Non-covalent poly (2-ethylhexyl acrylate) (P2EHA)/functionalized graphene/h-boron nitride flexible composites with enhanced adhesive and thermal conductivity by a facilitated latex approach

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1. Introduction

Efficient thermal management, which might prevent the heat accumulation to damage these devices or severely reduce their speed, reliability, and efficiency. Thermal interface materials (TIMs), applied between heat sources and heat sinks, are essential composition of thermal management [1]. Polymers have been widely used in almost all aspects of modern industrial filed because they are anti-corrosive, light, and easy to process [2]. However, low thermal conductivity of most polymers only around 0.1–0.3 W/m K, limiting their application in the area of thermal interface materials (TIMs) [3]. In order to improve the thermal conductivity of the polymer material, different kinds of thermally conductive fillers such as CNT, graphene, metals, and metal oxides had been utilized to manufacture polymer composites [4–7]. However, poor filler dispersion and high interfacial thermal resistance (TR), limiting the thermal conductivity of the resulting polymer composites reach the expectation [8,9].

Hexagonal boron nitride (h-BN) has a structural similar with graphene, also as a promising fillers in the preparation of thermally conductive and electrically insulative composites, owing to its good thermal conductor with the in-plane thermal conductivity ranging from 200 W/m K to 400 W/m K and the excellent electrical insulator [10]. In addition, the 2D h-BN is expected to introduce the anisotropic feature on the thermal conductivity of composites [11]. The in-plane thermal conductivity of composites was significantly higher than the through-plane thermal conductivity when the h-BN microplates were applied as fillers. The reason was that the oriented arrangement of fillers towards the in-plane direction [12]. Plenty of researches were mainly focusing on preparing the composites with high thermal conductivity by loading these 2D fillers. For example, one strategy to strengthened the phonon coupling in the composites was to improve the affinity between filler and polymer matrix [13]. However, because of the chemical inertness, the surface modification of h-BN was a trouble. Functional molecules with conjugate structures were applied in modifying h-BN nanosheets through π–π interaction. Shen et al. [14] used the dopamine chemistry to achieve the facile surface modification of h-BN microplates by forming the polydopamine (PDA) shell on its surface. In addition, Lewis acid-base
complexation was also used to modify h-BN. Lin et al. [15] modified h-BN with either octadecylamine (ODA) or amine-terminated oligomeric polyethylene glycol (PEG) to obtain ODA- or PEG-functionalized h-BN via the interaction between the amino groups and the boron (B) atoms. Hydroxyl groups can be introduced on the surface of h-BN by treatment with peroxide at high temperature [16]. Yu et al. [17] modified the surface of h-BN by γ-aminopropyl triethoxysilane and then linked a hyperbranched aromatic polyy- mide to improve the interaction between h-BN and epoxy matrix.

Several researches have been reported that suitable combinations of different fillers could significantly enhance the thermal conductivity of the composites compared to the individual filler. As the high in-plane strength, electrical and thermal conductivity, carbon-based materials (such as carbon nanotube or fiber and graphene sheet or foam) have become the priority [18–21]. Yu et al. [22] reported that the combination of single-walled carbon nanotubes (SWCNT, 2.5 wt%) and graphite nanolaminate (GNPs, 7.5 wt %), which improved the thermal conductivity from 0.2 to 1.75 W/m K in epoxy matrix. Considering the electrical insulation which avoid the short circuits when applied as thermal interface materi- als (TIMs) and the phonon energy damped by the substrate or matrix, which decreased the thermal conductivity of composites. The combination or hybridization of BN and graphene had attracted more attention [23]. For example, Cui et al. [13] reported a synergistic effect of few layered BN (s-BN) with few layered graphene (s-GH) nanosheets in the polystyrene (PS) and polyamide (PA) matrix, respectively. Small loading of s-BN enhanced the thermal conductive and mechanical properties of polymer composites due to the filling into the interspaces of s-GH, the stacked structure of s-BN/s-GH formed, and the flat surface of small s-BN. Tsai et al. [24] got a thermal conductivity ~2.21 W/m K of polyamide film with 50 wt% functionalized BN and 1 wt% glycyldimethacrylate-grafted graphene (g-TrG) composite filler. The graphene nanolayers filled into the voids between the BN filler and polymer matrix and thus led to the high thermal conductivity of the composites. Shtein et al. [25], and Liem et al. [26] also reported the similar effect of graphene with BN on the thermal conductivity enhancement in epoxy matrix.

Two-dimensional materials could be assembled into heterostructures that the monolayers were combined together by van der Waals forces [27]. Considering that a plethora of 2D mate- rials was available, creating a substantial variety of heterostruc- tures became possible. A large variety of novel experiments and prototypes such as mechanically-assembled stacks, (chemical vapor deposition) CVD, and physical epitaxy had already been car- ried out for van der Waals heterostructures. Currently, the most universal technique for heterostructures assembly was self-assembly in liquid [28]. Graphene was known to have usually high intrinsic thermal conductivity, which could arrived at 4840–

2. Experimental

2.1. Materials

The 2-Ethylhexyl acrylate (2EHA) (Aladdin Reagents (Shanghai, China)) and acrylic acid (AA) (Sinopharm Chemical Reagent (Beijing, China)) were purified via column chromatography over Al2O3. Sodium dodecyl sulfate (SDS) and octylphenylolethylene glycol (Triton X-100) (Aladdin Reagents (Shanghai, China)) were used as emulsifier. 2,2’-Azobis (2-methylpropionitrile (AIBN) (Sinopharm Chemical Reagent (Beijing, China)) was used as initia- tor. Diethyleneetriamine (DETA) (Aladdin Reagents (Shanghai, China)). Graphite was purchased from Qingdao Huatai Lubricant Sealing S&T Co., Ltd. Hexagonal BN powder (purity >99.5%) was provided by Dandong Rijin Science and Technology Co., Ltd. For all experiments, deionized (DI) water was used.

2.1.1. Preparation and functionalization of graphene oxide (GO/f-G)

GO aqueous solution was prepared by the modified Hummers’ method [37]. Firstly, 75 mL concentrated sulfuric acid charged into a 250 mL beaker equipped with a mechanical stirrer (Teflon impel- ler) with ice bath. Secondly, several grams of graphite were gradu- ally added under stirring to make a suspension. Then, 3 g potassium permanganate was added slowly to keep the mixture temperature under 10 °C. After 2 h, elevated the temperature to 35 °C and kept for 24 h. The mixture was subsequently diluted with deionized water. Then, the temperature was elevated to 80 °C and kept for 2 h. 30 wt% H2O2 was added slowly into the mix- ture. The suspension was centrifuged and washed by HCl solution and deionized water until the pH of the GO aqueous solution reached ~6.

According to our previous work [38]. Here, we reduced and functionalized GO by diethyleneetriamine (DETA) a weak base. 10 wt% of DETA was added into GO aqueous solution under vigorous stirring with reflux for 24 h at 80 °C. Finally, the functionalized graphene was code as E-G.

2.1.2. Preparation of surface negatively charged P2EHA latex via minimulsion polymerization

Minimulsion polymerization was used to prepare negatively charged P2EHA latex according to our previous work [39]. SDS/Triton X-100 compound emulsifiers were dissolved in water to prepare the aqueous phase. The monomers (2-ethylhexyl acrylate and acrylic acid) and initiator (AIBN) were mixed to prepare the oil phase. The aqueous phase and the oil phase were added to the glass reactor equipped with a N2 inlet and a tetrafluoroethylene impeller rotating at 1500 rpm for 30 min. The theoretical solid con-
tent of the miniemulsion was about 45 wt%. The continuous miniemulsion polymerization was carried out at a 250 mL glass reactor fitted with a reflux condenser, a N₂ inlet and a feeding inlet and the tetrafluoroethylene impeller rotating at 200 rpm. The reaction was carried out at 70 °C for 3 h, protected from light during the reaction. Finally, the latex was cooled to room temperature and filtered through a 30 mesh strainer to remove the emulsion particle agglomerated.

2.1.3. Preparation of the 2D stacked van der Waals heterostructures of h-BN/f-G

The h-BN (1 g) dispersed in 20 g ethanol was sonicated vigorously for 15 min to help peel away h-BN microplatelets. The functionalized graphene (f-G, 10 wt%, 20 wt%, and 30 wt%) was dropped in with the assistance of ultrasonic dispersion within 10 min and then stirring for 24 h at room temperature. These 2D stacked fillers was then dried in 50 °C for 24 h and 70 °C for 48 h, which were coded as 10%-f-G@h-BN (or f-G@h-BN), 20%-f-G@h-BN, and 30%-f-G@h-BN.

2.1.4. Preparation of the P2EHA/h-BN/f-G composite film

All the polymer composites were prepared by colloidal blending and casting method (shown in Fig. 1 blue box). In typical procedure, designed amounts of f-G@h-BN was mixed in deionized (DI) water, forming suspension with the help of ultrasonic. Then, above suspension was dropped into the P2EHA latex and stirred for 24 h at room temperature. Subsequently, these coagulation was poured into the Teflon mold quickly. The obtained P2EHA/f-G@h-BN were labeled for 30 min. Subsequently, these coagulation was poured into the Teflon mold quickly. Then, above suspension was dropped into the P2EHA latex and stirred for 24 h at room temperature. These 2D stacked fillers was then dried in 50 °C for 24 h and 70 °C for 48 h, which were coded as 10%-f-G@h-BN (or f-G@h-BN), 20%-f-G@h-BN, and 30%-f-G@h-BN.

2.1.5. Characterization

X-ray film and powder diffraction (XRD) patterns were taken on a Philips X’ Pert Pro MPD X-ray diffractometer (40 kV, 40 mA) with Cu-Ka radiation (λ = 0.154 nm). Measurement were done in 2θ mode using a bracket sample holder in continuous mode range of 5–80° at a scan speed of 10°/min. X-ray photoelectron spectroscopy (American Thermo) was used to obtain the XPS spectra. Raman spectra of samples were measured using a Confocal Raman Microscopy (Renishaw inVia Reflex). SEM (scanning electron microscopy) and TEM (transmission electron microscope) were used to characterize the morphologies of the materials. Atomic force microscopic (AFM) measurements was performed using SPA300HV AFM. Thermal property was characterized via a TGA (Q5000 IR) from 50 to 700 °C at a heating rate of 10 °C/min under nitrogen condition. With high volume resistivity beyond 10⁶ Ω cm at room temperature with the size of the specimen is 50 × 25 × 1 mm³, the high resistivity meter model LK2679A was used to measure the electrical conductivity with alignment and perpendicular direction. Data from five measurements were averaged. The bulk conductivity was calculated with the following equation [40]:

\[ \sigma = \frac{L}{R S} \]  

where L, R, and S are the length, resistance, and cross-sectional area of the specimen, respectively. The adhesive property tests were performed with the tensile machine (CMT, SANS) used the C-clamp at the speed of 50 mm/min. All adhesive measurements were carried out under the standard condition (T = 23 °C and humidity = 55%). The average force required to peel away the tape was recorded. Through-plane diffusivity was measured by laser flash method on LFA457 (Netzsch). The in-plane diffusivity was performed on DXF-500 thermal analyzer (American TA). All thermal diffusivity measurements were performed at room temperature. Differential scanning calorimetry (DSC) was used to measure the specific heat capacity of the composite at room temperature. The thermal conductivity (k) was calculated by the following equation:

\[ k = \frac{D \times \rho \times C_p}{S} \]  

where k, D, ρ and Cp are the thermal conductivity [W/mK], thermal diffusivity (mm²/s), density (kg/m³), and specific heat capacity [J/kg K]. Constant temperature resistance (12V 60 W) was mounted on the transformer (220 V to 12 V 60 W) to simulate the electronic devices and potted by a 1.5 mm thick composite layer. The operating temperature of the resistances were monitored by CEM DT-980 thermal camera and the thermal images were captured.

3. Results and discussion

3.1. Preparation of the functionalized graphene (f-G)

One of the obstacles which limited to achieve the improvement of desired properties for composites was that the maximum filler level that the composites can hold to achieve the best outcome

![Fig. 1. Schematic illustration of the procedure to prepare the functionalized graphene (f-G) sheet (red box), Lewis acid-base (δ⁺ · δ⁻) interaction and π-π stacking with h-BN sheet, the electrostatic self-assembly with negatively charged p(2-ethylhexyl acrylate) latex and the self-aligned layer by layer (blue box). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
In general, functionalization of the filler is an effective method to improve the compatibility between filler and polymer matrix. At the same time, excellent compatibility is also beneficial to the performance of composite materials.

The functionalization of graphene oxide sheet (GOs) by DETA would have significant influence on the performance of the surface functional groups. Fig. 2(a) shown the smooth surface of GOs compared with the wrinkles shown in Fig. 2(b) on f-G surface. As a weak base, DETA reduced the oxygen groups on GO surface and destroyed the hexagonal carbon skeleton, which introduced the loss of long-range ordering [42,43]. The lateral dimension of GO sheet and h-BN were around 21.84 \( \mu \)m and 26.31 \( \mu \)m (SI Fig. S1).

Raman spectroscopy could also be used to investigate the transformation of nanostructure from graphite to GO and f-G sheets. All samples were put on glass slides in powder without solvent. Fig. 2(c) shown the Raman spectra of graphite, GO sheets, and f-G sheets. The D-band which represents the disorder of defect mode that stems from disruption of planar sheet configuration at edges. The G-band represented primary Raman active mode in graphite and provided a good presentation of sp\(^2\) bonded carbon. Generally, the ID/IG ratio was used to measure the amount of disorder. Higher value of ID/IG ratio meant more defect or disorder and more binding sites in structure. The ID/IG ratio of GO was higher than f-G due to the reduction which resulted in the increase of sp\(^2\) planar carbon atoms.

Fig. 2(d) shown the atomic force microscopy (AFM) image of GO. The thickness of GO sheet was 1.41 nm which belong to the multilayers graphene and has excellent thermal conductivity after reduction [44]. The average thickness of h-BN was around 0.40 \( \mu \)m (SI Fig. S2).

After functionalization by DETA, some XPS characteristic peaks of GO were greatly weakened, disappeared or shifted and suggested the removal of oxygen-containing groups. Such as C-N (sp\(^3\)) bond appeared at 286.2 eV, C=C (sp\(^2\)) bond at 284.6 eV and some oxidized carbon functional groups including C-OH (285.2 eV), C=O (287.5 eV), and –O–C=O (288.1 eV)). Above characterization confirmed the efficient functionalization and reduction of GO (Fig. 2(e)).

The TGA curves of graphite, GO, and f-G were shown in Fig. 2(f). Two weight loss stages were observed for GO sample. The first weight loss stage below 230 °C was due to the decomposition of surface carboxyl or hydroxyl groups on the GO sheet. Some oxygen-containing functional groups remain on f-G that around 5 wt% weight loss of GO was obtained in the temperature range from 250 to 400 °C [45,46]. For the functionalized graphene (f-G), the weight loss below 230 °C was not obvious. However, the weight loss arranged from 250 to 400 °C was revealed obviously due to the decomposition of physically bonded amines (boiling temperature of DETA was 207 °C) which indicated the successful grafting of DETA molecule on graphene sheet [47].

3.2. The self-assembly of 2D van der Waals heterostructures between f-G and h-BN

Fig. 3(a) shown the smooth surface of h-BN compared with functionalized graphene (f-G) with the wrinkles on its surface shown in Fig. 3(b). As we know that graphene has excellent mechanical properties, low filler density (=1.2 g/m\(^2\)), great thermal properties (thermal conductivity = ~5000 W m\(^{-1}\)K\(^{-1}\)). Unfortunately, graphene-based fillers are electrically conductive (>10\(^7\) S/cm) [48]. If the loading of the filler exceeds the percolation threshold, it also significantly increases the electrical conductivity of the composite materials. Due to the atomically smooth surface and strong bonding of h-BN, it had been found to be an important 2D...
material and dielectric gate substrate for graphene, which led to weak electronic interlayer coupling between graphene and h-BN sheets, and thus preserved the electronic and optical properties intrinsic to graphene [49,50]. In this part of work, we proposed a simulation schematic for the van der Waals heterostructures as shown in Fig. 3(c), (g), and (h). Because of the repulsive force from the group (such as amino group) on f-G surface, the stacking of f-G displayed some wrinkles (Fig. 3(b)) compared to the h-BN with smooth surface (Fig. 3(a)). With the help of π–π stacking and Lewis acid-base interaction (B (d^-)–N (d^-)), f-G sheets would flat on the surface of h-BN as low ratio of f-G and h-BN. When the ratio of f-G and h-BN was high (2:10 or 3:10), the f-G would stacked between each other on the h-BN surface, therefore, we could find wrinkles in Fig. 3(e) and (f) and the conduction structure would form (Fig. 3(g)). However for the low ratio of f-G and h-BN (1:10), the insulation structure could be obtained (Fig. 3(h)). SEM images presented in Fig. 3(d) shown that f-G stacked on the surface of h-BN without contacting due to the similar hexagonal structure. The heat flow could be delivered by h-BN and would transfer between f-G through h-BN as the bridge via phono vibration. At the same time, the electronic would not pass through this channel [28]. Therefore, the 2D stacked filler of 10 wt%-f-G@h-BN has the ideal structure which would be chosen as the appropriate filler in polymer matrix.

The surface assembly were further confirmed by Raman spectra (Fig. 4(b)). Raman shift at 1367 cm\(^{-1}\) was the characteristic peak of h-BN ascribed to the high frequency intra layer E2 g vibration mode. A new peak at 1351 cm\(^{-1}\) and 1601 cm\(^{-1}\) appeared after functionalization, which was ascribed to G-band from f-G.

3.3. The film formation of P2EHA/f-G@h-BN composites film

Filler dispersion is the most common graphene precursor for the preparation of polymer composites. According to our previous work [38], because of the functional group on NH\(_2\)-rGO (f-G) sheet surface was amino group which was hydrophilic with positively charged. Modification of GO sheets increase the compatibility with P2EHA latex particles (negatively charged) through electrostatic interaction in the aqueous solution. After aqueous casting method, a remarkably improvement of properties was achieved in P2EHA/NH\(_2\)-rGO (f-G) system. Therefore, in this study, the f-G@h-BN dispersion was mixed with P2EHA emulsion to obtain a homogeneous colloidal dispersion of f-G@h-BN platelets and P2EHA latex particles.

Recently, many efforts have been focused on developing facile methods for preparing aligned graphene sheets in solvents and polymer with highly anisotropic characteristics. Therefore, to investigate the mechanism for formation of the stacked structure is necessary. The aspect ratios and filler content are the core factors to decide the stacked structure of the filler in the polymer matrix [51,52]. We have investigated that how the filler content have influence on the stacked structure of the P2EHA/NH\(_2\)-rGO (f-G) system in our previous work [38]. We used the filler with high aspect ratio which was beneficial for the self-alignment process. When the filler content was higher than 1 wt%, the NH\(_2\)-rGO (f-G) sheets would stacked with aligned structure. Due to the similar aspect
ratio and higher filler loading in this work, we believed that the filler would stack in the horizontal direction in this work with further characterization.

**Fig. 5** presented the SEM images of the surface and cross section for the composites. The 2D stacked filler of f-G@h-BN remained platelet-like shape after hard h-BN platelet coated by the flexible and thin f-G sheets. They are uniformly dispersed in the polymer matrix along the horizontal direction, which proved the effectiveness of aligned stacking with the assistance of the electrostatic interaction without the organic solvent. The latex particles or the polymer chains would result in the infiltration of the f-G@h-BN platelets. At high loading fractions, such as in P2EHA/20%-f-G@h-BN and P2EHA/30%-f-G@h-BN (shown in **Fig. 5**), the composite might contain some voids due to the limited maximum loading level, however, the effective continuous thermally conductive pathway could be considered to be formed as individual f-G@h-BN platelet overlapped with each other, which was important for heat conduction [41].

![Fig. 4](image-url)  
*Fig. 4.* (a) XRD patterns of h-BN, f-G, 10%-f-G@h-BN, 20%-f-G@h-BN and 30%-f-G@h-BN; (b) Raman spectra of h-BN and f-G@h-BN.

![Fig. 5](image-url)  
*Fig. 5.* The surface section and cross section SEM images of the composite and the yellow arrow for functionalized graphene; the blue arrow for h-BN. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
3.4. The adhesive properties of the composite film

Thermal interface adhesive (TIA) were polymer-based composites which improved contacts between the mating surfaces, offered good thermal conductivity of electronic packaging [53]. According to the actual characteristics of the composite, we made a correction to the common testing of the hold tack of the adhesive film. Tensile machine and C-clamp was used to carry out this testing. Fig. 6 shown the adhesive properties of the composite film. As we know, the filler have aspect ratio and self-alignment in polymer matrix. Obviously, the loading of 2D stacked filler decreased the maximum elongation due to the addition of rigid filler. Interesting, the change of yield stress and yield position had a V-shape. Small loading of filler would break the polymer matrix due to the high aspect ratio and less contact between each filler. However, high loading of filler enhanced the polymer matrix via the mutual restriction between each fillers.

3.5. Electrical conductivity of P2EHA/f-G@h-BN composite film

Fig. 7 shown the direct current electrical conductivity of the composites (σ) as a function of 2D stacked f-G@h-BN filler content. Compared to our previous work with the similar aligned structure and loading, the electric conductivity would arrive at 18 S/cm when the loading of graphene was 1.0 wt% [38]. However, the composite filled with hybrid f-G@h-BN in this work provided a high insulation property whether in the in-plane or through-plane direction, reflecting the rationality of the 2D stacked filler’s structural design shown in Fig. 3. When the loading of f-G@h-BN filler was 30 wt% (the content of graphene was around 3 wt%), the electrical conductivity of the composites would increase significantly and arrived at 9.09E–8 S/cm due to the overlapping of f-G@h-BN filler which the consecutive conductive pathway was originated from contact of graphene (f-G) sheets in the in-plane direction in the polymer matrix. However, the composite was still insulator [41,54].

3.6. Thermal conductivity of P2EHA/f-G@h-BN composite film and estimation with model

The aim of this paper was to develop a kind of 2D stacked heterostructures filler which could endow the polymer matrix with...
high thermal conductivity, high adhesive properties and insulation. Fig. 8 depicted the thermal conductivity of P2EHA/f-G@h-BN composites on in-plane and through-plane direction.

It was well known that transport of heat in nonmetals drives by phonon or lattice vibrations [55]. Compared to 1D filler such as CNTs, 2D filler (h-BN and graphene) probably provided larger area of interface contact than 1D filler due to the extended dimensionality and large contact area, which tend to decrease interfacial resistance between the fillers, also due to their much higher sound velocity and optical phonon energy [36,56].

The results of in-plane thermal conductivity ranged from 1.55 W/m K to 1.96 W/m K and 0.16 W/m K to 0.38 W/m K for through-plane thermal conductivity dependent on the loading fraction of 2D stacked filler. The thermal conductivity of the composite at ~10 wt% loading reached a value of ≈1.55 W/m K compared with currently available thermal interface materials (TIMs) which required about 5–10 times of the filler to achieve comparable thermal conductivities [57]. The reformed f-G@h-BN thermal conductive networks helped to enhance the thermal conductivity via contacting with each other (Fig. 8). The reason for thermal conductivity enhancement in the through-plane direction ascribed to the aligned stacked structure formed by the 2D stacked f-G@h-BN fillers in polymer matrix as shown in SEM cross section (Fig. 8). Low loading of filler enhanced the thermal conductivity slightly due to the polymer matrix separated the adjacent 2D stacked fillers. Therefore, when the dosage of 2D stacked filler increased, the less space between adjacent 2D stacked fillers was, the more contacting between them would form and improved the thermal conductivity in the through-plane direction.

Obviously, the integration of f-G@h-BN not only improved the thermal conductivity, but also diminish the increment of electrical conductivity significantly at the same time. Therefore, f-G@h-BN which combined with f-G and BN could effectively decouple the thermal/electrical conductivity.

3.7. Thermography of P2EHA/f-G@h-BN composite film

Fig. 9(a) shown the thermographic images of the composites. The thermographic images were acquired from the IR camera by captured the thermal images of the samples placed on a constant temperature resistance. The surface temperature was set at 60 °C. The heat conduction were considered and the surface temperature changes of the sample were measured as a function of elapsed time. (Fig. 9)
3.8. Thermal decomposition of P2EHA/f-G@h-BN composite film

Fig. 10 presented the thermal decomposition of the P2EHA/f-G@h-BN composite film via thermo-gravimetric analyses under nitrogen condition. The loading of 2D stacked f-G@h-BN filler improved the thermal stability of the composite when we compared the decomposition temperature at 50% weight loss due to the closed packing between 2D stacked filler and polymer matrix could be observed in SEM cross section images (Fig. 5). Interestingly, when the content of filler was >20 wt%, the initial decomposition temperature’s decreasing was non-negligible which could imply that higher content of the thermal conductivity filler accelerated the decomposition of the polymer [58]. For the decomposition of the f-G shown in Fig. 2(f), the residual decomposition of the composites was lower than the 2D stacked filler content we added.

4. Conclusion

In conclusion, we observed an extremely enhanced thermal conductivity in the P2EHA/f-G@h-BN composites. The h-BN platelets were connected by graphene sheets as the crosslinking agent which effectively reduced the amount of filler and obtained the best thermal conductivity \( \sim 1.96 \text{ W/m K} \) with excellent adhesive properties. And the special 2D stacked structure of the filler f-G@h-BN reduced the electrical conductivity for the composites with enhanced thermal stability and adhesive property. The latex blending and self-assembly method was carried out to achieve the dispersion of 2D stacked filler in polymer matrix. This functionality and fabrication methods were facile operating, environmentally friendly, and suitable for large scale production. Obtained polymer composite materials also have good insulation that could be applied in connectors, thermal interface materials (TIMs), and other high-performance thermal management systems.

Notes

The authors declare no competing financial interest.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.composites.a.2017.01.020.

References


