Superflexible Interconnected Graphene Network Nanocomposites for High-Performance Electromagnetic Interference Shielding

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ABSTRACT: Graphene-enhanced polymer matrix nanocomposites are attracting ever increasing attention in the electromagnetic (EM) interference (EMI) shielding field because of their improved electrical property. Normally, the graphene is introduced into the matrix by chemical functionalization strategy. Unfortunately, the electrical conductivity of the nanocomposite is weak because the graphene nanosheets are not interconnected. As a result, the electromagnetic interference shielding effectiveness of the nanocomposite is not as excellent as expected. Interconnected graphene network shows very good electrical conduction property, thus demonstrates excellent electromagnetic interference shielding effectiveness. However, its brittleness greatly limits its real application. Here, we propose to directly infiltrate flexible poly(dimethylsiloxane) (PDMS) into interconnected reduced graphene network and form nanocomposite. The nanocomposite is superflexible, light weight, enhanced mechanical and improved electrical conductive. The nanocomposite is so superflexible that it could be tied as spring-like sucker. Only 1.07 wt % graphene significantly increases the tensile strengths by 64% as compared to neat PDMS. When the graphene weight percent is 3.07 wt %, the nanocomposite has the more excellent electrical conductivity up to 103 S/m, thus more outstanding EMI shielding effectiveness of around 54 dB in the X-band are achieved, which means that 99.999% EM has been shielded by this nanocomposite. Bluetooth communication testing with and without our nanocomposite confirms that our flexible nanocomposite has very excellent shielding effect. This flexible nanocomposite is very promising in the application of wearable devices, as electromagnetic interference shielding shelter.

INTRODUCTION
The graphene-enhanced polymer matrix nanocomposites have attracted ever increasing attention because of their robust mechanical, electrical, and thermal properties.1−3 The improved electrical conduction property of nanocomposites is potentially applied in electromagnetic (EM) interference (EMI) shielding field.4−9 Normally, the graphene is not easy to be directly introduced into the polymer matrix because the graphene has very poor wettability with the matrix, thus the graphene tends to be aggregated. Therefore, the functionalization strategy is widely adopted to uniformly distribute the graphene or graphene oxide (GO) into polymer matrix. However, the electrical conductivity of these nanocomposites is normally weak either because the graphene oxide is not easy to be in situ reduced or because it is not easy to form three-dimensional (3D) conductive network when the graphene loading is small.4,7,10−13 For example, Ling et al. reported that the graphene-based EMI shielding poly(ether imide) (PEI) composite exhibited an EMI shielding effectiveness (SE) of 20 dB with loading of 10 wt %.12 To further increase the EMI effectiveness, higher graphene loading of 30 wt % is added (an improved 29 dB EMI SE of polystyrene (PS)/graphene composite).16 Unfortunately, high graphene loading results in brittleness problem of nanocomposites. To address this bottleneck, it is expected that three-dimensional conducting network could be constructed even though a small amount of graphene is introduced. Recently, graphene or reduced graphene-based three-dimensional architecture or network or aerogel has been widely explored.1−10 The reduced graphene aerogel (GA) demonstrates very good electrical conduction because of electrical conducting network. However, the reduced graphene aerogel shows obvious brittleness (quite different from that of carbon nanotube (CNT) sponge3) because the van der Waals interaction (very weak) exists among graphene nanosheets. The brittleness of graphene aerogels greatly limits their real application in electromagnetic interference shielding field, which requires the excellent flexibility sometimes. So far, the brittleness of graphene aerogel has not been addressed very
Here, we propose a novel strategy to fabricate flexible nanocomposite with excellent electrical property. This novel strategy includes two steps. In the first step, we construct a three-dimensional graphene network. In the second step, the flexible poly(dimethylsiloxane) (PDMS) matrix is directly infiltrated into graphene network using vacuum technique. The nanocomposite shows an excellent electrical conductivity and a high-performance EMI SE. Our thermal reduced graphene aerogel (TRGA)/PDMS nanocomposites can be used as a flexible EMI shielding material for portable equipment and wearable devices, potentially.

**RESULTS AND DISCUSSION**

The overall fabrication procedure of the TRGA/PDMS nanocomposite is shown schematically in Figure 1a. The graphene oxide aerogel (GOA) was prepared by the freeze-drying method at first. A following chemical reduction was carried out to convert GOA to graphene aerogel (GA) with the hydrazine hydrate vapor. The GA shows inelastic property after several cyclic compressions (Figure S1, Supporting Information). Subsequently, the TRGAs were thermally annealed at 1000 °C for 3 h. Finally, the PDMS was infiltrated into GA in a vacuum chamber. During infiltration process, the GA thickness could be adjusted by applying vertical force so that the reduced graphene oxide (rGO) percentage in nanocomposite could be tuned. As a result, the nanocomposites include two concentrations (1.07 and 3.07 wt %) of reduced graphene oxide.

The microstructures of TRGA and TRGA/matrix are observed by using scanning electron microscopy (SEM). Figure 1b shows a 3D architecture with abundant interconnected pores, thus PDMS can infiltrate into the TRGA. In this way, we obtained a PDMS nanocomposite, as shown in Figure 1c, the graphene network is embedded with PDMS matrix. The resulting nanocomposite maintains the same shape and size as TRGA network (Figure 1c), indicating that the TRGA network has not been interrupted during the infiltration process, which provides the electrical conduction channels in nanocomposites.

To investigate the chemical state and crystal structure, spectroscopic analyses were adopted using X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), and Raman spectroscopy. The deoxygenation of graphene was proved by the detailed characterization of X-ray photoelectron spectroscopy (XPS). Figure 2a,b illustrates that the C 1s scan spectra of GO is composed of four carbon components with different binding energies by fitting. The four carbon components include C−C/C≡C (284.8 eV), O−C−O (286.2 eV), C≡O (287.7 eV), and COO (289.2 eV). It is clear that GO contains a large amount of oxygenated functional groups. From the spectra of GA, we can conclude that the oxygenated functional groups have been removed almost, which is evident of the disappearance of the peak at 289.2 eV (COO) and the obvious decrease of the peak at 287.7 eV (C≡O). Furthermore, the peak of C≡O (287.7 eV) disappears in TRGA spectra, which reveals that the oxygen-containing functional groups on GA have been removed completely after the thermal annealing process.
The crystal structure of layered graphene can be determined by analyzing XRD patterns. As shown in Figure 2c, the XRD spectrum of GO has a peak at 11.1°, corresponding to an interlayer spacing of GO of 8.6 Å. After chemical reduction and thermal annealing of TRGA, a new peak (indexing to 002) at 26.5° appears, indicating an interlayer spacing of 3.3 Å, which is smaller than GO. It is concluded that almost all oxygen-containing functional groups disappeared from the interlayer space of graphene, generating a well-ordered lamellar structure. Finally, there is a sharp and narrow peak at 43−45°, as shown in the XRD pattern of TRGA. Li et al. reported that the (100)/(101) peak intensity is the evidence of more ordered stacking between layers (less rotation or translation). Therefore, the XRD shows that a less defective graphene aerogel emerges after heat treatment, and almost perfect graphene with single- or few-layer sheets is obtained by our method.

Raman spectroscopy was used to detect the defects, sp² bond, and the number of graphene layers. As shown in Figure 2d, the D-band peaking at 1360 cm⁻¹ is the characteristic of disorders and defects in graphene and G-band peaking at 1600 cm⁻¹ (originating from in-plane vibration of sp² carbon atoms) is the characteristic of the number of sp² carbon domains. The intensity ratio between them (I_D/I_G) can be used to confirm the number of defects in a graphene sample. Compared to GO, the G-band of GA shifted from 1610 to 1604 cm⁻¹, and its I_D/I_G intensity ratio is increased from 0.89 to 1.03, indicating that more numerous but smaller sp² carbon domains have partially recovered during the reduction. In contrast, the I_D/I_G ratio decreased from 1.03 (GA) to 1.00 (TRGA), suggesting the repairing of defects created by the attachment of oxygen groups and the preceding reduction. These results reveal that the defects of graphene were repaired by the thermal annealing process. As well known, the well graphitization is beneficial to improve the electrical property and the EMI shielding property.

To assess the mechanical properties, the TRGA/PDMS nanocomposites were cut into thin slabs, and neat PDMS thin slabs (made by the same curing condition with TRGA/PDMS nanocomposites) were also tested as reference samples. As seen in Figure 3a, the nanocomposite and neat PDMS show different tensile stress−strain (σ−ε) curves. The nanocomposite shows obvious mechanical enhancement. The tensile strength of pure PDMS is about 1.07 MPa, and the TRGA/PDMS composite increased to 1.75 MPa with loading 1.07 wt %. It is worth noting from Figure 3b that the TRGA/PDMS nanocomposites exhibit a tensile modulus of 11.73 MPa, 12 times of that of the neat PDMS (0.97 MPa). The significant improvement of mechanical strength can be attributed to two main factors: the uniform dispersion of the graphene nanofiller and the effective load transfer between the graphene nanofiller and PDMS matrix. Figure 3c,d shows the SEM images of the fractured surface of TRGA/PDMS composites after testing, which exhibit a uniform distribution of many short graphene segments extruding from the PDMS matrix. These graphene segments could prevent the crack from propagating through the PDMS matrix, thus consume more energy during fracture. At the same time, the tensile strain of nanocomposite is still very large, up to 25%, although it is decreased compared to neat PDMS. The large strain indicates that the nanocomposite is still flexible, which could also be reflected by optical images (Figure 3e). The nanocomposite could be twisted or tied as spring-like sucker, which double confirm that the nanocomposite is still flexible. Such excellent flexibility is very useful in wearable devices.
The electrical conductivity of GA, TRGA, and nanocomposites is evaluated by current−voltage (I−V) curve. Although the I−V characteristic of GA (before reducing and thermal annealing) displays typical linear ohmic behavior (Figure S2a, Supporting Information), the value of GA is only 0.11 S/m, which is not suitable to be used as EMI material (Figure S3, Supporting Information). This is because the GA contains a large amount of chemical groups and defects. By contrast, the electrical conductivity of TRGA is up to 10.66 S/m (Figure S2b, Supporting Information), 100 times that of GA. This is attributed to the removal of functional groups and defects recovery of graphene oxide, which is in good agreement with XPS and Raman results. However, the pure TRGA is so brittle that it is easy to break, which is not beneficial to wearable devices protection. When the PDMS is infiltrated into TRGA network and forms the nanocomposite, the electrical conductivity (Figure 4a) basically remains unchanged, in comparison with the same concentration of TRGA. This implies that the electrical conductive network was not destroyed during infiltrating process. The electrical conductivity increases as the TRGA density increases. That is to say, the electrical conductivity goes up when the reduced graphene concentration increases. As shown in Figure 4a,b, with 1.07 wt % of TRGA, the electrical conductivity of TRGA/PDMS nanocomposites is 9.2 S/m. As the TRGA loading increases to 1.73 and 1.95 wt %, the electrical conductivity of the nanocomposites increases to 39.96 and 65.6 S/m, respectively. Further increasing the content of TRGA to 3.07 wt % leads the rise of electrical conductivity to 103 S/m. Obviously, this phenomenon confirms that the conjunctions between the graphene sheets increased, the more pathways for electron transport are provided.\textsuperscript{26} The electrical conductivity of nanocomposites versus graphene concentration from references is compared with our result, as shown in Figure 4c. It indicates that our nanocomposite shows obviously advantageous electrical conductivity under the same graphene concentration.

As well known, the graphene nanosheets in polymer can change the diellectric properties of composite,\textsuperscript{27,28} thus the EMI
shielding property of nanocomposites could be tuned by changing the graphene content. We measured the EMI SE of pure PDMS and nanocomposites in the microwave frequency range of 8–12 GHz (X-band). The pure PDMS without graphene is transparent to electromagnetic waves and exhibits almost no shielding ability.29 Figure 5a shows the EMI SE of TRGA/PDMS nanocomposites at the different TRGA weight contents when the sample thickness is 2 mm. The EMI SE curve basically does not fluctuate along with the measured frequency range and the average EMI SE value is regarded as the EMI shielding effect usually. Even though the TRGA content is only 1.07 wt %, the average EMI SE value of composite can still reach to 30 dB, which implies that this nanocomposite is a very good candidate for EMI application. When the graphene loading is 3 wt %, the EMI shielding performance reaches 54 dB. Figure 5b shows the EMI SE of TRGA/PDMS nanocomposites at the different TRGA weight contents when the sample thickness is 0.8 mm. When the graphene loading is 3 wt %, the EMI shielding performance reaches 33 dB. Such excellent EMI performance is attributed to Table 1, which summarizes electrical conductivity and EMI shielding performance of the graphene or carbon nanotube-filled nanocomposites. It is obvious that at the same thickness (around 2 mm) of nanocomposite, our nanocomposite containing 3 wt % graphene only shows more advantageous EMI shielding performance in comparison with reported nanocomposites (the graphene content is up to 30 wt %).

When the incident EM wave arrives on the surface of material, the incident power \( P_i \) is divided into the reflected power \( P_r \), the absorbed power \( P_a \), and the transmitted power \( P_t \), corresponding to the reflectivity coefficients \( R \), transmissivity coefficients \( T \), and absorption coefficients \( A \), and their relationship can be expressed as

\[
A + R + T = 1
\]

The total EMI shielding effectiveness \( \text{SE}_{\text{total}} \) is the sum of absorption \( \text{SE}_a \), reflection \( \text{SE}_r \), and multiple internal reflections \( \text{SE}_m \), and the equation expresses as follow

\[
\text{SE}_{\text{total}} = -10 \log \left( \frac{P_i}{P_t} \right) = \text{SE}_a + \text{SE}_r + \text{SE}_m
\]

\[
\text{SE}_r = -10 \log (1 - R)
\]

\[
\text{SE}_a = -10 \log (T/1 - R)
\]

When \( \text{SE}_{\text{total}} > 10 \) dB, \( \text{SE}_m \) can be ignored.7 And the \( R \) and \( T \) coefficients can be expressed in terms of scattering parameters \( S_{11}, S_{22}, S_{21}, \) and \( S_{12} \), which are directly output from the vector network, expressed as follows

\[
R = |S_{11}|^2 = |S_{22}|^2
\]

\[
T = |S_{21}|^2 = |S_{12}|^2
\]

As known the value of \( R \) and \( T \), absorption coefficients \( A \) can be calculated by the former equation.
To analyze the EMI shielding mechanism of the TRGA/PDMS nanocomposites, $SE_{\text{total}}, SE_{\alpha}$, and $SE_{r}$ vary as the filler loading varies at 9 GHz in Figure 5c. As the filler increases, both $SE_{\text{total}}$ and $SE_{\alpha}$ improve significantly, whereas $SE_{r}$ increases slowly. When the TRGA content is 1.95 wt %, $SE_{\text{total}}, SE_{\alpha}$, and $SE_{r}$ are 43.6, 37.3, and 6.3 dB, respectively. It is noted that the $SE_{\alpha}$ is much higher than the $SE_{r}$ in the TRGA/PDMS nanocomposites. The results confirm that the TRGA/PDMS nanocomposites are the absorption-dominant shielding materials\cite{13,29} (Figure S4, Supporting Information). Additionally, the increased $SE_{\alpha}$ can be attributed to the increased electrical conductivity due to increasing graphene content. On the other hand, the 3D interconnected network structure of graphene formed amounts of multifaceted cells, and graphene can be regarded as the cell walls, so the EM waves can be reflected many times in our composites. As indicated in Figure 5d, the microscale cells in the matrix could attenuate the incident EM waves by reflecting and scattering between the cell walls. The nanocomposites have a large amount of interface resulted from the interconnected graphene nanosheet network, and the interfacial polarization could consume the EM waves.\cite{31,32} The EM waves were difficult to escape from the sample before being absorbed and transferred to heat.\cite{11,33} As a consequence, the $SE_{r}$ is attributed to the reflected EM only occurring on the surface of our nanocomposites. The interconnected graphene nanosheets have great attribution to the improved electrical conduction, which is highly related to electron hopping.\cite{33} According to the formula of microcurrent network, electron hopping will form microcurrent networks, resulting in conduction loss. The relaxation loss ascribed to the dipole polarization is another loss mechanism. The dipole polarization

Table 1. Comparison of EMI Shielding Performance of Graphene or CNT Nanocomposites

| composites                        | content (wt %) | thickness (mm) | EMI SE (dB) | frequency | conductivity (S/m) | ref  |
|-----------------------------------|----------------|----------------|-------------|-----------|--------------------|------|
| PS/rGO                            | 7              | 2.5            | 45.1        | 8–12 GHz  | 43.5               | 11   |
| PEI/rGO foam                      | 10             | 2.3            | 22          | 8–12 GHz  | 0.001              | 12   |
| PS/rGO                            | 30             | 2.5            | 29          | 8.2–12.4 GHz | 1.25             | 16   |
| S-doped rGO/PS                    | 15             | 2              | 24.5        | 12–18 GHz | 33                 | 5    |
| epoxy/rGO                         | 15             | 2              | 21          | 8.2–12.4 GHz | ≥5              | 13   |
| polyurethane/single-walled nanotube (SWNT) | 20             | 4.5            | 30          | 200–2000 MHz | 2              | 14   |
| poly(methyl methacrylate)/SWNT    | 20             | 4.5            | 30          | 200–2000 MHz | 2              | 14   |
| epoxy/SWNT                        | 15             | 2              | 23–28       | 8.2–12.4 GHz | 20              | 15   |
| PDMS/TRGA                         | 3.07           | 2              | 54.26       | 8–12 GHz  | 103                | this work |
arises from functional groups, defects, and multiple interfaces on the 3D graphene/PDMS nanocomposites.\textsuperscript{35}

We have conducted an experiment to measure the shielding effectiveness of our nanocomposite. The illustration is shown in Figure 6. The Bluetooth adapter is connected with the PC and then the radio signal strength is measured by testing software (BlueSoleil). The green bar in testing software represents the signal strength. The Bluetooth headset is covered by polypropylene (PP) box and nanocomposite during headset playing. When the Bluetooth headset is covered by PP box, the signal on PC is very strong. This means that the communication is excellent and there is no shielding. When the headset is covered by our nanocomposite, the signal strength is very weak due to shielding effect. This means that our nanocomposite has very excellent shielding property.

\section*{CONCLUSIONS}

We have fabricated a flexible, excellent electrical conductivity, and high-performance EMI shielding PDMS nanocomposites by impregnating PDMS into a highly porous graphene aerogel. The 3D network structure still remains after PDMS infiltration. The nanocomposite is so superflexible that it could be tied as spring-like sucker. Only 1.07 wt % graphene significantly increases the tensile strengths by 64\% as compared to neat PDMS. When the graphene weight percent is 3.07 wt \%, the nanocomposite has the more excellent electrical conductivity up to 103 S/m, thus more outstanding EMI shielding effectiveness of around 54 dB in the X-band are achieved.

Our flexible nanocomposite shows very excellent shielding effect from the Bluetooth communication testing. This flexible nanocomposite is very promising in the application of wearable devices, as electromagnetic interference shielding shelter.

\section*{EXPERIMENTAL METHODS}

Natural graphite powder (325 mesh) was provided by Alfa-Aesar and poly(dimethylsiloxane) (PDMS) was supplied by Dow Corning Co., Ltd. (China). KMnO\textsubscript{4}, H\textsubscript{2}SO\textsubscript{4} (98\%), H\textsubscript{3}PO\textsubscript{4} (85\%), H\textsubscript{2}O\textsubscript{2} (30\%), and HCl were purchased from Sinopharm Chemical Reagent Co., Ltd. (China). All these reagents were used without purification.

\textbf{Preparation of Graphene Oxide (GO).} Graphene oxide used in this work was synthesized according to the procedure reported elsewhere and described as follows. The concentrated H\textsubscript{2}SO\textsubscript{4}/H\textsubscript{3}PO\textsubscript{4} (360:40 mL) was added to a mixture of graphite flakes (3 g) and KMnO\textsubscript{4} (18 g), and then the reaction was heated to 50 °C for 11 h. After that, the mixture was cooled to room temperature and poured into ice bath. Then, further treated with 30\% H\textsubscript{2}O\textsubscript{2} (4 mL). The suspension was filtrated. And the remaining solid materials were first washed with 10\% HCl and then deionized water for three times, followed by centrifugation (8000 rpm for 1.5 h). The washing procedure was repeated several times. Then the GO suspension was gained.

\textbf{Preparation of Thermal Annealing Graphene Aerogel (TRGA).} Graphene oxide aerogel (GOA) was fabricated directly by the freeze-drying method. Typically, GO suspension (10
mg/mL) was continually stirred for 2 h in a beaker and then poured into the desired mold followed by freeze-drying for 72 h. The GOA was chemically reduced by hydrazine vapor at 90 °C for 24 h, followed by vacuum-drying at 160 °C for 24 h, then graphene aerogel (GA) was obtained. To tune the density, the GAs with different thicknesses were pressed to the same thickness. During press procedure, GAs were putted in a metal mold and pressed several times to a desired thickness. Subsequently, GAs were subjected to thermal annealing at 1000 °C in argon atmosphere for 3 h, then thermal annealed GA (TRGA) was obtained.

Preparation of TRGA/PDMS Nanocomposites. The PDMS solution was prepared by mixing PDMS base agent, curing agent in the ratio of 10:1, followed by degassing in a vacuum oven for 25 min. The TRGA/PDMS nanocomposites were fabricated by infiltrating the TRGA with PDMS under vacuum with repeated vacuum pumping for 10 times at time intervals of 20 min. Finally, the nanocomposites were thermally cured at 80 °C for 3 h.

Characterization. The morphologies and structures of the GA and TRGA/PDMS composites were characterized by field-emission scanning electron microscopy (Carl Zeiss, Supra 55). The electrical conductivity was measured by the two-probe method using an electrochemical workstation (PAR-STAT4000). The EMI shielding effectiveness was measured in the frequency range of 8–12 GHz, using an Agilent N5234A vector network analyzer. The composites were cut to fit the waveguide sample holder for EMI SE measurement. The tensile properties were tested at a crosshead speed of 0.5 mm/min, by Instron 5944. The geometries of sample used for tensile testing were 0.5 cm thickness, 1.5 cm width, and 10 cm length.

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