



Graphene Nanoarchitectonics: A New Material Horizon for Reinforcement of Sustainable Polymers

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Specialty section:

This article was submitted to
Smart Materials,
a section of the journal
Frontiers in Materials

Received: 21 April 2020

Accepted: 24 July 2020

Published: 18 August 2020

Citation:

Chen L, Zhang Y, Liu W and Liu Z
(2020) Graphene Nanoarchitectonics:
A New Material Horizon
for Reinforcement of Sustainable
Polymers. *Front. Mater.* 7:276.
doi: 10.3389/fmats.2020.00276

Using nanoscience to solve the issues that limit the development of a sustainable society is one of the most important topics at present. Sustainable polymers play an important role in the construction of a sustainable society, but its large-scale application is usually limited by the poor mechanical and shape-memory properties of the commonly used epoxy resin. The utilization of nanoarchitectures to improve the shape-memory performance of composite materials is an effective approach. Graphene, among the used reinforcement materials for epoxy resin, is undoubtedly a fascinating example. In this paper, the shape-memory and mechanical properties of graphene-modified epoxy resins are described respectively for different response types. In this mini-review, we present an overview of the research results of graphene nanoarchitectonics in the modification of shape-memory epoxy resin composites. Besides, this paper has reviewed the properties of different types of graphene/epoxy composites, looking forward to the development of graphene/epoxy composites and future prospects.

Keywords: graphene, nanoarchitectonics, composites, sustainability, polymers

INTRODUCTION

A sustainable polymer is a kind of material that can change shape and recover under certain conditions, such as heat, light, water, pH, and other external stimuli (Zhao et al., 2015; Yao et al., 2018). Shape-memory characteristics have broad application prospects in aerospace, biomedical, and other fields (Fabrizio et al., 2012; Li et al., 2012; Zhao et al., 2019). Among the sustainable polymer series materials, epoxy (EP) resin has attracted considerable attention both scholarly and popularly due to their excellent shape-memory properties, thermal properties, and mechanical properties (Liu et al., 2010).

Related researches pointed out that the interaction between nanoparticles and polymer matrix can accelerate the movement of molecular chains, improving the shape-memory performance of materials (Babaahmadi et al., 2017). Graphene has been considered as an irreplaceable reinforcing material because of its special structure and excellent properties and has great application value

in the field of composite modification. Wang et al. (2018) introduced graphene into EP–cyanate ester nanocomposites, and the results confirmed that graphene could promote the shape recovery of EP cyanate nanocomposites. It should be noted that the doped size, surface modification, and dispersion of graphene as a filler will have a significant impact on the performance of composite materials (Atif et al., 2016). For instance, functionalized graphene (FG) can obtain good bonding with the matrix, thereby enhancing the role of graphene as reinforcing filler (Choi et al., 2012). A novel slippery film with tunable wettability based on a shape-memory graphene sponge was presented by Wang et al. (2017). The shape-memory polymer was coated on the surface of porous graphene sponge, which was used to lock in inert lubricant and to construct a smooth surface to achieve good hydrophobicity. The superelasticity, high strength, and good conductivity of graphene sponge endow graphene/polymer hybrid films with good shape-memory properties. In fact, in addition to graphene, its derivatives, such as graphene oxide (GO) and thermally reduced graphene, are often used as fillers to modify composite materials (Kausar, 2019). At present, the use of graphene to modify shape-memory EP resins to prepare shape-memory graphene/EP resin composites with better shape-memory and mechanical properties has been widely investigated by scholars. Table 1 shows the literature summary of graphene-modified shape-memory EP resin. It can be seen from the table that different doping processes and dosage can improve the mechanical properties of shape-memory EP resin to a different extent. For different types of shape-memory EP resin, different graphene content can change its recovery performance correspondingly.

In this article, we summarized the recent developments of graphene nanoarchitectonics in a reinforcement of shape-memory polymers under various external stimuli like thermal, electrical, and other stimulations and discussed the problems of graphene in limiting the application of composites (Figure 1).

THERMO-ACTIVE GRAPHENE/EPOXY COMPOSITES

Thermo-induced shape-memory materials are the most widely studied type of shape-memory polymers. The shape-memory EP can be divided into the stationary phase and reversible phase, in which the stationary phase is used for shape-memory and recovery. When the temperature rises above the glass transition temperature (T_g), the material can deform under the effect of external load, and the reversible phase molecular chain is frozen when cooled, to obtain a stable morphology. When the temperature rises above T_g again, the reversible phase molecular chain recovers its activity and releases the stored strain energy, and the material returns to its original state gradually under the recovery of the stationary phase (Wang et al., 2019b; Willian and Cristina, 2020).

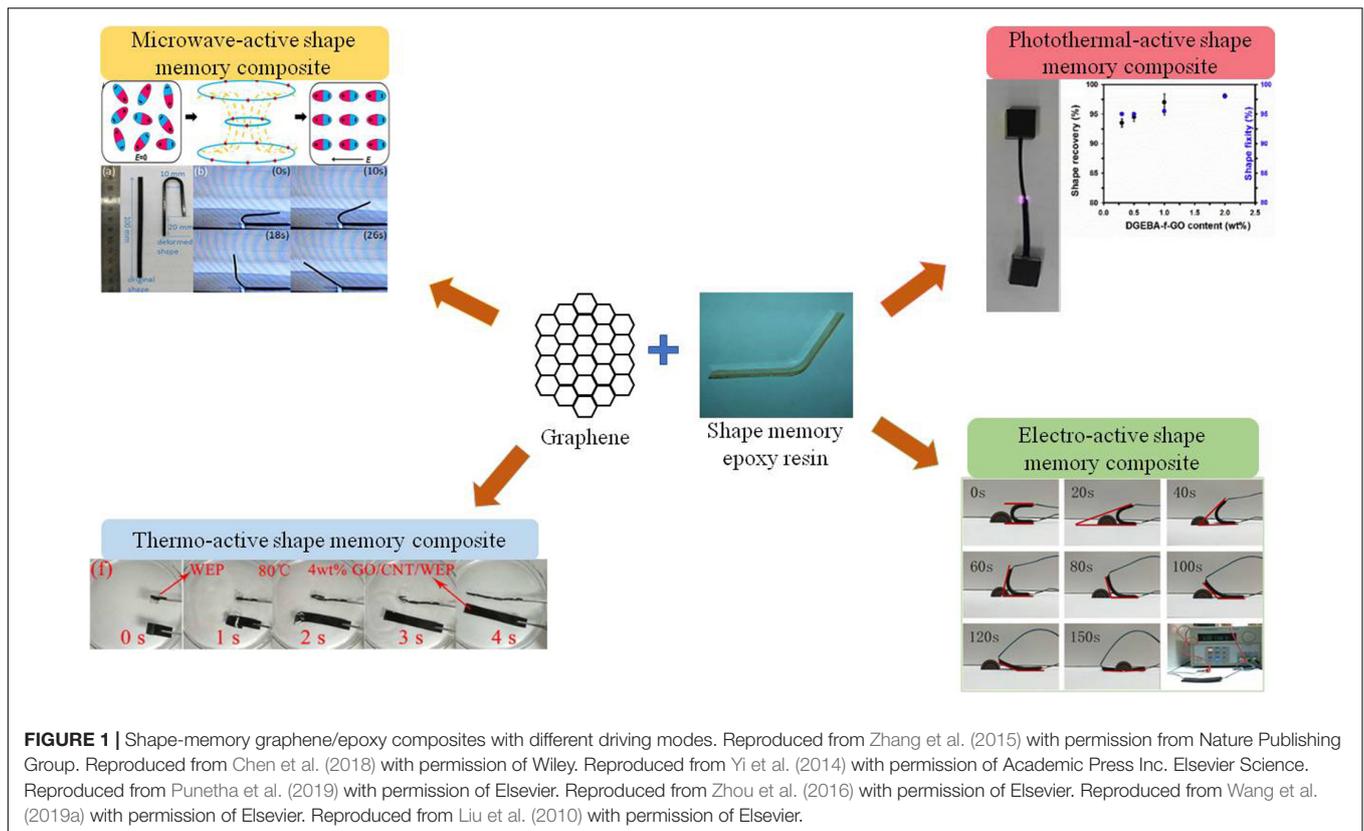
Recently, studies have shown that carbon nanomaterials can improve the thermally activated shape-memory properties of materials (Park et al., 2014; Liu et al., 2016). Ortiz et al. (2018) prepared a photocurable shape-memory EP/thiol-ene/graphene

nanoplatelete (GNP) composites that can be cured by light. By increasing the concentration of GNP and polythioethers in the co-network, the thermal conductivity increases almost linearly. According to the experimental data, the thermal conductivity of the sample with a concentration of polythioether (40% mol) and graphene (15 wt%) reaches the maximum, which is 0.39 W/mK. They also studied the shape-memory properties of the material, and the results showed that samples with 10% of GNP at all thiol-ene concentrations presented the best results in both shape recovery and shape fixity value, which was in the range of 95–99%. Meanwhile, the experimental results also found that the presence of soft domains (such as polythioethers) in the cross-linked co-network helps to enhance the shape-memory characteristics of GNP nanocomposites. GO/waterborne EP shape-memory composites were prepared by Wang et al. (2019a). In the experiments, they found that as the content of GO increases, the storage modulus of the material increases. The

TABLE 1 | Overview of shape-memory and mechanical properties of widely used graphene.

Doping methods	Graphene content	Performance	References
Ultrasonic dispersion	5–15 wt%	Tensile strength and elongation were increased by 5.3 and 33.5%, respectively	Zhang L. et al., 2016; Ortiz et al., 2018; Wang et al., 2019b
Vacuum-assisted resin transfer	2–12 wt%	Enhance the structure of the nanofiber web (increased 15%)	Fejos et al., 2013; Wang et al., 2015; Zhou et al., 2016
Solution casting	1–3 wt%	Young's modulus and tensile strength of nanocomposites prepared via R(GO-ISO) nanosheets have been improved (increased 51 MPa)	Nouri et al., 2020
Solution blending	0.1–4 wt%	Moduli of the imidized graphene nanocomposites were 25–30% higher than those of unmodified graphene nanocomposites. The addition of graphene improved the recovery rate	Yoonessi et al., 2012
Compound aerogels	0.5–0.8 wt%	Compared with pure epoxy, the electrical conductivity of the composite is improved by 13 orders of magnitude	Liu et al., 2015
Coupling reaction	2–6 wt%	A modulus of ~339 MPa and a shape recovery ratio of 98% were achieved	Zhang and Hu (2020)
<i>In situ</i> polymerization	0.5–1.5 wt%	Young's modulus of PU/GQD nanocomposites increases with the addition of GQDs content at both room temperature and 60°C	Sanaz et al., 2020; Sarabiyan et al., 2020
Condensation polymerization	0.1–1.0 wt%	The nanocomposite film containing 0.25wt% GNPs showed the highest shape recovery ratio (92.4%) since the net-point effect	Maryam et al., 2020

GQD, graphene quantum dot; PU, polyurethane; GNP, graphene nanoplatelete.



study showed that the addition of GO would improve the shape recovery rate and fixation rate of shape-memory EP materials and would increase the thermal response speed. Also, compared with pure WEP, the mechanical properties of the material have also improved. Subsequently, they also prepared GO/carbon nanotube (CNT)/WEP ternary hybrid polymeric shape-memory composites. The results show that GO and CNT show excellent synergistic enhancement effects in many aspects.

To solve the brittleness of the EP resin matrix, Zhang L. et al. (2016) used polyurethane (PU)/EP interpenetrating polymer network material as the matrix and introduced FG to prepare PU/FG/EP composites. The experimental results show that the FG has a strong binding force with the matrix. When 0.8 wt% FG was added, the tensile strength and elongation at break of the material were increased by 5.3 and 33.5%, respectively, and with the increase of FG load, the shape-memory property was enhanced continuously. When the FG content is 1.0 wt%, the values of shape fixity and shape recovery reach maximum, which are 96 and 94%, respectively. With the increasing of FG load, the shape-memory performance of the material decreased. This may be due to the agglomeration of high-content graphene and the interference of the increasing of graphene content on the fluidity of polymer in slender nanofibers. Other researchers have found similar phenomena (Das et al., 2016; Jiu et al., 2016; Chen et al., 2019). Graphene agglomeration is the result of van der Waals forces. Many processes have been proven to help avoid filler agglomeration and improve the dispersion of graphene (Atif and Inam, 2016).

In addition to the dual shape-memory effect, the triple-shape-memory composite materials have also attracted great interest from researchers. Triple-shape-memory polymers refer to shape-memory polymers capable of memorizing two temporary shapes in one shape-memory cycle (Zhang Q. et al., 2016; Fatemeh et al., 2017; Du et al., 2019). Fejos et al. (2013) used graphene nanosheets to reinforce polycaprolactone (PCL) nanofiber webs and then infiltrated EP into nanofiber webs by vacuum-assisted resin transfer molding to prepare triple-shape-memory EP/PCL system. The experimental results show that graphene filling can effectively enhance the structure of the nanofiber web, but it will also destroy the shape-memory properties of the PCL phase.

ELECTRO-ACTIVE GRAPHENE/EPOXY COMPOSITES

Thermal-active shape-memory polymer materials are limited by the direct heating temperature when excited, whereas electro-active shape-memory polymer is motivated by applying voltage, so it is more convenient to use than thermal-active shape-memory polymer materials (Lu and Gou, 2012; Meng and Li, 2013). Electromechanical shape-memory polymers use Joule heat generated by electrical current to trigger the shape-memory effect. However, due to the insulating properties of most polymers, researchers have introduced graphene into shape-memory material systems and improved the electrical conductivity of materials by building conductive paths to

improve the electro-active shape-memory effect (Li et al., 2016; Sabzi et al., 2016; Sofla et al., 2019).

Zhou et al. (2016) prepared three-dimensional PU-based graphene foam/EP/CNT (PGEC) shape-memory composites using shape-memory EP as a matrix, PU sponges as the stent, and graphene and CNTs as a conductive network. The conductivity of materials with different graphene contents is not the same. As the graphene content increases, a three-dimensional graphene network structure is gradually formed, and the electrical conductivity of the material is sharply increased. When the graphene content is 12 wt%, the average conductivity of the material reaches 3.33 S/m. A shape recovery test was performed on a PGEC composite containing 8% graphene and 3% CNTs. It was found that the shape fixation rate of the material was 98%. At 100 V, the sample almost restored its original shape in 150 s, with the shape recovery rate reaching 96%. Wang et al. (2015) prepared a reduced GO paper/EP-based shape-memory composite by using a resin transfer molding method to coat shape-memory-reduced GO paper on the surface of a shape-memory EP resin. Bending experiments tested the shape-memory performance of the material, and the surface temperature was detected with an infrared imager. The experimental results showed that when the voltage of 6 V is applied, the material is heated uniformly and can reach 98% shape recovery rate within 5 s. CNTs and graphene compound aerogels were prepared first, and then, nano-aerogel/shape-memory EP composites were obtained by impregnation with monomer EP resin by Liu et al. (2015). Compared with pure EP, the electrical conductivity of the composite is improved by 13 orders of magnitude, reaching a maximum of 5.2 S/m. In the shape-memory performance test, there was a shape recovery process of the compound aerogel (the weight ratio of CNTs to graphene is 3:5)/EP resin composite at 60 V. In the first 20 s, the recovery rate was only 0.5 degrees per second; subsequently, it gradually increased to 2.1 degrees per second. Finally, in the last 30 s, the recovery rate decreased to about 0 degrees, due to most of the stress released at an early stage.

OTHER TYPES OF GRAPHENE/EPOXY COMPOSITES

Shape-memory materials that can be remotely and wirelessly driven have broad application prospects in space engineering, which has attracted widespread attention. Microwave-active shape-memory material is one of them. Microwaves can trigger this kind of shape-memory materials because the dipoles in the materials are oriented and polarized under the action of microwaves. During the deformation process, there will be friction loss and heat generation during the excitation of the microwave-responsive shape-memory material (Zhang et al., 2015; Du et al., 2016). However, the microwave absorption capacity of general shape-memory materials is very low, so it is necessary to introduce enhanced absorbing fillers to improve the orientation polarization performance (Yu et al., 2014; Du et al., 2015; Likitaporn et al., 2018).

Chen et al. (2018) prepared shape-memory nanocomposites using EP-based shape-memory polymer (ESMP) as the matrix. This shape-memory nanomaterial is filled with zero-dimensional carbon black, one-dimensional multiwalled CNTs (MWCNT-COOH), two-dimensional graphene nanosheets (GNPs), and three-dimensional functional graphene sheets (FGSs). The nanofiller content was 2.15 wt% in all nanocomposites. The results of microwave heating experiments showed that GNP/ESMP nanocomposites heat the fastest under the same microwave power. In the shape-memory test, the shape fixation rate of the four nanocomposites was above 97% under 60 W microwave radiation. Except for MWCNT-COOH/ESMP nanocomposites, the shape recovery rate of other composite materials can reach 100% in 10-min microwave heating, and the FGS/ESMP nanocomposite material requires the shortest time, only 250 ± 15 s. This is because FGS/ESMP nanocomposites have the fastest heating speed.

In addition to microwave-active shape-memory materials, photo/thermal-active shape-memory material is also one of the materials that facilitate remote driving (Herath et al., 2019; Huang et al., 2019; Li et al., 2019; Kang et al., 2019). Photo/thermal actuators are made by doping fillers in a photo/thermal-active shape-memory polymer matrix (Lu et al., 2014; Yi et al., 2014). Punetha et al. (2019) prepared bisphenol A diglycidyl ether-functionalized GO (DGEBA-f-GO)-based PU/EP nanocomposites, and the photo/thermal behavior and shape-memory of the performance of the material were investigated. The research results show that with the increase of the content of DGEBA-f-GO, the shape recovery performance of the material under the driving of infrared laser is continuously improved. The shape recovery rate of 2.0 wt% DGEBA-f-GO reaches 98%, and the DGEBA-f-GO nanocomposites still exhibit good shape-memory performance after repeated cycles.

CONCLUSION

In summary, recent researches on graphene/EP composites have brought up a great deal of government and social attention and support. The research results showed that graphene/EP composite material is an intelligent material with important application potential. In addition to its excellent shape-memory function, it also has the characteristics of high strength, wide adjustable range of stiffness, and easy processing and forming. However, the current research on shape-memory graphene/EP composites also has many problems that need to be solved:

- The content of graphene has a great influence on the shape-memory performance of the material. Improper content of graphene can agglomerate in the shape-memory materials, and the aggregation of graphene significantly reduces the strengthening effect of graphene. Therefore, improving the dispersibility of graphene in EP resin and the interface compatibility of shape-memory composites is still a technical problem that needs to be urgently solved.
- To accurately realize the specific deformation of the shape-memory polymer structure in practical applications,

according to the special properties of the shape-memory composite material, the thickness dimension of the shape-memory graphene/EP resin should be limited. In the design of the shape-memory material structure, to prevent the fiber-reinforced material from falling off the resin matrix due to the irrational dimensional structure when the structure is deformed, it is necessary to limit the parameters such as outer size, thickness, and bending radius. Therefore, the application range of shape-memory composites will inevitably be affected.

- At present, the researches on shape-memory graphene/EP resin only focus on the static properties of the material. The dynamic changes of parameters such as glass transition temperature, elastic modulus, and thermal expansion coefficient caused by the preparation method and the graphene doping process are ignored. Also, the temperature change rate and temperature decrease rate in the preparation process may also have a certain effect on the memory performance of the material. Therefore, studying the properties of composite materials under dynamic conditions is also a problem that researchers need to solve.

Shape-memory graphene/EP composites still have a broad development space. If the previously discussed problems can be effectively solved, the application of shape-memory graphene/EP composites will be more extensive in the future.

AUTHOR CONTRIBUTIONS

LC and YZ produced and wrote the manuscript. LC and WL edited the manuscript. LC and ZL edited the manuscript and supervised. All authors read and approved the manuscript.

FUNDING

This work was supported by the 13th Five-Year Plan Equipment Pre-Research Fund under Grant No. 61402060404, the Natural Foundation of Shandong Province under Grant No. ZR2019BEE068, the State Key Laboratory of Mechanical System and Vibration under Grant No. MSV-2019-13, and the Fundamental Research Funds of Shandong University under Grant No. 2018GN034.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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