# Enhanced Toughness of Multilayer Graphene Filled Poly(vinyl chloride) Composites Prepared Using Melt-Mixing Method

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**In order to improve toughness of rigid poly(vinyl chloride) (PVC), we prepared multilayer graphene (MLG) filled PVC composites through conventional melt-mixing methods by taking advantages of easy dispersion and high flexibility of graphene. Microstructure, static, and dynamic mechanical properties of the MLG/PVC composites were investigated in details. We found that a small amount of MLG loadings (0.36 wt%) could greatly increase tensile fracture toughness and impact strength of the MLG/PVC composites, which is mainly attributed to high flexibility of the crumpled MLG throughout PVC matrix. Moreover, the presence of MLG can weaken intermolecular interactions and improve segmental motion of PVC chains, consequently resulting in low glass transition temperature and high toughness of the MLG/PVC composites. By virtue of its enhanced toughness and easy operation, the MLG/PVC composites show great potential to be used as high-performance composites in many fields. POLYM. COMPOS., 00:000–000, 2015. © 2015 Society of Plastics Engineers**

## INTRODUCTION

Poly (vinyl chloride) (PVC) has been widely used in many fields of building construction, packaging, transportation, and electric industry due to its good chemical stability, flame retardance, and low cost [1, 2]. The PVC materials are generally classified into two types, one is rigid PVC with high strength and stiffness, and the other is soft PVC in the presence of low-molecular-weight plasticizers which can greatly weaken interactions between PVC chains. Notably, the rigid PVC without addition of plasticizers has intrinsic disadvantages of low impact strength and low resistance to microcrack propagation, greatly hindering its practical application as high-performance structural materials [3]. In that case, improving toughness of rigid PVC has attracted much attention from industrial and academic fields, and it has been an important issue for developing high-performance PVC materials.

There are generally two methods of chemical and physical modifications for improving toughness of rigid PVC [1]. The chemical modification is to introduce flexible segments into PVC chains using chemically grafting copolymerization methods, but inevitably causing complicated processes and high cost. As a contrast, physical modification is to simply add some polymeric or inorganic modifier in a form of particles or fibers into molten PVC matrix during manufacture for obtaining modified PVC-based composites. This physical modification has been widely used in plastic industry by virtue of its easy operation, simple process, and high flexibility.

So far, some polymeric modifiers, such as chlorinated polyethylene (CPE), ethylene vinyl acetate copolymer (EVA), methylmetharylate-butadiene-styrene copolymer (MBS), acrylics (ACR), nitrile rubber, thermoplastic urethane (TPU), have been widely used in plastics industry for improving toughness of rigid PVC [4–7], but the addition of these polymeric modifiers inevitably results in a decrease in stiffness and heat resistance of the modified PVC. Recently, some inorganic nanoparticles, such as nano-$CaCO_3$ [8], montmorillonite [9], kaolinite [10], and nanocarbon materials [11, 12], have attracted great interest from industrial and academic fields. It has been

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proved that the presence of these nanoparticles can effectively improve stiffness and toughness of nanocomposites due to high strength and large specific surface areas of these nanoparticles. Notably, among all these nanoparticles, nanocarbon materials like carbon nanotubes (CNTs) and graphene exhibit the most suitable compatibility with PVC matrix by virtue of their carbon-atom constitution and similar density with polymer [13]. It has been reported that addition of CNTs can greatly increase tensile strength, modulus, and toughness of rigid PVC due to high strength of CNTs, good compatibility, and rich interface of nanocomposites [11, 14, 15]; however, there still exists a critical issue of dispersion of those entangled CNTs throughout PVC matrix [16].

Graphene, as promising multifunctional fillers, exhibit extremely high mechanical strength, extraordinary physical properties, and good compatibility with polymer matrix [17]. Especially, graphene show much better dispersion throughout polymer matrix than the entangled CNTs due to unique two-dimensional planar structure and large lateral size of graphene [16]. In that case, combining graphene with PVC matrix has been an effective method for obtaining significant mechanical reinforcement of graphene/PVC composites. Vadukumpully et al. used a solution blending method and obtained graphene/PVC composite films with good tensile strength, high electrical conductivity, and high thermal stability [18]. Salavagione et al. reported that the presence of chemically modified graphene oxide (GO) could remarkably improve glass transition temperature and storage modulus of the graphene/PVC composites due to strong interfacial bonding [19]. Wang and Deshmukh et al. found that the addition of GO could also improve tensile and thermal properties of GO/PVC films [12, 20, 21]. Notably, most of these graphene/PVC composites were fabricated by using solution methods due to easy dispersion of graphene in solution, but inevitably causing some problems of solvent residual, impurity, complex process, high cost, and unavailability for large-scale production of nanocomposites [22]. As a contrast, melt-mixing methods were widely used to prepared nanocomposites due to its easy operation and low cost. Recently, we have prepared commercial multilayer graphene (MLG) filled PVC composites by using melt-mixing methods and reported that the presence of graphene can increase tribological performance and tensile strength of the MLG/PVC composites [22, 23]. As far as we know, enhanced toughness of graphene/PVC composites prepared by using melt-mixing methods has not been reported yet.

The aim of this work is to improve toughness of rigid PVC using commercial multilayer graphene as toughening modifiers by taking full advantages of low density, good compatibility with polymer, easy dispersion, and high flexibility of the graphene. We prepared the MLG/PVC composites by using conventional melt-mixing method, and investigated microstructure, tensile strength, impact toughness, and dynamic mechanical behavior of the MLG/PVC composites in details. The results show that a small amount of MLG loadings can remarkably improve fracture toughness and impact strength of the MLG/PVC composites, which is mainly attributed to crumpled morphology and high flexibility of the MLG.

## EXPERIMENTAL

### *Raw Materials*

Multilayer graphene (MLG) powders, produced using interlayer catalytic exfoliation (ICE) method [24], were supplied from Sichuan Jinlu Group. Layer number of the used MLG is less than 10 graphitic layers, and its lateral size is as large as 10–15 μm and thickness is about 1–3 nm. The MLG powders possess higher structural integrity and less defects (its C/O ratio is as high as 20) than graphene oxide [23], and its electrical conductivity is over 700 S/cm. Poly(vinyl chloride) (PVC) resin powders (general type SG-5), rear-earth stabilizers (XT-1), and lubricants (ACR-401) were also supplied from Sichuan Jinlu Group.

### *Preparation of MLG/PVC Composites*

The weighed PVC powders, MLG, stabilizers and lubricants were mixed in advance at room temperature using a high-speed mixer, and there mass ratios of the stabilizers and lubricants to the PVC powders were 7 and 1.5 wt%, respectively. Thereafter, the obtained mixtures were molten at 165°C and mixed well at a rotation rate of 60 rpm for 5 min in a torque rheometer (type XSS-300, Shanghai Kechuang Rubber Production Machinery and Equipment). The obtained MLG-PVC blends were transferred into a two-roll mill, and further mixed at 170°C and squeezed into thin sheets at a rotation rate of 24 rmp (the speed ratio of front roller to rear roller was 1:1.2). Then, these thin sheets were stacked together, transferred into a hot-press machine, and pressed into flat MLG/PVC composite sheets of 2 mm in thickness at 165°C, 15 MPa for 10 min. The MLG/PVC composites with various MLG loadings of 0, 0.36, 0.72, 1.08, and 1.80 wt%, were obtained following same procedures.

### *Characterization*

Microstructure and morphology of the used MLG powders was characterized using scanning electron microscope (SEM, NavoSEM430, FEI) at 10 kV and high-resolution transmission electron microscope (HRTEM, Joel 2010) at 200 kV. X-ray photoelectron spectroscopy (XPS, ESCA LAB 250, Thermo) was used to detect chemical elements and composition of MLG surface, and atomic force microscope (AFM, CSPM400, Being Nano) was applied to characterize morphology and thickness of the MLG in a noncontact mode. Fracture surfaces of the MLG/PVC composites frozen and fractured in liquid

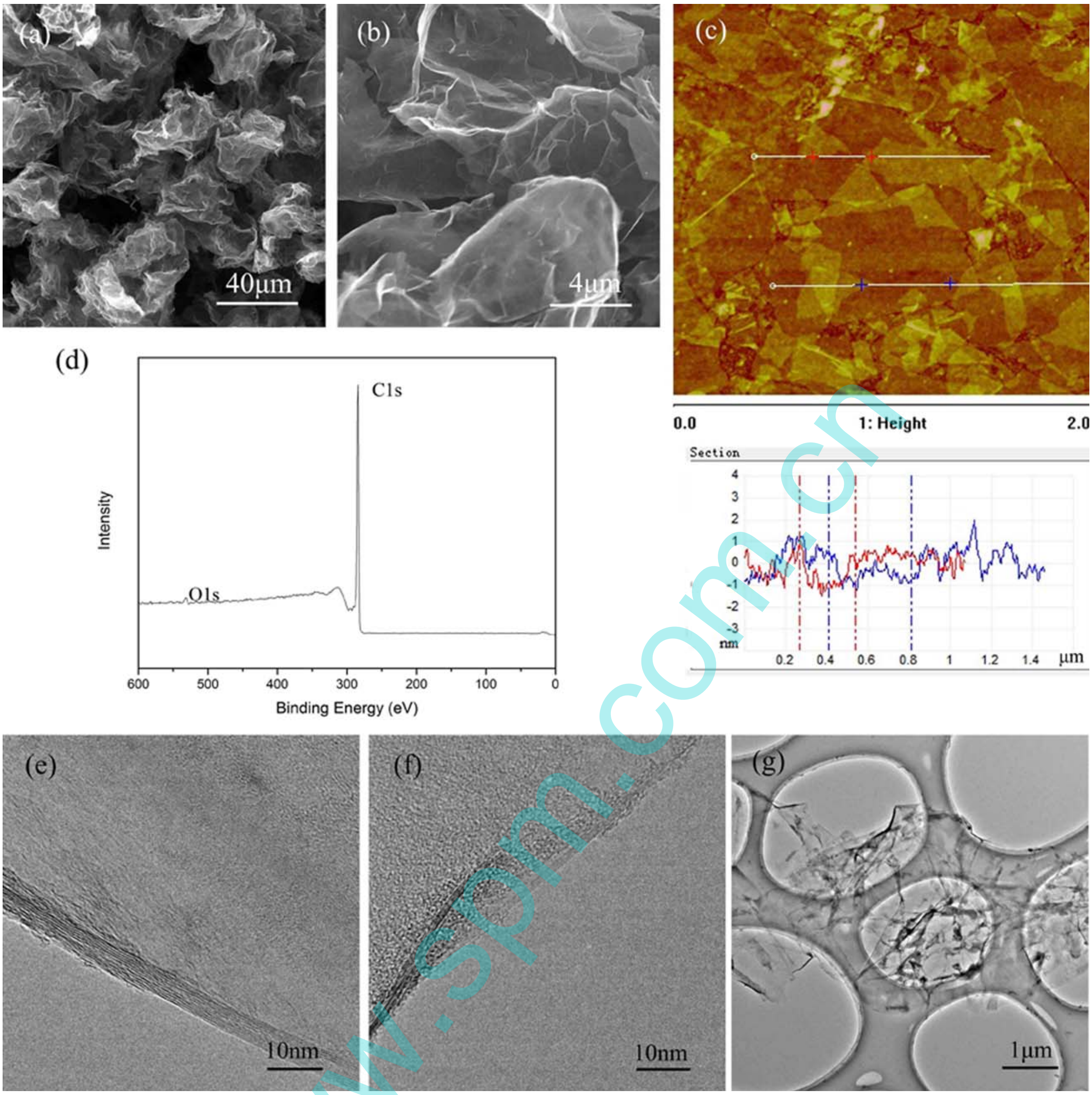


FIG. 1. Microstructure and morphology of the multilayer graphene. SEM images of the MLG at (a) 2,000× and (b) 20,000× magnifications, (c) AFM image, (d) XPS spectra, and (e–g) HRTEM images of the multilayer graphene. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

nitrogen were observed by using SEM technique. Tensile properties of the MLG/PVC composites were performed on a universal testing machine (type WSM-50KG, Changchun Intelligent Instrument Equipment) at a speed of 5 mm/min referring to ASTM D 638-10. Charpy impact tests of V-notch nanocomposites with a size of 40 mm × 3 mm × 2 mm (Type A notch, with notch-root radius of 0.25 mm and notch length of 0.5 mm, was made using a notching machine ZKY-1), were carried out referring to ASTM D 6110-10 using an impact testing machine (type JJ-20A, Changchun Intelligent Instrument Equipment). A dynamic mechanical analyzer (DMA Q800, TA Instrument) was used for measuring storage modulus, loss modulus, and loss factor of nanocomposites in a double-cantilever-beam mode at a heating rate of 3°C/min in air atmosphere, in order to investigate effect of MLG on dynamic mechanical behavior and glass transition temperature of the MLG/PVC composites.

## RESULTS AND DISCUSSION

### *Microstructure of the Multilayer Graphene*

Microstructure and morphology of the used MLG was observed and characterized by means of SEM, AFM, XPS, and HRTEM, as shown in Fig. 1. We can see from

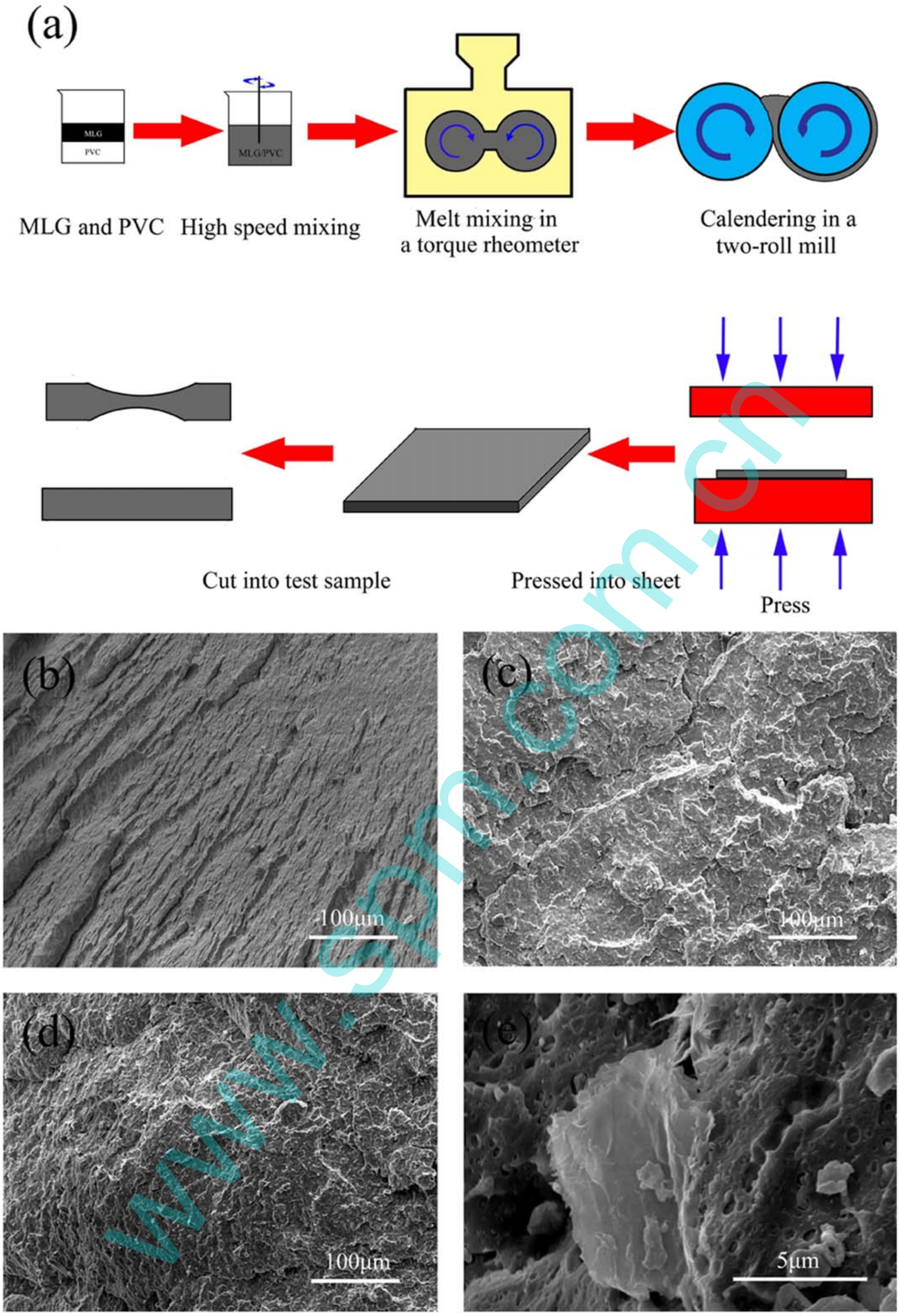


FIG. 2. Fabrication schematics (a) of the MLG/PVC composites and SEM images of (b) the neat PVC and the MLG/PVC composites with MLG loading of (c) 0.36 wt%, (d) 1.08 wt% at 500× magnification, and (e) 0.36 wt% at 10,000× magnification. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

SEM images of Figs. 1a and b that the MLG powders look like crumpled thin paper, totally different from so-called two-dimensional planar morphology for the graphene [17]. Such crumpled morphology of the MLG is mainly due to its extremely large lateral area and low thickness (its aspect ratio is more than 1000) [25]. AFM image of Fig. 1c shows that the thickness of MLG is about 1–3 nm, less than 10 graphitic layers. XPS spectra

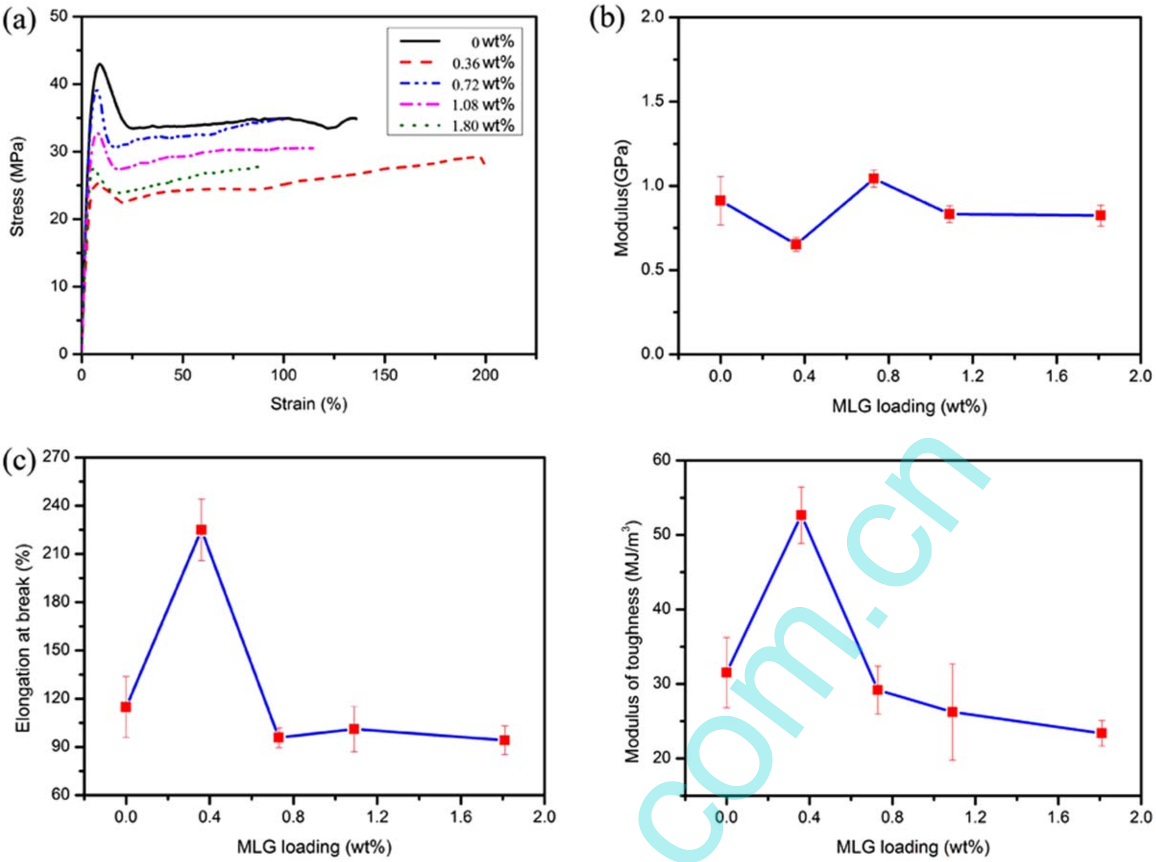


FIG. 3. Tensile properties of the MLG/PVC composites as function of MLG loadings. (a) stress–strain curves, (b) tensile modulus, (c) elongation at break, (d) modulus of toughness (strain energy density). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

shows that the C/O ratio of the MLG is about 20 (see Fig. 1d), implying less functional groups, higher structural integrity, and less defects than those graphene oxide. It is worth mentioning that the commercial MLG prepared by interlayer catalytic exfoliation (ICE) technique exhibit larger lateral size, higher quality, higher electrical and thermal conductivity, and even much lower cost than those graphene prepared by using traditional oxidation-reduction methods [24, 26], exhibiting great potential for realizing large-scale production of the MLG even at low cost. On the other hand, HRTEM images of Fig. 1e–g show few-layer microstructure and wrinkled morphology of the MLG, implying high flexibility of the crumpled MLG due to its nanoscale thickness and large lateral size [23, 25], which would be beneficial to enhancing toughness of the MLG/PVC composites.

### *Microstructure of the MLG/PVC Composites*

The MLG/PVC composites were prepared using conventional melt-mixing, calendering, and hot-pressing techniques (see Fig. 2a). Firstly, it can be clearly seen from the SEM images of Fig. 2b–d that no obvious aggregation of the MLG can be observed, indirectly implying uniform MLG-dispersion throughout PVC matrix. Although the dispersion degree of graphene by using melt-mixing method is rather lower than that by using solution method [18, 19], the obtained MLG-dispersion in our work is still acceptable, revealing that these conventional processing methods can be used to effectively disperse MLG throughout PVC matrix. It is because that the MLG are easier to be well dispersed throughout polymeric matrix than CNTs due to large lateral size, unique sheet-like microstructure, and little entanglement of the MLG [24, 27]. Secondly, coarse fracture surface of the MLG/PVC composites can be clearly observed from the SEM images of Fig. 2c and d in comparison with smooth fracture surface of the neat PVC, indicating a typical ductile fracture of the MLG/PVC composites. It means that the presence of MLG can increase fracture toughness of rigid PVC due to large specific surface area and high flexibility of the crumpled MLG [23]. Thirdly, we can see in Fig. 2e that the MLG dispersed throughout matrix exist in a form of crumpled clusters, which is because that the flexible and crumpled MLG powders are prone to be squeezed into clusters by shearing force in the melt-mixing process [16]. In that case, such high flexibility and crumpled morphology of the MLG would strongly influence mechanical properties of the MLG/PVC composites.

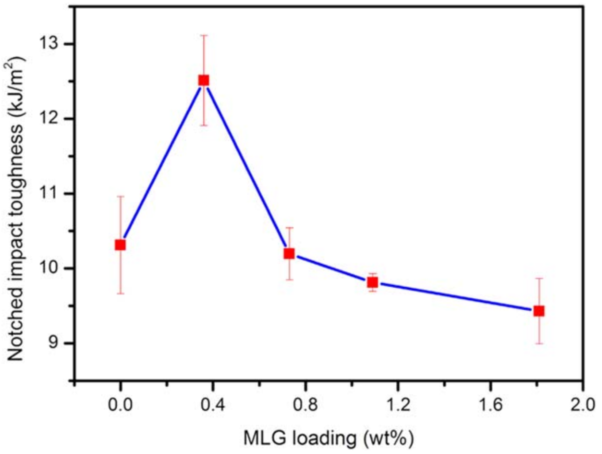


FIG. 4. Notched impact toughness of MLG/PVC composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

### *Tensile Properties of the MLG/PVC Composites*

Tensile properties of the MLG/PVC composites with various MLG loadings were measured and plotted in Fig. 3. It can be seen from the stress-strain curves of Fig. 3a that the neat PVC exhibits the highest tensile strength among all the MLG/PVC composites, implying that the presence of MLG causes a decrease in tensile strength of nanocomposites. Such little mechanical reinforcement of the MLG/PVC composites is mainly attributed to weak interactions between PVC and the MLG [28]. As mentioned above, the MLG used in our work exhibit higher structural integrity, less defect, and less active functional groups (its *C*/*O* ratio is more than 20) than those graphene oxide [18–20], indicating that there is little strong chemical bonding formed between PVC and MLG. Such weak PVC–MLG interactions inevitably result in unsatisfied load-transferring between components and low tensile strength of the MLG/PVC composites, as shown in Fig. 3a.

We can also see little mechanical improvement in tensile modulus of the MLG/PVC composites in Fig. 3b. Theoretically, the graphene possess extremely high modulus of about 1 TPa, but, in our work, the mechanical reinforcement of the MLG/PVC composites is rather weak. Such low modulus of the MLG/PVC composites is closely associated with the crumpled morphology of MLG and the weak PVC–MLG interactions. As shown in Fig. 2d, the MLG existing in nanocomposites are in a form of crumpled clusters due to its high flexibility and large aspect ratio, and these flexible and crumpled MLG are prone to be deformed under applied force, consequently resulting in low elastic modulus, even lower than that of the neat PVC [23]. In that case, the presence of crumpled MLG cannot remarkably improve elastic modulus of the MLG/PVC composites.

We can see clearly from Fig. 3 that the 0.36 wt% MLG/PVC composites exhibit the largest tensile strain at break and the largest area under strain–stress curves (strain energy density, modulus of toughness) among all the MLG/PVC composites, indicating that a small amount of MLG loadings can greatly improve tensile toughness of MLG/PVC composites. The elongation at break and modulus of toughness for 0.36 wt% MLG/PVC composites are 220% and 53 MJ/m$^3$, respectively, increasing by 95.6 and 71% in comparison with 115% and 31 MJ/m$^3$ for neat PVC. Such remarkable reinforcement in tensile toughness of the MLG/PVC composites is closely associated with high flexibility of the crumpled MLG. As mentioned above, the crumpled MLG existing in PVC matrix exhibit high flexibility and low modulus, and they can be easily deformed and orientated along axial directions in the tensile process, exhibiting large elongation at break and high strain-energy density (modulus of toughness), as shown in Fig. 3.

We can also see clearly that the tensile toughness of MLG/PVC composites strongly depends on MLG loadings. The 0.36 wt% MLG/PVC composites exhibit the highest tensile toughness among all the composites due to the high flexibility of the crumpled MLG. With the increment of MLG loadings, the elongation at break and the modulus of toughness for the MLG/PVC composites begin to decrease gradually, which is closely associated with aggregation of MLG at high loadings. At a low MLG loading of 0.36 wt%, the dispersed MLG remain isolated, crumpled, and highly flexible [23]. When the MLG loading increases, the flexible and crumpled MLG begin to contact and squeeze with each other, consequently resulting in low flexibility and high stiffness. As a result, the MLG/PVC composites with high MLG loadings exhibit high tensile modulus, low elongation at break, and low tensile toughness, as shown in Fig. 3. When the MLG loading further increases (up to 1.8 wt%), more structural defects were introduced into the MLG/PVC composites, inevitably resulting in low modulus and low tensile toughness of nanocomposites. The elongation at break of the 1.80 wt% MLG/PVC composites is as low as only 90% (see Fig. 3c). Therefore, we can see that the presence of MLG cannot remarkably increase load transferring and tensile strength due to the weak MLG–PVC interactions, but the tensile toughness of the MLG/PVC composites can be greatly enhanced, which is mainly attributed to high flexibility of the crumpled MLG.

### *Impact Toughness of the MLG/PVC Composites*

Notched impact strength of the MLG/PVC composites was measured by using a charpy impact tester, and experimental data were plotted in Fig. 4. It can be seen clearly that the 0.36 wt% MLG/PVC composites exhibit the highest impact strength of 12.0 kJ/m$^2$, increasing by 12.1% in comparison with that of the neat PVC. Such high impact strength of the MLG/PVC composites reveals that a small amount of MLG loading can greatly improve impact

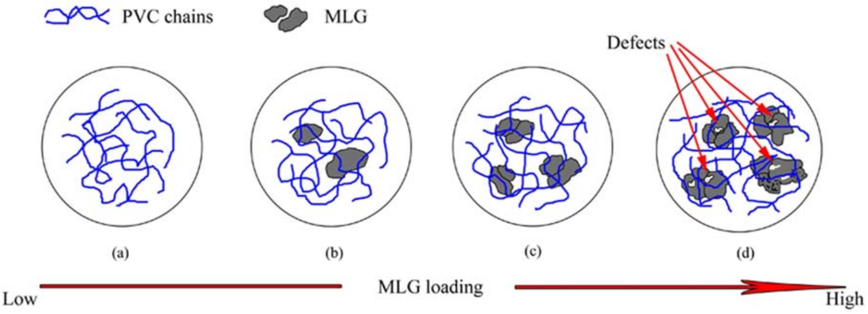


FIG. 5. Toughening mechanism of the MLG/PVC composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

toughness of nanocomposites, which is closely associated with high flexibility of the crumpled MLG. According to fracture mechanics theory [3], impact toughness of materials is closely related to energy consumed in the fracture process. Comparing with the neat PVC, the MLG/PVC composites possess much more interfaces and higher flexibility due to the presence of crumpled MLG, and they can consume much more energy in the process of deformation under applied impact loads, consequently exhibiting high impact strength of the MLG/PVC composites [23]. With further increment of MLG loadings, the impact strength of the MLG/PVC composites decreases accordingly as a result of lower flexibility and more structural defects of the MLG/PVC composites at higher MLG loadings, as shown in Fig. 4.

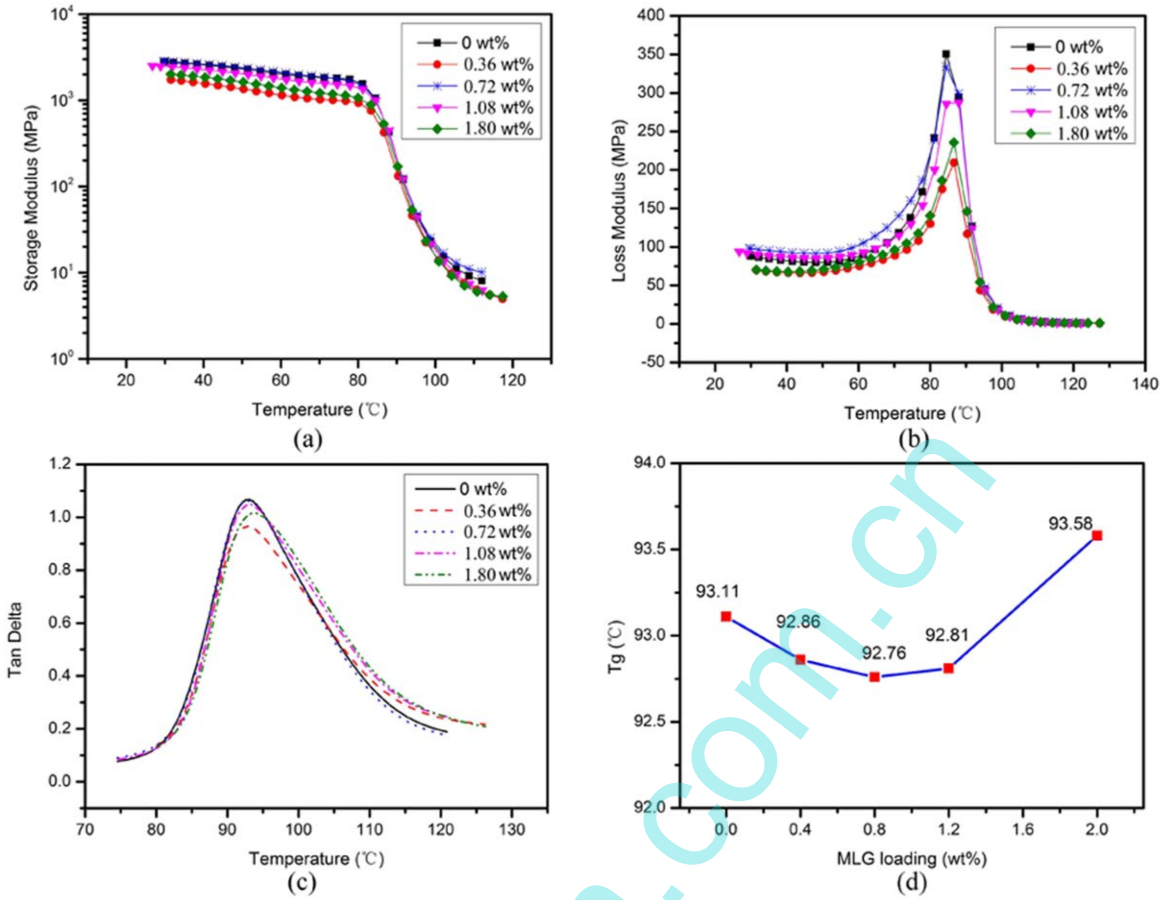
Toughening mechanism of the MLG/PVC composites can be further illustrated in Fig. 5. When a small amount of MLG loadings (0.36 wt%) is added into the neat PVC matrix, these MLG can be easily and well dispersed throughout the PVC matrix by shearing force in conventional melt-mixing processes, as shown in Fig. 2c. These well-dispersed MLG still remain isolated, crumpled, and highly flexible (see Figs. 5b and 2e), and they can be easily deformed under applied impact loads, consequently consuming much impact energy (high impact toughness shown in Fig. 4). On the other hand, the presence of the MLG between PVC chains can also enlarge intermolecular distance and weaken interactions between PVC chains [22]; thus, resulting in easy segmental motion of PVC chains and high impact strength of the MLG/PVC composites.

With the increment of MLG loadings, these isolated MLG begin to contact with each other (see Fig. 5c) and cause a decrease in flexibility of nanocomposites as a result of squeezing and compression between MLG and PVC chains, consequently showing low impact strength of the MLG/PVC composites at high MLG loadings. With further increment of MLG loading (up to 1.08–1.80 wt%), more structural defects were introduced into the MLG/PVC composites due to large specific surface area of the MLG (see Fig. 5d), inevitably resulting in low tensile strength and impact toughness of the MLG/PVC composites, as shown in Figs. 3 and 4. Therefore, in our work, the high impact strength of the 0.36 wt% MLG/PVC composites is mainly attributed to high flexibility of these isolated and crumpled MLG.

We further compared toughening effect of the MLG/PVC composites with that of other modified PVC materials. The toughening effect of the MLG/PVC composites in our work is comparative and even better than that of the kaolin/PVC [10] and montmorillonite/PVC composites [9]. For obtaining the same enhanced toughness, the MLG loading in our work is as low as only 0.36 wt%, much lower than 3 wt% for the kaolin or montmorillonite filled PVC composites, which is mainly attributed to low density and high flexibility of the crumpled MLG, and good compatibility between MLG and PVC [22]. On the other hand, the toughening effect of the MLG/PVC composites is lower than the modified CNT/PVC [15] and rubber impact modifier filled PVC composites [5, 29], which is closely associated with weak MLG-PVC interactions and low elasticity of the MLG [5]. Notably, as motioned in introduction section, the MLG/PVC composites still exhibit its advantages of easy MLG dispersion and high heat resistance in comparison with that CNTs or rubber filled PVC composites. Moreover, it is worth pointing out that the toughness of MLG/PVC composites strongly depends on many influencing factors, such as dispersion of graphene, interfacial bonding, intrinsic toughness of components, filler loadings, fabrication conditions, gelation degree of PVC matrix, and so on. In that case, optimal design and full consideration should be made for obtaining high-performance nanocomposites. Furthermore, with breakthrough of large-scale production of multilayer graphene at low cost [24], the cost of commercial multilayer graphene is decreasing drastically and will be gradually competitive and acceptable, and there is great potential for graphene to be utilized in some fields of high-performance composites.

### *Dynamic Mechanical Properties of the MLG/PVC Composites*

We further investigated influence of the MLG on segmental motion of PVC chains by using a DMA instrument, and the dynamic mechanical behavior of the MLG/



(a)

(b)

(c)

(d)

FIG. 6. Dynamic mechanical behavior of MLG/PVC composites. (a) Storage modulus, (b) loss modulus, (c) tan δ, and (d) the glass transition temperature ($T_g$) as function of MLG loadings. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

PVC composites was shown in Fig. 6. First of all, it can be seen clearly from Fig. 6a that all the MLG/PVC composites show lower storage modulus than the neat PVC, which is consistent with tensile modulus of the MLG/PVC composites shown in Fig. 3. Such low storage modulus of the MLG/PVC composites mainly results from the high flexibility of the crumpled MLG. On the other hand, the loss modulus (energy dissipation) and tan $\delta$ (damping ratio) of the MLG/PVC composites in the heating process were shown in Fig. 6b and c, directly reflecting responsive behavior of the PVC chains at different temperatures [18]. The corresponding temperature of the peak of tan δ is denoted as glass transition temperature ($T_g$), which is one of the most important parameters for evaluating chain segmental motion and heat resistance of polymeric materials. Low values of $T_g$ generally indicate easy segmental motion of chains, high toughness, and low heat resistance of polymeric materials [30].

Glass transition temperatures of the MLG/PVC composites against MLG loadings were plotted in Fig. 6d. It can be seen clearly that the $T_g$ values of the MLG/PVC composites with low MLG loadings (0.36–1.08 wt%) are lower than that of the neat PVC, implying that the presence of MLG can greatly increase the segmental motion of PVC chains. Notably, the $T_g$ value is closely associated with chain segmental motion and strongly dependent on interactions between polymer chains [30]. In our work, as mentioned above, the presence of the crumpled MLG with high flexibility and high structural integrity can enlarge intermolecular distance and weaken interactions between MLG and PVC chains [22, 23], consequently resulting in quick segmental motion of PVC chains and low $T_g$ values of the nanocomposites (92.76°C for the 0.72 wt% MLG/PVC composites versus 93.11°C for the neat PVC). Such quick segmental motion of PVC chains in the presence of MLG can consume quickly much energy and result in high toughness of the MLG/PVC composites. On the other hand, when the MLG loadings further increase, these MLG begin to contact and squeeze with each other and consequently hinder segmental motion of PVC chains, resulting in high $T_g$ values of nanocomposites (93.58°C for the 1.80 wt% MLG/PVC composites shown in Fig. 6). Therefore, in our work, a small amount of MLG loadings is beneficial to increasing segmental motion of PVC chains and consequently improving toughness of the MLG/PVC composites due to the high flexibility of the crumpled MLG.

## CONCLUSIONS

In this work, we prepared the MLG/PVC composites by using conventional melt-mixing techniques in order to improve toughness of rigid PVC by taking advantages of easy dispersion and high flexibility of the crumpled MLG. Microstructure, tensile properties, impact toughness, and dynamic mechanical behavior of the MLG/PVC composites were investigated in details. We found that that a small amount of MLG loadings can remarkably improve tensile fracture toughness and notched impact strength of the MLG/PVC composites, which is mainly attributed to the high flexibility of the crumpled MLG throughout PVC matrix. Moreover, the presence of MLG can also enlarge intermolecular distance and weaken interactions between PVC chains, consequently resulting in easy segmental motion of PVC chains and high toughness of the MLG/PVC composites. By virtue of its enhanced toughness and easy operation, the MLG/PVC composites show great potential to be applied as high-performance materials in many fields.

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