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REVIEW

Shape Memory Polymer Fibers: Materials, Structures, and Applications

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Abstract
Shape memory polymer (SMP) is a kind of material that can sense and respond to the changes of the external environment, and its behavior is similar to the intelligent reflection of life. Electrospinning, as a versatile and feasible technique, has been used to prepare shape memory polymer fibers (SMPFs) and expand their structures. SMPFs show some advanced features and functions in many fields. In this review, we give a comprehensive overview of SMPFs, including materials, fabrication methods, structures, multifunction, and applications. Firstly, the mechanism and characteristics of SMP are introduced. We then discuss the electrospinning method to form various microstructures, like non-woven fibers, core/shell fibers, hollow fibers and oriented fibers. Afterward, the multiple functions of SMPFs are discussed, such as multi-shape memory effect, reversible shape memory effect and remote actuation of composites. We also focus on some typical applications of SMPFs, including biomedical scaffolds, drug carriers, self-healing, smart textiles and sensors, as well as energy harvesting devices. At the end, the challenges and future development directions of SMPFs are proposed.

Keywords Shape memory polymer · Micro/nano fibers · Electrospinning · Microstructures · Multifunctional composites · Applications

Introduction

Shape memory polymers (SMPs) are important active deformation materials. Under the excitation of thermal, electrical, light, magnetic, microwave and other external environment, SMPs can remember the temporary shape and return to the initial state [1–4]. This ability of sensing environmental changes makes SMPs more widely used in aerospace, smart fabrics, biomedicine, intelligent robots, flexible substrates, optics and smart tools, as well as electronic information carriers [5–10]. Compared with shape memory alloys and shape memory ceramic, SMP shave the advantages of light weight, large deformation, and easy processing. Therefore, they have attracted extensive attention by scholars all over the world [11–15]. However, the current research on SMPs mainly focuses on the design and application of macro structures, while the preliminary research on microscale structure is relatively less, especially in the biomedical field. With the development of smart structures and manufacturing technology, SMPs with porous structure such as shape memory polymer fibers (SMPFs) play a key role in drug release, tissue engineering, and interventional therapy [16]. In addition, SMPFs also have important applications in smart clothing [17–20]. So, the research on its structure and performance is of great significance.

Electrospinning is an important method to prepare nano or microscale fibers. Due to the simple operation, the fiber structure and morphology can be controlled, and the fiber diameter can be changed by adjusting the spinning process parameters [21–23]. In addition, different fiber structures and arrangements can be obtained by using different syringe needles and receiving plates [24]. The development of electrospinning technology promotes the development of SMPs towards the direction of nano-/microfibers, and expands the structure of SMPs. These characteristics play a key role in the development of SMPs [25–27].

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In 2005, Cha et al. [28] reported for the first time that copolymerized shape memory polyurethane (SMPU) was prepared into nanofibers by electrospinning technology, and proved the shape memory effect (SME) of electrospun SMPU nanofibers. In 2007, Meng et al. [29] obtained the porous non-woven fiber structure by changing the electrospinning device and adjusting spinning process conditions. Later, researchers verified that the shape memory nanofiber membrane has faster recovery speed than the ordinary non-fiber structure membrane under thermal driving conditions, and the SMPF membrane can still maintain good shape memory performance after repeated cycles [30]. Moreover, the research group of Lendlein [31] and Mather [32] reported that SMPFs were applied in the biomedical engineering field, such as cell culture and tissue engineering. The thermoplastic SMPFs including polyurethane (PU) and polycaprolactone (PCL) have been prepared into nano-/micorfibers through electrospinning. The fiber membrane prepared by traditional electrospinning method has a porous non-woven structure, which has a certain permeability compared with porous foam materials [33, 34]. With the progress of SMPs and manufacturing technology, the SME displayed in SMPFs prepared by electrospinning can not only remember two shapes, but also can memorize multiple shapes [35].In addition, on the basis of adding functional fillers, the shape memory composite fiber solves the problems of traditional direct thermal driven method and the single function, breaks through the limitations in practical applications, and promotes the development of SMPFs. With the increasing application of porous materials in various aspects, the preparation of porous smart materials and the fast and controllable driving speed are also urgent problems to be solved. Based on this background, it is necessary to study multi-SME, remote driving methods and multifunctional nano-/microscale SMPs. The research on multi-shape memory micro-structure and multi-functional small-size porous materials controlled by electric and magnetic field [36–38], which greatly promote the development of SMPs and lay the foundation for the future practical applications.

Shape Memory Polymer Fibers

Shape Memory Polymers

SMP is a kind of smart material, which belongs to the category of functional polymer. SME is mainly achieved by designing physical or chemical cross-linking points between molecular chains and movable linear molecular chains. More and more SMPs have been endowed with new SEM and multi-functionality [39], breaking through the limitation of traditional concept, including multi-SME [40–42], two-way SME [43, 44], new actuation methods and multifunctional shape memory composites by adding functional particles to achieve enhanced functions and remote controlling actuation under different conditions [45–47]. In fact, the emergence of shape memory composites promotes the rapid development of SMPs. In particular, some new actuation methods [48], selective driving [49] and self-healing effects [50] make SMPs and their composites have a broader application space.

According to the internal molecular structure of SMPs, this smart material shows shape memory properties. The transition phase (soft segment) and the stationary phase (hard segment) are two important phases in the molecular structure of polymer. The soft segment has a movable and reversible molecular chain (crystalline structure). When heating above the transition temperature, the molecular chain moves freely. When it is lower than the transition temperature, the molecular chain is fixed, which determines the fixity of SMPs. The hard segment has chemical or physical crosslinking point, which determines the recovery performance of SMPs.

Figure 1a is a schematic diagram of the molecular network structure changes of SMPs in a typical shape memory cycle. The process is that when the polymer is heated to a temperature higher than its transition temperature (T_{trans}), the material changes from glass state to high elastic state, and the random winding molecular chain has mobility. When the external force is applied to make the material macroscopic tensile deformation, the winding molecular chain is stretched. While maintaining the external force, the temperature is reduced, and the temporary shape is fixed. The internal stress is frozen, and the oriented molecular chain is fixed after stretching. When the polymer is reheated to a temperature higher than the transition temperature, the melting of crystalline phase in the material softens and the molecular chain returns to the winding structure, which causes the macroscopic shape return to the initial state.

At present, most researches are focused on SMPs with dual SME, which can memorize a temporary shape and a permanent shape (Fig. 1b). With the in-depth study of SME, polymers with multiple SME have attracted attention [40, 51]. Unlike traditional SMPs, which can remember two shapes, polymers with multiple SME can remember multiple shapes at different transition temperatures (Fig. 1c), such as triple SME, quadruple SME and quintuple SME. When subjected to external stimuli, it can be restored to the initial state in turn. On the one hand, the polymer has two or more transition temperatures. On the other hand, the polymer has a wider transition temperature range, in which the deformation temperature can be set arbitrarily. This multiple SME provides the foundation for the development and application of SMPs.

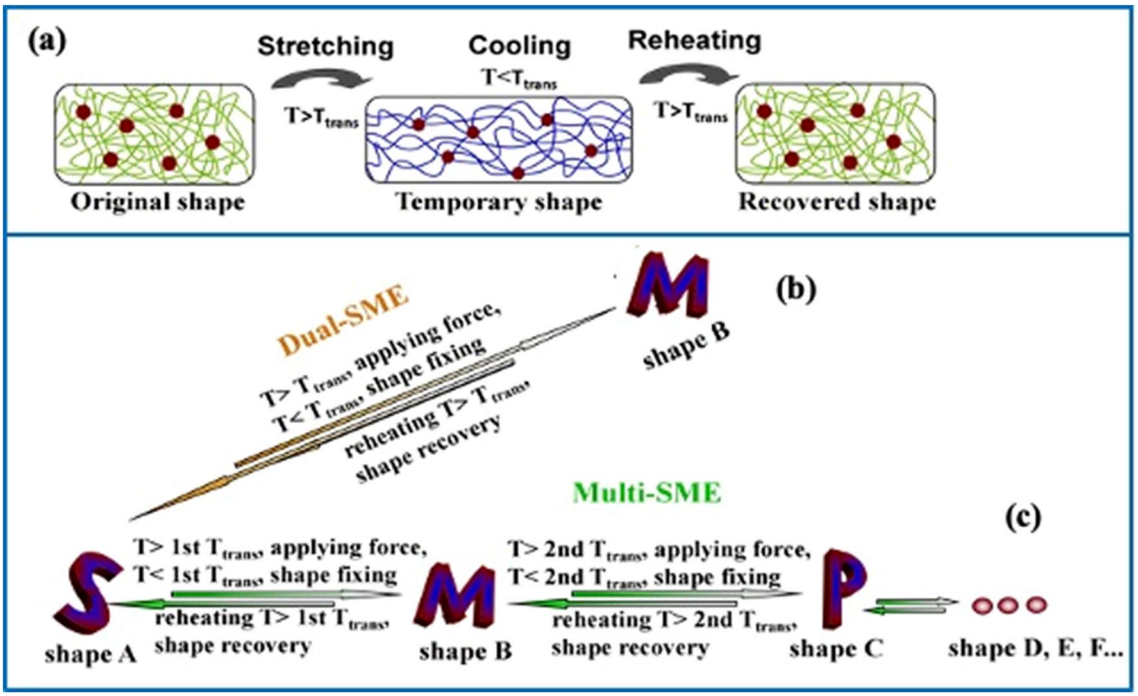


Fig. 1 SME: **a** mechanism, **b** dual SME, **c** multiple SME

Electrospinning Technology

In the early stage of SMPs, most research and applications are focused on large-scale bulk structures. In recent years, with the development and innovation of manufacturing technology, including the transfer printing, spin coating, electrospinning, 3D and even 4D printing technology, SMPs have been made into a variety of structures, especially small sizes complex structures, such as fibers, foam, particles, capsules, surface micro-patterned films and 4D printing complex structures, etc. [1, 2, 4]. These micro-structures meet the needs of practical applications of microscale devices, greatly promote the development of SMPs, and broaden the application range of SMPs.

As one of the various structures of SMPs, SMPFs have many advantages, so it has attracted extensive attention in various fields [52, 53]. SMPFs have large specific surface area, and the porosity of nonwoven structure makes it easier to penetrate other materials, and easy to achieve surface adsorption [54]. At present, SMPFs are mainly prepared by electrospinning, which has the advantages of easy operation and controllable fiber morphology [55–57]. The first SMP to be spun into nanofibers was PU [28]. After that, new SMPs were continuously prepared into fibrous structures, such as PCL [58], polylactic acid (PLA) [59] and other thermoplastic polymers [60]. These polymers not only have shape memory properties, but also have biocompatibility and biodegradability. Therefore, they have attracted wide attention in biomedical engineering, intelligent textiles, nanosensors and other fields [31, 32].

Electrospinning means that under the action of high-voltage electric field, polymer solution overcomes the surface tension of liquid to form a jet, and finally obtains fibers on the receiving plate. Figure 2a is a schematic diagram of a typical electrospinning device, which mainly includes a high-voltage power supply, a receiving plate, a syringe and a needle [61]. The principle of spinning is to form Taylor cone at the spinning port of polymer droplet under the action of electric field. When the electric field intensity is strong enough, the Taylor cone is stretched to form a jet flow, which extends to the receiving plate by the spiral track. During the spinning process, the solvent in the polymer solution evaporates, and then on-woven fiber membrane is obtained on the receiving plate. Figure 2b is a schematic diagram of a coaxial electrospinning device, which is mainly used to prepare core–shell structured fibers. The composite needle is used to realize the simultaneous injection of internal and external spinning solutions. Under the action of electric field, a composite Taylor cone is formed at the needle, and finally a composite fiber is obtained [62]. This coaxial electrospinning method can realize the multifunctionality of the fiber and its film, and fiber reinforcement function can be realized by adding functional materials into the core layer. In addition, the coaxial electrospinning method can also be used to prepare hollow structure microscale fibers, mainly volatilizing the core solvent in the core–shell structure fiber,

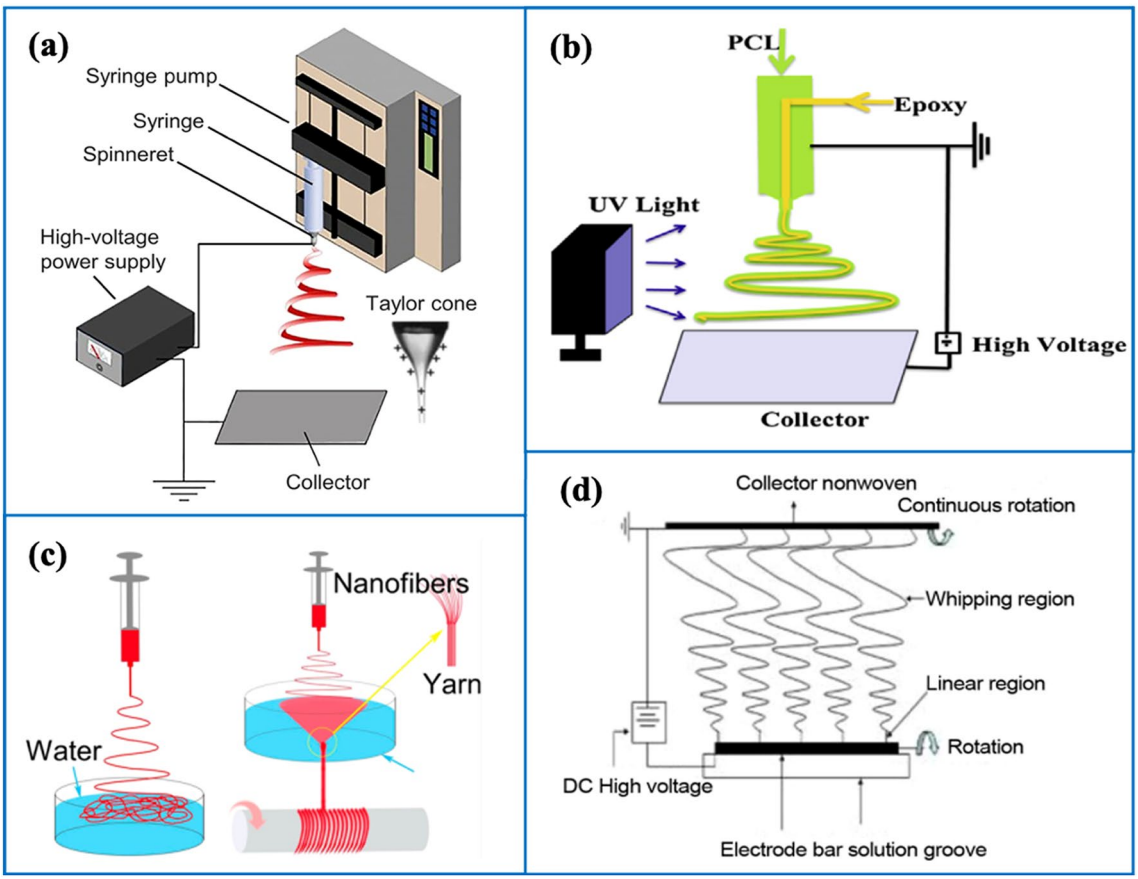


Fig. 2 Schematic diagram of the electrospinning device: **a** a typical electrospinning device [61], **b** a coaxial electrospinning device [62], **c** an electrospinning device using a roller shaft to receive and transpose as a receiving plate [63], **d** free liquid surface electrostatic spinning device [64]

and finally retaining only the shell polymer. Moreover, in order to obtain oriented fibers, the researchers used the roller receiving rotation as the rotating collector (Fig. 2c), and obtained continuous unidirectional fibers through high-speed rotation [63]. In recent years, electrospinning technology has developed rapidly and has been gradually industrialized. In order to improve the spinning yield, a new type of electrospinning machine called free liquid surface electrospinning device was developed by Elmarco Company of Czech Repblic. The charged liquid on the surface of the electrode rod forms numerous spray points in the high-voltage electric field, which increases the output and greatly improves the spinning efficiency. At the same time, it can effectively avoid needle blocking and other problems in the needle spinning device (Fig. 2d) [64]. Although the electrospinning technology has achieved fruitful results, its application in the preparation of SMPFs is still in its infancy. Most of the micro and nano fibers reported in the literature are nonwoven structures [65], and their single structure and function limit the development of SMPFs. Therefore, it is necessary to increase the breadth and depth of research on SMPFs.

Microstructures of Shape Memory Polymer Fibers

Currently, there are many types of polymers that can be used to make electrospun fibers (Fig. 3), including PU, PLA, PCL, and ethylene oxide. In the electrospinning process, solution concentration, feed rate, voltage, distance between needle to receiving plate, humidity and temperature can affect the fiber morphology [66–69]. In order to meet the needs of different applications, fibers with various morphologies can be obtained by adjusting the electrospinning process and parameters [70–72]. With the process of spinning equipment and technology, the fiber structure is becoming more and more diversified, and the shape memory micro/nano fibers with non-woven, core–shell, hollow, particle, and orientation structure have been successfully prepared [16].

Non-woven fiber is the most common form of electrospun fiber. The non-woven fiber adopts traditional flat receiving fiber. During the electrospinning process, the charged polymer at the tip of the nozzle flies to the receiving plate in a spiral shape, and the radius increases gradually, accompanied by uncontrollable factors such as solvent evaporation

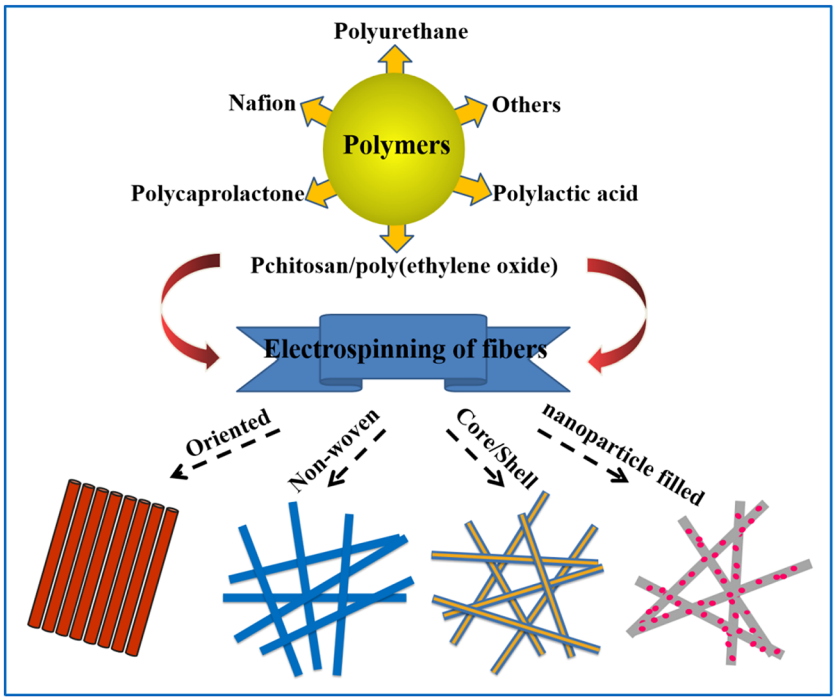


Fig. 3 Electrospinning fibers: materials and structures

and jet splitting. Finally, randomly oriented continuous fibers are obtained on the receiving plate [73–75]. Dan et al. [76] prepared SMPU fibers with non-woven structure by electrospinning. By changing the weight ratio of PCL and polydimethylsiloxane (PDMS), the mechanical properties and shape memory properties of the resulting fibers were investigated. Zhuo et al. [75] prepared nanofiers with nonwoven structure by dissolving PCL-based SMPU polymer (with 75% soft segment content) in dimethylformamide (DMF) at room temperature. The surface of SMPU nanofibers is smooth, but the distribution is uneven, with a diameter of 600–700 nm, forming a cross-linked fiber network structure (Fig. 4a).

The core–shell fibers prepared by coaxial electrospinning with unique structural advantages can be widely used in various fields. In the process of coaxial electrospinning, the external liquid and internal liquid are fed into the coaxial external and internal capillary tubes respectively. By adjusting the sample rate of the two, the coaxial line is formed under the high-voltage electrostatic field. Taylor cone forms a coaxial jet and receiving plate collects core–shell structure fibers. Hu et al. [77] used SMPU/DMF as the core solution and PySMPU/DMF as the shell solution to prepared new core–shell SMPFs by coaxial electrospinning, and tested their shape memory and antibacterial properties. In 2011, Cho et al. [78] prepared SMPFs with core–shell structure by coaxial electrospinning, using PU as core solution and PU/MWNT (multi wall carbon nanotube) as shell solution. Zhang et al. [62] prepared SMPFs with a core–shell structure by UV cross-linking with epoxy resin as core layer and PCL as shell layer. When the temperature is 70 ℃, the response speed of the fiber is faster, and the whole shape memory recovery process only takes 6.2 s, as shown in Fig. 4b.

On the basis of coaxial electrospinning, Zhang et al. [79] prepared shape memory hollow fibers with the same outer diameter and different hollowness, as shown in Fig. 4c. The shape memory hollow fiber was driven at 50 °C. After the first cycle, the results showed that the shape fixity ratio (R_f) with 13% and 33% hollowness were between 80% and 84%. When the hollow degree of the fiber was 0%, the R_f was more than 88% and the shape recovery ratio (R_r) was 59%. When the hollow degrees of the fiber were 13% and 33%, the R_r was around 66–67%. It can be concluded that the R_f coefficient decreased and the relative radiation coefficient increased with the increase of the SMPF hollowness.

In order to increase the functionality of SMPFs, multifunctional or modified SMPFs were prepared by adding functional fillers into the spinning solution [80–82]. Gong et al. [80] reported that the non-woven structure of shape memory PCL nanofibers. Magnetic Fe_3O_4 nanoparticles were added into the spinning solution to achieve SME and realize remote driving control. Increase the functionality of shape memory nanofibers, Zhang et al. [83] prepared Nafion/Fe_3O_4 composite nanofibers with particle filling structure by adding Fe_3O_4 nanoparticles to poly(ethyleneoxide) (PEO)/Nafion spinning solution through typical electrospinning technology (Fig. 4d). The composite fiber has excellent shape memory performances, and can also realize remote magnetic actuation control. Tan et al. [82] reported the preparation of functional SMPU composite nanofibers by adding

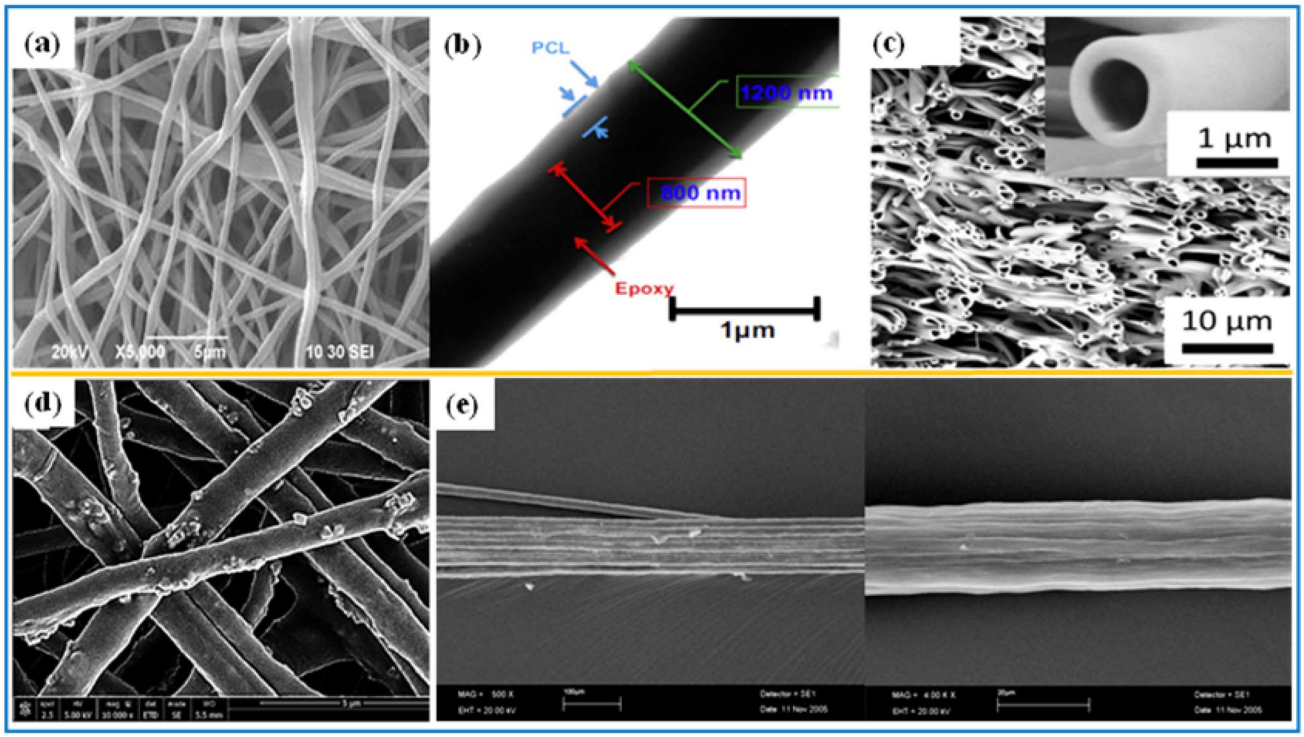


Fig. 4 SMPFs with different structures: **a** non-woven structure [75], **b** core–shell structure [62], **c** hollow structure [79], **d** particle-filled structure [83], **e** oriented structure [84]

graphene oxide fillers into the spinning matrix. The addition of graphene oxide improved the mechanical properties, thermal properties and surface wettability of the composites.

Gong et al. [80] changed the plate receiving device into a roller receiving device, and prepared the oriented nanofibers with SME for the first time. Later, Schneider et al. [31] obtained oriented SMPFs of poly(ether)esterurethane (PDC)/1,1,1,3,3,3 hexafluoro-2-propanol (HFP), poly(*p*-dioxanone) (PPDO)/HFP and polyetherimide (PEI)/dimethylacetamide (DMAc). Ji et al. [84] fabricated SMPU solutions with different hard segment contents into fibers. During the spinning process, PU molecules were oriented along the fiber axis to form electrospun nanofibers with oriented structure (Fig. 4e). Compared with PU film, this PU fiber has better shape recovery property, and the fiber has the strong shape recovery ability in the axial direction. In 2013, Mather et al. [85] dissolved poly(vinyl acetate) (PVAc) in MeOH/DMF mixed solution for electrospinning, and chemically cross-linked fibers with different orientations with PDMS to obtain a glass transition temperature (T_g) of 55 °C. The R_r and R_f of SMPFs were 98% and 100%, respectively. In 2015, PCL and PVAc were used to prepare a fiber with SME assisted self-healing property. The resulting fibers have up to 99% strong healing ability [86]. In 2016, the experiments related to the bi-directional electrospinning of PLA and PCL were carried out [87]. Although the oriented fibers are still in their infancy, the appearance of this structure has greatly enriched the structure of SMPs and plays a positive role in promoting the development of SMPs.

Multifunctional Shape Memory Composite Fibers

SMPFs can be combined with some functional materials, showing excellent shape memory performance and versatility [88]. In recent years, SMPs as a kind of stimuli-response polymers have shown excellent material properties, especially the actuation method, which plays a very important role in the development of SMPs and their composites. According to the needs of practical applications, more and more driving methods have been extensively studied. At present, most traditional SMPs are unidirectional and can only remember two shapes, one permanent shape and one temporary shape. In order to achieve the multiple deformations of materials and structures, researchers have investigated the multiple SME and reversible SME. Moreover, adding different function fillers can make the shape memory composite fiber have different actuation methods, such as remote control.

Multiple SME means SMPFs can remember more than two shapes. Zhang et al. [89] reported that electrospun Nafion microfiber membranes have multiple SME. Nafion fiber membrane has a wide transition temperature from 55

to 170 °C, which enables the fiber membranes to remember three, four or five shapes at the same time, and can return to the original state at the corresponding deformation temperature (Fig. 5a). Subsequently, Zhang et al. [83] added magnetic nanoparticles into the Nafion system and electrospun multiple shape memory nanocomposite fibers with remote actuation control. Recently, Sabzi et al. [90] successfully prepared a triple SMP scaffold with a phase-separated structure using PLA and PVAc miscible polymers as raw materials by co-spinning, and adding graphene nanosheets to SMP to characterize the changes in shape memory performances. The experimental results show that a series of composite fibers are woven into a network with smooth surface. At the

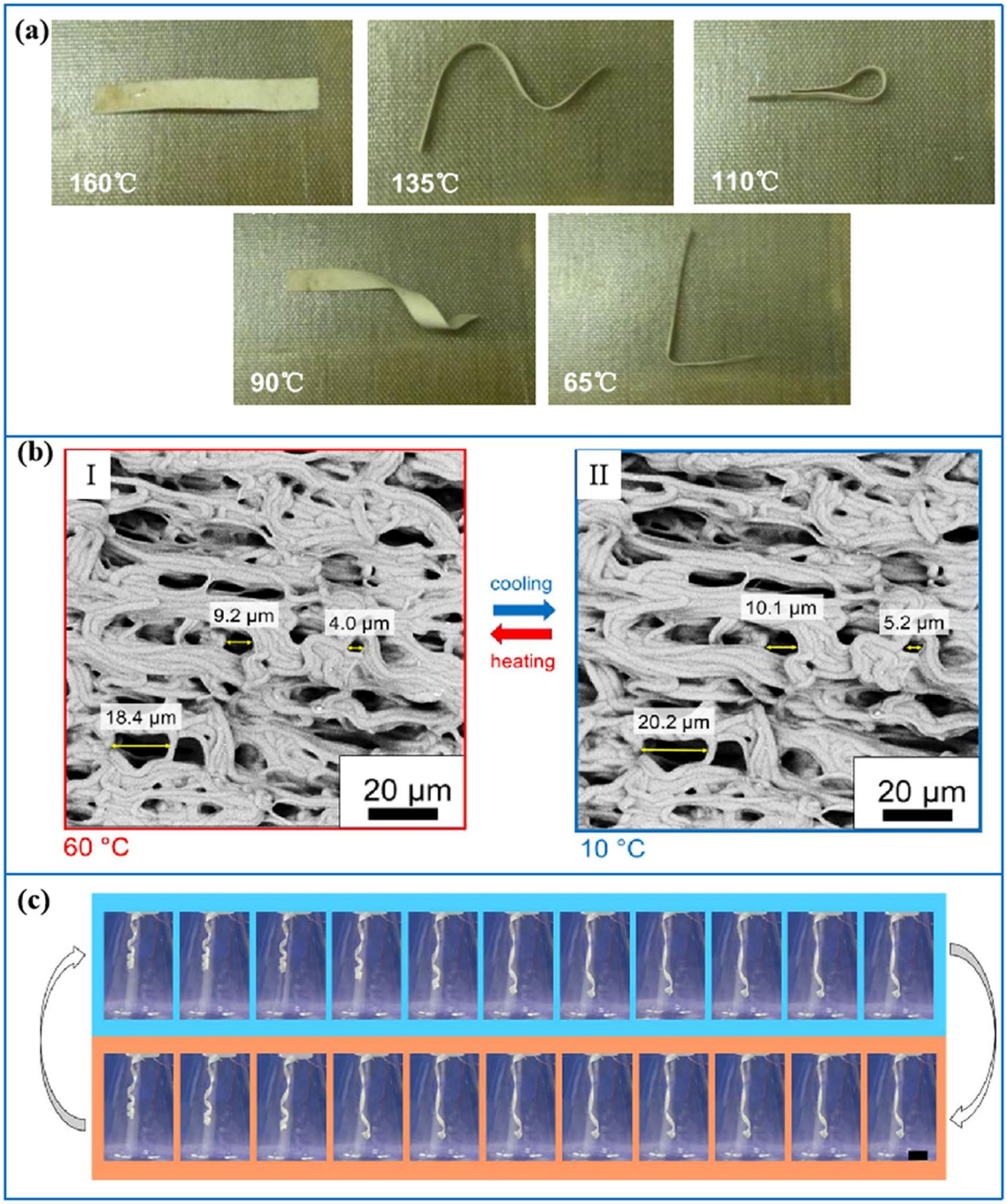


Fig. 5 SMPFs with multifunction: **a** quintuple-shape memory property of electrospun Nafion microfiber membranes [89], **b** SEM images of cPCL [92]: (I) heating to 60 °C, (II) cooling down to 10 °C, **c** series of images extracted from a video of sequential heating (lower, orange) and cooling (upper, blue) for a twisted specimen investigated in the range of 60–10 °C [92]

same time, the addition of graphene nanosheets can improve the conductivity and elastic modulus of polymer solution.

Reversible SME plays an important role in many applications. However, most SMPFs are unidirectional. How to realize the bidirectional function of SMPFs is very important to the practical applications. Reversible SMPs can realize the repetitive conversion of two different shapes driven by external stimulation. Chen et al. [91] reported that SMPU nanofibers had reversible SME. The SMPF membrane exhibited different morphologies before and after deformation. When the fibers were stretched along the direction of the arrow, they are oriented and maintain good shape memory performance after several shape memory cycles. Zhang et al. [92] studied the self-reversible programmable pore size change of electrospun fiber webs, and changed the pore size by stretching the fiber. Microfibers prepared without external stress. When the temperature changed, the pore size changed reversibly with temperature (Fig. 5b), from 10.5 ± 0.5 μm at 60 °C to 11.8 ± 0.6 μm at 10 °C. Figure 5c shows the deformation behavior of the sample during heating and cooling process.

In addition to the above function of SMPFs, the actuation methods of SMPFs are also becoming more and more diversified, including heat, magnetic, water, electric, and light. At the same time, some polymers have excellent properties, but their shape memory ability is limited. By blending with other materials, the shape memory properties and other properties of SMPs can be improved. Based on the traditional thermal actuation, the new actuation methods make the SMPs universal and remote controllable. Although many researchers have conduced in-depth research on the new actuation method, in order to meet the requirements of SMPs in a wide range of practical applications, to a certain extent, SMPs still need to be improved.

PLA has been widely concerned because of its high tensile strength and elastic modulus, good biocompatibility and non-toxic degradation. However, its T_g of 60 °C is much higher than that of human body, which limits its application in many aspects. Adrian Leonésa et al. [93] added lactic acid oligomer (OLA) as plasticizer to PLA-based electrospun fiber to reduce its T_g and broaden its application in the biomedical science. The results show that the PLA electospun fiber with OLA has better toughness, lower T_g and closer to the Young's modulus of human tissue, which can be better applied in tissue engineering, wound healing and other fields.

Chen et al. [94] processed one-dimensional (1D) gelatin/polylactic-glycolic acid (PLGA) electrospun fibers into ink suitable for 3D printing, and then combined with 3D printing, freeze-drying and cross-linking technology, successfully prepared water-driven 3D printing scaffolds (3DP) with precise shape control and large pore size. Compared with pure gelatin scaffold, the scaffold had better mechanical properties, water absorption and shape recovery performance. The square scaffold was freeze-dried and fixed into tubular scaffold, which was restored to its original square shape within 20 s after absorbing water. The rectangular scaffold was folded and fixed into a wave shape in freeze-dried state, and was restored to its original state after immersed in water for 30 s (Fig. 6a).

Magnetic actuation is to add magnetic particles into the fiber to generate heat in the magnetic field, which triggers the recovery of the fiber. By adding Fe_3O_4 magnetic particles into SMP matrix, electrospun SMPFs can be driven remotely by magnetic field, avoiding the damage of normal tissues around the lesion by direct thermal driven method. In addition, the long distance actuation makes the fibers triggered in the body more intelligent and convenient. Gong et al. [80] fabricated Fe_3O_4@CD-M oriented nanofibers with shape memory properties by coaxial electrospinning. The shape memory properties of nanofibers were tested by magnetic and hot water actuation. The R_r was 92% when T_g was 45 °C. The fiber membrane can expand under both conditions, but the expansion speed in hot water was much higher than that in magnetic field.

Zhang et al. [83] prepared Nafion/Fe_3O_4 composite fiber membranes by electrospinning. The R_f and R_r of the three mechanical cycles tests were all above 90%. Figure 6b shows a magnetic field induced shape recovery process. For the recovery of samples with different shapes, the surface temperature was always lower than 40 °C. Therefore, the composite fiber membrane has potential applications in biomedical field.

In electric field induced recovery process, conductive polymers can be used to form ideal manufacturing materials. Zhang et al. [95] investigated a conductive shape memory microfiber membrane with core–shell structure. The core was prepared by electrospinning different concentrations of PLA, and the shell was prepared by depositing conductive PPy on electrospun microfibers by chemical vapor polymerization. The shape recovery speed of conductive microfiber film was very fast. Figure 6c shows the shape recovery process within 2 s. By applying different voltages to observe the shape recovery behavior of the conductive fibers, it can be found that the maximum limit voltage was 40 V.

Applications of Shape Memory Polymer Fibers

SMPFs have the advantages of diverse structures, large specific surface area, and adjustable porosity, which can be widely used in various fields [6], especially in biomedical field [96–98]. At present, SMPFs have been successfully applied in drug release, cell culture and tissue scaffolds. The structural characteristics of SMPFs make it suitable for the

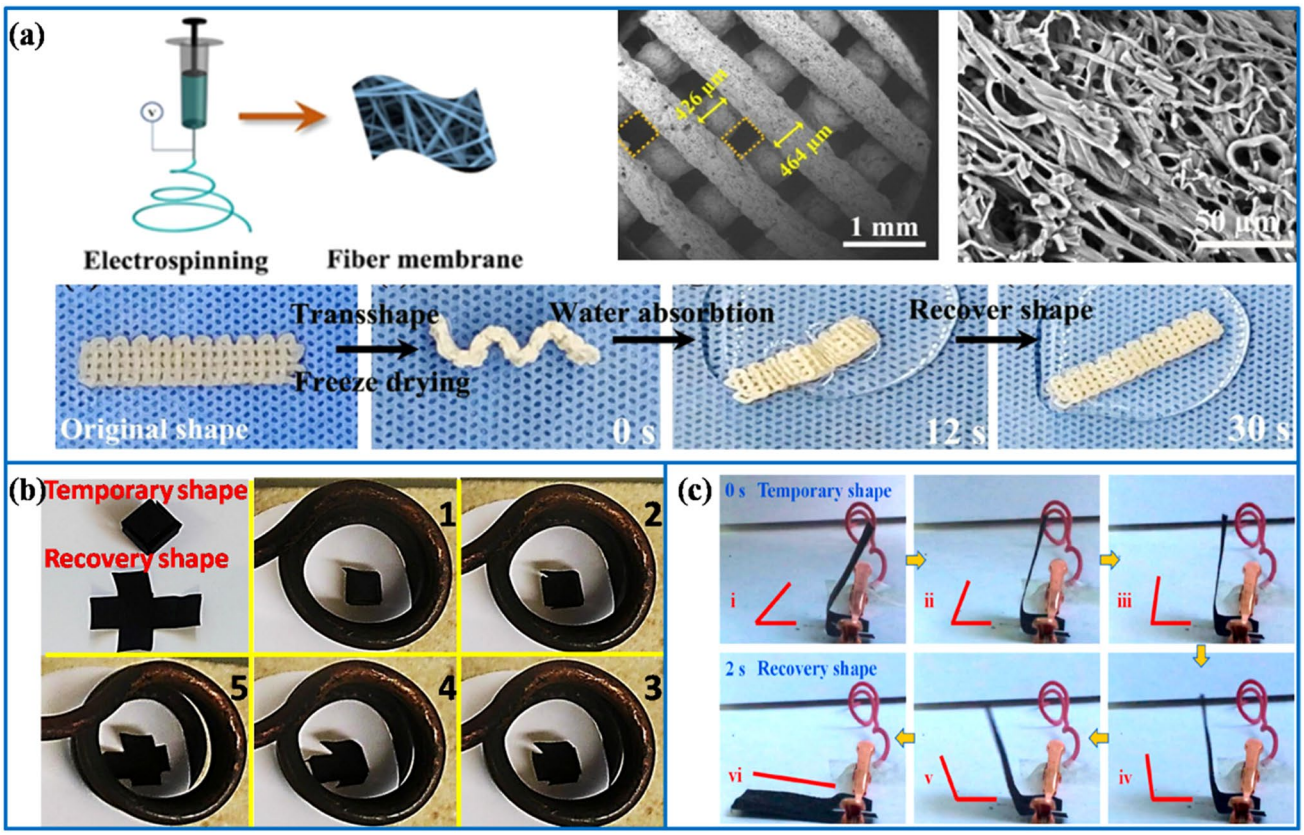


Fig. 6 Shape recovery process of SMPFs exposed upon different stimuli conditions. **a** Water [94], **b** magnetic field [83], and **c** electrical field [95]

delivery and release of different drugs. It can simulate the extracellular matrix used in tissue engineering and provide more opportunities for fixing functional groups in medical diagnosis and other applications [99, 100]. Furthermore, the research on SMPFs in other areas has also attracted wide attention, including energy harvesting and stress sensors.

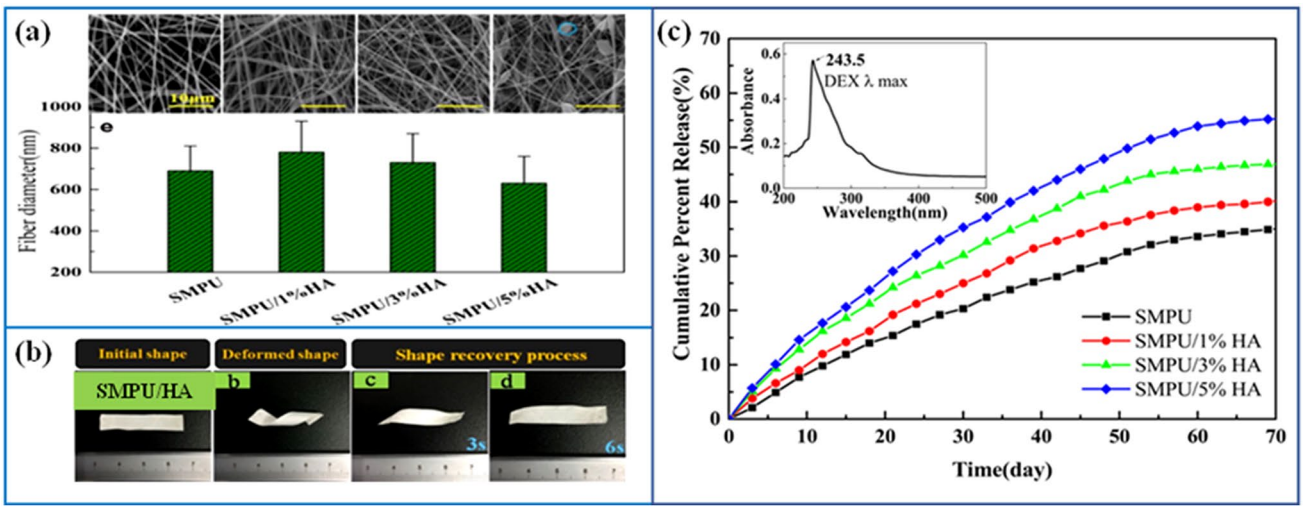


Fig. 7 Application of SMPFs in drug release [101]: **a** microscopic characterization of electrospun fibers, **b** macroscopic deformation of electrospun fibers: optical image of the shape recovery process of SMPU/3% HA composite fiber, **c** DEX cumulative release curve of SMPU fibers and SMPU/HA composite fibers

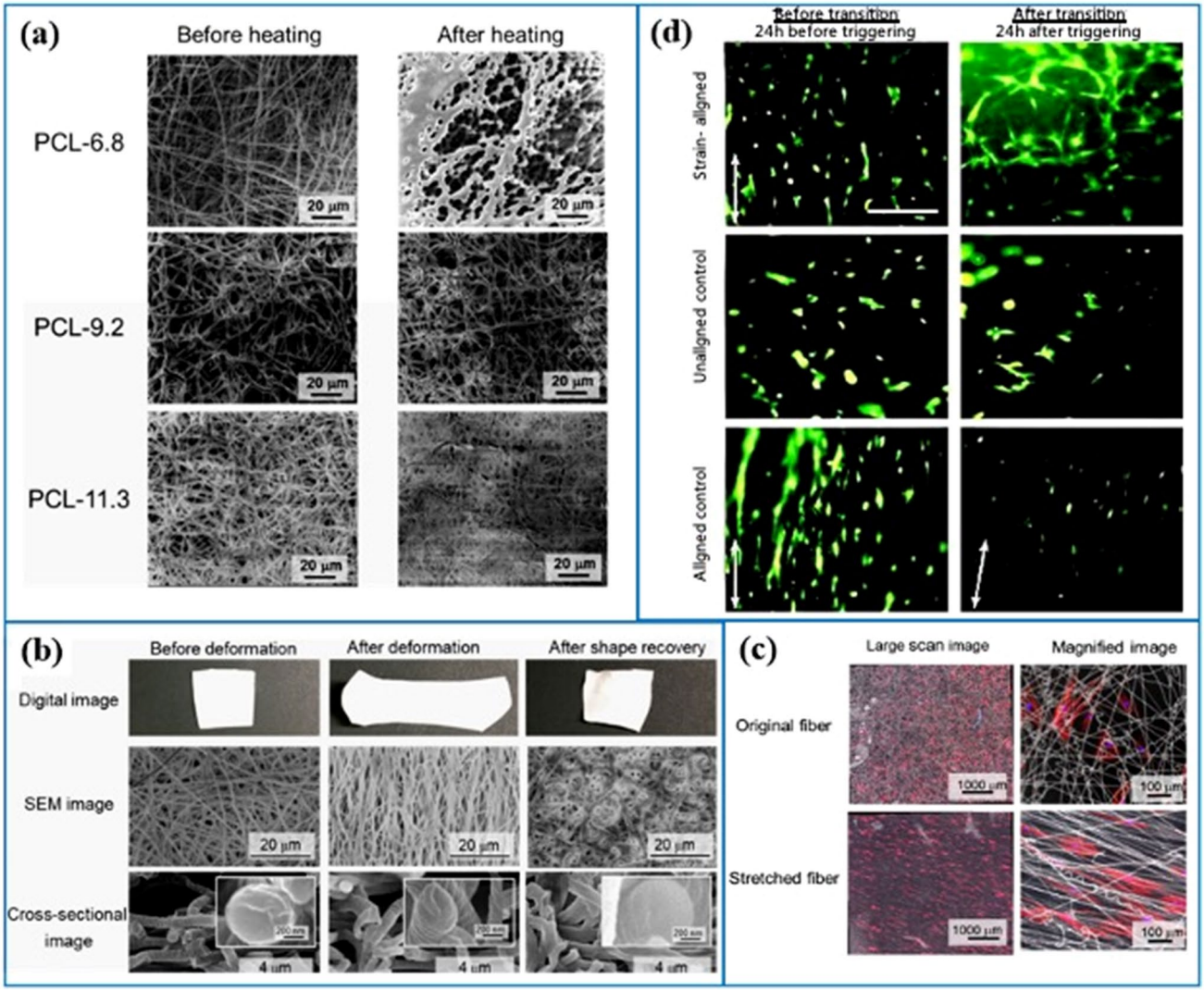


Fig. 8 Application of SMPFs in cell culture: **a** SEM images of electrospun fibers before and after heating at 60 °C [102], **b** digital, SEM and cross-linked images of nanofibers before and after deformation and recovery process [102], **c** adhesion morphology and arrangement of human bone marrow mesenchymal stem cells [102], **d** before and after the shape and structure of the scaffold is changed, the cell viability on the scaffold with active strain arrangement is still very high [31]

Drug Release

The structure of SMPFs has obvious advantages in drug release. It can not only adapt to different physical and chemical properties of drugs, but also has high drug loading. SMPU has attracted much attention because of its excellent properties, large deformation and low cost. SMPU has high elasticity, and its modulus and biocompatibility need to be improved. Hydroxyapatite (HA) nanocrystals are the main component of bone. It has excellent biological activity, compatibility and conductivity. The addition of HA can improve the mechanical properties and SME of the material. Lv et al. [101] prepared a new type of SMPU/HA bionic composite fiber through electrospinning. The fiber has excellent biological properties and shape memory properties, and has a good application prospect in biomedical field. When the HA content was 1%, the fiber diameter increased from 690 ± 120 to 780 ± 150 nm, but with increase of HA content, the fiber diameter began to decrease. There are tiny particles on the surface of SMPU/3%HA composite fiber. When the HA content increased to 5%, the particles number increased (Fig. 7a). In addition, the incorporation of HA has a positive effect on the shape memory function of the composite fiber. The initial shapes of SMPU fiber and SMPU/3%HA composite fiber were all strips and they were deformed and fixed at 40 °C. SMPU/3% HA composite fiber can return to the original strip shape within 6 s, while SMPU fiber cannot fully recover within 6 s (Fig. 7b). The drug release behavior

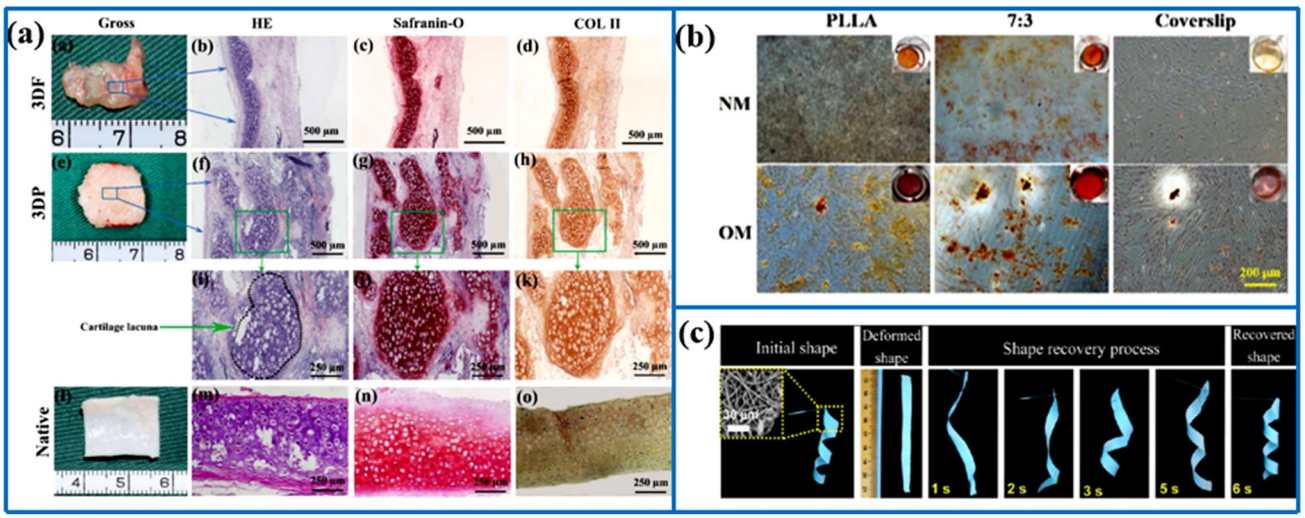


Fig. 9 Application of SMPFs in bone tissue scaffold: **a** in vivo cartilage regeneration process [94], **b** ARS stained image is used to express the bone formation of BMSCs cultured on different scaffolds/matrices for 14 days under osteoinductive and non-osteoinductive conditions [104], **c** macroscopic view of the shape recovery process of spiral PLMC film [73]

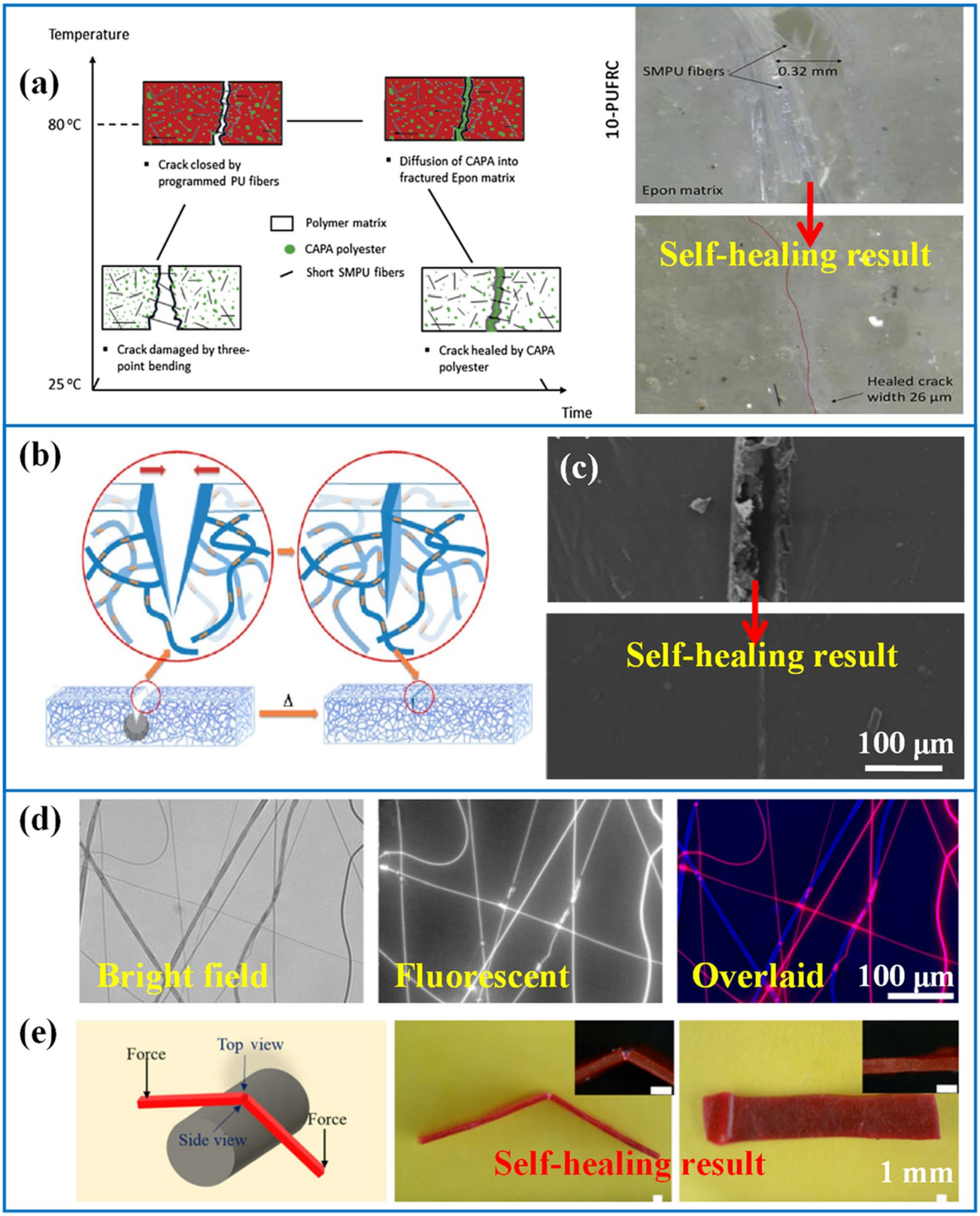
of SMPU/HA composite fibers was studied with DEX as drug model. The release amount increased with the increase of HA content, which was consistent with the change trend of degradation rate, as shown in Fig. 7c.

Cell Culture

Most of the traditional bionic scaffolds are static structures, which cannot accurately simulate the dynamic microenvironment of living organisms. This is one of the challenges of biomimetic fiber scaffolds. However, some studies have shown that cell culture on fiber scaffolds can change its morphology and better simulate the dynamic microenvironment. Niiyama et al. [102] prepared a series of poly(ε-caprolactone) (PCL)-based PU nanofiber nets with different ratios of soft and hard segments through electrospinning. SEM images before and after heating showed that the three samples were uniformly distributed in the form of fibers before heating (Fig. 8a). Figure 8b shows the deformation behavior of PCL-9.2 nanofibers. The structure of fibers was different before and after deformation. Even if the deformation reached 400%, the recovery rate was as high as 89%. These shape memory nanofiber networks have similar environment to extracellular matrix and can provide a platform for cell culture. Human bone marrow mesenchymal stem cells were respectively inoculated on PCL-9.2 nanofibers before and after stretching (300%). The cells on the unstretched fibrous network were widely distributed and randomly oriented, and the cells on the stretched fibrous network elongated along the fiber axis (Fig. 8c). Then the cells were cultured on the nanofibers before deformation and after shape recovery. The diffusion of cells is limited by the geometry, so the directional culture of cells can be realized. Two days later, the cells were still alive, and the cell survival rate was similar to that of the control group.

Tseng et al. [32] prepared thermal sensitive PU biomimetic fiber scaffolds with good shape memory properties ($R_r > 94$ to 96%, $R_f > 90$ to 100%). The effects of deformation on the cell structure and nucleus of human adipose stem cells (hASCs) were studied by changing the macro and microstructure of fibers. It was found that after the cells were seeded on the stretched fibers, the active filaments and nuclei arranged along the fiber strain direction. After shape recovery and activation, the cells still attached to the surface of fiber scaffold, and the activity was more than 86%. However, the cells were no longer orderly arranged and randomly arranged into random fibers after recovery, as shown in Fig. 8d. The cell morphology can be controlled by changing the arrangement of fiber scaffold.

In a recent study, Pandinietal et al. [103] used PCL and hydrogel to prepare directional and unidirectional SMPF membranes, and tested their cytotoxicity, adhesiveness and proliferation. The strain fixation rate and recovery rate are more than 80% and 99%, respectively. In vitro cell experiments proved that the original fiber, high-PCL and low-PCL fiber membranes were not cytotoxic. High-PCL fiber membrane had a good ability of cell adhesion and proliferation, which may be due to its greater stiffness than low-PCL fiber membrane, indicating that the stiffness of scaffold had an important influence on cell adhesion, proliferation and extracellular matrix synthesis.



Bone Tissue Scaffolds

Biological scaffolds play an important role in tissue engineering. Its structure and chemical composition need to be similar to extracellular matrix. The unique three-dimensional structure of SMPFs is conducive to cell migration,

◂**Fig. 10** Self-healing of SMPFs: **a** schematic of the two-step healing process and crack narrowing due to SME of the short programmed SMPU fibers [107, 108], **b** schematic illustration of the coating morphology and the shape memory assisted self-healing(SMASH) concept [9], **c** the surface of the coating was heated at 80 °C [110], **d** representative optical micrographs showing: bright field, fluorescent and overlaid images of the dual-electrospun PVAc60:PCL40 fibers. Blue and pink colors correspond to PCL and PVAc fibers, respectively [86]. **e** Schematic illustration of the crack opening method and photographs of the damaged and self-healed samples, respectively [86]

adhesion and proliferation. The size of fiber pore size is conducive to the transport of nutrients and the regeneration of blood vessels in tissue repair. Therefore, it has great potential in applications requiring active structures and functional components. Chen et al. [94] successfully fabricated gelatin/PLGA electrospun fiber scaffolds with controllable pore size by combining 3D printing, freeze-drying, and cross-linking techniques. The scaffold can be well used for chondrocyte regeneration. Cartilage-like tissue cells were formed in both groups after 6 weeks of culture. However, the shape of 3DF group was irregular and the tissue was thin, while 3DP group retained the original regular squares and thick tissues. Consistent with the above results of in vitro culture (Fig. 9a), irregular shape and very thin tissue appeared in 3DF group after 8 weeks of stent implanting. Histological examination revealed that cartilage-like tissue was only found in the surface area of the specimen. In 3DP group, the tissue was white, similar to the natural cartilage tissue, and retained the original regular squares, and the tissue was very thick. Histological examination showed that the formation of cartilage-like tissue and matrix deposition can be observed in the whole sample area, which was also similar to that of natural cartilage. In short, 3DP fiber scaffolds can achieve better cartilage regeneration in vivo.

Wang et al. [104] incorporated poly(3-hydroxybutyrate-3-hydroxyvalerate) (PHBV) into poly(L-lactide) (PLLA) and spun PLLA-PHBV blend into superfine fibers by electrospinning and evaluated the mechanical, shape memory and osteogenic properties with mouse bone marrow mesenchymal stem cells (BMSCs). Figure 9b shows the ARS staining results of BMSCs cultured on different fiber scaffolds and substrates for 14 days. BMSCs cultured on PLLA-PHBV (7:3) composite fiber scaffolds were bright red. Therefore, PLLA-PHBV (7:3) composite fiber had excellent ability to induce BMSCs to differentiate into bone.

Bao et al. [74] explored the potential application of shape memory poly(D, L-lactide-co-trimethylene carbonate PDLLA-*co*-TMC, PLMC) fiber in bone screw hole filling or bone tissue repair membrane. The study found that by changing the ratio of PDLLA:TMC, the diameter of PLMC fiber decreased from 1500 to 680 nm, and T_g decreased from 44.2 to 19.2 °C. The R_r was above 94%, and the R_f was more than 98%. When the temporarily elongated fiber was immersed in hot water at 39 °C, the original spiral fiber can be restored in only 6 s (Fig. 9c), indicating that PLMC biomimetic fiber scaffold had great application potential in repairing bone screw hole or guiding bone regeneration barrier membrane.

Self-Healing

The unique SME of SMPs is an approach to realize self-healing. It can not only repair microscopic cracks, but also fill macroscopic cracks [100]. For example, embedding SMPFs into traditional thermosetting engineering plastics can make raw materials have self-heating function, which is of great significance to improve performance [105, 106]. Li et al. [107, 108] developed a stimulus-responsive SMPF/thermoplastic particle/epoxy composite. The self-healing system made the network formed by SMPFs as the skeleton and thermoplastic resin particles as the healing agent. The healing process was divided into two steps, as shown in Fig. 10a. When the material was damaged, heat it. When the heating temperature exceeded the T_g of SMPFs, due to the SME, the pre-stretched fiber contracted, resulting in shrinkage stress and pulling the matrix material to close the macroscopic crack. After that, the molten thermoplastic resin particles flowed to complete the crack filling and repair. Moreover, Zhang et al. [109] further introduced SMPFs into foam and other structures to achieve the self-healing function of fiber reinforced composites.

In addition, Mather group [9, 86] developed another self-healing material system, mainly using electrospun fibers as repair layers to form SMP composites. The self-healing mechanism (Fig. 10b) and repair effect (Fig. 10c) were investigated. Besides, Nejad et al. [110] further developed a dual electrospinning system (Fig. 10c–e), and simultaneously spun PVA and PCL to obtain composite fibers with mixed non-woven structure. This material system not only had good self-healing performance, but also had good shape memory performance, which expanded a new idea for the application of SMPFs. Furthermore, Yao et al. [111] prepared PCL fiber by electrospinning and compounded it with epoxy resin. The composites not only had good shape memory properties, but also had specific self-healing function.

Other Applications

SMPF is a kind of smart polymer, which can be applied in many fields according to its functionality. The fiber itself has excellent structural characteristics such as large specific surface area and volume, adjustable surface morphology, and diverse structure. Therefore, the application of SMPFs in energy collection, stress sensor, and smart textiles has also been widely concerned.

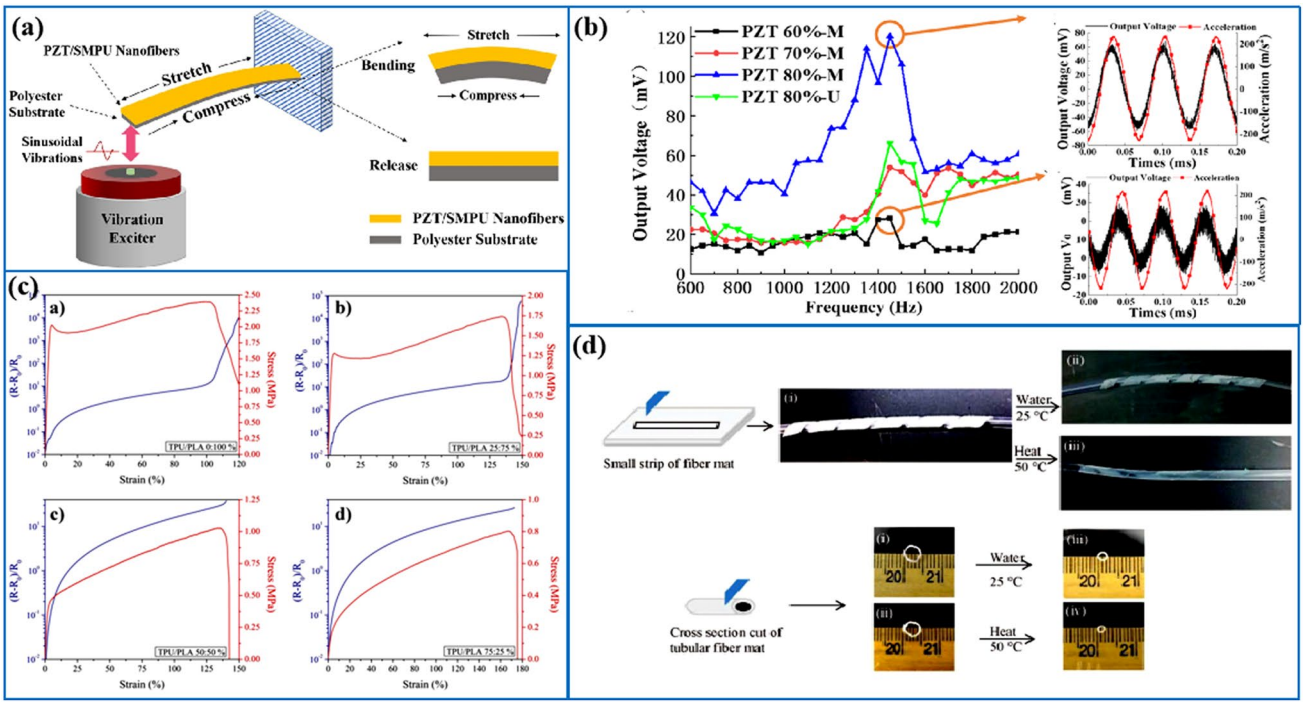


Fig. 11 Application of SMPFs: **a** schematic diagram of energy harvesting device [112], **b** PZT/SMPU nanofiber energy harvesting performance: PZT/SMPU nanofiber output voltage at a fixed acceleration of 233 m/s^2 and different frequencies, PZT 60%-M and PZT 80%-M output voltage at 1750 Hz frequency [112], **c** pressure resistance and tensile test response: **a** PU/PLA 0:100 wt%, **b** PU/PLA25:75 wt%, **c** PU/PLA 50:50 wt%, and **d** PU/PLA 75:25 wt% [113], **d** shrinkage of shape memory polyvinyl acetate fiber membranes with indocyanine green added [116]

Guan et al. [112] added zirconate titanate (PZT) particles into SMPU to prepare multifunctional composite nanofibers by electrospinning. Nanofibers had both piezoelectric effect and SME, and possessed potential applications in the field of energy collection. As shown in Fig. 11a, the modified nanofibers had better shape fixation and recovery ability, and the shape recovery process can be completed in 4 s, with a recovery rate of more than 94%. Through the study of energy harvesting performance of PZT/SMPU nanofibers, it was found that the collector can generate continuous sinusoidal voltage and had repeatability. Then, the output voltage decreased as the frequency increased (Fig. 11b). When the frequency was 1450 Hz, the output voltage of PZT/SMPU nanofibers increased with the increase of acceleration, and the increase of PZT 80%-M was the largest.

In addition, SMPFs can also be used as strain sensors. Compared with traditional metal strain sensors, electrospun nanocomposite fibers have higher sensitivity and multidirectional sensing ability, and lighter structure to allow greater deformation. Khalili et al. [113] used electrospinning technology to mix PLA and thermoplastic PU to make nanofiber mesh, and then sprayed single-arm carbon nanotubes on the electrospun fiber mesh to make a strain sensor, and nanotubes were sensing elements. Under the action of high strain (50–100%), the resistance change of the fiber increased sharply, and the mutation occurred at the beginning of fiber fracture, indicating that the resistance and strain were normalized responses (Fig. 11c). Due to the shape memory property of electrospun fiber, the extensibility of strain sensor can be improved, and the reversible stretching can be realized at the maximum stretching.

Moreover, SMPFs also show excellent characteristics in smart textiles [114]. Hu et al. [9] prepared SMPU fibers by electrospinning. In addition to good shape memory property, this SMPF also had good mechanical properties and elasticity, which can be used to prepare various shape memory textiles. Smart clothing is one of the applications of SMPs. It uses SMPFs to prepare deformable clothes, and realize the shape change process by changing the temperature [115].

Furthermore, in order to promote the development of SMPFs, researchers have improved the preparation methods of SMPFs. Torbati et al. [116] systematically studied the luminescent shape memory polyvinyl acetate fiber membrane prepared by electrospinning, and added in docyanine green into the spinning solution. As shown in Fig. 11d, this material can be immersed in water at 25 °C or heated to

Table 1 Materials, structures, driving methods and applications of SMPFs

Material	Structure	Driving method	Application	Research group
PU	Non-woven	Heat	–	Cha et al. [28]
PU	Non-woven	Heat	Cell culturing	Tseng et al. [32]
PCL/PDMS	Non-woven	Heat	Tissue engineering	Dan et al. [76]
PU/PCL	Non-woven	Heat	–	Zhuo et al. [75]
PU/PCL	Core–shell	Heat	Antibacterial	Hu et al. [77]
PU/MWNTs	Core–shell	Heat	–	Cho et al. [78]
PCL/epoxy	Core–shell	Heat	Biological scaffold	Zhang et al. [62]
PDC/PEG	Hollow	Heat	Biological scaffold	Zhang et al. [79]
Nafion/Fe_3O_4	Particle-filled	Magnetic	Tissue engineering	Zhang et al. [83]
PU/graphene	Particle-filled	Heat	–	Tan et al. [82]
PCL/MWNTs	Oriented	Heat	Cell culturing	Gong et al. [80]
PCL	Oriented	Heat	Biological scaffold	Schneider et al. [31]
PU	Oriented	Heat	Actuator	Ji et al. [84]
PVAc/PDMS	Oriented	Heat	Biological scaffold	Mather et al. [85]
PVAc/PCL	Oriented	Heat	Food packaging	Mather et al. [86]
PLA/PCL	Oriented	Heat	Biological scaffold	Mather et al. [87]
PLGA	–	Water	Biological scaffold	Chen et al. [94]
PLA/PPy	Core–shell	Electric	–	Zhang et al. [95]
PU/PCL	Non-woven	Heat	Drug release	Lvet al. [101]
PU/PCL	Non-woven	Heat	Cell orientation	Niiyama et al. [102]
PLLA	Non-woven	Heat	Biological scaffold	Wang et al. [104]
TP/epoxy	Non-woven	Heat	Self-healing	Li et al. [107, 108]
PVA/PCL	Non-woven	Heat	Self-healing	Mather et al. [110]
PU/PZT	Particle-filled	Heat	Energy harvesting	Guan et al. [112]
PLA/TPU	Non-woven	Heat	Strain sensor	Khalili et al. [113]
PU	Non-woven	Heat	Smart textiles	Hu et al. [9]
PVAc/DMF	Non-woven	Heat/water	Medical device	Torbati et al. [116]

50 °C to achieve shrinkage, which provides potential applications for medical devices (gastro esophagus and catheters). In summary, we list the materials, structures, actuation methods and applications of SMPFs in recent years, as shown in Table 1.

Summary and Perspectives

SMP has many advantages, such as light weight, easy processing, controllable deformation temperature, good biocompatibility and biodegradability, and so on. Among them, SMPF has high surface area to volume ratio, high interconnected pore size, adjustable surface morphology and other significant characteristics, which is similar to the three-dimensional structure of extracellular matrix. It can be designed as a scaffold for cell proliferation and differentiation in tissue repair engineering. SMPFs can also be used in other fields. However, there are still some challenges in the application of SMFs and their composites. In order to meet the needs of practical application, it is necessary to further develop smart fiber structures. For tissue engineering, different cells may need different scaffolds for better growth and differentiation. The most important thing is to accurately control the size of the fiber, and the most ideal is to control the fiber in the nano size, so as to more accurately simulate the natural extracellular matrix. In addition, the types of polymers that can be made in fibers are still very limited. In the process of tissue culture, the mechanical properties of fibers and the hydrophilic and hydrophobic properties of fiber surface are highly required. It is expected to develop more materials suitable for fiber forming in various fields. Moreover, the microstructure of SMPFs is very important for cell differentiation, but the structure of fibers is very random, which is difficult to compare with the natural extracellular matrix structure. The emerging 4D printing technology brings hope for this problem. Through the combination of electrospinning and 4D printing, the bionic fiber scaffolds with different fiber sizes and orientations can be customized. Furthermore, in addition to electrospinning technology, other fiber manufacturing techniques can also be used to prepare fibers to break through the limitations of electrospinning [117, 118], such as thermal drawing technique [119, 120]. The diversity of manufacturing methods can produce

fibers composed of a variety of materials with different properties [121, 122]. At the same time, it can make fibers complex structures [123]. With the deepening of research work, it is believed that the SME and biological characteristics of SMPs will continue to improve, and its breadth and depth of application in various fields, especially in biomedicine, will continue to expand, so as to realize personalized and intelligent precision medicine.

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Declarations

Conflict of interest There are no conflicts to declare.

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