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Localized modulus-controlled PDMS substrate for 2D and 3D stretchable electronics

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Abstract

Stretchable electronics have great importance for applications of wearable devices and electronic skin. Balancing and improving mechanical stretchability and electronic performance are the key challenges that restrict further development of stretchable electronics. In order to achieve stretchable electronics, it is crucial to choose appropriate substrates, among which PDMS is the most commonly used polymer due to its easy fabrication and low cost. In this paper, we propose a novel strategy and fabricate localized modulus-controlled PDMS for both 2D and 3D stretchable electronics. Based on a secondary cross-link effect, the modulus of cured PDMS can be enhanced and controlled by spin-coating different masses of curing agent. Using a laser-cut PI mask, the modulus-enhanced region can be defined by users. Through this simple method, the functional conductive thin-film materials (gold/Ag nanowires/reduced graphene oxide) can be well protected when the structural layer is stretched and the 'barrel effect' of a multi-material film (where different material films possess different stretchability) on one piece of substrate can also be solved. Besides this, compared with uniform PDMS, different 3D buckling structures can be formed on locally modified PDMS as a substrate by a pre-stretching and releasing process, illustrating a new way to control the 3D buckling structure.

Keywords: PDMS, stretchable conductive films, stretchable 3D structures, local modulus-controlling

 Supplementary material for this article is available [online](#)

(Some figures may appear in colour only in the online journal)

1. Introduction

With the development of wearable electronics [1–3] and bioelectronics [4–6], designing and fabricating devices that can be stretched without simultaneous function failure is a meaningful challenge, and becoming the development trend for the next generation of intelligent electronics [7–9]. Due to its great significance, much research on this topic has been reported. Hong *et al* fabricated a metal nanowire percolation

network for wearable and transparent heaters [1]. Song *et al* designed a high-efficiency self-charging smart bracelet using Ecoflex substrate, which is lightweight and customized as a type of novel wearable electronics [3]. Han *et al* demonstrated a stretchable and flexible 3D structure on soft substrate and its potential application value in energy harvesting and biomedical researching [5]. A large number of advanced materials such as metal nanowires [1, 2], polymer substrates [3], carbon nanotubes [7], and advanced structures such as arch structures

[4] and porous structures [9] have been studied, and demonstrated stretchability and efficiency for electronic devices.

Among these works, the deposition of functional materials, such as conductive materials, directly onto a substrate to form a conductive film is one of the most common methods in the fabricating of stretchable electronics [10, 11]. However, most of the conductive films crack and become non-conductive when the substrate is stretched to a certain degree [12]. To avoid this problem, various strategies and methods concerning functional materials themselves (special patterns, manufacturing factors or other property improvements) or interaction of functional materials with substrate (depositing, hybrid or dispersing) have been proposed. Special patterns such as serpentine patterns have been designed and demonstrated which can ensure the conductivity under a large stretching strain [13]. Wrinkle structure as a quasi-3D structure also provides directional stretchability for conductive materials [14, 15], and can be well controlled [16] as reported by Rogel *et al*. In addition, fabricated 3D structures such as arc-shaped 3D bridges or 3D wavy metallic micro-wires are able to accommodate stretching by changing wavelength and amplitude [17, 18]. However, these technologies require specific processing for specific materials, which means complex fabrication processes and higher costs, and are not universal.

Since stretchable electronics usually require a stretchable substrate to support other kinds of electronics, and the soft region underneath the functional material or structure leads to the crack and failure of the device, it is important to locally modify the stretchable substrates so that they are more suitable for supporting protection for the functional materials or structures when being stretched [19–26].

Directly and locally embedding stiffer materials into the soft substrate to form a gradient of modulus is a type of technology that can effectively protect the stiff electronic device encapsulated in the substrate or on the surface [19–22]. Oh *et al* embedded Ni/silicone resin mixture into PDMS to form a core-shell structure which locally enhanced the modulus of the PDMS substrate [19]. As for the stiffer material, PDMS with a relatively higher modulus (compared with the substrate) is a more widely-used choice. Matsuhisa *et al* used a sacrificial layer in the fabrication process to simultaneously form a locally modulus-enhanced PDMS substrate for embedding organic thin film transistors [20], while Naserifar *et al* chose to etch a piece of PDMS with a high modulus and encapsulated this stiffer PDMS and the device with soft PDMS [21]. Similarly, Park *et al* fabricated an elastomeric substrate with a locally-tailored structure to protect transistors [22].

Moreover, splicing a polymer stripe with different moduli can realize three or more types of modulus distribution on one piece of polymer [23]. Wu *et al* mixed polyethylenimine ethoxylated (PEIE) with PDMS mixture and gained softer PDMS (S3PDMS). Before the curing process, pouring three types of modulated S3PDMS liquid into a space-separated mold, a stripe of locally modulus-controlled PDMS was fabricated.

In contrast to embedding external stiffer materials, local chemical treatment on the original soft substrate is another common method. Cao *et al* implemented local oxidation on the surface of a pristine film realizing extreme enhancement

of local modulus [24]. Liu *et al* proposed a local stiffening strategy through additional crosslinking on hydrogels for spatial modulation of hydrogel stiffness [25]. Assisted by 3D printing and controlling the mass of resins in a crosslink process, Kokkins *et al* proposed a novel strategy to achieve printed materials with tunable modulus gradient and tunable failure [26].

Compared with various processing technologies for different materials and structures, a locally modulus-controlled substrate is superior and more universal. However, these technologies for inhomogeneous modulus distribution for stretchable electronics still have some drawbacks. Directly and locally embedding a type of stiffer material into a soft substrate requires complex MEMS technology, such as photolithography [20] and etching [21], which also limits to some extent the application for stretchable electronics. 3D printing also needs to carefully control two kinds of resins and the velocity, volume and height to ensure the complete crosslink reaction [26]. Some methods influence the transparency of the original soft substrate as a result of the mixing of other materials such as Ni [19] or PEIE [22]. Besides this, the aforementioned methods for local modification of a substrate have not demonstrated the ability for various modulus controlling of the substrate, which is significant because of the unknown modulus of materials in or on the soft substrate [24, 25]. Compared with pristine film or hydrogel, PDMS, as one of the most widely used silicone-based polymers, has its unique advantages such as stretchability, transparency, chemical robustness and is non-toxic for stretchable electronics. Meanwhile, the crosslink effect in the fabrication process of PDMS also indicates the possibility of modifying its modulus by adjusting the mass ratio of prepolymer to curing agent [27].

Based on consideration of the problems mentioned above, here we report a simple method that can locally control the modulus of PDMS to improve the stretchability of directly fabricated 2D conductive film, or 3D structures on it. Without etching or lithography technology, the Young's modulus of the material can be controlled from 0.169 to 0.583 MPa by spin-coating more curing agent on the user-defined region of a cured PDMS following heating. The enhancement of the modulus of the specific region of PDMS can protect a 2D conductive film above when stretched. Moreover, defining the modulus of different regions on PDMS for different conductive materials can also effectively avoid the 'barrel effect', which can maximize the integral stretchability. More importantly, this localized modulus-controlled PDMS shows great ability for fabricating and controlling stretchable 3D structures by compressive buckling. In contrast to already reported works which focus on the design of a 2D precursor, this method of substrate modification is firstly proposed for buckling 3D structures.

2. Experiment

The processes for the experiment are as follows:

Fabrication of localized modulus-controlled PDMS: The original PDMS membrane is fabricated by mixing the prepolymer and curing agent of commercial PDMS (Sylgard 184, Dow Corning Corporation) with a quantity ratio of 20:1.

The mixture is heated at 65 °C for 4 h to form a cured easily stretchable PDMS film. The thickness of the PDMS is about 400 μm . The speed I and time I of spin-up for dispersing the curing agent are 500 rpm and 10 s, respectively. The speed II and time II of the spin-off process for reducing the height of film are 500–5000 rpm (controlled spacing: 500 rpm) and 30 s. After heating at 65 °C for a certain time (15, 30, 45, 60, 120, 180, 240 min), the PI mask is peeled off carefully.

Fabrication of gold film on PDMS: First, a 5 nm-thick adhesion interlayer of chromium is deposited. Then 100 nm-thick layers of gold are deposited on the top by a magnetron sputtering process, for about 20 min at 200 W sputtering power.

Fabrication process for the AgNWs film (or RGO film) on PDMS: Oxygen plasma treatment is carried out onto a defined region, to make its surface hydrophilic, by Corona (Electro-Technic Products, Inc. USA). After repeating a spray coating of ethanol solution of AgNWs (or isopropanol solution of RGO), the membrane is annealed at 100 °C for 10 min to enhance conductivity and stability (the RGO film does not involve this process). Finally, the PI mask is carefully peeled off.

Fabrication process for buckling 3D structure on PDMS: Laser-cut 25 μm -thick PI film (2D precursor) is screen-printed with a thin layer of adhesive at bonding sites. The 2D precursor is carefully fixed onto the corresponding region of the pre-stretched locally treated PDMS (the strain of the whole piece of PDMS is 25%). The adhesive is heated to dry, and then releasing the PDMS, the controlled 3D structure appears.

Fabrication process for electrodes: The electrodes for the conductors on the PDMS are thin lines of commercial conductive silver adhesive, which is drawn by a syringe and then heated at 80 °C for 5 min.

Characterization setup: The structure and morphology of the gold, AgNWs and RGO films were characterized by using scanning electron microscopy (SEM, Quanta 600F, FEI Co.). The mechanical response including strain and stress of the PDMS was measured at room temperature by a tension tester (HSV, HANDPI). The electrical characteristics (resistance) of the conductors were collected at room temperature using a digital multimeter VICTOR VC9808+ from Double King Industrial Holdings Co.

Finite element method (FEM) analysis: The software used for FEM simulation is COMSOL Multiphysics 5.2 © 2019 BY COMSOL INC. The Young's modulus of the untreated and treated PDMS is 0.17 MPa and 0.5 MPa, respectively.

3. Results and discussion

As reported, the inter-chain bonding of the PDMS prepolymer is mainly controlled by chemical curing agent [28, 29]. The higher the degree of the PDMS network's crosslinking, the higher the stiffness [27]. Different from the common ratio (10:1) of PDMS pre-polymer and curing agent, we started by mixing them at the ratio of 20:1 (figure 1(a)), because of its lower Young's modulus, meaning the PDMS is softer and

more stretchable which is more suitable for stretchable electronics as a substrate. To realize the modulus control, extra curing agent is spin coated on the 20:1 PDMS as a secondary curing. The treated PDMS (T-PDMS) shows higher modulus and stiffness. By covering the patterned PI layer as a mask, the modulus of the defined region can be enhanced by being treated with a certain mass of curing agent, as shown in figure 1(b).

This method shows great potential for application to 2D stretchable conductive films and 3D stretchable structures. Figure 1(c) shows the simulation and optical photos of the untreated PDMS and locally treated PDMS (40% of the area is treated at the center) under 60% stretching strain. Although the strain of the untreated part increases, the treated part is less stretched, which can provide protection for materials on its surface. Figure 1(d) shows the application of localized modulus-controlled PDMS in the fabrication of different stretchable 3D structures, such as a 3D centipede-like structure, cross-shaped structure and movable leaf structure.

3.1. Controlling of the modulus of PDMS

Considering the secondary absorbing and curing process of PDMS membranes, there are two main factors in accurately controlling the modulus of treated PDMS: the first is the absorbing mass of curing agent and the second is heating time. The mass of the curing agent is controlled by adjusting the rotation speed of spin-coating, which decreases as the rotation speed increases, as shown in figure S2(a). Figure 2(a) shows the stress–strain curves of samples with different rotation speeds for the spin-coating process, and figure 2(b) shows the enlarged and relatively linear region of the stress–strain curves. The lower the rotation speed, the larger the Young's modulus, as shown in figure 2(c), which makes the treated PDMS stiffer and more difficult to stretch. Under the same stress, the strain of the PDMS after spin-coating at a lower rotation speed is smaller.

Figure 2(d) shows the stress–strain curves of samples with different heating time and figure 2(e) shows the enlarged and relatively linear region of the stress–strain curves. A long heating time deepens the absorbing process of the curing agent and enhances the crosslink effect in the inter-chain reaction, leading to the increase in the Young's modulus as shown in figure 2(f). After the first 60 min, the Young's modulus obviously increases, while it changes little after 120 min.

It is worth mentioning that an important property of PDMS, high transparency, is not influenced by either of the two factors mentioned above. As shown in figures S3(a) and (b), the transmittance is not changed by this method regardless of the mass of the curing agent and heating time, which will be useful in some soft optical electronics. Meanwhile, figure S4 shows the effect of curing agent treatment on the bending state of the PDMS. The untreated PDMS bends smoothly as a naturally arched shape (red frame in figure S4) while the treated PDMS shows a relatively flat region (black frame in figure S4) at the bending state.

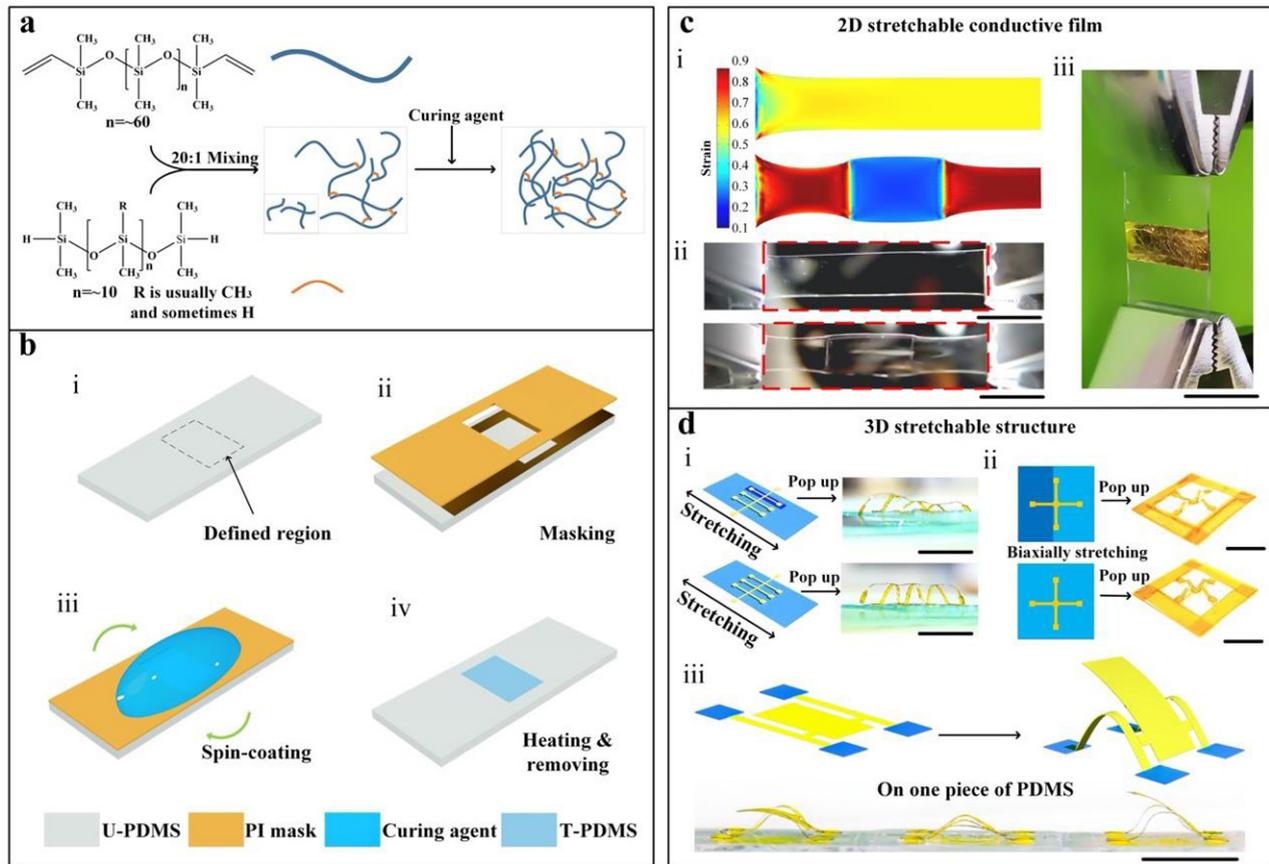


Figure 1. (a) Structural formula of typical linear PDMS prepolymers and curing agent, and schematic illustration of low crosslink reaction and high crosslink reaction. (b) Schematic illustration of modulus control at specific area on PDMS. (c) Locally controlling the modulus of the PDMS for 2D stretchable conductive film. (i) Finite element simulation of in-plane strain maps of untreated PDMS and treated PDMS in the stretching direction. (ii) Corresponding optical photo of untreated PDMS and treated PDMS under 60% stretching. (iii) Treated part coated with gold for stretchable conductivity test. (d) Localized controlling of the modulus of PDMS for 3D stretchable structure. (i) 3D centipede-like structure. (ii) 3D cross-shaped structure. (iii) Differing shapes of 3D movable leaf structure on one piece of PDMS. Scale bar: 10mm.

3.2. Localized modulus-controlled PDMS for 2D conductive films

For depositing 2D conductive film, the localized modulus-controlled PDMS as a stretchable substrate is a good choice, dispensing with complex patterning or wrinkle fabrication. A rectangular region of PDMS is treated by different masses of curing agent controlled by spin-coating at different rotation speeds. As demonstrated, the greater the mass of the curing agent, the stiffer the PDMS. Gold, AgNWs and RGO, chosen as representational materials for 2D stretchable conductive film, are deposited on the rectangular region (see red frame in figures S5(a) and (b) in the online supplementary material (stacks.iop.org/JMM/30/045001/mmedia)) by sputtering (gold) and spraying (AgNWs and RGO) technologies (see figures S5(c) and (d)). Figure 3(a) shows a schematic illustration of various conductive films fabricated on the modulus-controlled region of the PDMS. Figures 3(b)–(g) show the SEM images of three kinds of conductive films directly on the treated region of the PDMS and the cracks of the film when stretched.

The results show that the localized modulus enhancing works well in protecting the 2D conductive film’s conductivity

directly on the surface of PDMS, when stretched to a certain limit. Additionally, the conductivity protection improves with the increase of mass of the curing agent for all three samples, as shown in figures 4(a)–(c).

By simplifying the model as three parts (see figure S5(b) in the supplementary material), the strain of total elongation can be modeled as the equation

$$\epsilon_{total} = \frac{\Delta l}{l} = \frac{E_2}{E_1} \epsilon_{crack} \frac{x_1 + x_3}{x_1 + x_2 + x_3} + \epsilon_{crack} \frac{x_2}{x_1 + x_2 + x_3} \tag{1}$$

where E_1 , E_2 represent the modulus of the untreated PDMS and treated PDMS, respectively. ϵ_{crack} represents the critical strain by which the 2D conductive film is stretched to crack (non-conductive state). x_1 , x_2 , x_3 represent the lengths of the three parts of locally treated PDMS. Δl and l represent the increased length and initial length of the whole piece of locally treated PDMS, respectively. The larger ϵ_{total} means the whole piece of locally treated PDMS can be stretched further, until a non-conductive state. The derivation of this equation can be seen in the supplementary information.

From equation (1), the higher the modulus of the rectangular region, the more the whole PDMS can be stretched.

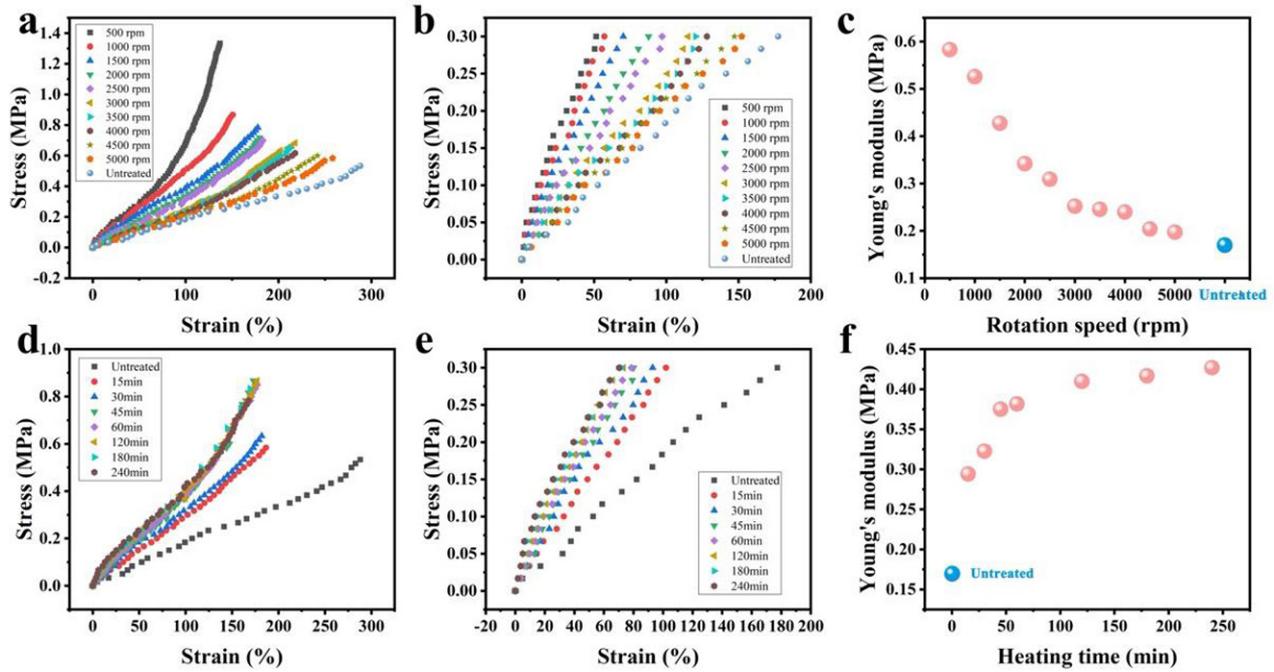


Figure 2. (a) Stress–strain curve of treated PDMS with different rotation speeds of spin-coating of curing agent. (b) Enlarged linear part of strain–stress curve at 0–0.3 MPa. (c) Young’s modulus of treated PDMS as a function of rotation speed. (d) Strain–stress curve of treated PDMS with different treating time of curing agent. (e) Enlarged linear part of strain–stress curve at 0–0.3 MPa. (f) Young’s modulus of treated PDMS as a function of treating time.

If the rotation speed is 1000 rpm when depositing a layer of gold film on the rectangular region and stretching the PDMS, the untreated PDMS can be stretched to a maximum strain of 30% (exceeding this leads to non-conductivity of the gold film), while the locally treated PDMS can be stretched to a maximum strain of 75%.

The strategy is as follows: by letting the softer part undertake more strain and the stiffer part undertake less when the whole piece of PDMS is stretched, this supports a better protection for the 2D conductive film compared with untreated PDMS, and the protection can be even accurately controlled by adjusting the mass of the curing agent. Figure 4(a(iv)) shows that compared with untreated PDMS, at the same elongation for the whole piece of PDMS, the elongation of the gold part of locally treated PDMS is less. However, the elongation of other part becomes larger, which demonstrates that softer part ‘protects’ the stiffer part when the PDMS is stretched.

Figure S6 demonstrates that regardless of different aspect ratios (the same length, with different width), this method always provides protection to 2D conductive films. Gold film with larger width shows less changes in resistance when stretched [30]. This indicates that gold film with larger width can be stretched more before reaching a non-conductive state, meaning a larger ϵ_{crack} for larger width. According to equation (1), the larger the width, the larger the strain ϵ_{total} that gold film on treated PDMS can afford.

3.3. Localized modulus-controlled PDMS for multiple 2D conductive films on one substrate

Multiple materials/structures on one piece of stretchable substrate also face the ‘barrel effect’ problem of the different

stretching limitations of each kind of material/structure, which fails to maximize the stretchability of the integral device.

With the expectation of multiple functions, or better electro-mechanical performance, stretchable electronics require multiple functional materials fabricated on one stretchable substrate [31, 32]. A localized modulus-controlled PDMS can play a better role as a stretchable substrate for a multiple conductive material films system. As shown in figure S7(a), gold film, AgNWs film with a percolation network, and RGO film are fabricated on one piece of PDMS. For different kinds of conductive materials and different fabricating methods and conditions, the maximum strain to non-conductive state of 2D conductive film is also different. As a summary, in figure 5(a), three types of conductive material films show different stretching performance on the same treated PDMS (controlled by mass of curing agent). Thus, a localized and separately controlled modulus of PDMS for every type of conductive material is necessary. Using stretchable 2D conductive film as a conductor, when it is at the initial state (conductive state), the battery can successfully drive the LED. However, when stretched to 20%, the multiple conductive materials on the uniformly treated PDMS (every region is treated with same mass of curing agent and the rotation speed is 5000 rpm) become non-conductive. This is because different conductive materials require different degrees of protection from their corresponding substrate. At the condition of 5000 rpm rotation speed, the Ag nanowire film and graphene film on treated PDMS can afford more tensile strain than gold film. Stiffer PDMS at the region of gold film is therefore required. By reducing the rotation speed to 1000 rpm, the mass of the curing agent in the region of the gold film increases and the modulus is enhanced. In figures 5(b)–(d), the comparison

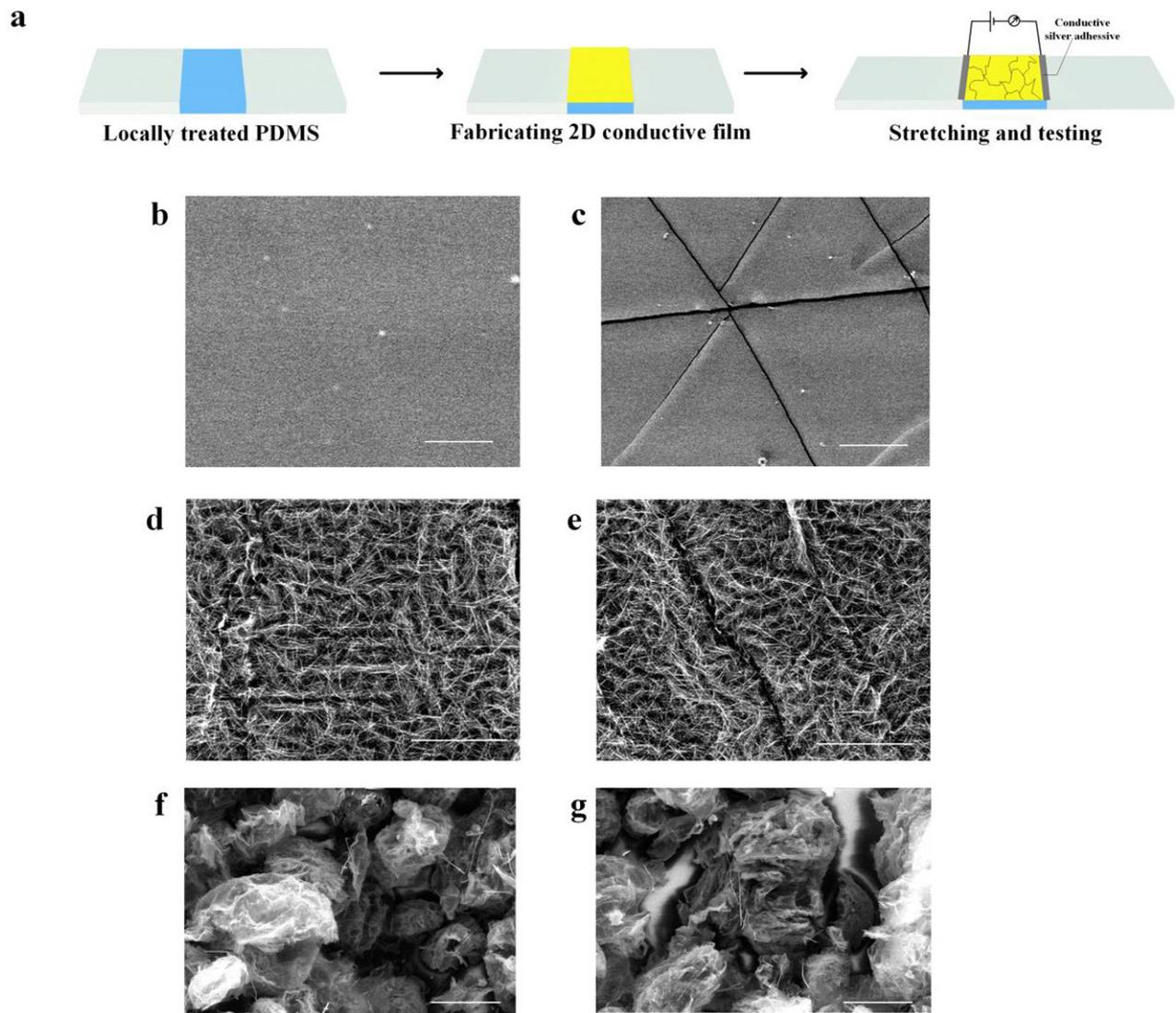


Figure 3. (a) Schematic illustration of various conductive films fabricated on the modulus-controlled region of PDMS. (b), (d) and (f) SEM images of Gold, AgNWs and RGO film directly fabricated on the localized modulus-controlled PDMS before being stretched, respectively. (c), (e) and (g) SEM images of cracks on conductive films (gold: (c), AgNWs: (e), RGO: (g)) after being stretched. Scale bar: 50 μm for (b) and (c), 10 μm for (d) and (e), 20 μm for (f) and (g).

of on and off LEDs demonstrated that localized modulus-controlled PDMS for 2D stretchable conductive films with multiple conductive materials can be stretched two directions, and even biaxially stretched. Figures S7(b)–(d) show the different rotation speeds of the spin-coating process for different stretching directions.

3.4. Localized modulus-controlled PDMS for 3D stretchable structures

3D stretchable structures have been demonstrated to play an important role in stretchable electronics and possess wide application prospects. The realization of 3D stretchable structures is mainly achieved by ‘popping up’ from a patterned kirigami 2D film (also called a 2D precursor) on a pre-stretched soft substrate. In previous work, a soft substrate with uniform modulus was used as a deformation supporter. Kim *et al* fabricated microscale arc-shaped ‘ribbon cables’

of metal and plastic in neutral mechanical layouts, interconnecting silicon devices located at the nodes of the mesh on a pre-stretched PDMS substrate with uniform modulus [17]. Han *et al* transferred a 2D pattern onto an Ecoflex substrate with uniform modulus to form a 3D piezoelectric microsystem for harvesting mechanical energy [5]. Similarly, a wide range of 3D deformable graphene hydroelectric generators that can automatically assemble and transform into a specific geometry configuration were realized by a pre-stretched Ecoflex substrate by Yang *et al* [33]. Kofod *et al* utilized a piece of pre-stretched dielectric elastomer to form a 3D actuator based on minimization of the hyperelastic free energy of a stretched elastomer and a plastic frame. Meanwhile, the research in [34] focuses on how localized modulus-changed PDMS substrate changes 3D structures.

Patterned PI films by laser cutting were used as 2D precursors (the design of the 2D precursors can be seen in figure S8 in the supplementary material). Soft PDMS with a 20:1 ratio

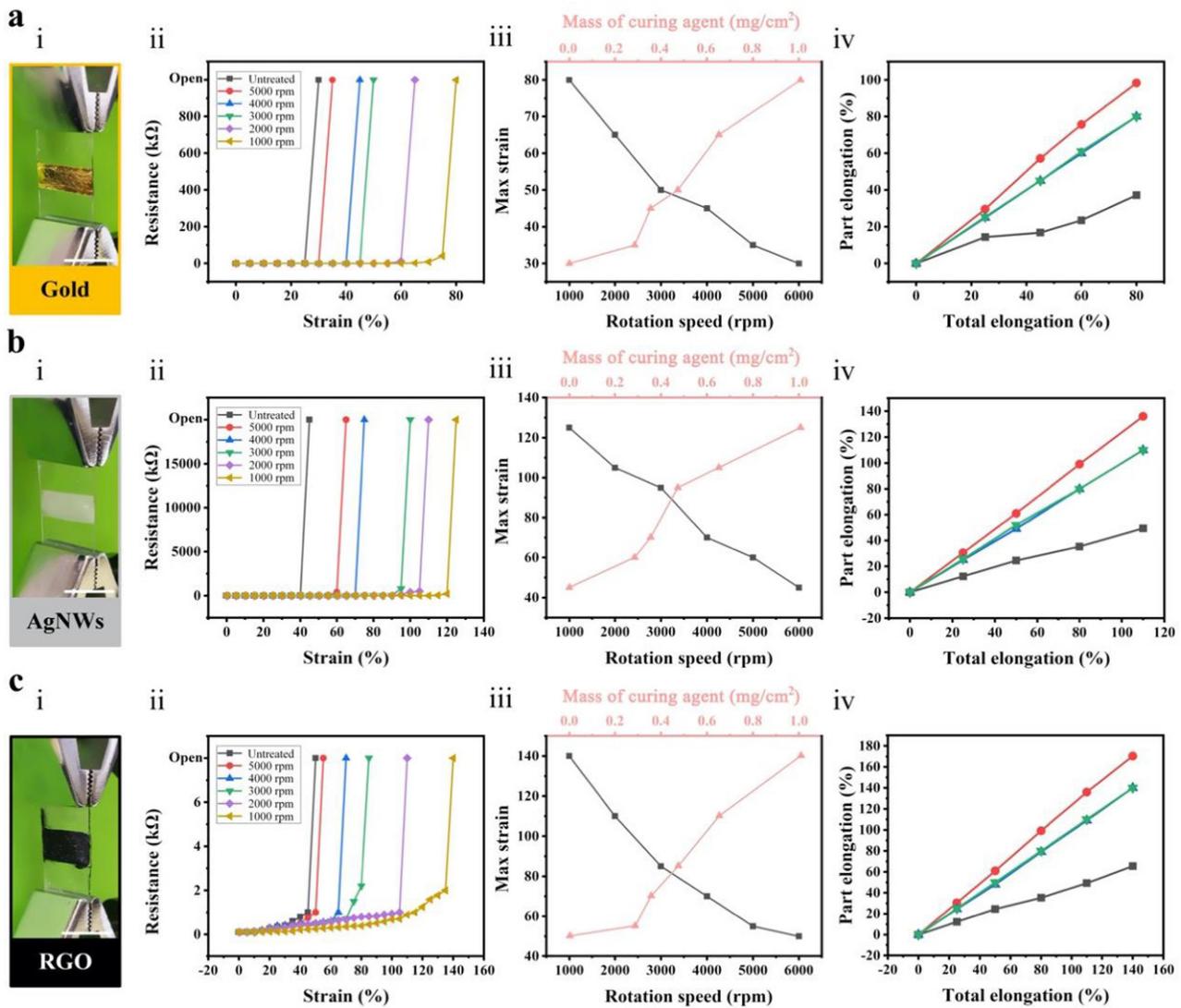


Figure 4. Resistance test of localized modulus-controlled PDMS for stretchable conductive film. (a), (b) and (c) Stretchability of gold, AgNWs and RGO film directly fabricated on the localized modulus-controlled PDMS, respectively. (i) Practical photos of localized modulus-controlled PDMS with gold, AgNWs and RGO directly on the defined region. Scale bar is 10 mm. (ii) Resistance–strain curves of three samples with different rotation speed of spin-coating in the modifying process of PDMS. (iii) Max strain for non-conductive (open) state as a function of rotation speed of spin-coating and mass of curing agent. (iv) Elongation of different PDMS parts with different total elongation. Red and grey lines represent the gold and other part of the locally treated PDMS, respectively. Blue and green lines represent the gold and other part of the untreated PDMS, respectively. Rotation speed is 1000 rpm.

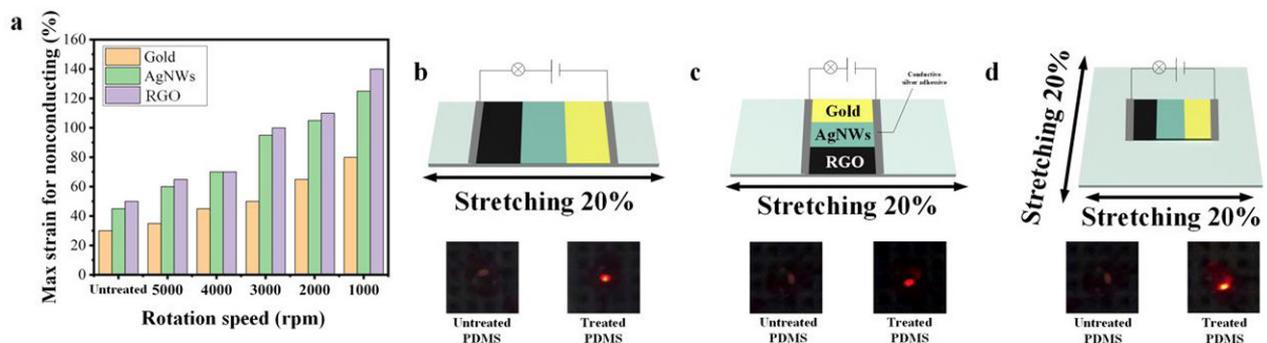


Figure 5. (a) Max strain for non-conducting state varies differently with RGO, AgNWs and gold at every rotation speed. (b), (c) and (d) Schematic illustration of one piece of PDMS with RGO, AgNWs and gold simultaneously coated on the surface before stretching and after stretching at different directions. The light turns on (with treated PDMS) and off (with untreated PDMS) after stretching the PDMS substrate.

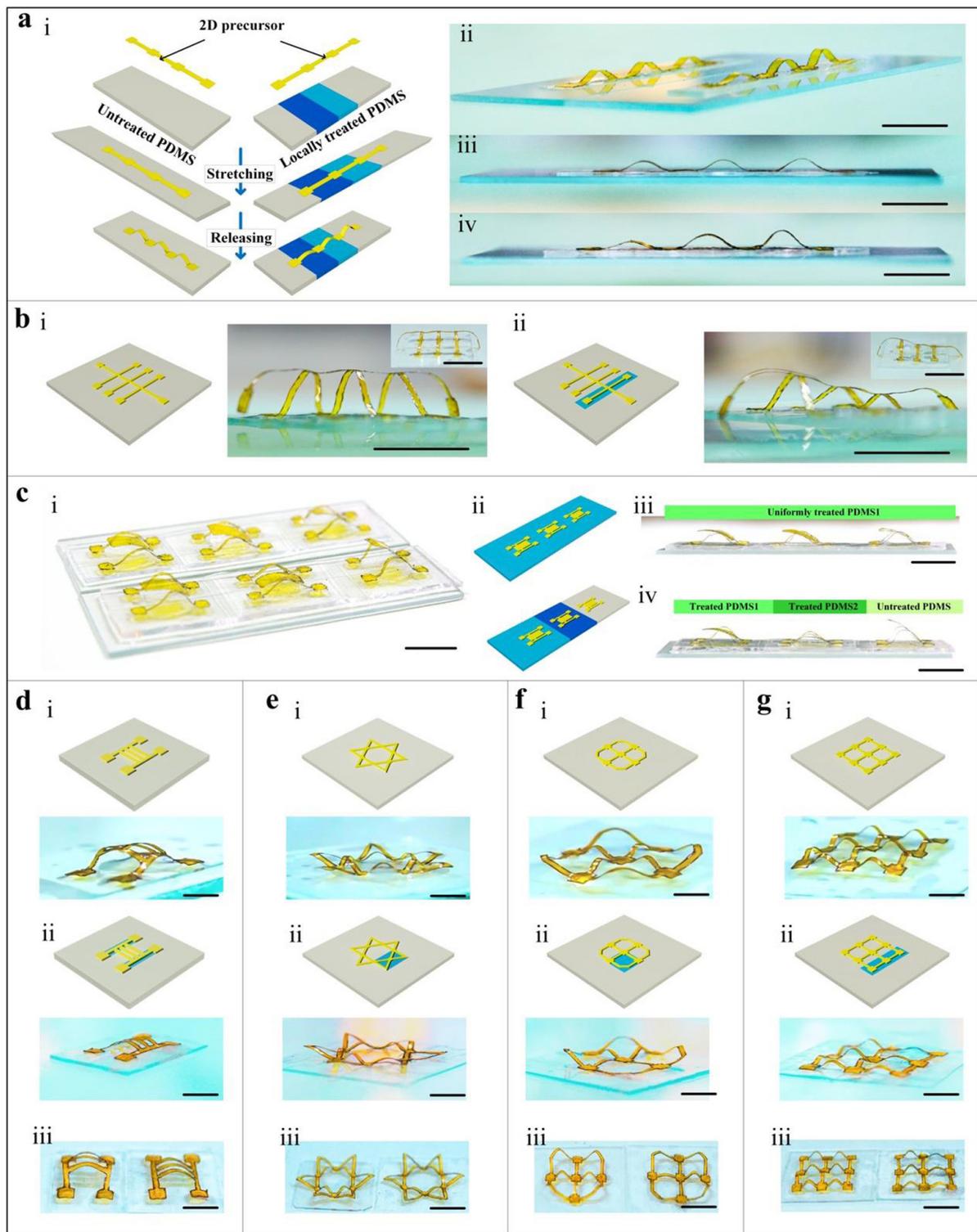


Figure 6. (a) Schematic illustration of untreated PDMS and localized modulus-controlled PDMS for wavy-like structure. (i) Fabrication process comparison. Left: untreated PDMS. Right: locally treated PDMS. Gray, blue and deep blue regions represent untreated PDMS, treated PDMS and highly treated PDMS, respectively. (ii) Optical image of two types of wavy-like structure. (iii) and (iv) Section views of wavy-like structure on untreated PDMS and locally treated PDMS. (b) Optical images of centipede-like structure. (c) 3D movable leaf structure array on uniformly treated PDMS and locally treated PDMS. (a), (b) and (c) Scale bar: 10 mm. (d)–(g) Different 3D structures on PDMS fabricated by uniaxial pre-stretching and releasing. (i) 3D structures on untreated PDMS. (ii) 3D structures on locally treated PDMS. (i) and (ii) Scale bar: 5 mm. (iii) Front views of 3D structures on untreated PDMS and locally treated PDMS. Scale bar: 10 mm.

of pre-polymer to curing agent was used as the pre-stretched substrate. As shown in figures 6(a(i) and (ii)), a uniform wavy structure was fabricated by releasing the untreated PDMS, while a stepped wavy structure was realized by using locally treated PDMS. The gray, blue and deep blue regions correspond to the untreated PDMS, treated PDMS and highly treated PDMS. The stiffer region of PDMS is more difficult to stretch than the softer region, which supports a smaller strain to ‘pop up’ the part of the 2D precursor after total release. Figures 6(a(iii) and (iv) show the section view of the uniform wavy structure whose amplitude and wavelength are consistent, while those of the stepped wavy structure are different from left ‘wave’ to right ‘wave’: the amplitude increases and the wavelength decreases with the modulus of the corresponding region decreasing gradually. The amplitude of each wave increases from 1.71 mm to 3.06 mm and the wavelength of each wave decreases from 8.82 mm to 7.28 mm, with the modulus decreasing from 0.427 35 MPa to 0.169 11 MPa (see figure S9(d) in the supplementary material). The relationship between the amplitude and wavelength of the wave can be expressed as

$$A = \frac{2W_0}{\pi} \sqrt{\frac{W_0 - W}{W_0} - \varepsilon_c} \quad (2)$$

where W_0 and W represent the wavelength at the stretching state and releasing state, respectively, and ε_c is the critical buckling strain (the derivation can be seen in the supplementary material). The tested amplitude is quite similar to the theoretical value (see figure S10(b) in the supplementary material).

Through uniaxial stretching, the more complex and centipede-like 3D structure can be realized by enhancing the modulus of the PDMS at the third pairs of ‘feet’ (the deep blue region) in figure 6(b). The localized modulus controlling method can also control every single structure in a 3D structure array, as shown in figure 6(c). A movable leaf is specially designed into 2D precursors, stuck onto the PDMS and popped up into a 3D movable leaf-shaped structure array. One of these is on a localized modulus-controlled PDMS substrate, and every leaf shows a different bend angle and elevation height because of different releasing strain at its substrate, caused by the modulus controlling method, which is different from that on uniform PDMS.

By biaxial stretching, a more 3D stretchable structure can be achieved and controlled by localized modulus-controlled PDMS, containing shapes such as a beetle (figure 6(d)), two-layer flower (figure 6(e)), and folded box with cross-shape (figure 6(f)) on center and wavy mesh (figure 6(g)). Figures 5(d)–(g) also show obvious comparisons between 3D structures fabricated on locally treated PDMS and untreated PDMS. The method of locally modifying the substrate—defining region, controlling modulus, and releasing to construct a different 3D structure, regardless of the material of the 2D precursor itself—is quite simple and universal, and can be widely used in stretchable 3D structures.

4. Conclusion

In summary, this work illustrates a simple method for locally controlling the modulus of cured PDMS which is applied to 2D and 3D stretchable electronics. The local modification of PDMS can effectively solve the crack problem when the conductive film on PDMS is stretched. Moreover, the ‘barrel effect’ of the stretchability of multi-films on one piece of PDMS substrate can be avoided through selectively enhancing the modulus of different regions corresponding to different functional materials. This method also proposes a new way of building and controlling stretchable 3D structures. Via changing the modulus of the releasing region, the degree of buckling is also controllably changed. Rather than focusing on the processing technology of functional materials and structures on PDMS, this work focuses on how to modify one of the most used soft substrates—on PDMS itself, and analyses the relationship between the stretchability of 2D films or the shape of 3D structures and the degree of modulus enhancement on the specific region of PDMS, which shows great potential in the application of stretchable electronics.

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References

- [1] Hong S, Lee H, Lee J, Kwon J, Han S, Suh Y, Cho H, Shin J, Yeo J and Ko S 2015 *Adv. Mater.* **27** 4744
- [2] Shi M, Zhang J, Chen H, Han M, Shankaregowda S, Su Z, Meng B, Cheng X and Zhang H 2016 *ACS Nano* **10** 4083
- [3] Song Y, Wang H, Cheng X, Li G, Chen X, Chen H, Miao L, Zhang X and Zhang H 2019 *Nano Energy* **55** 29–36
- [4] Zhang X, Han M, Wang R, Zhu F, Li Z, Wang W and Zhang H 2013 *Nano Lett.* **13** 1168
- [5] Han M et al 2019 *Nat. Electron.* **2** 26
- [6] Song Y, Chen H, Chen X, Wu H, Guo H, Cheng X, Meng B and Zhang H 2018 *Nano Energy* **53** 189–97
- [7] Yamada T, Hayamizu Y, Yamamoto Y, Yomogida Y, Izadi-Najafabadi A, Futaba D and Hata K 2011 *Nat. Nanotechnol.* **6** 296
- [8] Liu Y, Pharr M and Salvatore G 2017 *ACS Nano* **11** 9614
- [9] Song Y, Chen H, Su Z, Chen X, Miao L, Zhang J, Cheng X and Zhang H 2017 *Small* **13** 1702091
- [10] Chen X, Song Y, Su Z, Chen H, Cheng X, Zhang J, Han M and Zhang H 2017 *Nano Energy* **38** 43
- [11] Xu S et al 2015 *Science* **347** 154
- [12] Lacour S, Wagner S, Huang Z and Suo Z 2003 *Appl. Phys. Lett.* **82** 2404
- [13] Zhang Y et al 2014 *Adv. Funct. Mater.* **24** 2028

- [14] Zhang J, Song Y, Chen H, Cheng X, Chen X, Meng B, Yuan Q and Zhang H 2017 *2017 IEEE 30th Int. Conf. on Micro Electro Mechanical Systems*
- [15] Yang S, Khare K and Lin P 2010 *Adv. Funct. Mater.* **20** 2550
- [16] Rogel R, Borgne B, Mohammed-Brahim T, Jacques E and Harnois M 2017 *Adv. Mater. Interfaces* **4** 1600946
- [17] Kim D, Song J, Choi W, Kim H, Kim R, Liu Z, Huang Y, Hwang K, Zhang Y and Rogers J 2008 *PNAS* **105** 18675
- [18] Ahn B, Duoss E, Motala M, Guo X, Park S, Xiong Y, Yoon J, Nuzzo R, Rogers J and Lewis J 2009 *Science* **323** 1590
- [19] Oh E, Byun J, Lee B, Kim S, Kim D, Yoon J and Hong Y 2017 *Adv. Electron. Mater.* **3** 1600517
- [20] Matsuhisa N, Kaltenbrunner M, Yokota T, Jinno H, Kuribara K, Sekitani T and Someya T 2015 *Nat. Commun.* **6** 7461
- [21] Naserifar N, LeDuc P and Fedder G 2016 *Adv. Mater.* **28** 3584–91
- [22] Park C, Jung S, Na B, Oh J, Park N, Lee S and Koo J 2016 *Semicond. Sci. Technol.* **31** 025013
- [23] Wu Z, Zhang S, Vorobyev A, Gamstedt K, Wu K, Guo C and Jeong S 2018 *Mater. Today Phys.* **4** 28–35
- [24] Cao Y, Zhang G, Zhang Y, Yue M, Chen Y, Cai S, Xie T and Feng X 2018 *Adv. Funct. Mater.* **28** 1804604
- [25] Liu H, Li M, Liu S, Jia P, Guo X, Feng S, Lu T, Yang H, Li F and Xu F 2019 *Mater. Horiz* **1** 203–13
- [26] Kokkinis D, Bouville F and Studart A 2018 *Adv. Mater.* **30** 1705808
- [27] Wang Z, Volinsky A and Gallant N 2014 *Appl. Polym. Sci.* **131** 41050
- [28] Berean K et al 2014 *Separation Purification Technol.* **122** 96
- [29] Dollase T, Spiess H, Gottlieb M and Yerushalmi-Rozen R 2002 *Europhys. Lett.* **60** 390
- [30] Adrega T and Lacour S P 2010 *J. Micromech. Microeng.* **20** 055025
- [31] Liang J, Li L, Chen D, Hajagos T, Ren Z, Chou S, Hu W and Pei Q 2005 *Nat. Commun.* **6** 7647
- [32] Jing M, Han C, Li M and Shen X 2014 *Nanoscale Res. Lett.* **9** 588
- [33] Kofod G, Wirges W, Paajanen M and Bauer S 2007 *Appl. Phys. Lett.* **90** 081916
- [34] Yang C, Huang Y, Cheng H, Jiang L and Qu L 2019 *Adv. Mater.* **31** 1805705