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Future Prospects: Shape Memory Features in Shape Memory Polymers and Their Corresponding Composites

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Abstract

Shape memory polymers and their related composites are formally known as SMPs and SMPCs have been classified as innovative categories of smart materials, in which they affected by a particular stimulus and consequently memorize the original shape. As one of the most vital feature of shape memory characteristics, Shape Memory Effect (SME) that been attracted significant attention from the shape memory researchers and scientists. On the other hands, there are abundant approaches can be implemented to actuate the SMPs and SMPCs deformation, whereby the features of the electro-or thermal response associated with the structural changes are predominant. In this chapter, a particular emphasis on how the incorporation of micro/nano-fillers and particles or fibers do affect in the SMP matrices, which it is intentionally carried out to improve the mechanical properties and their related shape memory features of various types of shape memory polymers. In the summary, the shape memory effect is been sustained to be an intrinsic feature for the SMPs and based on this property, the implementation of the SMPs have covered a wide range of applications according to the required functions and performances.

Keywords: shape memory polymer, shape memory effect, shape fixity, thermomechanical cyclic, CNTs, noble metals/fiber-based reinforcement

1. Introduction

As being a kind of smart materials, shape memory polymers (SMPs) and their composites (SMPCs) are significantly attracting consideration [1, 2]. An excellent type of polymers seems to have been disclosed to demonstrate shape memory attributes [2–4], however, the various characterizations or even analysis techniques of shape memory characteristics along with the changing circumstances among various scientists (see **Figure 1**), which they possess intention that the claimed characteristics of SMPs are not identical. Consequently, the correlation between shape memory characteristics and the structures is not entirely recognized for certain categories of SMPs. Thus, this can probably obstruct the growth and
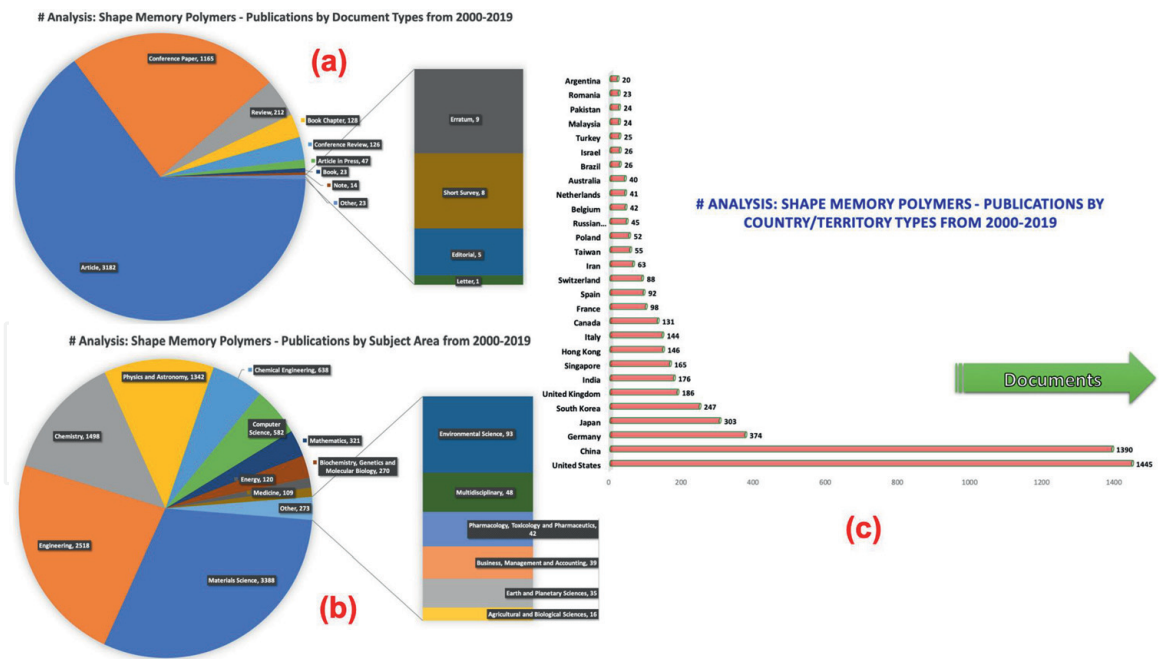
Figure 1.
Shape memory polymer publications based on (a) document type, (b) subject area, and (c) country/territory.
"Scopus source accessed on January, 2019".

development of high-performance of SMPs. Aside from that, in comparison with the prompt boost of the variety of SMPs, the main uses of SMPs lags much behind. One among the key factors is certainly utilized the characterization of the SMPs is not going to produce the extensive properties for scientists. For that reason, the analysis of SMPs is vital for the enhancement as well as implantations of SMPs. Based on the Scopus database, a literature analysis was carried out through using the keywords of "Shape Memory Polymers" and/or "SMPs" and the analysis graphs are presented in **Figure 1.** This chapter presents a basic overview up to the date the main employed characterization of the shape memory characteristics of polymers.

2. Shape memory effect in shape memory-alloys over -polymers

The shape memory effect in the shape memory alloys is typically execute based on the test temperature of the austenite ↔ martensite transformation temperature, in which it occurs with the deformation of the SMAs in the martensitic phase during the loading and unloading at temperatures below M_f. After heating these deformed alloys to a temperature above A_f, the austenite phase forms, and thus, the original shape is recovered. In addition, these temperatures are typically will be indicated based on the type of alloys. There are three main based-types of shape memory alloys; Titanium-based, Copper-based and Iron-based SMAs. **Figure 2(a)** shows a typical loading path $1 \rightarrow 2 \rightarrow 3 \rightarrow 4 \rightarrow 1$, wherein the property of SME is observed [5]. The parent phase transforms into the twined martensite $(1 \rightarrow 2)$ when it undergoes the cooling process. The stress induced detwinning and inelastic strains can occur when the materials are loaded $(2 \rightarrow 3)$. The maternsite phase is in the same state of the detwinned structure without obtaining any recovered inelastic strains even after the unloaded process $(3 \rightarrow 4)$. In the final step, the materials are returned to the original shape by recovering the inelastic strains after being heated above Af $(4 \rightarrow 1)$. On the other hands, there are two types of shape memory effects can be occurred in the SMAs, namely, one-way and two-way SME. On the contrary, the SME of SMPs (see **Figure 2b**) is mainly influenced by the presence of phases
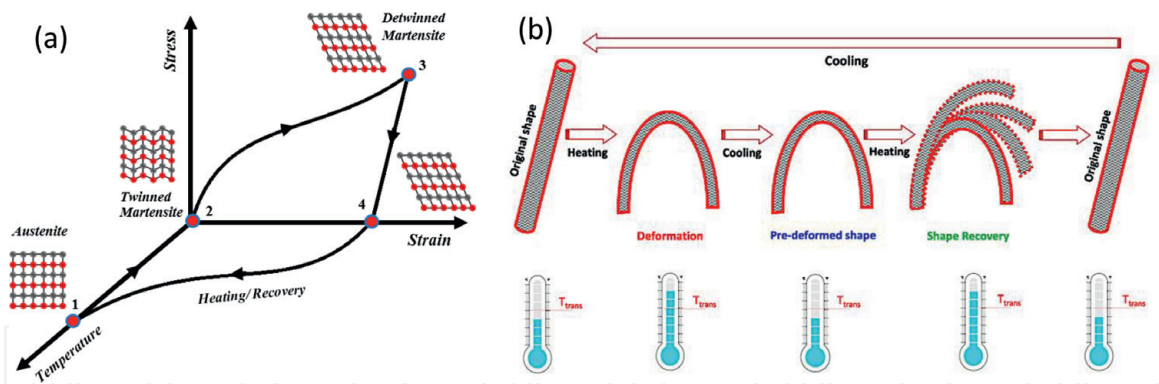
Figure 2.
Schematic diagram of (a) stress-strain-temperature for the involved crystallographic changes during the phenomena of SME [5], (b) one-way SME for SMPs.

Property	SMAs (Ti-based)	SMPs (Polystyrene)
Density/g.cm^{-3}	6–8	0.9–1.1
Deformation strain (%)	<8	≥800
Young Modulus at Temp.> Trans (GPa)	83	0.01–3
Recovery speed (min)	Based on type of alloy	<0.1 min- several min
Deformation stress (MPa)	50–200	1–3
Thermal conductivity (W.m^{-1}.K^{-1})	18	0.15–0.3
Cost ($)	~ 250 per pound	~ 10 per pound

Table 1.
The main comparison between the characteristics of SMAs over SMPs [1, 8].

that linked to the coiled or cross-linked polymer structure. The SMPs is deformed at a temperature below the glass temperature (Tg), and the percentage of deformation is mainly depending on molecular chains of polymer, in which they are controlled by the chemical composition and physical cross-linked structure of SMPs. After preheating the deformed polymers, these molecular chains are able to return back to the original coiled-shape structure. The shape-memory transformation varies according to the apparatus in which polymer molecules transpose between the restricted together with random entangled conformations. As comparison with the SMAs, the SMPs are able to exhibited only one-way SME, whereby, the SMPs deformation at the called phase "soft" only along with the incorporation of the external force [6]. The main benefits of SMPs over SMAs is dependent mainly on their inherent attributes, for instance, they are lower cost and/or density, easier manufacturing process associated with higher percentage of strain [7]. **Table 1** details the principal differences in the SMPs and SMAs characteristics, in which the SMPs are able to obtain up to 800% strain compared with the lower strain in the range of 0.1–20% for SMAs or other types of materials.

3. Shape memory polymer features and properties

To give details about the shape memory characteristics of polymers, a number of variables are essential. Initially, the variables can certainly reveal the characteristics of polymers. Following by the differentiation of them from other sorts of attributes of materials, shape memory capabilities are demonstrated by means of a variety of thermomechanical cyclic procedures. Consequently, the specifications must be able

to describe the natural shape memory functions too. Finally, the structure of the variables must look into the prospective purposes. With taken the consideration of these kinds of aspects, a number of variables were presented and also quantified [9–11]. The variables are presented in the following subsections:

3.1 Feature of shape fixity in SMPs

Like appears to have been explained in the foregoing parts, the shape memory behavior is initiated by heating the shape memory polymer to a temperature above the transformation temperature (T_{trans}) [12], it could actually cultivate considerable deformations and this can be mainly predetermined by cooling the materials to a temperature below the T_{trans}, whereby, this parameter was advocated to define the severity of a brief shape becoming fastened in a pattern of shape memorization [12–14]. It ought to be pointed out that the numerous perplexed utilizations as well as illustration appear in the characterization of SMPs. Regarding to the shape fixity, further sorts of capabilities for instance strain fixity [9] and also shape preservation [15, 16], stand for the exact same actual physical indication, in which the shape fixity (R_f) is comparable of the extension ratio of the predetermined deformation to the total deformation, which can be prearranged as:

$$Shape\ fixity = {permanent\ deformation}/{total\ deformation} \qquad (1)$$

Furthermore, the main mechanism of the shape fixity is attributed to the structure and thermomechanical conditions of the shape memory characteristics. The latter condition is significantly been implemented in shape fixity determination as well as the properties of shape memory materials. Wu et al. [17] shows the shape recovery of commercial ether-vinyl acetate copolymer (EVA) with a 300% of pre-stretching at the room temperature, as shown in **Figure 3**. It was revealed that as the number of cycles increased the strain recovery reduced, in which the residual strain starts from 136% and dropped to 112% after 20 min, and within the 9 h, it reaches to
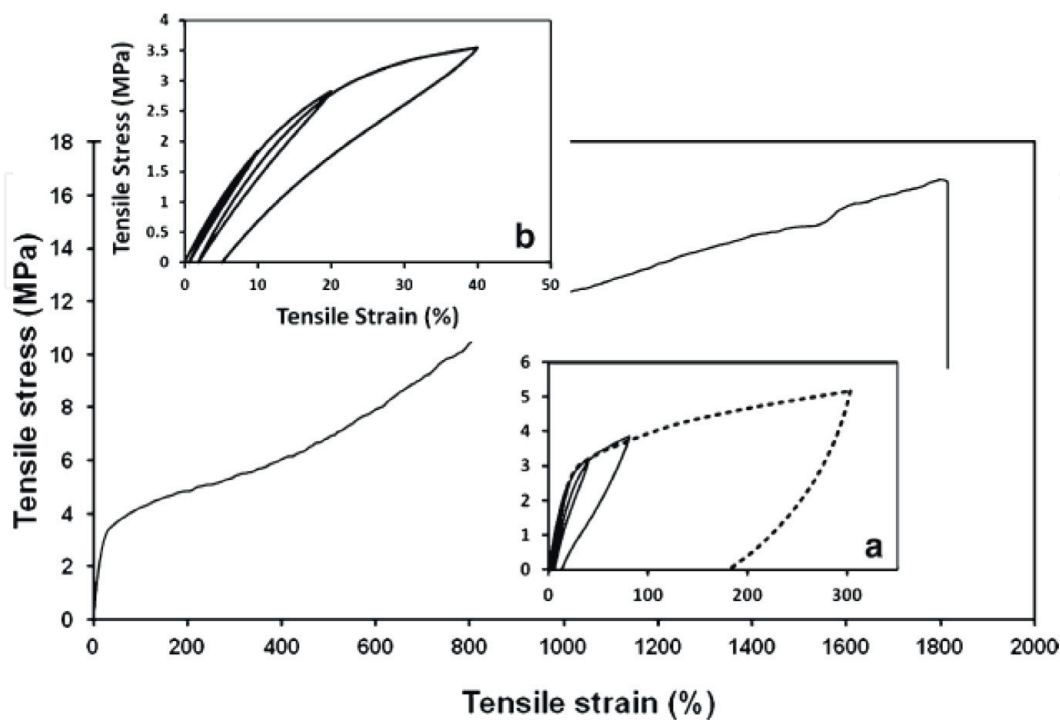

Figure 3.
Relationship curve of stress-strain of EVA at the room temperature. Insets: (a) multiple cycles at different strains; (b) cyclic test [17].

94% and with increasing the time to 72 h, it ends up with 88%. It would be proven that as the time increased; the rapid creep turns to be gradual creep within the first 9 h. Therefore, a long term of the shape fixity ratio was described according to the value of residual strain. Julie et al. [18] demonstrated the shape fixity (R_f) of the epoxy network based on the torsion test, whereby the R_f was found based on the ratio of angle of torsion after unloading to the angle of torsion after loading. It was found that the shape fixity of epoxy within a dimension of $100 \times 10 \times 1$ mm^3 is about 95% at a deformation angle of 360°.

3.2 Feature of shape recovery in SMPs

Shape memory recovery (R_r) is mainly reflect the ability of any substance to recover the memorized shape after being deformed at low temperature and subsequently heated above the transformation temperature (T_{trans}) [9, 12, 13]. It is significant to notice that, in a shape recovery event, the full strain energy is emitted by means of the two-recovery strain and stress. To a first approximation, nevertheless, the recovery stress to stain ratio is consistent for an identical material [19]. The stored strain energy, alternatively, ought to be influenced by the internal material energy needed in the deformation, despite the fact that the particular relationship is not recognized, in another words, the quantity of energy loss throughout the shape-fixing stage is not identified. Hence, there needs to be a minimum of a qualitative correlation between recovery stress together with deformation energy (or even input energy). Tobushi et al. [9] and Kim and coworkers [13] have performed the thermomechanical test with multi-cycles in purpose of evaluating the performance of SMPs and found the main determination of strain/shape recovery, in which can be calculated using the following interpretation:

$$Shape\ recovery = \frac{Deformation\ recovered\ in\ a\ certain\ cycle}{Total\ deformation\ in\ one\ cycle} \times 100\% \qquad (2)$$

While the shape recovery rate was determined by Li and Larock [20] after been utilized a bending test on SMPs and came with the following formula:

$$Shape\ recovery = \frac{Deformation\ recovered\ within\ heating\ process}{Fixed\ deformation} \times 100\%. \qquad (3)$$

According the above-mentioned equations of (2) and (3), it can be proven that there are different mechanisms referring to the shape recovery in different perspectives. Julie et al. [18] obtained that the kinematic of the shape recovery of epoxy networks is a function of the applied deformation angle. It was found that a complete recovery (100%) was obtained and as the deformation increased the recovery ratio tends to decreased. They also found that the lower heating rate is able to attain a full recovery compared with the high heating rate. As both of two types of recovery are mainly related to the molecular mobility that been coincides with the variation of polymer viscoelasticity properties. They were also proven that the torsion test provides a useful interpretation on the molecular mobility with the glass transition when a uniformed deformation is applied. Moreover, the speed of recovery process and deformation of recovery speed were named by Li et al. [20] and Luo et al. [21], respectively, thereby, both terminologies were reflected the shape memory characteristics of polymers. The shape recovery process of different types of shape memory polymers was studies by Liu et al. [22] using video camera records within a rate of 20 frames per seconds. The results of their experiment revealed that the polymer was capable to obtain a full recovery after 0.7 s. Whilst, Luo et al. [21]

found that the curve of shape recovery of SMPs as function of temperature and then the shape recovery speed was determined based on the following equation:

$$V_r = \frac{dR_r}{dT} \times \frac{dT}{dt} \qquad (4)$$

whereas V_r is representing the shape recovery speed, dR/dT is the ratio of shape recovery Vs temperature, and dT/dt is the heating rate. On the other hands, Tobushi et al. [9], Takahashi et al. [23], and Kim [13] were performed the tensile test as thermomechanical cycling via a specially designed machine as shown in **Figure 4a** to study the shape memory characteristics of polymers, in which the tensile test process was isolated under a certain temperature and an extensometer was attached to record the stress–strain data and a programmable software was used to plot the final behavior of one or multicycle of relationship of stress versus strain versus temperature and then the shape recovery and memory effect were determined. **Figure 4b** shows the final curve behavior of shape memory effect, thereby the curve can be classified into four stages, as the first stage, the polymer sample is heated to a higher temperature (i.e. > transition temperature), which often temperature would be in range of 15–25°C. Second stage describes the strain behavior that maintained a constant strain (ε_m) followed with a cooling to a temperature lower than the transition temperature (room temperature) to obtain the permanent shape. The unloading process and the elastic recovery stress turns to reach a zero value at a certain strain (ε_u), as presented in stage 3. An external heating process was applied at a higher temperature to recover the original shape with a minimum value of residual strain (ε_p) as demonstrated in stage 1′ or 4 based on the type of polymer.

Tobushi et al. [21] have demonstrated the shape recovery (R_r) of polymer at different temperatures and the R_r can be calculated based on the following equation:

$$R_r(N) = \left[\frac{\varepsilon_m - \varepsilon_p(N)}{\varepsilon_m - \varepsilon_p(N-1)} \right] \times 100\% \qquad (5)$$

The $R_r(N)$, ε_m, and ε_p are the shape recovery, residual strain, and plastic strain, respectively under a number of cycles (N), in which found that as the number of
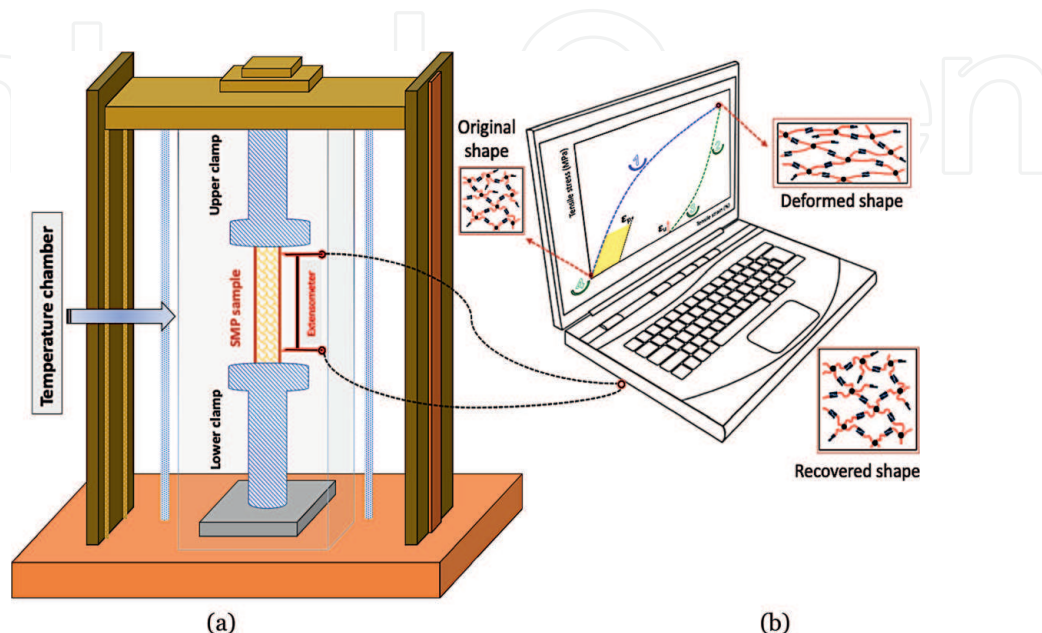

Figure 4.
(a) Tensile test as thermomechanical cyclic machine; (b) shape memory effect curve [9, 13, 23].

cycles increased, the shape recovery maintained to attain 100%. While, Kim and Lee [13] have found that the shape recovery using the following equation:

$$R_r(N) = \left[\frac{\varepsilon_m - \varepsilon_p(N)}{\varepsilon_m}\right] \times 100\% \qquad (6)$$

Thereby, the shape recovery tends to decrease as the number of cycles increased and shown a stabilized behavior after a number of cycles. Another studies by Liu et al., [22] Lin and Chen [24] and Li and Larock [20] found that the employing of the bending test is much easier and more approachable than tensile test for the thermomechanical test. **Figure 5** shows the mechanism of thermomechanical cycles using the bending test, whereas the shape memory polymer sample with a strip shape is bent to angle θ_{max} at a higher temperature > transition temperature (T_{trans}). The deformed sample followed by cooling process at a temperature < T_{trans}, in which the sample been unloaded and shape recovery started to an angle represented by θ_{fixed}. However, with preheating the deformed sample, the original shape recovered gradually associated with number of $\theta(T)$ recorded. At the final stage, the sample turns to recover the final shape at angle of θ_{final}. Therefore, the shape recovery is determined using the following equation:

$$R_r = \left[\frac{\theta_{fixed} - \theta_{final}}{\theta_{fixed}}\right] \times 100\% \qquad (7)$$

A compression test was also utilized to obtain the shape memory behavior of polyurethane shape memory polymer foam MF5520 at a nominal glass temperature of 63°C. The shape recovery behavior was obtained as the foams were compressed at T > Tg, then cool it to the room temperature, and finally the shape recovery SMP foam was investigated upon heating after different period time of hibernation. The results revealed that at 80 and 93.4% pre-strain and 1 N applied load, the recovery curves acquired the same trends without a full recovery as shown in **Figure 6a** and **b**, conversely, removing the applied load, the shape recovery was attained. On the other hands, the sample without hibernation displayed a reduction in the shape recovery as the load increased. Gall et al. [25] was investigated the shape recovery of the thermosetting polymer (CTD-DP7 SMP) along with their corresponding composites using a Dynamic Mechanical Analyzer (DMA). The prepared samples were placed in a three-point fixture and the tip of probe was in contacted to the inner surface of SMP
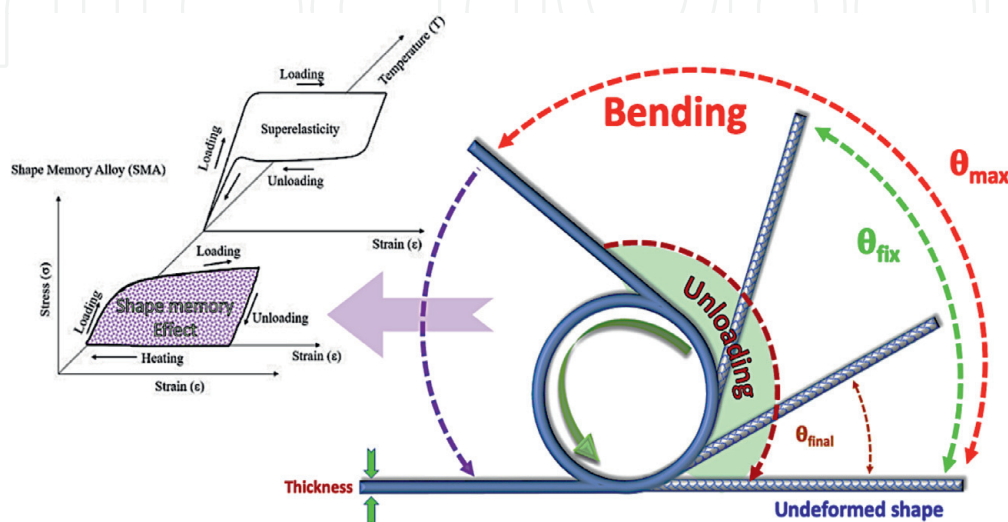

Figure 5.
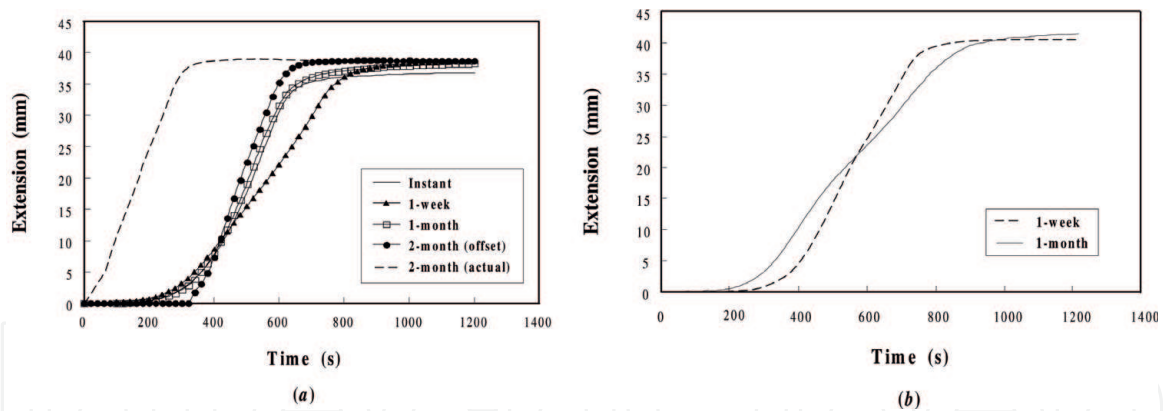*Schematic drawing of bending test as thermomechanical cycles test.*

Figure 6.
Extension behavior representing the shape recovery SMP foams hibernated at different time for the pre-strain;
(a) 80% and (b) 93.4%.

sample. With fixed probe contact, the shape recovery was recorded as the tempera-
ture was increased.

4. Parameters affects the shape memory effect in SMPs

4.1 Thermo-mechanical behavior

Lendlein and Kelch [12] described that the behavior of the shape memory for
SMPs is not only linked to the polymer properties, but it is also mainly controlled by
the structure and morphology associated with the manufacturing processes. There-
tofore, the shape memory effect of any type of polymers is demonstrating the
thermomechanical cyclic performance includes, shape recovery, deformation, and
shape fixing and each of these processes associated with the condition of
thermomechanical process are able to vary the shape memorization and thus affect
the shape memory characteristics. Hence, it is essential for further development and
advanced applications of SMPs to give a complete characterization. For instance,
Wang et al. [26] have carried out the thermomechanical cyclic for SMP composite
using the cyclic tensile test within 30 mm/min as displacement rate for 5 cycles, as
shown in **Figure 7a** and **b**. They revealed in their results that there is a huge
hysteresis between 1st and 2nd cycle, while these differences are attainment smaller
from 2nd, 3rd, 4th and 5th cycles. These variations are mainly attributed to the
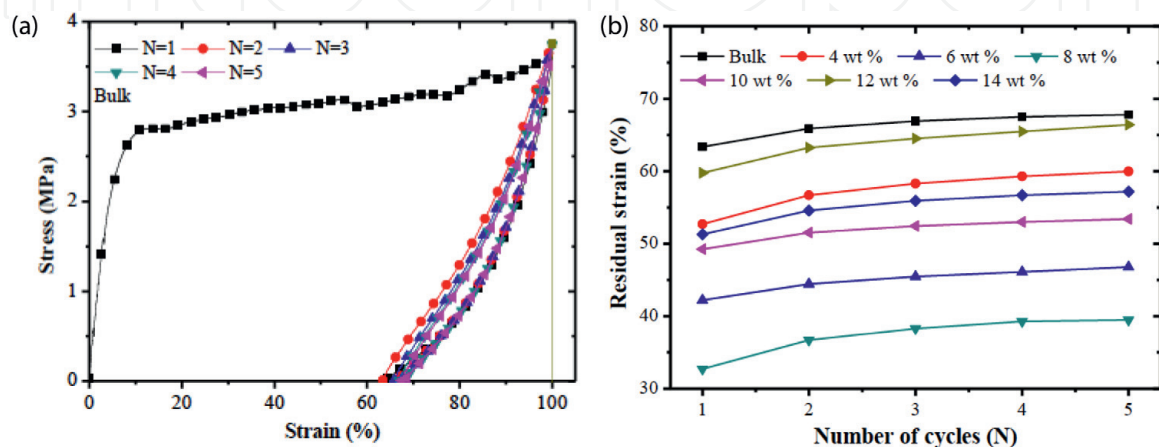existence of deformation, composite structure failure along with initial training

Figure 7.
(a) Stress-strain curves of SMP composite under different loading-unloading cycles; (b) residual strain versus
number of cycles of the reinforced SMP with different percentage of chopped carbon fiber [26].

effect for the 1st cycle. **Figure 7b** illustrates that the variation of residual strain at the room temperature versus the number of cycles, in which the residual strain tends to increased precipitously with final stabilization behavior as the number of cycles increased due to the resistance of the modified particles against the deformation, which will be explained in details in Section 3.2.

Pieczyska et al. [27] have been studied the thermomechanical properties of polyurethane theoretically and experimentally under different mechanical loadings at temperature of 20°C above and below T_g at a strain rate of 2 per second within a strain range of 0.6/s. It was observed that when the temperature been slightly dropped, the thermoelastic effect stepped affected. On the other hands, the thermal images (see in **Figure 8**) are referring to the variations in the strain vales during the loading-unloading process, whereby the uniform distribution of the temperature replicated the deformation process in a macroscopically homogenously presented.

4.2 Particle/fiber reinforcement

The design of SMPs and SMPCs thermomechanical behavior can be vary based on the changes in the polymer molecular structure and/or addition of functional particles or fillers in purpose of forming multi-phases composite SMPs. The reinforcement categorization for the SMP composite can be inserted under the particle/filler according to the type of applications. There are various types of particles can be incorporated with SMP, for instance, Silicon carbide (SiC), carbon nanotubes (CNT), nickel, carbon black (CB), clay, and Fe_3O_4 [28–31] along with some others different fibers based on the application requirements [32, 33]. Thus, these types of additions or reinforcements are trigger to enhance the electrical and mechanical properties of SMPs.
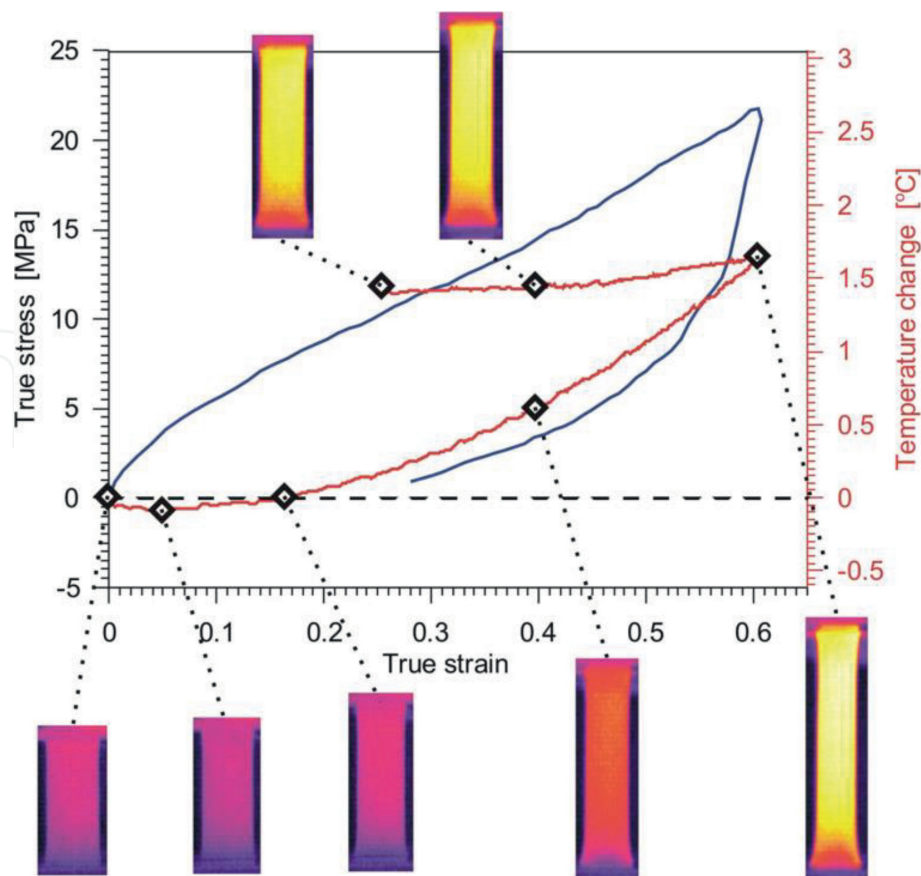

Figure 8.
True stress-strain curve during loading and unloading of PU-SMP along with thermal images that represented different values of strain [27].

4.2.1 Carbon-based reinforcement

Thermally induced shape memory polymers have been characterized as a remarkable substance with a high recovery and shape memory effect is required, however, their mechanical properties such as modulus and strength are still low. Therefore, the incorporation of carbon nanotube (CNTs) with different types of polymer is essential and their inadequate properties are able to improve after a certain modification been considered [8, 34, 35]. On the other hands, the carbon nanotubes have the presented a potentially implementation the nanoelectronics devices, for example, electrochemical energy storage and artificial muscles. According to the above-mentioned deliberations, Kai Yu et al. [36] have studied the effect of carbon nanotubes on the shape memory effect of SMPs after being exposure to microwave radiation. It was found that the CNTs particles have been absorbed the electromagnetic radiation and converted to be an internal heating source, thus lead to induce a shape recovery for SMCs, as shown in **Figure 9**. Furthermore, increasing the frequency of radiation and/or amount of CNTs lead to enhance the shape recovery, whereas the SMPs composite within 3 and 5 wt.% of carbon nanotubes have shown a fully recovery in their shape, while, the SMPs within 1 wt.% addition has a lowered recovery with 80% less than high amount and the unrecovered shape has been resulted due to the insufficient amount of radiation to overcome the caused friction between the CNTs and polymer matrix. From the same point of view, another research was conducted poly(vinyl alcohol) (PVA) filled by CNTs, in which the results revealed that there is a wide boarding in the glass transition temperature and the initiated stress during the recovery was almost double value compared with the conventional polymer [37]. Conversely, Raja et al. [38] found that there no apparent for the shape recovery of PU/PVDF polymer blend nanocomposites after been deformed in "U" shape and preheated using a hot water with a temperature of 60°C for 2 h and followed by a direct quenching in cold water, as demonstrated in **Figure 10a**. An external heating source using a DC controller with a 40 V was attached to the ends of modified polymer strips to activate/initiate the shape recovery. In spite of this, the modified PU/PVDF nanocomposites with CNTs fillers, namely as PUPF-NTM10 has been recovered after 15 s and others samples filled with pristine CNT, namely as PUPF-NTP10 has been recovered the complete shape after 30 s with the external applying of electrical impulse. It was also found that repeating the shape memory test (i.e. increase the number of cycles) led to reduce the shape memory ratio as depicted in **Figure 10b**.

The influence of multi-walls CNTs on the shape memory effect of epoxy nanocomposites was deliberate by Abishera et al. [39] as implementation in the self-healing systems applications under different programming conditions. It was
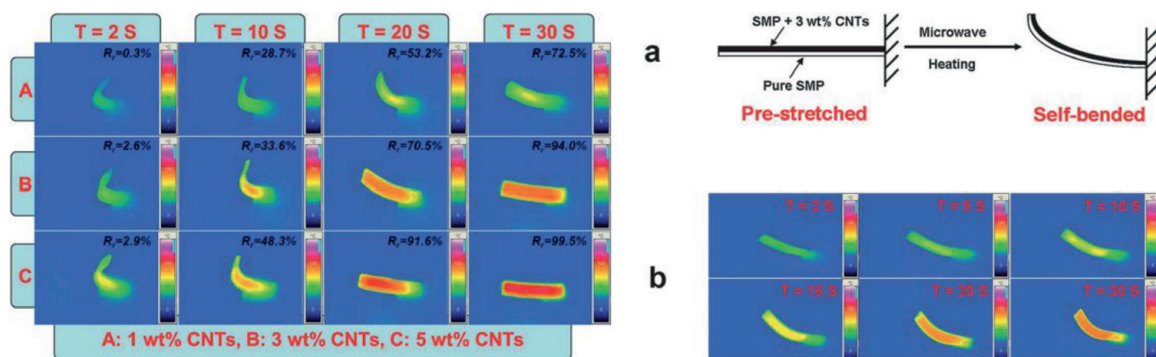

Figure 9.
Sequence of the shape recovery process of the SMP composites under microwave radiation (2.45 GHz): (a) deformation process and (b) modified SMP/CB/CNT sample shape recovery and temperature distributions.
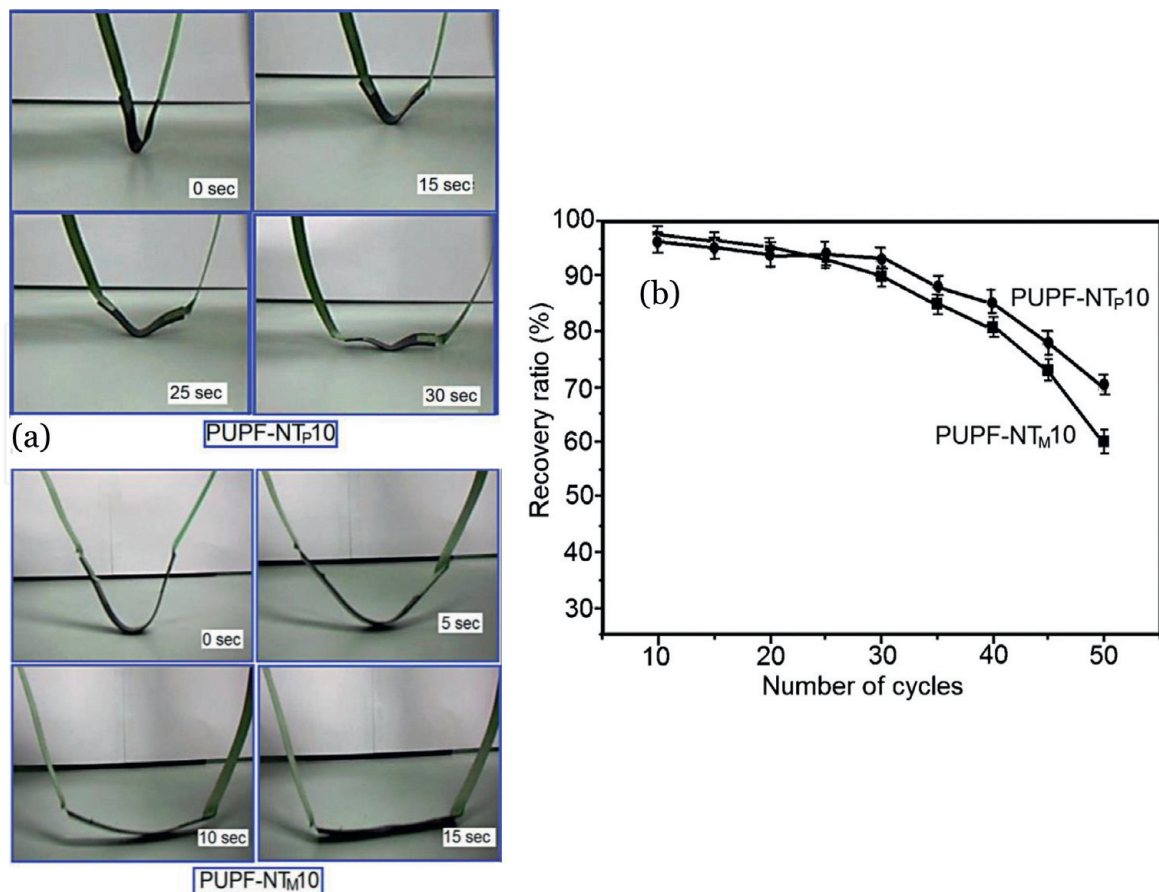
Figure 10.
(a) SME of unmodified PUPF-NTP10 pristine and CNT modified PUPF-NTS10 filled with PU/PVDF nanocomposites; (b) recovery ratio versus number of cycles [38].

revealed that the changing in the programming conditions obtained an excellent shape memory behavior, as well as, the incorporation of the multi-walls of CNTs has indicated an improvement in the young modulus, strength, recovery speed and shape fixity of epoxy associated with drops in the failure strain. Qi et al. [40] have investigated the shape memory properties of polylactide (PLA)/thermoplastic poly (ether)urethane (TPU) composites after been reinforced with carbon black (CB) nanoparticles within the blending ratio of 70:30 by weight. Due to the continuous phase of thermoplastic poly(ether)urethane (TPU), an outstanding shape memory behavior was acquired for the novel ternary structure of PLA70/TPU30/CB, wherein it resulted in the occurrence of the persuasive recovery driving force. In addition, the addition of CB with different percentages displays a slight improvement in the shape fixing ratio (R_f) of 90% (see in **Figure 11**), this may relate to the elongated TPU phase retraction, in which resulted in a total rigid of polylactide phase at 25°C. As the temperature increased more than Tg, the amorphous chains of PLA started to move and thus release constrained TPU phase. The principal contributor in the enhancement of shape recovery ratio (Rr) is the strong pliability of TPU phase and addition of CB, as it was shown only 59% for the binary phase of PLA70/TPU30 and increased to 80.2% as the CB was added, as shown in **Figure 11a** and **b**. Moreover, the increment in the heating time under a consistent 30 V led to increase the shape memory ratio of the ternary phases of PLA70/TPU30/CB6 and PLA70/TPU30/CB8, as shown in **Figure 11c**. The fastest shape recovery response for PLA70/TPU30 after 8 wt.% addition of CB with 90% ratio in 80 s, on the other hands, the PLA70/TPU30 with 6 wt.% of CB approached the same ratio in 150 s, as illustrated in **Figure 11d**. Haibao Lu et al. [41] presented the shape memory behavior of shape memory polymer nanocomposite (SMPs) after carbon nanotube
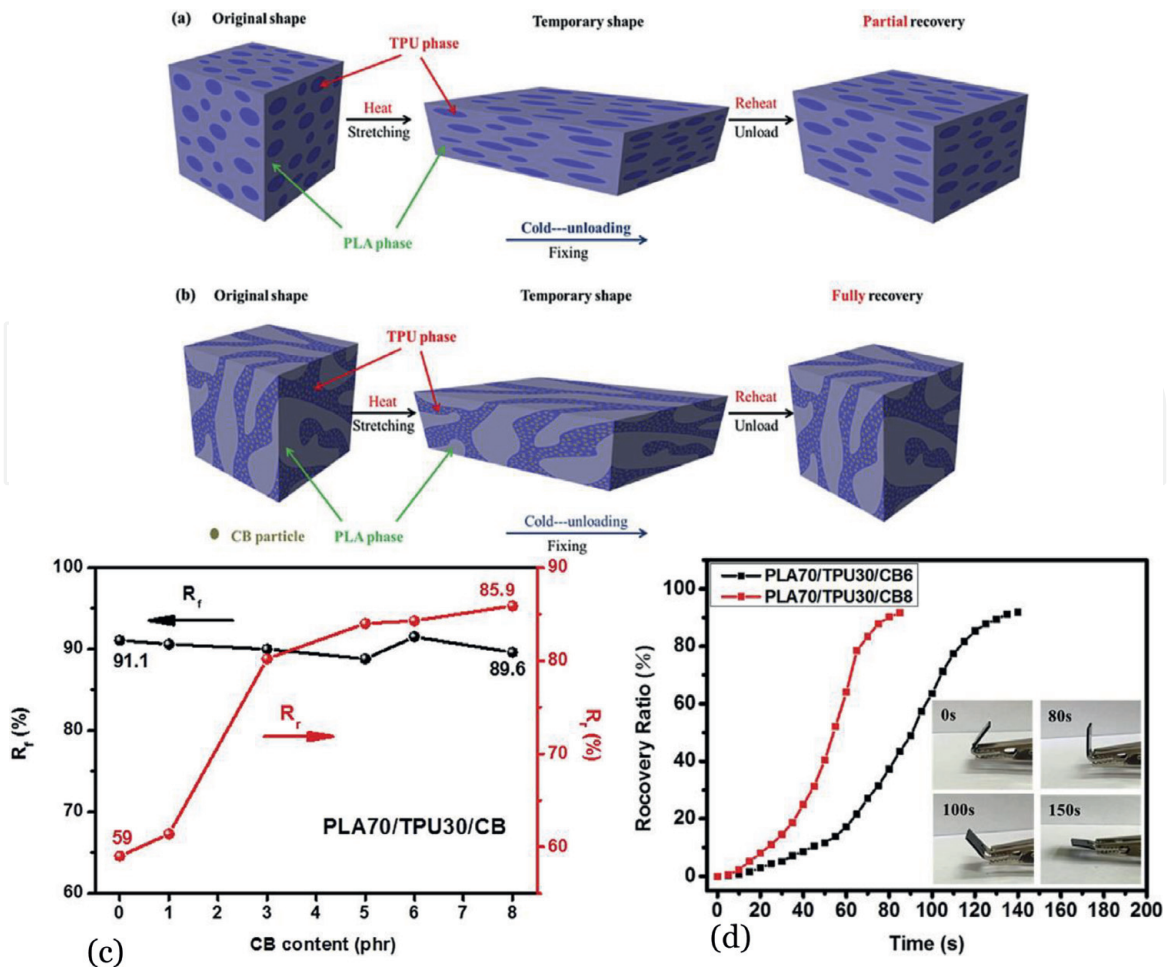
Figure 11.
Representation design of the shape recovery of (a) binary blends of PLA70/TPU30 and (b) ternary blends of PLA70/TPU30/CB5; (c) recovery ratio (R_r) versus CB contents and (d) shape fixing (R_f) versus time PLA70/TPU30/CB$_{6/8}$ [40].
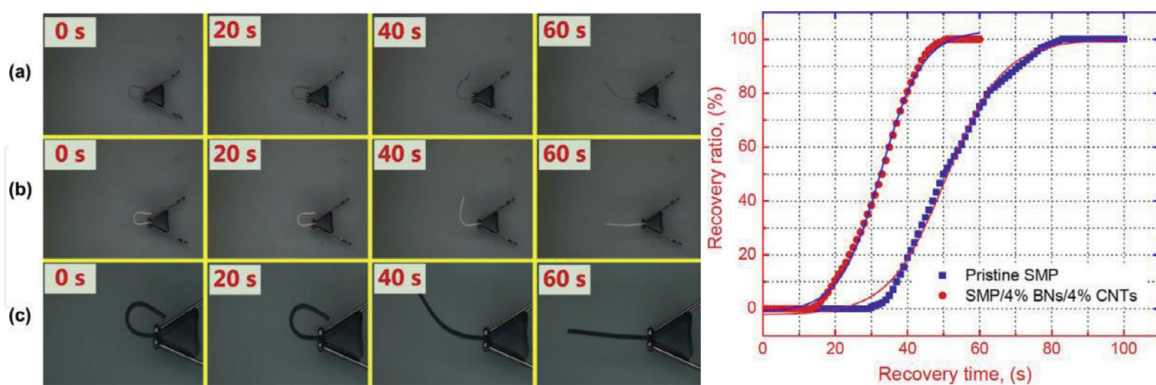

Figure 12.
Optical shape recovery of (a) pristine SMP, (b) nanocomposite SMP reinforced with 4 wt.% of BN, (c) nanocomposite SMP reinforced with 4 wt.% of BN and CNTs, and (d) shape recovery ratio versus time for pristine SMP with and without BN/CNT reinforcements [41].

and boron nitride additions using a bending testing with an Infrared light-induced as heating source. A full recovery 100% in 60 s was recorded for the modified SMP with 4 wt.% boron nitride and CNTs, as shown in **Figure 12(a-c)**, in which the unmodified sample was obtained shape recovery lower than 80% in the same exposure heating time, as shown in **Figure 12d**. This kind of improvements is mainly attributed to the particle of boron nitride that have been improved the thermal conductivity through the facilities of the heat transfer in the composite

polymers [44, 45], and thus, CNTs and boron nitride additions have been drastically superior the infrared light-induced shape recovery.

4.2.2 Noble metals-based reinforcement

The effects of the nanosized noble metals-based, such as gold (Au) and silver (Ag) nano-particles/wires on the structure and properties of SMPs have shown a great interest for the researchers and scientists as a multi-responsive shape memory polymer in composite form [8, 42, 43]. Due to the large surface and high plasmonic resonance, these nanosized metals been offered a structure that able to absorb the specific wavelength and convert it into heat energy, thus produce a remarkable type of polymer with high shape actuation and wavelength activation.

4.2.2.1 Ag addition

The cross-link structure between the polymer and ion-metals as ligand coordination have shown a promising method to produce shape memory polymers with noteworthy properties. An isonicotinate-functionalized polyester (PIE) was studied by Wang et al. [44], which they described the effect of silver (Ag) addition to the coordination of the polymer network structure to produce the strip specimens. The shape memory effect was measured based on the DMA test, whereas the strips was heated to 50°C for 1 min, followed by bending process into spiral shape. The shape fixation was carried out by frozen the deformed strips at 0°C for 1 min. Lastly, the strips were recovered the initial shape after being heated to 40°C using hot air. It was found that the modified strip was shown an excellent shape recovery at 37°C for 60 s, as shown in **Figure 13**. Another study on producing a film by Lu et al. [45], the shape memory behavior was investigated using bending test, where the sample was bent in U-like shape at 160°C and cooled to the room temperature (which it was about 22°C). it was found that the sample contains the Ag particles decorated GO has recovered 100% after 36 s within an electric power of 3.87 Watt, as shown in **Figure 14**.

The behavior of the electro-response shape recovery of the surface modified of SMP with Ag nanowires layer was investigated by Luo et al. [46]. The recoded data of shape recovery versus time as shown in **Figure 15a**, the results exhibited that the Ag modified samples were not only able to recover the full initial shape but also with fastest speed recovery in shorten time compared with un-modified samples. It was also found that the higher addition of Ag led to reduce the shape recovery, and therefore, the external heating source was essential. As the heating increase, the thermal transition was occurred and consequently the shape recovery developed. **Figure 15b** shows that with the applied external heat of 5 volt, the bent angle turns to be change within 3 s, this because the presence of nanofillers is capable of conductive network reduction and subsequently increase the resistivity of the strain sensitivity and conductivity [47].
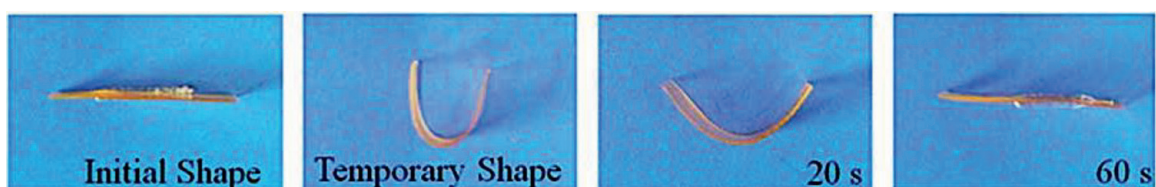

Figure 13.
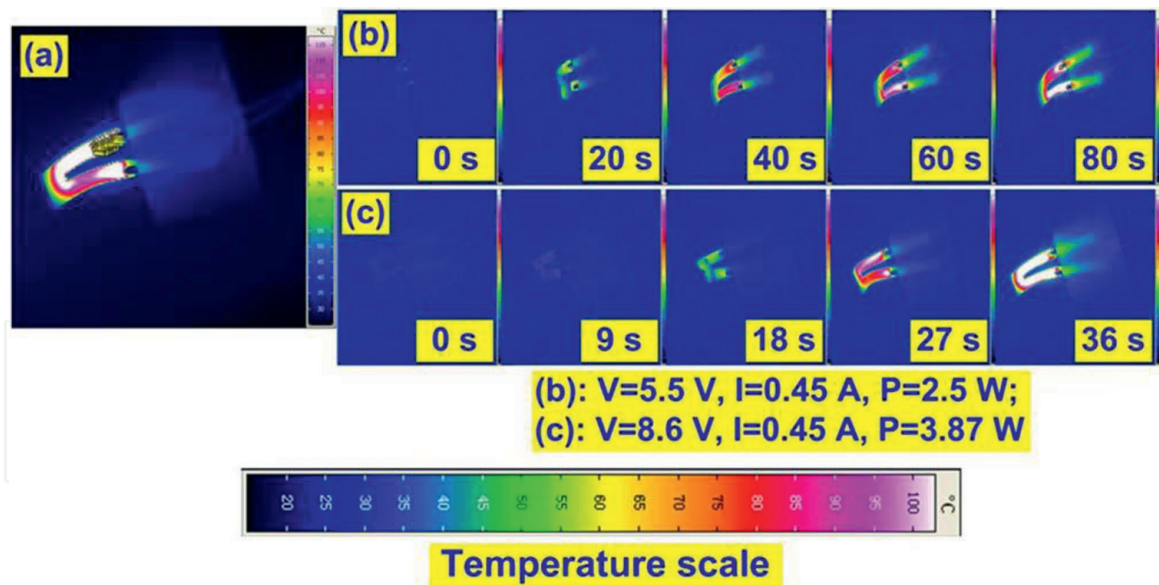*Shape memory effect of Ag–PIE in PBS at 37°C [44].*

Figure 14.
Shape memory effect of SMPs composite induced by joule heating based on (a) trend scale of temperature, (b) reinforcement of carbon fiber grafted with graphene oxide, (c) reinforcement of carbon fiber grafted with nanoparticles of Ag associated with GO decoration [45].
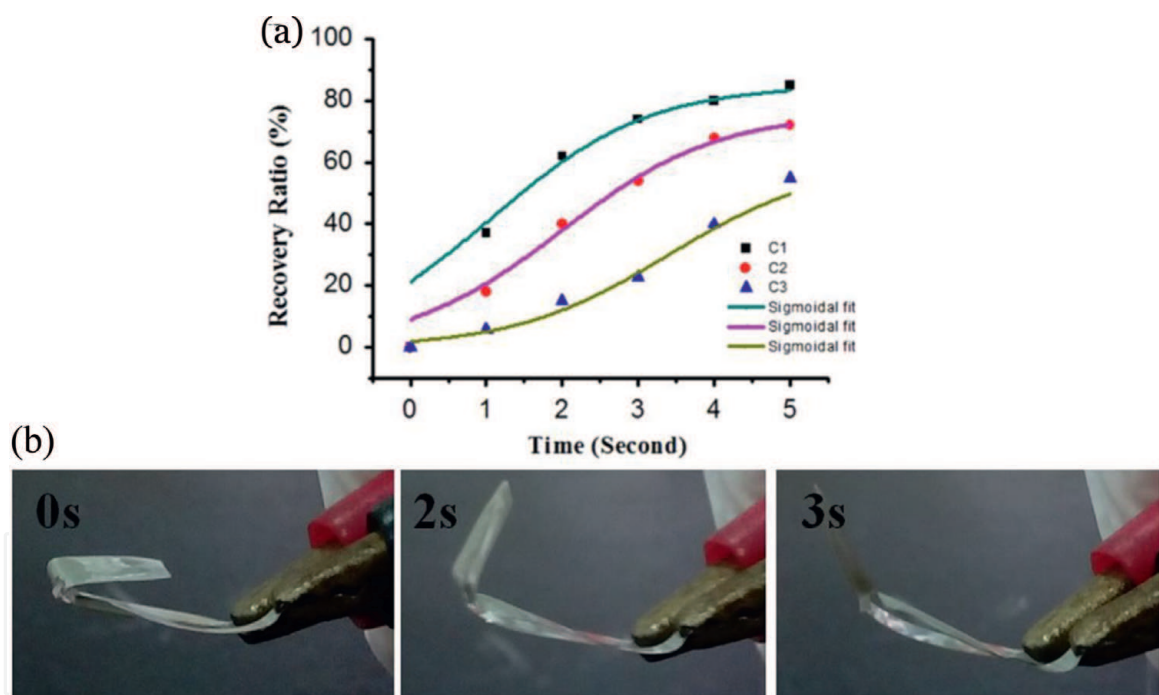

Figure 15.
(a) Shape recovery ratio versus exposure time under a voltage of 5 V; (b) shape recovery demonstration under different exposure times [46].

4.2.2.2 Au addition

A semi-crystalline PEO_{20K} polymer was prepared by Zhang et al. [42] using a cross-linked loaded with nano-particles, whereas found that the gold addition is not utilized to obtain/control the shape memory effect, however, it has also enhanced the properties of self-healing of SMPs. An exceptional concern in such a form of substances is the fact that SMPs necessitate an everlasting system structure (characteristically cross-linked) that could be in confrontation with the substantial a string movability and also inter-diffusion intended for the manufacturing of self-healing properties (SHP) polymers. Within this purpose, the configuration of the
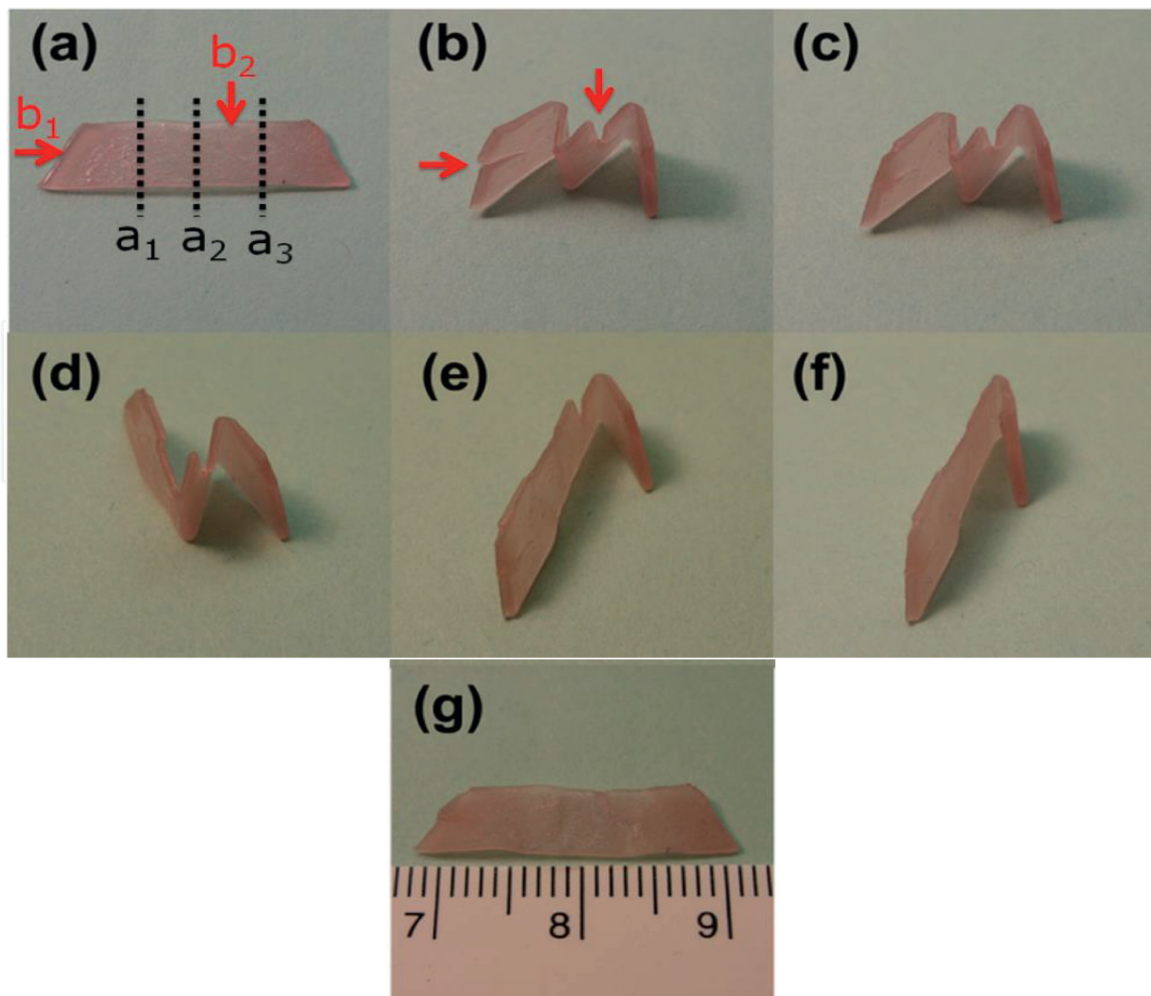
Figure 16.
Optical healing/recovery procedure of a film made from a cross-linked PEO/AuNP using light controlled as a source of heating: (a) Original film, (b) temporary shape obtained by folding the film along the lines a1, a2, a3 at 80 °C followed by cooling to room temperature; then two cuts were made as indicated by red arrows (b1 and b2 in photo a), (c) the b1 cut was healed by exposing the crack to laser (12 W/cm2) for 5 s; (d) the first unbending after 10 seconds laser scanning along the fold a1 at a power of 6 W/cm2, followed by the second unbending under the same condition along the fold a2; (e) the other cut b2 remained in the film of an intermediate temporary shape; (f) the cut b2 was optically healed under the same condition as for the cut b1; (g) the third light-triggered unbending along the fold a3 completed the permanent shape recovery.

SHP it can be more suitable to develop materials upon one single-polymer with the two light-controlled shape-memory and optical recovery features. It was found that the addition of small amount of AuNPs of 0.003 wt.% to the cross-linked poly (ethylene oxide) (PEO) films was sufficient enough to deliberate the main shape memory properties of produced films. The shape memory effect of the produced films is shown in **Figure 16**.

In 2009, Hribar et al. [48] have been developed a modified polymer network, namely β-amino esters, whereas the modified nanocomposites SMPs has demonstrated shape memory effect using IR light as heating source within a temperature above glass transition temperature (Tg). The results exhibited that the prompt shape recovery was spotted until the transformation turn out the path of the beam associated with a thermal transition in the polymer from glassy ⇒ rubbery networks. There are new perspectives for the actuated and functional shape memory polymer that recently been used in different application, for examples: dry adhesives, panels of light-tracking solar, light-guided for smart windows and even actuators. Zheng et al. [49] have been produced a light responsive SMPs as micropillar with a diameter of 10 μm mixed with 0.1–0.2 mol% of AuNRs in a hexagonal array
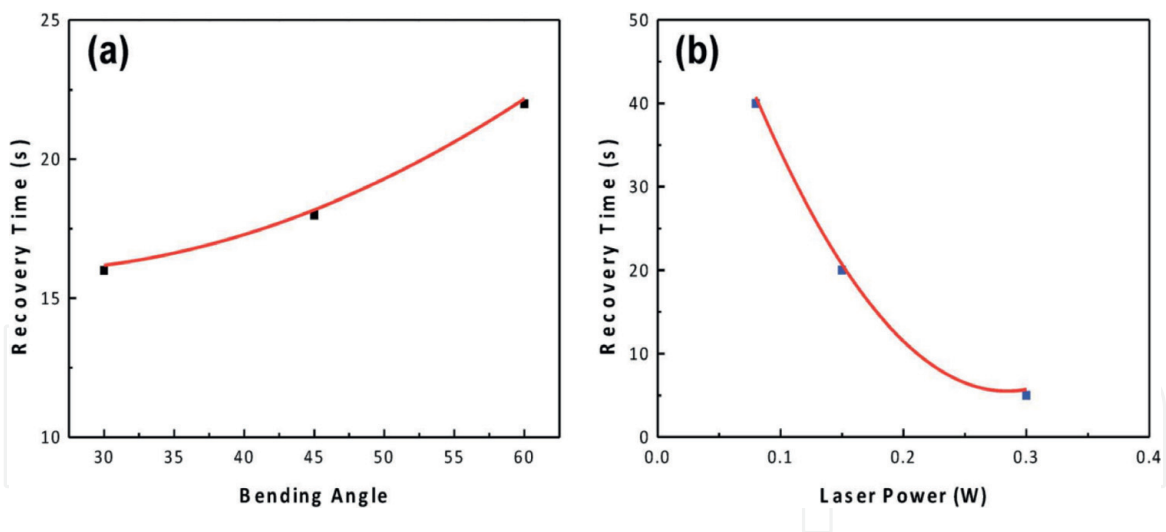
Figure 17.
Recovery time versus (a) bending angle and (b) laser power, of the reinforced SMPs with AuNRs at 45° pillar angle [49].

using poly(dimethylsiloxane) mold as replica molding. To obtain a temporary shape, the pillars were bent at different angles (θ = 30, 45, and 60°) at a temperature above the glass transition temperature (i.e., T = 20°C) and then following by cooling process to the room temperature, as shown in **Figure 17a** and **b**. A heating source was applied using a green laser with a wave-length of 532 nm for different period of times and the shape recovery was monitored by optical microscopy. The results revealed that within 0.08 W, as a laser power, the pillars need 40 s to recovery their temporary shape, however, with raising the laser power to 0.3 W, they grabbed the original shape within only 5 s. It was also found that when the pillars are ground to be collapsed, the shape recovery will not be able to obtain a full recovery (i.e.,100%), and this may attribute to the large force of adhesion between the substrate and pillars, in which it has particularly a higher value than the stored elastic energy of the deformed pillars.
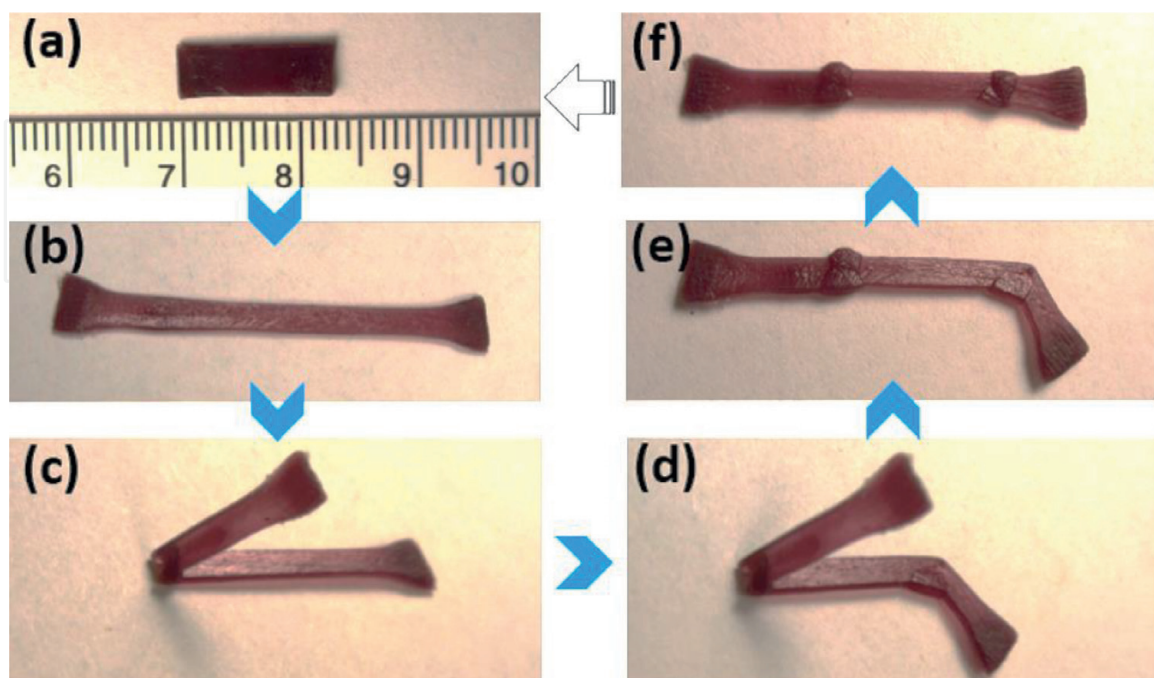

Figure 18.
Optical observation of the shape transformation of cross-linked PEO film reinforced with AuNPs using a laser as a temperature source; (a) original film, (b) deformed sample at 80 °C, (c) large-out of plane bending, (d) gradient chain relaxation and (e,f) shape recovery after applying the laser heating source.

A cross-linked poly(ethylene oxide) (PEO) loaded with 0.5 wt.% AuNPs was experienced to bending loading and unloading along with an exposure to a laser source within a wave-length of 532 nm and power of 0.15 W [50]. The deformation process involved a stretching of the modified PEO/AuNPs to 90% (approximately 200% strain) at 80°C above the Tm, followed by cooling to the room temperature. The top surface of the film was exposure to the laser in purpose of maintaining the anisotropic relaxation in the polymer chains, followed by cooling a temperature below Tm and consequently, resulted in fixing the temporary shape, as shown in **Figure 18(a–f)**, in which the main principles of the technology to exploited the unique shape memory effect property in numerous types of photothermal based shape memory polymers. On the other hands, the adaption of the light polarization has also shown a significant effect on the shape memory property, in which it led to control the photo-based thermal effect, as shown in 2013 study by Zhang et al. [51], whereas the cross-linked network polymer of PVA containing of 0.02 wt.% AuNRs film was stretched and heated to a temperature of 80°C, i.e., above the T_g to maintain the shape permanent transformation, as shown in **Figure 19(a–c)**. A laser in a linear polarization featured with a wave-length of 785 nm and 0.2 W/cm^2 was applied. The results revealed that there is no shape recovery was obtained when the polarization was perpendicular to the deformed film, even after 2 minutes of exposure. Whilst, the deformed shape was fully recovered in 10 s when the polarization turned to be in parallel direction aligned with stretching direction. The reason behind the directional effects can be elucidated that there was no longitudinal absorption in the perpendicular direction, thus resulted with no heat released. However, with the parallel direction, the longitudinal absorption reached to the maximum, and subsequently, the temperature increased above the glass transition temperature (T_g) that lead to reactivate the shape recovery.
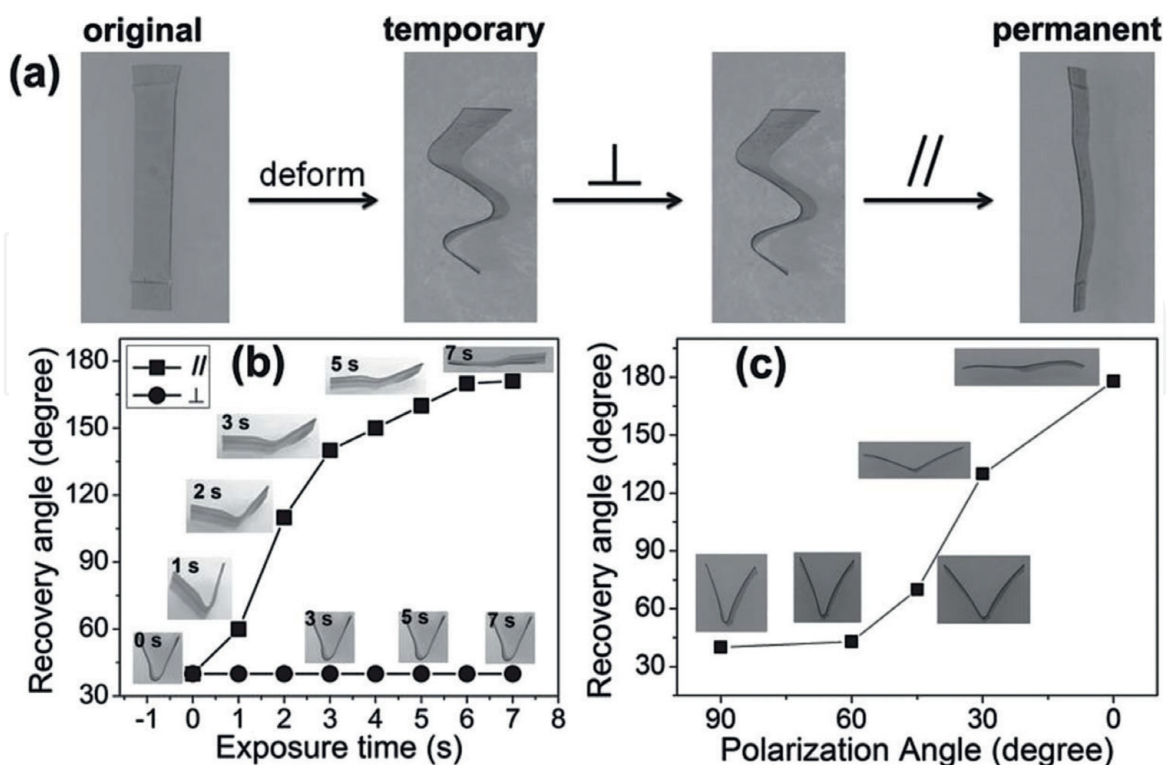

Figure 19.
Shape memory characteristics of PVA/0.02 wt.% AuNR with the light polarization-dependent at the room temperature; (a) optical images of the shape memory behavior; (b) recovery angle verse exposure time under different period of times in two directions, parallel and perpendicular; (c) relationship curve of recovery angle and polarization angle in 1 minute exposure using a light polarization [51].

4.2.3 Fiber reinforcement

Generally, the enhancement in the shape memory polymer mechanical properties is still limited and their reinforcement with short fibers and/or particles is irrationally to be proposed as structural substances [52]. Therefore, the reinforcement with continuous fiber has been attentionally employed to improve the mechanical properties of SMPs [53, 54]. Due to the highly potentialized SMPs, their usages are widely fulfilled for various advanced applications, such as solar arrays, trusses and antennas, which they essential with no moving segments [32]. Furthermore, the majority of researches with regards to SMPs composites are involved in thermoplastic SMPs resins such as polyurethane SMPs. Nevertheless, the comparatively inadequate thermal together with mechanical properties, for instance, moisture, temperature and/or chemical resistance of thermoplastic SMPs are not able to fulfill sensible demands [2]. Thermosetting SMPs, in spite of this, established a marked improvement in the latter characteristics which enable it to be extremely popular for many practical and or structural resources. The development of the fabricated polyurethane SMPs reinforced with carbon-based fiber was implemented for the industrial applications [55–57], a larger ratio of the bending recovery was exhibited in the reinforced polymers compared with pure SMPs sheets, from the same point of view, the epoxy-based SMCs namely EMC (Elastic Memory Composite) was potentially executed for the structure of spacecraft applications, as this EMC was developed in the early 1990s by Composite Technology Development (CTD) [58, 59]. Gall et al. [60] have been investigated the deformation micro-mechanisms of EMC and highlighted the development interaction reinforcement between the epoxy EMC laminate as shape memory polymers resins and fibers and found that because of the changing in the surface of the neutral-strain and micro-buckling effects, the reinforced SMP was able to produce a large value of compression strain compared with the traditional resin composite. Furthermore, the development of composite of thermosetting styrene-based reinforced with fiber was studied by Leng et al. [61], and found due to the good strain capabilities and their relative properties, these types of reinforced-polymers have been potentially chosen to take a part of the structure's applications. From the same perspectives, as a comparison with the pure SMPs, the carbon-based fibers present better thermomechanical properties, and thus been proposed to be use as multi-functional materials [32, 62]. The reinforcement of the glass and kevlar fibers SMPs composite have obtained a superior improvement in the stiffness associated with decrement in the recoverable strain, has been premeditated by Liang et al. [52]. On the other hands, the chopped glass fibers were added to the thermoplastic SMPs and their influences on the shape memory characteristics were examined by Ohki et al. [55]. It was found that the reinforcement by 50 wt.% glass-fiber entertains increment in the failure stress to 140% and reduced the recovery rate to 62%. Another study by Wang et al. [26] demonstrated the shape memory effect of TPI SMPCs under the effect of different mass fraction of the chopped carbon-fibers at three experimental temperatures of 299, 319, and 339 k, as shown in **Figure 20a**. it was found that both experimental variables; temperatures and fibers reinforcement have been remarkable affected the shape memory effect of TPI polymers. The shape recovery ratios tend to rapidly increased as the applied temperatures increased and reinforcement ratio of carbon fiber decreased (see **Figure 20b**), which can be justified these variations that as the cross-linked structure are getting dense, their movements are required more energy and the chain segment movements would be required a higher free space. Therefore, as the temperatures increased, the chain segments will have enough energy to release and thus perform the shape recovery.
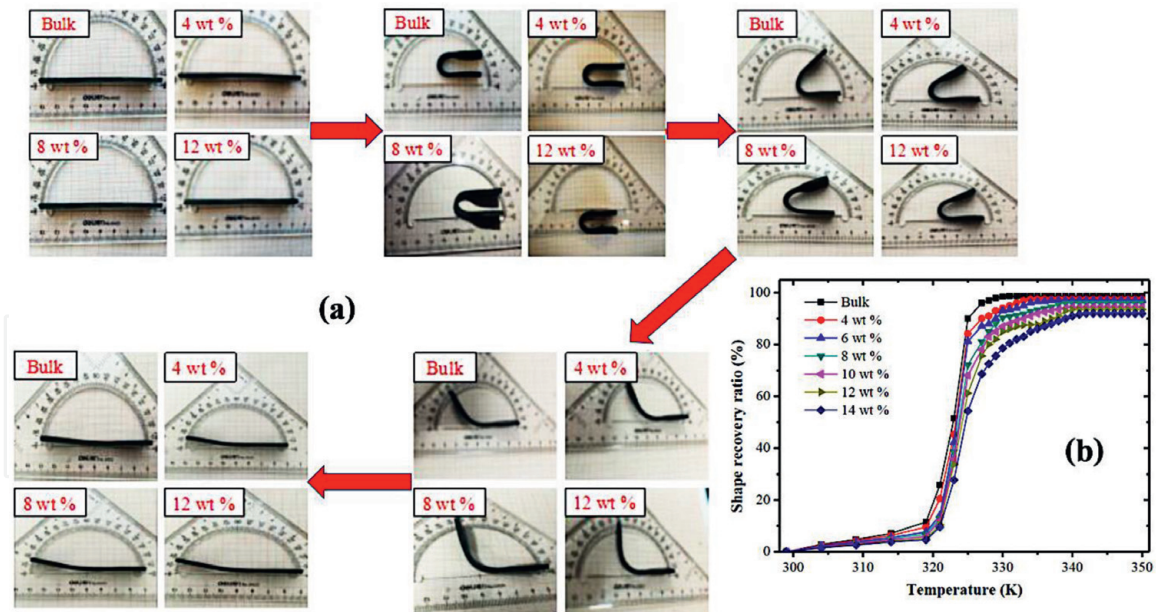
Figure 20.
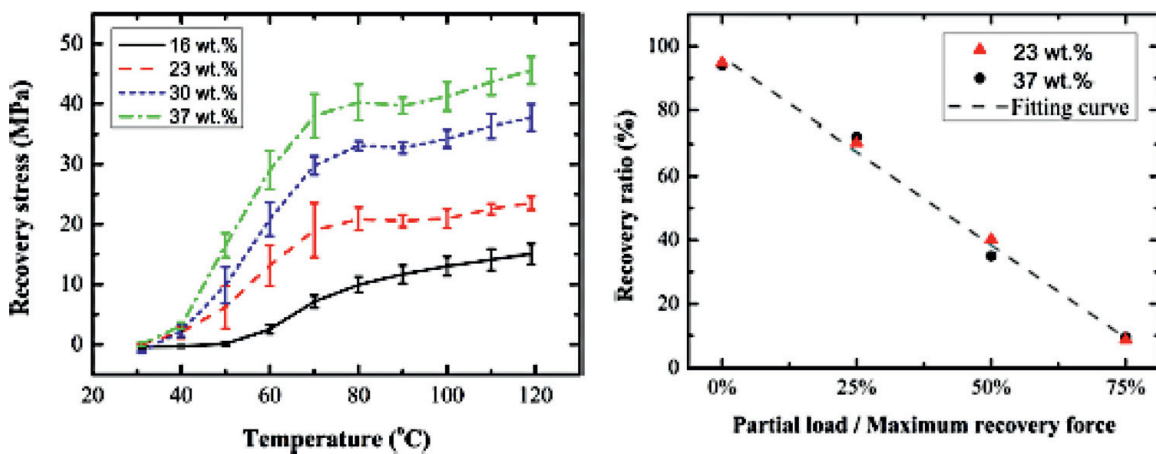*(a) Shape memory effect behavior with the reinforced SMP along with (b) the shape recovery ratio [26].*
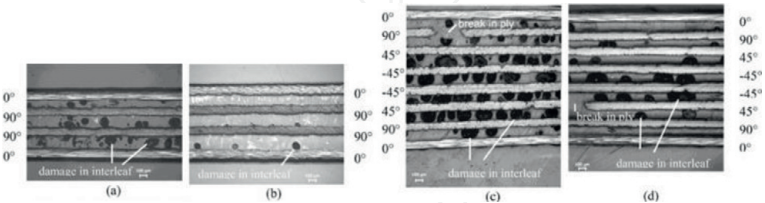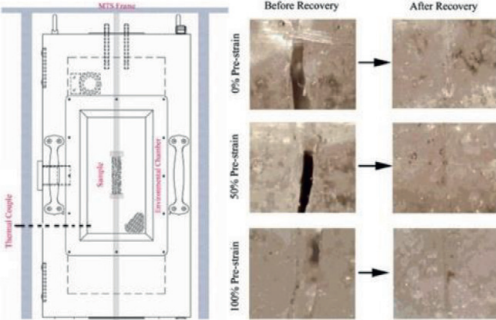

Figure 21.
(a) Stress recovery curves for four percentages of fiber reinforcement; (b) recovery ratio of the reinforced SMP composites with 23 and 37% mass fraction within different partial loads [63].
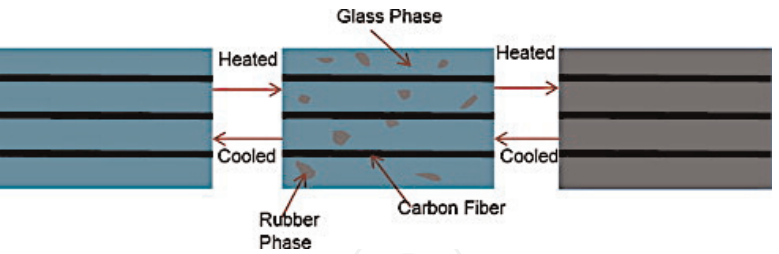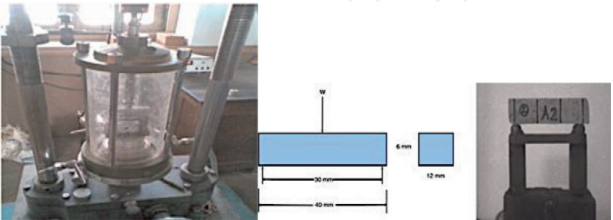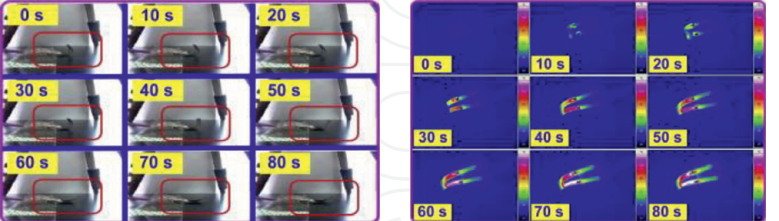
Figure 21a and **b** displays the epoxy-based SMPs reinforced of carbon fibers with four percentages ratio of 16, 23, 30, and 37% were designed by Li et al. [63]. The designed composites have demonstrated an excellent shape recovery at 120°C within a ratio of 90% and obtained a full recovery 100% after 20 min at the same temperature. The recovery ratio behaved proportionally with the mass fraction of fibers and inverse proportional with the partial load level. Extensive researches were carried out on different types of SMPs matrix and various fiber reinforcements because of the increasing demands for different applications as been summaries in **Table 2**.

5. Conclusions and remarkable observes

Shape memory polymers (SMPs) and their composite (SMPCs) have exhibited exceptional features that led to proposed them to be implemented as advanced materials for the current and potential applications. However, the traditional shape

Author	Substrate	Type of fiber	Recovery ratio (%)	Shape fixity ratio (%)	Comments	Shape memory recovery method/demonstration
Alvaro Iregui et al. [64]	Polymerized epoxy/ polycaprolactone blends	Blends	88–100	95–99	• A remarkable shape memory effect was obtained from the designed mat of the reinforced SMPs.	 (c-e) Shape recovery mechanisms for three times folding at 60°C for 30 min
Paul Robinson et al. [65]	Thermoplastic Interleaf Layers	Carbon Fiber Laminates	fully	—	• The recovery times were significantly longer for the quasi-isotropic laminates (8–9 min) than for the cross-ply laminates (2–3 min).	 (a-d) Micrographs of the cross-sections after the shape memory recovery process under different bending angles
Jianming Guo et al. [66]	Trans-1, 4-Polyisoprene (TPI)	Carbon Fiber	30–70	—	• The shape recovery ratio decreases as the carbon-fibers fractions increases. • The temperature effects have shown a significant enhancement, as the temperature increased, the shape recovery increased.	Bend and fold method
Guoqiang Li [67]	EPOXY EPON828	Polyurethane	24	30–66	• The stress–strain recovery decreases nonlinearly. • The applied force to close the initiated crack in the matrix led to narrow down the area of the cracks.	Three-point bending test

Author	Substrate	Type of fiber	Recovery ratio (%)	Shape fixity ratio (%)	Comments	Shape memory recovery method/demonstration
Guoqiang Li [68]	EPON™ Resin 828 (thermosetting polymer)	Polyurethane	—..	—..	• The relationship between the healing efficiency and pre-strain level is indirectly proportional. • The addition of 50% fibers caused an improvement in healing efficiency to double value compared with nonmodified SMPs. While, further pre-straining did not show any significant improvement due to the saturation behavior of the cracks in the healing efficiency.	 Schematic and optical images of the healing test and shape recovery
Herath et al. [69]	Epoxy Prepreg	Carbon Fiber	86	100	• An excellent shape memory characteristic attained in the modified shape memory polymer composite with carbon fibers without any existence for the effects of spring-back.	 Steps of the shape recovery for the modified SMPCs at different bending angles.

Author	Substrate	Type of fiber	Recovery ratio (%)	Shape fixity ratio (%)	Comments	Shape memory recovery method/demonstration
Qiao Tan et al. [70]	Epoxy Based	T700 Toray Carbon Fiber	—	—	• The glass ↔ rubbery transition concepts have presence in harmony behavior as the temperatures and amount of fiber changed.	 Glass ↔ rubbery transition state for the SMPCs/CF.
Shankar A.Hallad [71]	Epoxy	Nano- and Micro-Graphene and Carbon Fibers	—	—	• The SMP beams have revealed an enhancement in the mechanical properties as the reinforcement with GO and CF existed comparing with the nonmodified SMP beams.	 Three-Point Load Set-up for Shape Recovery Test
Haibao Lu [72]	Epoxy-Based Thermoset SMP Resin	Carbon-Fibers	95%.	—..	• The combination of GO and CF additions have been significantly the reliabilities during the bonding procedure associated with assisted the heat transfer resistivity between the matrix of polymer and fibers. • A synergistic effect of rGO and CF on the induced shape recovery during the Joule heating.	 Shape recovery of nanocomposite SMPs induced by Joule heating after been incorporated with 0.03 g of GO and 0.14 g of carbon fiber. Shape recovery behavior in SMP nanocomposite induced by Joule heating and infrared video camera was utilized for recording.

Author	Substrate	Type of fiber	Recovery ratio (%)	Shape fixity ratio (%)	Comments	Shape memory recovery method/demonstration
Kun Wei [73]	Thermoset Hydro-Epoxy SMP	Glass Fiber, 437H, Taishan Fiberglass	Full recovery	—	• No any noticeable variations in the glass temperature of SMP after the addition of short glass fiber. • Within an amount less than 4.5 wt.% short glass fiber, the hydro-epoxy resin SMP acquired an excellent shape recovery with a 100% recovery after several minutes of different temperature exposures.	 Shape memory properties: A visual perform of SMP after 4.5 wt.% addition of short glass fiber

Table 2.
Summary of various types of fibers and their shape memory effect.

memory characteristics and features are quite limited to due to their abilities of recovering the original shape using the heating source only. Therefore, the reinforcements with micro/nano-fillers and particles or fibers are essentially important to be considered to meet the needed functions and performances. Furthermore, the reinforced SMPs and SMPCs are not only demonstrated a significant mechanical and shape memory properties, but also obtained noble features after being exposure to any electro-or-thermal heating source. Based on the previous researchers, the foreseeable future concepts of SMPs and SMPCs may well-rely on how to adopt the benefits of this kind of properties, in addition to, exceptional attributes as advanced alternatives. On the positive front, the practical applications probability of SMPs and SMPCs are found extensively when displayed in the remarkably distinct application principles which may have seemed in the recent peer-reviewed publications and also patents. Moreover, we believe that the investigation of the advanced features of these types of materials is still in development of new design and/or incorporations. The actual vital, hence, can be found in the finding of substantial beneficial functions wherefore SMPs and SMPCs are enablers or even no less than tremendously excellent substitutes.

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Conflict of interest

The authors declared without any conflict of interest.

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