Review

Shape Memory Graphene Nanocomposites—Fundamentals, Properties, and Significance

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Abstract: Shape memory nanocomposites are excellent smart materials which can switch between a variable temporary shape and their original shape upon exposure to external stimuli such as heat, light, electricity, magnetic fields, moisture, chemicals, pH, etc. Numerous nanofillers have been introduced in shape memory polymers such as carbon nanotubes, graphene, nanodiamonds, carbon nanofibers, etc. Among nanocarbons, graphene has attracted research interest for the development of shape memory polymer/graphene nanocomposites. Graphene is a unique one-atom-thick two-dimensional nanosheet of sp^2-hybridized carbon atoms. Graphene has been used as an effective nanofiller in shape memory polymeric nanocomposites owing to its remarkable electrical conductivity, flexibility, strength, and heat stability. Thermoplastics as well as thermoset matrices have been used to form the shape memory nanomaterials with graphene nanofiller. In shape memory polymer/graphene nanocomposites, their shape has been fixed above the transition temperature and then transformed to the original shape through an external stimulus. The inclusion of graphene in nanocomposites can cause fast switching of their temporary shape to their original shape. Fine graphene dispersion, matrix–nanofiller interactions, and compatible interface development can lead to high-performance shape memory graphene-derived nanocomposites. Consequently, this review focuses on an important class of shape memory graphene-based nanocomposites. The fabrication, physical properties, and shape memory actuation of polymer/graphene nanocomposites are discussed. The stimuli-responsive polymer/graphene nanocomposites mostly revealed heat-, electricity-, and light-induced effects. The inclusion of graphene enhanced the physical/covalent linking, shape recovery, shape fixity, flexibility, and crystallization effects in the polymers. Furthermore, potential applications of these materials are observed in the aerospace/automobile industries, civil engineering, and biomaterials.

Keywords: graphene; nanocomposite; shape memory; thermoresponsive; electroactive; aerospace

1. Introduction

Shape memory materials have been identified as a unique category of smart materials [1]. In the 1980s, this class of smart materials, i.e., shape memory materials, was discovered with the capability to recover their original shape (from a deformed shape) when exposed to external stimuli such as heat [2], light [3], electric fields [4], magnetic fields [5], water [6,7], chemicals [8], solvents [9], pH [10], etc. The shape memory effect is a phenomenon in which a material recovers its original form through external stimuli. The shape memory materials that respond to a heat stimulus are referred to as thermoresponsive materials, which initially gained research attention owing to their facile processing

and applications [11]. Afterwards, the shape recovery of materials using electric fields, light, moisture, and other stimuli was studied. Various polymers revealed the shape memory effect including polyurethane, epoxies, polyesters, and other thermoplastics and thermosetting polymers [12]. The most widely studied shape memory materials include thermoactive, electroactuated, and light-responsive materials. Segmented polymers with various blocks in the main chain show a good shape memory effect. The stimuli-responsive properties of polymers have been enhanced through the incorporation of carbon nanoparticles such as carbon nanotubes, graphene, carbon black, etc. [13,14]. The design, features, and utilization of shape memory polymers and nanocomposites have been explored [15]. Moreover, the mechanism of shape memory materials and their actuation effects have been investigated [16].

Graphene is a unique two-dimensional nanocarbon nanostructure, which is made up of sp^2-hybridized carbon atoms [17]. Graphene has a large surface area and physical properties that are suitable for forming high-performance shape memory alloys and polymeric nanomaterials [18,19]. Consequently, the shape memory features of metal alloys (such as copper alloys) have been improved by doping with minor amounts of graphene [20]. Graphene has been included in metal alloys to enhance their shape recovery, superelasticity, ductility, and strength properties. In polymeric nanocomposites, the inclusion of graphene may cause fast switching of their temporary shape to their original shape [21]. In this regard, graphene dispersion, matrix–nanofiller interactions, and compatible interface formation have led to high-performance stimuli-responsive graphene nanocomposites. Moreover, the fabrication, properties, and actuation methods of shape memory graphene nanocomposites have been investigated. Shape memory nanocomposites have applications in a wide range of fields such as aerospace, automobiles, electronics, textiles, biomedicine, etc. [22]. In this review, polymer- and graphene-derived nanocomposites were surveyed for their physical characteristics and shape memory effects. Numerous polymers were studied to determine their shape memory effects with graphene nanofillers. Their indispensable features and the significance of stimuli-responsive polymer/graphene nanocomposites were considered. In this regard, some previous studies on shape memory graphene nanocomposites were reviewed; however, the literature is not in an updated form to portray the current state of these materials. Nevertheless, future developments in the field of shape memory graphene nanocomposites are not possible for related researchers without prior knowledge of the recent literature. Accordingly, this review includes the significant literature found between 2016 and 2023 (Figure 1). To the best of our knowledge, a specific review on shape memory graphene nanocomposites has not been performed before that arranges, interprets, and outlines the recent literature.
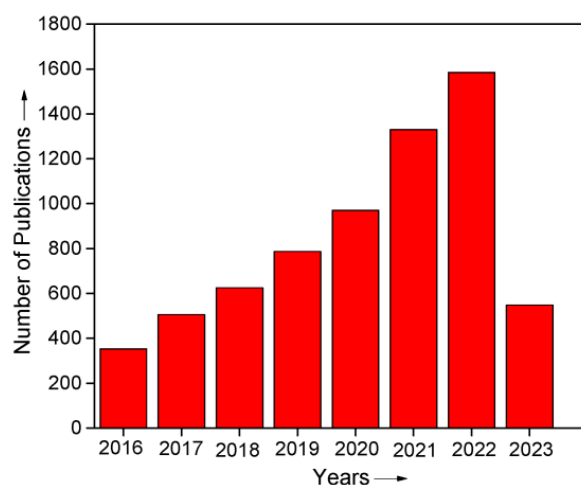

Figure 1. Trend in year-wise publications in the field of graphene-based nanocomposites.

2. Shape Memory Behavior of Polymers

In shape memory polymers, the shape change effect has been observed due to their transformation from a temporary shape to a permanent shape upon exposure to an external stimulus. Various stimuli can cause shape changes in polymers including heat, electricity, light, water, solvents, and other external sources. Heat-responsive shape memory polymers have been studied [23]. Thermoresponsive shape memory polymers show actuation at temperatures higher than their transition temperature (T_{trans}) such as glass transition temperature (T_g) or melting temperature (T_m) [24]. At temperatures above T_{trans}, shape memory polymers can be easily heated and molded to a desired temporary shape [25]. Similarly, electroactive shape memory polymers revealed shape transitions upon the application of an external electric field [26]. In order to understand the shape memory phenomenon, it is important to study the mechanism behind this effect [27]. Most importantly, not all polymers show shape memory effects, for example, rubbers [28]. Rubbers can be deformed elastically under load and return to their original shape upon removal of the load; however, these polymers cannot be fixed to a temporary shape. Therefore, the elastic deformation and recovery of rubbers cannot be considered a shape memory effect. The polymers exhibiting a shape memory effect may have net points and switch segments. The net points in shape memory polymers usually determine the original shape which is usually achieved through covalent or physical cross-linking, crystalline phase formation, molecule entanglement, and interpenetrating network formation. The net points are stable at high temperatures to preserve a shape with stable polymer network formation. On the other hand, switch segments are flexible parts of polymer chains responsible for reversible switching transitions. Moreover, switch segments are important for fixing the temporary shape of polymers. The switch segments may have a crystallization/melting transition, a glass transition of amorphous phase, reversible molecule cross-linking, hydrogen bonding, supramolecular association, and liquid crystal transitions. Figure 2 shows different interactions and cross-linking in shape memory polymers. Various molecular mechanisms can lead to a comprehensive architecture of shape memory polymers.
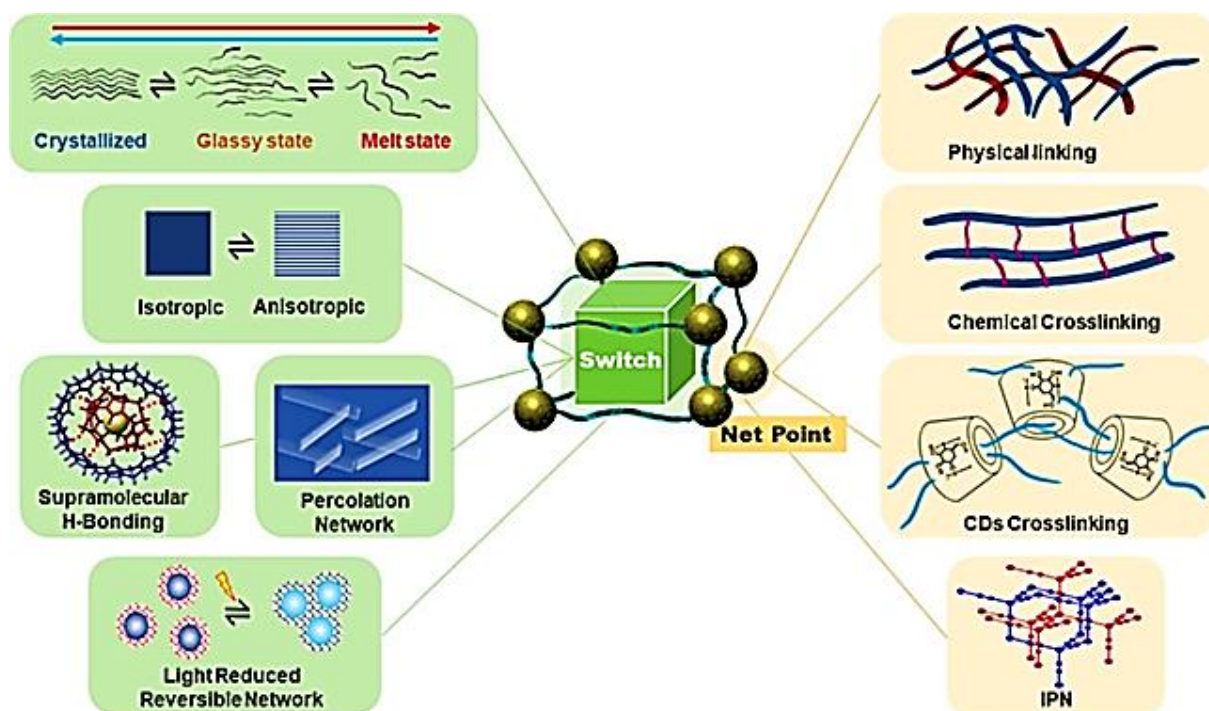

Figure 2. Schematic of comprehensive architecture of shape memory polymers [27]. Reproduced with permission from Springer.

The cross-linking phenomenon due to chemical bond formation between the polymer chains is reversible for shape recovery effects. The physical linking of polymer chains may involve van der Waals forces, hydrogen bonding, and the entanglement phenomenon. Shape memory polymers have significant potential applications in automotive, electronics, and biomedical fields [29]. The inclusion of nanoreinforcements in polymers has enhanced the shape memory effect, mechanical properties, and thermal stability [30]. Among shape memory polymers, segmented polymers such as polyurethanes and polyesters have gained research attention [31–33]. In these polymers, the shape memory effect has been observed due to the formation of reversible interactions between the polymer chains [34].

3. Stimuli-Responsive Polymer Nanocomposites

Thermoplastic and thermoset polymers have been widely used as shape memory materials [35–37]. Among nanofillers, carbon nanoparticles and inorganic nanofillers have been filled in polymeric matrices to cause the shape memory effect. Consequently, nanocarbon [38], nanoclays [39], and inorganic nanoparticles [40] have been investigated. Carbon nanoparticles (graphene, carbon nanotubes, fullerene, etc.) have been used as effective reinforcements in thermoresponsive and electroactive shape memory materials (Figure 3) [41]. For high-performance shape memory materials, homogeneous nanoparticle dispersion has been considered important [42]. Shape memory nanocomposites have been fabricated using facile techniques such as solution casting, melt blending, and other synthesis techniques [43]. Functional nanoparticles have been used to enhance the dispersion and compatibility in polymeric matrices [44,45]. In nanocomposites, the major types of shape memory effects include electroactive, thermoresponsive, light-responsive, moisture-sensitive, and magnetic-responsive effects [46,47].
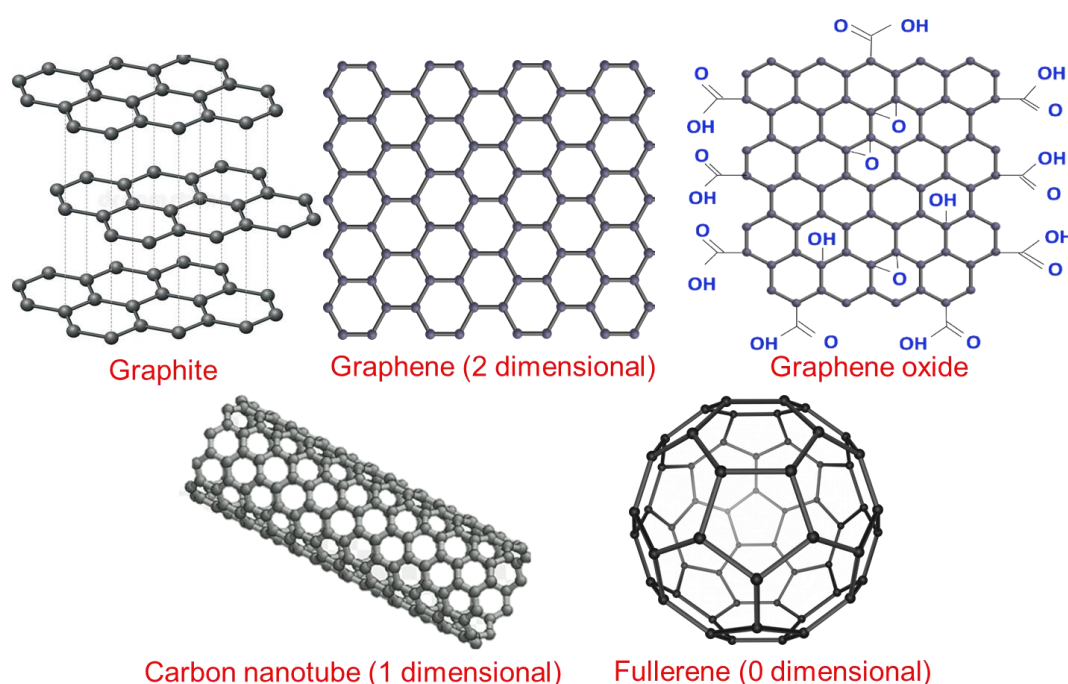

Figure 3. Various carbon nanofillers in shape memory nanocomposites.

3.1. Electroactive Shape Memory Nanocomposite

Conductive nanofillers such as carbon nanoparticles have been used to produce the electroactive shape memory effect in nanomaterials [48]. Carbon nanotubes have been known as an effective nanoreinforcement for shape memory polymers [49]. Functional carbon nanotubes can develop covalent cross-linking with polyurethanes to enhance the shape memory effect [50]. Nanofiller loading also influences the shape memory properties of these materials [51]. Shape memory polyurethane/carbon nanotube nanocomposites have been

fabricated using the spray deposition modeling (SDM) technique, in which nanofillers were deposited on the polymer films [52]. Using SDM, 10, 20, 30, 40, and 50 carbon nanotube layers were printed on polymers. The rectangular, semi-circle, and spiral line areas of the samples were studied for the applied voltage, and thermal images were scanned. Upon the application of voltage, the nanocomposite stability was observed at constant temperature. Furthermore, shape memory polyurethane/nanoclay and polyurethane/metal nanoparticle nanocomposites have been designed and studied [53,54].

Commonly studied shapes of electroactive shape-responsive polymers include rectangular, semi-circle, and spiral forms. In addition, more complex shapes have been observed in these shape memory polymer nanocomposites. For example, Xie et al. [55] designed star-shaped electroactive shape memory networks derived from polylactide and aniline. The polymer chains were chemically cross-linked to synthesize a shape memory nanocomposite. Lu et al. [56] studied octagon-shaped shape memory polymer/carbon nanofiber nanocomposites. Joule heating-triggered shape recovery was observed in the three-dimensional template of self-assembled nanocomposites, and the electroactivated recovery of the nanocomposites was monitored.

Moreover, the alignment of conductive nanofillers plays an important role in shape memory actuation. Lu et al. [57] studied a shape memory polymer nanocomposite based on self-assembled multi-walled carbon nanotubes and nickel nanostrands. The vertical alignment of conductive nanofillers in the shape memory matrix enhanced the electroactive recovery behavior. Good nanofiller alignment resulted in a 100% recovery ratio of the shape memory nanomaterials.

3.2. Thermoresponsive Shape Memory Nanocomposite

In addition to the electroactive effect, heating-triggered shape memory polymer nanocomposites have been reported [58]. Thermoresponsive shape memory polymeric nanocomposites have been widely studied for a range of potential applications from the engineering to biomedical sector [59,60]. Microwave heating has been used for rapid, uniform, and controlled actuation of nanocomposites [61]. Gopinath et al. [62] developed thermoresponsive polycaprolactone/polystyrene-block-polybutadiene-block-polystyrene/carbon nanofiber nanocomposites. The shape memory effect in these nanocomposites was stimulated by a change in temperature. Due to the good miscibility of the polymer chains and nanofiller dispersion, good shape recovery and fixing performance were observed. Epoxy resins have been studied for the shape memory effect [63]. The shape memory effect in epoxy resins has been observed due to network formation and reversible switching transitions [64]. Consequently, shape memory epoxy/carbon nanotube [65], epoxy/nanoclay [66,67], and epoxy/metal nanoparticle nanocomposites have been investigated [68]. In poly(vinyl alcohol)/carbon nanotube nanocomposites, thermoresponsive and electroactive effects have been observed [69,70]. Flexibility and shape recovery properties were investigated for low nanofiller contents in the poly(vinyl alcohol) matrix [71]. Several other thermoplastic polymeric nanocomposites revealed an effective shape memory phenomenon [72].

3.3. Light-Responsive Shape Memory Nanocomposite

Light-responsive stimuli-responsive nanocomposites have been effectively developed [73]. The light-driven actuation of shape memory nanocomposites, with light of different wavelengths, has been found efficient relative to other shape memory effects [74,75]. Chen et al. [74] formed polycaprolactone/polyurethane-based light-sensitive nanomaterials. The polydopamine nanospheres were used as photothermal nanofiller. The irradiation of 150 s caused light-sensitive recovery of >78%. Light-responsive nanocomposites have been applied for fabricating intelligent structures, biomedical microdevices, artificial muscles, robotics, etc. [76].

3.4. Moisture-, Chemo-, Or Solvent-Responsive Shape Memory Nanocomposites

Moisture-sensitive, chemoresponsive, and solvent-responsive shape memory nanocomposites have also been developed [77–79]. The shape recovery in these materials does not require external heating. The moisture-sensitive shape recovery is an environmentally friendly process with sufficient structural and chemical flexibility [80,81]. Moisture-sensitive or chemoresponsive nanomaterials also possess the advantage of low-temperature actuation [82]. Moreover, glass transition temperature, intermolecular hydrogen bonding, and plasticizing do not affect the moisture-sensitive recovery process [83]. Wang et al. [84] fabricated chemoresponsive polycaprolactone/graphene oxide nanomaterials due to the uniform network formation. The inclusion of 0.5% nanofiller enhanced the strain by 300%.

3.5. Magnetic-Responsive Shape Memory Nanocomposite

Magnetically sensitive shape memory polymer nanocomposites have been reported, in which covalent integration of nanoparticles improved the shape memory effect [85,86]. Magnetic metal oxide nanoparticles such as Fe_3O_4 (size: ~100 nm) have been used to create a magnetic response in the nanocomposites [87]. Pekdemir et al. [88] prepared magnetic-responsive polylactide/poly(ethylene glycol) with magnetic Fe_3O_4 nanoparticles. These magnetic-responsive materials have been applied in data storage and microfluidic devices [89]. Moreover, pH-sensitive effects have been observed in the polymer/nanocarbon nanocomposites.

The shape memory polymer/nanocarbon nanocomposites have been applied in engineering, civil, and technical devices and systems [90,91]. Shape recovery effects in these materials have also been explored through modeling and simulation approaches [92]. The thermoviscoelastic properties of shape memory polymer composites have been examined [92]. The shape memory nanocomposites usually consist of reinforcements (carbon nanoparticles, carbon black, carbon fibers, glass fibers, etc.) embedded in the matrix. Nanofiller addition has been found to alter the thermomechanical properties and shape memory performance of the nanomaterials. To understand the shape recovery phenomenon, the mechanisms of the thermomechanical properties of nanocomposites need to be investigated. Changes in thermomechanical properties were found to be dependent on the glass transition and viscoelastic properties of the nanocomposites. To explore the shape memory behavior of the nanocomposites, finite-deformation and time-dependent thermoviscoelastic models have been studied [93]. Zeng et al. [94] developed the thermoviscoelastic finite-deformation constitutive model for thermoresponsive shape memory nanocomposites. The model was based on the modified temperature-dependent laminate analogy theory to predict thermal-dependent effective elastic properties of nanocomposites. The constitutive model was applied to study the thermoviscoelastic properties of shape memory nanocomposites with different filler loadings. The simulation results revealed agreements with the experimental data.

4. Shape Memory Effect in Polymer/Graphene Nanocomposites

Graphene is a one-atom-thick nanosheet made up of sp^2-hybridized carbon atoms [95]. It has a honeycomb lattice structure [96]. Graphene is a derived form of graphite with stacking graphene layers with van der Waals interactions [97]. High-surface-area graphene has high electron transportation, thermal conductivity, Young's modulus, and strength properties. Graphene gained special position among nanocarbon nanoparticles due to its unique structure and properties [98]. Graphene has been used to form various derived nanomaterials [99]. Moreover, graphene can be further modified or functionalized to form graphene oxide and reduced graphene oxide-like structures [100]. Graphene, graphene oxide, and reduced graphene oxide have been used as efficient nanofillers for polymers [101]. Applications of graphene and its derived materials have been found in the fields of engineering structures, energy devices, electronics, biomedicine, and nanocomposites [102–104].

The inclusion of graphene in polymers has been found to enhance shape recovery and self-healing properties [105]. Generally, superior shape recovery properties were

observed with low graphene contents of 0.005–3 wt.% [106]. The inclusion of graphene has been found to improve the physical properties and shape memory features of polymers. Several types of graphene have been produced such as CVD graphene, modified graphene, graphene oxide, graphene foams, reduced graphene oxide, mechanically exfoliated graphene, etc. The physical and chemical characteristics of different graphene types are different. Graphene oxide and reduced graphene oxide also have different chemical and structural features owing to variances in their chemical compositions. The synthesis of graphene oxide has been attained using a top-down approach such as the treatment of graphite with strong oxidants and subsequent exfoliation. In this process, the sp^2 graphite structure is disrupted to form different oxygen-containing functionalities (carboxyl, hydroxyl, or epoxy groups) on graphite layers. The disruption of sp^2 bonding causes low electrical conductivity properties. The oxidation of graphite layers also enhances the interplanar spacing of the graphite structure. The reduction of graphene oxide also causes graphene-like behavior. Chemical, thermal, or photothermal reduction methods have been applied to attain reduced graphene oxide structures. However, reduced graphene oxide cannot achieve a pristine graphene structure. Residual oxygen and structural defects exist on the reduced graphene oxide surface due to chemical oxidation synthesis. CVD graphene has been fabricated as single-layer graphene with a fine structure and the least impurities. All the graphene and derivative forms of graphene have been successfully used to form shape memory nanocomposites.

Liu et al. [107] formed CVD graphene and applied it to fabricate shape memory epoxy/graphene nanocomposites through the vacuum infusion technique. The nanocomposites had high conductivity of 16 Sm^{-1}. The electroactive shape memory effect was studied at 60 V. The shape recovery rate was found to be 0.5 degrees per second in 20 s. Rong et al. [108] also produced single-layer graphene by the CVD method. Then, graphene foams were developed to form shape memory nanomaterials. The derived electroactivated shape memory nanocomposites had a fast response of 53 ms.

High-performance shape memory polyurethane/graphene systems have been designed [109]. Jung and co-workers [110] considered shape memory polyurethane/graphene nanocomposites with high mechanical properties and shape recovery force (1.8 MPa cm^{-3}) due to fine nanofiller dispersion. Modified graphene has been developed to form shape memory nanocomposites. Kim et al. [111] developed acrylate-terminated polyurethane and allyl isocyanate-modified graphene-based nanocomposites. The shape recovery ratio was measured to evaluate the effect of nanofillers on the shape memory behavior of the nanocomposites. The shape recovery ratio was improved with the inclusion of graphene, up to 1.5 phr content, due to fine nanofiller dispersion. The graphene nanofiller was found to control polymer segmental movement during the recovery motion of the sample, leading to a high shape recovery ratio. However, the shape memory properties were decreased at higher graphene contents due to aggregation. Consequently, graphene addition caused a high yield strength, modulus, and glass transition temperature of the nanocomposites.

Graphene oxide has been obtained using facile methods for fabricating shape memory nanocomposites. Yan and co-workers [112] formed shape memory polyurethane/multi-layer graphene oxide nanocomposites. A shape recovery ratio of 83% in 7.6 s was attained. The superior shape recovery ratio of the nanocomposites was due to the restriction of polymer chain mobility and limitation of stress transfer on interfacial domains. In addition to graphene oxide, reduced graphene oxide has also been effectively employed as nanofiller in shape memory nanomaterials. Yoo et al. [113] reinforced graphene oxide, reduced graphene oxide, and poly(ε-caprolactone) functional graphene oxide nanofillers in the polyurethane matrix. A solution processing method was used to develop the shape memory nanocomposites. The formation of poly(ε-caprolactone) functional graphene oxide is shown in Figure 4. The shape recovery of neat polyurethane and polyurethane/graphene nanocomposites was studied in different cycles (Figure 5). The shape recovery of the polyurethane/poly(ε-caprolactone) functional graphene oxide nanocomposite was >95% relative to neat polyurethane (88.3%) and other nanocomposites. It was suggested that

the functional graphene oxide develops better interactions and reversible cross-linking to facilitate the shape recovery process. The thermoresponsive behavior of the samples was analyzed at 50 °C under 6.5 gf loading (Figure 6A). The nanocomposite revealed a fast shape recovery of 50% in 8 s relative to the neat polymer sample tested. Hence, the polyurethane/graphene oxide samples revealed fast shape recovery along with mechanical stability properties. Gupta et al. [114] produced shape memory polyurethane/graphene nanoplatelets through the melt-blending method. The shape recovery was considered under microwave irradiation. The nanocomposites were developed with 0.2, 0.4, 0.6, and 0.8 phr nanofiller loadings. Figure 6B shows the shape recovery behavior of the nanocomposites with time. The inclusion of graphene nanoplatelets in the polyurethane matrix enhanced its dielectric and magnetic properties by acting as heating nodes. The sample recovered 80% of its shape in 60 s, which is slower than the reported functional graphene oxide-based nanocomposites [113].
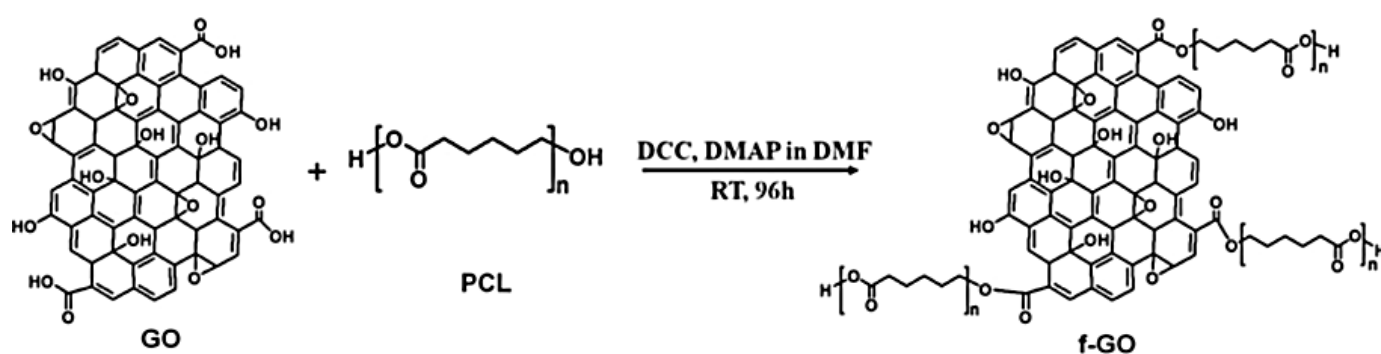

Figure 4. Synthesis scheme of poly(ε-caprolactone) functional graphene oxide [113]. Reproduced with permission from ACS.
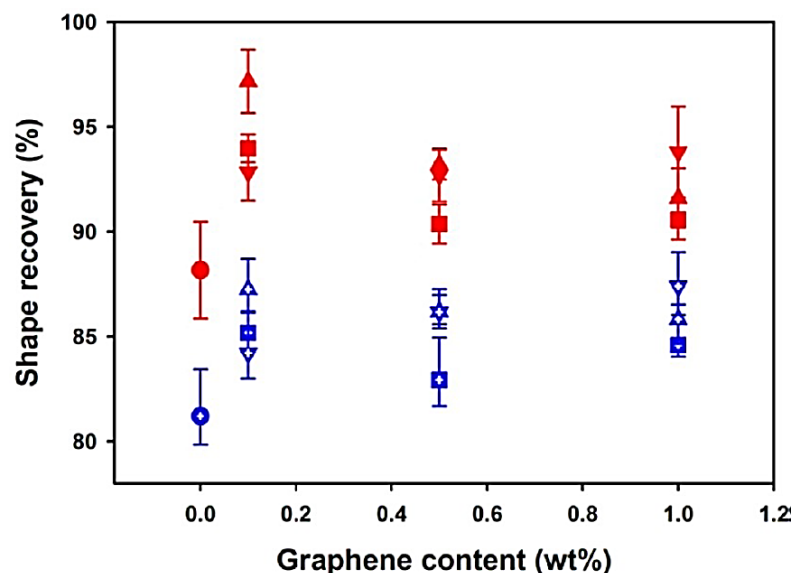

Figure 5. Shape recovery of pure PU and PU/graphene nanofiber webs with different cycles (red and solid symbols: first cycle; blue and open symbols: fifth cycle; ● PU; ■ PU/GO; ▲ PU/f-GO; and ▼ PU/r-GO) [113]. PU = polyurethane; PU/GO = polyurethane/graphene oxide; PU/f-GO = polyurethane/functional graphene oxide; PU/r-GO = polyurethane/reduced graphene oxide. Reproduced with permission from ACS.
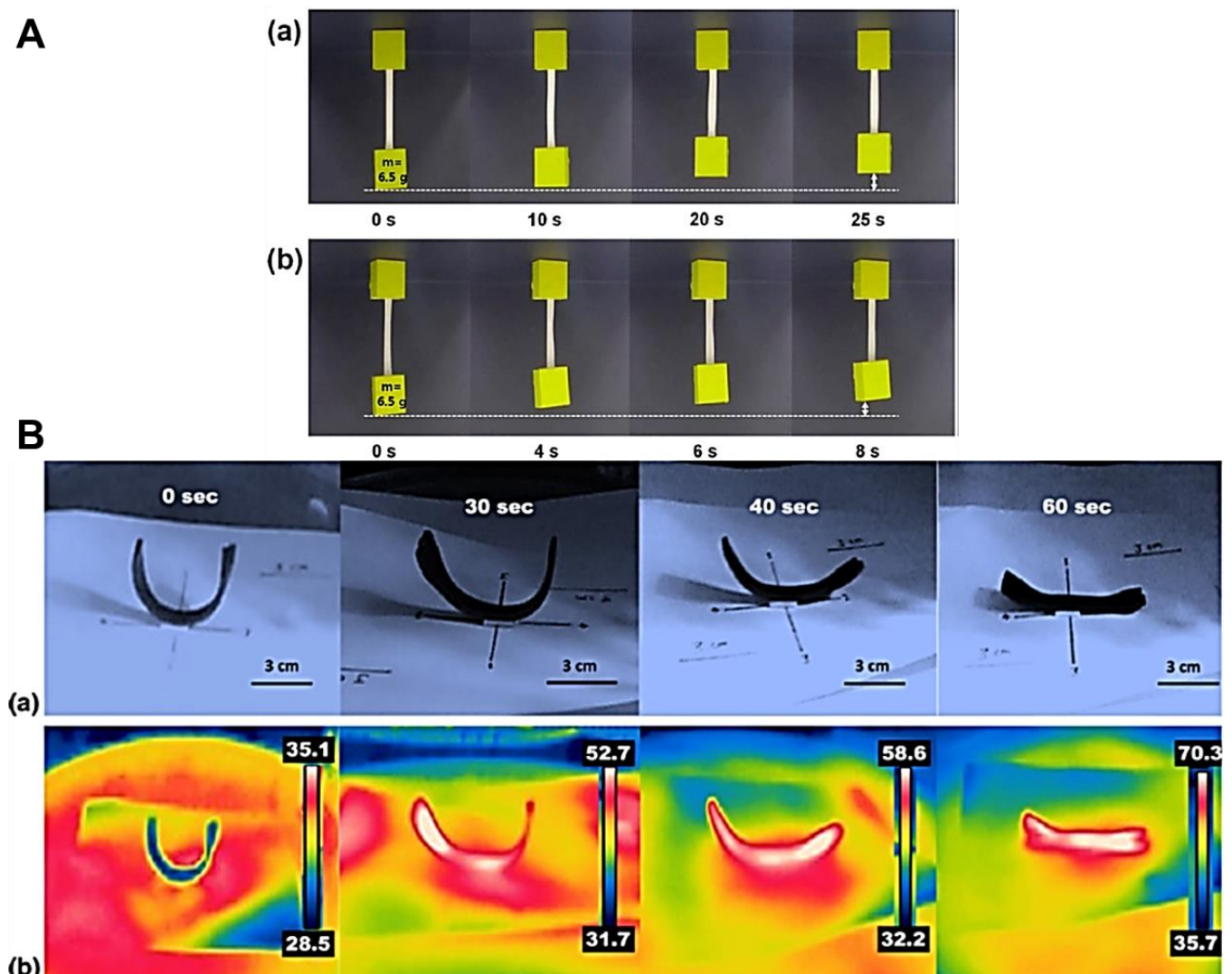
Figure 6. (**A**) Shape recovery images of (**a**) PU and (**b**) PU/GO nanocomposite, where shape recovery test was carried out at a constant temperature of 50 °C with increasing time [113]. PU = polyurethane; PU/GO = polyurethane/graphene oxide. Reproduced with permission from ACS. (**B**) Sequence of shape recovery of shape memory polyurethane/graphene nanoplatelets under microwave irradiation: (**a**) digital images; (**b**) infrared thermal images [114]. Reproduced with permission from Springer.

Sofla et al. [115] fabricated electroactive shape memory polyurethane and CVD graphene-based nanocomposites. The polyurethane was prepared using polycaprolactone, hexamethylene diisocyanate, and 1,4-butanediol in the solution processing. Figure 7 displays variations in the flowing electrical current with respect to the applied voltage for the prepared nanocomposites. The electrical conductivity was enhanced with the graphene addition, and a percolation threshold of 1.5 wt.% was obtained. For the shape recovery of the fixed samples, 75 V was applied for 60 s (Figure 8A). The inclusion of 1–1.5 wt.% graphene in polyurethane did not cause complete shape recovery of the samples. The addition of 2–3 wt.% graphene nanofiller caused shape recovery within 60 s. Table 1 shows the shape memory properties of neat polyurethane and the resulting nanocomposites. Increasing graphene content improved the shape fixity (83.4%) and shape recovery (100%) of the nanocomposite samples. Thus, the inclusion of graphene in segmented polyurethane enhanced the shape memory parameters due to better matrix–nanofiller compatibility. Kim et al. [116] developed electroactive shape memory polyurethane nanocomposites with various amounts of thermally reduced graphene. The thermally reduced graphene was chemically modified with allyl isocyanate. The electroactive shape recovery behavior is

shown in Figure 8B. Thermally reduced graphene was loaded up to 25 wt.%. At low nanofiller contents, the electric current did not induce any shape change due to the inadequate electrical dissipation. On the other hand, at high nanofiller contents (20 and 25 wt.%), up to 97% of the original shape was recovered. However, the electroactuation response was slower than for the reported polyurethane and graphene-based samples [115].
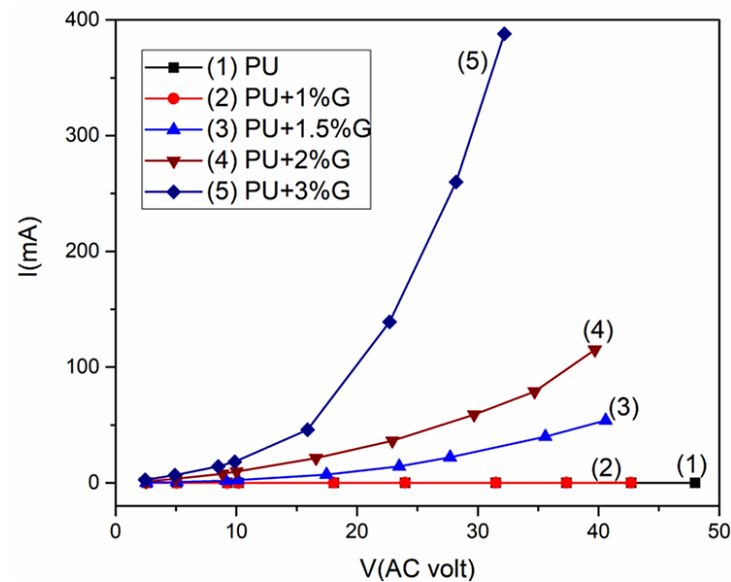

Figure 7. Variation in flowing electrical current vs. applied voltage for PU and PU/graphene nanocomposites [115]. PU = polyurethane; G = graphene. Reproduced with permission from Elsevier.

Table 1. Shape memory properties of PU and PU/graphene nanocomposites [115]. PU = polyurethane; G = graphene. Reproduced with permission from Elsevier.

Sample	Shape Fixity (%)	Shape Recovery (%) (Thermal: 60 °C)	Shape Recovery (%) (Electrical: 75 V at 60 s)
PU	65 ± 2	90 ± 2	Not Recovered
PU + 1% G	71 ± 3	94 ± 2	Not Recovered
PU + 1.5% G	78 ± 2.2	94 ± 3	Not Recovered
PU + 2% G	82.2 ± 4	98 ± 2	95
PU + 3% G	83.4 ± 1	100	100

Epoxy nanocomposites with graphene nanofiller have been investigated for their thermoresponsive shape recovery effects [117]. Williams and co-workers [118] developed thermally actuated shape memory epoxy/graphene nanocomposites. The influence of nanofiller content and polymer–nanofiller interactions on shape recovery was examined. Yu et al. [119] designed shape memory epoxy and graphene oxide-derived nanocomposites. Figure 9 depicts the thermal deformation mechanism for the shape memory epoxy material. A temperature above T_{trans} was used to mold the material under applied force, followed by cooling the material to fix the shape. Then, reheating the materials above T_{trans} and removing the applied force recovered the original shape of the material.
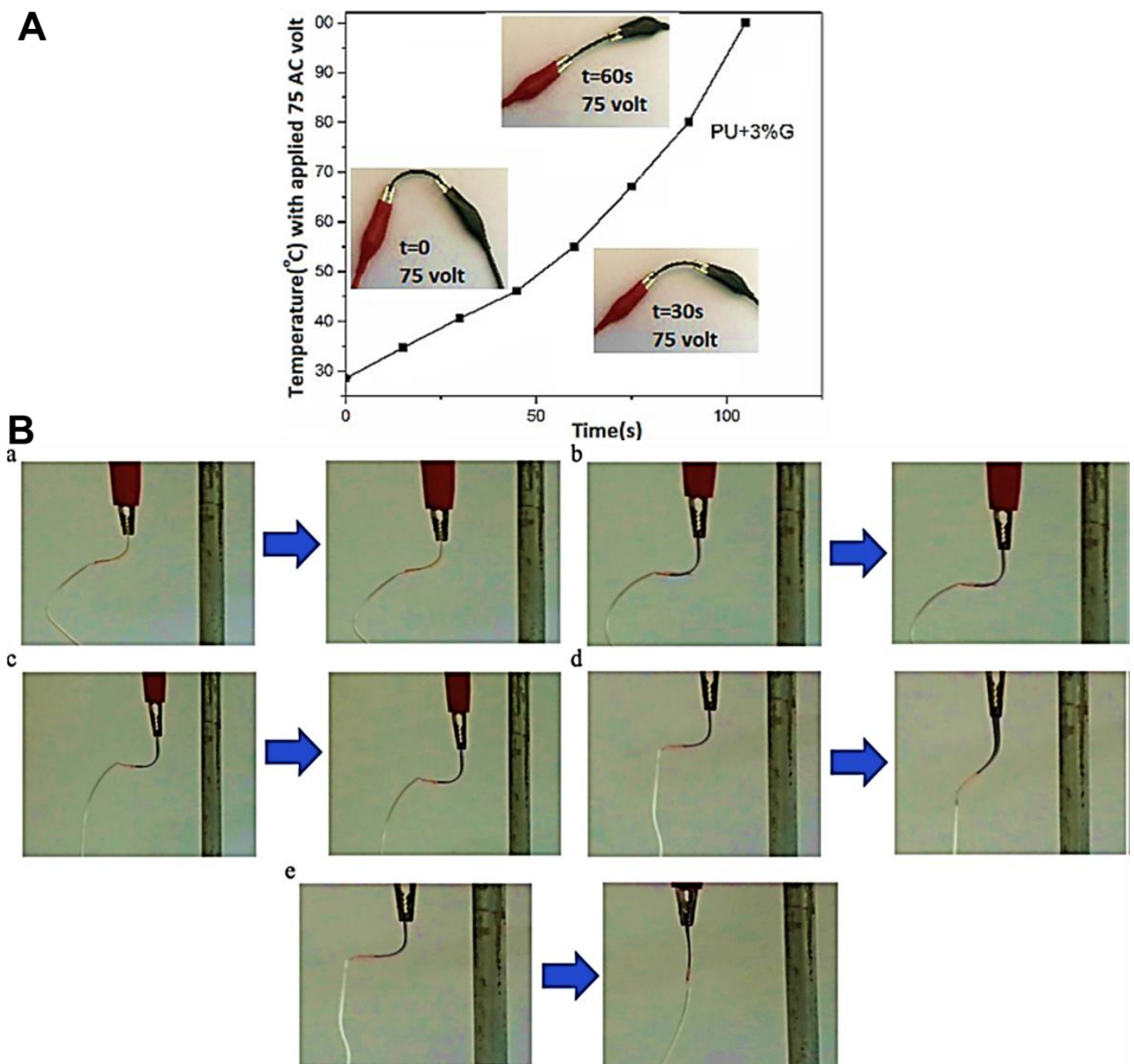
Figure 8. (**A**) Electroactuation of polyurethane/CVD graphene samples [115]. Reproduced with permission from Elsevier. (**B**) Electroactive shape memory behavior of polyurethane with 0 (**a**), 10 (**b**), 15 (**c**), 20 (**d**), and 25 (**e**) wt.% thermally reduced graphene. The as-cast straight line is deformed (left) and recovered (right) partially with 20 and almost completely with 25 wt.%, whereas 0 and 10 wt.% do not respond to the electrical current [116]. Reproduced with permission from Elsevier.
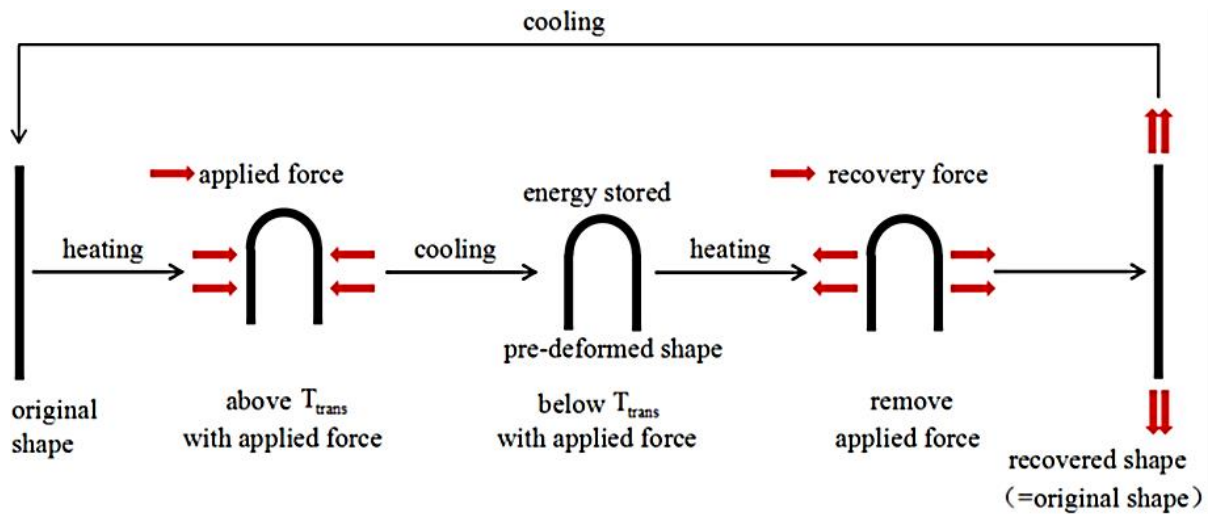
Figure 9. Schematic of thermal deformation shape memory epoxy material [119]. Reproduced with permission from MDPI.

Lu and co-researchers [120] fabricated shape memory epoxy-derived nanocomposites. Graphene oxide-coated carbon fibers were used as reinforcement [121]. The interfacial interactions between the epoxy and graphene oxide-modified carbon fiber are shown in Figure 10. These interactions enhanced the matrix-filler compatibility. The electrical-current-induced Joule heating effect was also observed in the epoxy/graphene oxide-coated carbon fibers (Figure 11). Up to 95% shape recovery was observed. Reduced graphene oxide was applied to form shape memory nanocomposites.
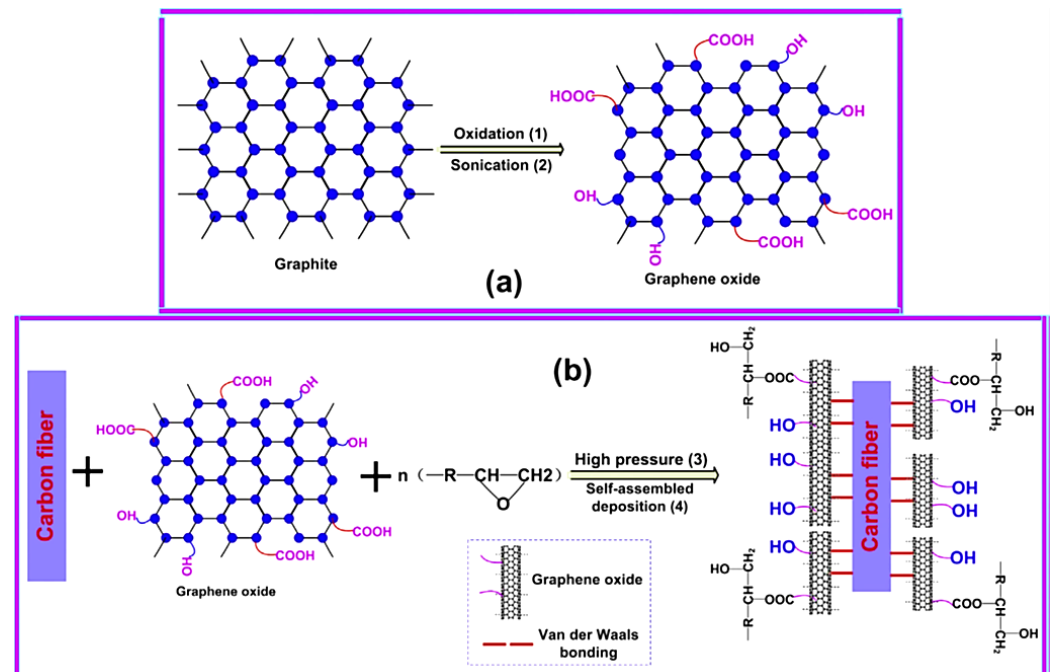

Figure 10. (**a**) Illustration of graphite stack oxidized to separate individual layers of GO; and (**b**) role of GO in interfacial bonding between carbon fiber and epoxy-based SMP matrix via van der Waals bonding and covalent cross-linking, respectively [120]. GO = graphene oxide; SMP = shape memory polymer. Reproduced with permission from Elsevier.
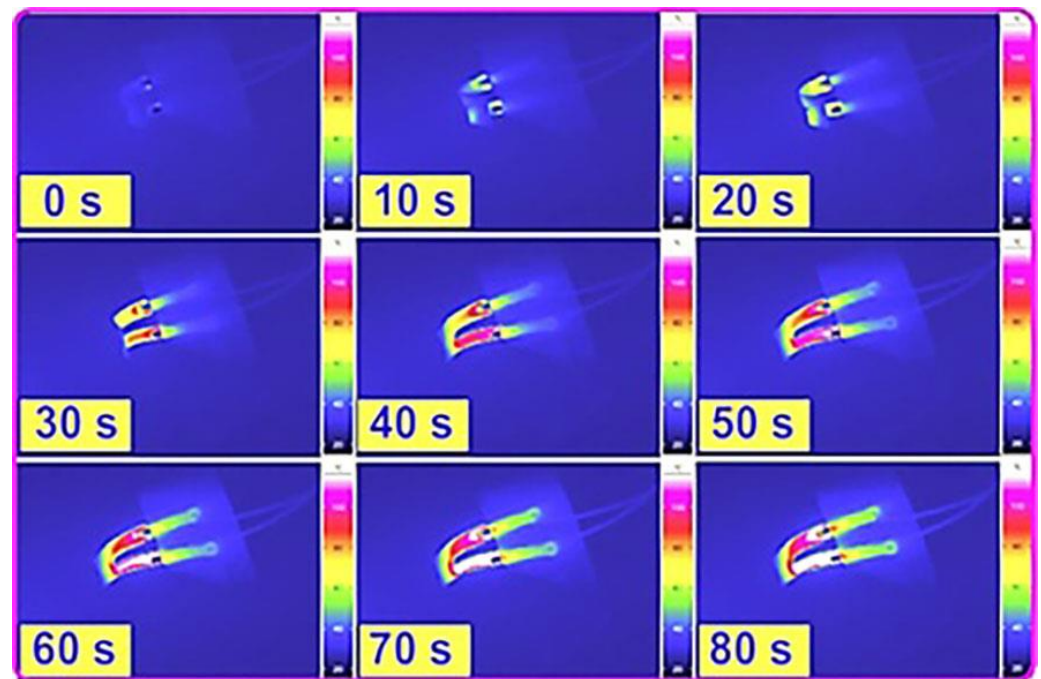
Figure 11. Snapshot of Joule heating-induced shape recovery in SMP nanocomposite recorded by infrared video camera [120]. SMP = shape memory polymer. Reproduced with permission from Elsevier.

Wang and co-workers [122] formed electroactive shape memory epoxy/reduced graphene oxide nanocomposites. The applied voltage was used to enhance the shape recovery effect of these materials. Figure 12A shows the shape memory process in the epoxy and reduced graphene oxide-derived nanocomposites. Initially, the nanocomposite was heated to 113 °C and fixed to a temporary 'U' shape. Then, the temporary shape was transformed to the original shape at 6 V. The shape recovery properties of the nanocomposite are given in Table 2. Wang et al. [123] prepared a waterborne epoxy/reduced graphene oxide nanocomposite. The sample was fixed into a 'U' shape above T_g and then cooled to 25 °C. Electroresponsive shape recovery was observed at 8 V (Figure 12B). The nanocomposite film had a response within 8 s, i.e., slower than the epoxy/reduced graphene oxide shape memory nanocomposite under 6 V [122]. Liu et al. [107] designed epoxy thermally reduced graphene/carbon nanotube hybrid nanocomposites. Due to the high conductivity of the nanocomposites, the original shape was actuated at 60 V (Figure 12C). The recovery rate was 0.5 degrees per second during the first 20 s. The recovery rate was enhanced to 2.1 degrees per second. However, the shape recovery was slower than that reported for epoxy/reduced graphene oxide nanomaterials [123].
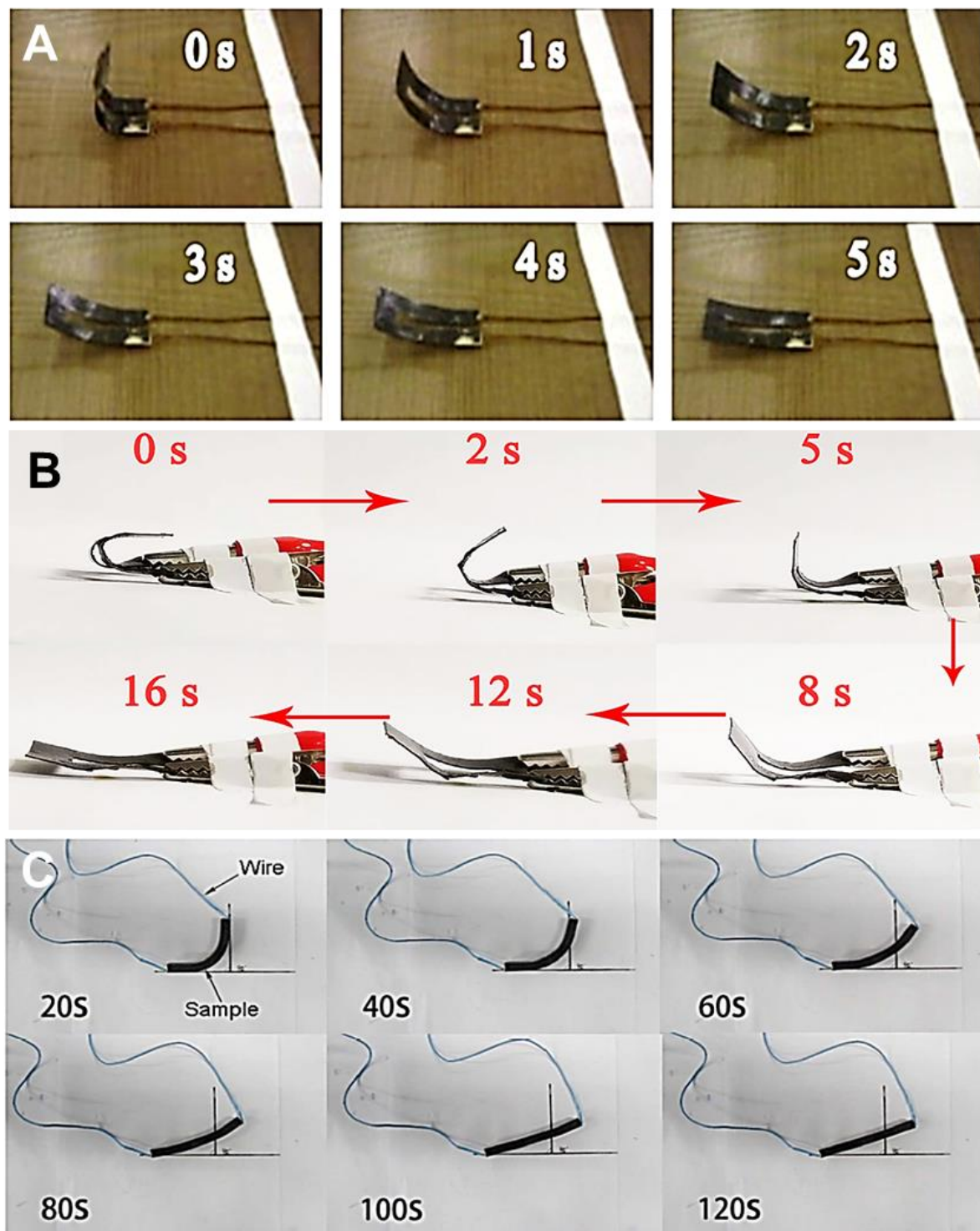
Figure 12. (**A**) Shape recovery process of epoxy/reduced graphene oxide paper-based shape memory nanocomposite under 6 V [122]. Reproduced with permission from Elsevier. (**B**) Electroresponsive shape memory effect of the waterborne epoxy/reduced graphene oxide nanocomposite film at 8 V applied voltage [123]. Reproduced with permission from Elsevier. (**C**) Shape recovery process of the compound aerogel (the weight ratio of carbon nanotubes and graphene is 3:5)/epoxy resin composite under voltage of 60 volts [107]. Reproduced with permission from RSC.

Table 2. Epoxy/graphene oxide or modified graphene oxide-based shape memory nanocomposites.

Sample	Nanocomposite Dimensions	Applied Voltage or Current	Shape Recovery Time or Ratio	Highest Temperature (°C)	Ref.
Epoxy/reduced graphene oxide paper	Thickness, 0.1 cm	6 V	5 s	240	[122]
Epoxy/graphene oxide	$80 \times 6 \times 2$ mm^3	-	90%	74	[119]
Epoxy/graphene	5 mm	5–10 mA	60 s	60	[124]
Epoxy/graphene oxide/carbon fiber	$50 \times 15 \times 0.30$ mm^3	-	20 min/100%	80	[125]
Waterborne epoxy/graphene oxide	$30 \times 4 \times 0.25$ mm^3	2–9 V	3–7 s/> 90%	25	[123]
Polyurethane/epoxy resin/functional graphene	$25 \times 5 \times 1$ mm^3	-	96%	50	[126]

The shape recovery ratio measures the capability of the shape memory material to recover its original shape [127]. It is usually calculated as total deformation recovered vs. maximum deformation attained during programming. For electroactive shape memory epoxy/reduced graphene oxide nanocomposites, a shape recovery ratio of up to 98% was achieved. Similarly, actuation force is the force exerted by the sample on an object. The optimization of actuation force has been found important to achieve an efficient shape memory effect [128]. The assessment of applied actuation force over time is also important to analyze the shape memory behavior. At the maximum actuation force, usually all energy stored within the polymer chains is released. In shape memory nanocomposites, the inclusion of nanoparticles affects the actuation force. Better interfacial interactions between the polymer and graphene requires a large actuation force for the chain movement. In particular, the electric-field-driven shape recovery revealed a good actuation force and recoverable strains, thus providing opportunities to form high-performance actuating systems.

Moreover, the shape memory phenomenon has been observed in polyacrylonitrile/graphene nanocomposites [129]. Cross-linking in polyacrylonitrile/graphene nanocomposites revealed good stimuli-responsive effects. Shape memory polylactic acid/graphene nanomaterials have also been developed [130]. Uniform graphene dispersion has enhanced the crystallization and shape memory effects of polylactic acid [131]. Graphene has been filled in polyethylene elastomer for the shape memory effect [132]. Increasing the nanofiller loading was observed to enhance the shape recovery effect. Subsequently, the nanofiller dispersion, loading, and cross-linking have been found to increase the stimuli-responsive phenomenon in these polymers. Table 3 details the shape memory effect in significant graphene-based nanocomposites.

Table 3. Shape memory effect in graphene-based nanocomposites.

Polymer	Nanofiller	Actuation Type	Properties	Ref.
Polyurethane	Graphene	Electroactive	Shape recovery force: 1.8 MPa cm^{-3}	[110]
Polyurethane	Graphene	Light-active	Nanofiller dispersion; shape recovery ratio	[111]
Polyurethane	Multi-layer graphene oxide	Electroactive	Shape recovery ratio: 83%; Shape recovery time: 7.6 s	[112]
Polyurethane	Graphene oxide, reduced graphene oxide, poly(ε-caprolactone functional graphene oxide	Thermoresponsive	Shape recovery > 95%	[113]
Polyurethane	Graphene	Electroactive	Percolation threshold: 1.5 wt.%; shape recovery: 100% in 60 s; shape fixity: 83.4%	[115]
Epoxy	Graphene oxide	Thermoresponsive	Thermal deformation mechanism; T_{trans}	[119]
Epoxy	Graphene oxide-coated carbon fiber	Electroactive	Electrical-current-induced Joule heating effect; shape recovery: 95%	[120]
Epoxy	Reduced graphene oxide	Electroactive	Shape recovery at 6 V	[122]

5. Applications of Graphene-Based Shape Memory Nanocomposites

Due to superior shape memory properties, multiple triggering strategies, and fast responses, graphene-based shape memory nanocomposites have been used in various fields ranging from research to industry [133]. Significant applications have been observed in electronics, sensors, energy devices, etc. [134]. Shape memory graphene materials have high electrical conductivity for electroactive actuation [135]. Gao and co-workers [136] formed shape memory polycaprolactone/graphene oxide with a millisecond response. The nanocomposites have been applied in a high-speed fuse to avoid current overloading. The shape memory graphene-based fuse easily recovered its original shape and circuit breaking. These nanocomposites have also been used as micro-oscillators by coupling with a high-frequency electromagnetic field. Xie et al. [137] formed shape memory graphene-based cyclic actuators, which were triggered by heat or light stimuli. The nanocomposites have been applied in smart devices for automatic restoring, due to cyclic sensitivity. In addition to electronics, stimuli-responsive graphene nanomaterials have been used for energy storage purposes [138]. This application relies on the excellent photothermal conversion and thermal insulation potential of the materials [139].

Shape memory polymer nanocomposites have essential civil engineering applications due to their high mechanical properties [140–142]. Civil-engineered structures demand high bending, twisting, and fluctuating capacities [143]. Consequently, civil structures with shape memory graphene-based nanomaterials have been formed [144]. Thermoresponsive, electroactive, and moisture-active polymer/graphene nanomaterials have been used in civil engineering [145–147]. Self-healing polymer and graphene nanocomposites have also been used for civil structures [148]. Graphene-based civil materials have high strength, large recoverable strain, and high recovery stress [149]. Stimulation studies have also been performed to explore shape memory graphene-derived civil engineering structures.

In addition, stimuli-responsive polymer/graphene nanocomposites have been studied for aerospace and automotive components [150–152]. Space craft/automobile outer body, wings, inner seats, airflow controls, lenses, and other structures have been developed using polymer/graphene nanomaterials [153,154]. Shape memory nanocomposites have

the advantages of inexpensiveness, a light weight, fast actuation, facile processing, and mechanical robustness [155,156]. In the space sector, self-healing nanocomposites have gained recent interest [157]. The stimuli-responsive polymer/graphene nanocomposites have also been exploited in the actuators, sensors, and microelectronics related to aircrafts and automobiles [158].

Moreover, the application of shape memory nanocomposites has been observed in biomedical devices [159,160]. The stimuli-responsive polyurethane nanocomposites have been used in biomedical implants and other devices [161]. The switchable segments in polyurethane/graphene nanocomposites have enhanced their function for biomedical applications [162–164]. A few attempts of applying shape memory polymer/graphene nanocomposites in smart textiles and fabrics have been observed [165].

6. Conclusions

Among thermoplastic polymers, polyurethane, polyacrylonitrile, and other polymers have been used as matrices for shape memory graphene-based materials. Among thermosets, epoxy resins and graphene-based materials have been designed to observe the shape memory phenomenon. T_{trans}, strain recovery, strain fixity, and the cross-linking phenomenon have been studied as important factors to enhance the shape memory effect in these materials. The response towards a particular stimulus depends upon the formation of physical and covalent cross-linking in the polymer/graphene nanocomposites. Hence, this review article considered the shape memory polymer and graphene-derived nanocomposites. Shape memory nanocomposites of different polymers (polyurethane, epoxy, polylactic acid, etc.) with graphene and modified graphene were considered. In this regard, the significant thermally, electrically, and light-active stimuli-responsive materials were investigated. Shape memory polymer/graphene nanocomposites have potential for electronics, civil structures, aerospace, and biomedical fields. High-performance shape memory polymer/graphene nanocomposites need to be further explored for functional polymers, modified graphene, and actuation mechanisms. Combinations of different polymers with graphene and modified graphene nanocomposites revealed that the shape memory effect depends upon the optimization of the actuation force and a high shape recovery ratio. High graphene nanofiller loadings significantly influenced the shape recovery ratio. In addition, the presence of optimal nanofiller and the interaction with the polymer affect the actuation force required for the shape recovery of the nanocomposites. More actuation force is required to overcome the chain recovery due to strong interfacial interactions between the polymer chains and the graphene nanosheets. Future attempts must focus on the exploration of graphene-based nanocomposites for unexplored high-tech applications.

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