Research Article

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Preparation and characterization of a graphene hybridizing polyurethane damping composite

Abstract: A graphene hybridizing polyurethane/polyethyl methacrylate (GR-PU/PEMA) damping composite was synthesized using the sequential interpenetration method. The effects of the graphene content and the microphase separation structure on the damping properties, thermal stability, and mechanical properties have been studied in detail. The dynamic mechanical analysis indicated that graphene could improve the damping peak value of PU/PEMA, and the microphase separation structure could be beneficial for broadening the damping temperature range. The damping peak (tan $\sigma_{\text{max}}$) of PU/PEMA hybridizing with 0.5 wt% graphene reached 0.82, and the temperature range of the loss factor (tan $\sigma \geq 0.3$) was expanded to 88.3°C. Analysis of scanning electron microscopy, transmission electron microscopy, and small-angle X-ray scattering reveals that graphene is uniformly dispersed in the polymer matrix, and the composite with interpenetrating polymer network (IPN) shows more microphase separation structures. Fourier transform infrared analysis indicated that there is strong interaction between graphene and IPN matrix. Furthermore, the addition of graphene improved the mechanical properties and thermal stability of composites.

Keywords: polyurethane, graphene, IPN, damping properties, microphase separation

1 Introduction

With the rapid development of industry, the problems of noise and vibration pollution have become increasingly severe, which has a negative impact on human health, industrial production, and environmental security. Various damping systems [1,2] have been developed to reduce the harm caused by the vibrations.

In the damping system, the damping materials play crucial roles. A perfect damping material not only has a higher damping peak but also a broader damping temperature range (tan $\sigma_{\text{max}} \geq 0.3$). Various preparation methods, such as polymer blending, copolymerization, and interpenetrating polymer networks (IPNs) [3,4], have been used to develop damping materials. Among them, IPN is a polymer alloy consisting of two or more polymers in a network form, entangling together by occasional covalent bonds [5,6]. IPNs have received extensive attention in recent years, because of their relatively high mechanical properties and excellent damping properties around the glass transition temperature, such as polyurethane/vinyl ester resin [7], polyurethane/epoxy [8], polyurethane/polyactic acid [9], etc. Chen et al. [10] investigated a series of polyurethane/polyacrylate IPNs, and discovered that the maximum damping factor (tan $\delta$) increased from 0.41 to 0.78 and the IPN with longer side monomer had better damping properties. Jin et al. [11] studied the synchronous interpenetration network of polyurethane (PU)/polyethylene dimethacrylate with a damping factor of 0.8. It can be seen that the structure of IPN plays a very important role in improving the damping properties of materials.

Besides the above methods, the interaction and friction between inorganic particles and polymers can also be used to improve the damping properties of materials [12]. In recent years, the addition of inorganic particles to
polymers has been recognized as an effective way to improve composite properties. Chen et al. [13] studied multi-walled carbon nanotubes (CNTs) reinforced the castor oil-based PU/EP graft IPN composites, of which the damping temperature range was 53.1–102.2°C, and the corresponding peak value of tan δ reached 1.04 when the content of CNTs reached 0.1 wt%. Kim et al. [14] synthesized PU/graphene foams. The results suggested that graphene was an effective additive for the formation of PU foams, able to increase the acoustic damping properties. The outstanding damping properties were attributed to the large specific surface area and Young’s modulus of graphene sheets [15–17]. In the past few years, the graphene has been incorporated into the various polymer matrices mainly in order to enhance its thermal or mechanical properties, while few studies have been reported on the influence of graphene on the damping properties of IPN composites.

In this study, a novel kind of graphene hybridizing polyurethane/polyethyl methacrylate (GR-PU/PEMA) damping composite was synthesized by the sequential interpenetration method. The damping properties of this damping composite were studied. In addition, the effects of the graphene content and the microphase separation structure on the damping properties, thermal stability, and mechanical properties were also studied by dynamic mechanical analysis (DMA), small-angle X-ray scattering analysis (SAXS), scanning electron microscope (SEM), thermogravimetric analysis (TGA), tensile test, etc. This work aims to provide a reference for developing high-performance damping composites.

### 2 Experimental program

#### 2.1 Materials

Polytetrahydrofuran (PTMG, $M_n = 1,000$) was obtained from Zibo Defu Chemical Co., Ltd. 2,4-Disocyanatotoluene (TDI), 3-aminopropyltriethoxysilane (KH-550, coupling agent), and 3,3′-dichloro-4,4′-diaminodiphenyl methane (MOCA, chain extension agent) were supplied by Sinopharm Group Chemical Reagent Shenyang Co., Ltd. Ethyl methacrylate (EMA), azobisisobutyronitrile (AIBN, initiator), dibutyltin dilaurate (T-12, accelerator), and sodium hydroxide (NaOH) were purchased from Tianjin Kemiou Chemical Reagent Co., Ltd. Graphene (purity: 98%) was purchased from Nanjing Xianfeng Nano Material Technology Co., Ltd. Triazine hexabasic quaternary ammonium chloride (TH$_2$QC, dispersant) was prepared by the method described in our previous paper [18]. TDI and PTMG were dehydrated at $85^\circ$C for 24 h before use.

#### 2.2 Preparation of GR-PU/PEMA composite

##### 2.2.1 Pretreatment of graphene

Graphene was dissolved in sufficient excess of aqueous solution containing TH$_2$QC and KH-550. In order to obtain better dispersion effect, the weight ratio of TH$_2$QC to KH-550 was controlled at 1:2, and the total content of TH$_2$QC and KH-550 accounted for 10.0 wt% of graphene. The mixed solution was stirred for 30 min at room temperature, then the graphene dispersion solution was freeze-dried at $-50^\circ$C for 48 h. The modified graphene was obtained by sublimating the water [19].

##### 2.2.2 Preparation of PU pre-polymer

The pre-polymer of isocyanate-terminated PU was prepared with 1.5 equivalents of TDI and 1 equivalent of PTMG at $80^\circ$C. The reaction was carried out until the theoretical isocyanate (–NCO) content determined by di-n-butyl amine titration reached. When the percentage of –NCO reached the theoretical value, the mixture was rapidly lowered to the room temperature to terminate the reaction and degassed under vacuum.

##### 2.2.3 Preparation of GR-PU/PEMA IPN composite

The pre-polymer PEMA was prepared by the following procedure: 100 g distilled EMA monomer and 0.5 wt% AIBN were added in a four-necked bottle equipped with a dry nitrogen inlet, stirrer, thermometer, and reflux condenser, and the reactor was heated to $80^\circ$C. The mixture was stirred until AIBN was dissolved completely. The reaction progress was monitored by adding a drop of the mixture in isopropyl alcohol to precipitate solid PEMA. The reaction was terminated by rapid cooling to room temperature when the conversion reached 10–15 wt%, and the pre-polymer PEMA was obtained [20,21]. After
that, the different contents of modified graphene sheets were added into the pre-polymer PEMA and dispersed by ultrasonic irradiation (70 kHz) for 30 min. Then the pre-polymer PEMA containing the modified graphene sheets was mixed with PU pre-polymer (the weight ratio PU/PEMA was 7:3). A small amount of T-12 and 0.5 equivalent of MOCA were added into the mixture and reacted at 60°C for 30 min to prepare the IPN composite. Finally, the reaction products were poured into Teflon mold and cured in a vacuum oven at 80°C for 36 h. Figure 1 shows the synthesis process of the modified GR-PU/PEMA damping composite.

2.3 Characterization

The Fourier transform infrared (FT-IR) spectrum analysis was performed using a Nicolet IS 10 FT-IR spectrophotometer (ThermoFisher, USA). The FT-IR spectra were acquired by scanning the specimens in the wavenumber range from 4,000 to 500 cm\(^{-1}\). The test adopted the attenuated total reflection mode with 16 scans.

TGA tests were carried out over the temperature range from 40°C to 600°C with a DTG-60 H thermogravimetric analysis instrument (Shimadzu, Japan) under nitrogen atmosphere, and the heating rate was 10°C min\(^{-1}\). The temperature at which the weight loss reached 3% was defined as the onset decomposition temperature.

DMA data were obtained using Diamond DMA (Perkin Elmer, USA) in a dual cantilever bending mode, over a temperature range from −75 to 150°C with a heating rate of 3°C min\(^{-1}\) at 1 Hz. The samples were rectangular bars (2.0 cm × 1.0 cm × 0.2 cm).

SAXS analysis was performed at the 1W2A small-angle scattering experimental station of the Beijing Synchrotron Radiation Facility (BSRF, Laboratory of the Beijing Institute of High Energy Physics). The X-ray wavelength is 0.154 nm, the detector used is Mar165 CCD, the pixel size is 79.1 μm, the spot size is 2,048 × 2,048 pixels, the exposure time is both 10 s, and the scanning range is 0–10°.

Transmission electron microscope (TEM) observations were carried out using a JEM-1230 electron microscope (JEOL, Japan) with an acceleration voltage of 90 kV. EM UC7 ultra-thin microtome (Leica, Germany) was used to cut the sample into ultra-thin sections with a thickness of about 100 nm, and PU/PEMA specimens were stained with 2% OsO4 vapor for 48 h.

SEM examinations were performed using an S-3400 microscope (HITACHI, Japan) on liquid nitrogen fractured surfaces of the composites coated with a gold layer. The tensile properties were obtained with H10K-S universal testing machine (Hounsfield, England). The dumb-bell specimens (115 mm × 6 mm × 2 mm) were tested at a crosshead speed of 500 mm min\(^{-1}\) according to the Chinese national standard GB/T 528-2009. The tensile strength and elongation at break were reported as

![Figure 1: Schematic diagram for the synthesis of GR-PU/PEMA IPN damping composite.](image-url)
average values of five specimens. Hardness testing was carried out using a TH200 shore A hardness tester (Beijing TIME Co., Ltd, China), and the average of values from five measurements was used.

3 Results and discussion

3.1 FT-IR analysis

Figure 2 shows the FT-IR spectra of graphene (GR) and modified graphene (M-GR). As seen from Figure 2, compared with that of GR, the FT-IR spectrum of M-GR has two new characteristic peaks at 1,110 and 1,028 cm\(^{-1}\) corresponding to the stretching vibration of C–N and the bending vibration of Si–O–Si, respectively. The hydrogen bond peak at around 3,380 cm\(^{-1}\) in the FT-IR spectra of M-GR becomes significantly broader and more intense. The appearance of new absorption peaks and the change of hydrogen absorption peak indicate that there is a strong interaction between GR and the modifier, and GR was successfully modified by KH550 and TH\(_{12}\)QC.

FT-IR spectra of PU, PU/PEMA, and GR-PU/PEMA are shown in Figure 3. It is clearly observed from Figure 3 that the characteristic peaks of the isocyanate group (–NCO) at 2,250–2,280 cm\(^{-1}\) disappear in all specimen spectra, indicating that the reaction between –NCO groups of TDI has reacted completely with –OH groups of PTMG. The characteristic absorption peak at 3,295 and 1,725 cm\(^{-1}\) corresponds to the stretching vibration of N–H and C=O in the carbamate. In the FT-IR spectrum of PU/PEMA, the absorption peaks at 1,270 and 1,176 cm\(^{-1}\) can be ascribed to the symmetric and asymmetric stretching vibration of C–O in PEMA. The absorption peaks at 2,946 and 2,861 cm\(^{-1}\) are the stretching vibration peaks of C–H in methyl and methylene. Furthermore, with the addition of M-GR, a strong stretching vibration peak of the Si–O–Si bond appears at 1,025 cm\(^{-1}\) in the FT-IR spectrum of GR-PU/PEMA, which can be ascribed to the modification by KH550. The absorption band around 3,300 cm\(^{-1}\) becomes broader, indicating that there exists an enhanced interaction of carbamate and M-GR [22].

3.2 Morphologies and microphase structure analysis

The morphology of PU/PEMA and GR-PU/PEMA composite was observed by TEM. Figure 4(a) and (b) shows the TEM micrographs of PU/PEMA matrix, and Figure 4(c) and (d) are TEM micrographs of 0.1 wt% GR-PU/PEMA composite. Because the samples were stained by OsO\(_4\), the dark areas on the micrograph represent PU phase. It can be seen from Figure 4(a) and (b) that a lot of bright sphere-like PEMA disperse in the phase of PU. As shown in Figure 4(e), this sphere-like structure is just a two-dimensional representation of a certain section of the IPN structure. Similar morphology of IPN was also observed by Trakulsujaritchok and Hourston and Chen et al. [9,10]. Moreover, the boundaries of the two phases diffuse, which indicate that the PU phase and PEMA phase are closely integrated by the IPN structure. The properties of the composite depend largely on the dispersion of graphene in the
polymer matrix. TEM was used to reveal the dispersion of graphene in the polymer matrix. As shown in Figure 4(c) and (d), it can be observed that the length of graphene sheet is about 0.5 μm and M-GR is evenly embedded in the polymer matrix without obvious agglomeration, illustrating the strong interfacial interaction and excellent compatibility between M-GR and polymer matrix. In addition, it can be seen from Figure 4(c) and (d) that the graphene sheets possess wrinkled surfaces, which can enhance the adhesion of nanofiller and matrix through the interlocking effect.

The fractured surface morphologies of PU, PU/PEMA IPN, and GR-PU/PEMA were also analyzed by SEM, as shown in Figure 5. It can be seen from Figure 5(a) that pure PU has characteristic toughness features with smooth fractured surfaces. With the addition of PEMA, the
fractured surfaces of PU/PEMA IPN become rougher and more uneven (Figure 5(b)). Meanwhile, partial phase separation appears in SEM micrographs. Combined with the results of TEM, SEM micrographs provide more direct evidence for the formation of the IPN structure.

The phase morphology of composites with 0.5 wt% graphene hybridizing IPN composite is shown in Figure 5(c). Graphene sheets are well-dispersed in the PU/PEMA matrix at this content. It can also be observed that the composites possess rough fractured surfaces, and most of the graphene sheets are coated with polymer matrix, which can be attributed to strong interfacial adhesion and good compatibility between the polymer matrix and graphene. However, when the content of graphene reached 1.0 wt%, a small amount of graphene aggregated can be observed (Figure 5(d)), which is not beneficial to the mechanical and damping properties.

SAXS technology was used to analyze the aggregation structure of PU/PEMA and GR-PU/PEMA. The SAXS curve reflects the scatter information of the regular structure [23]. The peak position in the SAXS curve is related to the length of the regular structure in the composite. The SAXS scattering patterns and Lorentz-corrected curve of PU/PEMA and GR-PU/PEMA are shown in Figure 6. As shown in SAXS scattering patterns, the scattering pattern of GR-PU/PEMA exhibits great differences with the increase in graphene content. In order to show this change more clearly and intuitively, the curve of q vs iq² can be obtained by performing absorption correction, subtracting air scattering, and using Lorentz correction. The length of long period d was calculated by equations (1)–(3).

From the Prague formula:

\[ \lambda = 2d \sin \theta, \]  
\[ Q = 4\pi \sin \theta / \lambda. \]

The long period was calculated by the following formula:

\[ d = 2\pi / q_{\text{max}}. \]

In these formulae, \( \lambda \) is the incident light wavelength (0.154 nm), \( \theta \) is the diffracted angle, \( Q \) is the scattering vector, \( q_{\text{max}} \) is the scattering vector corresponding to the peak in the scattering curve, and \( d \) is the length of long period.

It can be observed from Figure 6 and Table 1 that all the curves have \( q_2 \) peaks. However, not all the Lorentz correction curves of the samples have \( q_1 \) peak, and only the PU/PEMA IPNs filled with M-GR have the peak of \( q_1 \). It can be preliminarily inferred that the \( q_2 \) peak is related to the polymer matrix, and the \( q_1 \) peak is caused by the new periodic structure due to the addition of graphene. This new periodic structure may be caused by the epiphytic crystallization of the polymer matrix on the surface of graphene [24]. As shown in Figure 6, only when the content of graphene reaches or exceeds 0.5 wt%, can the peak of \( q_1 \) be observed, indicating that epiphytic crystallization is affected by the content of graphene. The Bragg equation was further used to calculate the long period of the periodic structure. For 1.0 wt% GR-PU/PEMA IPN composite, the calculation results are \( d_1 = 62.8 \) nm \( (q_1 = 0.10 \text{ nm}^{-1}) \) and \( d_2 = 10.30 \) nm \( (q_2 = 0.61 \text{ nm}^{-1}) \), in which \( d_1 \) and \( d_2 \), respectively, represent the long period of the periodic structure related to epiphytic crystals and

**Table 1:** Characteristic SAXS data of GR-PU/PEMA IPN composites

| Sample            | \( q_{1\text{max}} \) (nm\(^{-1}\)) | \( q_{2\text{max}} \) (nm\(^{-1}\)) | \( d_1 \) (nm) | \( d_2 \) (nm) |
|-------------------|---------------------------------|---------------------------------|----------------|----------------|
| Pure PU/PEMA      | —                               | —                              | —              | 10.13          |
| 0.1 wt% GR-PU/PEMA| —                               | 0.62                           | —              | 9.81           |
| 0.5 wt% GR-PU/PEMA| 0.13                            | 0.58                           | 48.3           | 10.83          |
| 1.0 wt% GR-PU/PEMA| 0.10                            | 0.61                           | 62.8           | 10.30          |
non-epiphytic crystals. The size of epiphytic crystals is much larger than that of non-epiphytic crystals. Generally, the longer the period structure of the crystal, the more complete the crystal structure, and the perfect epiphytic crystal structure makes polymer tightly adhere to the surface of graphene, which benefits to improve the mechanical properties of the composites [25, 26].

3.3 Damping property analysis

The storage modulus ($E'$) of GR-PU/PEMA composite as a function of temperature is shown in Figure 7(a). The storage modulus decreases as the temperature increases throughout the entire scanning temperature range for each sample. It can also be found from Figure 7(a) that the storage modulus increases significantly with the incorporation of graphene. When the content of graphene is 1.0 wt%, the storage modulus increases from 1.23 GPa (pure PU/PEMA) to 1.89 GPa at the initial temperature. The storage modulus can reveal the stiffness of the molecular chain. The changes of storage modulus, on one hand, are due to the polymer epiphytic crystals (as described in SAXS analysis), and on the other hand, are caused by the higher Young’s modulus of graphene.

Figure 7(b) shows the temperature dependence curve of the loss modulus ($E''$). When the content of graphene is less than or equal to 0.5 wt%, the peak value of the loss modulus increases with the increase of graphene content. The increase in loss modulus is mainly due to the friction between the graphene and PU/PEMA matrix and the

![Figure 7: Temperature dependence of (a) storage modulus, (b) loss modulus, and (c) tan $\delta$ for pure PU/PEMA, 0.1 wt% GR-PU/PEMA, 0.5 wt% GR-PU/PEMA, and 1.0 wt% GR-PU/PEMA.](image-url)
restriction effect of the graphene on the molecular chain motion, which will increase the energy consumed during molecular chain motion.

Figure 7(c) shows the loss factor (tan δ) curve of the GR-PU/PEMA IPN composite, and the damping data are shown in Table 2. It is noted from Figure 7(c) and Table 2 that the DMA curve exhibits only one loss factor peak when the content of graphene is 0.5 wt% and the maximum loss factor reaches 0.82. The damping properties of GR-PU/PEMA have great improvement compared to pure PU [27] or nano-organoclay modified PU/PEMA [28], in which the loss factors are 0.36 and 0.62, respectively. In addition, it can be found that the temperature at which tan δ peak occurs becomes higher with the increase of graphene content, indicating the movement of polymer chains becomes more difficult. It is noteworthy that when the content of graphene reaches 0.5 and 1.0 wt%, there is a “shoulder peak” in the DMA curve of the composite, indicating that an appropriate compatibility is formed among the components and the effective damping temperature range is broadened greatly. Due to the “synergistic effect” produced by IPN structure, the damping peaks in the high and low temperature regions increase at the same time, and the effective damping temperature range (tan δ ≥ 0.3) is, respectively, broadened to 88.3°C (−59.6 to 28.7°C) and 83.9°C (−58.9 to 25.0°C).

As shown in Figure 6, GR-PU/PEMA IPN composites have excellent damping properties. In fact, the damping properties of the material are evaluated by the rate of energy dissipation reflecting the degree of the internal friction between the components and the restriction degree of the molecular chain motion. The more intense the internal friction and restriction, the better the damping properties. The increase of graphene content reduces the free volume of the composite and increases the system viscosity. The friction between graphene and the friction between the polymer matrix and graphene all increase the energy dissipation rate of the composites. Although the introduction of graphene as the reinforcing particle can enhance the rigidity of the material, the addition of graphene can greatly increase the loss modulus of the material. Meanwhile, the loss factor is the ratio of the loss modulus to the storage modulus. Therefore, when the graphene content is less than or equal to 0.5%, the loss factor increases significantly, and the damping properties are significantly enhanced. However, when the content of graphene reaches 1.0 wt%, the improvement effect of graphene on the storage modulus is more significant than that of the loss modulus, so the peak of the loss factor shows a slight downward trend.

### 3.4 Mechanical properties

Figure 8(a) shows the tensile strength of the composite is proportional to the graphene content, and the elongation at break is inversely proportional to the graphene content. When the graphene content reaches 1.0 wt%, the tensile strength of the composite reaches the optimal value of 15.9 ± 0.4 MPa, and the tensile strength increases by 51.1% compared to pure PU/PEMA. The results may be attributed to the well-dispersed graphene in the polymer matrix, as seen from the SEM analysis and epiphytic crystals mentioned in the SAXS scattering analysis. Graphene has the inherent high mechanical strength and specific surface area. In the process of tensile fracture, the generation and propagation of cracks play an important role [29]. Well-dispersed graphene can more effectively resist the propagation of cracks. At the same time, epiphytic crystals enhance the interaction between the graphene and the IPN matrix. As a result, with the increase of graphene content, the tensile strength of composite also increases. However, this increase of tensile strength cannot provide large deformation, so the elongation at break decreases with the increase of graphene content. As with the hardness, as the content of graphene increases, the shore hardness of the composite continues to increase (Figure 8(b)). And the hardness value reaches 80.5 ± 1.3 when the content of graphene is 1.0 wt%. The results revealed that the addition of M-GR can effectively increase the tensile strength and hardness of the material.

| Sample                 | Temperature range (°C) of tan δ ≥ 0.3 | tan δ (max) | Temperature (°C) of tan δ (max) |
|------------------------|---------------------------------------|-------------|----------------------------------|
| Pure PU/PEMA           | −61.7 to 1.9                          | 0.68        | −39.0                            |
| 0.1 wt% GR-PU/PEMA     | −61.2 to −6.8                         | 0.72        | −36.1                            |
| 0.5 wt% GR-PU/PEMA     | −59.6 to 28.7                         | 0.82        | −34.5                            |
| 1.0 wt% GR-PU/PEMA     | −58.9 to 25.0                         | 0.77        | −26.7                            |
3.5 Thermal stability properties

TGA was used to evaluate the thermal stability properties of composites. The TGA and DTG curves of pure PU/PEMA and GR-PU/PEMA are shown in Figure 9, and the obtained data are listed in Table 3. It can be seen that GR-PU/PEMA presents higher thermal stability compared to pure PU/PEMA. Below 250°C, the addition of graphene has no obvious effect on the thermal decomposition process, and there is obviously no weight loss process. Then the pure PU/PEMA is decomposed quickly at 269.1°C. However, the onset decomposition temperature of 1.0 wt% presents higher thermal stability compared to pure PU/PEMA. Below 250°C, the addition of graphene has no obvious effect on the thermal decomposition process, and there is obviously no weight loss process. Then the pure PU/PEMA is decomposed quickly at 269.1°C. However, the onset decomposition temperature of 1.0 wt%

![Graph showing thermal stability properties](image)

**Table 3: Characteristic DTG data of GR-PU/PEMA IPN composites**

| Sample                | $T_{\text{onset}}$ (°C) | $T_{\text{20%}}$ (°C) | Residual weight at 600°C (%) |
|-----------------------|--------------------------|------------------------|----------------------------|
| Pure PU/PEMA          | 269.1                    | 334.8                  | 6.8                        |
| 0.1 wt% GR-PU/PEMA    | 273.0                    | 337.7                  | 5.9                        |
| 0.5 wt% GR-PU/PEMA    | 273.8                    | 340.3                  | 6.3                        |
| 1.0 wt% GR-PU/PEMA    | 279.7                    | 343.4                  | 6.6                        |

**Figure 8:** The effect of graphene content on (a) tensile properties and (b) hardness.

**Figure 9:** (a) TGA and (b) DTG curves for GR-PU/PEMA, 0.1 wt% GR-PU/PEMA, 0.5 wt% GR-PU/PEMA, and 1.0 wt% GR-PU/PEMA composites.
GR-PU/PEMA increases up to 279.7°C, more than 10°C higher than that of pure PU/PEMA. The enhancement of the thermal stability may be due to the formation of hydrogen bonds between hydroxyl groups on the graphene surface and carbonyl groups of PU. As shown in Figure 9(b), three thermal weight loss processes are in the tested temperature range. The first stage corresponds to the decomposition of PEMA. The second and the third stages of weight loss occurred in the range of 300.0–490.0°C, corresponding to the decomposition of PU soft segment and hard segment. Following the increase of graphene content, the two decomposition stages also move to high temperatures. Furthermore, it is demonstrated from TGA and DTG that incorporating graphene enhances the thermal stability of PU/PEMA composite.

4 Conclusion

This study demonstrated a feasible approach to prepare high damping composites based on IPN (PU/PMMA) structure and graphene. The influence of graphene on PU/PEMA IPN composite properties was investigated. DMA indicated the addition of graphene could effectively improve the damping peak of the composites, and the temperature range of effect damping was obviously broadened due to the existence of IPN structure. The maximum tan δ of the composite with 0.5 wt% graphene reached 0.82, and the damping properties were the best. The results of TGA and tensile tests showed that the thermal stability and tensile strength increased with the increase of graphene content. Compared with polymer matrix, the onset decomposition temperature of composites containing 1.0 wt% graphene increased by more than 10°C, and the tensile strength increased by 51.1% compared to pure PU/PEMA. SEM, TEM, and SAXS analyses revealed the graphene was evenly dispersed in the polymer matrix, the composites with IPN structure showed more microphase separation, and the periodic structure became longer due to the existence of epiphytic crystals. FT-IR analysis revealed a strong interaction between graphene and polymer matrix. The alteration of material microstructure caused by the addition of graphene improved the damping properties, mechanical properties, and thermal stability of the composites. It can be believed that the simple and efficient preparation method will display a vast potential in the manufacture of high-performance damping materials.

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