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Engineering Foam Skeletons with Multilayered Graphene Oxide Coatings for Enhanced Energy Dissipation

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Abstract: This work shows how to improve the energy dissipation of open-cell polyurethane (PU) foams by creating multilayered graphene oxide (GO) nano-architectures onto the struts via a modified dip-coating process. Pristine PU foams are alternately dip-coated with GO coatings and water-based polyurethane dispersions (PUD) for a given number of times. The GO coating morphologies are carefully adjusted and the inner energy dissipation mechanisms reach the optimized interfacial frictions of GO-PU and GO-GO. Along with the synergistic effect of the multiple interpenetrating structure of GO/PU coating phases, these engineered composite foams with extremely low GO content (~0.12wt%) afford a significant increase of quasi-static energy dissipation (52%) and dynamic damping (76%) when compared with counterpart foams coated with the same number of pure PUD layers. The specific Young's modulus and strength of the designed foams also show remarkable enhancements of 310% and 490% respectively compared with those of pristine PU foams.

Keywords: graphene oxide; interfacial friction; nanocomposite foams; energy dissipation

1. Introduction

Synthetic cellular materials like structural foams and honeycombs possess energy dissipation characteristics and are widely used for packaging, cushioning, acoustic and vibration damping [1,2]. In recent years the use of polymer nanocomposite foams for vibration and noise reduction has attracted much interest within the research community to increase the energy dissipation efficiency in classical viscoelastic polymer foams [3,4]. Experimental and numerical studies have been carried out to evaluate the interfacial sliding mechanism for damping performance in nanocomposites [5-9]. Two main interface coupling mechanisms have been proposed to describe this phenomenon in polymer-filler and filler-filler interfacial frictions [8,10]. Polymer-filler sliding is determined by the interface bonding of polymer/filler and the dispersion quality of fillers [7-9], while the filler-filler sliding is closely related to the filler content [7,8,11]. Carbon nanotubes (CNTs), for example, have been used as reinforcements inside the cell struts of polymer foams produced by direct foaming. The overall enhancement in damping exhibited by thus type of porous nanocomposite is however quite limited (no more than 30%), because it merely takes advantage of the polymer-CNTs interfacial sliding mechanism [3,12,13]. Although the increase of the CNTs content can introduce more filler-filler sliding, it can also causes a virulent aggregation of the nanoparticles, therefore leading to a deterioration of the mechanical properties [12].

Among various types of carbon nanofillers, 2D graphene nanoplatelets possess ultrahigh mechanical strength and extremely large specific surface area, which are proposed to be favorable characteristics for application in damping polymers [14,15]. The damping properties of traditional graphene nanocomposites fabricated by directly mixing matrix and fillers show however a very limited enhancement (less than 15%) or even a decrease after adding graphene [16-19]. This is mainly caused by the low dispersion capacity of 2D graphene nanoplatelets in the matrix, as the natural phenomena of re-aggregation and self-scrolling largely decrease the contribution of the high specific surface area for interface friction [16,17]. The dispersion of graphene can be improved by chemical modifications, and the resulting strong graphene-matrix

interface can also enhance the Young's modulus and the strength of the composites. However, an increase in the level of bonding between the graphene and the matrix dramatically restricts the interface sliding performance and therefore reduces damping [18,19]. Moreover, a more uniform dispersion of the modified graphene nanoplatelets tends to restrict the mobility of the polymer matrix chains, and that inhibits the natural energy dissipation capacity of the matrix [10,20]. As a consequence, graphene nanoplatelets have been seldom used as reinforcements in polymer composite for damping applications, especially the nanocomposite foams fabricated from directly foaming technology. To address this issue, the skeleton microstructure of the foams should therefore be carefully designed to make full use of the large specific surface area of graphene, without causing a deterioration of the base mechanical properties of the polymer matrix.

A dip-coating technique using graphene nano inks has been recently developed to generate 3D graphene-polymer nanocomposite foams for enhanced electrical, thermal, mechanical (Young's modulus and strength) and microwave absorbing properties [21-23]. This particular fabrication process can not only overcome drastic reaggregation, restacking and self-scrolling of the 2D graphene nanoplatelets, but also maintain the integrity of the polymer as a network configuration. The work in this paper presents a modified dip-coating method to engineer PU foams for vibration damping applications where foam skeletons possess multilayered sandwich architectures. The unfolded GO nanoplatelets provide the same effects of functional damping cores in sandwich nano-structures, and meanwhile allow to preserve the pristine 3D foam network of struts by superimposing the sandwich coating layers. Within this configuration, the damping capacities of the foams are significantly enhanced by the synergistic effect of the multiple interpenetrating network structure of GO/PU coating phases. The internal energy dissipation mechanisms of GO-PU and GO-GO interfacial frictions responsible for the increase in damping are carefully described. Meanwhile, the specific Young's modulus and strength of the fabricated foams also increase due to the coated configuration compared with those of pristine PU foams. The functions of GO nanoplatelets on the mechanical properties of the foams are detailedly illuminated under the deformations of the skeletons with different coating layers.

2. Experiments

2.1 Design and fabrication of the nanocomposite foams

The manufacturing process is illustrated in **Fig. 1a** and the details are supplied in the Supporting Information (Fig. S1). The pristine open-cell PU foams (PPI: 60-80, purchased from Changzhou Daye Tengfei Sponge Factory, China) are used as the skeleton templates and Step 1 produces foams coated with the required wt% GO nano inks. A water-based aliphatic polyurethane dispersion (PUD, 715, obtained from Jining Huakai Resin Co. Ltd. China) is then used as a top coating and Step 2 produces structural foams with one-layer sandwich struts, with the GO core coating acting as the damping layer. Foams with multi-layer sandwich skeletons can be successfully fabricated by repeating Steps 1 and 2 for several times (*i.e.*, Steps 3 and 4). The obtained samples were denoted by $n(A+B+C)$, where n , A, B and C stand as the number of sandwich coating layers, pristine PU foam skeleton, GO core layer and PUD constraint layer respectively.

The O₂ plasma treatment applied here has a double function. On the one hand, the treatment hydrophilizes the to-be coated foams with oxygen-contained groups so that, the inks or the water-based PUD can be easily absorbed into the foams due to improved wettability; on the other hand, the treatment can also strengthen the adhesion of the coating interface by forming chemical bonds with the assistance of silane coupling agents (KH-550) [11,24]. The vacuum and application of constant pressure are exerted here to provide close contact between the base material and the coating solutions (including GO ink and water-based PUD). The GO inks were prepared based on our previous work [24] and the as-prepared GO nanoplatelets (Figure 1b, c) are ~10 μm in lateral size and ~6 nm in thickness. XRD and XPS characterizations (Fig. S2) show that the GO nanoplatelets are successfully fabricated and functionalized with oxygen-contained groups. The three-dimensional configuration of pristine PU foams (Fig. 1d) illustrates an interconnected framework with pore sizes ranging between 200 μm and 450 μm . The cross-section of the struts presents a triangular shape with an equivalent diameter of ~50 μm . The large flat surface of the struts can ensure enough space for the deposition of the GO nanoplatelets (~10 μm) in an unfolded status, which is designed to fully utilize the large specific surface areas of GO. By sandwiching GO damping core layers between two constrained layers of PU matrix, we can

create an efficient damping mechanism that dissipates mechanical energy into heat through the nano-scale deformation of the GO sheets with relative displacements.

2.2 Characterizations

Surface morphologies were investigated by a cold Field Emission Scanning Electron Microscopy (FE-SEM, Hitachi S-4800) operating at 3.0 kV. Before observation, the samples were sprayed with Au for 90 s. The dimension (edge size and thickness) of the as-prepared GO nanoplatelets are measured by a Scanning Probe Microscope (SPM-9700, SHIMADZU) conducted at room temperature. Raman spectra registered on a Micro Raman spectrometer (109907hk, Renishaw plc) were carried out to identify the GO coatings and PU surfaces. X-ray photoelectron spectroscopy (XPS) equipment supplied by Kratos Analytical Ltd, was employed to analyze the oxygen-contained functional groups of GO and the X-Ray Diffraction (XRD Empyrean 200895 PANalytical B.V.) was used to define the lamellar distance of GO sheets. The viscosities of coating inks with different GO concentrations were measured by Digital Viscometer NDJ-8S at room temperature.

The quasi-static (cyclic) compression tests were performed at room temperature using Zwick/Roell Z5.0 universal testing machine with a 5kN load cell. Compression tests were taken under displacement control with a maximum strain of 18% at a crosshead speed of 3 mm/min. Cyclic compression tests were conducted with a maximum strain of 10% in a triangular waveform at a loading-unloading speed of 5 mm/min. The dimension of samples is 30×30 ×15 mm, which is based on the test standard for plastic compressibility (GB/T1041-2008). The dynamic damping properties as frequency-dependent viscoelastic properties (storage modulus E' , loss modulus E'' and $\tan \delta$) of the designed nanocomposite foams were evaluated by a Dynamic Mechanical Analyser (DMA-TAQ800) under compression mode. The specimens were subjected to a frequency scan ranging 1-30 Hz with a fixed amplitude of 80 μm (1% deformation) at room temperature.

3 Results and discussion

3.1 Foam skeletons with one-layer sandwich coating

3.1.1 Morphologies of foam skeletons with one-layer sandwich coating.

As the content of GO nanoplatelets can largely determine the performance of the interfacial sliding mechanisms [3,12], samples coated with GO inks of various concentrations (0.05wt%, 0.1wt%, 0.2wt%, 0.3wt% and 0.4wt%) have then been prepared. It can be clearly observed from **Fig. 2a** that the surfaces of the struts belonging to the pristine PU foams are smooth, while the same surfaces gradually become rougher with the increase of the GO ink concentrations. The surfaces of the ink-coated foams exhibit island-like GO coatings when relatively low GO dispersions are adopted (0.05wt% and 0.1wt%). The unfolded and wrinkled GO nanoplatelets almost cover the whole strut surface when the ink concentration reaches 0.2wt%, forming an entangled GO coating network with some sparsely uncovered PU areas (Fig. 2b, c). The GO coating and PU surface are identified with Raman spectroscopy analysis as GO coating reveals the typical D and G peaks while PU shows its intrinsic characteristic peaks (Fig. S3) [24, 25]. A more pronounced and continuous deposition of GO gradually appears on the strut surface with increasing coating ink concentrations. The 0.3wt% (Fig. 2a₅) and 0.4wt% (Fig. 2a₆) ink-coated surfaces show similar morphologies, with the unfolded and overlapped GO nanoplatelets successfully coated on the whole surfaces of the PU skeletons. Apart from the preparation of the quality GO ink solutions based on our previous work that produce inks with desired viscosity (Fig. S4) [24], another crucial point is the use of a centrifugation process that ensures even coatings (Fig. S5).

After the GO inks coating, foams were then coated with a water-based PUD acting as a top constrained layer. SEM images of the pristine PU foams after only PUD coating (**Fig. 3a, b**) show that the foam skeletons are fully wrapped with a smooth and uniform PUD layer of thickness $\sim 2\ \mu\text{m}$ due to the applied centrifugation process. The close bonding of this PUD layer to the interface is explained by the similar solubility of the two PU matrixes, and also by the increased surface wettability obtained through the O_2 plasma treatment. The cross-section of a single strut coated with 0.2wt%GO+PUD (Fig. 3c, d) show that the strut is similar to a coaxially coated sandwich beam, with the GO nanoplatelets acting as the core layer. The GO core coating is not completely continuous and this is consistent with the 0.2wt% ink-coated morphology because the struts are not fully covered with the GO nanoplatelets (Fig. 2b, c). Despite the presence of some

holes caused by the pull-out of the GO nanoplatelets during the preparation of the samples for the SEM characterization, the PU and PUD phases appear to be directly bonded with the GO core coating, indicating the good compatibility between PU and GO due to the O₂ plasma treatment. The morphology of the separated PU and PUD layers (Fig. 3e, f) shows that GO nanoplatelets are still attached to the PU skeleton and also partly appear on the PUD surface. This is due to the chemical bonds amongst the PU-GO and the PUD-GO interfaces created by the coupling agents, which are stronger than the interlaminar Van der Waals' forces between the GO nanoplatelets [26]. The GO nanoplatelets at the overlapped areas are therefore exfoliated and delaminated when the PU and PUD phases are separated from each other [24].

3.1.2 Mechanical, quasi-static energy dissipation and dynamic damping properties of the foams.

The stress-strain curves from quasi-static uniaxial compression tests are shown in **Fig. 4a**. In this work, we have chosen the slope within the linear elastic range of 2%~3% strain to calculate Young's modulus (E), and the plateau stress outside the linear elastic range at 10% strain as the corresponding strength (σ_{10}) [11]. The pristine PU foam has the smallest E of 48 kPa and σ_{10} of 3.4 kPa among all the tested foams (Fig. 4b, c). The Young's Modulus E first increases with the GO ink concentration reaching the peak at 265 kPa for the PU+0.2wt%GO+PUD sample, and then starts to decrease. The decrease can be explained by the easier slippage between GO-GO interfaces compared with strong chemical bonded GO-PU interfaces [24]. This is in consistent with the coating morphology, with plenty of overlapped GO nanoplatelet attachments when the ink concentration goes beyond 0.2wt% (Fig. 2). On the contrary, the strength of these foams monotonically increases with the increase of the coating ink concentration, and finally reaches a maximum value of 16.5 kPa. This is because of the self-locking of the wrinkles when the foams are subjected to a large compressive strain (σ_{10} at 10%). The GO nanoplatelets related to the 0.4wt% ink coatings are unfolded, but they still present some wrinkles and overlap together (Fig.2). The interactions between wrinkles of different nanoplatelets contribute to the energy dissipation via GO-GO interfacial friction under relatively small deformations. For large applied strains, too much interactions between wrinkles occur and cause however the self-locking of the same wrinkles [24]. In this case, the load can be efficiently transferred between the GO sheets with a consequent increased strength of the foams versus the increased GO

content.

The effect of the GO coatings on the energy dissipation of the foams is firstly evaluated by the hysteretic loading-unloading curves from the cyclic quasi-static compression. The energy dissipation (ΔW) can be derived from the area of hysteresis loop and the corresponding loss factor (η) can be given by the following equation [11,27] (Fig. S6):

$$\eta = \frac{\Delta W}{\pi U} \quad (1)$$

where U is the maximum energy stored during a cycle and can be calculated from the area under the middle line of the hysteresis loop. The tests were conducted for five cycles and the fifth hysteresis loop was used for the calculations [28]. The results (Fig. 4d) show that both the energy dissipated ΔW and the loss factor η of the samples with the GO core coatings are higher compared with those of pristine PU foams, and the optimum amplitudes are achieved when the concentration of the GO coating ink reaches 0.2wt%.

The dynamic viscoelastic performance of these samples was also characterized by dynamic mechanical analysis (DMA) to evaluate their damping properties. The storage modulus E' and damping loss factor $\tan \delta$ under different frequencies (Fig. 4e, f), which are derived from the frequency sweep curves (Fig. S7), present a variation of those values versus the ink concentration similar to the Young's modulus and loss factor in quasi-static compression (cyclic) tests. Both E' and $\tan \delta$ experience an increase-decrease process versus the ink concentration, with the optimal performance reached by the PU+0.2wt%GO+PUD sample. The slightly increased value of E' between 1-30 Hz is likely due to the frequency-dependent viscoelasticity of the PU polymer. In addition, the values of E' (~0.30 MPa) and $\tan \delta$ (~0.16) at 1 Hz are within the same order of magnitude (slightly higher) of the Young's modulus (~0.25 MPa) and loss factor (~0.11) from the quasi-static compression tests. These data confirm the consistency and reliability of our measurements.

3.1.3 Mechanisms of quasi-static and dynamic energy dissipation affected by GO content.

There are two main types of energy dissipation mechanisms facilitated by the GO coating under vibration (cyclic) deformations here: the GO-PU and the GO-GO interfacial frictions. The GO-PU interfaces have a strong adhesion because of the chemical bonds from the silane coupling agents, and therefore require large forces to activate the interfacial slide

[29, 30]. On the opposite, the GO-GO interfaces are mainly kept together by much weaker Van der Waals' interactions, with significantly smaller forces and energies required to activate the interfacial slide [31]. Moreover, these two types of interfacial friction mechanisms are in mutual competition and significantly affected by the GO content.

As illustrated in **Fig. 5a**, the GO-PU interface friction plays a predominant role at low GO contents (0.05wt% ink coating, for example) [32], contributing to the enhanced energy dissipation of the samples compared with the PU+PUD foams. Because of the affinities between GO, few GO nanoplatelets may still be stacked onto the PU surface, even in the case of ultra-low ink coating. With the GO content increasing (0.05wt%-0.2wt% inks), more GO-PU interactions are generated and the GO-GO interactions gradually increase due to the increasing number of overlapped GO nanoplatelets, resulting in the continuous enhanced energy dissipation capacities. When the content reaches 0.2wt% ink coating (**Fig. 5b**), both the GO-PU and GO-GO interface frictions are fully activated and show an optimized energy dissipation behavior. For this particular ink loading more GO-PU interfaces on both sides of the stacked GO nanoplatelets are also present; the GO-GO interfaces that require lower driving forces tend however to slip first, and this type of GO-PU interfaces cannot be activated. With larger GO loading (0.2wt%-0.4wt% inks, **Fig. 5c**), the overlapped GO core coating gradually covers the whole PU strut surfaces and the GO-GO sliding begins to prevail over the GO-PU one. The GO-PU mechanism gradually comes to be dead and the net result is the decrease of the energy dissipation performance in these samples. The PU+0.2wt%GO+PUD samples therefore show the highest loss factor increase of 31% (quasi-static test) and 45% (DMA) compared with the PU+PUD foams. It should also be mentioned that the energy dissipation properties of the GO coated sandwich foams are always higher than those of the pristine and PUD coated foams regardless of the GO content.

3.2 Foam skeletons with multi-layer sandwich coatings

3.2.1 Morphologies and characteristics of foam skeletons with multi-layer sandwich coatings.

The cross-sections of the foam struts dip-coated multiple times (five in the present case) with the optimal 0.2wt% GO ink (**Fig. 6a**) show an evident five-layer sandwich structural coating with strongly combined interfaces between the different layers. The five quasi-connected GO core layers can be easily identified and can be defined as the repeated

configuration of the one-layer sandwich structure. The uniform thickness of $\sim 2 \mu\text{m}$ for different coating layers is also evidence of the robustness of the dip-coating process. Considering the slight changes of the to-be coated skeleton surfaces for each coating, the PUD contents are $\sim 13.7\text{wt}\%$ for 1st, $\sim 15.4\text{wt}\%$ for 2nd, $\sim 17.1\text{wt}\%$ for 3rd, $\sim 19.6\text{wt}\%$ for 4th and $\sim 21.2\text{wt}\%$ for 5th. The sizes of the struts inside the foams with different coating layers are measured and analyzed and show an expected decrease of the length/diameter ratio (getting thicker) with the increasing number of coating layers (Fig. 6b, c). The densities of $n(\text{PU}+\text{PUD})$ and $n(\text{PU}+\text{GO}+\text{PUD})$ foams continuously increase with increased n and are always similar for a constant n value (Fig. 6d). This indicates a negligible added weight provided by the GO coatings compared with the PUD coating layers.

3.2.2 Mechanical properties of foams under different coating layers.

The compression stress-strain curves of the multi-coated foams are shown in **Fig. 7a** and samples with various layers of pure PUD coating (Fig. 7b) were also tested for comparison. Both Young's modulus E and strength σ_{10} (Fig. 7c, d) increase monotonically with the number of coating layers n , which can be explained by the continuously increased density [33]. However, the increasing rates (slopes of the curves) of the Young's modulus and strength in $n(\text{PU}+\text{PUD})$ samples are slightly larger than those in $n(\text{PU}+\text{GO}+\text{PUD})$ specimens. The Young's modulus and strength of GO+PUD coated samples are higher than those of PUD coated ones when only one or two coating layers are present, and appear to be lower when n reaches four or five. This indicates that the added GO coatings in $n(\text{PU}+\text{GO}+\text{PUD})$ specimens tend to act as reinforcements in samples coated with low layer numbers (one and two), however to be detrimental to the whole mechanical performance for samples with high numbers of coating layers (four and five). The number of layers for which the properties of multi-layer sandwich strut foams transition from reinforcement to detriment is 2~3 for the Young's modulus, and 3~4 for the strength. This may be due to the different compressive strain ranges used to identify the properties (2~3% for E and 10% for σ_{10}). Although the GO coatings impair the basic compression properties of the foams with high numbers of deposited nano-ink layers, the 5(PU+GO+PUD) foams still exhibit quite remarkable 310% and 490% enhancements of the specific Young's modulus and strength (E and σ_{10} divided by the density) respectively

compared with the pristine PU foams (Fig. 7c,d, inserted).

A number of micromechanical models have been developed to understand the mechanical response of foam materials under compression [33,34]. For example, Gibson and Ashby demonstrate that the Young's modulus E and plastic yield strength σ_{pl}^* of an open-cell foam are related to its relative density ρ/ρ_0 when the foam struts bend, and described by the following equations [33]:

$$\frac{E}{E_0} = A \left(\frac{\rho}{\rho_0} \right)^2 \quad (2)$$

$$\frac{\sigma_{pl}^*}{\sigma_{ys}} \approx 0.3 \left(\frac{\rho}{\rho_0} \right)^{3/2} \quad (3)$$

where A is ~ 1 , ρ is the density of the foam, E_0 , ρ_0 and σ_{ys} are the corresponding Young's modulus, density and yield strength of the solid material used to make the foam. The first-order linear fitting curves of $E \sim \rho^2$ and $\sigma_{pl}^* \sim \rho^{3/2}$ (Fig. 7e, f) show that the linear relationships match well for samples with low numbers of coating layers, however tend to deviate for samples with high numbers of coatings. Moreover, the deviations of E and σ_{pl}^* belonging to the $n(\text{PU}+\text{GO}+\text{PUD})$ samples are larger than those related to the $n(\text{PU}+\text{PUD})$ specimens. This means that the experimental results cannot agree well with Eqs. (2) and (3) for samples with high numbers of coating layers, and the larger deviation in the $n(\text{PU}+\text{GO}+\text{PUD})$ samples could be expected by the presence of the GO core coatings. It must be however noticed that Eqs. (2) and (3) do not consider stretching and transverse shear deformations of the struts [34-36], and therefore these assumptions are not suitable for foams with thick skeletons (with high numbers of coating layers).

3.2.3 Deformation mechanisms of foam skeletons and effect of GO on compressive mechanical properties.

The inner structure of the open-cell foams with different coating layers after being subjected to a 10% compression strain (where the strength σ_{10} records), have been observed *via* SEM to identify the deformations of the skeletons. The beam-like ligaments of the foams show some evident residuals of bending and buckling deformations in the weak spots of the struts (**Fig. 8a-f**, the marked red circles). The deformations, clearly identified here, mostly happen in the major load-bearing struts whose angles α (between the applied load direction and the ligament orientation) are within $-45^\circ \sim +45^\circ$ [37,38]. These deformations are mitigated by increasing the number of coating layers and finally almost

disappear in samples with five coating layers. That is because the getting thicker struts can withstand a higher compression load due to their corresponding increased bending stiffness and buckling strength [33]. Meanwhile, the shear compliance of the cross-section of the struts will increase and may outweigh the increase in bending stiffness (buckling strength), leading to the additional shear deformation (Fig. 8g, h) [36,39]. The intrinsic strut deformations of the foams at 10% strain therefore gradually transform from bending and buckling to axial compression and transverse shear (difficult to mark these deformations) when the number of coatings increases from zero to five. This type of shift of deformation mechanism can also be identified from the behavior of the stress-strain curves (Fig. 7a, b). The plateau following the elastic region is the typical compression response of pristine PU foams (enlarged figure, inserted) since the skeletons buckle and collapse [36,40]. As the density of the foams gradually increases with the number of coating layers, the plateau becomes less pronounced and the curves tend to be the classical compression curves of dense bulks [41,42].

Other works have referred to the fact that graphene nanoplatelets can act as stretched reinforcement nanofillers in composites to enhance the strength under tensile loading, however they could also be in-plane contracted to weaken the compression strength of the composites [43,44]. The GO nanoplatelets here are designed to be unfolded and to be distributed along the axis of the foam struts, but also to be embedded at a depth in the ligaments. The GO will be stretched when the high aspect ratio struts (Fig. 8g, “slender” ones with low numbers of coatings) bend and buckle. The stiffness and strength of the foams can be enhanced in that case because the stretched GO nanoplatelets can transfer the load and therefore function as reinforcements of the matrix. On the contrary, the GO will be contracted (shrunk) when the low slenderness struts (Fig. 8h, “stubby” ones with high numbers of coatings) are axially compressed and transversely sheared (still axially compressed under the axial partial stress). This will weaken the stiffness and strength of the foams because the contracted GO nanoplatelets cannot withstand a compression load, and therefore act as triggers for defects in the matrix. Fig. 7c and 7d consequently reflect such reversion relationships of the mechanical properties between $n(\text{PU}+\text{PUD})$ and $n(\text{PU}+\text{GO}+\text{PUD})$ samples versus the number of coating layers.

3.2.4 Quasi-static energy dissipation properties of multi-layer coated foams

The quasi-static energy dissipation properties of the composite foams (**Fig. 9**) under cyclic compression show that the loss factors of the $n(\text{PU}+\text{PUD})$ samples increase first and then decrease as the coating layers (n) increase, with the highest η at $n=3$. This could be explained by the transition between different deformation forms at strut levels (from bending/buckling to axial compression/transverse shear) during the cyclic compression of the foams. The cyclic bending/buckling of the skeletons is a more effective energy dissipation mechanism for foams than the cyclic compression/shear of the struts [45,46]. The enhanced loss factors of the samples with increasing PUD coatings (1-3) are ascribed to the more efficient energy dissipation caused by the bending/buckling of the getting thicker ligaments. However, the bending/buckling gradually recedes and transforms to the compression/shear deformation, leading to the decreased η for samples with high PUD coatings (3-5). Besides, according to Eq. (1), the decreased values of η are also caused by the increased stored maximum energy U owing to the increased stiffness (Young's modulus E) of the foams with higher PUD coatings. On the contrary, the loss factors η of the $n(\text{PU}+\text{GO}+\text{PUD})$ samples are dramatically larger than those of the $n(\text{PU}+\text{PUD})$ specimens and monotonically increase with the coating layers (n). This demonstrates that the GO core coatings can enhance the energy dissipation of the foams and the contribution surpasses the reduction due to the high PUD coatings. The increments $\Delta\eta$ of the loss factors between the $n(\text{PU}+\text{GO}+\text{PUD})$ and the $n(\text{PU}+\text{PUD})$ foams continuously increase with the coating layers from 27% ($n=1$) to 52% ($n=5$).

3.2.5 Dynamic damping properties of the multi-layer coated foams

The dynamic viscoelastic properties of the multi-layer coated foams show that the storage modulus E' (**Fig. 10a-c**) increases with the number of coating layers, and this is coincident with the Young's modulus E from the quasi-static compressive tests (Fig. 7c). Moreover, the magnitudes of storage modulus E' (~ 10 MPa in the 5(PU+GO+PUD) samples, for example) are larger than the Young's modulus E (~ 5 MPa for the same type of multilayer foams) that further confirms the reliability of our measured results. The E' of the $n(\text{PU}+\text{GO}+\text{PUD})$ samples are obviously smaller than those of the $n(\text{PU}+\text{PUD})$ specimens, especially when the coating layer n reaches three or higher. Unlike the reversion phenomenon of E versus the number of coatings (Fig. 7c), this behavior for E' seems to disappear here (Fig. 10c). Combined with the

variation of the strength (Fig. 7d), the transition of the dependence of the storage modulus versus the number of nano-ink coatings tends to shift to lower numbers of coating layers and finally disappears with the decrease of the compressive strain of the foams (10% for σ_{10} , 2-3% for E and 1% for E'). The deformation responses of the 5(PU+GO+PUD) foams under different compressive strains (Fig. S8) show that the ligaments most endure axial compression/transverse shear (before elastic bending/buckling) under small strain (less than 5%) and bending/buckling under large strain (more than 15%). The ligaments of the foams for the DMA test under 1% strain (less than 5%) therefore tend to be subjected to the compression/shear deformations. As a result, the GO nanoplatelets are contracted in the struts of the compressed foams for all different coating layers and play a weakening role to reduce the values of the storage modulus. The reduction becomes more obvious because of the higher content of GO nanoplatelets in the foams when larger numbers of coating layers are applied. Despite all this, the 5(PU+GO+PUD) foams still exhibit a 670% enhancement of the specific storage modulus (modulus divided by the density) compared with the pristine PU foams (Fig. 10c, inserted).

Similarly, the variation of the damping $\tan \delta$ values for the multi-layer coated foams (Fig. 10d-f) also consists well with that of the quasi-static loss factors η (Fig. 9). The $\tan \delta$ of n (PU+PUD) samples versus the numbers of coating layers present an increase-decrease trend and the decrease is mainly caused by the substantial increase of the storage modulus, as also observed by Zhang *et al* [11]. For the n (PU+GO+PUD) foams, both the loss factors $\tan \delta$ and the relative enhancements ($\Delta \tan \delta$) relative to the n (PU+PUD) foams increase continuously with the number of coating layers. This means that the GO core coatings help to enhance the vibration energy dissipation notwithstanding the decrease of $\tan \delta$ provided by the PUD coatings in samples with large numbers of coating layers ($n > 3$). The $\tan \delta$ of the 5(PU+GO+PUD) samples with an extreme low GO content (an equivalent 0.12wt% in the five-coated foams) is 76% higher than the one of the 5(PU+PUD) specimens.

3.2.6 Energy dissipation mechanisms of foam skeletons with multi-layer sandwich coatings.

The energy dissipation mechanisms related to the quasi-static and dynamic damping for the foams are depicted in **Fig.**

11. There are three major cyclic deformation behaviors of the struts under dynamic loading: a) the bending/buckling for

slender (high slenderness ratio) ligaments, b) the axial compression/transverse shear for stubby (low slenderness ratio) ligaments, and c) the secondary torque/twisting for all ligaments [34-35]. The single ligament of the foams in this work presents a coaxial multilayered sandwich structure with the multiple quasi-continuous GO coatings and the PU matrix layers distributed spacing each other at the microscale (Fig. 11b, d). The multilayered PU matrix locates in a surface depth of the ligaments and experiences a varying degree of cyclic micro-deformation for different layers under dynamic loading [11,24,47], which can efficiently activate the optimized interfacial frictions of GO-GO and GO-PU (Fig. 11e, f). Considering the preexisting 3D network struts of the pristine PU foams, the engineered nano-foams present here a multiple continuous 3D network configuration with the interpenetrated GO/PU phases along with the skeletons. This type of foam can not only dissipate energy through the interfacial slidings of GO-GO and GO-PU, but can also preserve the continuity of the PU matrix that maintains its intrinsic damping capacities [48]. The interfacial slidings and the interpenetrated GO/PU phases are in a synergistic way [49] to contribute to the remarkable energy dissipation performance of the foams, with a large 76% damping enhancement with only 0.12wt% GO content. For comparison, Wang *et al.* [50] reported an impressive damping enhancement of 53% for graphene/nickel foams with a 0.24wt% graphene coating by chemical vapor deposition (CVD). However, since no constrained top-layers are present, the damping enhancement is less efficient compared with that in our work and the exposed GO coatings featured in Wang's work result in many microcracks produced after several cyclic compressive tests [51]. Furthermore, the designed architectures here are dissimilar from the microstructure with uniformly dispersed GO, in which GO sheets can reduce the crystallinity of the hard segment of the PU and confine the mobility of the soft segment that results in the limited or decreased damping capability of those nanocomposites [52]. For example, the damping capacities in the cellular foams with 0.1wt% graphene uniformly embedded in the skeletons merely exhibits an enhancement of 18% [35].

The results of this work make apparent the fact that the energy dissipation capacities (quasi-static and dynamic damping loss factors) and the load-bearing mechanical properties (Young's modulus, storage modulus and strength) are driven by different deformation mechanisms at microscale level. The load-bearing mechanical characteristics depend upon the

interfacial bonding strength between GO-PU and the in-plane loading modes of the GO nanoplatelets, which behave differently for different slenderness ratios of the struts. The stretched nanoplatelets combined with strong interfaces enhance the mechanical properties of the foams with low numbers of coating layers because of the bending/buckling of the slender struts. The contracted (shrinked) nanoplatelets weaken the mechanical performance of the foams made of large numbers of coating layers due to the compression/shear of the stubby struts. On the contrary, the energy dissipation performance considerably relies upon the interfacial sliding of GO nanoplatelets and the cyclic deformation-recover processes. The multilayer-decorated PU matrix under cyclic micro-deformations can drive the cyclic interface slides of the sandwiched GO nanoplatelets [24], regardless of the slenderness ratio of the struts. Thus, the loss factors η and $\tan \delta$ of the $n(\text{PU}+\text{GO}+\text{PUD})$ samples are larger than those of the $n(\text{PU}+\text{PUD})$ specimens for all different numbers of coating layers considered here. As an example, the Young's and storage moduli of the $5(\text{PU}+\text{GO}+\text{PUD})$ samples are smaller than those of the $5(\text{PU}+\text{PUD})$ specimens, however their loss factors η and $\tan \delta$ are larger. This is because the high loss factor polymeric damping materials usually do not possess large high modulus, which commonly goes against the damping capacity [18,19]. Although the GO coatings weaken the Young's and storage moduli of the $5(\text{PU}+\text{GO}+\text{PUD})$ samples, the corresponding enhancements of the moduli due to the presence of PUD coatings are still 310% and 670% compared with the pristine PU foams. This can guarantee the strong elastic recovery of the foams under vibration and dynamic loads, which is very important for the continuous cyclic interfacial sliding of the GO nanoplatelets to strengthen efficiently the energy dissipation performance.

4 Conclusions

In this work, PU foam skeletons with multi-layer sandwich GO coatings are successfully designed and fabricated by using a modified dip-coating method. The GO coatings work as the core layers of the sandwich architecture covering the whole surfaces of the foam skeleton and provide a satisfying energy dissipation mechanism of the GO-GO and GO-PU interfacial micro sliding under cyclic deformation. When compared with the counterpart foams without GO layers, the

energy dissipations (loss factors) of composite foams with only ~0.12wt% GO content increase by 52% and 76% respectively from quasi-static cyclic compression tests and DMA tests. These enhancements are attributed to the inner energy dissipation mechanisms performed by the optimized interfacial frictions of GO-PU and GO-GO and the synergistic effect of the multiple interpenetrating structure of GO/PU coating phases. Besides, the introduction of the PUD coating layers significantly enhances the Young's modulus (310%) and strength (490%) of the foams compared with the pristine PU foams. This work provides an effective route to design and manufacture graphene nanocomposite foams for damping applications and gives insightful analyses for the structural mechanisms of this material under static and dynamic load.

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Fig. 1. a) Schematic diagram of the preparation process of PU composite foams with skeletons of multi-layer sandwich structure. b) AFM image of the as-prepared GO nanoplatelets and c) height profile pattern derived from line 1, revealing that the nanoplatelets are $\sim 10 \mu\text{m}$ in lateral size and $\sim 6 \text{ nm}$ in thickness. d) SEM image showing the cellular macrostructure of the pristine PU foams, the inserted image shows the cross-section of one of the foam struts.

Fig. 2. SEM images of a) PU foam struts coated with GO inks of different concentrations, a₁) pristine PU, a₂) 0.05wt%, a₃) 0.1wt%, a₄) 0.2wt%, a₅) 0.3wt% and a₆) 0.4wt%, the rougher surfaces with the increasing concentrations of coating inks demonstrate more GO nanoplatelets attached on the surface. The surface morphologies of the struts coated with 0.2wt%GO-ink under b) low and c) high magnifications, showing that the unfolded GO nanoplatelets are quasi-uniformly coated on the whole struts of the foams and form an entangled coating network with some sparsely uncovered PU areas.

Fig. 3. SEM images of the sandwich structural struts of the foams. a) The smooth surface of the PUD coating and b) cross-section of PU+PUD samples brittlely fractured by an external force after liquid nitrogen treatment. Cross-sections of PU+0.2wt%GO+PUD samples c, d) brittlely fractured after liquid nitrogen treatment and e, f) gradually fractured at room temperature.

Fig. 4. The characterizations of the mechanical, quasi-static energy dissipation and dynamic damping properties of the sandwich structural foams coated with GO inks of various concentrations. a) Stress-strain curves from compression tests and the derived b) Young's modulus (E) at 2%~3% strain and c) strength (σ_{10}) at 10% strain. d) Energy dissipations (ΔW) and loss factors (η) obtained from quasi-static cyclic compression tests. e) Storage modulus (E') and f) damping loss factor ($\tan \delta$) obtained by DMA under different frequencies.

Fig. 5. The schematic of the evolution of interfacial friction mechanisms with the increase of GO coating contents (ink concentrations). The GO nanoplatelets are sandwiched by the PU matrix (PUD is the polyurethane dispersion), and the interfacial friction can be activated for the foam skeletons under the vibration (cyclic) microscale deformations.

Fig. 6. Cross-sections of a) a ligament of 5(PU+GO+PUD) samples with brittle failure after liquid nitrogen treatment and the zoomed-in image showing the obvious five-layer sandwich structure. b) Micrograph showing the measured length and diameter of a ligament. c) Curves of lengths, diameters and length/diameters ratios of the ligaments versus coating layers (the data are the averages with error bars based on 50-60 measurements). d) Density variation of the foams with the increase of numbers of coating layers n .

Fig. 7. Stress-strain curves of a) $n(\text{PU}+\text{GO}+\text{PUD})$ and b) $n(\text{PU}+\text{PUD})$ samples under compression. The inserted figure is the enlarged stress-strain curve of pristine PU foams under large compression rate, containing three distinct and typical deformation regions. c) Young's modulus E at 2~3% strain and d) strength σ_{10} at 10% strain of the multi-layer coated foams. The inserted figures are the corresponding specific Young's modulus and strength divided by the density. Linear fittings for the relationships of e) the Young's modulus E versus ρ^2 and f) the strength σ_{10} versus $\rho^{3/2}$. The fitting lines are derived from the four data of samples with 0-3 coating layers because the values of Adj. R^2 are almost 1 (0.97-0.99). The data points for samples with 4 and 5 coating layers do deviate significantly from the linear fitting.

Fig. 8. SEM images of $n(\text{PU}+\text{GO}+\text{PUD})$ foams with different coating layers at 10% compression strain as a) pristine, b) one, c) two, d) three, e) four and f) five coating layers. Red circles show the observable deformations of the struts as the bending and buckling. The deformation schematics of supporting cell-bars under the applied compression for g) the slender struts and h) the stubby struts, and their derived loading and deforming forms of GO nanoplatelets.

Fig. 9. Quasi-static energy dissipation of foams with different coating layers.

Fig. 10. Damping properties of the foams with different coating layers. Storage modulus E' of the foams a) without and b) with GO core coatings and c) their compared storage modulus at 10 Hz, the inserted figure is the specific storage modulus divided by the density for multi-layer coated foams. Damping loss factor $\tan \delta$ of the foams d) without and e) with GO core coatings and f) their compared $\tan \delta$ at 10 Hz.

Fig. 11. Schematic of the damping mechanisms in the multilayer-coated foams. The three major cyclic deformation formats of the foam ligaments under dynamic loading as a) bending/buckling, b) compression/shear and c) torque/twisting. d) The distribution of GO nanoplatelets in the PU matrix, revealing that the multilayer quasi-continuous coatings of GO are sandwiched by the PU matrix. The optimized damping mechanism as the energy dissipations in e) GO-PU and f) GO-GO interface frictions.