Materials Today Advances 7 (2020) 100089

Contents lists available at ScienceDirect

Materials Today Advances

journal homepage: www.journals.elsevier.com/materials-today-advances/

Materials and manufacturing strategies for mechanically transformative electronics

S.-H. Byun [a, c], J.Y. Sim [b, c], K.-C. Agno [a], J.-W. Jeong [a, *]

[a] School of Electrical Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon, 34141, Republic of Korea
[b] Welfare & Medical ICT Research Department, Electronics and Telecommunications Research Institute, Daejeon, 34129, Republic of Korea

ARTICLE INFO

Article history:
Received 30 April 2020
Received in revised form
30 May 2020
Accepted 2 June 2020
Available online xxx

Keywords:
Mechanical mode conversion
Stimuli-responsive materials
Phase change
Stiffness tuning
Reconfigurable electronics

ABSTRACT

The static mechanical properties of conventional rigid and emerging soft electronics offer robust handling and interfacing mechanisms and highly compliant and adapting structures, respectively, but limit their functionalities and versatility. Mechanically transformative electronics systems (TESs) have extensive potential applications beyond these existing electronics technology owing to their ability to achieve both rigid and soft features as a result of bidirectional reconfiguration of their mechanical structure under the influence of stimuli (e.g. heat, electric/magnetic field, light, stress). In this article, we review recent advances in materials and fabrication methods as well as their applications for the development of TESs. We present key requirements for TESs and cover a range of stimuli-responsive materials and design strategies. Potential applications with demonstrated utility in wearables, implantable devices, sensors, and robotics, alongside key challenges and opportunities in the development of this emerging technology, are also discussed.

1. Introduction

Modern electronics are manufactured either in a flat and rigid form, which is common in conventional electronics, or in a soft and flexible form, which is typically used in recent wearable and implantable devices. Generally, conventional rigid and soft electronics [1,2] are application-specific, and their design and material properties are fixed over their entire life span. A rigid form factor is commonly used for consumer electronics, such as laptops and smartphones, owing to their static structure that facilitates handling and interfacing. However, when conventional rigid electronics are integrated with bodily tissue, their hard interface can cause discomfort [3,4] and inflammation [5–7] owing to the mechanical mismatch between soft tissue and the hard electronic structure. On the other hand, soft electronics are optimal for wearables [1–4,8–14], implantable devices [15–21], and soft robotics [22,23] because of their high compliance that enables seamless interfacing. However, their inherent softness and limited load-bearing capability make handling and interfacing difficult and inconvenient.

To overcome the physical limitations of both rigid and soft electronics, recent research in materials and manufacturing strategies has established a new class of electronics, referred to as transformative electronics systems (TESs), which can tune their shape, rigidity, and stretchability upon receiving a stimulus to match the intended application (Fig. 1). TESs can hold their shape and withstand loads and contact forces in rigid mode. When the same device becomes soft, it can curve, stretch, and adapt to dynamic deformation. This reconfigurability of TESs can open new opportunities for a wide variety of applications including personal electronics, biomedical devices, sensors, and robotics, by offering advantageous features of both rigid and soft electronics, which make them highly versatile. A key to the transformation of TESs is their variable stiffness enabled by stimuli-responsive materials [24–49]. When these responsive materials, such as phase-change metals [24–34,49], shape-memory polymer (SMP) [35–40], electroactive polymer [41–44], and smart fluids (magnetorheological fluid [MRF] and electrorheological fluid [ERF]) [45–47], are exposed to an appropriate stimulus (e.g. heat [25,28,31,49], light [40], magnetic field [45,47], voltage [41–44], or stress [48]), their mechanical properties can be significantly changed, altering from either rigid-to-soft or soft-to-rigid state.

Overviews of stimuli-responsive materials were reported previously, highlighting either a particular stiffness-variable material

* Corresponding author.
E-mail address: jjeong1@kaist.ac.kr (J.-W. Jeong).
[c] These authors contributed equally to this work.

https://doi.org/10.1016/j.mtadv.2020.100089

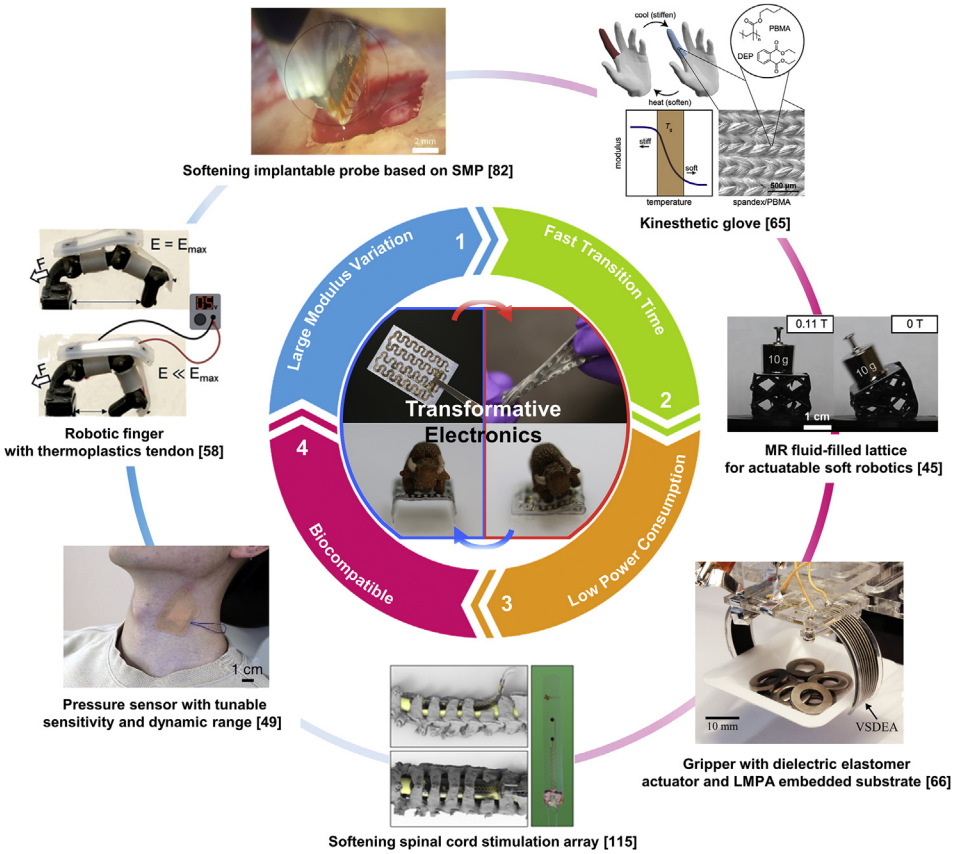

Fig. 1. Overview of transformative electronics systems, illustrating their key properties, design parameters, and applications [45,49,58,65,66,82,115]. All the images are reproduced with permission from Jackson et al [45], Byun et al [49], Rich et al [58], Carpenter et al [65], Shintake et al [66], Zátonyi et al [82], and Garcia-Sandoval et al [115]. LMPA, low-melting-point alloy; SMP, shape-memory polymer; MR, magnetorheological.

(e.g. SMP [50] and liquid metal [51–53]) for various potential applications or a range of materials intended for a specific application (e.g. soft robotics [54,55] and medical devices [56]). While some views in previous review articles can be applied to TES design, unique aspects of TESs that integrate stimuli-responsive materials with advanced electronics design have not been reviewed yet. In this article, we review recent technologies that present materials and fabrication techniques and their applications to the design and implementation of TESs. The article begins with key requirements for TESs and then reviews various stimuli-responsive materials and design approaches. The subsequent section presents potential applications of this technology with demonstrated functionality in wearables, implantables, robotics, and sensors. Finally, this review concludes with a discussion of key challenges and opportunities of this technology, which will encourage continuous research and development toward future electronics.

2. Requirements for transformative electronics

For practical utilities in various real-world applications, ideally, TESs should have the following capabilities: (i) large stiffness tuning, (ii) fast phase transition between rigid and soft modes, (iii) energy-efficient operation, and (iv) biocompatible integration with biological tissue (Fig. 1). The capability of large elastic modulus tuning with a ratio of more than 10^4 between soft mode (<MPa) and rigid mode (>GPa) is a crucial feature for transformative electronics to fully leverage key attributes of both emerging soft electronics and traditional rigid electronics. In soft operation mode, TESs that can lower the elastic modulus to the extent comparable with that of soft tissue (i.e., order of tens to hundreds kPa) are desired to allow not only conformal integration with the curvilinear surface of the body but also accommodation to natural tissue deformation to facilitate wearable and implantable applications. On the other hand, in rigid mode, the ability of electronics to increase their hardness to the level of rigid electronic or packaging materials (e.g. silicon, metal, glass, etc.) is required to enable the unique features of conventional rigid electronics that are optimal for load-bearing tasks and user-electronics interfaces. For this reason, TESs should be designed to achieve the maximum stiffness-tuning ratio. Together, advanced functions, such as stiffness tuning between multiple states [24,25,29] and gradual stiffness modulation [45], can enable new opportunities for broad applications. Besides, rapid mode conversion between soft and rigid states and low power consumption during mode transition are other indispensable characteristics required for TESs. The former attribute makes TESs highly adaptable for various needs, enhancing productivity as well as convenience of use, while designs optimized for energy-efficient operation enable electronics to be operable for long periods of time. These factors must be taken into account in the choice of materials and the design of device structures to maximize the versatility and practicality of TESs. Biocompatibility is also an essential aspect when using TESs in biomedical applications. Electronics that do not cause any skin irritation, allergic reaction, or severe inflammatory response will ensure biologically safe operation while performing their desired functions on or within the body. Various stimuli-responsive materials have been investigated to enable mechanically reconfigurable electronics with the aforementioned characteristics. In the following section, we discuss materials and design

strategies for the emerging transformative electronics that may provide insights for the development of future electronics beyond the present rigid or flexible/stretchable form of electronics.

3. Materials and design approaches

3.1. Comparison of stimuli-responsive materials

Stimuli-responsive materials play a central role to meet the requirements of TESs discussed in the previous section. These materials change their mechanical properties through physical and chemical transformations, which can be activated by thermal transports or the application of electric/magnetic fields. Table 1 summarizes the key characteristics of stimuli-responsive materials with respect to activation methods, modulus-tuning range, transition time, and their applications. Thermally activated materials include gallium [24–26,49], low-melting-point alloys (LMPAs) [27–34,57], SMPs [35–39], and thermoplastics [44,58,59]. Materials that undergo field-induced transformations incorporate magnetorheological elastomers (MREs) [60,61], dielectric elastomers [42,43], and elastic composites filled with ERF or MRF [45–47,62]. In this section, we discuss both thermally activated and field-activated materials regarding the principle of their transformation and describe the advantages and drawbacks of each material for use in TESs.

3.1.1. Thermally activated materials

Thermally activated materials can be categorized by the type of state transition, namely, phase transition and glass transition. Phase-transition materials include gallium and LMPAs in which the phase is converted between solid and liquid at the melting temperature (T_m). Glass-transition materials are SMPs and thermoplastics that undergo glass transition between rubbery and glassy states at the glass transition temperature (T_g). When gallium and LMPAs are integrated with elastomers, their phase transition allows the effective elastic moduli of devices to switch in a wide range between hundreds of kPa and a few GPa (Fig. 2a). As gallium and LMPAs become liquid above T_m, the effective elastic moduli decrease approximately to the modulus of encapsulants (e.g. tens or hundreds of kPa) because the contribution of the liquid metals becomes negligible. However, phase-transition materials require a relatively long transition time and large energy consumption to meet the demand of latent heat and sensible heat (Fig. 2b and c). In contrast to gallium with the T_m of 30 °C, the T_m of LMPAs varies from 45 °C to 62.5 °C (Table 1). The high melting temperature of LMPAs ensures reliable operation in rigid mode at room temperature in the daily life, but a disadvantage is that the higher the sensible heat to reach T_m, the longer the switching time and the larger the power expense. Furthermore, such a high temperature would be undesirable in wearable and skin-attachable applications owing to the potential of skin damage.

In comparison to LMPAs, thermoplastics are of low cost, lightweight (~1 g/cm^3), and compatible with three-dimensional (3D) printing techniques, enabling easy fabrication and manufacturing of geometrically complex structures [37,63,64]. However, their slow transition time with a small modulus-tuning range (between 1 MPa and tens of MPa) [44,58,59] limits their range of application. SMPs show a relatively large modulus-tuning range comparable with LMPAs, but their low thermal conductivity, which impedes the glass transition temperature being reached, prevents rapid rigidity modulation. On the other hand, SMPs show lower power consumption (few watts) than LMPAs owing to the second-order transition that glass transition typically presents [35,39]. In general, thermally activated materials present a relatively slow state-transition speed in comparison with field-activated materials (Fig. 2b).

Table 1
Comparison of various stimuli-responsive materials in terms of stiffness-tuning performance.

Material	Modulus	Modulus tuning ratio	Transition point	Transition time (rigid to soft)	Transition time (soft to rigid)	Application	Ref
Gallium	0.1–10000 MPa	100000	30 °C	1 s	200 s	Implantable	[24]
Gallium	4.5–1250 MPa	278	30 °C	>15 s (body heat)	15 s (ambient air)	–	[25]
Gallium	0.05–0.16 MPa	3.2	30 °C	1 s (4.56 W)	1 s (ambient air)	–	[26]
Gallium	0.022–9 MPa	409.1	30 °C	–	–	Sensor	[49]
LMPA	0.1–3200 MPa	32000	62 °C	–	–	–	[27]
LMPA	–	4000	55 °C	12 s	70 s (0 °C water)	Actuator	[28]
LMPA	347–35700 N/m	103	47 °C	150 s (18 W)	240 s (ambient air)		[29]
LMPA	–	–	62 °C	3.5 s (28 W)	58 s (4 °C water)	Biomedical	[30]
LMPA	1.25–888 MPa	710	62 °C	25 s	30 s	Robotics	[31]
LMPA	1.5–40 MPa	26.7	45 °C	1 s (0.5 W)	10–60 s (ambient air)	–	[32]
LMPA	0.1–3.1 MPa	31	62 °C		79 s (ambient air)	Robotics	[33]
LMPA	–	–	47 °C	>5 s (1.47 W, NiCr wire)	>40 s (ambient air)	Actuator	[34]
LMPA	–	–	50 °C	60 s (0.47 W)	-(0 °C water)	Actuator	[57]
SMP	6.73–167.73 N/m	24.9	45 °C	7 s	10 s (ambient air)	Robotics	[35]
SMP	9.0–1400 MPa	155.6	60 °C	10 s (2.02 W, AgNP)	22 s (4 °C water)	Actuator	[36]
SMP	0.08–11.6 MPa	139.8	71 °C	–	–	–	[37]
SMP	3.33–125.65 MPa	37.7	55 °C	20 s (2.48 W, NiCr wire)	75 s (ambient air)	Actuator	[38]
SMP	1.40–2150 MPa	1535.7	45 °C	40 s (0.75 W)	15–30 s (ambient air)	Actuator	[39]
Thermoplastics	1.5–37 MPa	24.7	75 °C	6 s (3.3 W)		Robotics	[44]
Thermoplastics	0.7–10.4 MPa	14.9	73 °C	2–40 s (5 W)	–	Wearable	[58]
Thermoplastics	0.86–30.12 MPa	35	75 °C	15 s	–	Robotics	[59]
MRE	3.10–3.4 MPa	1.1	0–130 mT	–	–	–	[60]
MRE	0.85–1.31 MPa	1.5	0–940 mT	–	–	–	[61]
MRF	0.047–0.076 MPa	1.6	0–180 mT	0.23 s	0.83 s	–	[45]
MRF	300–900 N/m	3	0–36 mT	–	–	Biomedical	[47]
MRF	0.5–5 MPa	10	10–35 mT	–	–	–	[62]
ERF	0.043–302 kPa	7023.3	3 kV/mm	<2 s	5 s	–	[46]
Electroactive unit	0.5–1.6 N/mm^2	3.2	0–1.3 V	–	–	Artificial muscle	[41]
Electroactive unit	12.6–39.1 N/m	3.1	0–5.6 kV	–	–	Wearable	[42]
Electroactive unit	2.9–79.7 N/m	27.5	0–500 V	–	–	–	[43]

ERF, electrorheological fluid; MRF, magnetorheological fluid; MRE, magnetorheological elastomer; SMP, shape-memory polymer; LMPA, low-melting-point alloy.

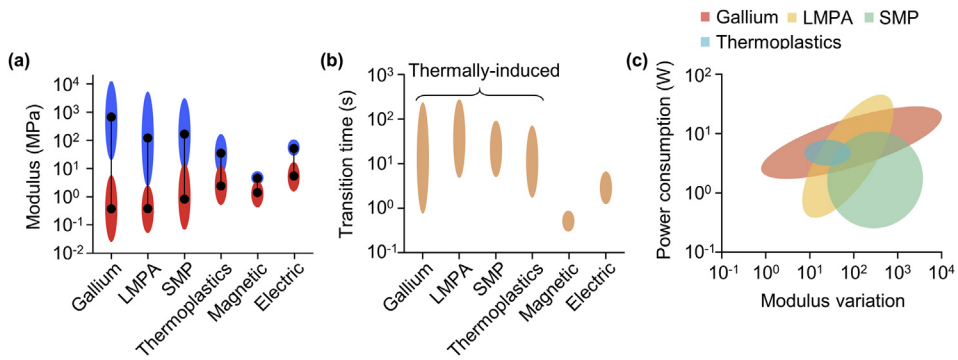

Fig. 2. Summary of key properties of stimuli-responsive materials (Table 1) that can be used for the construction of TESs. (a) Moduli of stimuli-responsive materials in rigid (blue) and soft (red) modes. (b) Transition time for mechanical transformation. (c) Correlation between consumed power to change the modulus and modulus variation. LMPA, low-melting-point alloy; SMP, shape-memory polymer; TES, transformative electronics system.

To establish a standalone system, heating units are commonly integrated with thermally activated materials to build TESs. The assistive heating element can be conductive fabrics, wires [28,31], or nanoparticles [36]. In the case of conductive stimuli-responsive materials, such as gallium and LMPAs, the material itself can be used not only to generate heat by Joule heating but also to provide a stiffness-tuning platform [30,32]. Therefore, these conductive materials are favorable for the simplification of the TES architecture. In contrast, SMPs and thermoplastics are electrically non-conducting; therefore, they require an additional heating layer to enable stiffness tuning. Blending conductive additives, such as carbon black, enables Joule heating, but mediocre electrical conductivity of the resulting composite requires a high operating voltage [39,58,59]. The assistive heating element accelerates switching of the operation mode from rigid to soft, but cooling components enabling soft-to-rigid transition have not been extensively utilized for TESs yet. Applying cold water enables rapid transition from soft mode to rigid mode, but the need for a cold water supply inhibits TESs from working as independent electronics [28,30,57]. A recently reported flexible thermoelectric device can be considered as an assistive cooling element for integration with TESs to form a compact, standalone system [65].

3.1.2. Field-activated materials

Magnetic/electric field—activated materials are potential candidates for agile stiffness tuning in TESs. Magnetic activation exploits microsized/nanosized ferromagnetic particles in either a liquid (MRF) or solid (MRE) phase carrier. For instance, polymer structures filled with MRFs have been reported [45,47,62] in which applied magnetic fields align particles and form chains, increasing mechanical resistance to deformation of the structure. However, an MRF has several drawbacks, such as the agglomeration and sedimentation of magnetic particles, leakage of fluid, and environmental contamination [60]. MREs are promising alternatives, which consist of an elastic matrix with embedded magnetic particles [60,61]. MREs are preferable to enable stiffness tuning because they do not suffer from the aforementioned shortcomings of MRFs [55]. Electroactive polymers, a class of materials activated by electric fields, are electric counterparts of magnetically activated materials. Dielectric elastomers [42,43] and ionic electroactive polymers [41] are representative examples. In dielectric elastomers that consist of an elastomeric dielectric film sandwiched between two compliant electrodes, a voltage applied across the electrodes results in Maxwell stresses, expanding the area and reducing the thickness of the film. Actuation forces have been used to modulate stiffness by changing the film shape [42] or inducing electrostatic chucking [43].

Field-induced mechanisms are preferable to thermal activation when rapid transition is needed (Fig. 2b). Jackson et al. presented three-dimensionally printed hollow struts filled with MRFs whose response times were 0.23 s for softening and 0.83 s for hardening [45]. Field-induced materials are also favorable to modulate the effective elastic modulus continuously. However, one drawback of this approach is a small modulus-tuning ratio (1.1~10 in magnetic activation; 3.1~27 in electric activation. See Fig. 2a and Table 1). Magnetically activated materials additionally require an external apparatus to apply magnetic fields, which can be bulky, energy-inefficient, and difficult to scale [55]. Electroactive polymers are superior to magnetically activated materials in this regard owing to their lightweight, compact dimensions, and convenience in control [41].

3.2. Design and fabrication approaches for transformative electronics

The underlying scheme to achieve TESs is incorporating a stiffness-tuning function into soft electronics. A simple but effective design approach is to integrate flexible/stretchable electronics with a mechanically transformative platform, which is a variable-stiffness structural composite embedded with stimuli-responsive materials as described in the previous section (Fig. 3a). Functional electronic layers can be assembled onto the transformative platforms by using adhesive bonding [49], oxygen plasma bonding [66], or direct transfer printing [67] (Fig. 3b). A silicone elastomer (e.g. poly(dimethylsiloxane) [PDMS] and series of Ecoflex) is one of the most useful base materials for transformative platforms. It allows the bonding of soft electronics built with silicone onto the platform via oxygen plasma treatment [68,69] or by using uncured elastomers as adhesives [70]. Using these integration methods, inorganic semiconductors, sensors, and metallic device structures can be directly transfer-printed from a donor substrate (e.g. silicon wafer) to a silicone-based transformative platform.

Various fabrication approaches can be applied to implement transformative platforms, depending on their base stimuli-responsive material (Table 2). In the case of solid-type stimuli-responsive materials (e.g. SMPs, thermoplastics, and MREs), a transformative platform can be manufactured by direct 3D printing [38,39], molding [71], melt spinning [72], and hydraulic heat pressing [61] (Fig. 3c). For photosensitive SMPs and thermoplastics, stereolithographic 3D printing (Fig. 3c, ①) is applicable to form complex shapes precisely [63] (Fig. 3d). If a fiber configuration is desired, these materials can also be processed using the melt spinning method (Fig. 3c, ②). Magnetorheological elastomers can be fabricated by mixing magnetic particles into an uncured elastic

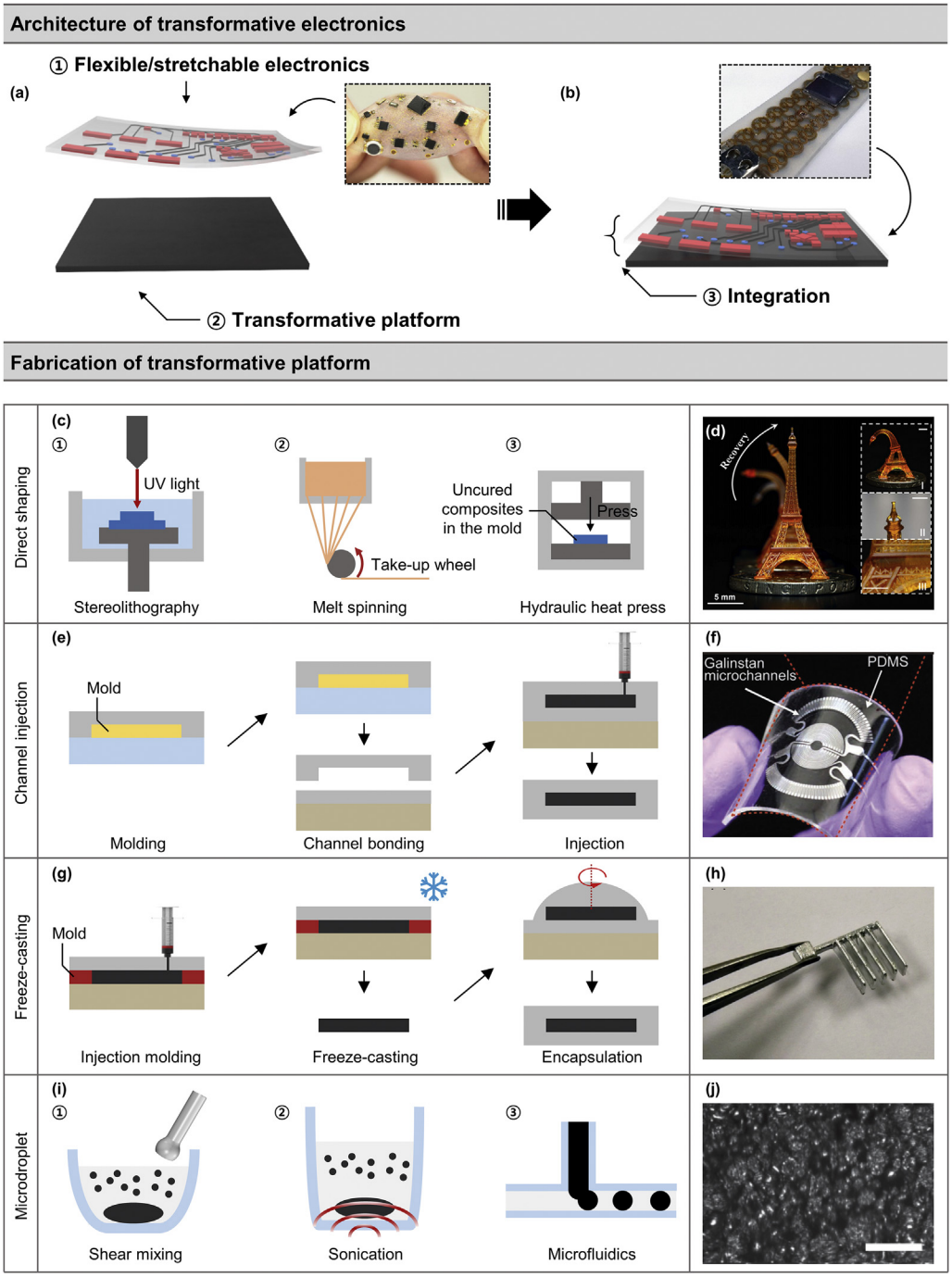

Fig. 3. Schematic diagrams illustrating the architecture and fabrication of TESs. (a) Exploded-view diagram that highlights the major components of TESs: that is, flexible/stretchable electronics and a transformative platform. The inset shows an image of a representative flexible/stretchable electronic device [13]. (b) Illustration of a complete TES, which integrates flexible/stretchable electronics with a transformative platform. The inset shows an image of an example TES built by combining a gallium-based transformative platform with stretchable electronics. (c–j) Illustration of various fabrication approaches for transformative platforms incorporating stimuli-responsive materials. (c, d) Solid-phase composite approaches and an optical image showing three-dimensionally printed structures with photo-curable SMPs [63]. (e, f) Channel injection method and the resulting device filled with gallium alloy [68]. (g, h) Freeze-casting method and the corresponding image highlighting a freeze-cast gallium alloy structure that allows easy handling [73]. (i, j) PCM microdroplets-elastomer composite approaches and a scanning micrograph showing PCM microdroplets mixed with elastomer by sonication [74]. All the images are reproduced with permission from the authors of the studies mentioned. PCM, phase-changing material; SMP, shape-memory polymer; TES, transformative electronics system.

matrix to prepare base composite materials, followed by extrusion, molding, and curing (Fig. 3c, ③) [61]. Transformative platforms can also be constructed using phase-changing stimuli-responsive materials, such as gallium, LMPAs, or MRFs, by injecting them into polymeric fluidic channels (Fig. 3e and f) [68], by freeze-casting them (Fig. 3g and h) [73], or by mixing microdroplets with elastomers (Fig. 3i and j) [49,74]. For the channel injection approach, fluidic channels are manufactured by creating channel patterns on a planar substrate [32,49], extruding tubular fibers [25,29,30], or three-dimensionally printing hollow struts [45]. Planar channel patterns can be created by using photolithographically defined molds for micron-resolution patterns [32] or three-

Table 2
Comparison of various fabrication processes for TESs.

Fabrication method	Direct shaping			Channel injection	Freeze-casting	Microdroplets		
	Stereolithography	Melt spinning	Hydraulic heat press			Shear mixing	Sonication	Micro-fluidics
Material	SMP, thermoplastics	SMP, thermoplastics	SMP, thermoplastics, MRE	Gallium, LMPA, MRF	Gallium, LMPA	Gallium, LMPA, MR, ER	Gallium, LMPA, MR, ER	Gallium, LMPA
Resolution (μm)	1	100	100	10	100	20	0.1	75
Speed (production time scale[a])	− (min–hr)	++ (min)	+ (min)	− (hr[b])	− (hr)	+ (min)	+ (min)	− (hr)
Manufacturability of complex 3D structure	++	−	−	+	++	+	+	+
Advantages	• Dexterous manufacturing	• Mass producibility	• Even surface • Mass producibility	• Simple process	• Simple process	• Simple process	• Simple process	• Uniform droplet size • Low production speed
Disadvantages	• Material dependency • High-cost equipment	• Limited form (fiber) • High-cost equipment	• High-cost equipment • Difficult maintenance	• Limited geometry	• Vulnerable to deform • Low reproducibility	• Heterogeneous droplet size	• Severe supercooling of nanoscale droplets	
References	[35–39,63,64]	[116]	[58,60,61]	[24,25,29–34,41,45,47,57,62,68,117]	[26,28,73]	[27,49,118,119]	[46,118,120,121]	[122,123]

Qualitative evaluation scale: −, +, ++ from the lowest to highest values.
ER, electrorheological elastomer/fluid; LMPA, low-melting-point alloy; MR, magnetorheological elastomer/fluid; MRE, magnetorheological elastomer; MRF, magnetorheological fluid; SMP, shape-memory polymer; TES, transformative electronics system.
[a] Time scale for producing a batch of devices.
[b] Time is mostly taken to build channel.

dimensionally printed molds for larger feature sizes [49] (Fig. 3e). The patterned devices are then bonded to a flat elastomeric layer to form fluidic channels, which allow the injection of phase-changing materials (PCMs) to create transformative platforms with variable stiffness (Fig. 3f). The freeze-casting method (Fig. 3g) is another technique that can be used to fabricate the same type of transformative platforms. The difference from the previous approach is that it uses solid-state core structures (Fig. 3h) created by injection molding and freezing to make elastomer-encapsulated platforms. Although the transformative platforms constructed via channel injection or freeze-casting methods provide good stiffness tuning, they are not capable of uniform mechanical modulation across the platform owing to the discrete frame structures that do not cover the entire area. Creating transformative platforms using PCM microdroplet-elastomer composites is an alternative strategy that can address this issue. Shear mixing, sonication, or microfluidics-based droplet generation method (Fig. 3i) facilitates the dispersion of uniform PCM microdroplets in elastomers, thus helping to establish PCM microdroplet-elastomer composites with consistent characteristics across the platform (Fig. 3j) [74]. Shear mixing and sonication are simple and straightforward methods that can be used for this approach, but they result in a relatively large distribution of microdroplet sizes compared with the microfluidics-based scheme.

4. Applications of transformative electronics

4.1. Wearables

TES technology have opened new possibilities of making wearable electronics that are more effective to guide motions [31], give haptic feedback [75], and record physiological signals [49]. Moreover, they are convertible between flexible and rigid forms [58,76]. To achieve conformal contact on curvilinear body surfaces, soft electronics technology has been widely adopted to minimize the mechanical mismatch at the device-skin interface [1,10,12,77,78]. However, their intrinsically soft nature has limited their off-body usage because soft wearable devices are incapable of bearing loads and holding their shape by themselves. In our recent work published in an article called '*Mechanically transformative electronics*,' we presented a personal electronic system that is convertible between handheld and wearable configurations (Fig. 4a) [49]. A key material for stiffness tunability was a low-melting-point metal, gallium, encapsulated in silicone elastomer. Owing to the high elastic modulus of gallium (9.8 GPa) in the solid phase, the demonstrated device accomplished a change of more than two orders of magnitude in bending stiffness from 0.023 mN m^2 in soft mode to 2.6 mN m^2 in rigid mode. When the device was mounted on the skin, the skin temperature (~32 °C), which is above the melting temperature of gallium (29.8 °C), could convert it to soft electronics without an additional power supply.

Likewise, advanced wearable rehabilitation devices could be developed by integrating the tunable load-bearing ability of TESs to address the difficulty of securing body parts in a fixed position with soft wearable devices. For instance, Tonazzini et al. [31] developed a variable-stiffness fiber with an LMPA (Field's metal) core and demonstrated a reconfigurable finger splint by knitting the fiber with a cotton thread (Fig. 4b). When the LMPA core was solid, it provided sufficient strength to hold the finger posture during the healing process. When operating a built-in heater, the molten LMPA core allowed the finger movement to adjust the posture. Ramachandran et al. [75] demonstrated a wearable haptic device with a stiffness-tuning function, which consists of electroadhesive clutch plates woven into a textile (Fig. 4c). Using an electric-field trigger mechanism, the authors achieved rapid transformation (15 ms) and

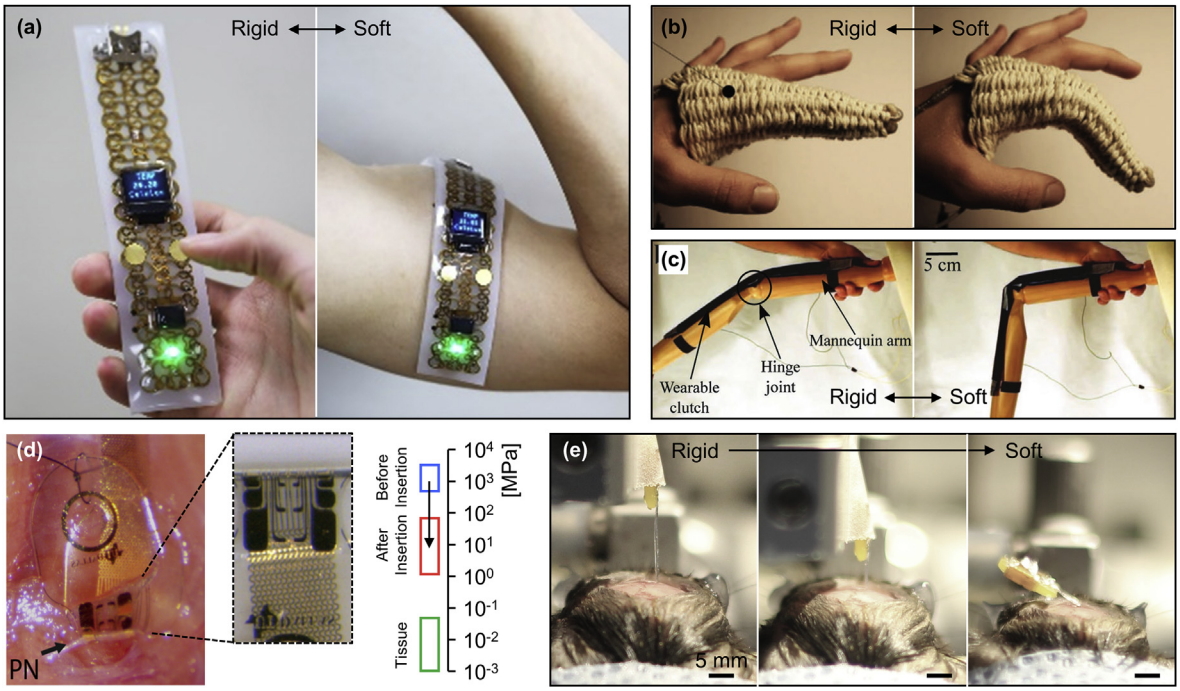

Fig. 4. Examples of TESs for wearable and implantable applications. (a) Personal electronics built on gallium-elastomer composite, which can convert between handheld rigid electronics and wearable flexible physiological sensors [49]. (b) Rehabilitation device based on LMPAs, which restricts unnecessary movements of the finger in rigid mode and allows the change of posture in soft mode [31]. (c) Haptic device based on electrostatic adhesion, which can control mechanical resistance to tensile load [75]. (d) Cuff electrodes based on SMPs, which can soften and wrap around nervous systems upon implantation [79]. (e) Neural probe built on a gallium needle that spontaneously softens upon being inserted into brain tissue to minimize inflammation response [49]. All the images are reproduced with permission from the authors of the studies mentioned. LMPA, low-melting-point alloy; PN, pelvic nerve; SMP, shape-memory polymer; TES, transformative electronics system.

low power consumption (1 mW) to control mechanical impedance to tensile loads up to 10 kg. Because the stiffness-tuning function allows users to perceive various amounts of mechanical impedance, the applications of the demonstrated haptic device can be extended to augmented reality.

4.2. Implantable devices

Similar to wearable devices, the ability of TESs to modulate stiffness within tissue expands the potential applications for implantable devices. One of the most representative implantable devices that can benefit from TES technology is neural probes. Neural probes inserted into brain tissue have allowed neuroscientists to directly investigate the neural circuitry of the deep brain. Albeit powerful and innovative, conventional rigid probes cause significant inflammation response over time owing to mechanical mismatch between the rigid probes and soft tissue, making them unsuitable for long-term use. To address this long-standing challenge, researchers have applied the mechanically transformative approach to the design of neural probes. TES probes that are rigid before implantation soften upon integration with tissue, thereby increasing biomechanical compatibility. SMPs have been widely used to develop softening neural implants [79–82]. For instance, Ware et al. [80] presented SMP-based cortical probes with large variation of modulus (between 18 MPa and 1 GPa). González-González et al. [79] recently demonstrated softening SMP cuffs with multielectrodes that enabled conformal contact with rat sciatic and pelvic nerves (Fig. 4d). Traditional cuffs require thick silicone walls (200–600 μm) to maintain intimate contact with the nerves, but this structural design exerts a large pressure on neural tissue, provoking an inflammation response at the neural interface. To reduce this adverse response, the authors used SMPs to make a smart cuff that softens upon implantation. The substantial decrease in the device modulus (from 1.8 GPa to 41 MPa) within tissue enabled the non-destructive yet intimate integration of electrodes with nerves. Gallium is another attractive material to enable TES implants. Recently, Byun et al. [49] reported a gallium-based microscale inorganic light-emitting diode (μ-ILED) neural probe for chronic *in vivo* optogenetics (Fig. 4e). In this work, a thin gallium needle (50 μm in thickness) encapsulated in silicone was used as a transformative platform to enable spontaneous softening upon injection into brain tissue. When implanted, the modulus of the probe was tuned from 50 MPa to 50 kPa, leading to significantly reduced inflammatory glial responses and lesion size compared with rigid tungsten probes of the same dimension. By using the TES probes, researchers successfully performed optogenetic experiments to control the feeding behavior in mice for more than 6 weeks.

4.3. Robotics

Soft actuators and robots made of intrinsically soft and elastic materials are rapidly gaining attention owing to their excellent ability to enhance interaction with the complex 3D geometry of the human body and delicate objects [83,84]. However, the low stiffness of their constituent materials makes the soft robotic systems unsuitable for tasks that require the ability to bear large loads, such as the manipulation of heavy objects [85], rehabilitation [86,87], and precise surgical operations [88]. To address this challenge, substantial efforts have been made to apply stimuli-responsive materials and structural design strategies to soft actuators and robots. For example, researchers have developed pneumatic soft actuators, one of the most widespread forms of soft robotics, integrated with stimuli-responsive materials, such as LMPAs

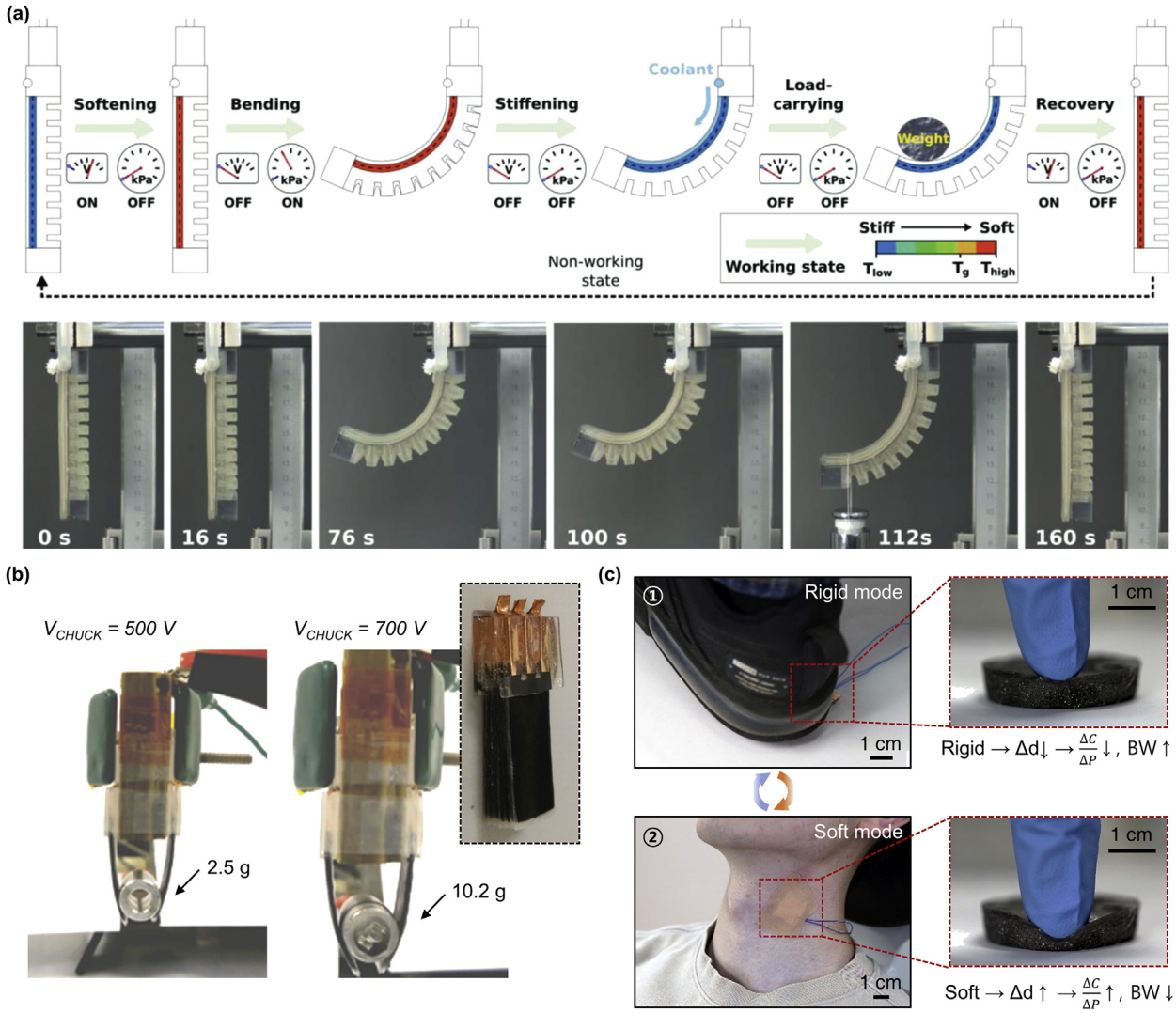

Fig. 5. Examples of TESs for applications in robotics and sensors. (a) Robotic actuators based on shape-memory polymers, which can control the load-carrying capacity through stiffness modulation [36]. (b) Dielectric elastomer actuators based on electric-field actuation, which can control the stiffness through modulation of an applied voltage [43]. (c) Transformative pressure sensor with tunable bandwidth and sensitivity, which is fabricated using gallium microdroplets-elastomer composite. The sensor can measure a large pressure (foot stepping) in rigid mode (1) and a small pressure (pulsation) in soft mode (2) [49]. The insets show the degree of deformation for each mode. All the images are reproduced with permission from the authors of the studies mentioned. TES, transformative electronics system.

[28–31,34,57], SMPs [35,36,38,39], and thermoplastics [58], to enable thermal tuning of their stiffness. Zhang et al. demonstrated SMP-based soft pneumatic actuators via hybrid multimaterial 3D printing that could stiffen by up to 120 times and lift objects with complex shape and a weight of 1.5 kg (Fig. 5a) [36]. The authors accelerated the transition speed by introducing printed Joule heating circuits and fluidic cooling channels, so that it could accomplish softening-stiffening conversion within 32 s.

Transformative designs also have been used not only to increase the maximum payload capacity but also to change the mechanical degree of freedom of soft pneumatic actuators. For instance, Hao et al. [28] developed an LMPA-based soft pneumatic actuator that allowed three distinct segments to selectively melt and reduce the average bending force by 35 times within a transition time of 12 s. The tunable degree of freedom of the individual segments resulted in six motion patterns, and the device was able to grasp objects weighing 20 times more than the actuator. Soft pneumatic actuators that can be selectively pressurized to control the number of bending points have also been reported, which use stimuli-responsive materials, such as LMPAs [31] and SMPs [35,36,39] to eliminate the need for multiple internal chambers and independent inlets and valves for control. Although the thermally tunable actuation is effective in achieving dramatic changes in payload capacity and bending stiffness, the major drawbacks are a long transition time and bulky design requiring a large pneumatic cavity and auxiliary pumping equipment.

Dielectric elastomers are promising alternatives for variable-stiffness actuators owing to their fast response and compact dimensions [42,43]. Exploiting these features, researchers have applied dielectric elastomers for tunable load capacity [42,43], adaptable mechanical impedance [89], and reconfigurable hand rehabilitation orthosis [90]. For instance, Li et al. [42] presented a dielectric elastomer actuator with a tunable load capacity that could increase the maximum relative stiffness by 71.8% and critical load-bearing capacity by 75.6%. Here, electric field actuation could modulate the longitudinal bending stiffness of the polymethyl methacrylate film structure by changing the transverse curvature of a curved strip. Imamura et al. [43] demonstrated another

mechanism of variable-stiffness actuators using electrostatic chucking between multiple dielectric layers (Fig. 5b). By clamping or releasing the adjacent layers, the authors showed that the actuator could increase maximum stiffness up to 39.2 times and lift a 10-g object with 0.6 g of its weight.

4.4. Sensors

Physical sensors such as strain and pressure sensors are another exciting area to which TESs can contribute with their mechanical tunability. Recent e-skin research has made remarkable advances in the development of soft tactile sensors that can be seamlessly integrated onto the skin [68,91–94]. Despite the extremely high sensitivity of such soft sensors [91,93,95], their broad application is restricted owing to the lack of a wide sensing bandwidth. To overcome the trade-off between the sensitivity and the dynamic range, researchers have explored *in situ* modulation of the sensitivity and the dynamic range of mechanical tactile sensors [96,97]. For instance, Zarate et al. [97] reported a vibrissae-type tactile sensor with variable stiffness that comprises two antagonistically working actuators connected via spring-like structures. The mechanical components of the joints and motors were, however, too bulky and rigid to meet the requirements of soft electronics adapting to complex 3D surfaces and bodily tissues.

Recent innovations in materials and manufacturing approaches addressed this issue by applying variable-stiffness technology to pressure sensors (Fig. 5c) [49]. A key component of these pressure sensors is an active layer made of gallium microdroplets dispersed in silicone elastomers, which is sandwiched between indium-tin-oxide electrodes to enable capacitive pressure sensing. This active layer can convert between soft and rigid states via the phase transition of gallium to enable the pressure sensor to change its operation mode between the soft mode with an ultrahigh sensitivity and a narrow dynamic range and rigid mode with a large dynamic range and low sensitivity. When it was switched from the rigid mode to the soft mode, the onset sensitivity of the tunable sensor increased from 0.96 kPa^{-1} to 15.77 kPa^{-1} with a tuning ratio of 18.3, compromising on the dynamic range from 1 MPa to 80 kPa. In this study, the adoption of a gallium microdroplet-elastomer composite for the construction of the transformative active layer enabled homogenous sensing across the entire sensing area [49]. In a demonstration experiment, the tunable pressure sensor was able to sense heavy loads generated by foot stepping in rigid mode and could be reconfigured to soft mode to capture sophisticated pressure generated by carotid arterial blood pulsation.

5. Challenges and outlook

TESs, based on stimuli-responsive materials, a mechanics concept of variable stiffness, and advanced manufacturing processes, are a new class of electronics that can possibly break the boundary between traditional rigid electronics and emerging soft devices. It holds immense potential to bring new opportunities for consumer electronics, biomedical devices, sensors, and robotics. To advance TES technology for practical utility, further research and development efforts need to be made. First of all, investigation of new stimuli-responsive materials is necessary to overcome the limitations of currently available ones, such as the large power requirement of gallium and LMPAs for phase transition and the small modulus variation of SMPs and field-activated materials (Fig. 2). One major cause of large power consumption in TESs constructed with gallium or LMPAs is supercooling [98], which makes them require excessive energy and time to initiate phase transition from liquid to solid. One solution would be to add nucleating agents to minimize the degree of supercooling, thereby lowering the energy requirement for TES mode conversion [99,100]. Similarly, by dispersing and incorporating nanoparticles chemically or physically into SMPs and field-activated materials, the relatively small stiffness-tuning range presented by TESs based on SMPs or field-activated materials can be improved [101–103]. Another essential aspect for future TES technology is to enable standalone, compact designs to make these systems easily portable and biointegratable. Integration of flexible thermoelectric devices [65] (for thermally responsive TESs) or electric/magnetic field actuators [104,105] (for field-responsive TESs) would facilitate such designs. In addition, it would enable rapid mechanical transformation for TESs. For biomedical applications, the biologically safe operation of TESs should be ensured in wearable [31,47,49,65,75,76] and implantable [49,79–82] devices. Silicone elastomers (e.g. PDMS [19,106], Ecoflex [107], and Silbione) and other flexible polymers (e.g. Parylene and polyimide) [108,109] can be used as compliant packaging materials for TESs to provide biocompatible interface with biological tissues [1]. However, these encapsulating materials still hold the potential of long-term foreign-body responses such as inflammation and fibrous-tissue formation. Applying new biomaterial composites [110] or coating the surface with biological polymers [111,112] can improve the biocompatibility over long periods of time. Moreover, robust yet compliant packaging is desired to securely contain embedded stimuli-responsive materials and to support electronics operation both in rigid and soft modes. Silicone elastomers provide sufficient flexibility but need improvement in their toughness. For example, the poor tear strength (~3 N/mm) of PDMS makes it unsuitable for long-term practical use. For this purpose, further investigation is needed on tough composite materials [113] and polymers [114] to replace them. Such innovative future technology developments addressing these challenges will enable the realization of highly versatile TESs that can go far beyond what existing electronics technology can offer to users.

Funding

This work was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (NRF-2018R1C1B6001706 and NRF-2018025230).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The images in Figs. 1 and 3–5 are reproduced with permission from the references cited in the figure captions.

References

[1] H. Lim, H.S. Kim, R. Qazi, Y. Kwon, J. Jeong, W. Yeo, Advanced soft materials, sensor integrations, and applications of wearable flexible hybrid electronics in healthcare, energy, and environment, Adv. Mater. 32 (2020), 1901924.
[2] S. Zhang, S. Li, Z. Xia, K. Cai, A review of electronic skin: soft electronics and sensors for human health, J. Mater. Chem. B. 8 (2020) 852–862.
[3] Y. Liu, J.J.S. Norton, R. Qazi, Z. Zou, K.R. Ammann, H. Liu, L. Yan, P.L. Tran, K.-I. Jang, J.W. Lee, D. Zhang, K.A. Kilian, S.H. Jung, T. Bretl, J. Xiao, M.J. Slepian, Y. Huang, J.-W. Jeong, J.A. Rogers, Epidermal mechano-acoustic sensing electronics for cardiovascular diagnostics and human-machine interfaces, Sci. Adv. 2 (2016) e1601185.
[4] J.-W. Jeong, W.-H. Yeo, A. Akhtar, J.J.S. Norton, Y.-J. Kwack, S. Li, S.-Y. Jung, Y. Su, W. Lee, J. Xia, H. Cheng, Y. Huang, W.-S. Choi, T. Bretl, J.A. Rogers,

Materials and optimized designs for human-machine interfaces via epidermal electronics, Adv. Mater. 25 (2013) 6839–6846.
[5] J.-W. Jeong, G. Shin, S.I. Park, K.J. Yu, L. Xu, J.A. Rogers, Soft materials in neuroengineering for hard problems in neuroscience, Neuron 86 (2015) 175–186.
[6] J.Y. Sim, M.P. Haney, S.I. Park, J.G. McCall, J.-W. Jeong, Microfluidic neural probes: in vivo tools for advancing neuroscience, Lab Chip 17 (2017) 1406–1435.
[7] R. Qazi, C.Y. Kim, S.-H. Byun, J.-W. Jeong, Microscale inorganic LED based wireless neural systems for chronic in vivo optogenetics, Front. Neurosci. 12 (2018) 764.
[8] L. Dejace, N. Laubeuf, I. Furfaro, S.P. Lacour, Gallium-based thin films for wearable human motion sensors, Adv. Intell. Syst. 1 (2019) 1900079.
[9] M.D. Bartlett, E.J. Markvicka, C. Majidi, Rapid fabrication of soft, multilayered electronics for wearable biomonitoring, Adv. Funct. Mater. 26 (2016) 8496–8504.
[10] D.-H. Kim, N. Lu, R. Ma, Y.-S. Kim, R.-H. Kim, S. Wang, J. Wu, S.M. Won, H. Tao, A. Islam, K.J. Yu, T. Kim, R. Chowdhury, M. Ying, L. Xu, M. Li, H.-J. Chung, H. Keum, M. McCormick, P. Liu, Y.-W. Zhang, F.G. Omenetto, Y. Huang, T. Coleman, J.A. Rogers, Epidermal Electronics, Science 333 (2011) 838.
[11] M. Kaltenbrunner, T. Sekitani, J. Reeder, T. Yokota, K. Kuribara, T. Tokuhara, M. Drack, R. Schwödiauer, I. Graz, S. Bauer-Gogonea, S. Bauer, T. Someya, An ultra-lightweight design for imperceptible plastic electronics, Nature 499 (2013) 458–463.
[12] S. Wang, J. Xu, W. Wang, G.-J.N. Wang, R. Rastak, F. Molina-Lopez, J.W. Chung, S. Niu, V.R. Feig, J. Lopez, T. Lei, S.-K. Kwon, Y. Kim, A.M. Foudeh, A. Ehrlich, A. Gasperini, Y. Yun, B. Murmann, J.B.-H. Tok, Z. Bao, Skin electronics from scalable fabrication of an intrinsically stretchable transistor array, Nature 555 (2018) 83–88.
[13] K.-I. Jang, K. Li, H.U. Chung, S. Xu, H.N. Jung, Y. Yang, J.W. Kwak, H.H. Jung, J. Song, C. Yang, A. Wang, Z. Liu, J.Y. Lee, B.H. Kim, J.-H. Kim, J. Lee, Y. Yu, B.J. Kim, H. Jang, K.J. Yu, J. Kim, J.W. Lee, J.-W. Jeong, Y.M. Song, Y. Huang, Y. Zhang, J.A. Rogers, Self-assembled three dimensional network designs for soft electronics, Nat. Commun. 8 (2017) 15894.
[14] G. Constantinescu, J.-W. Jeong, X. Li, D.K. Scott, K.-I. Jang, H.-J. Chung, J.A. Rogers, J. Rieger, Epidermal electronics for electromyography: an application to swallowing therapy, Med. Eng. Phys. 38 (2016) 807–812.
[15] Y. Song, J. Min, W. Gao, Wearable and implantable electronics: moving toward precision therapy, ACS Nano 13 (2019) 12280–12286.
[16] G. Schiavone, F. Fallegger, X. Kang, B. Barra, N. Vachicouras, E. Roussinova, I. Furfaro, S. Jiguet, I. Seáñez, S. Borgognon, A. Rowald, Q. Li, C. Qin, E. Bézard, J. Bloch, G. Courtine, M. Capogrosso, S.P. Lacour, Soft, implantable bioelectronic interfaces for translational research, Adv. Mater. (2020), 1906512.
[17] L. Xu, S.R. Gutbrod, A.P. Bonifas, Y. Su, M.S. Sulkin, N. Lu, H.-J. Chung, K.-I. Jang, Z. Liu, M. Ying, C. Lu, R.C. Webb, J.-S. Kim, J.I. Laughner, H. Cheng, Y. Liu, A. Ameen, J.-W. Jeong, G.-T. Kim, Y. Huang, I.R. Efimov, J.A. Rogers, 3D multifunctional integumentary membranes for spatiotemporal cardiac measurements and stimulation across the entire epicardium, Nat. Commun. 5 (2014) 3329.
[18] R. Qazi, A.M. Gomez, D.C. Castro, Z. Zou, J.Y. Sim, Y. Xiong, J. Abdo, C.Y. Kim, A. Anderson, F. Lohner, S.-H. Byun, B. Chul Lee, K.-I. Jang, J. Xiao, M.R. Bruchas, J.-W. Jeong, Wireless optofluidic brain probes for chronic neuropharmacology and photostimulation, Nat. Biomed. Eng. 3 (2019) 655–669.
[19] I.R. Minev, P. Musienko, A. Hirsch, Q. Barraud, N. Wenger, E.M. Moraud, J. Gandar, M. Capogrosso, T. Milekovic, L. Asboth, R.F. Torres, N. Vachicouras, Q. Liu, N. Pavlova, S. Duis, A. Larmagnac, J. Voros, S. Micera, Z. Suo, G. Courtine, S.P. Lacour, Electronic dura mater for long-term multimodal neural interfaces, Science 347 (2015) 159–163.
[20] K.N. Noh, S.I. Park, R. Qazi, Z. Zou, A.D. Mickle, J.G. Grajales-Reyes, K.-I. Jang, R.W. Gereau, J. Xiao, J.A. Rogers, J.-W. Jeong, Miniaturized, battery-free optofluidic systems with potential for wireless pharmacology and optogenetics, Small 14 (2018), 1702479.
[21] C. Howe, S. Mishra, Y.-S. Kim, Y. Chen, S.-H. Ye, W.R. Wagner, J.-W. Jeong, H.-S. Byun, J.-H. Kim, Y. Chun, W.-H. Yeo, Stretchable, implantable, nanostructured flow-diverter system for quantification of intra-aneurysmal hemodynamics, ACS Nano 12 (2018) 8706–8716.
[22] E.J. Markvicka, M.D. Bartlett, X. Huang, C. Majidi, An autonomously electrically self-healing liquid metal–elastomer composite for robust soft-matter robotics and electronics, Nat. Mater. 17 (2018) 618–624.
[23] F. Schmitt, O. Piccin, L. Barbé, B. Bayle, Soft robots manufacturing: a review, Front. Robot. AI. 5 (2018) 84.
[24] X. Wen, B. Wang, S. Huang, T. "Leo" Liu, M.-S. Lee, P.-S. Chung, Y.T. Chow, I.-W. Huang, H.G. Monbouquette, N.T. Maidment, P.-Y. Chiou, Flexible, multifunctional neural probe with liquid metal enabled, ultra-large tunable stiffness for deep-brain chemical sensing and agent delivery, Biosens. Bioelectron. 131 (2019) 37–45.
[25] S. Park, N. Baugh, H.K. Shah, D.P. Parekh, I.D. Joshipura, M.D. Dickey, Ultra-stretchable elastic shape memory fibers with electrical conductivity, Adv. Sci. 6 (2019), 1901579.
[26] R. Poon, J.B. Hopkins, Phase-changing metamaterial capable of variable stiffness and shape morphing, Adv. Eng. Mater. 21 (2019), 1900802.
[27] T.L. Buckner, M.C. Yuen, S.Y. Kim, R. Kramer-Bottiglio, Enhanced variable stiffness and variable stretchability enabled by phase-changing particulate additives, Adv. Funct. Mater. 29 (2019), 1903368.
[28] Y. Hao, T. Wang, Z. Xie, W. Sun, Z. Liu, X. Fang, M. Yang, L. Wen, A eutectic-alloy-infused soft actuator with sensing, tunable degrees of freedom, and stiffness properties, J. Micromech. Microeng. 28 (2018), 024004.
[29] D. Zappetti, S.H. Jeong, J. Shintake, D. Floreano, Phase changing materials-based variable-stiffness tensegrity structures, Soft Robot. 7 (3) (2020) 362–369.
[30] F. Alambeigi, R. Seifabadi, M. Armand, A continuum manipulator with phase changing alloy, in: 2016 IEEE International Conference on Robotics and Automation (ICRA), 2016, pp. 758–764.
[31] A. Tonazzini, S. Mintchev, B. Schubert, B. Mazzolai, J. Shintake, D. Floreano, Variable stiffness fiber with self-healing capability, Adv. Mater. 28 (2016) 10142–10148.
[32] B.E. Schubert, D. Floreano, Variable stiffness material based on rigid low-melting-point-alloy microstructures embedded in soft poly(-dimethylsiloxane) (PDMS), RSC Adv. 3 (2013) 24671.
[33] I.M. Van Meerbeek, B.C. Mac Murray, J.W. Kim, S.S. Robinson, P.X. Zou, M.N. Silberstein, R.F. Shepherd, Morphing metal and elastomer bicontinuous foams for reversible stiffness, shape memory, and self-healing soft machines, Adv. Mater. 28 (2016) 2801–2806.
[34] S. Yoshida, Y. Morimoto, L. Zheng, H. Onoe, S. Takeuchi, Multipoint bending and shape retention of a pneumatic bending actuator by a variable stiffness endoskeleton, Soft Robot. 5 (2018) 718–725.
[35] Y. Yang, Y. Chen, Y. Li, M.Z.Q. Chen, Y. Wei, Bioinspired robotic fingers based on pneumatic actuator and 3D printing of smart material, Soft Robot. 4 (2017) 147–162.
[36] Y.-F. Zhang, N. Zhang, H. Hingorani, N. Ding, D. Wang, C. Yuan, B. Zhang, G. Gu, Q. Ge, Fast-response, stiffness-tunable soft actuator by hybrid multi-material 3D printing, Adv. Funct. Mater. 29 (2019), 1806698.
[37] C. Yang, M. Boorugu, A. Dopp, J. Ren, R. Martin, D. Han, W. Choi, H. Lee, 4D printing reconfigurable, deployable and mechanically tunable metamaterials, Mater. Horiz. 6 (2019) 1244–1250.
[38] W. Wang, C.Y. Yu, P.A. Abrego Serrano, S.-H. Ahn, Shape memory alloy-based soft finger with changeable bending length using targeted variable stiffness, Soft Robot. 7 (3) (2020) 283–291.
[39] Y. Yang, Y. Chen, Y. Li, Z. Wang, Y. Li, Novel variable-stiffness robotic fingers with built-in position feedback, Soft Robot. 4 (2017) 338–352.
[40] F. Pilate, R. Mincheva, J. De Winter, P. Gerbaux, L. Wu, R. Todd, J.-M. Raquez, P. Dubois, Design of multistimuli-responsive shape-memory polymer materials by reactive extrusion, Chem. Mater. 26 (2014) 5860–5867.
[41] I. Must, E. Sinibaldi, B. Mazzolai, A variable-stiffness tendril-like soft robot based on reversible osmotic actuation, Nat. Commun. 10 (2019) 344.
[42] W.-B. Li, W.-M. Zhang, H.-X. Zou, Z.-K. Peng, G. Meng, Bioinspired variable stiffness dielectric elastomer actuators with large and tunable load capacity, Soft Robot. 6 (2019) 631–643.
[43] H. Imamura, K. Kadooka, M. Taya, A variable stiffness dielectric elastomer actuator based on electrostatic chucking, Soft Matter 13 (2017) 3440–3448.
[44] W. Shan, S. Diller, A. Tutcuoglu, C. Majidi, Rigidity-tuning conductive elastomer, Smart Mater. Struct. 24 (2015), 065001.
[45] J.A. Jackson, M.C. Messner, N.A. Dudukovic, W.L. Smith, L. Bekker, B. Moran, A.M. Golobic, A.J. Pascall, E.B. Duoss, K.J. Loh, C.M. Spadaccini, Field responsive mechanical metamaterials, Sci. Adv. 4 (2018) eaau6419.
[46] S. Lee, J. Lee, S.H. Hwang, J. Yun, J. Jang, Enhanced electroresponsive performance of double-shell SiO_2/TiO_2 hollow nanoparticles, ACS Nano 9 (2015) 4939–4949.
[47] A. Coon, T.-H. Yang, Y.-M. Kim, H. Kang, J.-H. Koo, Application of magnetorheological fluids for investigating the effect of skin properties on arterial tonometry measurements, Front. Mater. 6 (2019) 45.
[48] B.S. Chang, R. Tutika, J. Cutinho, S. Oyola-Reynoso, J. Chen, M.D. Bartlett, M.M. Thuo, Mechanically triggered composite stiffness tuning through thermodynamic relaxation (ST3R), Mater. Horiz. 5 (2018) 416–422.
[49] S.-H. Byun, J.Y. Sim, Z. Zhou, J. Lee, R. Qazi, M.C. Walicki, K.E. Parker, M.P. Haney, S.H. Choi, A. Shon, G.B. Gereau, J. Bilbily, S. Li, Y. Liu, W.-H. Yeo, J.G. McCall, J. Xiao, J.-W. Jeong, Mechanically transformative electronics, sensors, and implantable devices, Sci. Adv. 5 (2019) eaay0418.
[50] H. Gao, J. Li, F. Zhang, Y. Liu, J. Leng, The research status and challenges of shape memory polymer-based flexible electronics, Mater. Horiz. 6 (2019) 931–944.
[51] L. Zhu, B. Wang, S. Handschuh-Wang, X. Zhou, Liquid metal–based soft microfluidics, Small 16 (2020), 1903841.
[52] J. Yan, Y. Lu, G. Chen, M. Yang, Z. Gu, Advances in liquid metals for biomedical applications, Chem. Soc. Rev. 47 (2018) 2518–2533.
[53] Y. Yu, E. Miyako, Recent advances in liquid metal manipulation toward soft robotics and biotechnologies, Chem. Eur J. 24 (2018) 9456–9462.
[54] L. Wang, Y. Yang, Y. Chen, C. Majidi, F. Iida, E. Askounis, Q. Pei, Controllable and reversible tuning of material rigidity for robot applications, Materials, Today Off. 21 (2018) 563–576.
[55] M. Manti, V. Cacucciolo, M. Cianchetti, Stiffening in soft robotics: a review of the state of the art, IEEE Robot. Autom. Mag. 23 (2016) 93–106.
[56] L. Blanc, A. Delchambre, P. Lambert, Flexible medical devices: review of controllable stiffness solutions, Actuators 6 (2017) 23.
[57] L. Zheng, S. Yoshida, Y. Morimoto, H. Onoe, S. Takeuchi, Pneumatic balloon actuator with tunable bending points, in: 2015 28th IEEE International Conference on Micro Electro Mechanical Systems (MEMS), 2015, pp. 18–21.
[58] S. Rich, S.-H. Jang, Y.-L. Park, C. Majidi, Liquid metal-conductive thermoplastic elastomer integration for low-voltage stiffness tuning, Adv. Mater. Technol. 2 (2017).

[59] A.M. Nasab, A. Sabzehzar, M. Tatari, C. Majidi, W. Shan, A soft gripper with rigidity tunable elastomer strips as ligaments, Soft Robot. 4 (2017) 411–420.
[60] M. Kukla, J. Górecki, I. Malujda, K. Talaśka, P. Tarkowski, The determination of mechanical properties of magnetorheological elastomers (MREs), Procedia Eng. 177 (2017) 324–330.
[61] C.W. Lee, I.-H. Kim, H.-J. Jung, Fabrication and characterization of natural rubber-based magnetorheological elastomers at large strain for base isolators, Shock Vib. 2018 (2018) e7434536.
[62] C. Majidi, R.J. Wood, Tunable elastic stiffness with microconfined magnetorheological domains at low magnetic field, Appl. Phys. Lett. 97 (2010), 164104.
[63] Q. Ge, A.H. Sakhaei, H. Lee, C.K. Dunn, N.X. Fang, M.L. Dunn, Multimaterial 4D printing with tailorable shape memory polymers, Sci. Rep. 6 (2016), 31110.
[64] J. Wu, C. Yuan, Z. Ding, M. Isakov, Y. Mao, T. Wang, M.L. Dunn, H.J. Qi, Multi-shape active composites by 3D printing of digital shape memory polymers, Sci. Rep. 6 (2016), 24224.
[65] C.W. Carpenter, S.T.M. Tan, C. Keef, K. Skelil, M. Malinao, D. Rodriquez, M.A. Alkhadra, J. Ramírez, D.J. Lipomi, Healable thermoplastic for kinesthetic feedback in wearable haptic devices, Sensor Actuator A Phys. 288 (2019) 79–85.
[66] J. Shintake, B. Schubert, S. Rosset, H. Shea, D. Floreano, Variable stiffness actuator for soft robotics using dielectric elastomer and low-melting-point alloy, in: 2015 IEEE/RSJ International Conference on Intelligent Robots and Systems (IROS), 2015, pp. 1097–1102.
[67] A. Orita, MarkR. Cutkosky, Scalable electroactive polymer for variable stiffness suspensions, IEEE ASME Trans. Mechatron. 21 (2016) 2836–2846.
[68] Y. Gao, H. Ota, E.W. Schaler, K. Chen, A. Zhao, W. Gao, H.M. Fahad, Y. Leng, A. Zheng, F. Xiong, C. Zhang, L.-C. Tai, P. Zhao, R.S. Fearing, A. Javey, Wearable microfluidic diaphragm pressure sensor for health and tactile touch monitoring, Adv. Mater. 29 (2017), 1701985.
[69] M.A. Eddings, M.A. Johnson, B.K. Gale, Determining the optimal PDMS–PDMS bonding technique for microfluidic devices, J. Micromech. Microeng. 18 (2008), 067001.
[70] Yong-Lae Park, Bor-Rong Chen, R.J. Wood, Design and fabrication of soft artificial skin using embedded microchannels and liquid conductors, IEEE Sensor. J. 12 (2012) 2711–2718.
[71] Y. Hao, Z. Liu, Z. Xie, X. Fang, T. Wang, L. Wen, A variable degree-of-freedom and self-sensing soft bending actuator based on conductive liquid metal and thermoplastic polymer composites, in: 2018 IEEE/RSJ International Conference on Intelligent Robots and Systems (IROS), IEEE, Madrid, 2018, pp. 1–9.
[72] Q. Meng, J. Hu, Self-organizing alignment of carbon nanotube in shape memory segmented fiber prepared by in situ polymerization and melt spinning, Compos. Appl. Sci. Manuf. 39 (2008) 314–321.
[73] A. Fassler, C. Majidi, 3D structures of liquid-phase GaIn alloy embedded in PDMS with freeze casting, Lab Chip 13 (2013) 4442.
[74] A. Fassler, C. Majidi, Liquid-phase metal inclusions for a conductive polymer composite, Adv. Mater. 27 (2015) 1928–1932.
[75] V. Ramachandran, J. Shintake, D. Floreano, All-fabric wearable electro-adhesive clutch, Adv. Mater. Technol. 4 (2019), 1800313.
[76] T.P. Chenal, J.C. Case, J. Paik, R.K. Kramer, Variable stiffness fabrics with embedded shape memory materials for wearable applications, in: 2014 IEEE/RSJ International Conference on Intelligent Robots and Systems, IEEE, Chicago, IL, USA, 2014, pp. 2827–2831.
[77] J.-W. Jeong, M.K. Kim, H. Cheng, W.-H. Yeo, X. Huang, Y. Liu, Y. Zhang, Y. Huang, J.A. Rogers, Capacitive epidermal electronics for electrically safe, long-term electrophysiological measurements, Adv. Healthc. Mater. 3 (2014) 642–648.
[78] K.-I. Jang, H.N. Jung, J.W. Lee, S. Xu, Y.H. Liu, Y. Ma, J.-W. Jeong, Y.M. Song, J. Kim, B.H. Kim, A. Banks, J.W. Kwak, Y. Yang, D. Shi, Z. Wei, X. Feng, U. Paik, Y. Huang, R. Ghaffari, J.A. Rogers, Ferromagnetic, folded electrode composite as a soft interface to the skin for long-term electrophysiological recording, Adv. Funct. Mater. 26 (2016) 7281–7290.
[79] M.A. González-González, A. Kanneganti, A. Joshi-Imre, A.G. Hernandez-Reynoso, G. Bendale, R. Modi, M. Ecker, A. Khurram, S.F. Cogan, W.E. Voit, M.I. Romero-Ortega, Thin film multi-electrode softening cuffs for selective neuromodulation, Sci. Rep. 8 (2018), 16390.
[80] T. Ware, D. Simon, C. Liu, T. Musa, S. Vasudevan, A. Sloan, E.W. Keefer, R.L. Rennaker, W. Voit, Thiol-ene/acrylate substrates for softening intracortical electrodes: substrates for intracortical electrodes, J. Biomed. Mater. Res. 102 (2014) 1–11.
[81] D.M. Simon, H. Charkhkar, C. St John, S. Rajendran, T. Kang, R. Reit, D. Arreaga-Salas, D.G. McHail, G.L. Knaack, A. Sloan, D. Grasse, T.C. Dumas, R.L. Rennaker, J.J. Pancrazio, W.E. Voit, Design and demonstration of an intracortical probe technology with tunable modulus: design and demonstration of an intracortical probe technology, J. Biomed. Mater. Res. 105 (2017) 159–168.
[82] A. Zátonyi, G. Orbán, R. Modi, G. Márton, D. Meszéna, I. Ulbert, A. Pongrácz, M. Ecker, W.E. Voit, A. Joshi-Imre, Z. Fekete, A softening laminar electrode for recording single unit activity from the rat hippocampus, Sci. Rep. 9 (2019) 2321.
[83] S. Kim, C. Laschi, B. Trimmer, Soft robotics: a bioinspired evolution in robotics, Trends Biotechnol. 31 (2013) 287–294.
[84] P. Polygerinos, N. Correll, S.A. Morin, B. Mosadegh, C.D. Onal, K. Petersen, M. Cianchetti, M.T. Tolley, R.F. Shepherd, Soft robotics: review of fluid-driven intrinsically soft devices; manufacturing, sensing, control, and applications in human-robot interaction, Adv. Eng. Mater. 19 (2017), 1700016.
[85] H.K. Yap, H.Y. Ng, C.-H. Yeow, High-force soft printable pneumatics for soft robotic applications, Soft Robot. 3 (2016) 144–158.
[86] P. Polygerinos, Z. Wang, K.C. Galloway, R.J. Wood, C.J. Walsh, Soft robotic glove for combined assistance and at-home rehabilitation, Robot. Autonom. Syst. 73 (2015) 135–143.
[87] J. Kwon, S.J. Yoon, Y.-L. Park, Flat inflatable Artificial muscles with large stroke and adjustable force– length relations, IEEE Trans. Robot. (2020) 1–14.
[88] T. Ranzani, G. Gerboni, M. Cianchetti, A. Menciassi, A bioinspired soft manipulator for minimally invasive surgery, Bioinspiration Biomimetics 10 (2015), 035008.
[89] G. Palli, G. Berselli, On the control of a dielectric elastomer artificial muscle with variable impedance, in: American Society of Mechanical Engineers Digital Collection, 2014.
[90] F. Carpi, G. Frediani, C. Gerboni, J. Gemignani, D. De Rossi, Enabling variable-stiffness hand rehabilitation orthoses with dielectric elastomer transducers, Med. Eng. Phys. 36 (2014) 205–211.
[91] C.M. Boutry, Y. Kaizawa, B.C. Schroeder, A. Chortos, A. Legrand, Z. Wang, J. Chang, P. Fox, Z. Bao, A stretchable and biodegradable strain and pressure sensor for orthopaedic application, Nat. Electron. 1 (2018) 314–321.
[92] J.C. Yang, J. Mun, S.Y. Kwon, S. Park, Z. Bao, S. Park, Electronic skin: recent progress and future prospects for skin-attachable devices for health monitoring, Robot. Prosthet. Adv. Mater. 31 (2019), 1904765.
[93] K. Meng, J. Chen, X. Li, Y. Wu, W. Fan, Z. Zhou, Q. He, X. Wang, X. Fan, Y. Zhang, J. Yang, Z.L. Wang, Flexible weaving constructed self-powered pressure sensor enabling continuous diagnosis of cardiovascular disease and measurement of cuffless blood pressure, Adv. Funct. Mater. 29 (2019), 1806388.
[94] G.-H. Lee, J.-K. Park, J. Byun, J.C. Yang, S.Y. Kwon, C. Kim, C. Jang, J.Y. Sim, J.-G. Yook, S. Park, Parallel signal processing of a wireless pressure-sensing platform combined with machine-learning-based cognition, inspired by the human somatosensory system, Adv. Mater. (2019) n/a 1906269.
[95] J.C. Yang, J.-O. Kim, J. Oh, S.Y. Kwon, J.Y. Sim, D.W. Kim, H.B. Choi, S. Park, Microstructured porous pyramid-based ultrahigh sensitive pressure sensor insensitive to strain and temperature, ACS Appl. Mater. Interfaces 11 (2019) 19472–19480.
[96] W.-C. Lai, M.-Y. Lin, Y.-C. Lee, W. Fang, In-process and in-use modulation of sensitivity and sensing range for CMOS-MEMS tactile sensor with dielectric PDMS nanocomposite, in: 2019 20th International Conference on Solid-State Sensors, Actuators and Microsystems & Eurosensors XXXIII (TRANSDUCERS & EUROSENSORS XXXIII), IEEE, Berlin, Germany, 2019, pp. 2523–2526.
[97] J. Zárate, T. Helbig, H. Witte, Design of a bioinspired variable stiffness sensor, in: 2019 IEEE International Conference on Mechatronics (ICM), 2019, pp. 376–381.
[98] V.B. Kumar, Z. Porat, A. Gedanken, DSC measurements of the thermal properties of gallium particles in the micron and sub-micron sizes, obtained by sonication of molten gallium, J. Therm. Anal. Calorim. 119 (2015) 1587–1592.
[99] R. Al-Shannaq, J. Kurdi, S. Al-Muhtaseb, M. Dickinson, M. Farid, Supercooling elimination of phase change materials (PCMs) microcapsules, Energy 87 (2015) 654–662.
[100] C. Zhang, L. Li, X. Yang, J. Shi, L. Gui, J. Liu, Study on the nucleating agents for gallium to reduce its supercooling, Int. J. Heat Mass Tran. 148 (2020) 119055.
[101] E. Wisse, L.E. Govaert, H.E.H. Meijer, E.W. Meijer, Unusual tuning of mechanical properties of thermoplastic elastomers using supramolecular fillers, Macromolecules 39 (2006) 7425–7432.
[102] J.T. Kim, H.J. Jeong, H.C. Park, H.M. Jeong, S.Y. Bae, B.K. Kim, Electroactive shape memory performance of polyurethane/graphene nanocomposites, React. Funct. Polym. 88 (2015) 1–7.
[103] T. Hu, S. Xuan, L. Ding, X. Gong, Stretchable and magneto-sensitive strain sensor based on silver nanowire-polyurethane sponge enhanced magnetorheological elastomer, Mater. Des. 156 (2018) 528–537.
[104] S. Sun, J. Yang, W. Li, H. Deng, H. Du, G. Alici, Development of a novel variable stiffness and damping magnetorheological fluid damper, Smart Mater. Struct. 24 (2015), 085021.
[105] Y. Yang, Y. Wu, C. Li, X. Yang, W. Chen, Flexible Actuators for Soft Robotics, Adv. Intell. Syst. 2 (2020), 1900077.
[106] J.-W. Jeong, J.G. McCall, G. Shin, Y. Zhang, R. Al-Hasani, M. Kim, S. Li, J.Y. Sim, K.-I. Jang, Y. Shi, D.Y. Hong, Y. Liu, G.P. Schmitz, L. Xia, Z. He, P. Gamble, W.Z. Ray, Y. Huang, M.R. Bruchas, J.A. Rogers, Wireless optofluidic systems for programmable in vivo pharmacology and optogenetics, Cell 162 (2015) 662–674.
[107] G. Park, H.-J. Chung, K. Kim, S.A. Lim, J. Kim, Y.-S. Kim, Y. Liu, W.-H. Yeo, R.-H. Kim, S.S. Kim, J.-S. Kim, Y.H. Jung, T. Kim, C. Yee, J.A. Rogers, K.-M. Lee, Immunologic and tissue biocompatibility of flexible/stretchable electronics and optoelectronics, Adv. Healthc. Mater. 3 (2014) 515–525.
[108] R. Herbert, J.-H. Kim, Y. Kim, H. Lee, W.-H. Yeo, Soft material-enabled, flexible hybrid electronics for medicine, healthcare, and human-machine interfaces, Materials 11 (2018) 187.
[109] Y. Sun, S.P. Lacour, R.A. Brooks, N. Rushton, J. Fawcett, R.E. Cameron, Assessment of the biocompatibility of photosensitive polyimide for implantable medical device use, J. Biomed. Mater. Res. 90A (2009) 648–655.

[110] H. Yuk, B. Lu, X. Zhao, Hydrogel bioelectronics, Chem. Soc. Rev. 48 (2019) 1642–1667.
[111] A. Mahapatro, K. Jensen, S.-Y. Yang, Effect of polymer coating characteristics on the biodegradation and biocompatibility behavior of magnesium alloy, Polym. Plast. Technol. Mater. 59 (2020) 301–310.
[112] W.-J. Cherng, Y.-H. Pan, T.-C. Wu, C.-C. Chou, C.-H. Yeh, J.-J. Ho, Hemocompatibility and adhesion of heparin/dopamine and heparin/collagen self-assembly multilayers coated on a titanium substrate, Appl. Surf. Sci. 463 (2019) 732–740.
[113] N. Kazem, M.D. Bartlett, C. Majidi, Extreme toughening of soft materials with liquid metal, Adv. Mater. 30 (2018), 1706594.
[114] S. Zhu, J.-H. So, R. Mays, S. Desai, W.R. Barnes, B. Pourdeyhimi, M.D. Dickey, Ultrastretchable fibers with metallic conductivity using a liquid metal alloy core, Adv. Funct. Mater. 23 (2013) 2308–2314.
[115] A. Garcia-Sandoval, A. Pal, A.M. Mishra, S. Sherman, A.R. Parikh, A. Joshi-Imre, D. Arreaga-Salas, G. Gutierrez-Heredia, A.C. Duran-Martinez, J. Nathan, S.M. Hosseini, J.B. Carmel, W. Voit, Chronic softening spinal cord stimulation arrays, J. Neural. Eng. 15 (2018), 045002.
[116] J. Kaursoin, A.K. Agrawal, Melt spun thermoresponsive shape memory fibers based on polyurethanes: effect of drawing and heat-setting on fiber morphology and properties, J. Appl. Polym. Sci. 103 (2007) 2172–2182.
[117] M.D. Dickey, R.C. Chiechi, R.J. Larsen, E.A. Weiss, D.A. Weitz, G.M. Whitesides, Eutectic gallium-indium (EGaIn): a liquid metal alloy for the formation of stable structures in microchannels at room temperature, Adv. Funct. Mater. 18 (2008) 1097–1104.
[118] R. Tutika, S. Kmiec, A.B.M.T. Haque, S.W. Martin, M.D. Bartlett, Liquid metal–elastomer soft composites with independently controllable and highly tunable droplet size and volume loading, ACS Appl. Mater. Interfaces 11 (2019) 17873–17883.
[119] S.H. Jeong, S. Chen, J. Huo, E.K. Gamstedt, J. Liu, S.-L. Zhang, Z.-B. Zhang, K. Hjort, Z. Wu, Mechanically stretchable and electrically insulating thermal elastomer composite by liquid alloy droplet embedment, Sci. Rep. 5 (2016), 18257.
[120] Y. Lin, J. Genzer, W. Li, R. Qiao, M.D. Dickey, S.-Y. Tang, Sonication-enabled rapid production of stable liquid metal nanoparticles grafted with poly(1-octadecene- *alt* -maleic anhydride) in aqueous solutions, Nanoscale 10 (2018) 19871–19878.
[121] S.-Y. Tang, R. Qiao, S. Yan, D. Yuan, Q. Zhao, G. Yun, T.P. Davis, W. Li, Microfluidic mass production of stabilized and stealthy liquid metal nanoparticles, Small 14 (2018), 1800118.
[122] J. Thelen, M.D. Dickey, T. Ward, A study of the production and reversible stability of EGaIn liquid metal microspheres using flow focusing, Lab Chip 12 (2012) 3961.
[123] S.-Y. Tang, I.D. Joshipura, Y. Lin, K. Kalantar-Zadeh, A. Mitchell, K. Khoshmanesh, M.D. Dickey, Liquid-metal microdroplets formed dynamically with electrical control of size and rate, Adv. Mater. 28 (2016) 604–609.