# Mechanical, toughness and thermal properties of 2D material- reinforced epoxy composites

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## ARTICLE INFO

## ABSTRACT

Developing epoxy composites with high thermal conductivity and excellent mechanical properties becomes imperative in electronic and aerospace industries. This study investigates and compares the effect of adding boron nitride (BN) sheets and graphene platelets (GnPs) on the mechanical properties and thermal conductivity of epoxy resin. The study shows that incorporation of BN or GnPs into epoxy matrix significantly enhanced both mechanical properties and thermal conductivity of epoxy composites. At fractions ranging 1–4 wt%, GnPs/epoxy composites provide higher Young's modulus, fracture toughness ($K_{1c}$) and critical stress energy release rate ($G_{1c}$) compared to BN/epoxy composites. The thermal conductivity of the epoxy composites is up to the maximum of 0.33 $Wm^{-1}K^{-1}$ at 4 wt% of GnP loading, which is much higher than that of the composites filled with the same loading of BN (0.23 $Wm^{-1}K^{-1}$). The study emphasizes the importance of adding thin nanosheets (thickness 3–5 nm) at low loadings in developing epoxy composites to achieve desired mechanical and thermal properties.

## 1. Introduction

Epoxy resins are widely used as adhesives and coatings in electronic and aerospace industries due to their high specific strength, high processability, cost-effectiveness, excellent thermal stability and outstanding weather resistance. However, epoxy resins are inherently poor in thermal conductivity, which limits their applications. Also, epoxy resins are highly crosslinked polymers and thus are very brittle adhesives where they are prone to fast crack propagation under dynamic loads. Enhancing fracture toughness of epoxy resins is one of the ongoing research using various approaches [1,2] such as adding nanofillers into epoxy resins including (but not limited to) rubber nanoparticles [3,4], nanosilica [5], nanoclay [6] and metal nanoparticles [7]. Adding high thermally conductive nanofillers into epoxy is proved to be an effective and economical approach to improve its thermal conductivity [8–12], toughness [13–15] *and thus widen its applications.*

Employing micro or nanofillers with intrinsically high thermal conductivity into epoxy (in general polymers) creates heat-conducting paths through which the composite would be able to dissipate heat efficiently. In the past two decades, typical fillers including graphite, carbon black, carbon fibres and ceramics with high thermal conductivity were added at high loadings (~50 wt%) to enhance polymer's thermal conductivity [16,17]. High loadings of microfillers result in low mechanical properties and processing difficulties of the host matrix. Therefore, reducing filler's fraction to a low level in composites for achieving both desired thermal conductivity and mechanical properties is a challenge.

Owing to the outstanding specific surface area of nanofillers, they can form heat-conducting paths across the composite at substantial low concentrations. Two-dimensional graphene has pinched intensive attention from academia to fabricate highly thermal conductive polymers due to its exceptional thermal conductivity (~5300 W/mK) through phonons transport [16,17]. Their potential incorporation in

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polymers for thermal conductivity enhancement has been reported in many studies [18–21]. Haddon et al. [22] demonstrated a ~600% enhancement in thermal conductivity of epoxy resin when 10 wt% of graphite nanoplatelets were added. It reached even higher of ~800% when graphite nanoplatelets and single-walled carbon nanotubes were added together at 3:1 ratio of total 10 wt%.

In our previous works, graphene platelets (GnPs) of thickness ~3.57 ± 0.5 nm were fabricated using thermal shock at high temperature followed by ultrasonication [23]. Comparing to graphene oxide and reduced graphene oxide, GnPs have following advantages [24,25]: (a) high electrical conductivity of 1460 S/cm due to less defect concentration [26], and thus chemical reduction is not needed, (b) a thickness around 3 nm provides large contact surface area to interface with the matrix for efficient stress, electron and phonon transfer, and (c) scalable and cost-effectiveness (~$20/kg) in fabrication. Therefore, GnPs prove to be a promising candidate for developing multifunctional polymer nanocomposites.

Boron nitride nanosheets (BN), two-dimensional nanofillers, possess high thermal conductivity (250–300 $Wm^{-1}K^{-1}$) at 25 °C [27], excellent electrical insulation, superb chemical resistance and low coefficient of friction. Recently, BN attracted much attention for further studies on its properties [28,29] and its role to promote the polymer properties for multifunctional purposes [30–33]. Yao et al. [34] incorporated BN sheets into epoxy resin to enhance thermal properties of the epoxy. The results showed that at 2.7 vol%, the thermal conductivity of epoxy nanocomposites was improved by 27.5% when BN sheets were modified by hyperbranched aromatic polyamide compared to only 2.7% when pristine BN was added.

There is number of studies available in the literature, which investigated the mechanical and thermal properties of the epoxy/GnP composites [35], while few studies have focused on epoxy/BN composites. Despite both GnPs and BN are two-dimensional nanomaterials, the behavior of BN and GnPs and their interfaces with polymers are dissimilar owing to the contrast in their intrinsic physical and chemical properties. In this work, we investigated and compared the effect of adding BN and GnP into epoxy matrix on its mechanical and thermal properties at a range of loading 0–4 wt%. The results showed that GnPs perform better than BN in both thermal conductivity and mechanical properties.

## 2. Experiments

### 2.1. Materials

Asbury Carbons, Asbury, NJ kindly supplied graphite intercalation compound (1721). Boron Nitride nanosheets (hexagonal crystal, 30–50 nm in size, purity >99.9%) were purchased from *Shanghai Chaowei Nanotechnology Co., Ltd., China*. Epoxy resin (WSR 618, diglycidyl ether of bisphenol-A) was provided from Nantong Xingchen Synthetic Material Co., China. Jeff-amine 400 (J-400) hardener was supplied by *Huntsman, China*. All materials were used as supplied without any further purification.

### 2.2. Fabrication of epoxy composites

Boron nitride sheets (BN) were used as received. Epoxy/BN composites were produced as follows. BN was added into acetone at 1 wt% in a metallic container and magnetically stirred for 10 min, followed by ultrasonication for 1 h below 25 °C. Then, DGEBA was added and fully dissolved into the mixture after 10 min magnetic stirring followed by 30 min ultrasonication to entangle epoxy molecules with the BN sheets. Using a hot plate at 70 °C and magnetic stirring, acetone was completely evaporated from the mixture leaving a viscous mixture of DGEBA and BN. To confirm the mixture is free of trapped gasses or acetone, it is degassed in a vacuum oven at 100 °C for 10 min. When the mixture cooled down to 30 °C, the hardener–J400 was slowly poured into the

mixture, and manually stirred for 5 min. Finally, the epoxy/BN composites were poured into the molds for curing. Preparation of GnPs is fully described elsewhere [36]. Epoxy/GnP composites were prepared by following the same procedures used for the epoxy/BN composites.

Epoxy/BN and epoxy/GnP viscous mixtures were then poured into silicone rubber molds. The filled rubber molds were carefully moved to a fan oven for curing. The curing process has two stages: 80 °C for 2 h and then 120 °C for 10 h. The final samples were polished to suppress any visible flaws or marks. At last, the polished samples were treated in a common oven at 100 °C for one hour to diminish any flaws arisen during polishing.

### 2.3. Characterizations

Although the thickness of boron nitride sheets is provided by the manufacturer and thickness of graphene platelets was confirmed in previous studies [21,37], atomic force microscopy was used to confirm the thickness of both sheets under identical conditions. Atomic force microscopy (AFM) measurements and micrographs were obtained by using NT-MDT NTEGRA SPM instrument equipped with NSG03 non-contact "golden" cantilever. Each sample was prepared by suspending the sheets in N-methyl-2-pyrrolidone (NMP) at 1 wt% and then diluted sequentially to 0.0004 wt% by 15-min ultrasonication to avoid stacking. The 0.0004 wt% suspension was dropped on silicon wafer followed by drying at 150 °C in a fan oven. Then, the samples are ready for AFM measurements.

Transmission electron microscopy images were taken using (Philips CM200, TEM) at 200 kV as an accelerating voltage. 50 nm-thick sections of both GnP and BN epoxy composites at 2 wt% were microtomed using Leica Ultracut S microtome with a diamond knife and collected on 200-mesh copper grids. The fracture surfaces were observed by scanning electron microscope (SEM, Philips 505) at 10 kV. Thin layers of platinum were coated over the fracture surfaces before SEM imaging.

Dynamic mechanical analysis (DMA) was conducted at frequency of 1 Hz on DMA 2980 TA Instrument, Inc., USA. DMA was run on a single cantilever clamped-rectangle samples of cross-section (4 × 12 mm) with free span 20.00 mm at temperature ranging 20–150 °C. Samples were tightened on the clamp using a torque of 1 N·m. Tensile testing was carried out using dumbbell samples at speed of 0.5 mm/min. An extensometer was attached to the middle part of the dumbbell sample to capture the elastic strain; Young's moduli were determined at strain range 0.0005–0.0015. The fracture toughness ($K_{IC}$) of the neat epoxy and its composites was calculated using compact tension (CT) test according to ISO13586 standard. The CT samples have dimensions of 30 × 30 × (5–6) mm. At least three replicates were used for each test and fraction. The records are the average of the results from the tested replicates.

Thermal conductivity of the composite was measured using TA Instruments DTC300 analyzer–Fig. 1a. The test samples were placed between the upper and lower heating plates. The temperature difference between the two plates is 30 °C. The lower plate is part of the calibration heat flow meter. An axial temperature gradient is formed when the heat is passed from the upper plate through the sample to the lower plate. The tests are in accordance with the ASTM E1530 Standard. A schematic diagram of thermal conductivity measurement is shown in Fig. 1b.

## 3. Results and discussions

### 3.1. Thickness of graphene platelets and boron nitride sheets

Atomic force microscopy (AFM) was employed to examine the thickness of fillers; graphene platelets (GnPs) and boron nitride sheets (BN) in this study. For each sample, 20 randomly selected platelets were measured, and their representative micrographs are shown in Fig. 2 a&b. The BN nanosheets show thickness 15 ± 2.47 nm in Fig. 2a while GnPs have far lower thickness of 3.12 ± 0.45 nm as in Fig. 2b. This
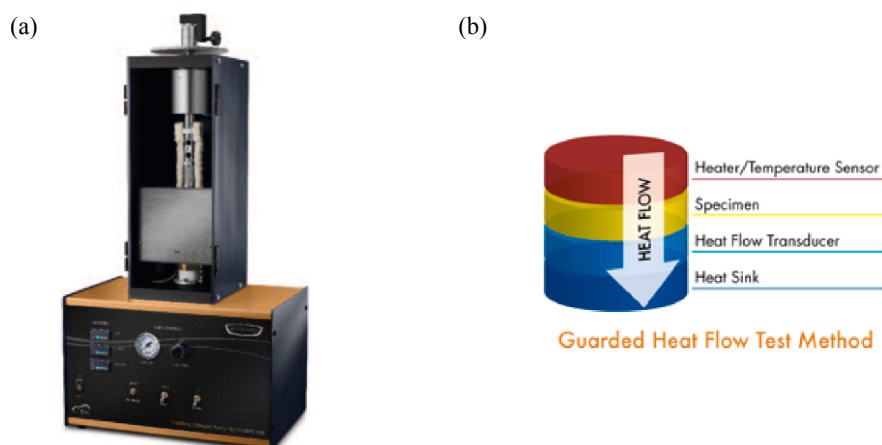
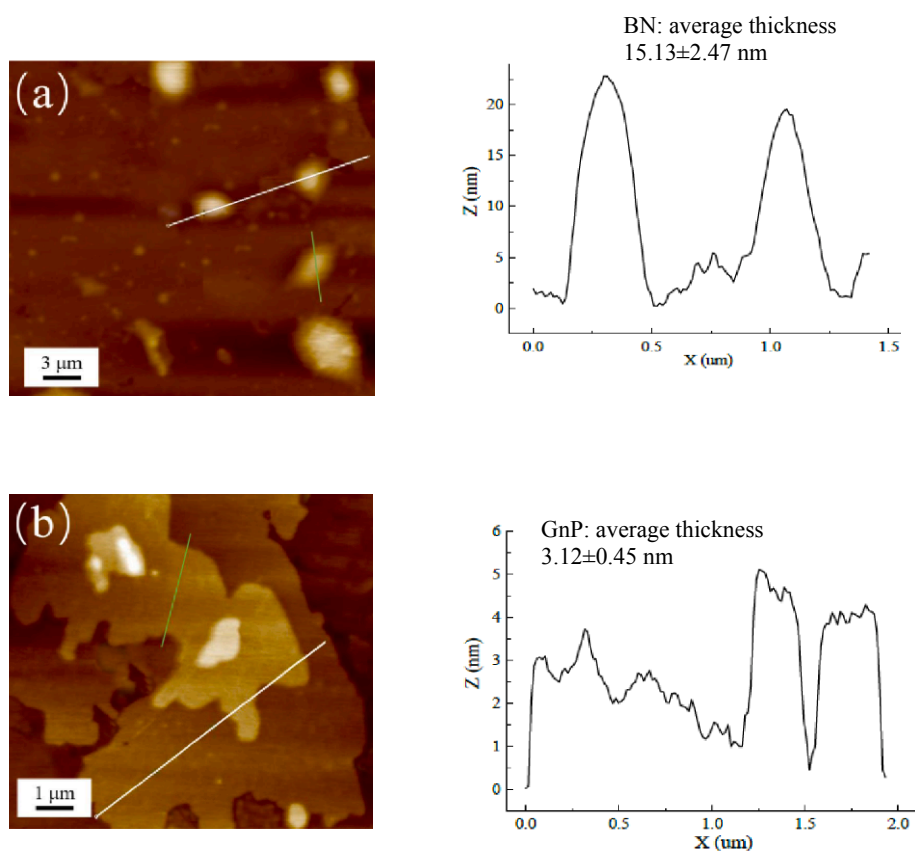Fig. 1. (a) Thermal conductivity tester DTC300 and (b) test procedure [38].



Fig. 2. AFM micrographs and thickness measurements of boron nitride nanosheets and graphene platelets.

thinner thickness of GnPs reflects outstanding specific surface area with small inter-distance between the platelets inside the matrix leading to strong physical interface with the matrix compared to BN.

### 3.2. Filler dispersion in epoxy matrix

Quality of filler dispersion and its interface with the host matrix are crucial factors to define the mechanical and functional properties of the final product. *The thickness of GnP is measured as 3.12 ± 0.45 nm which confirms that each GnP contains 3–4 graphene layers* [15,39]. *The result is consistent with previous studies* [26,39,40]. Boron nitride sheets (BN) were employed into epoxy resins as received; its thickness is 30-50 nm (the information is provided by the supplier). However after sonication for

30 min– the AFM measurement showed that BN sheets possess 15.13 ± 2.47 nm, yet it is far larger compared to GnP's thickness. TEM analysis is used to examine the dispersion quality of epoxy/GnP and BN composites as displayed in Fig. 3. Generally, BN and GnPs are uniformly dispersed and properly implanted into the epoxy, demonstrating strong interactions between the fillers and matrix. Fig. 3a represents a typical TEM micrographs of the composite containing 2 wt% BN. A selected area was magnified in Fig. 3b showing that BN forms aggregates (white arrows). However, these thick aggregates are equally dispersed in the entire matrix.

Fig. 3c contains low magnification micrograph of 2 wt% epoxy/GnP composite, where separate-dispersed GnPs are observed. Fig. 3d shows a high magnified area, and the dispersion of GnPs in the epoxy resin is
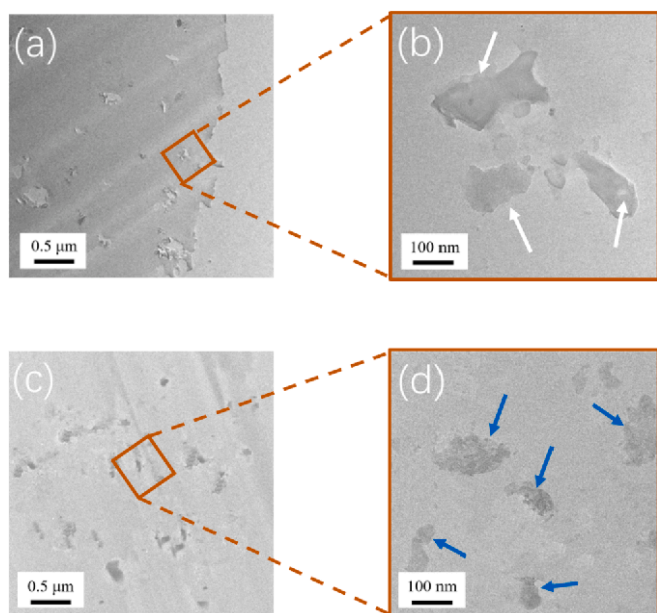
**Fig. 3.** TEM images of (a,b) 2 wt% epoxy/BN composite and (c,d) 2 wt% epoxy/GnP composite.

relatively uniform as depicted by blue arrows. The TEM analysis proves that GnPs disperse better in the matrix in comparison to BN. This difference is a key factor in determining the mechanical and thermal properties of the epoxy composites as discussed in the following sections.

Moreover, chemical structure of both fillers has influence on the dispersion and interface of fillers with the matrix. Graphene consists of carbon atoms arranged in a honeycomb structure with hybrid $sp^2$ atoms; polymers are hydrocarbon materials where carbon atoms are the backbone of most of them. This makes graphene and graphene derivatives are compatible with polymers which in turn enhances the physical interface between graphene fillers and the host matrix. On the other hand, boron nitride is a compound of boron and nitrogen elements which does not match with the chemical structure with most of polymers leading to poor physical interface with the matrix unless the BN surface is chemically modified.

### 3.3. Fracture toughness and mechanical properties

Fracture toughness ($K_{1c}$) – which measures the absorbed energy to propagate sharp crack – and critical strain energy release rate ($G_{1c}$) are primary properties to evaluate the toughness of epoxy resins. Fig. 4a&b contains both fracture toughness $K_{1c}$ and critical strain energy release rate $G_{1c}$ of the prepared composites as a function of the filler's content. Both $K_{1c}$ and $G_{1c}$ of epoxy composites are significantly improved by adding BN and GnPs up to 2 wt% at different increments; for example, $K_{1c}$ and $G_{1c}$ of epoxy/GnP composites are improved by 160% and 338%, respectively, while 121% and 280% increase in case of BN. The increment of these properties slows down once the content of the fillers is higher than 2 wt%; at 3 wt%, $K_{1c}$ and $G_{1c}$ start to decline but still higher than that of neat epoxy.

Fig. 4 c&d shows Young's moduli and tensile strengths of epoxy/BN and epoxy/GnP composites. The Young's moduli steadily increase with the individual addition of BN and GnP. Tensile strengths of both epoxy/BN and epoxy/GnP composites slowly decline with increasing of fillers' content. The drop in tensile strength is well-known for brittle
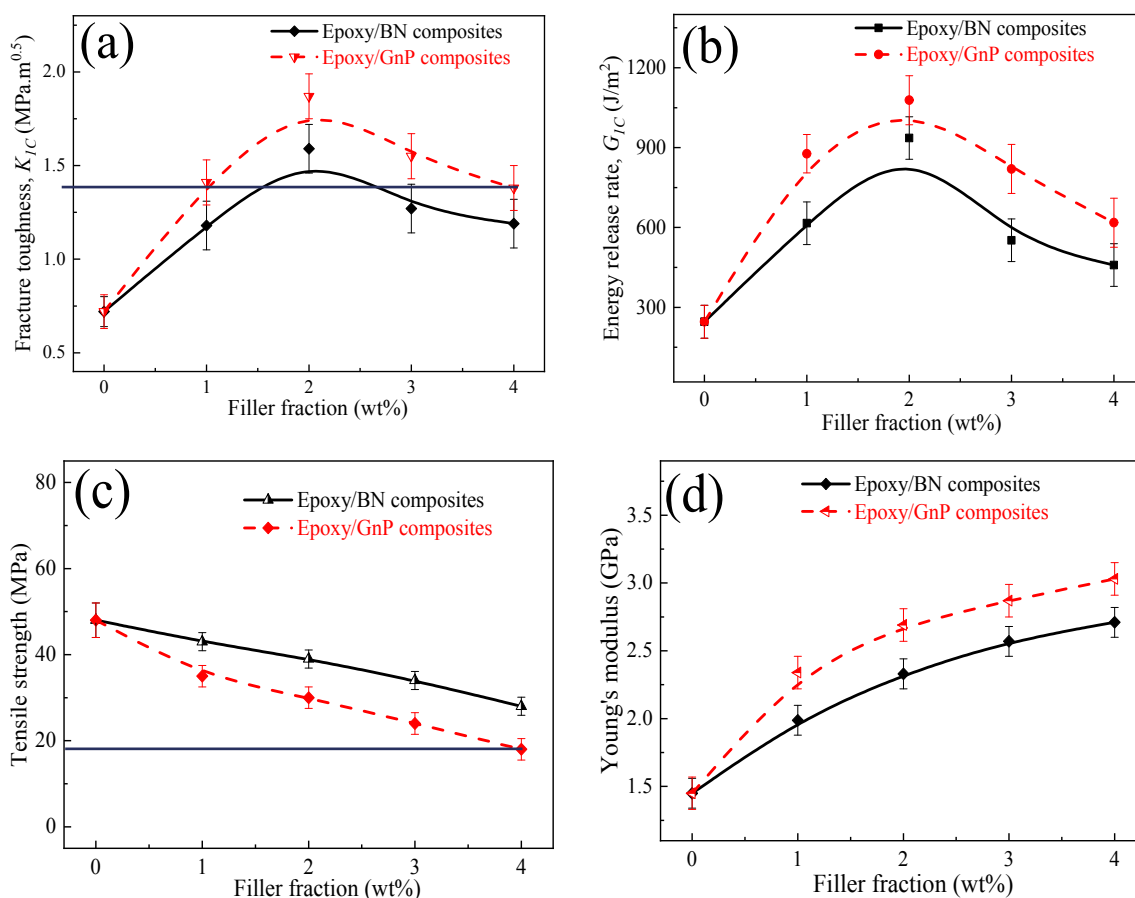


**Fig. 4.** Toughness and Mechanical properties of epoxy/BN and epoxy/GnP composites.

crosslinked polymers such as epoxy due to stiffening effect and debonding between the filler and polymer. This conforms with the previous studies [39–41].

Fig. 4 shows that the enhancements achieved by adding GnP into epoxy resin are much higher compared to those obtained by adding BN, in all tested properties including fracture toughness ($K_{1c}$), critical strain energy release rate ($G_{1c}$) and Young's modulus; while the drop in tensile strength is more in case of adding GnPs compared to that of adding BN. For example, at 2 wt%, the $K_{1c}$, $G_{1c}$, Young's modulus and tensile strength of epoxy/GnP are 1.87 MPa m$^{0.5}$, 1078 J/m$^2$, 2.69 GPa and 30 MPa compared to 1.59 MPa m$^{0.5}$, 936 J/m$^2$, 2.33 GPa and 39 MPa in epoxy/BN composites, respectively.

The results are explained as follows. Adding BN and GnP restricts the mobility of epoxy's chain resulting in high stiffness. At low fraction (<2 wt%), fillers have much space to disperse and thus can achieve strong interactions with the matrix; therefore, stress transferring and load sharing between the filler and matrix are improved. While at high fraction, the fillers become swarming and aggregating so that clustering has high chance to be formed. These aggregates create stress concentration locales and hence cracks can be easily initiated.

Thickness of the nanofiller is a key factor in developing a strong interface between the matrix and filler [37]. Giving that GnP has a thickness of ~3.12 nm compared to 15.13 nm (30–50 nm as provided by the manufacturer) in case of BN, the contact surface area with the matrix is substantially larger in GnP providing that dispersion degree is similar. This promotes the interfacial strength resulting in high mechanical performance. Also, thicker fillers give thick aggregates which weaken the stress sharing between the matrix and filler and act as stress concentration locales. This explains why the higher increments in mechanical properties happen in case of GnP than epoxy/BN composites.

### 3.4. Dynamic mechanical analysis (DMA)

Dynamic mechanical analysis (DMA) is an effective tool to evaluate the viscoelastic properties of polymers and their composites including storage modulus, loss modulus and mechanical loss factor (tan δ). DMA examines the molecular relaxation processes of the phases inside a polymer composite [42]. These properties are vastly dependent on the fillers: their dispersion quality in the matrix, fraction, geometry and dimensions, and stress transfer between the matrix and fillers [43].

Damping ratio is important to define the glass transition temperature of the polymer. The glass transition temperature ($T_g$) is the temperature where the polymer molecules transit from glass phase to viscoelastic (rubbery) phase, and it is determined at the peak of tan δ curve. Fig. 5 contains the damping ratio (tan δ) for the neat epoxy and its composites as a function of temperature. Table 1 records the $T_g$ values of neat epoxy

and its composites. $T_g$ is significantly improved at low wt% of BN and GnP; for example, at 1 wt%, $T_g$ increases from ~56 (neat epoxy) to 68.3 °C when BN is added, and to 70.2 °C in case of using GnP. When more filler (>1 wt%) is added to epoxy, $T_g$ steadily reduces.

Fig. 6 presents the storage modulus ($E'$) of neat epoxy and its composites versus temperature. The value of storage modulus indicates the stiffness of epoxy composites before and after glass transition temperature. Storage modulus increases with the addition of BN and GnPs at 30 °C, and it shows that GnPs have better reinforcement efficiency than BN at all filler contents as previously noticed in Section 3.3. The maximum storage modulus is obtained at 1 wt% with an increment of ~46% and ~53% when BN and GnP incorporated into epoxy, respectively. At fraction >1 wt%, the storage modulus declines as the agglomeration and clustering reduce the filler-matrix interaction.

At low fractions, high storage modulus and $T_g$ are due to the following main reasons: (i) dispersion is uniform and the entanglements of the fillers with epoxy molecules are strong; this gives the composites high ability to fight against mechanical deformation and (ii) when the fillers are uniformly dispersed, they reinforce the crosslinking density increasing the stiffness of the polymer with restricted chain mobility. When the temperature is beyond the glass transition temperature, the matrix becomes viscoelastic material and therefore, the matrix molecules have high mobility over the fillers with a low resistance to mechanical loads. This explains the low storage modulus observed at temperatures range 60–90 °C.

It is also depicted from Fig. 5 that the peak value of tan δ is minimum at 1 wt% and maximum for the neat epoxy. The peak value of tan δ signifies the amount of energy dissipated under cyclic load which relates to the interaction between the filler and polymer molecules [34]. The minimum value of tan δ at 1 wt% confirms the strong interaction between the filler and epoxy molecules at low fractions. The difference between the values obtained for epoxy/GnP and epoxy/BN composites are explained in the previous section.

**Table 1**
Glass transition temperature of neat epoxy and its composites.

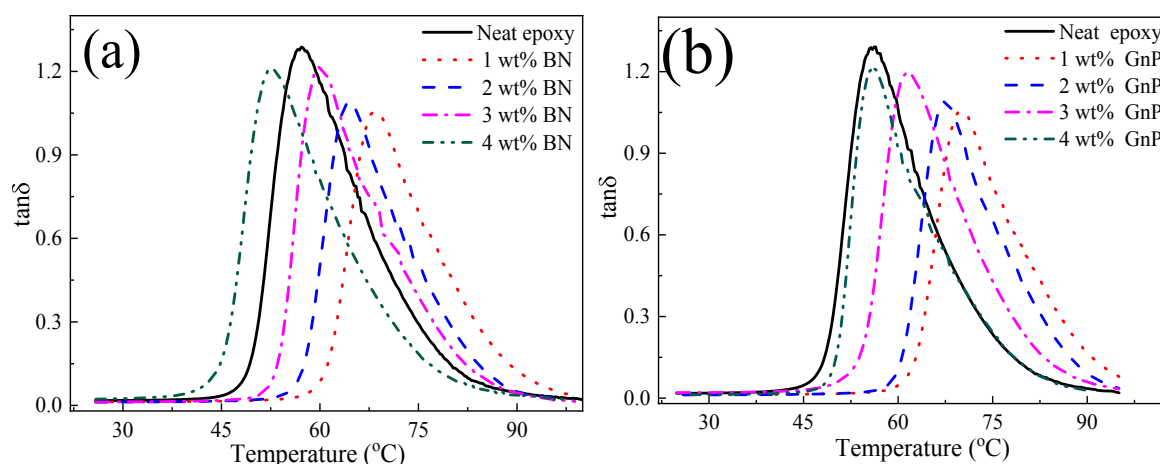| Epoxy composite (wt%) | Glass transition temperature, $T_g$ (°C) | |
|---|---|---|
| | Epoxy/BN composites | Epoxy/GnP composites |
| Neat epoxy | 56.4 | 56.4 |
| 1 | 68.3 | 70.2 |
| 2 | 62.5 | 65.4 |
| 3 | 57.8 | 61.5 |
| 4 | 51.5 | 55.9 |



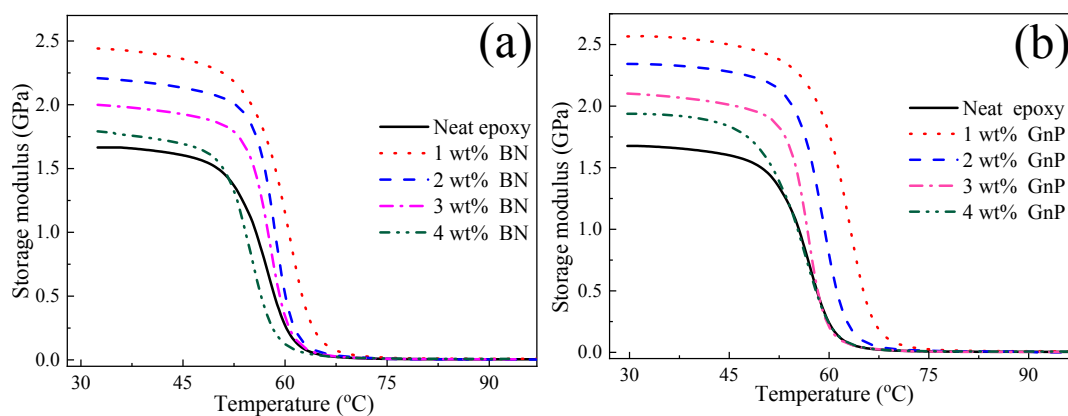**Fig. 5.** Tan delta for (a) epoxy/BN and (b) epoxy/GnP composites.

**Fig. 6.** Storage modulus for (a) epoxy/BN and (b) epoxy/GnP composites.

### 3.5. Thermal conductivity

It is well-known that epoxy (in general polymers) is poor in thermal conductivity due to the inter-chain spaces. Adding high thermal conductive fillers into polymers is one way to enhance their thermal properties. Graphene and boron nitride possess thermal conductivity of 5300 and 300 W/m·K, respectively. Fig. 7 shows the thermal conductivity of neat epoxy and its composites as a function of the filler fraction. It is clear that adding GnP or BN enhances the thermal conductivity of epoxy at different increments. For example, 4 wt% of GnP elevates thermal conductivity of epoxy from 0.15 to 0.353 W/m·K recording 135% increment compared to only 64% in case of BN at the same fraction. Also, it is noted at low filler fraction, the increments are not as substantial as at high fractions.

At low filler content, the filler is segregated inside the epoxy matrix, therefore, the thermal conductivity does not substantially improved. While at 4 wt%, the thermal conductivity of composites increases significantly because the amount of filler is sufficient to form thermal network pathways so that phonons can transfer across the matrix. The epoxy/GnP composites exhibit higher thermal conductivity than epoxy/BN composites, mainly due to the fact that the inherent thermal conductivity of GnPs is much higher than that of BN. Moreover, the GnPs with high specific surface area form plethora heat conductive pathways in the epoxy matrix compare to BN. This leads to a reduction in the thermal interfacial resistance (Kapitza resistance) [21].

### 3.6. Fractography analysis

When a composite is prone to high mechanical deformation leading to failure, it loses its structural integrity at both microscopic and macroscopic levels. Herein, we studied the fracture toughing mechanism by investigating the fracture surface of compact tension (CT) specimens using SEM analysis. The SEM micrographs of epoxy composites at 2 wt% of the fillers are presented in Fig. 8. The fracture surface of neat epoxy is a mirror-like surface as provided in previous studies [1,24] and it is not provided here.

Fig. 8 (a1–a3) shows the fractography of epoxy/BN composites from high to low magnification. Image (a1) shows a relatively rough surface (compared to neat epoxy) which indicates energy absorbed during crack propagation till fracture. Continuous ridges are observed over the surface, indicating that the energy absorption process was not even during
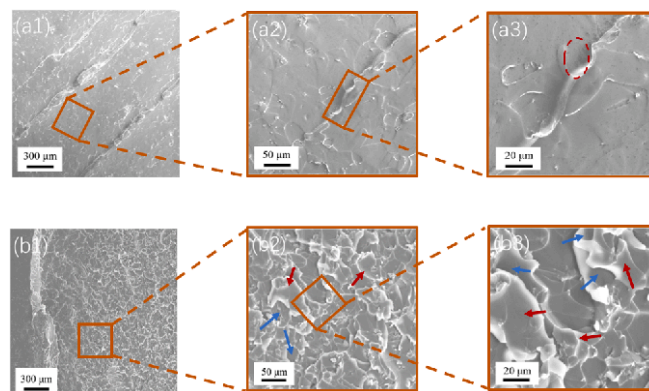


**Fig. 8.** (a1–a3) epoxy/BN composite and (b1–b3) epoxy/GnPs composite at 2 wt%.
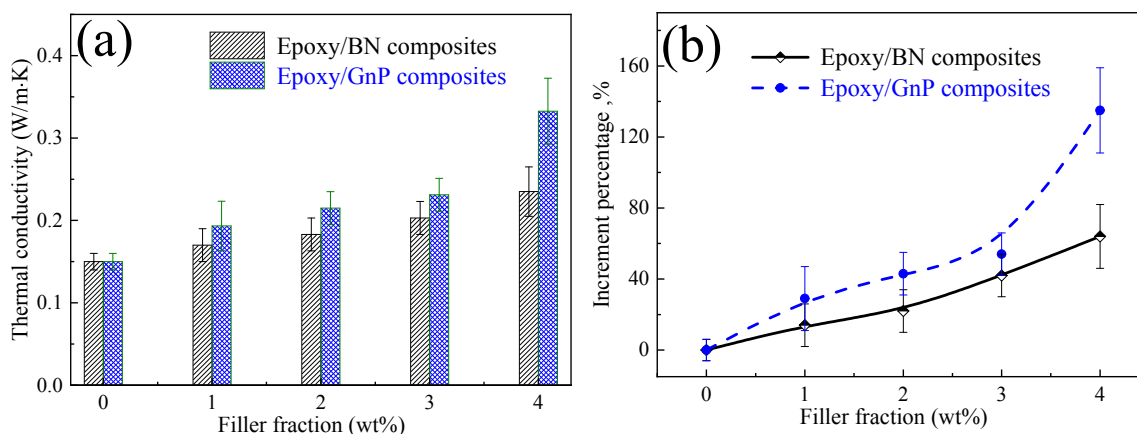


**Fig. 7.** Thermal conductivity of epoxy/BN and epoxy/GnP composites.

the fracture process. Images (a2) and (a3) are high magnification SEM micrographs for the fracture surface. They show irregular and rugged surfaces in both images (a2) and (a3). Also, voids (red circle) are noticed on the surface indicating weak interfacial bonding between BN and epoxy.

Fig. 8 (b1−b3) displays the fractography of compact tension surface of the 2 wt% epoxy/GnP composite. Image (b1) shows an overview of a large area from the fracture surface. It is obvious that the fracture surface is rougher than that of epoxy/BN composite. Also, it unveils that roughness is evenly distributed over the surface which confirmed in images (b2) and (b3). This means large amount of energy was absorbed during the crack propagation till fracture and higher than that consumed in the epoxy/BN composite. This supports the high fracture toughness of epoxy/GnP observed in Fig. 4. Moreover, in images (b2) and (b3), the voids and clusters are observed, which are indicated by blue and red arrows, respectively; most of the cracks propagate near the clusters. However, SEM micrographs of epoxy/GnP prove that GnP has better dispersion and stronger physical bonding interface with the matrix compared to BN. These were reflected in mechanical performance and thermal conductivity of the prepared composites.

In brief, much thinner GnPs (3 nm) have advantages over BN (30–50 nm in thickness). Thin thickness means a high surface area, which leads to a strong interfacial bond to the matrix. This is the key factor in determining the mechanical performance and most of the physical properties. This recommends that further studies need to be conducted to achieve thinner BN sheets.

## 4. Conclusions

This study investigated the mechanical and thermal properties of graphene platelets (GnPs) epoxy composites and boron nitride nanosheets (BN) epoxy composites. The results reveal that BN and GnPs are able to enhance the mechanical properties and thermal conductivity of the epoxy resin. However, although BN and GnP are two-dimensional nanofillers, they enhance epoxy resin at different increments. For example, at 2 wt% filler loading, $K_{1c}$, $G_{1c}$ and Young's modulus of epoxy/BN composites are improved by 121%, 260% and 61% whereas in case of GnPs, they are increased by 160%, 338% and 86%, respectively. At high fraction such as 3 wt%, the mentioned properties are improved by small increments compared to low filler loadings due to filler aggregation and nonuniform dispersion which lead to relatively weak interface between the fillers and matrix. However, the thermal conductivity of epoxy composites recorded its maximum increment at 4 wt% for both GnP and BN; at 4 wt% GnP, the thermal conductivity of epoxy/GnP composite increased by 135% compared to 64% in case of BN at the same fraction.

The results indicate that both GnPs and BN contribute to the composite's mechanical properties, and thermal conductivity. Graphene platelets as a structural reinforcement additive is more effective than boron nitride nanosheets.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgments

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