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The research status and challenges of shape memory polymer-based flexible electronics

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Shape memory polymer-based flexible electronic devices (SMPFEDs) that incorporate novel features and functions of shape memory polymers (SMPs) and their composites (SMPCs) represent a rapid development in flexible electronics. The shape memory effect and variable modulus of SMPs could endow more functions with flexible electronic devices and thus enable some novel applications. This paper summarizes state-of-the-art of SMPFEDs. Some examples of SMPFEDs, like shape memory polymer light-emitting diodes, shape memory polymer thin film transistors and optical devices, are introduced. The printing electronic technology and 4D printing are discussed as potential fabrication technologies for SMPFEDs. Furthermore, the actuation methods for SMPFEDs, including chemo-, electro-, magnetic-, and light- actuation methods, are detailed. Finally, the technical challenges and development directions of SMPFEDs are proposed.

1. Introduction

In the past decades, flexible electronics has been developed and attracted tremendous attention from various fields. Different from the traditional electronic devices, flexible electronic devices are fabricated on the flexible or soft substrates, rather than brittle silicon materials or printed circuit board, to achieve flexibility or even stretchability, resulting in potential applications of such devices in medical devices, soft robotics, wearable electronics, photonic devices, etc. 1-11. In the beginning stage of the development of flexible electronic devices, huge challenges lied in the realization of flexibility and the maintenance of stable electrical performance after deformation. Some research utilized transparent and conductive ceramic indium tin oxides (ITO) as electrodes in flexible electronic devices after laminated with plastic substrates 12-15. However, the functional properties and electrical performances of ITO based devices could be deteriorated after large deformation or prolonged usage since ITO is intrinsically a brittle electrode material 16-19. Thus, alternative electrode materials of ITO like graphene 20-22, carbon nanotubes 23-25, metal grids 26-28, random metal nanowires 22, 29, 30, and conducting polymers 31-33 have been used, and flexible solar cells, light-emitting diodes and thin film transistors based on these electrodes have been demonstrated 32, 34-37. Besides development of novel electrode materials, another effective approach is the design of deformable conductive structures, like wavy structures, island-bridge structures with straight, serpentine, or coiled interconnects, to achieve flexibility, bendability or stretchability while maintaining good electrical performances 18, 19, 38-40.

After solving the technical problems in realizing flexibility, more functions are expected for flexible electronic devices. Some researchers have turned their sights on SMPs since SMPs could not only satisfy the material requirements of flexible electronics, but also have some special characters that traditional polymers do not possess, like shape memory effect and variable modulus. SMPs are smart stimuli-responsive polymeric materials that can memorize permanent shapes and recovery from temporary shapes to permanent shapes under external stimuli 41-49. The shape memory performance of SMP looks similar to that of shape memory alloys (SMAs) 50, 51. But different from the reversible martensitic transformation mechanism of SMAs, the shape-memory effect in SMPs stems from a dual-segment system: stable netpoints to determine the permanent shape and switching segments to enable fixation of the temporary shape 50, 51. The netpoints can be made of molecule entanglement 52, crystalline phase 53, chemical cross-linked system 54, interpenetrated network 55, or cyclodextrin (CD) polymer inclusion 56, 57. The switching segments are including reversible crystallization/melting transition 58, vitrification/glass transition 59, liquid crystal anisotropic/isotropic transition 47, 60, reversible molecule cross-linking transition 46, 61, 62, or supramolecular association/disassociation transition 63. The modulus of SMP drops dramatically by two or three orders of magnitude around transition temperatures, so that the desired temporary shape could be easily programmed at higher temperature and then fixed after cooling down 64. Compared with SMA, SMPs possess many advantages in some practical applications. SMP is lighter, more flexible and have larger recoverable strain (up to 400%)

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than SMA 65, 66. There are more fabrication technologies for SMPs and SMPCs, similar to that of traditional polymers, like compression molding, injection molding, extrusion molding and 3D printing 67. Thus the cost of SMPs is cheaper. For example, the price of the SMPs (~$10 per 1b) is much cheaper than that of shape memory nickel-titanium (NiTi) alloy (~$250 per 1b) 68 but comparable with that of traditional polymers, like PET (~$3 per 1b) 18. Due to the special properties and some technical advantages, SMPs have potential applications in medical devices 43, 69, textiles 70, aerospace 71, building materials 72, robotics 73, daily life 74, electronics 75 and information 76, etc., as illustrated in Fig. 1. The details could be found in some other reviews 64, 66.

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This review focuses on some issues in the application of SMP on flexible electronics, including materials, fabrication technologies and actuation methods. In the first part of this review, we will introduce advances in the SMPFEDs including shape memory polymer light-emitting diodes (SMPLEDs), shape memory polymer thin film transistors (SMPTFTs) and optical devices. The second part discusses some available fabrication techniques for SMPFEDs, especially the 4D printing. Then the potential actuation methods of SMPFEDs are also described. Finally, the challenges and further developments of SMP-based flexible electronics are described.

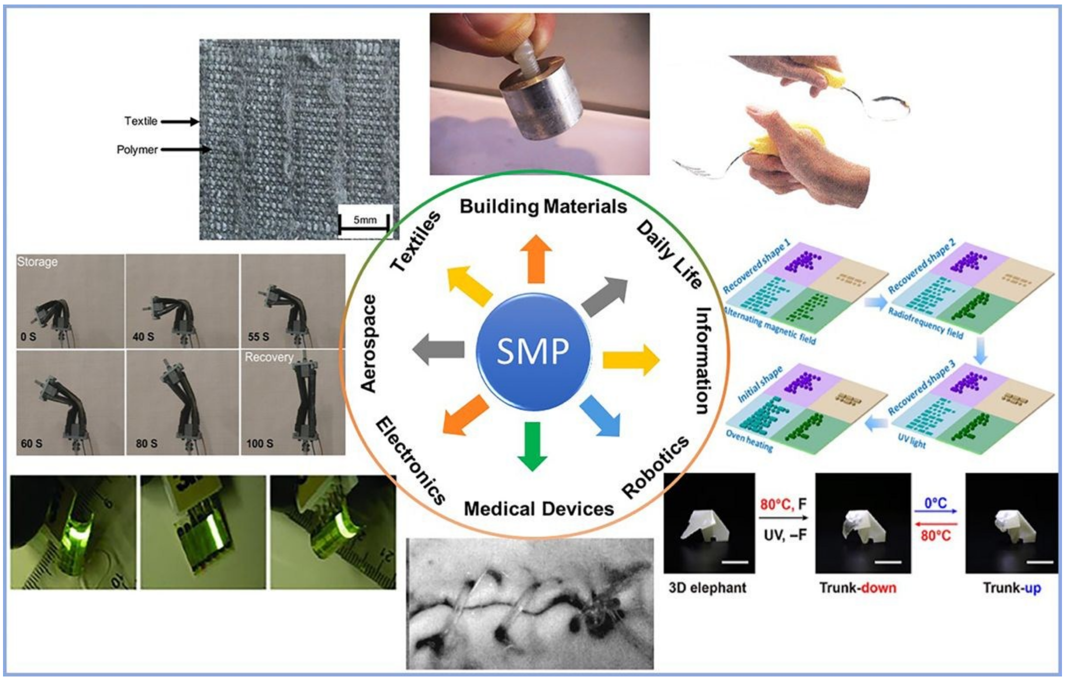

Fig. 1 The applications of SMP in the areas of medical devices 43, textiles 70, aerospace 71, building materials 72, robotics 73, daily life 74, electronics 75 and information 76. Reproduced from ref. 43 and 73 with the permission of The American Association for the Advancement of Science. Reproduced from ref. 70 with the permission of Springer Nature. Reproduced from ref. 71 and 74 with the permission from IOP Publishing. Reproduced from ref. 72 with the permission of Elsevier. Reproduced from ref. 75 with the permission of John Wiley and Sons. Reproduced from ref. 76 with the permission of American Chemical Society.

2. The state-of-the-art of SMP-based flexible electronics and their applications

Several shape memory polymer flexible electronic devices, including shape memory polymer light-emitting diodes (SMPLEDs) and shape memory polymer thin film transistors (SMPTFTs), have been fabricated using SMP as substrate or encapsulation layer. The transparent SMPs, like thiol-ene/acrylate SMP (T_r (recovery temperature): 70 °C) 77, crosslinked shape memory polyacrylate (T_r: 120 °C) 75 and shape memory poly(tert-butylacrylate) (PtBA, T_r: 70 °C) 78 have been used as the substrates in some LED devices. Such SMPLEDs could maintain stable electroluminescent properties under large deformation or after multiple bending-recovery cycles and have the potential in applications of wearable displays. For example, a metal-free SMPLEDs fabricated on PtBA SMP can withstand 45% strain in response to body movements. The electroluminescent properties at different strain are shown in Fig. 2A 78. The shape memory film (T_r: 150 °C) from NIPPON MEKTRON Ltd. 79 is a commercially available substrate for SMPFEDs and was utilized in the first reported SMPTFT. This SMPTFT could be formed into a helix shape at high temperature and be used to measure the spatial distribution of pressure inside long, narrow tubes 79.

Another commonly used substrate for SMPTFTs is thiol-ene/acrylate SMP with a T_r of 70 °C 11, 80-86. However, when put into physiological environment, the solvent-induced plasticization would dramatically reduce its T_r to 37 °C, which is close to the body temperature. The plasticization also lowers the modulus of this material to reduce the mismatch between devices and the biotissues, as can be seen from the modulus change in Fig. 2B 11, 84, 87. This particular property makes this material extremely suitable for implantable biomedical devices. The devices based on this material could be implanted in a relatively rigid state and then the softening could make the devices comfort to the tissue and improve the interface as well 11, 87-89. Fig. 2C illustrates the configuration change of a SMPTFT with thiol-ene/acrylate SMP substrate before being implanted into a rat and after being taken out after 24 h implantation 11. The thiol-ene/acrylate SMP has also been used to fabricate the adaptable nerve cuffs 90, 91 and cortical probes 87, 92, 93, 94, 95 that demonstrated the capabilities of recording and monitoring of some biosignals, including temperature, blood pressure, neural activity, etc.. In addition, the thiol-ene/acrylate SMP has a low water absorption in the physiological environments (2-3% volume increase after implantation for 77 days) 11, 87, 92, 93, which ensures the prolonged signal monitoring of the thiol-ene/acrylate SMP based devices in biomedical applications 11, 82, 94, 96. Besides, researches concerned with the fabrication and performance improvements of SMPTFTs for biomedical applications have been conducted, such as approaches to realize solution deposition on thiol-ene/acrylate SMP film 83, 85 and modification of the structure and functional layers of the SMPTFTs 81, 82, 84, 86.

In the aforementioned devices, SMPs serve as the substrates or encapsulation layers. However, a single SMP film could realize some particular functions, like directed water shedding 96-100, controllable wettability 101, information and energy storage 102, cell manipulation 103-105, when patterned with pillar arrays, microprism arrays, microlens arrays, gratings or holograms 106-109 by hot embossing, nanoimprinting, transfer printing or photolithography. Such approaches are also effective in fabricating tunable optical devices for optoelectronics 110-112. For example, the shape memory crosslinked poly(ethylene-co-vinyl acetate) (cEVA SMP) film with micro-prism arrays could be transmitted from transparency to opacity 110. As shown in Fig. 2D, the optical property of each area of the film could be selectively tuned by the attached 3×3 transparent conductive ITO glass array. More complex patterns would enable more functions like the beam power splitters fabricated by Li et al. 111 that could realize the switch between different diffraction patterns through the transition of different micro-patterns upon heating, as illustrated in Fig. 2E.

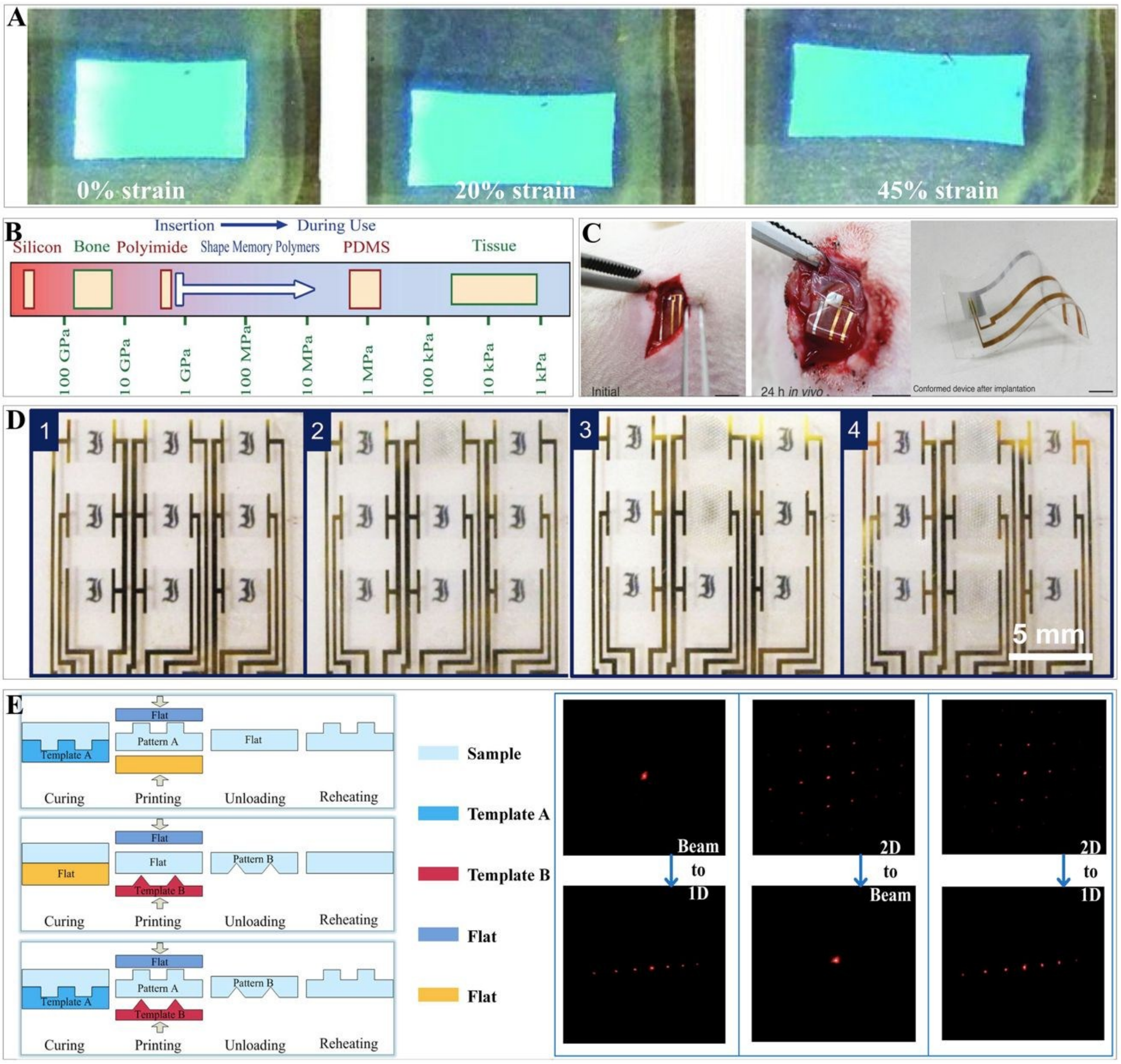

Fig. 2 (A) Electroluminescent performances of the PLED at 8 V with different strains 78. (B) Schematic indicating the modulus change of thiol-ene/acrylate SMP before and after implantation 87. (C) Configuration change of a SMPTFT before and after implantation into a rat 11. (D) Micro-optic devices with selectively tunable transparency 110. (E) Different micropatterns in beam power splitters and corresponding variations in the diffraction pattern distributions 111. Reproduced from ref. 11, 78, 87, and 110 with the permission of John Wiley and Sons. Reproduced from ref. 111 with the permission of Springer Nature.

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3. Fabrication technologies for SMP-based flexible electronics

Optical lithography, shadow mask, transfer printing and ink-jet printing technologies have been well developed and designed for patterning of microelectronic devices. Although optical lithography having high resolution that is beneficial for patterning small size devices, such technologies are not suitable for patterning large-area flexible electronics 113. It is because that the preparation process is complicated, high-cost, time-consuming, materials wasting, and also requires complex steps to wipe out resists, solvents and developers, which result in poor compatibility with plastic substrates 113, 114. More discussions about optical lithography technology for flexible electronics can be found in some published articles 115, 116. Compared with the optical lithography patterning method, the shadow mask technology can be regarded as a "dry" process without solvent and shows lower resolution which will affect the properties of the organic semiconductors 114, 117. Because of the limitations of these two methods, transfer printing technology is currently the key method for preparation of flexible electronics, especially for the inorganic flexible electronics 118, 119. The most important part of transfer printing technology is to precisely transfer required architectures onto target substrates using stamps. Although successfully applied in the fabrication of devices with unusual constructs 120-125, transfer printing technology is still not the most effective approach to fabricate circuits directly on the flexible substrates.

As a versatile, fast and promising manufacturing method, the printing electronic technology, which prints electronic components directly on flexible substrates to connect active electronic components, has become a hot spot in materials, electronics, and manufacturing in recent years 126. Compared with photolithography, shadow mask, and transferring printing technology, printing electronic technology is effective, low cost and has better compatibility with roll-to-roll production technology in the fabrication of flexible electronic devices. This technology could be divided into contact and non-contact printing technology. Contact printing technology contains gravure printing, screen printing, rotary screen printing, and flexorgraphic printing. Spray coating and ink-jet printing belong to non-contact printing technologies. Among the mentioned technologies, ink-jet printing technology is an environmentally friendly patterning method with lower material wastes and it also has better compatibility with substrates with different mechanical properties 127. Ink-jet printing technology has been utilized in the fabrication of various electronic components, like transistors 127, 128, solar cells 129, 130, light-emitting diodes 131, 132, sensors 133, 134, and multifunctional composite structures 135-137.

The emerging 4D printing, which is generated through adding another dimension, the time dependent shape or volume transformation, to 3D printing, would bring new opportunities to SMP-based flexible electronics 67. The self-assembly of 4D printed structures could benefit the function realization of the SMPFEDs, like conforming to bio-tissues or autonomously folding and unfolding. The SMP-based 4D printed structures have demonstrated potential applications in robotics 138, 139, flexible electronics 140, medical devices 141-144, etc. For example, Yang et al. 138 fabricated a SMP gripper using shape memory polyurethane (SMPU, T_r: ~45 ºC) by fused deposition modeling and the gripper can successfully grab a cap of a pen, as illustrated in Fig. 3A. Zarek et al. 140 printed a shape memory construct (Fig. 3B) based on shape memory polycaprolactone (SMPCL, T_r: ~55 ºC) by stereo lithography and then deposited silver nanoparticle inks as the conductive coatings on the surfaces of 3D SMPCL structures. The recovery of the temporary shape of printed structure could form a conductive path to light up a LED as illustrated in Fig. 3B. Wei et al. 142 fabricated a remotely actuated 3D scaffold (Fig. 3C) by direct-writing of the shape memory polylacticacid (SMPLA)/Fe_3O_4. Most of the currently printable polymer materials are thermoplastic because thermoset materials are hard to be directly printed. Though, a lot of efforts have been made to construct complicated structures with thermoset polymers based on 3D printing 144, 145. From the reports, the available SMP and SMPC materials for 4D printing include SMPU, SMPCL, SMPLA and their composites 138, 140-143, 146, thermoplastic SMPLA with post-crosslinking structures 147, photocurable SMP materials and composites 139, 140, 148, 149, digital materials with good shape memory functions 150-156, thermoset SMPs and SMPSCs that can be pre-cured at room temperature 142, 157, etc. However, there are still some challenges in using 4D printing technology in flexible electronics, like printing multi-functional materials at macro and micro level to manufacture 3D structures to achieve functional integration. Besides, the combination of 4D printing pattern method with roll-to-roll production technology (R2R) for manufacturing large-scale SMP-based 3D structural electronics will undoubtedly improve the production efficiency 158.

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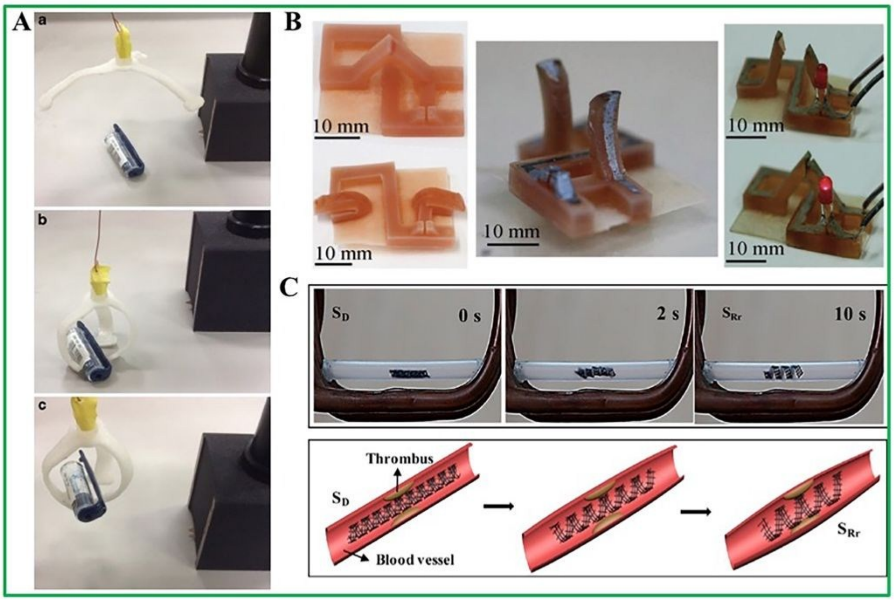

Fig. 3 (a) A 4D printed SMP-based gripper 138. (b) 4D printed construct for flexible electronic devices 140. (c) Deployment of a 4D printed biomedical scaffold 142. Reproduced from ref. 138 with the permission of Springer Nature. Reproduced from ref. 140 with the permission of John Wiley and Sons. Reproduced from ref. 142 with the permission of American Chemical Society.

4. Actuation methods for SMP-based flexible electronics

The following parts of this paper will introduce potential actuation methods of triggering SMPFEDs. In most studies, the shape change of SMPs is triggered when the temperature is higher than the transition temperature of SMPs. However, how to obtain the required temperature is a problem in many practical scenarios. Therefore, many investigations have been conducted to actuate the SMPs or SMPCs by solution (water or organic solvent), electricity, magnetism or light 41-49.

In some specific SMP-solution systems, the transition temperature of SMP could be significantly reduced due to the disruption of intermolecular hydrogen bonds and plasticization of SMP. As mentioned before, this will provide an alternative approach to drive implantable biomedical SMPFEDs since the shape recovery of SMPFEDs could be realized without additional heating, due to the transition temperature reduce of the SMP substrate upon immersion into physiological conditions. Fig. 4A demonstrates a nerve cuff conformed to the vagus nerve after being implanted in an anesthetized rat 91. The reported SMP/SMPC-solution systems include water-driven SMPU 48, 159, dimethylformamide-driven thermosetting styrene-based SMP 160, water-actuated sodium dodecyl sulfate/thermosetting epoxy-based SMP 161, and water-induced graphene oxide reinforced polyvinyl alcohol nanocomposites 162, which have application potentials in SMP-based flexible medical devices.

Compared with direct heating, the joule heating method is more convenient, efficient, and controllable in driving SMPFEDs. To realize heating by electricity, SMP is required to be made conductive. An effective approach is to fill conductive materials into SMPs. After being incorporated into SMPs, those conductive materials, including carbon nanofibers 163, carbon nanotubes 164, 165, metal nanoparticles 166, and graphene 167, could greatly improve the electrical performances of SMPs without reducing the shape recovery performance, which would possibly broaden the potential applications of SMPFEDs into electro-driven shape memory display, optical devices and other areas. An example of the shape recovery by Joule heating of 4D printed electronic components is illustrated in Fig. 4B 157.

Similarly, the shape changes of SMPFEDs can also be actuated through induction heating by alternating magnetic field when the magnetic nanoparticles, such as Fe_2O_3 168 and Fe_3O_4 44, are incorporated with SMPs 169. The induction heating could enable remotely controlled recovery of SMPFEDs through locally or selectively induced heat 170, which is meaningful in some biomaterial applications such as contactless treatment of tumour and removal of clot in blood vessel 171. As shown in Fig. 4C, the magnetic-actuated SMPCL/Fe_3O_4

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nanocomposite could deploy under the alternating magnetic field 44.

Another approach to realize remotely and spatially control of the recovery of SMPFEDs is through light 170. In order to achieve the actuation of SMPFEDs under an appropriate light radiation, the photo-responsive groups of light-activated SMPs, such as cycloaddition of cinnamate (or coumarin) groups and isomerization of azobenzene, could be used as the switching molecules 46, 172-174. Lendlein 46 et al. have illustrated the mechanism of molecular switches of cinnamate (or coumarin) groups under UV light and the UV-irradiated specimen can have shape recovery after exposed to the UV light (<260 nm) due to the cleavage of the cross-links. In addition to molecular switches, it is also possible to heat the SMP through light absorption by adding functional fillers such as nanocarbon, gold nanoparticles, and nanorods to SMPs 175-178. The shape recovery of SMP under light is illustrated in Fig. 4D 178.

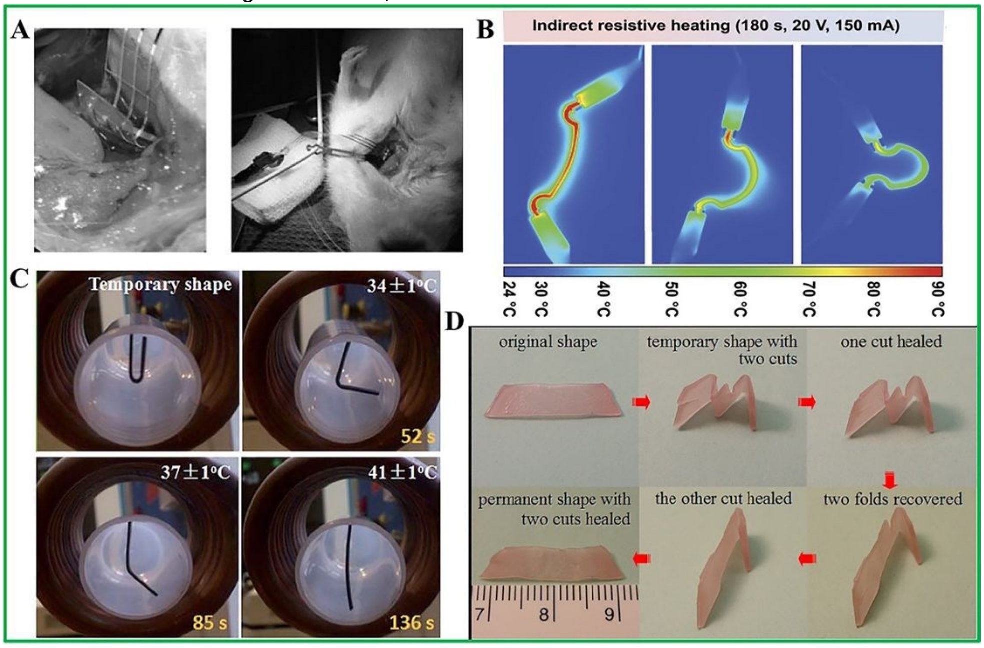

Fig. 4 (A) A SMP-based nerve cuff conforming to the vagus nerve in physiological environment after implantation 91; (B) Shape recovery of a 4D printed SMP composite structure under direct heating (left) and Joule heating (right) 157; (C) Shape recovery of SMPCL/Fe_3O_4 nanocomposites in alternating magnetic files 44; (d) the light-controlled shape recovery process of cross-linked PEO/AuNP 178. Reproduced from ref. 44 with the permission of Royal Chemistry of Society. Reproduced from ref. 91 with the permission of John Wiley and Sons. Reproduced from ref. 157 with the permission of Springer Nature. Reproduced from ref. 178 with the permission of American Chemical Society.

5. Challenges and developments for SMP-based flexible electronics

From the above discussions, it can be seen that SMP-based flexible electronic devices are capable of memorizing pre-deformed shapes and recovering to permanent shapes when exposed upon environment changes. Endowed with features like shape memory effect and variable modulus, SMPFEDs could realize more functions and thus have potentials in some applications. With the development of SMPs and SMPCs, the structural optimization and mature processing technique of flexible electronics, remarkable achievements have been obtained in SMP-based flexible electronics during last few years.

The state-of-the-art of SMPFEDs including SMP types, T_r, fabrication technology, devices and applications is illustrated in Table 1. The maximum T_r of the above SMPs is 150 °C, which means that those SMPs could not be used in high temperature that is required in some fields like aerospace. To meet different application requirements, it is necessary to develop novel SMPs and SMPCs with broader transition temperature range. Although thiol-ene/acrylate SMP substrates are suitable for implantable devices, some technical problems are to be solved for biomedical applications. The reduced modulus (~80 MPa) of the substrate is still much higher than the modulus (~100 KPa) that biological tissues, which may lead to bio-incompatibility. The high driving temperatures make some SMPs unsuitable for using in wearable electronic products due to the possible damages to body. Thus for the biomedical or wearable SMPFEDs, the SMPs with lower transition temperature range and modulus will be required. Except for the chemo-actuation of thiol-ene/acrylate SMP substrates, SMPFEDs trigged by other methods should be developed to realize the programmable and remotely controllable actuation. Moreover, SMPs with two-way or multiple shape memory effect could be used in SMPFEDs to realize more complicated functions 179-185.

The emerging 4D printing technology provides another promising method for fabricating SMPFEDs. As can be seen from Table 1, the SMP materials that can be used for printing are generally thermoplastic SMP, photosensitive SMP, post-crosslinking SMP or normal-temperature cured SMP. However, thermoset SMPs cannot be directly printed by the additive

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manufacturing equipment. Syntheses of novel 4D printable shape memory materials with more functions will greatly accelerate the development of SMP-based flexible electronics. Suitable technology to realize the quantity production is also a challenge in practical applications and commercialization of SMPFEDs.

Table 1 The SMPs, driving methods, T_r, fabrication technologies, applications and research teams of SMPFEDs

SMP materials	Driving methods	T_r (°C)	Fabrication technologies	Applications	Research teams
NIPPON MEKTRON	Heat	150	Shadow mask	Pressure sensors	Sekitani [79] et al
			Photolithography	Biomedical devices	Voit [91] et al
Thiol-ene/acrylate SMP	Heat, solvent	70	Shadow mask	Biomedical devices	Avendano-Bolivar [80] et al
			Shadow mask	Biomedical devices	Reeder [11] et al
			Shadow mask	Wearable electronics	Gaj [77] et al
Crosslinked poly(acrylate)	Heat	120	Shadow mask	Flexible display	Yu [75] et al
cEVA SMP	Heat	100	Nanoindentation	Optical devices	Xu [110] et al
SMPS	Heat	100	Transfer printing, hot embossing	Optical devices	Li [111] et al
SMPU	Heat	45	4D printing	Robotics	Yang [138] et al
Photo-curable methacrylate based SMP	Heat	-50~+180	4D printing	Biological gripper	Ge [139] et al
Methacrylated SMPCL	Heat	55	4D printing	Biomedical devices	Zarek [141] et al
Digital SMPs	Heat	0~70	4D printing	Active structures	Wu [153] et al
SMPLA/Fe_3O_4	Heat, magnetism	7~66	4D printing	Biomedical devices	Wei [142] et al
SMPLA	Heat	60	4D printing	Smart textiles	Schmelzeisen [70] et al
SMPCL, SMPCL/CNT	Heat, electricity	55	4D printing, deposition	Actuators	Zarek [140] et al

Conclusions

The shape memory effect and variable modulus of SMPs could endow more functions with flexible electronic devices, leading to a new research area of SMP-based flexible electronics. The light-emitting diodes, thin film transistors and optical devices have been developed based on different SMP substrates. Some special propertied of the SMPs will be beneficial for particular applications. For example, the decrease in transition temperature and modulus of thiol-ene/acrylate SMP after immersed in physiological environment make this material suitable in SMPFEDs with biomedical applications. The currently used fabrication technology for flexible electronics, like transfer printing technology and printing electronic technology, could be transferred in the fabrication of SMPFEDs. The emerging 4D printing technology is promising to build SMPFEDs with more complicated functions. The actuation of SMPFEDs could also refer to the common actuation methods of SMPs to meet different application requirements. However, there are still many challenges in SMP-based flexible electronics for practical applications or commercial production. The synthesis of SMP material systems, with broad transition temperature ranges, large modulus spans and two-way or multiple shape memory effect, development of novel fabrication methods for electronic devices with complicated structures and combination of different actuation methods to realize the programmable and remotely controllable actuation would obviously broaden the application of SMPFEDs.

Conflicts of interest

There are no conflicts to declare.

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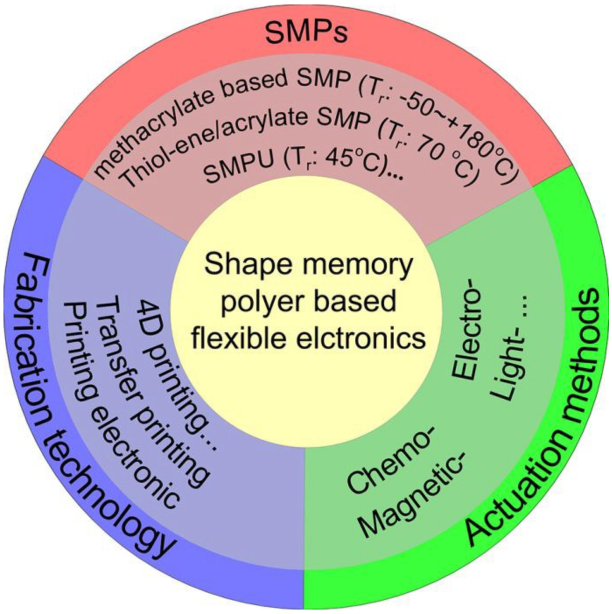

Highlight:

This review summarizes the advances and challenges of shape memory polymer based flexible electronic devices