Enhanced dispersion of carbon nanotube in silicone rubber assisted by graphene

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We report a novel, scalable and inexpensive approach to fully disperse carbon nanotubes in silicone rubber by the addition of graphene. In comparison to graphene, the dispersion of multi-walled carbon nanotubes (MWNs) in silicone rubber matrix is extremely difficult although both of them possess similar physical structure. The different dispersion behavior of graphene and MWNs could be contributed to the difference in their interaction with polymer matrix and their geometry. Based on SEM, TEM and XRD analysis, we find that the dispersion of MWNs in silicone rubber is dramatically improved by the addition of graphene. Graphene acts as a compatilizer since it shows strong interaction with both polymer matrix and MWNs. This method provides a simple route to enhance the dispersion of carbon nanotubes and improve the electrical property of the polymer composites. The synergic effect of the hybrid materials may not be limited to the applications in polymer composites.

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1. Introduction

Since the discovery of carbon nanotubes (CNTs) [1] and graphene [2], they have received increasing interest in various fields such as chemistry, physics, materials science, and electrical engineering [1–16]. Both of the carbon materials, possess unique nanostructures with remarkable physical and mechanical properties [1–8]. They are widely used as nanofillers to improve electrical, thermal and optical properties of polymer composites [9–12]. The prospect of obtaining advanced nanocomposites with multifunctional features and materials used for structures and electrical conductors has attracted researchers’ attention in both academic and industrial communities [13–16], especially as electro statically dissipative materials and aerospace structural materials [17–20].

Although the research on carbon nanotube-based polymer composites has achieved significant progress, two major issues remain challenging for the fabrication of carbon nanotube reinforced polymer composites: (1) the dispersion of CNTs, i.e. the tendency of nanotubes to aggregate during processing; (2) the limited availability of high-quality carbon nanotubes in large quantities and high cost of their production. Graphene, a hexagonal network of carbon atoms, is actually the basic unit of a carbon nanotube. Rolling graphene into a seamless, hollow cylinder forms the carbon nanotube [21,22]. In addition, although graphene and carbon nanotubes are similar in chemical composition, different topology distinguishes carbon nanotubes from graphene and offers them unique properties. One of the major advantages of graphene over carbon nanotubes lies in the precursor of graphene—graphite, which is natural abundant [23]. It is essential for industrial applications in composite fields, since high yield production and a cost-effective and scalable process is possible [24,25].

The investigations on graphene or carbon nanotube-based polymer composites are mainly focused on plastics matrices such as polyamide 6 (PA6) [26], polycarbonate (PC) [26,27], epoxy [28], polyurethane (PU) [29], polystyrene (PS) [30,31], poly (methyl methacrylate) (PMMA) [32,33] or poly (styrene-co-isoprene-co-styrene) (SIS) block copolymer [34]. There are only a few of studies on graphene or CNTs/rubber composites [35–38]. One important reason is that it is difficult for nanoparticles to be dispersed in rubber matrices because of their high viscosity or low surface energy [36]. Conductive rubbers are critically needed in the fields such as ESD (electrostatic discharge) and EMI (electromagnetic interference) shielding materials, electronics packaging, telecommunication antenna, mobile phone parts and frequency shielding coatings for aircraft and electronics [28,35–38]. However, the physical interaction between rubbers and nanoparticles are weaker than that between plastics and nanoparticles. It is of importance to understand the mechanism of the dispersion of nanoparticles in rubbers.
Silicone rubber offers good tolerance to extremely low and high temperatures, being able to operate normally from \(-55\) °C to \(+300\) °C. The carbon backbone of organic rubbers makes them susceptible to ozone, UV, heat and other aging factors while silicone rubber can perform well. This feature makes silicone rubber become one of the choices of elastomers in many extreme environments. Silicone rubber also has other superior properties such as the ease of changing its shape and filling the seals in a matrix. Therefore, silicone rubber was selected as the polymer matrix in this paper. One critical issue for the fabrication of nanocomposites is how to uniformly disperse nano fillers in polymer matrix. Many methods including physical and chemical methods were investigated to assist in the dispersion of CNTs [39]. Many reports investigated the synergistic effect of different fillers to make balance between properties and cost. Graphite (G) together with carbon fibers (CF) in polymer composites [40–47] has been studied for many years and excellent electrical conductivity of the composites has been obtained. Carbon nanotubes also have been investigated when blended with other fillers such as clay [48,49], single-walled carbon nanotubes (SWNTs) [50], graphite (G) and carbon black (CB) [51]. Nanoplatelets/carbon black/carbon nanotubes three-system in epoxy resin has also been reported [52]. Balberg et al. [53,54] proposed models to explain electrical percolation based on excluded volume theory. Sun et al. [51] extended the excluded volume theory and obtained an equation to predict the amount of fillers used to get the percolation when two fillers were used. It is commonly believed that two (or three) types of fillers can form co-supporting conductive networks in which the fibrous CF filler act as long distance charge transporters and the particulate fillers such as CB serves as an interconnection between the fibers by forming local conductive paths [42,43]. However, the dispersion of nanofillers in a synergetic system is rarely explored.

In this work, we report a simple and effective way to dis perse carbon nanotubes and graphene in silicone rubber. The excellent dispersion of CNTs can be achieved with the aid of graphene because of strong interaction between graphene and silicone rubber matrix as well as strong interaction between graphene and CNTs. The mechanism of dispersion will be discussed in detail in this paper.

2. Experimental section

2.1. Materials

Silicone rubber (Methyl-Vinyl-Silicone, MVQ 110-2) was provided by Jiangsu Hongda Materials Company, China. The molecular weight of the transparent silicone rubber is about 4.50–5.9 × 10⁶ g/mol. Natural flake graphite (LC50-9999, particle size 40 μm) was purchased from Qingdao Ruisheng Graphite Co. Ltd. China. Carboxylated multi-walled carbon nanotubes (MWNTs, MC5, purity > 95%, OD: 20–30 nm, Length: 10–30 μm, -COOH content: 1.23 wt%, resistivity: 2.8 Ω cm) was purchased from Chengdu Institute of Organic Chemistry Co., Ltd. China. Curing agent 2,5-dimethyl-2,5-di(tert-butyl peroxy) hexane (AkzoNobel, Tianjin), is commercially available and used as received.

2.2. Preparation of graphene

Graphite oxide (GO) was prepared according to the modified Hummers method first [55]. 5 g of flake graphite (40 μm), 3.75 g of NaNO₃, and 230 mL of H₂SO₄ were mixed at 0°C. 15 g of KMnO₄ was slowly added into the mixture to give a mixture solution with pH value was close to 7, followed by stirring and drying in a vacuum oven for one week. The dried GO samples were thermally treated at 1050°C for 5 min in a tubular furnace to obtain thermally reduced graphene (TrG). The detailed process and properties of graphene can be obtained elsewhere (see reference 10).

2.3. Preparation of graphene/carbon nanotubes/silicone rubber composites

The rubber composites were fabricated as follows: (1) Silicone rubber (MVQ, before crosslinking) was dissolved in THF to obtain homogeneous solution. (2) Graphene (or MWNTs) was dispersed in THF. The mixture was sonicated for 1 h to get stable dispersion. (3) The above (1) and (2) suspensions were mixed together and sonicated for 30 min to obtain homogeneous suspension. (4) THF was evaporated at 50°C, and the mixture was dried in vacuum oven. (5) Curing agent was added into the mixture, and then the resulting compound was moved to vulcanizing machine for curing followed by Vulcanizing at 160°C for 12 min to obtain the composites.

2.4. Characterizations

2.4.1. X-ray diffraction (XRD)

X-ray diffraction measurements (D-MAX 2500/PC, 18 kW rotating anode, Rigaku Corporation) were carried out to characterize the structure of graphene, and the composites with Cu Kα radiation (40 kV, 100 mA, wavelength, 1.5406 Å). X-ray diffraction patterns were obtained at room temperature with a scanning rate of 4°/min and the step size of 0.02°.

2.4.2. Scanning electron microscopy (SEM)

Cryogenic fracture surfaces of the specimens were coated by sputtering with gold and then observed with field emission scanning electron microscopy (FESEM, Japan Electronics Co., Ltd., JSM-6700F).

2.4.3. Transmission electron microscopy (TEM)

Ultrathin specimen (thinner than 100 nm) for TEM observation were cryogenically cut with a diamond knife using an ultra thin microtome (UC7, Leica Ultracut, EMFC7) and collected on 200-mesh copper grids. The exfoliation of graphene nanosheets in silicone rubber was observed with transmission electron microscope (TEM, JEOL, JEM2100).

2.4.4. X-ray photoelectron spectroscopy (XPS)

XPS experiments were carried out on a RBD upgraded PHI-5000C ESCA system (Perkin Elmer) with Mg Kα radiation (hv = 1253.6 eV) or Al Kα radiation (hv = 1486.6 eV). Binding energies were calibrated by the containment carbon (C1s = 284.6 eV). The data analysis was carried out using RBD AugerScan 3.21 software (RBD Enterprises, USA).

2.4.5. Resistivity analysis

PC-68 high resistance meter (Shanghai Baiye Instrument Technology Co., Ltd, China) was used to test the resistance of silicone rubber composites. The resistivity was calculated through the formula:

\[ \rho = \pi d^2 R / \ln2 \]
where \( \rho \) is resistivity, \( d \) is the thickness of the sample, and \( R \) is the resistance of the sample.

2.4.6. Thermal diffusivity

Thermal diffusivity was measured by NETZSCH LFA 447 at 25 °C. Xenon lamp as heating source launched a bunch of pulse to the surface of sample, the increasing temperature was measured by infrared detector.

3. Results and discussion

3.1. Characterization of graphite oxide (GO) and thermally reduced graphene (TrG)

FTIR, XRD and XPS were used to characterize graphite oxide (GO) and thermally reduced graphene (TrG). TrG was prepared as reported previously [10]. As shown in Fig. 1(a), the band at 1732 cm\(^{-1}\) appeared in the FTIR spectrum of GO, indicating that carbonyl groups (vibration modes of C=O at 1732 cm\(^{-1}\) and C–O–C at 1065 cm\(^{-1}\) were anchored on GO during the oxidation of graphite. The oxygen-containing groups of TrG almost disappeared, and the vibration peaks of graphene (~1385, ~1446, and ~1635 cm\(^{-1}\)) appeared, indicating that GO was fully reduced to TrG after thermal treatment.

XRD (Fig. 1(b)) pattern shows that the interlayer spacing of GO (0.794 nm, 11.13°) was larger than that of pristine graphite (0.335 nm, 26.6°) [56] because of the intercalation of functional groups on GO. After thermal reduction of GO, the diffraction peak (11.13°) of GO disappeared and a broad diffraction peak of about 25° appeared, implying that the distance between the intercalated graphite layers decreased and the layer structure was reconstructed during thermal reduction although remarkable amount of graphene was fully exfoliated by thermal expansion [57].

Fig. 1. The characterization of GO and TrG, (a) FTIR spectra of GO and TrG, (b) XRD profiles of GO and TrG, (c) C1s XPS spectrum of GO, (d) C1s XPS spectrum of TrG, (e) SEM image of GO, (f) TEM image of TrG.
X-ray photoelectron spectroscopy (XPS) was used to probe functional groups on graphite oxide and thermally reduced graphene. C1s XPS spectrum of GO (Fig. 1(c)) shows the existence of oxygen-containing groups (C=O at 286.2 eV, and C=O at 287.8 eV) due to the oxidation of graphite. The doublet area for C−C (284.5 eV) in the XPS spectrum of GO accounts for about 64 percent of total peak area, indicating that graphite has been fully oxidized. C1s XPS spectrum of TrG (Fig. 1(d)) shows that the peaks of C=O and C=O have dramatically decreased after 5 min pyrolysis reduction at 1050 °C, but a few of C=O and C=O groups were still retained on graphene, which can not be detected in the FTIR spectrum of TrG. These oxygen-containing groups play an important role in the dispersion of graphene.

Fig. 1(e) is the SEM image of GO, and Fig. 1(f) is the TEM image of TrG. It can be seen that the surfaces of GO and TrG are not smooth but wrinkled. The original sp² bonding of two-dimensional graphene evolved into a sp³ hybrid non-planar bonding, resulting in the disruption of the original plane structure. As a result, a wrinkled morphology was formed. This morphology is very favorable for the dispersion of graphene in rubber, which will be discussed in the following section.

3.2. Dispersion of multi-walled carbon nanotubes (MWNTs) in silicone rubber

The agglomeration of carbon nanotubes (labeled by white circle) in silicone rubber/carbon nanotube (5 wt%) were observed by SEM and TEM as shown in Fig. 2 (a) and (b). Although ultrasonic treatment of carbon nanotubes have been carried out during the preparation of MWNTs/silicone rubber composites, MWNTs still can not be fully dispersed in the polymer matrix due to intrinsic strong attractive forces (Van der Waals force) of carbon nanotubes.

Fig. 2 (c) shows XRD profiles of MWNTs, MVQ and MVQ/MWNTs (5 wt%) composite. The diffraction peaks of MWNTs were observed at 24.4° (002) and 43.9° (100), which were assigned to interlayer space in the radial direction and in-plane graphitic structure of MWNTs, respectively. The MVQ showed a diffraction peak at 12.1°. In the XRD profiles of MVQ/MWNTs composite, the relative intensity and peak position of diffraction peak at 12.1° were almost not changed and the peaks at 24.4° and 43.9° were very weak, which indicated that the MWNTs were contained in the nanocomposite and did not influence the structure of MVQ.

3.3. Dispersion of graphene in silicone rubber

SEM, TEM and XRD of silicone rubber/graphene (3 wt%) composite were shown in Fig. 3. Fig. 3 (a) and (b) showed SEM images of silicone rubber/graphene composite. It could be clearly seen that almost all of graphene was well dispersed in the silicone rubber matrix, consistent with the results of TEM observation of silicone rubber/graphene composites (Fig. 3 (c)). The XRD profile of graphene (TrG) (Fig. 3 (d)) exhibited a broad diffraction peak at about 25.4°. When graphene was blended with silicone rubber (1 wt% or 3 wt%), the diffraction peak of TrG in those composites disappeared. It implies that graphene was almost exfoliated by silicone rubber and very well dispersed in the silicone rubber.

Graphene and carbon nanotube have similar sp² chemical structure, but they show dramatically different dispersion behavior in silicone rubber. The main reasons can be found in the following two aspects:

The first aspect is that the morphology of the two carbon materials is quite different. Fig. 3 (b) is the amplification of the region of black dashed circle in Fig. 3 (a). We can find that the surface of graphene is not flat. Instead, a worm-like wrinkled morphology of graphene was observed. The original sp² bonding of two-dimensional plane graphene evolved into a sp³ hybrid non-planar bonding, resulting in the disruption of the original plane structure. As a result, a wrinkled morphology was formed. This worm-like structure can prevent the restacking of graphene layers when mixing with silicone rubber. The ripples of graphene are very beneficial for the formation of occluded rubber in the silicone rubber composites, which limits the movement of molecular chains of silicone rubber. As a result, the strength and hardness of the graphene/silicone rubber composites are improved. Meanwhile, the wrinkled morphology renders the silicone rubber to be better integrated with graphene, which facilitates the dispersion of graphene in silicone rubber.

The second aspect is that the interaction between graphene and silicone rubber is much stronger than that between MWNTs and silicone rubber. Schniepp [57] reported that the surface area of graphene is about 800 m²/g as measured by BET surface adsorption. The huge surface area of graphene offers numerous active adsorption sites, which render better interaction between the graphene and silicone rubber. In contrast with MWNTs, the two planes of graphene possess more contact area with silicone rubber. Also, a few of oxygen-containing groups were still retained on graphene (XPS spectrum in Fig. 1(d)). Hydrogen bonds may form when graphene contact with the oxygen-containing groups on the silicone rubber [36]. Thus, an effective interface bonding forms between the graphene and the silicone rubber.

The mixing energy in the blend system can be described by eq. (1):

\[
\Delta G_M = \Delta H_M - T \Delta S_M
\]

When graphene or MWNTs is mixed with silicone rubber, the entropy changes little, because conformation entropy change little.

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Fig. 2. MWNTs agglomerated in silicone rubber/MWNTs (5 wt%), (a) SEM image of MVQ/MWNTs (5 wt%) (10000×), (b) TEM image of MVQ/MWNTs (5 wt%) (150000×), (c) XRD profiles of MWNTs, MVQ and MVQ/MWNTs (5 wt%) composite.
and entropy change mainly arises from the position change of graphene or MWNTs. In the mixing process, the entropy is positive, which is beneficial for the dispersion of graphene or MWNTs into silicone rubber. The main factor that prevents nanoparticles from being dispersed in silicone rubber or other matrices is the effect of enthalpy. The interaction between the nanoparticles is strong due to the huge surface energy of nanoparticles. There are three competing interactions in the system when nanoparticles are mixed with polymers. The first is the interaction for a polymer-polymer contact ($\Delta H_{p-p}$), the second is the interaction for a nanoparticle–nanoparticle contact ($\Delta H_{n-n}$), and the third is the interaction for a polymer-nanoparticle contact ($\Delta H_{p-n}$). The first and second interactions will cause the phase separation of polymer and nanoparticles, which is conducive to the aggregation of nanoparticles. The third interaction will favor the dispersion of nanoparticles into the polymer. The difference in the dispersion of graphene and MWNTs is caused by the interaction between the nanoparticles and polymers. $\Delta H_{p-p}$ and $\Delta H_{n-n}$ are almost the same for graphene and MWNTs as they have the same elements—carbon and the $sp^2$ structure of C–C are similar for both graphene and MWNTs. The difference arises from the $\Delta H_{p-n}$. Good dispersion of Graphene in polymers implies that the $\Delta H_{p-c}$ (G represents graphene) is much higher than $\Delta H_{p-c}$ (C represents carbon nanotube).

Suppose that the interactions among graphene and polymer chains are the sum of Van der Waals forces. The force is weak for only one carbon atom interacting with a silicone atom. There are numerous interactions in a sample, so the sum is huge enough. Let us compare the difference between graphene and carbon nanotubes. The elements of graphene and carbon nanotubes are the same, meaning that they have the same Van der Waals forces between the carbon elements and other elements of the polymer. The difference lies in the shape of these two nanoparticles. Both sides of a graphene plane can interact with the polymer, but only the outer layer of MWNTs can do. The total Van der Waals forces among graphene and the polymer are doubled as compared to that among the polymer and MWNTs. The total $\Delta H$ of the graphene/polymer system will be much smaller than that of the MWNTs/polymer system. As a result, it is easier for graphene to be dispersed in the polymer.

### 3.4. Graphene-assisted dispersion of MWNTs in silicone rubber

When Graphene/carbon nanotubes/silicone rubber were blended together, an interesting phenomenon was observed. Fig. 4 shows the SEM, TEM images and XRD profiles of the system. It can be seen from the SEM and TEM images (Fig. 4 (a)–(e)) that graphene and MWNTs were well dispersed in the silicone rubber. And the graphene are dispersed around carbon nanotubes. Carbon nanotubes and graphene together form a conductive network (as indicated by the black lines in Fig. 4 (a) and (b)) in the silicone rubber matrix. Graphene is well dispersed in the silicone rubber matrix, and the aggregation of carbon nanotube in the matrix was not found in SEM or TEM images.
XRD profiles (Fig. 4(f)) showed that when the graphene was mixed with MWNTs and silicone rubber (silicone rubber/graphene/MWNTs = 100/1/2.5), the diffraction peak of interlayer structure of TrG (25.4°) was not found, whereas the diffraction peak of MWNTs (24.4°) was still observed as showed in Fig. 4(f), indicating that graphene was still well dispersed in the silicone rubber matrix regardless of the presence of MWNTs. The dispersion of graphene in silicone rubber was not influenced by the presence of MWNTs. The dispersion of MWNTs can not be assessed by XRD measurement. However, the SEM and TEM images indicate that both MWNTs and TrG are well dispersed in silicone rubber matrix.

The excellent dispersion of MWNTs and graphene in the blend system may be attributed to the following reasons:

The diameter of graphene is in the range of tens of nanometer to several micrometers, while the dimension (length) of MWNT is 10–30 μm. Graphene is smaller in particle size as compared with MWNTs, and it can act as a separator between MWNTs. The force that the sonication provides results in the intercalation of graphene into nanotube bundles. A graphene sheet may “adhere” to many carbon nanotubes due to the strong interaction between carbon nanotubes and graphene, leading to the disruption of carbon nanotube bundles. Once graphene “adheres” to MWNTs, it is not

Fig. 4. Silicone rubber/graphene/MWNTs = 100/1/2.5, (a), (b) SEM images, (c) TEM image, the insert is the enlargement of carbon nanotube in the white rectangle, (d) TEM image, (e) TEM image of silicone rubber/graphene/MWNTs = 100/1/3, (f) XRD profiles of MWNT, TrG, MVQ and MVQ composites. (The arrows point to the MWNTs and graphene are in white circles).
easy for graphene to be “driven out” from MWNTs because graphene possesses the extended π electron system similar to that of carbon nanotubes.

There exists a good interaction between graphene and silicone rubber matrix. On one hand, the –OH– on graphene may form hydrogen bonds with the –O– on silicone rubber. On the other hand, there is huge contact surface area between silicone rubber and graphene because graphene has very large specific surface area [57] and each plane of graphene can contact with silicone rubber. Graphene can be dispersed uniformly in silicone rubber as discussed above.

Since graphene has good interaction with silicone rubber and carbon nanotubes, graphene plays a role of compatilizer for MWNTs and silicone rubber, which facilitates the dispersion of MWNTs in silicone rubber.

Based on the aforementioned analysis, we believe that the most important factor for the excellent dispersion of MWNTs assisted by graphene is the strong interaction between graphene and the matrix as well as the strong interaction between graphene and MWNTs. Graphene has a large surface area and carbon nanotubes have a great aspect ratio. One dimensional and two-dimensional materials cooperate, forming a “conductive bridge” in the silicone rubber as shown in the schematic diagram (Fig. 5).

3.5. Effect of the dispersion on electrical properties of silicone rubber composites

With the increase of filler content, the volume resistivity of silicone rubber decreased (Fig. 6). The percolation threshold for graphene is about 2 wt% with volume resistivity of \(1 \times 10^3 \Omega \text{cm}^{-1}\). By contrast, the percolation threshold for MWNTs is about 5 wt% with volume resistivity of \(4.21 \times 10^2 \Omega \text{cm}^{-1}\). The graphene with two-dimensional planar structure is better dispersed than MWNTs, which favors the formation of conductive network. MWNTs tend to aggregate in silicone rubber, leading to larger percolation value.

When both graphene and carbon nanotubes are filled in silicone rubber composites, carbon nanotube is well dispersed with the aid of graphene and the conductive network forms. Graphene is distributed around the carbon nanotubes. Carbon nanotubes play as a “conductive bridge” connection between graphene layers and the matrix. At the graphene content of 1 wt% and the MWNT content of 2 wt%, the volume resistivity of the composite was \(4.91 \times 10^3 \Omega \text{cm}^{-1}\). At the graphene content of 1 wt% and the MWNT content of 2.5 wt%, the volume resistivity was reduced to \(8.0 \times 10^3 \Omega \text{cm}^{-1}\).

3.6. Effect of the dispersion on thermal diffusivity of silicone rubber composites

Thermal diffusivity of silicone rubber composites was shown in Fig. 7. The thermal diffusivity of pure silicone rubber was 0.105 m²/s, which was increased by the addition of graphene or MWNTs or two fillers together. Thermal diffusivity of silicone rubber composites were 0.113 and 0.119 m²/s respectively by filling graphene (1 wt%) and MWNTs (5 wt%). By contrast, thermal diffusivity of silicone rubber composite was enhanced to 0.123 m²/s by filling 1 wt% graphene and 3 wt% nanotubes together. Graphene has strong interaction with silicone rubber but also breaks up the agglomeration of MWNTs. The remarkable enhancement is due to the synergistic effect of graphene and MWNTs.

4. Conclusion

In summary, we studied the dispersion of MWNTs and graphene in silicone rubber matrix. It is difficult to disperse MWNTs in silicone rubber alone, whereas it is much easier for MWNTs to be dispersed in the matrix with the aid of graphene. The strong interaction between graphene and silicone rubber as well as the strong interaction between graphene and MWNTs accounts for the good dispersion of MWNTs. Graphene behaves as a compatilizer. Both physical isolation between graphene and MWNTs and some hydrogen bonding between graphene and MWNTs may form during the dispersion of MWNTs assisted by graphene. Addition of
graphene provides a simple route to disperse carbon nanotube and the synergetic effect facilitates the fabrication of high performance polymer composites.

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