron X-ray diffraction (2d-WAXD). Figure 6 and Figure S3 shows the 2d-WAXD patterns of all nanocomposite films. The isotropic rings of PE (110) and (200) lattice planes are clearly shown for the non-stretched samples, suggesting

the PE lamellae were randomly positioned within the composite films. With increasing λ , the isotropic rings became short arcs along the equatorial direction, suggesting the lamellae start to orient along the stretching direction (lamella perpendicular to the stretching direction). In addition, a weak isotropic ring of (002) BNNP lattice plane is visible which aligns along the stretching direction as λ increases. These results collaborate well with the SEM observations.

Figure 5 Wide angle X-ray diffraction (WAXD) patterns for nanocomposite films with $\phi_{BNNP} = 0, 5, 10$ and 15 w.t.% at $\lambda = 1, 2, 3, 4$ and 5 , respectively. White arrows highlight the characteristic Bragg scattering of the (110) and (200) planes of PE and (002) plane of BN. The PE lamellae are perpendicular to the stretching direction.

Herman's method [39] was used to calculate degree of orientation of the PE lamellae by quantifying the average orientation between the normal of lattice plane and the stretched direction, see Table 1.

Table 1 | Degree of lamellar orientation in nanocomposite films stretched at various λ

BN loadings (w.t.%)	Degree of lamellar orientation				
	SR = 1	SR = 2	SR = 3	SR = 4	SR = 5
0	0	0.86	0.974	0.98	0.985
5	0	0.958	0.967	0.974	0.982
10	0.04	0.96	0.966	0.971	0.98
15	0.11	0.955	0.965	0.97	0.98

For neat PE, the degree of lamellar orientation increased rapidly with increasing λ and the values stabilized when λ reached 3. For the nanocomposite films, the presence of BNNPs (e.g., $\phi_{BNNP} = 10$ and 15 w.t.%) has a pronounced effect on enhancing the lamellar orientation, particularly at lower stretching ratios. This could be because during the stretching, the rigid 2D BNNPs were being aligned by the shear stress, their alignment can also facilitate the orientation of the surrounding lamellae. At larger stretching ratios ($\lambda \geq 2$), the enhancing effect of BNNP on the lamellae orientation is negligible, and the shear stress induced by the stretching process predominates the lamellae orientation. Supplementary Table S1 shows the degree of crystallinity for all testing samples. Given the range of errors (i.e. 60 % (± 5 %)), the variation of crystallinity for different samples are negligible. This suggests that the crystallinity does not play a significant role in the drastic κ enhancement. This is in contrast to the past theory where crystallinity-dependent thermal conductivity in polymers was emphasized [22].

Figure 7 shows the schematic illustrating the potential thermal conduction mechanisms within the uniaxially stretched PE/BNNP nanocomposites. Multiple thermal transport pathways are possible with the oriented lamellae, stretched polymer chains and BNNP forming a co-continuous network structure. When heat is applied from one end of the stretched nanocomposite film, the atoms within the lamella gain the energy and diffuse it to the whole lamellar crystal through phonon transfer. Stretched single PE chain with high κ ($1400 \text{ W m}^{-1} \text{ K}^{-1}$) or BNNP ($300 \text{ W m}^{-1} \text{ K}^{-1}$) can both bridge the folded lamella, capturing the thermal energy and transferring it rapidly to an adjacent heat conducting element. The melt processing followed by uniaxial stretching of the nanocomposites has thus created a unique co-continuous network structure where the thermal energy can diffuse easily and rapidly through the composite film, resulting in a metal-like κ of the resulting material.

Figure 7 Schematic showing potential thermal conduction mechanisms within the stretched nanocomposite film.

4. Conclusions

Through melt-processing and uni-axial stretching, we designed and manufactured PE/BNNP composite films the thermal conductivity of which can be tailored by manipulating the BNNP loading and the stretching ratio λ . By optimizing the process conditions ($\phi_{BNNP} = 15$, $\lambda=5$), a metal-like thermal conductivity ($106 \text{ W m}^{-1} \text{ K}^{-1}$) can be achieved by the nanocomposite produced. Such remarkable thermal performance has been attributed to the effective phonon transfer within the co-continuous network structure consisting of interconnected PE lamellae, stretched PE chains and oriented BNNP. In contrast to other thermally conductive polymer composites which consist of much greater loading of thermally conductive filler materials, our composites offer better filler dispersion, lighter weight and lower costs. In addition, the easy/ scalable processing of our composites would allow its promising future applications that require effective thermal managements and electrical insulation.

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