Shape memory polymers for composites

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ABSTRACT

Shape memory polymers (SMPs) are a class of active, deformable materials that can switch between a temporary shape, which can be freely designed, and their original shape. With their large deformation, low density, various stimulation methods, good biocompatibility and other advantages, SMPs have become widely accepted as smart materials. However, SMPs have many limitations and weaknesses that are exposed in engineering applications. For this reason, the significance of SMP composites (SMPCs) has been analyzed in terms of four aspects: reinforcement, innovation and improvement of driving methods, the creation of specific deformations and the creation of multifunctional materials. We then introduce the constitutive theory of SMPs and the post-buckling analysis of SMPCs. Afterward, we introduce the extensive applications of SMPCs in the fields of aerospace, biomedical equipment, self-finishing, deformable mandrels and the 4D printing of active origami structures, demonstrating their ability to undergo active driving and deformation, their adaptiveness, their ease of transport and their rapid production capacity, which fully demonstrate the unique advantages of SMPs in solving application problems. Finally, the advantages and disadvantages of SMPCs in applications are summarized, and the prospects for new SMPCs and new SMPC structures are described.

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1. Introduction

The shape memory effect (SME) is a special mechanical phenomenon usually described by the shape memory cycle (SMC). Fig. 1.1 shows a highly common shape memory cycle. SMC
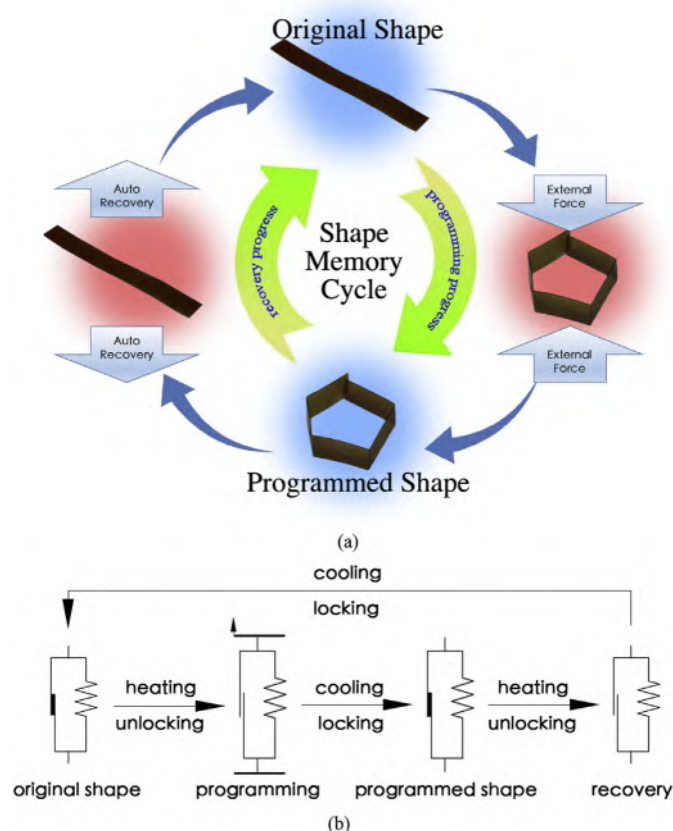

(a)

(b)

Fig. 1.1. Once flowers have opened for the day, can the material revert to its original form again? This return can be achieved by shape memory effects. (a) This shape memory cycle consists of two hot stages (red background) and two cold stages (blue background), and the shape changes occur during the hot phase. First, the original shape (straight) is heated and can be adjusted by an external force into a special shape (pentagonal). This shape can be fixed after cooling. This stage is called the programming process, and the temporary shape is called the programming shape. When heated again, the material will return to its original shape and output a certain recovery force. This stage is called the recovery process. (b) Shape memory effect is easy to understand by an analogy. A spring and a telescopic rod with a lock are connected in parallel. When locked, the spring can be stretched without external force. Conversely, when unlocking, the spring restores the original length. We are about to find that the spring (entropy-based elastic network) and the shape lock have corresponding features in the polymer. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

represents the mechanical process that embodies the shape memory effect of that kind of material, and a more exact definition will be explained later. The shape memory effect is common in polymers, but most examples are inferior. One class of polymers can provide excellent shape memory effects and they are predominantly used in shape memory applications. They are called shape memory polymers (SMPs). Shape memory polymers are active deformable materials that undergo large deformations, first mentioned by Vernon et al. in a dental patent in 1941 [1]. In the sixties, heat-shrinkable tubes entered the market [2,3]. Their applications have drawn substantial research attraction. As the seventies began, a number of commercial companies developed their own shape memory polymers [4]. Since the end of the last century, researchers have begun to systematically study shape memory polymers. The principle of shape memory has been increasingly elucidated, and diverse shape memory effects have been observed [5].

Taking the initiative to change shape is vitally necessary for animals and even other organisms. Compared to inorganic ceramics and metal materials, polymer materials show natural advantages such as lower density, better biological and organic compatibility, and easier modification and processing. Accordingly, shape memory polymers are blossoming in radiant splendor in the field of active polymers. Actively moving materials can be effectively deformed in shape by external stimulation [6—9]. Examples such as shape memory polymers, electroactive polymers [10,11], photo-induced polymers [12], and hydrogels [13,14] have been the subject of substantial research. Actively moving materials are often categorized by response behavior, and the differences in properties between different actively moving materials are significant.

Shape memory polymers are materials driven by external stimuli that actively switch between multiple shapes. Compared with other materials, shape memory polymers have the advantages of high stress tolerance [15], the ability to undergo large deformations [2], a rich selection of driving methods (including heat 3456, light [7,8], electricity [9—11], magnetism [12,13] wetting [14], and pH [16]), excellent radiation resistance and good biocompatibility [17], which make them a research hotspot in the field of actively moving materials.

At present, shape memory polymers have many applications in aerospace [18], medicine [19—21], self-finishing smart textiles [22,23] and electronic devices [24], and self-assembling structure [25—27]. Specific applications, including low-impact release mechanisms in the aerospace field, large spatial deployable structures, shape memory polymer sutures, minimally invasive surgical instruments with good biocompatibility, the active deformation or self-finishing of textiles, electronic devices, and variable mandrels, which effectively solve the thorny problems of the corresponding fields, show the great power of shape memory polymers. The above applications involving composites will be described in detail in Section 5.

2. Basis of shape memory effect in polymers

In this section, the basic characteristics of shape memory polymers are reviewed, and the topic of shape memory polymer composites is not addressed. The majority of this section is thus a classic topic, which can be found in a standard shape memory polymer review. Therefore, this section is brief. More detailed description can refer to the supplementary documents.

This section does not discuss an important property of the shape memory polymer, the stimulation method. Because taking into account shape memory polymers that used in engineering are commonly based on phase change. Unless the composite material is made, the driving method is scarce. So please refer to the relevant review of shape memory polymers [28], and Section 3.2.

Shape memory polymers can be divided into physical bonding and molecular/supramolecular switching through the mechanism of switching, but also through the shape memory effect classification: Shape memory polymers can be divided into one-way and two-way from directionality of the shape memory effect (So far, two-way SME is reported only in shape memory alloys [29—31]), or divided into dual SME and multi SME from the number of stable shapes. There are many delicate distinctions involved. Since our focus is on dual SMP based physical bonding which is convenient and mature for engineering applications, we will not cover them in detail in the text, please refer to the supplementary document.

2.1. Process of shape memory effect in polymers

Therefore, the shape memory effect involves comprehensive effects of material and process. We then describe the principle of phase transition shape memory polymers, which allows us to quite accurately describe the shape memory cycle of polymers with a standard linear solid (SLS) model [32—34], as shown in Fig. 2.1 (a). The points on the corresponding stress-strain-temperature curves for each state are shown in Fig. 2.1 (b). We describe a hot

programming, hot recovery, no-load dual-SME. We identify the most obvious mechanical characteristics of the shape memory polymers. At high temperature, the polymer shows viscoelastic behavior, while it shows general elastic behavior at low temperature, which shows that viscosity has a great dependence on temperature, i.e., the viscosity changes between the maximum and a smaller measurable value during the phase transition process. This is the essence of the shape memory effect. First, upon heating, the viscosity of the dashpot is reduced to be barely measurable, and creep occurs under an external load to obtain a programmed shape. When the shape is maintained by the external force and the temperature is decreased below the transition temperature range, the viscosity of the dashpot increases greatly. Then, when the external force is removed, apart from a small general elastic response, the programmed shape stays the same, and the shape creation process is now complete. When heated again above the transition temperature range, the viscosity of the dashpot decreases, and the system creeps back to its original properties. This description summarizes the main mechanical processes of the shape memory effect, but it is difficult to provide an accurate description of the details of the process, which makes improving the linear viscoelastic model challenging. A new description (the phase transition method) has been proposed, and a more precise constitutive theory of shape memory polymers will be presented in Section 4.1.

The recovery process of the shape memory polymer can exert a force to the outside, so the shape memory polymer has a driving ability. But in this process, the energy is stored through mechanical deformation in programing process, while the external stimulus is only used to open the switch. So we do not use the word "drive method", which is prone to ambiguity, when talk about the stimulus later.

The above shape memory cycles (i.e., hot programming, hot recovery, dual-SME without load) are used to evaluate whether the shape memory effect is good or bad, which is usually achieved by describing some representative parameters of the shape memory
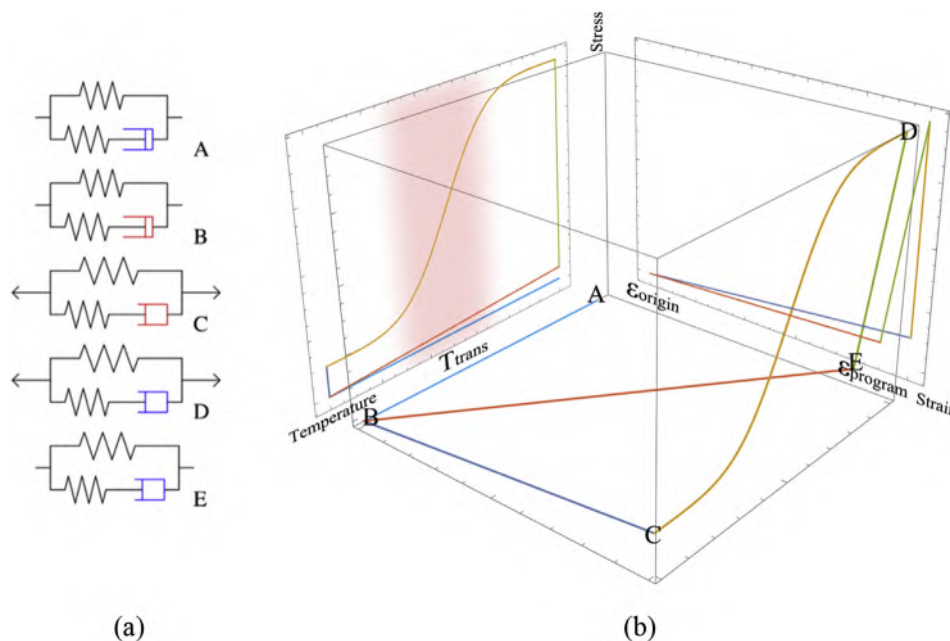

(a)   (b)

Fig. 2.1. A typical hot programming, hot recovery, unloaded dual-shape memory cycle and the SLS model corresponding to the state of each stage. In this figure, the blue dashpot indicates that the viscosity of the dashpot is at its maximum (at room temperature) and that the material has ordinary elasticity, while the red dashpot indicates that the viscosity of the dashpot is barely measurable (at high temperature) and that the material is highly elastic. Symbol legend: A, the original shape; B, the heated original shape; C, the original shape distorted under external load into a temporary shape; D, application of an external load to maintain the temporary shape as the material cools; E, removal of the external load and the occurrence of a small ordinary elastic recovery, not substantially altering the temporary shape. A-B-C-D-E, programming process; E-B-A, recovery process. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
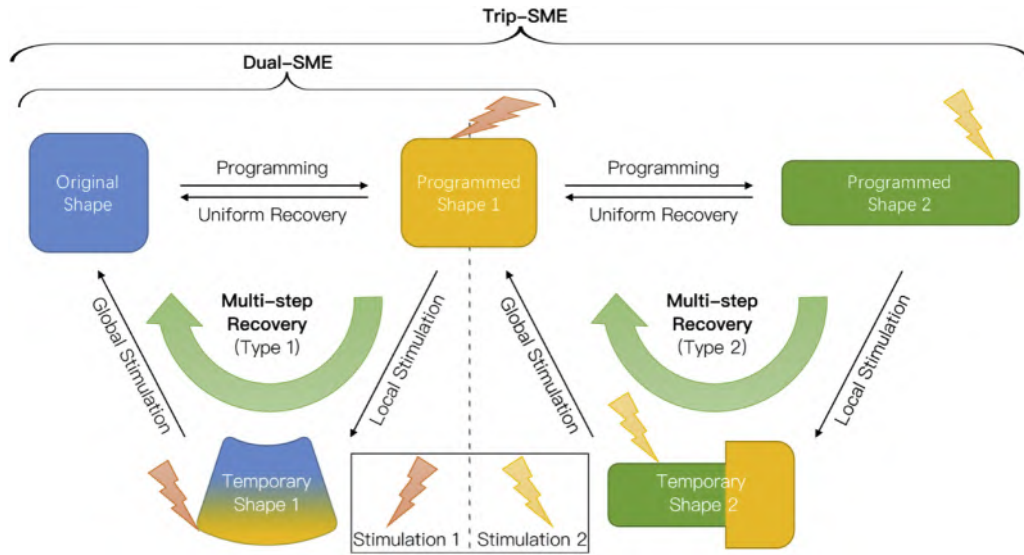
Fig. 2.2. Some cases of global deformation of shape memory polymers. In this figure, different colors represent different tensile states. The order of recovery can be designed to take advantage of different transition temperatures (or other stimuli) and the material can be kept in a certain state of local recovery, thus enriching the shapes of the shape memory polymer that can be fixed. For the sheet, the creation of complex shape memory behavior has two basic ideas, namely, local recovery in the direction of thickness and local recovery in the direction of length or breadth. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

cycle, namely, the fixation rate and recovery rate. Through the shape memory cycle diagram, they can be expressed as follows (considering only the one-dimensional case):

$$R_f = \frac{\varepsilon_{tem,t} - \varepsilon_{ori}}{\varepsilon_{tem,0} - \varepsilon_{ori}} \times 100\%$$

$$R_r = \frac{\varepsilon_{tem,t} - \varepsilon_{rec}}{\varepsilon_{tem,t} - \varepsilon_{ori}} \times 100\%$$

where $\varepsilon_{tem,0}$ is the strain at the last moment before the external force is removed, $\varepsilon_{tem,t}$ is the strain at the moment of measurement, ε_{ori} is the strain before the programming, and ε_{rec} is the strain after the recovery. This is also true for each SMC of multi-SME.

It should be noted that the reduction in the fixation rate consists of two parts, the general elastic shape recovery due to the removal of external loads at the end of the program and the cold recovery after long-term storage. Cold recovery is related to the material and its storage conditions, such as temperature, external force, and chemical environment. The higher the temperature and the greater the external forces that tend toward the original shape, the more rapidly cold recovery will occur [28,35,36]. The chemical environment may also trigger a recovery, such as by wetting [36]. The recovery rate is dependent on the temperature. Additional, the fixation rate and recovery rate reflect the performance of the material in a specific shape memory cycle. Obviously, they are process-dependent values, which are related to strain, temperature, time and even strain rate [37].

2.2. Principle of shape memory effect in polymers

For further explanation from the perspective of molecular mechanics, the shape memory effect in a shape memory polymer-based glass transition is mainly due to the two-phase structure of the polymer: the stationary phase to maintain the original shape of the macrostructure and the reversible phase of softening and hardening. When the temperature is lower than the Tg range, the shape of the reversible phase can be frozen; otherwise the entropic elasticity drives the polymer network to recover to the initial state.

The stationary phase ensures that the network deformation is affine without viscous flow due to external forces. In general, physical crosslinking and chemical crosslinking correspond to thermoplastic shape memory polymers and thermosetting shape memory polymers, respectively. When the Tg range of the stationary phase is high, and softening and relaxation do not occur in the temperature range of use of the material, so the memory of the original shape can be ensured. When the Tg range of the reversible phase is low, softening and hardening can easily occur as the temperature changes, and the segment of material has a high deformation capacity at higher temperatures. When the temperature is low, the polymer network is in a low-energy state, the segment cannot rotate freely, and the polymer is generally elastic at the macroscopic scale. When the temperature gradually increases to the Tg range, the segment rotation is unlocked, and then the polymer is highly elastic on the macroscopic scale, with a high deformation capacity. When the temperature is lowered again to below the Tg range, the segment does not return to the low-entropy state as the segment rotation locks, so the material shows a temporary shape on the macroscopic scale. The origin of the shape memory effect described above has been supported by recent experiments [38–42].

2.3. Shape memory behavior under global view

Finally, it is emphasized that the above discussion focuses on the local mechanical properties of shape memory polymers. The global shape of the shape memory polymer is the result of the superposition of each local deformation. Therefore, because the temperature and stress distribution can be designed in time and space, the shape memory behavior of the global shape is quite complex and rich, as shown in Fig. 2.2. In the case of an average dual shape memory polymer, when the material is uniformly heated, the shape of all the different parts of the material is restored together, that is, a one-step recovery process; when non-uniform heating occurs, the recovery of the different parts of the material is carried out separately, i.e., "pseudo multi-SME" [43–45]. (When a deformation of the material is observed whose macroscopic behavior resembles a kind of shape memory effect but does not accord with all the

stability requirements of the shape memory effect, for example, if its shape is dependent on the amount of stimulation, we use the prefix "pseudo".) The shape-memory behavior will be further complicated when external constraints occur; for example, "pseudo two-way SME" may appear [46,47]. These complex shape memory behaviors will create a series of smart shape memory polymer structures, which will be described in Section 3.3.

3. Why use SMPCs instead of SMPs?

In general, the significance of composite materials is that several components complement each other, result in synergies, or improve or enrich the function of the matrix material. For shape memory polymers, composites are made with two basic objectives, i.e., reinforcement and finding new and effective stimulation methods. (For shape memory polymer composites, the stimulation can be more precise and highly selective, so more sophisticated shape memory behaviors can be achieved, such as the example in Section 3.3.). In addition, multifunctional and smart shape memory polymer materials are promising research directions, with abilities including self-healing [48–50], drag reduction [51], and optical properties [52–55].

This section describes shape memory polymer composites, which can be used as a matrix, filler, or layer of composites. When a shape memory polymer is used as a matrix, the doping can consist of one or more of zero dimensions (particles), one dimension (fibers and vessels) and two dimensions (layers and films) of carbon materials, polymers, metal and ceramic particles, liquid or even gas, to achieve reinforcement, novel stimulation methods, self-healing and other functions. A shape memory polymer can also be used as a functional filler in the form of fibers or the like to form a multifunctional composite material. A well-designed composite structure can achieve two-way recovery and other interesting and useful functions.

The study of shape memory polymer composites is driven by application, according to the need to design the type and proportion of filler reasonably. Too much filler will often reduce the thermal and mechanical properties and lead to a lower transition temperature. On the other hand, only a small number of fillers can enhance the shape memory effect; most fillers will reduce the shape memory effect of a polymer, as reflected in the fixation rate and recovery rate (see Fig. 3.1).

memory polymers have greater deformation capacity, lower density, lower production costs, richer properties and modification methods, but because of limitations of their stiffness, strength and driving forces, they are difficult to use as structural materials. Given the need for structures with greater deformability and lower weight, shape memory polymer composites are beginning to be studied.

We identify the differences between shape memory polymers and shape memory alloys. The shape memory effect of an SMA depends on a martensitic transformation, i.e., martensite at low temperature and austenite at high temperature [58,59], which leads to a higher Young's modulus at high temperature [57]. The shape memory effect of a shape memory polymer is the diametric opposite, which allows the shape memory polymer to be programmed much more easily than the shape memory alloy. In addition, the deformation of the lattice of a shape memory alloy gives it clear superelasticity, as its restoring force is less than the Young's modulus [57], while the shape memory polymer has high elasticity above the transition temperature, and its restoring force and modulus are similar. In terms of their shape memory effect, a shape memory alloy has a unique two-way shape memory effect [60–64], while a shape memory polymer can have a multi-shape memory effect [65,66]. In terms of the stimulation method, shape memory alloys can be stimulated by heat or magnetism [67–70], while shape memory polymers have very diverse stimulation methods, including heat, electricity, magnetism, light, ultrasound, solvents, metal ions, pH and others. Moreover, polymers can more easily be used to produce composites to achieve better properties and richer functionality, and this article will detail them.

A shape memory polymer is reinforced mainly to increase the driving force of the recovery process and to provide better carrying capacity, which correspond to a high elastic modulus and a general elastic modulus, respectively. In general, the reinforcement filler improves these two indicators at the same time. Reinforced shape memory polymer composites mainly include particle [71–74], nanofiber [75,76], short fiber [77,78] and long fiber [79,80] reinforcement. Carbon materials have been widely used in shape memory polymer composites because of their good reinforcement, rich physical and chemical properties and good combination with polymer-based materials. Examples include carbon nanotubes [71,74], graphene [72,73], and carbon fiber [75–78]. Carbon materials can also form self-assembled structures, such as carbon nanotube arrays [74] and carbon nanopaper [81,82]. In addition to carbon materials, glass
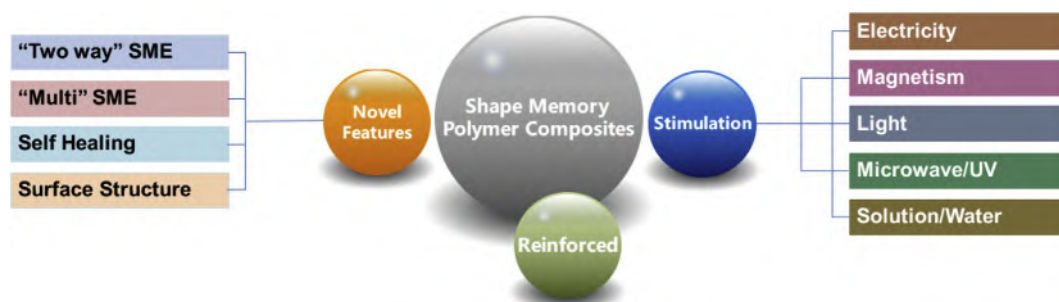

Fig. 3.1. Why make shape memory polymer composites? Reinforcing and providing more stimuli are two basic purposes. Recently, there has been a new purpose in making composites.

3.1. Reinforced SMPCs

Shape memory polymers are relatively recent compared to shape memory alloys (SMA). Shape memory polymers were first proposed and noted in the biomedical field for their polymer-based properties [56]. In the field of engineering, shape memory alloys were extensively studied in US Navy laboratories in the 1960s [57]. Shape

fiber [77,83], silica [84,85], Kevlar fiber [77] and spandex fiber also provide excellent reinforcement, and doping with metals such as Ni has also been shown to provide reinforcement [11]. Generally, the filler has a better reinforcing effect when it exhibits an ordered structure. Usually, in shape memory polymer composites, the reinforcing ability of long-fiber filler is greatest, followed by that of short fibers and particles [86–88] (see Table 3.1).

Table 3.1
Effect of doping in shape memory polymers.

	pure shape memory epoxy polymers	carbon black, CNT	long carbon fiber
Transition temperature	~105 °C	slight reduction (slight rise in polystyrene)	constant
Modulus at normal temperature	2.8 GPa	slight rise, <3.5 GPa	significant rise, >8 GPa
Modulus at high temperature (recovery force)	~10 MPa	slight rise, ~10 GPa	significant rise, >3 GPa
Limit strain	>50%	slight reduction	significant reduction (mainly through buckling)
Fixed rate and response rate	~100%	~100%	reduction, difficult to exceed 95%

The tensile strain rate in the longitudinal direction of the long fiber is generally less than 2%, which restricts the tensile deformation of the shape memory polymer. However, when the material is bent, part of the system shows compressive deformation, and the fiber may undergo great geometric nonlinear deformation, i.e., a post-buckling phenomenon, to achieve greater longitudinal compression deformation [89–93]. Compared to the traditional reinforcement method, the theoretical basis of long fiber-reinforced soft matter is different [94–96]. Based on buckling and post-buckling, long-fiber shape memory polymer composites, which are commonly used to integrate deformation and structure in one material, have been applied in spatial deployable structures and actuators, such as spatial deployable hinges [82], trusses [97] and antennas [98].

In general, too much filler or filler with too strong a reinforcement effect can also reduce the shape memory effect. In general, too much or too strong reinforcing materials tend to reduce the shape memory ability while enhancing the mechanical properties of the polymer. After adding the reinforcing filler, the reinforcing material in the temporary shape is in the elastic deformation state. This is a cold recovery process that reduces the fixation rate. On the other hand, the reinforcing filler can be prone to plastic yield in a large deformation, forming a permanent plastic deformation and thereby reducing the recovery rate. In addition, the influence of doping on the glass transition temperature is uncertain. For example, doping with spherical nanoparticles such as Ni powder will reduce the glass transition temperature of epoxy-based shape memory polymer [11], while CNF/MWCNT doping in styryl shape memory polymer has the opposite effect [99]. Chen et al. incorporated C60, carbon nanotubes and graphene into shape memory polymers to test their shape memory properties. The results showed that C60 and CNT significantly increased the recovery rate but had little effect on the shape recovery stress, while the graphene reduced the material recovery rate but improved the recovery stress [100]. It is necessary to refer to the application requirements to balance the appropriate form and proportion of filler. Ni et al. demonstrated that 10–20 wt% is the optimum fiber weight fraction with very low residual strain during cyclic loading.

However, some fillers can perform dual functions, both providing reinforcement and increasing the shape memory effect. Such fillers generally rely on supramolecular systems. For example, cellulose whiskers and graphene oxide are both reinforcing materials and both interact with the polymer network through hydrogen bonds in a reversible supramolecular effect, fixing the programmed shape [101,102]. In addition, the addition of self-organized nanoparticles, such as carbon black, to the polymer [103] may also increase the fixation rate and recovery rate of the material.

3.2. Novel stimulation based SMPC

In addition to improving thermal and mechanical properties, shape memory polymer composites are often responsible for providing new stimulation methods for the recovery process and programming process. In the wide range of applications of shape memory polymers, there is a demand for non-thermal triggers, such as in aerospace structures and actuators and biomedical devices. More accurate and non-contact stimulation methods are important steps in the application of shape memory polymers, including electrical, magnetic, and optical responses. One solution is to fabricate molecular switch shape memory polymers, but it is difficult to design such materials from scratch, their properties are unpredictable, and their uses are constrained by production costs and processes. The second solution is indirect stimulation using a filler, which is effective and easy to achieve by doping with conductive materials, magnetically conductive materials, or absorptive materials. In addition, shape memory polymers can be stimulated by solvent, ultrasonic, etc.

We introduce a typical solution to novel stimulation. The development process and specific examples can be found in the supplementary document.

3.2.1. Electric stimulation

Electric stimulation is the most convenient and precise of all kinds of stimulation. The use of current-driven shape has produced a great increase in the applications of shape memory polymers. Because of their restricted electronic structure and low conductivity, intrinsically conductive polymers have never been reported. Conversely, polymers mixed with conductors have many advantages, such as high efficiency, stability and ease of design. The heat is fed into the polymer through the current, enabling the endogenous initiate recovery process. Carbon materials such as graphene, nanotube and nanofiber as well as nanoscale granules of metal can be used for the conductive doping of polymers.

There are two issues that need to be addressed when designing an electrical stimulation of shape memory polymer composites [9,28]. First, the doped conductor is formed into a conductive three-dimensional network structure. For example, nickle powder is formed to chain under magnetic field [10,11], or using carbon black to connect short carbon fibers can increase the conductance by 100 times [104]. Second, to make the doped conductor more uniformly and firmly incorporated into the polymer matrix, surfactant [105] and chemical modification are generally used. A review of specific examples can be found in the supplementary document.

3.2.2. Magnetic and light stimulation

Magnetic driving and light driving are two of the most typical and practical methods of non-contact stimulation and are extremely suitable for applications such as biological and medical instruments where stimulation by direct contact is difficult. These two means of driving have two further types of approach, direct driving and indirect driving (mainly heating). Light-reactive shape memory polymers generally involve reversible chemical bonds,

such as the cycloaddition reaction of cinnamic acid [7,106,107]. However, the polymer itself does not have a magnetic heating effect, and as a result, magnetic heat driving can only be achieved indirectly. Other methods of magnetic driving have never been reported. Magnetic drives are similar to electric drives and also address the problem of doping and matrix connection.

Magnetic driving is a typical approach to stimulation without direct contact. The principle is the indirect heating of shape memory polymers by doping with magnetic materials, such as ferrite and soft magnetic materials. This heating method is highly suitable for medical instruments such as implanted shape memory polymer supporters. In 2006, Lendlein et al. added magnetic nanoscale particles to poly(ε-caprolactone) with shape memory effects and thereby made a new type of shape memory polymer stimulated by an alternating magnetic field12. Further, this method can be improved in a heating mode which mixes magnetic induction heating and direct heating [108]. Since the magnetic heating is controllable, the required for ambient heating is tunable. In this way, the material exhibits its apparent switching temperature being varied. They also added magnetic materials to shape memory polymer systems with temperature memory effects and obtained shape memory polymer nanoscale composites with magnetic memory effects [109]. Leng and Smoukov et al. reported the magnetic driving of a composite thin membrane made of Nafion and Fe3O4, with the controllable realization of as many as four programmed shapes13. This magnetic driving was highly controllable. Even when the local temperature exceeded 80 °C, the surface temperature remained near the body temperature (38–40 °C), which offered great potential in medical applications.

To address the problem that doped materials such as the magnetic medium ferrite oxide had obvious boundaries with the composite and were unevenly distributed, modifications of the ferrite oxide surface and crosslinking it into the composite network provided an effective solution. Lendlein et al. covered the surface of magnetic particles with oligo(u-pentadecalactone) (OPDL), covalently incorporating nanoscale metal granules into the composite, thereby obtaining even doping distributions, and achieved precise control of a two-segment shape effect and a reversible shape memory effect through magnetic fields [110]. There are other ways to combine. For more examples, see the supplementary document.

Light driving is splendid for enabling remote driving and control. Reactions can occur under different wavelengths of light depending on the doping, and light from all wave bands has the potential to serve as stimulation. Table 3.2 lists some examples. For a brief introduction to the following examples, please see the supplemental documentation.

Infrared light and microwaves have good penetration ability, which makes them suitable for the driving of shape memory polymer biomedical instruments. Electromagnetic waves in this wave band mainly produce heat effects, and therefore the essential prerequisite for using infrared light and microwaves is to establish a wave-absorbing material. These materials are also frequently researched as wave-absorbing materials for radar. The wave-absorbing agents of wave-absorbing materials mainly act through resistance, an electric medium or a magnetic medium. Resistance forms include simulated circuit dampers, carbon materials, metal powders, silicon carbide, and other conductive materials. Electric medium forms include barium titanate, conductive polymer materials and other materials with high dielectric constants. Magnetic medium forms include ferrite, carbonyl iron, soft alloy magnets and other materials with self-resonance and hysteresis loss. Other reviews present more detailed discussions of microwave-absorbing materials [100,101]. Shape recovery by wave-absorbing agents such as nanoscale carbon materials [102,103], silicon carbide [123], nanoscale ferrite materials [104], water [105] and rare earth organic complexes 106 have been reported. Magnetic medium materials such as ferrite have the best wave-absorbing ability, but the composition boundary remains obvious because they are metal, and they decrease the thermomechanical ability of the shape memory polymer to undergo large deformations. Taking the reverse consideration, carbon materials, such as carbon nanotubes, carbon black, and carbon fibers have a wide absorption range. Additionally, their combination with polymer bases enhances the dynamic properties of shape memory polymers to some extent, which gives them abundant applications as wave-absorbing fillers.

3.2.3. Wetting stimulation

Additionally, solvents can serve as a method of stimulating the shape recovery process. The doped solutions reported to drive shape memory polymers include a shape memory polymer with a glass state transformation and a hydrogen bond-locked shape memory polymer. For a shape memory polymer with a glass state transformation, the essence of solution driving is plasticization. As moisture increases, the polymer chain parts are softened gradually, and the temperature of the glass state transformation decreases because of the swelling effect and the polarization effect. For most plastics such as epoxy resin, phenylethylene and other engineering plastics, the speed at which solvents permeate them is very limited. In 2014, Leng et al. reported an epoxy resin shape memory polymer doped with SDS that could undergo moisture driving [124].

On the other hand, the shape memory effect can be created from scratch using composites. Qi et al. made a nanoscale polymer of

Table 3.2
Doping absorption and the corresponding wave band and wavelength.

Doping	Absorption band	Absorption wavelength	reference
Free radical reaction network	UV	~302 nm	[111]
Gold nanoparticles/nanorods	Visible	~532 nm	[112–116]
Carbon material	Radar	Various (Broad absorption)	[117–119]
Ferrite	Radar	Various (Narrow absorption)	[120]
Water	IR	~1.9 μm	[121]
Yb(TTA)3Phen and Nd(TTA)3Phen	IR	980 and 808 nm	[122]
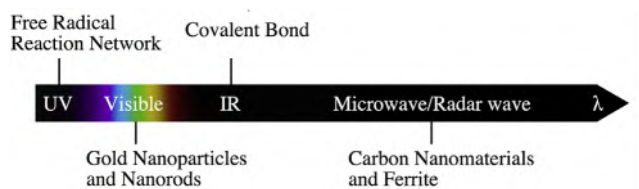
Poval and oxidized graphite, which adopted temporary shapes upon heating and underwent controlled shape recovery in moisture [87]. The switch phase of this shape memory polymer was based on hydrogen bonds, and both heating and solvation could destroy hydrogen bonds, unfreezing the macromolecular chain parts fixed at the surface of the oxidized graphene and allowing shape recovery. Zhu et al. added nanoscale cellulose whiskers to elastic thermoplastic polyurethane, and the material system showed shape memory effects with similar principles to the previous example [88]. Zhang et al. designed a novel shape memory polymer composite that adheres to a layer of cellulose nanocrystal-supported carbon black (CB@CNC) in the PU micrometer skeleton [125], which is equivalent to introducing a shape lock. This can be achieved by heating, moisture and various other means of stimulation. In the three instances above, Poval and polyurethane themselves are entropic elastic networks, but they did not show shape memory effects. After the addition of nanoscale structures that could form hydrogen bonds, the system was frozen, and it had shape memory effects at that time, which illustrates the origin of shape memory effects to some extent.

3.3. Creating SMPCs with novel shape memory behaviors

As described at the beginning of this paper, the shape memory effect is a process-dependent phenomenon that depends on various thermodynamic processes, especially the spatiotemporal distribution of temperature and external force. In the absence of load, this diverse deformation process can be attributed to the zonal recovery of the material. The recovery of its global shape goes through several specific temporary shapes, and sometimes these temporary shapes can even remain stable. Phenomenologically, this example seems to be summarized as a multi-SME, but it must be noted that according to the definition of a multi-SME, there must be some temporary shape in the recovery process that remains stable at a certain temperature. In this well-designed recovery process, the shape is sensitive to the temporal and spatial distribution of heating, so we call it pseudo multi-SME. The most common method is to achieve non-uniform heating during the recovery progress, which will be described for two typical representatives. First, for pre-stretched (uniaxial or biaxial) shape memory polymer sheets, heating one surface will cause the material to appear warped, resulting in a warping of the sheet. Second, for pre-programmed (including stretching and bending) shape memory polymer strips, different portions of the strip are heated to achieve controlled and segmented shape recovery.

For the creation of a specific temperature spatiotemporal distribution, the simplest approach is partitioned heating [126–128]. However, partitioned heating is difficult to control, so it is not easy to use. Therefore, it is important to introduce an accurate heating mechanism, and the previously described shape memory polymer composite, which is indirectly heated, again demonstrates its utility. When these dopants exhibit a particular distribution, such as distribution at a surface or only in a certain area, they are heated in a fixed way. Dickey et al. stained a biaxial pre-stretched transparent shape memory polymer (Shrinky-Dinks) with dye at a specific location. When the polymer was heated by light, the black portion was heated first, the painted portion unilaterally contracted, and the sheet showed curvature45. This kind of bending is designable, so this method can create the required three-dimensional origami structure. When heated further, the material will be fully recovered, showing a smaller planar shape. This group then analyzed the effects of the power, intensity, width and shape of the laser used on the folding [129]. Further, the group used different colors to dye different parts and then used different colors of light for stimulation, thus achieving a selective response that facilitates the creation

of more complex shapes [130] (Fig. 3.2(a)). Xie et al. reported a segmented shape memory polymer composite strip constructed sequentially by ferrite doping and undoped and carbon nanotube doping. Ferrite is heated by low-frequency electromagnetic waves, while carbon nanotubes are heated by high-frequency electromagnetic waves. The undoped portion is finally recovered by heating, resulting in four different shapes [43] (Fig. 3.2(b)). In addition, through a similar construction process, segmentation by light and heat control is also reported [131] (Fig. 3.2(c)). Guo et al. deposited carbon nanotubes on the shape memory polymer film and described the local recovery and control response rates, depending on the location and number of layers deposited [132].

On the other hand, shape memory behavior will be further complicated if we consider external constraints. We review some valuable applications that have been reported. It is worth noting that unlike the no-load case, an ingeniously designed external constraint can achieve stable temporary shapes. Light-induced shrinkage is an important means of introducing these constraints, that will enrich the number of temporary shapes for opaque polymers with photocrosslinking or photo-isomerous [133,134]. Constraints can of course also be introduced by additional construction features, such as a laminate. Felton et al. reported a series of shape memory polymer origami structures [26,135,136]. Both sides of the pre-stretched shape memory polymer are constrained by a film that is only bendable, and a notch is formed in the portion to be folded. This topic will be explained in detail in Section 5.3.

In addition, a specially designed laminate can be used to fabricate a two-way deformation actuator whose two-way deformation is facilitated by the great changes in the modulus of the shape memory polymer during the process of phase transition. Note that the two-way deformation is not programmable. We introduce several representative designs. In 2010, Hu et al. first proposed the idea of two-way bending laminates and laminated polystyrene with shape memory polyurethane, which was equivalent to adding a temperature-dependent constraint to the surface of a uniaxially pre-stretched shape memory polymer sheet [46,137] (Fig. 3.2(d)). Imai designed a new two-layer structure, including one or two layers of shape memory polymer and composite aluminum to achieve electrical driving [138]. Zhang et al. also achieved two-way bending with an electroactive polymer and a shape memory polymer. The curvature was also calculated and verified by finite element simulation [139] (Fig. 3.2(e)). Leng et al. used shape memory polymer attached to an electroactive polymer and achieved a degree of freedom for programmable two-way deformation by the double control of heating and voltage47. However, these methods can only achieve two-way bending, limited by the design of the special drive. Achieving designs with more complex two-way drive structures remains our future direction.

Recognizing that shape memory behavior is process dependent, we can design richer deformation processes for shape memory polymers. Considering the design of the heating conditions and external constraints can greatly broaden the imagination space of smart shape memory polymer structures. Because of the high potential for the design of smart structures and of temperature and confinement distribution in space and time, although shape memory behavior design is rich, even endless, its design and application to date have only exposed the tip of the iceberg and await further development.

3.4. Multifunctional material systems based on SMPCs

Reinforcement and the search for new effective stimulation methods are two basic goals of shape memory polymer composites. In addition, the filler can also confer new functions on the shape memory polymer, resulting in novel multifunctional composites.
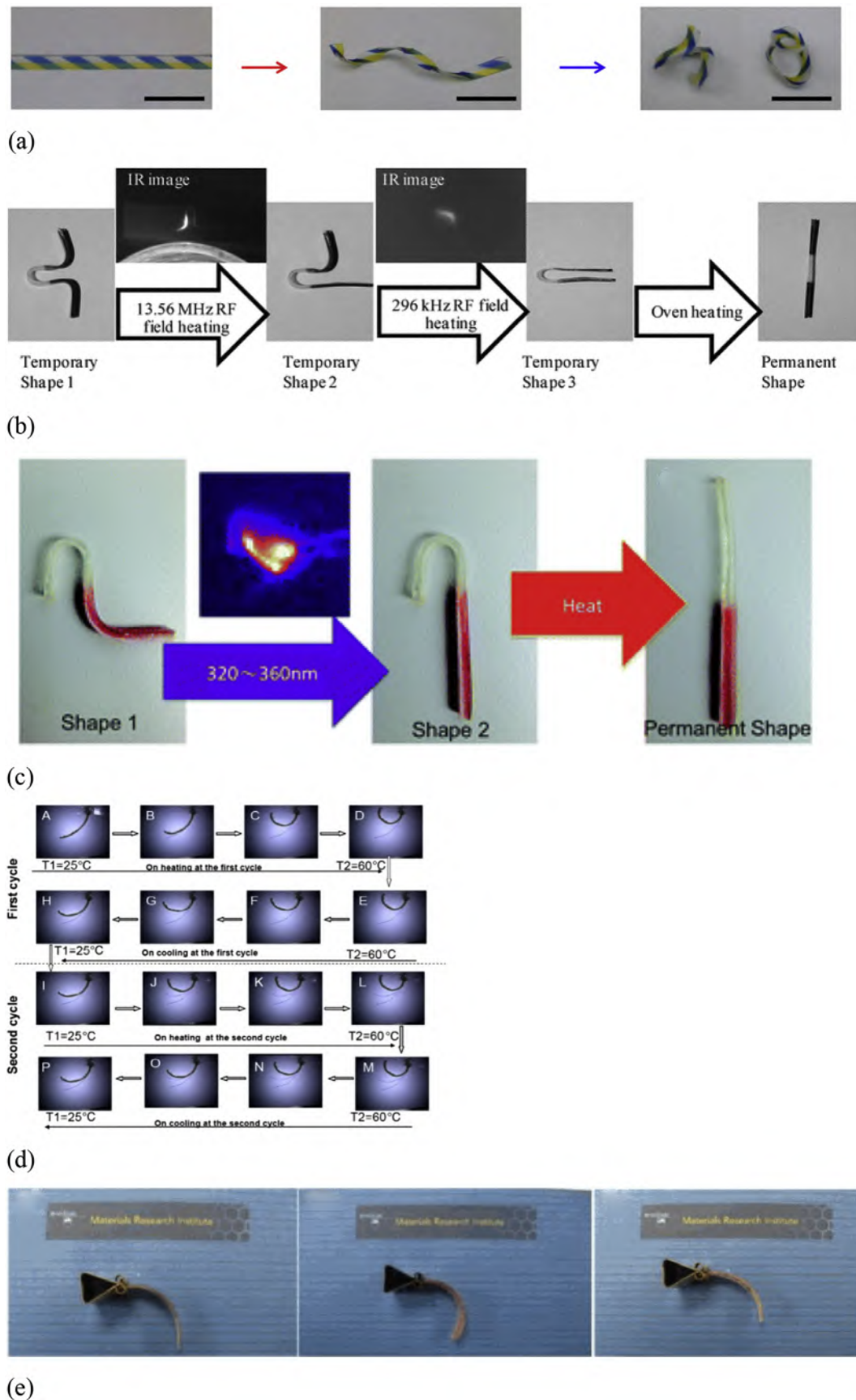
(a)

(b)

(c)

(d)

(e)

Fig. 3.2. List of several ways to create novel shape memory behaviors using dual-SME. (a–c) Pseudo multi-SME. (d,e) Pseudo two-way SME. (a) Different forms dyed with light. (b,c) Different doping. (d) Laminate polystyrene with SMP. (e) laminate SMP with EAP [43,46,114,115,123].

Multifunctional materials and structures are highly compact due to the integration of different materials or devices and have the advantages of saving weight and space, simplifying structural design and broadening the application scope of the materials [140].

Inspired by biology, researchers began to study smart materials and structures, that is, multifunctional material systems that integrate perception, processing and feedback. When external stimuli such as electricity are applied as input, shape memory polymer composites can process them into heat, as described above, and output the feedback form as shape recovery. Therefore, electric-stimulated shape memory polymer composites can be considered smart materials.

In the following review, we consider the shape memory effect as a single function and focus on shape memory polymer composites with additional functions. As a composite is still a polymer material, the material or phase transition process will have many properties and functions, such as insolubility, insulation, refractive index, and thermal expansion and change. These properties are inherent, but in general, inherent properties do not attract our interest. However, in addition to these phenomena, the properties of artificially designed phase transitions are also significant, such as changes in color [141], permeability [142] and transparency [143].

As with phase transition properties, most of the properties of composites are ordinary. However, the design of multifunctional composite systems often exploits such properties. Typical examples include self-healing materials, soft electronics, material health monitoring, energy collection and management, and radar absorption. Because of the excellent properties of the shape memory polymer itself and its working environment, self-healing properties are the most noticeable, the most compelling and the most extensively studied, and therefore, self-healing shape memory polymers and composites are reviewed first.

3.4.1. Self-healing SMPCs

Self-healing is a classic biological concept. Every animal and plant has excellent self-healing ability to resist various possible injuries from the living environment. Similarly, materials are faced with the threats of fatigue damage and accidental damage, such as impact, bending, stretching, electric shock or chemical corrosion. Damage to traditional materials is irreversible, which increases cost and also introduces many hidden problems. Therefore, self-healing materials are of continuous research interest, and materials such as concrete [144,145], polymers [146,147] and ceramics [148,149] are extensively studied. Polymer systems (especially soft polymers) have inherent self-healing advantages, i.e., there are long segments that can be moved and modified, so for a self-healing polymer, two concepts are essential: first, the polymers achieve re-crosslinking through their own physical and chemical properties [150], and second, damage repair can be achieved by doping with a healing agent [151]. Polymer nanocomposites are a common approach and either provide healing agents through microcapsules or microtubules or introduce a framework to provide an external force that can bring the fracture surface back into contact.

A large number of reviews have summarized the principles and examples of self-healing [152,153] so this article only describes the principles and several examples related to shape memory polymers.

Shape memory polymers have a close relationship with self-healing polymers, which is manifested in the shared requirements of the shape memory effect and self-healing ability. The intersection of the two behavioral principles is shown in Fig. 3.3. A shape memory polymer undergoes a large deformation in its work process and often plays the role of a structural material in applications, which makes damage to the shape memory polymer more probable. On the other hand, the shape memory polymer has the
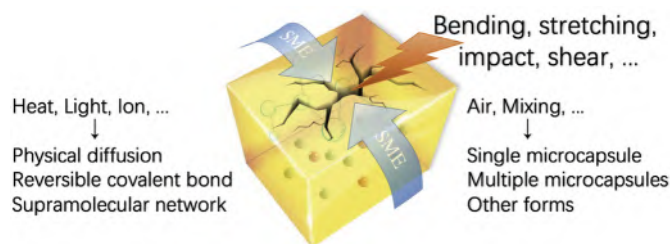

Fig. 3.3. Self-healing of shape memory polymers: sources of damage, self-healing stimulation and principles and assistance by shape memory polymer properties.

inherent advantage of self-healing without an external force, i.e., the shape can be recovered after injury, and the fractured surface will recombine, allowing the material to achieve adhesion of the wound without external force after a limited degree of damage. This ability solves the problem that the traditional self-healing process requires external force bonding and thus offers high potential for engineering applications. In summary, the shape memory and self-healing effects are promoted by similar properties, and the development of self-healing polymers with a shape memory effect is highly attractive.

For a given polymer, the healing of cracks involves two phases. First, the surfaces of the cracks must remain in contact with each other [154] and form adhesions between the surfaces by physical or chemical processes, which can be provided by the material or by added healing chemicals. Due to their intrinsic well-behaved shape memory effects, non-destructive damage to shape memory polymers (where the effect on undamaged parts can generally be regarded as a process of cold programming) can often be recovered by the intrinsic shape recovery properties in the first phase. We now introduce the second phase.

Self-healing can be achieved by the polymer itself, which has already been addressed by many summaries [130–132] and will be discussed only briefly here. Generally, self-healing is based on a reversible physical effect, that is, expansion occurring near a crack, or on reversible bonds, including reversible covalent bonds [155,156], ionic bonds [157], metal coordinate bonds [158,159] and supramolecular interactions [160,161].

Self-healing materials operating by this kind of principle often require triggering conditions, such as heating, to promote the quick expansion of the contact surface or to trigger chemical reactions. It is noteworthy that the reversible bonds available for self-healing effects are similar to the ones in shape memory polymers based on reversible bonds, which enables many kinds of shape memory polymers based on reversible bonds to exhibit self-healing ability. Below, Table 3.4 summarizes whether various kinds of photochemical principles can be applied to self-healing or shape memory effects. As illustrated previously, the principles of shape memory polymer composites stimulated by new methods are similar to those of this kind of light-stimulated self-healing material system [97,162].

Table 3.4
Summary of different reversible interactions as well as their applicability to self-healing and shape memory/change.

Reversible Interaction	Self-healing	Shape memory/change
Covalent crosslinking	Yes	Yes
Isomerization	No	Yes
Metathesis	Yes	No
Metal coordination crosslinking	Yes	Yes
Hydrogen bonding crosslinking	Yes	Yes
Salt crosslinking	Yes	Yes
Heat[a]	Yes	Yes

[a] Heat includes diffusion and phase transition.

Self-healing materials that rely on physical expansion are generally soft thermoplastic materials or poorly crosslinked rubber materials whose chain parts exhibit good expansion ability. In real applications, many thermosetting shape memory polymers with high crosslinking density form a complex solid network using chemical crosslinking. The interaction between the surfaces after they are broken apart is relatively weak, and the speed of expansion of the chain parts is low, which hinders self-healing. Therefore, the use of adhesive agents that are not part of the material itself is a promising idea. Through the reasonable design of the expansion of the adhesive agent, artificial healing could be transformed into self-healing of the material system.

Next, we discuss the effect of shape memory polymers on self-healing properties, i.e., the spontaneous completion of the splicing process. This can be achieved by a shape memory polymer matrix or by shape memory polymer fiber doping. A large number of molecular switch shape memory polymers have been previously reported to be self-healing, such as Diels-Alder reaction-based shape memory polymers [163] and thermoplastic shape memory polymers. For general thermosetting shape memory polymers, self-repairing abilities can be obtained by doping with a thermoplastic polymer such as PCL. Luo et al. reported the addition of a PCL-based shape memory polymer fiber with electrospinning to epoxy resin to obtain a self-healing shape memory polymer composite [48].

Depending on the actual application conditions, shape memory-assisted self-healing (SMASH) and close-then-heal (CTH) [50] are two kinds automatic splicing in self-healing polymers. The difference between these two methods is mainly whether the shape memory part is programmed beforehand. SMASH is simpler to create than CTH, but CTH has a wider range of applicability. The shape memory polymer is effective (close to 100%) when no ambient load is applied, and therefore, the SMASH method can be used. On the other hand, when there is an external load, the shape memory polymer has difficulty in completing recovery, so the CTH method is very effective [130]. The basic process of CTH is shown in Fig. 3.4. When a shape memory polymer matrix is used, the shape of the polymers will recover to the original shape when there is no external constraint. Therefore, the preprogrammed shape memory polymer base requires an external constraint upon recovery. A fabric] or grid can provide external compression constraints to achieve splicing.

In addition, CTH can be extended to other matrices. Doping other active materials can also be the driving force for fracture joints, including shape memory alloys [164,165], artificial muscles [166], and shape memory polymers [167–171]. Shape memory alloy doping was the first to be reported [148,149]. Shape memory polymers have better compatibility with polymer-based composites than SMAs but have less driving force and require more stretching. There are several reports on self-healing stimulated by doping with shape memory polymer short fibers [151–155]. Li et al. have predicted the mechanics of CTH by adding pre-programmed shape memory polymer short fibers [172,173].

3.4.2. SMPC surfaces

The development of shape memory polymer surfaces is an attractive direction. Surface microstructure has been a topic of great concern in recent years. The concept is biomimetic, that is, the nature and phenomena of biological or natural nanomaterials, devices and processes are derived from complex surface interactions and physical and chemical properties [174,175]. These microstructures greatly expand the specific surface of the materials, and the structure even reaches photon size, thus achieving special mechanical and optical properties that are geometrically related, i.e., geometric deformation will produce a property change. Shape memory polymers have the basic property of variable geometry, based on which a large number of surface microstructure materials with variable properties will be generated. The variable properties caused by geometric deformation can be divided into two categories, affine deformation and non-affine deformation. Please note that the criteria are local. The former is mainly based on tension or compression, which leads to tuning properties [176], while the latter is based on the creation or erasure, of surface microstructure, which leads to properties that either exist or disappear [52,160,177]. Superficial hydrophobicity [178], fluid drag reduction [51], transparency [179] and one- or two-dimensional photonic structures [161,162] can be achieved by creating surface microstructures of shape memory polymers such as waves, bumps, tree protrusions, and column arrays.
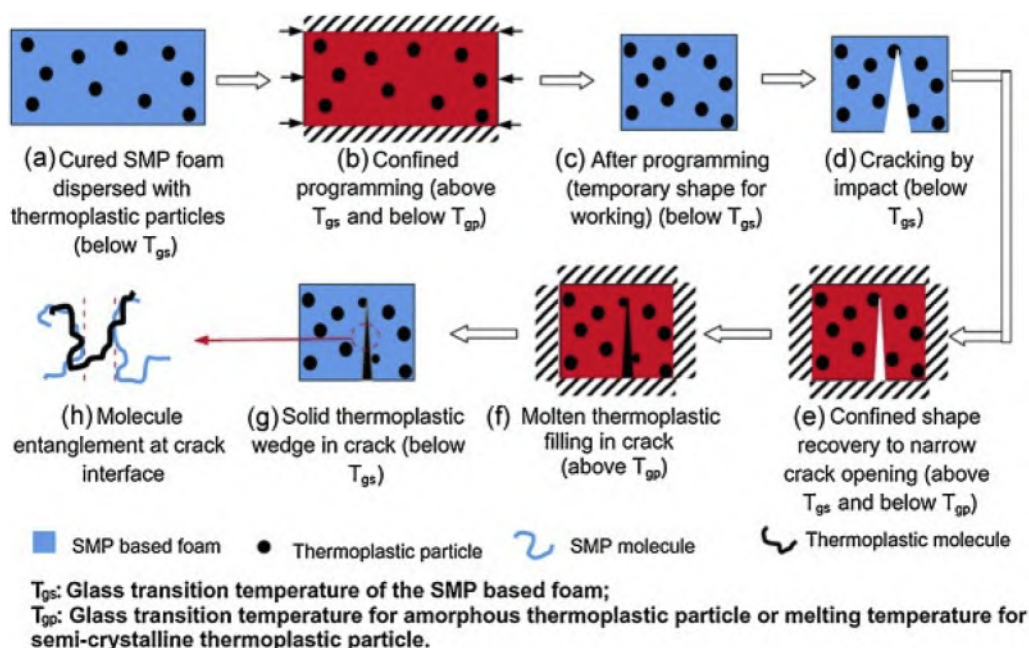

T_{gs}: Glass transition temperature of the SMP based foam;
T_{gp}: Glass transition temperature for amorphous thermoplastic particle or melting temperature for semi-crystalline thermoplastic particle.

Fig. 3.4. Schematic of self-healing progress of shape memory polymer-based smart foam, showing an example of close-then-heal (CTH) [50].
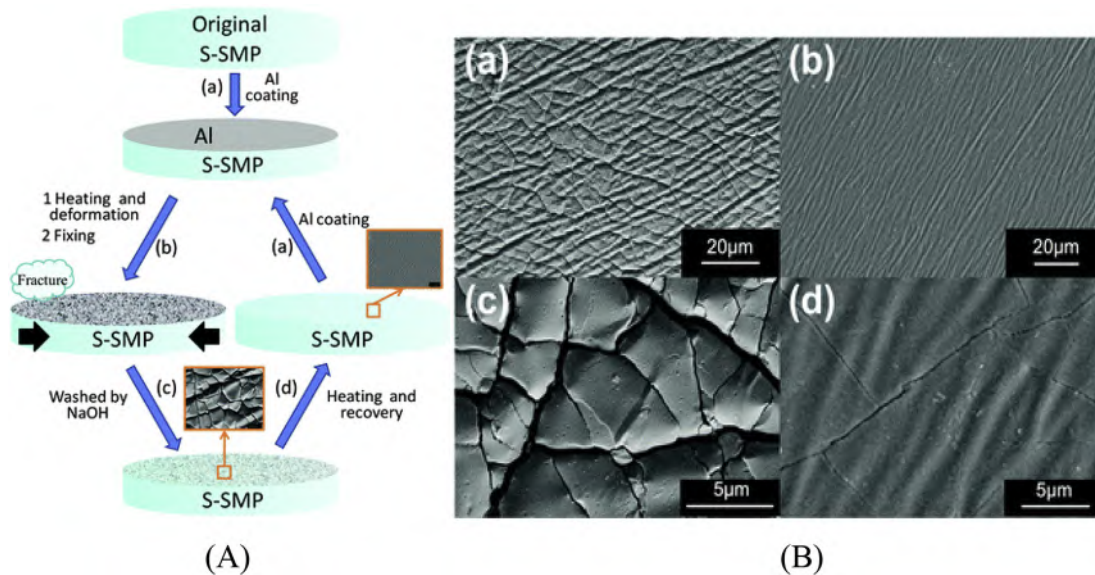
Fig. 3.5. Controlled wettability based on reversible micro-cracking on a shape memory polymer surface. (A) Schematic of the process of hydrophobicity change. (B) Microstructure photos. (a,c) As the film cracks, the polymer substrate is exposed, so the surface shows a hydrophilic nature. (b,d) Metal cracks in contact with the surface of the material showing the hydrophobicity of the metal [164].

When another material is attached to the surface of the polymer, such as a metal film, main surface microstructure is made of the film, the shape memory polymer is mainly provided geometric deformation to achieve the change of the film. Some novel properties can be achieved by composites, which is difficult for pure polymers. Leng et al. aluminized a shape memory polymer surface with artificial cracks to achieve tunable hydrophobicity [180]. The process is shown in Fig. 3.5(A). In the original shape, a hydrophobic aluminum film completely covers the polymer surface. When the polymer substrate is stretched, the aluminum film is separated by the presence of cracks and is insufficient to cover the whole surface. Therefore, the hydrophilic polymer matrix is exposed, and the contact angle is significantly increased. Fig. 3.5(B) shows some evidence of this phenomenon using microstructures.

Polymer surface microstructures have three basic methods of creation: top-down creation, bottom-up creation and the combination of the two. In the past, the top-down approach has dominated, that is, microstructures have been obtained by macroscopic treatment of the polymer surface, including lithography, imprinting, and the like. With the aid of the shape memory effect, bottom-up creation is possible, mainly by buckling of the film or lines on the polymer substrate.

Buckling is a long-studied structural stability problem. In past practice, buckling has been avoided because it severely degrades the load carrying capacity of the structure. In recent years, this view has been changing. This strongly avoided phenomenon has unique energy and geometric characteristics, and with the rise of smart structures in sensors, energy collection, damping, drivers and high-deformation structures, the subject has gained wide attention. The use of shape memory polymers can effectively produce buckling of a film or line to facilitate the creation of surface microstructures, such as one-dimensional sinusoidal corrugated surfaces [54] or more complex two-dimensional structures [55] (Fig. 3.6). This is an important case of buckling mechanics, which will be followed by a detailed introduction to its theory and creation method.

4. The mechanics of SMPs and SMPCs

As mentioned earlier, the shape memory effect is a polymer mechanical behavior. With the principle of shape memory effect explained, increasing numbers of people are aware of the universality of this effect. Once it is recognized that the shape memory
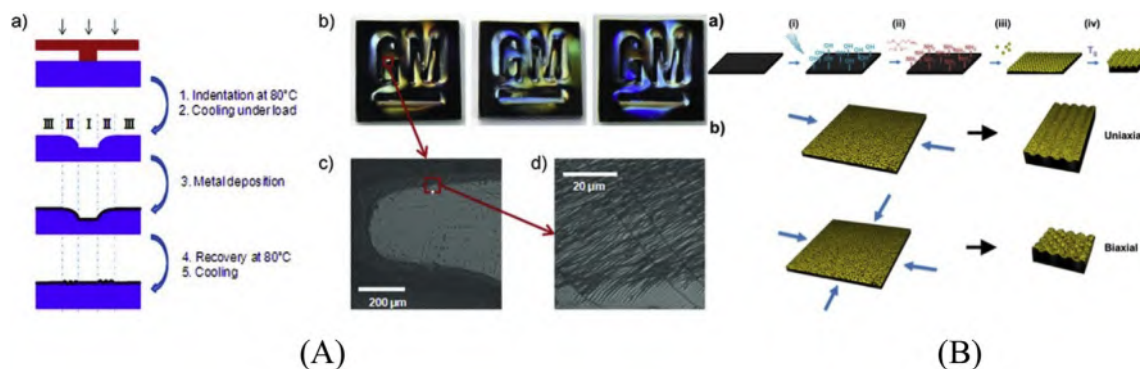

Fig. 3.6. Surface structure of shape memory polymer created by buckling. (A) one-dimensional sinusoidal corrugated surfaces. (a) Schematic of production progress, (b) photo and (c) microstructure photo. (B) One-/two-dimensional sinusoidal corrugated surfaces. (a) Schematic of production progress. (b) Uniaxial and biaxial recovery create different shapes [54,55].

effect is to a considerable extent a process property, the description of the mechanical behavior becomes very urgent. There are significant differences in the shape memory behavior of polymers with respect to different programming and recovery processes, which are manifested in the arbitrariness of the temporary shape, the length of the recovery time and the different forms of damage to the material.

For the calculations required in the application field, it is necessary to supplement the constitutive relation of the shape memory polymer. In addition, for specific structures, special simplification is required for use. In this section, we first discuss the constitutive model of pure shape memory polymers. The mechanical performance of shape memory polymers in a typical shape memory cycle is presented in the Introduction, so we will not repeat the description here. A suitable constitutive theory of shape memory polymers should successfully describe the shape memory cycle but, of course, is not limited to this. The modeling of shape memory polymers includes thermodynamic testing and mathematical description. Although the testing process is important, in the application of the model, the conclusion is even more important. Therefore, this paper introduces several constitutive models for shape memory polymers, including the viscoelastic model and the phase transition model.
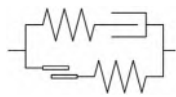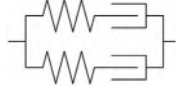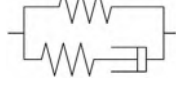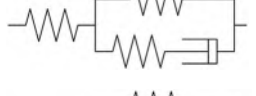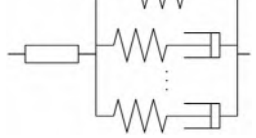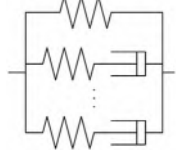
Compared with the research on constitutive models of shape memory polymer, the modeling of shape memory polymer composites began much later, with little research on their mechanics. Except for a few special structures, such as beams [181–185] or sheets [186–188], shape memory polymer composites are poorly or even incompletely modeled, especially for dynamic predictions, which are difficult. For the further application of extension, it is urgent to establish models for accurately describing geometrical and mechanical behavior and for the dynamic analysis of shape memory polymer composites. It is hoped that this field can receive corresponding attention.

4.1. Constitutive theory for shape memory polymers

The modeling began much later than the discovery of shape memory effects. In the 1990s, Tobushi et al. first modeled the shape memory behavior of polyurethane [189] using a four-element model that adds a residual deformed element into a standard linear solid model, describing the one-dimensional mechanical behavior of the shape memory polymer with incomplete recovery. Further, a non-linear correction can be made to the model [32]. Chen et al. proposed the use of dual Maxwell elements in parallel to describe this incomplete response [190]. Now, the viscoelastic model has been extended to 3D [191–194]. In 2006, Diani et al. first proposed a three-dimensional constitutive model [178], which suggests that the elasticity is mainly generated by entropy or cohesion and gives a mechanical explanation of the stress-strain-temperature variation, but the details of the process are not sufficiently accurate. In 2008, Nguyen and Qi et al. proposed a new theory that describes the behavior of shape memory more accurately, taking into account the effects of incorporating structure and stress relaxation from the perspective of the molecular mechanism [195]. In 2011, Chen and Nguyen developed the previous model and proposed a more complex description of the molecular mechanism [196]. In 2013, this group used multiple discrete relaxation processes to describe stress relaxation to study the effect of deformation temperature and physical aging on the shape memory behavior of amorphous networks [197]. In another work, multiple relaxation times were considered [177,198], that is, multiple Maxwell elements were included to account for the time and

Table 4.1
Several multi-component models of shape memory polymers.

Reference	Model diagram
Tobushi 1997 [173]	
Chen 1999 [174]	
Diani 2006 [178]	
Nguyen 2008 [179]	
Westbrook 2011 [177]	
Yu 2012 [182]	

temperature dependency of the shape memory effect. Some typical component models are listed in Table 4.1. The finite element modeling of shape memory polymer structures has also been proposed [199].

Another modeling approach, which was first used in shape memory alloys, is based on the phase transition process [200]. Compared with metal, which has a clear phase boundary, the physical and chemical properties of polymers change gradually, so the polymer phase transition is a relatively fuzzy concept but does indeed occur in the shape memory cycle as a phenomenological description, which allows us to use this modeling approach. According to the discussion of Section 2.3, we know that the process of shape memory polymers across the glass transition temperature range can be described by the proportional function of the frozen phase ($\Phi(T)$). As shown in Fig. 4.1.

In 2006, Liu et al. first introduced the phase transition method for the shape memory behavior of polymers [201]. It is maintained that the shape memory polymers are a two-phase mixture near the transition temperature. The strain is composed of mechanical strain and thermal strain, and the mechanical strain is composed of storage strain and elastic strain. Using the composite method, the shape memory polymer is described at different temperatures, but the model is only suitable for one-dimensional small-strain processes. In 2008, Chen et al. considered large strain and extended the model to 3D [202,203]. In 2009, Wang et al. introduced parameters related to heating rate and hysteresis into the improved model to describe the complex shape memory behavior [204].

On the other hand, three-phase models have also been developed to avoid the introduction of stored strain and to provide a
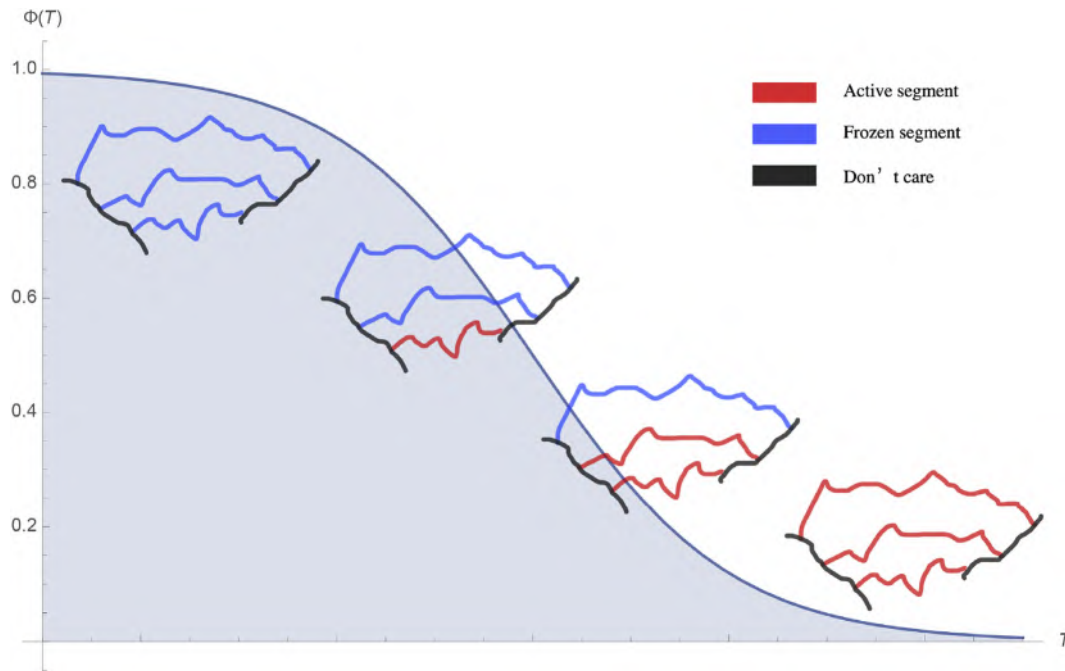
Fig. 4.1. Proportional function of the frozen phase ($\Phi(T)$). By this way, the process of shape memory polymers across the glass transition temperature range can be described. The longer segments are softer, and generally easier to unlock from the frozen phase. The black parts are also segments, their status is not investigated, but can be considered similarly.

Table 4.2
Frozen phase volume fraction functions proposed in the existing literature. 191.

Reference	Frozen phase volume fraction function
Liu et al. (2006) [185]	$\Phi(T) = 1 - \frac{1}{1 + c_f(T_h - T)^n}$
Qi et al. (2008) [189]	$\Phi(T) = \frac{1}{1 + \exp\left(-\frac{T - T_r}{a}\right)}$
Wang et al. (2009) [188]	$\Phi(T) = \alpha \exp\left(-\left(\frac{T_s}{T}\right)^m \beta^{-n}\right)$
Reese et al. (2010) [210]	$\Phi(T) = \frac{1}{1 + \exp\frac{2w}{T - T_r}}$
Volk et al. (2011) [211]	$\Phi(T) = \frac{b - \tan\frac{T - A}{B}}{b - a}$
Gilormini and Diani (2012) [212]	$\Phi(T) = \left(1 - \left(\frac{T - T_{min}}{T_{max} - T_{min}}\right)^m\right)^n$
Guo et al. (2015) [213]	$\Phi(T) = \int_{T_s}^{T} \frac{1}{S\sqrt{2\pi}} \exp\left(-\frac{T - T_g}{2S^2}\right) dT$
Li et al. (2016) [191]	$\Phi(T, \dot{T}) = 1 - \left(1 - \left(1 - \exp\left(-\frac{\Delta H_a(T)}{k_B T}\right)\right)^{\frac{dt}{\tau_0}}\right) \cdot \int_{r_c(T)}^{\infty} p(r) dr$

more accurate description of shape memory behavior. In 2008, Qi et al. proposed a three-phase model of the three-dimensional finite strain, which divides the glass phase in the traditional two-phase model into the original glass phase and the frozen glass phase [205]. This model provides a better prediction of shape memory polymer properties during the cooling process. In 2010, Kim et al. developed a three-phase model (a hard segment, an active segment and a frozen soft segment) based on the microstructure variation principle [206]. The stress and strain relationships of each phase are mathematically described using a three-element viscoelasticity equation and two Mooney-Rivlin hyperelastic equations to more accurately describe the shape memory behavior of polyurethane.

Table 4.2 lists a number of expressions for frozen volume fractions that introduce parameters to fit the shape memory behavior, but these parameters do not have strong physical significance, that is, they do not obviously correspond to the known properties of the material. Li et al. proposed a physical model in 2016 [207]. But this

area still needs to continue to study, more accurate and universal model to be developed. Finding the physical explanation of the volume fraction distribution is an important challenge for the future development of the phase transition method. In addition, phase transitions are also related to other mechanical conditions, such as stress [208] and strain rates [209].

4.2. Post-buckling analysis of SMPC

Driven by applications, surface-adhered thin films or wires, layered composite materials and fiber-doped shape memory polymer composites have drawn attention for use in substitution structures and in driving bi-functional materials. Shape memory polymers doped with long fibers and boards (thin films) must be explained by bending theory because of the property of large deformations (see Fig. 4.2).
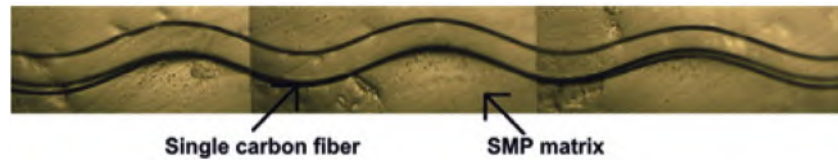
Fig. 4.2. A typical post-buckling phenomenon: single carbon fiber in the shape memory polymer buckling, showing a similar sine wave form [214].

Bending refers to the phenomenon that when the structure loses stability, that is, when loading reaches some threshold, the structure will suddenly switch to another neutral balance. This is a problem of structure stability that was discovered long ago and has been researched for centuries [215]. Theoretical studies in this field are highly mature, dating back to the 18th century, when Euler et al. researched the stability of rods under compression. However, bending analysis in geometry and mechanics, specifically post-bending theory, has been a widespread focus in recent decades and has become the common focus of subjects including applied math, biology, material science, mechanics, physics and engineering [216]. Early studies on bending mainly sought to avoid bending in the engineering of architectural structures. On the other hand, in recent decades, scientists have found numerous fantastic cases of bending in nature. The extensive geometric deformation can achieve new functions [217–219], such as the bending of thin beams and shells in stretchable electronics [220,221], complex 3-dimensional structures [222,223], materials with negative Poisson ratio [224–226] and adjustable acoustic super materials [227,228]. Furthermore, the changes in surface curvature produced by wrinkling and folding, facial chemistry [229], wetting [230], force of cohesion [231] and resistance can be controlled. With the increase in research pursuing smart structures, bending is a common focus because of its unique energetic and geometric properties, which are useful in applications related to energy (such as sensors, energy collection, damping accessories and drivers) and in the field of structures that undergo large deformations.

There are multiple cases in which elasticity loses stability [232–240]. In addition to compression, stretching can also cause elasticity to lose stability to establish a backward-bending shape. At present, smart structures consisting of shape memory polymers mainly use the compression bending of fibers or boards. This essay mainly discusses the conditions of bending the and geometric properties of backward-bending, which have broad applications in the design of driving devices and in establishing structures using shape memory polymers.

A thin membrane with surfaces consisting of large-deformation material will be compressed as the matrix is compressed, and the elasticity will lose stability when the reaction reaches a certain extent, which manifests as corrugation of the surface. However, this structure strengthens shape memory polymers and does not sacrifice their applications in structures that undergo large deformations. On the other hand, nanoscale surface corrugation structures will introduce new properties to the material, such as photon structures, anti-drag in fluids, and super-hydrophobicity [241]. Traditionally, this surface structure was achieved by top-down methods, such as photoetching and impression. These methods are relatively comprehensive, but they require complex machines and are expensive. However, using bottom-up methods could produce this surface structure efficiently and precisely [54] [242–246]. The corrugation and thickness of thin membranes are related to dynamic properties such as the shearing modulus, and thus this method is highly designable. Huang et al. reported the bending of semi-conductor membranes on periodically attached silicon boards [205,247–251]. In addition, when the boards were warped during compression, nanowires and tubes were fixed to pre-loaded boards. In the work of Huang and Zhang et al., this method has undergone numerous theoretical studies [252–255], which had significant and broad applications in many fields of science, including biomedical devices, MEMS systems and memory storage systems.

When a thin membrane is compressed on a single axis, it performs like a sinusoidal wave in the backward-bending phase. The performance on two axes is more complicated and can take the form of a chessboard, a hexagon, a "Y" and various other shapes. In addition, the shear force also influences the shape. Shape memory polymers have special advantages in establishing this kind of surface shape, such as unloaded long-term storage and controllable shape recovery. It is obvious that shape memory polymers have particularly high potential for application among all kinds of smart structures that use active deforming materials.

There have been many studies on the bending dynamics of tough surfaces on thin membranes of soft substrates and on backward-bending shapes [201,256–259]. In 2012, Chen et al. examined the corrugation of coating films of shape memory polymers and used viscoelastic models and the principle of lowest energy to analyze the backward-bending shape of surfaces of composite polymers [260]. After experiments examining the influences of the programming and the membrane thickness on the wavelength and amplitude of corrugation, the theory gave good predictions within a wavelength range of 930 nm to 5 μm. Leng et al. analyzed the influence of the curvature and thickness of the membrane on backward-bending shapes for shape memory polymers with their surfaces strengthened by metal films and subjected to large deformations, and they analyzed the surface bending by the finite-variable method [261]. Furthermore, this group analyzed the dynamics of one-way fiber-strengthened shape memory polymer composite materials after local bending [262]. As shown in the figure, the horizontal sections under curved bending can be classified into non-bending stretching, non-bending compression and bending compression sections, which are characterized by the critical position of bending, the position of the neutral surface and the half-wavelength of optical fibers. Additionally, the critical curvature of bending, critical position of bending, position of the neutral surface, wavelength of the fiber bending, amplitude of the fiber bending and macroscopic reactions of the structure were calculated.

It should be noted that bending could occur only in thin films. For thicker laminates or a larger modulus, the overwhelmingly large shear force prevents the bending of the fibers or boards. The materials appear to bend as a whole, which departs from the research focus on bending. However, these cases are still of interest, since they can be used for origami structures and have bright futures in rapid production and self-loading. The dynamic theorems are relatively simple and will not be introduced here, whereas the details of application will be introduced in Section 5.3.

5. Applications of SMPCs

Applications have always been the focus of studies on shape memory polymers and their composites. During the 1960s, shape memory polymers gained their first large-scale applications, namely, the use of PE thermal contraction tubes [263,264], which now are made from nylon [265] or polystyrene [266] materials. Thermal contraction tubes have good flame retardance, insulation and temperature tolerance properties and have been widely used in insulated joint protection, harnesses and erosion prevention, as well as having broad applications in electrical work. This was the most meaningful large-scale application of shape memory polymers and thus attracted scientific research attention. Since then, studies of shape memory polymers have undergone rapid development.

To develop applications in which shape memory polymers are central, we need to know the properties of shape memory polymers. First, we consider two-phase shape memory polymers, which have abundant useful properties and have been researched most thoroughly. The essential information is that they exhibit single-direction recovery, that is, in an unloaded recovery process, unless we reprogram them, shape memory polymers can transform only from temporary shapes to their original shapes. Even though heating or other stimulations during the recovery process are highly designable, their one-way dynamic behavior still greatly limits their applications. As a result, we consider using shape memory polymers and their composites only in structures with mainly one-way deformations. One major type of such applications aims at solving transportation problems by enabling transportation in a convenient shape and recovery to shapes needed for functions at specified times, as in the case of spatially expanding structure and minimally invasive surgical instruments. Another significant application of shape memory polymers is self-arrangement, in which the operation conditions perform cold programming. In this case, shape recovery can be utilized to let instruments and structures recover from plastic deformations accumulated during use, thus extending their lifespans.

Additionally, shape memory polymers are a type of elastic-adhesive elastic material, that is, they show normal elasticity without stimulation and show adhesive elasticity under stimulation, which must be taken into consideration in engineering applications. As shape memory polymers are macromolecular materials, their programming and recovery are both creeping processes, which makes their shape memory behaviors time-consuming. On the one hand, the reaction time gives smart structures based on shape memory polymers the property of low impulse, but on the other hand, it limits applications involving quick reactions.

Shape memory polymers are limited to their lower thermodynamic properties and single stimulation mode, so making shape memory polymer composites are extremely valuable. In the aerospace field, shape memory polymers have a tendency to replace shape memory alloys and incorporate structural materials with their light weight, high deformability and high fixation rate. Through composite fiber materials, the hardness, hardness ratio, recovery forces and other thermodynamic properties could be greatly enhanced while maintaining the above advantages. In the field of biomedical instruments, shape memory polymers are mainly used in appliances, brackets and occluders. Shape memory polymer smart structures applied inside the body, such as minimally invasive surgical instruments, require methods of stimulation without direct contact, which are often realized by the indirect heating of doping materials. Furthermore, shape memory polymers could be used to make substrates, sensors, energy gathering devices and soft electronic accessories. Shape memory polymer composite materials also have special value for applications in the fields of smart fabrics, 4D printing, etc.

5.1. Applications in aerospace and aviation

Because of their stable single-deformation ability, shape memory materials have attracted broad attention in all kinds of aerospace expansion structures and in driving machine-based locking-release structures with limited deformation times. Early studies of variant structures, mainly using shape memory alloys to provide driving forces, included a series of applications of spatially expanding hinges [267,268] and variant structures in aircrafts [269–271]. Their relatively large density (6–8 g/cm3) made it difficult to use shape memory alloys as structural materials, and they were necessarily accompanied by other light materials in the form of wires or boards, which increased the difficulty of design and introduced the inconvenience of choosing the accompanying structural materials. On the other hand, the shape memory effects of the shape memory alloys themselves were not satisfactory. Therefore, shape memory polymers became a rising star in solving problems.

By comparing the properties of fiber-strengthened shape memory polymers to those of shape memory alloys, we directly obtained ideas of the necessary differences in application design. Because of the low mass, high toughness and large deformation of polymer-based shape memory composites, they could be made into matrices of large expansion structures (such as reflective antennas [84]), which achieved the combination of driving devices and structural materials.

In our previous review, the use of shape memory polymers in the aerospace and aerospace fields has been exhaustively analyzed. In this article, we further clarify the design principles, and introduced some typical applications, especially large-scale expansion of the structure.

The environment in space is much worse than that in the atmosphere. Polymers and other organic materials will be more severely eroded than metal or ceramic materials by atomic oxygen and ultraviolet radiation and will be subjected to extreme temperature differences, which will lead to loss of mass of polymers, reduction in their dynamic properties, or even complete loss of function [272–276]. As a result, shape memory polymers must be carefully selected and tested for applications in space. For shape memory polymers, these tests should not only consider conventional factors such as losses in mass and changes in components and modulus but also the loss of the shape memory ability. It has now been proven that epoxy resin-based and cyanate ester-based shape memory polymers have good tolerance of the outer-space environment. Furthermore, because of their outstanding thermodynamic properties (especially the tolerance of high and low temperature cycling) and stable chemical properties, polyimide-based shape memory polymers have been identified as ideal candidates for future applications of shape memory polymers in space. Similarly, fillers with good space tolerance such as carbon material and glass fiber must be chosen when making shape memory polymer composites. When doping with materials with active chemical properties, such as healing agents, special design considerations are necessary to prevent these active properties from being influenced by space radiation (see Fig. 5.1).
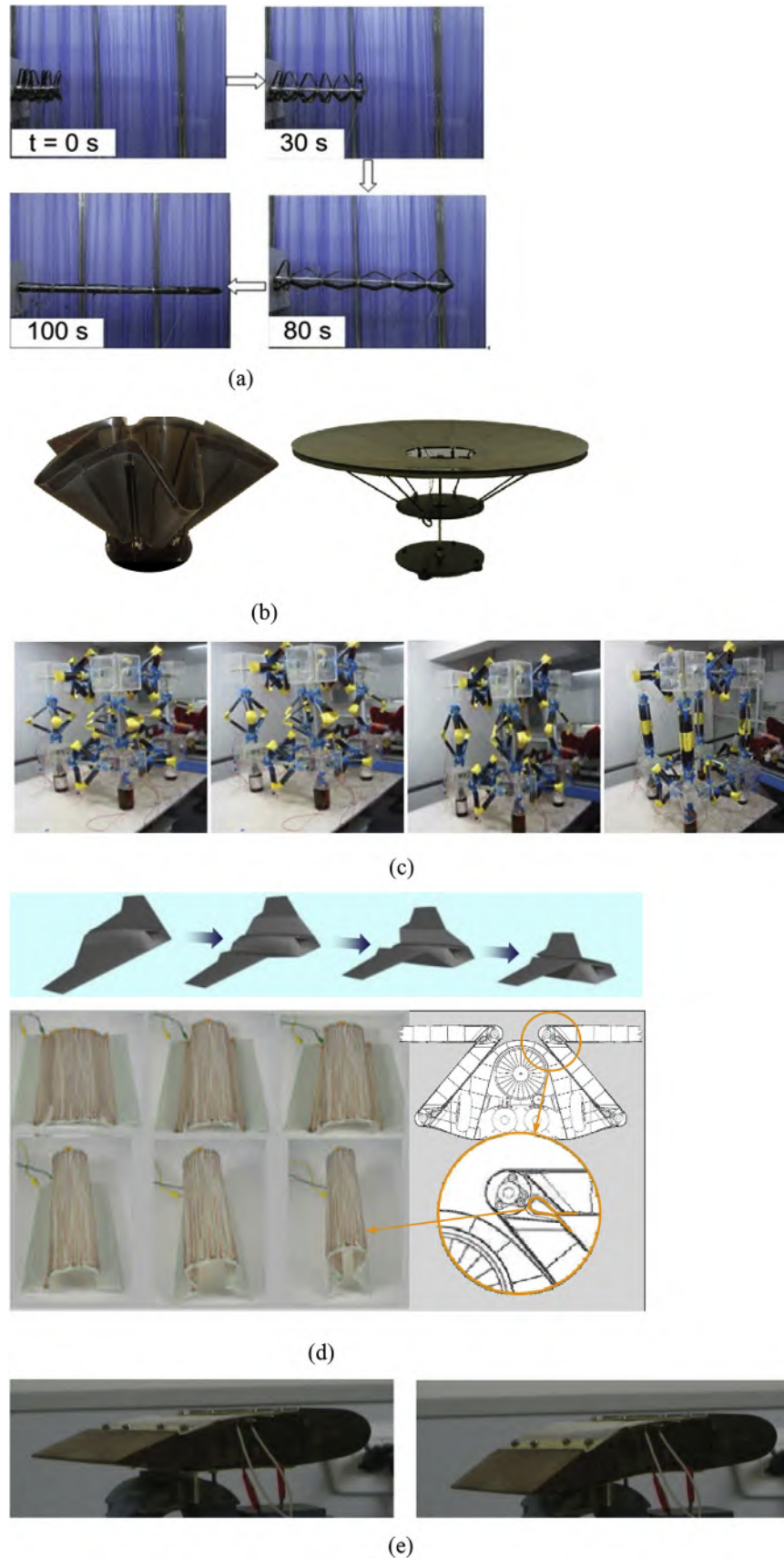
(a)

(b)

(c)

(d)

(e)

Fig. 5.1. (a) Modular shape memory polymer composite space deployment truss. A variety of trusses can be easily produced by this unit [83]. (b,c) Large deployable shape memory polymer composite structure. (b) Inflatable antennas based on SMPC-based reflector substrate [277].(c) Expandable space accommodation [278].(d,e) Application of shape memory polymers and composites in morphing aircraft. (d) Morphing aircraft with foldable wings. The skin of the deformable part is made of SMP [279,280]. (e) Steel wire springs are incorporated into the SMP to provide the dual functions of strengthening and heating [281].

5.2. Biomedical devices based on SMPCs

Shape memory polymers have been of interest to the medical field from the beginning, which can be dated back to the fourth decade of the last century when Vernon et al. first introduced shape memory polymers in a dentistry patent. Compared with metal materials, polymers offer suitable recovery force as well as good biological adaptability. Many shape memory polymers also have the ability to be decomposed in the body, such as poly-urethane [282], polycaprolactone [17] polyether urethane [12] and polylactic acid [283,284]. On the other hand, by making composite materials, the driving methods of shape memory polymers can be enriched. Among them, light driving (especially microwave) and magnetic driving offer the ability of remote driving, whose principles and examples have already been introduced in Section 3.2 and will not be restated. These two properties give shape memory polymer composites excellent potential for use in medical instruments used inside the body. Shape memory polymers and their composite materials have been used in many biomedical instruments, as described in the reports of prototypes and experiments. Their uses include thrombus cleaners, surgical sutures, intravascular stents, aneu-rysm occluders and orthodontic fixers.

Buckley et al. made original flower-shaped and inflated ball-shaped operation instruments using SMP and SMP foam with magnetic doping, which could be controllably expanded in the body through magnetic driving [285] (Fig. 5.2(a)). Small et al. wrapped a layer of shape memory polymer around fiber to achieve IR-stimulated shape recovery [286]. These indirect driving methods laid the foundations for the use of shape memory polymers for in vivo surgical instruments. Ali et al. used a double-sided Cu-coated polyimide to produce a magnetically stimulated shape memory polymer drug release mechanism based on simple heating stimulation [287].

Lendlein et al. reported a surgical suture made of shape memory polymers, whose temperature of transformation was near body temperature [17]. Experiments on animals illustrated that the suture underwent shape recovery when the temperature rose to 41 °C, closing the surgical incisions. Wache et al. devel-oped a new concept of stents using shape memory polymers [20]. This stent could be inflated to eliminate the problem that tradi-tional metal stents limited the transportation of medicine by making vessels narrower. Shandas developed a porous stent, which further minimized the necessary size of the trans-formation and provided convenience for minimally invasive cardiovascular operations [288]. Lendlein et al. reported a ureter stent made of decomposable shape memory polymers, which could provide directional dosing and decomposition [21]. This group also designed a directional dosing structure made of shape memory polymers, whose shape recovery could be realized by single-step or multi-step methods and which achieved the pre-dictable release of an effective loading of 90% or more in 80 days [289,290]. Cho et al. produced shape memory polymer floss for orthodontics using polyurethane base shape memory polymer by melt spinning. The balance recovery force was approximately 50 gf, which was sufficient to correct unadjusted teeth in ortho-dontic tests [291]. Yakacki et al. reported a decomposable shape memory polymer with a modulus as high as 23.0 MPa and used it to make soft tissue repair materials [292].

Hampikan et al. made aneurysm coils from shape memory polymers to replace traditional platinum coils, eliminating aneurysm re-opening caused by biological inertia [293] (Fig. 5.2(b)). The tantalum filler is compounded in a shape memory polymer, increasing the X-ray impermeability, thereby obtaining an X-ray image of the tantalum filled SMP coil. Through the simulation experiment, it is proved that the response of the coil does not affect the blood flow. Maitland et al. designed a medical stent to increase the visibility of X-ray or magnetic resonance (MR) imaging by doping with iron nano-particles or Gd chelates [294]. Metcalfe et al. reported a polyurethane-based foam occluder, which was proven by animal experiments to occlude aneurysms effectively [264]. The major-ity of aneurysm necks were sealed by the formation of a thick new internal membrane on the surface of foam bubbles. Small et al. designed a new embolization structure made of stents and foam [295]. Both parts consisted of shape memory polymers, and the structure was used to address and cure more difficult wide-necked aneurysms. Maitland made a new kind of aneurysm embolization structure using an inflatable SMP foam [296,297]. Platinum coils or a deployable alloy stent was used for anchorage, and the SMP foam could be inflated to over one hundred times its original volume (Fig. 5.2(c)). Hernandez et al. have designed a new hemostatic device made of shape memory foam that can rapidly fill wounds to achieve hemostasis and sterilization. The device expansion force is very small and will not cause secondary damage to the wound [298] (Fig. 5.2(d)).

5.3. Two types of enlightening applications: smart textiles, microelectronics

In addition to the particular focus on the aerospace and biomedical fields, shape memory polymers could also be applied to fields such as material shaping, smart fabrics, sensors, energy collecting devices, and soft electronic device substrates, among others.

For complicated structures, especially for shaping bent com-posite material structures, the making of dabbers is a challenge. Traditionally, there are two solutions: multi-piece metal man-drels and water-soluble mandrels. The former can ensure good precision and repeatability, but the design, production, installa-tion and uninstallation are expensive. The latter are seriously limited because of disposability and hazardous residues. At this time, shape memory polymers have become a candidate for solving the problem. Compared with alloys, shape memory polymers and their composite materials have greater deforma-tion ability, which makes it easier to produce more complex structures. The CRG company used shape memory polymers to produce bottle-shaped and S-shaped air duct structures, whose dabbers could be used repeatedly and were easy to knock out, and offered ideas for making complex bent structures [299—301]. Leng et al. further designed shape memory polymer dabbers using the dynamics of composite materials [302,303]. Finite variable analysis and illustrative experiments showed that shape memory polymer dabbers had good deformation and repeat-ability. However, the transformation temperature of shape memory polymers, which has been researched somewhat
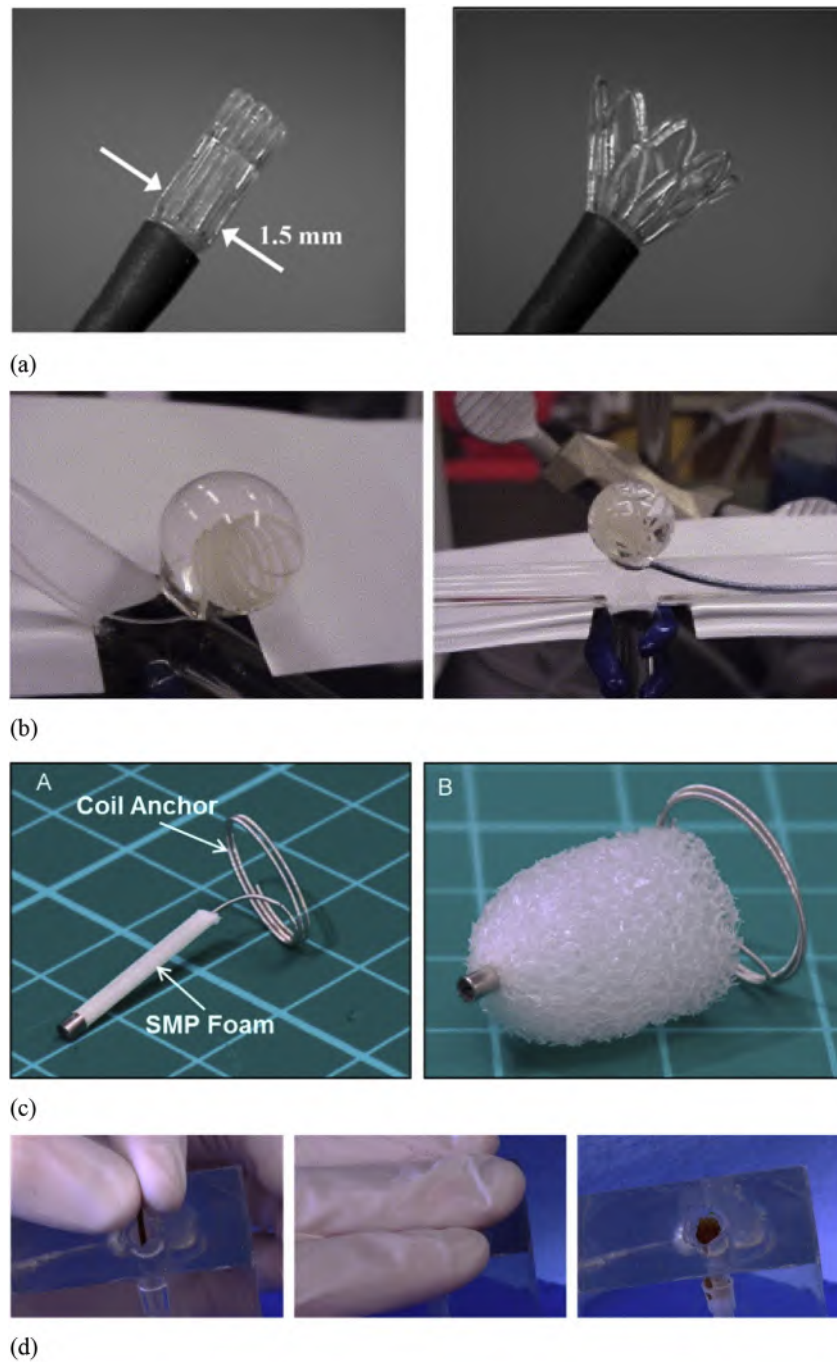
Fig. 5.2. Biomedical devices based on SMPCs. (a) Flower-shaped surgical instruments made of SMPC with magnetic doping. (b) Aneurysm coils based on shape memory polymers without platinum coils. (c) Aneurysm embolization structure based on inflatable SMP foam, which can expand more than a hundred times. (d) Hemostatic device based on shape memory foam that can rapidly fill a wound to achieve hemostasis and sterilization [267,275,278,280].
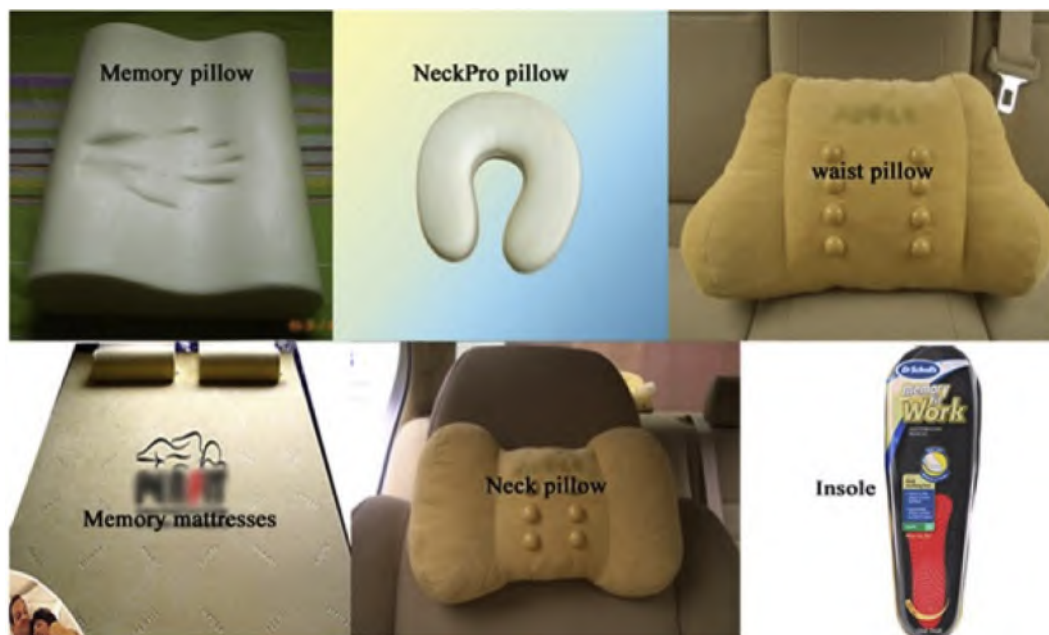
Fig. 5.3. Items made of shape memory polymers: pillows and insoles [22].
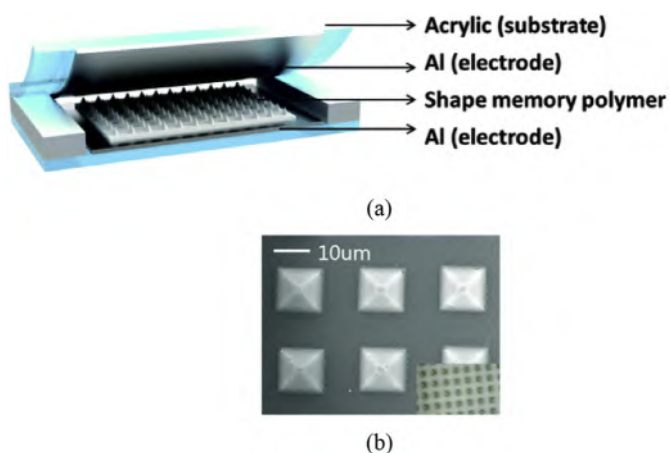

Fig. 5.4. Shape memory polymer-based self-healing triboelectric nanogenerator. (a) Schematic of the PU–TENG structure. (b) PU pyramid pattern on the PU triboelectric layer [24].

thoroughly, still presents difficulties when applied to materials that require higher temperatures for shaping. The choice of materials is a further direction for research on shape memory polymer dabbers.

Shape memory materials have the property of self-finishing, that is, the fatigue and injury introduced in the use of shape memory material devices or by various complex physical or chemical factors in the working environment, especially the material deformation caused by external forces, can be regarded

to some extent as programming processes of the shape memory materials. This means that the materials could be recovered to their original shapes under stimulations such as heating, or as the saying goes, "the phoenix reaches nirvana and rises from the ashes". This property has been applied to the smart fabrics field and to self-healing devices. Shape memory polymer fibers can be obtained by moist spinning [304], melt spinning [305] or electric spinning [306—308] to make textiles or non-woven fabrics. The main reason for wrinkling in textiles is the lack of adjustment of hydrogen bonds. By making textiles from shape memory polymers, wrinkles could be automatically removed through stimulations such as heating [22,309]. Furthermore, self-finishing insoles and pillows could be made using shape memory polymer foam [22,291], Some examples are shown in Fig. 5.3.

Kim et al. reported a new kind of triboelectric nanogenerator (TENG), with a frictional electric layer made of shape memory polymers [24]. A diagram and a photo of the structure are shown in Fig. 5.4. After its pyramid structure is worn down by long-term use, shape memory effects could allow the devices to regain their original properties, extending the lifespans of the TENGs. This approach is an effective way to extend the lifespans of electronic or mechanic devices.

In addition, shape memory polymers are used in soft electronics. Soft electronics is a field focused on biological electronic techniques. Traditional hard electronic devices cannot adjust to complex and soft biological surfaces, and soft electronic devices exhibit better properties and cause lower discomfort in use [291—311]. Compared to normal soft electronic devices, there is a benefit to using shape memory polymers. That is, their hardness is changeable. When heated, the shape memory polymer is in a soft and programmable state, which could allow the adjustment of its properties or its self-finishing. When the temperature is
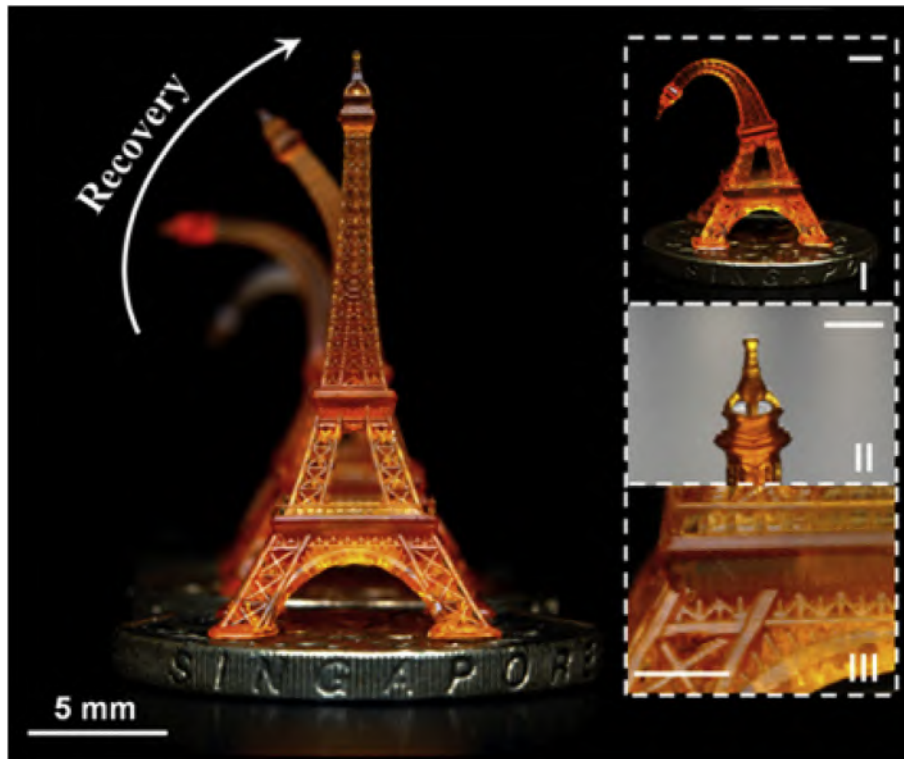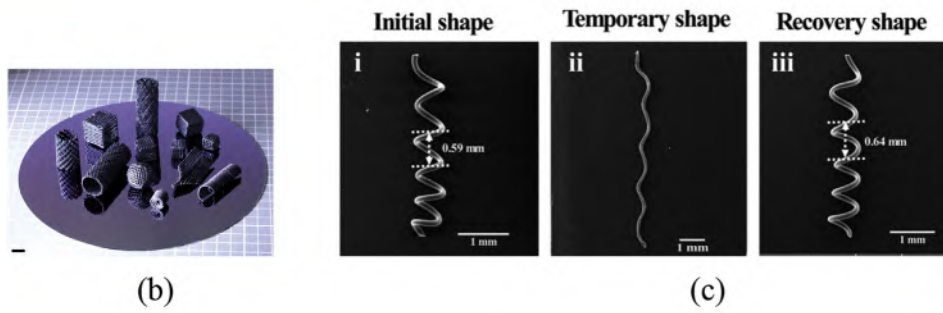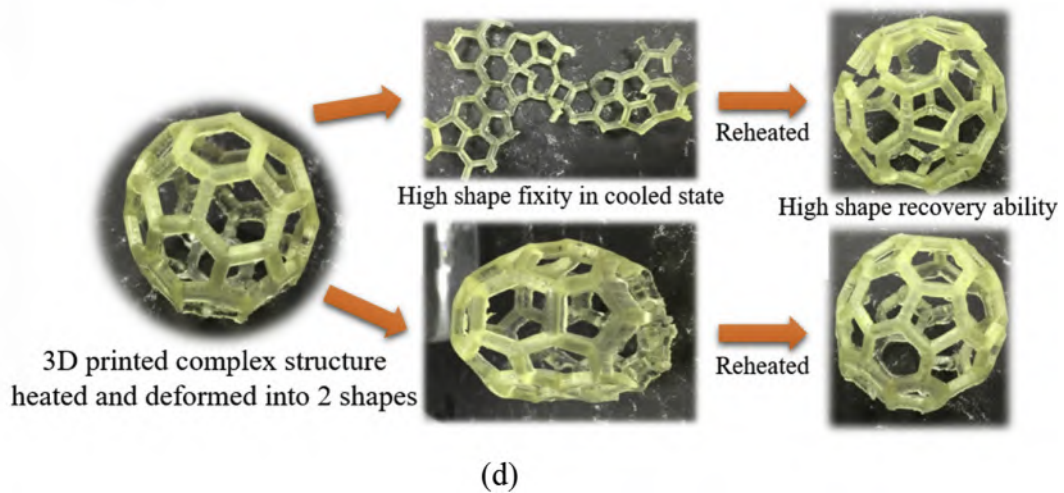
Fig. 5.5. 3D printing product of shape memory polymers and composites. (a) 3D printing of the Eiffel Tower based on shape memory polymers, with meticulous detail and outstanding ability to recover. (b) SMPC 3D printing based on ESBO and BFDGE, with carbon nanofibers (CNFs) added as fillers to tailor the mechanical properties and Tg while inducing electrical conductivity. The scale bar is 1 cm. (c) 3D printing of SMPC doped with ferrite to achieve remote control of the shape change. (d) Twenty truncated printing surface created by stereolithography apparatus (SLA) showed good shape memory properties under both cutting in flattening and flattening cases [302–323].

reduced to room temperature, it enters a tough and stable state, which can stabilize its properties and offer structural supports. Voit et al. made 3-dimensional stimulation electrodes using shape memory polymer substrates whose hardness could change by over 100 times for automatic adaptation to the geometric shapes in the bodies of creatures [312]. Animal experiments proved that this electrode effectively avoided inconsistency between biological and non-biological surfaces [294]. This group further made crystal tubes with organic thin membranes using shape memory polymers through full photolithography [313]. This system exhibited excellent electric properties and reliability and retained its function after 100 cycles of softening and bending. Leng et al. incorporated gold electrodes into shape memory polymers. Experiments showed that the resistance was an exponential function of the reactance. These structures could be used to make adjustable resistors or sensors [314]. In addition, Luo et al. reported that resistors containing carbon nanotubes and shape memory polyurethane composite material showed sensitivity to water and thus could be used to make moisture sensors [315]. Furthermore, this group combined surface layers of shape memory polyurethane with silver nanoscale wire (AgNW) and observed that the resistance of the composite materials was sensitive to temperature, enabling the construction of soft temperature sensors [316]. Zhong et al. used layer-by-layer (LBL) techniques to make a new type of super capacitor from multi-walled carbon nanotubes and shape memory polymers, which provided excellent stretchability, shape memory capacity and energy storage properties [317]. Kipplen et al., using wearable biologically consistent shape memory polymers as substrates, developed a green electroluminescent phosphorous organic diode, which had excellent lighting and self-finishing ability [318].

5.4. A potential share: 4D printing and origami

Shape memory polymers also have some status in self-installation fields. Macroscopic independent installation is a concept with strong prospects. This creative thought not only is highly interesting but also would reduce storage space and transportation, and its low cost and fast production give it great potential for development. The changes in shape of most shape memory polymers are permanent and stable. Even though they lose the ability to perform repeated driving, this property also makes them irreplaceable structural materials that achieve permanent shapes in independent installation. Using convenient, quick and large-scale handling methods, simple structures could be processed into complicated shapes. One idea is 4D printing.

4D printing is an improvement of 3D printing, introducing time into printed matter. And the change involves not only shape, but other properties tuning. They are all design directions (e.g. optics property). This change is also active, so 4D printing is actually the printing of smart materials containing time-dependent.4D printing has gradually become a hot point, just

recently, International Journal of Precision Engineering and Manufacturing-Green Technology [319] 4D published a monograph of 4D printing, which includes more than 6 articles about 4D printing review, so this paper will not repeatedly introduce in detail.

4D prints have unique or even irreplaceable advantages in many fields. For example, through the rational design shapes of idle and application, efficiency of production and transportation can be improved. Another example is the production of soft drive components, 4D print product can be used directly without the need to introduce complex mechanical components [27,320,321].

Magdassi et al. pioneered 3D printing of shape memory polymers [27] (Fig. 5.5(a)), and Lewicki et al. achieved 3D printing of shape memory composites [303] (Fig. 5.5(b)). Choong et al. used stereolithography apparatus (SLA) and modified two-component shape memory polymer to print a truncated twenty face body frame. Cut it off and flatten it, they get a pretty recovery result [194], which makes the practitioner feel confident (Fig. 5.5 (d)). Leng et al. achieved the 3D printing of polymers with excellent shape memory ability by the direct-write printing of UV crosslinking poly(lactic acid)-based inks doped with ferrite to achieve remote control of the shape change [322] (Fig. 5.5(c)).

Paper art is a kind of paper processing, or more abstractly, is the processing of the plane, so that the plane will get structure beyond the plane: Origami makes the structure develop from two dimensions to three dimensions, and kirigami increases the degree of freedom of plane. So paper art can be said a 3D printing process.

Traditional origami needs to be folded manually, allowing the material to yield at creases. It's a passive origami, it must be done step by step. And more importantly, it can't be printed. When a programmable material or composite system is used, origami becomes active, and becomes a printing deformation material. (Active completion does not mean that it should be done by one step, and active folding step by step can create complex structures such as riveting [26].) When printing deformable materials are introduced, origami has shown new vitality in the field of rapid flexible manufacturing. We can say that origami is such a simple and practical 4D printing method.

For 4D printing, the concept of active origami for flats has been reported, which is the idea of producing 3-dimensional structures from a 2-dimensional sheet. Quite complicated functional structures and microscopic structures have been made. Qi et al. incorporated shape memory polymer fibers into elastic polymer substrates, whose curvature was controlled by adjusting the reactance of the stretching [25,324—329]. Now the structure can realize complex structure, such as Miura origami and delicate origami [330]. Using this basic idea, a series of 4D printing structures were designed. Inspired by pop-up structures in MEMS [331—333], Felton et al. made multi-layer composite boards using pre-stretched shape memory polymers and created a series of origami structures [26,119,334] whose sizes reached as low as microscale [120]. We are looking forward to the development and maturity of self-installation (see Fig. 5.6).

(a)

(b)

(c)
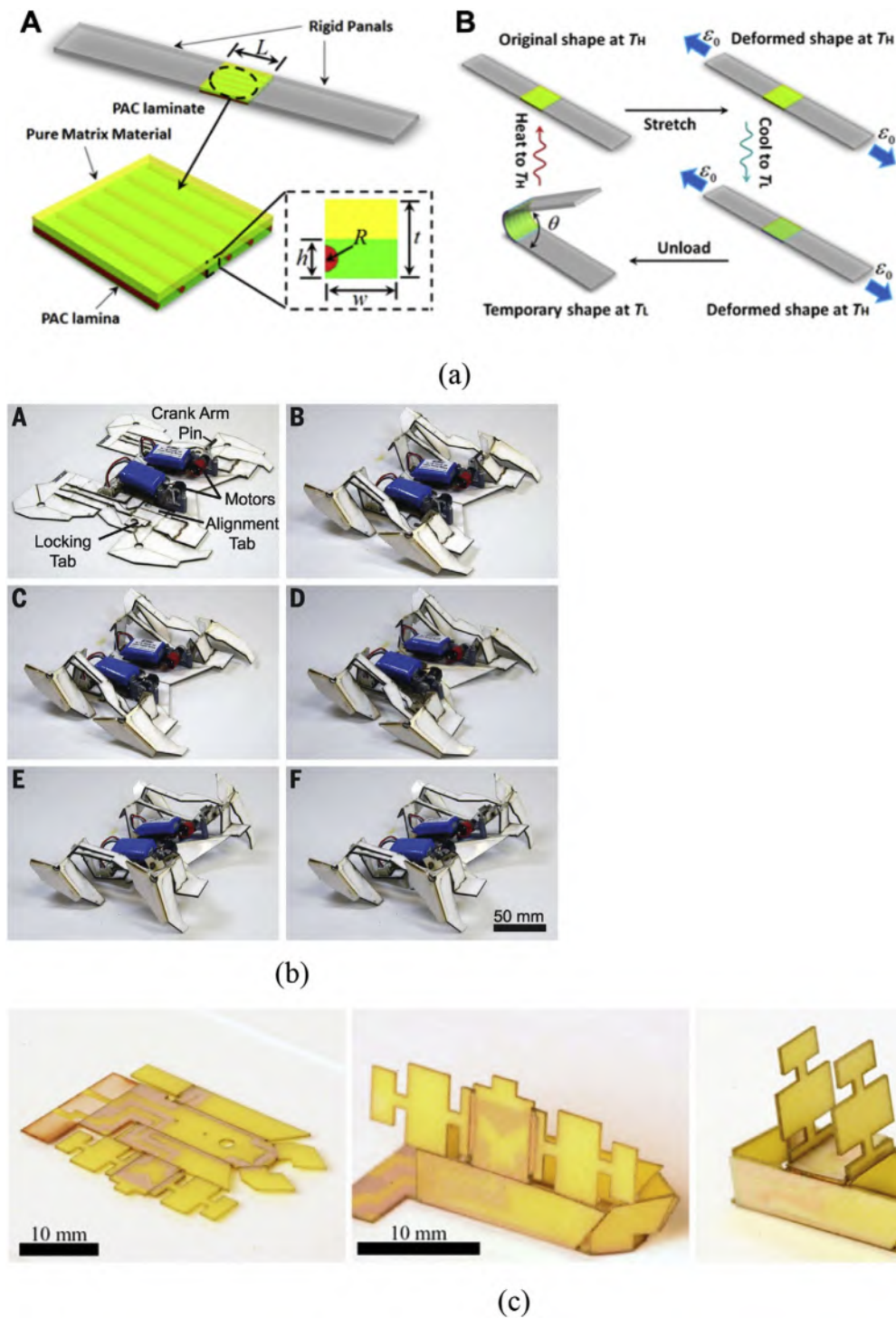
Fig. 5.6. Origami is achieved by laminating an SMPC. (a) Schematics of a PAC hinge and programming progress. In essence, this folding is also a loaded shape memory cycle produced by stretching at high temperatures. After the external load is released, the hinge is bent to the designed angle because of the competition between the base material and the shape memory material. Afterward, the hinge can be reheated and reused. (b) Creation of a movable robot based on origami. The figure shows the robot at each stage of assembly, three of which are self-folding. (c) A boat is made by a similar method, and its size is as low as the millimeter level [26,120,309].

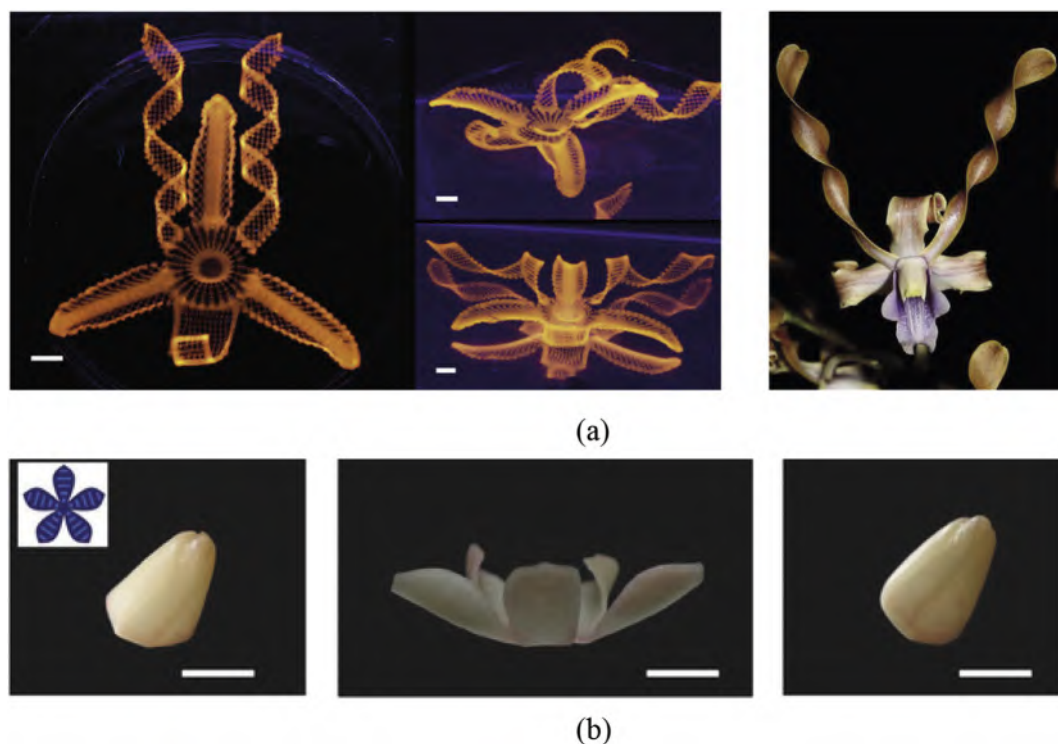**Fig. 5.7.** (a) Several shapes inspired by the native orchid on the right are printed by hydrogel composite with cellulose and swollen in water. The scale is 5 mm(b) Tulip, printed with shape memory polymer. From left to right are permanent shape, temporary shape and shape of recovery. Which light blue and dark blue corresponding to 2 s and 4 s of exposure, respectively. The scale is 1 cm [317,318]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Lewis et al. inspired by the plant, designed a printing method through cellulose-based hydrogel composite [335]. In different locations, the proportion of cellulose in the axial and normal orientation is different, so water swelling capacity is different. In this way, the flat print can be designed to swell into the corresponding shape (Fig. 5.7 (a)). Unlike above origami, the three-dimensional building obtained in this case is a non-developable surface. Zhao and Xie et al. also used the global deformation to design a 4D print based on swell equilibria [336], which utilizes ultraviolet light for different exposure time, allowing the hydrogels to achieve different cross-linking conditions, thereby changing the swell equilibria. The technology exposes the plane simultaneously, no longer rely on one-dimensional print head. Further, the group replaced the hydrogel with shape memory polymer (hydrophobic lauryl acrylate (LA) was used as the monomer, 1,6-hexanediol diacrylate (HDDA) as the cross-linker), and swelled in a wax (Fig. 5.7 b)). Benefited from the shape memory effect, which is obtained by the shape that can be fixed while the hydrogel cannot. Shape memory performance is excellent, the fixation rate of up to 100% and recovery rate of up to 95%.

The above methods can be designed by program calculation and manufactured by programmatic printing. Their production is convenient and fast. Therefore, we have reason to believe that this approach will lead to a revolution in production techniques.

6. Conclusion and outlook

It is not appropriate to discuss shape memory polymers without discussing shape memory effect, so I hope that increasing numbers of people will be able to learn about the shape memory effect as a general phenomenon of polymer mechanics that occurs through the material learning of shape memory polymers, i.e., it is determined by the properties of the material and the external factors together. This concept can guide their design, especially in cases of active deformation behavior. We distinguish and classify shape memory polymers and shape memory behavior mainly because of different focuses in materials science and engineering.

Shape memory polymers and their composites are excellent smart materials with outstanding properties and rich functionality. The development of composite materials (SMPCs) has greatly expanded the functionality and application range of shape memory polymers. In this paper, the flourishing of SMPC materials has been reviewed from the perspective of material function and application. Doping has been found to overcome the defects of shape memory polymers, including the minor restoring force and the limited stimulation methods, which enables the application of SMPCs in many fields from aerospace to biomedicine.

While appreciating these developments, we must also address many daunting challenges:

1. Although we have seen a wealth of development ideas involving shape memory behavior, the shape memory polymer is essentially limited by the great shortcoming of its own one-way deformation. The shape memory effect must be extended to two-way or combined with other active deformation principles, or shape memory polymers will be trapped in a small application area, such as deployable structures. We have seen that composites have made some initial progress in solving this problem (See Section 3.3 "Creating SMPCs with novel shape memory behavior"), and it is important to examine how composite concepts can achieve greater freedom or even full two-way recovery.

2. The electric driving of SMPCs is the stimulation method with the greatest potential in application, and its research is also the most abundant. Despite substantial encouraging progress, we must admit that electric driving is still in the experimental stage. Its

reliability is not high enough, and the cost is relatively high. Currently, we still use paste or buried resistance film for electrical heating, as we lack reports on the electrically stimulated devices and structures using the shape memory polymer nanocomposites. How to produce electrically stimulated SMPCs with high reliability and low cost is still the challenge we face.

3. We see in Section 3.1, "Reinforced SMPCs", that in most cases, when SMPs are changed to SMPCs, we can obtain better mechanical properties, but the shape memory effect will be reduced, mainly reflected in the fixation rate and the recovery rate. Especially for long-fiber-reinforced shape memory polymer composites, the most promising SMPC in engineering, a strong fiber will lead to a substantial decline in the fixation rate and recovery rate, a decrease in the transition temperature, and the development of non-negligible creep during storage. This situation is often difficult to solve with limited strain. However, sufficient stiffness is vital to the application of shape memory polymers in the field of drives. Therefore, a solution to this problem is essential for engineering applications of SMPCs.

In addition to these long and arduous challenges, we have seen some opportunities whose research is still in its infancy, such as the following:

1. The shape memory polymer itself and its auxiliary self-healing have great attraction in abundant multifunctional shape memory materials, but it is still a young research direction. Although there have been many studies of principle, today, due to the limitations of mechanical properties, cost, and healing effects, the progress of self-healing is still in the experimental stage, so the self-healing route in use in practical engineering applications remains long. In addition, other multifunctional materials and composites, including combinations of the shape memory effect with soft electronics, material health monitoring, energy collection and management, and radar-absorbing materials, have unique applications in electronic technology, aerospace, defense and so on. They offer promising future directions.

2. The use of shape memory polymers to create microstructures remains an area to be developed, particularly for the creation of microstructures inside shape memory polymers, as well as the incorporation of a polymer basis or polymer doping into new functional materials. Shape memory polymers have the ability to deform from the nanoscale to the macroscopic scale, so micro-materials designed with shape memory polymers will have tunable properties. In addition, shape memory polymer composites are unique in creating microstructures, such as by the use of composite filler materials as sacrificial components, and by the creation of microstructures by buckling. 3D printing techniques for shape memory polymers and their particle-doping composites have been studied. This is an important method for creating complex 3D structures, and the multi-component printing of SMPC can be used to produce novel functional materials.

3. For engineering applications, not only do shape memory polymers and their composites provide an alternative to shape memory alloys, which have been extensively designed and used in the past, but also their large deformability allows SMPs or SMPCs to fabricate structural materials from large deformable structures that were unimaginable in the past, such as parabolic reflection antenna. We should note that the greatest advantage of shape memory polymers is not the replacement of the bending mechanism in traditional structure but their benefits in curve and surface structures. We should break the constraints of traditional design ideas to design more ingenious and flexible structures and applications.

In recent years, shape memory polymers and the shape memory effect have become increasingly familiar to materials scientists and have attracted their research interest. SMP and SMPC research has undergone rapid and even explosive growth. We have reason to believe it will soon be even more vigorous.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.compscitech.2018.03.018.

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