

1 **Bio-Inspired Ultrathin Piecewise Controllable Soft Robots**

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18 Keywords: Soft robotics, Bio-inspired electronics, Thermal actuators, Flexible electronics,
19 Piecewise control

20 **Abstract**

21 In nature, animals or plants often use soft organs to move and hunt. Research works on
22 bio-inspired materials and devices have attracted more and more interest as which show the
23 potential for future intelligent robots. As key components of soft robots, biomimetic soft
24 actuators are adapted to greater requirements for convenient, accurate, and programmable
25 controlling robots. Here, we report a class of materials and processing routes of ultrathin
26 actuators for bio-inspired piecewise controllable soft robots, where the actuators associate with
27 thermal-responsive soft silicone thin film with thickness as thin as 45 μm and electrically
28 driven by well mechanical designed metallic thin film electrodes. Multiple electrodes in the
29 robots in charge of individual segments control allow the soft robots exhibiting similar
30 functionalities of animals or plants (for example, imitating the tongue of a reptile, such as
31 chameleon to hunt moving preys, and mimicking vines to tightly wind around objects). These
32 bionics results in the soft robots demonstrate their advantages in precise and flexible operation,
33 which provides a good reference for the future research of intelligent soft actuators and robots.

34

35 **Keywords:** Bio-inspired robots, Soft robots, Piecewise Control, Bi-layer film, Flexible
36 electrode

37 **1. Introduction**

38 Intelligent robots are gradually playing crucial roles in the engineering and medical
39 fields.(1)(2)(3)(4)(5) Inspired by natural creatures, bionics research on robots has attracted great
40 of attentions as which could make robots more intelligent.(6) The trend of developing bio-
41 inspired robots tends to be exploring materials and devices to act as “soft body” or “soft organs”
42 in robots for realizing advanced mechanical attributes, great environmental adaptability and
43 sophisticated applications. Through scientists’ efforts from multidisciplinary fields,
44 technologies in flexible electronics haven been proved to be a good foundation for future
45 intelligent soft robots.(7)(8)(9)(10)(11) The intrinsic nature of “organs” in soft robots, such as
46 hands, legs, tongue, etc. is various kinds of flexible actuators. Functional materials that can be
47 responsible to electricity, temperature, magnetic field, and light, i.e. dielectric elastomer,(12)
48 bilayer graphene(13)(14), thin magnetic membranes,(15) and liquid crystal elastomer
49 (LCE),(16)(17) enable the corresponding flexible actuators generating obvious deformations to
50 serve as the active organs in robots.(18) For instance, imitating animals’ behaviors from insects’
51 crawling and quadrupeds’ walking allows scientists realizing insect-sized soft robots and three-
52 dimensional (3D) robots with flexible limbs or body;(19)(20) mimicking the twining
53 phenomena of the growing vines stimulates the development of screw-typed soft
54 actuators.(21)(22)(23) The key point of developing these bionic soft robots is properly
55 coordinating materials, structures and actuation modes.

56 To date, various actuation methods including magnetic field, light, heat, etc. have been
57 adopted for driving soft actuators or robots.(16)(24)(25) Among these methods, magnetic field
58 is one of the most used one, that associates with developing the main body of a robot by
59 advanced manufacturing techniques, i.e. 3D or 4D printing to create programmed magnetization
60 configuration that allows the robot deforming or moving under a unidirectional magnetic
61 field.(25)(26) For actuators that respond to temperature or light, heat or light radiation can

62 converting thermal or optical energy to mechanical forces for deforming the structure of the
63 actuator.(27)(28) Despite the progress of soft actuators based on the above mentioned actuation
64 methods, the specific requirements such as external magnetic field controlling systems or
65 accurate light sources are needed and thus significantly limit their applications.(29) Compare
66 with these passive actuation methods, electrically control is an excellent active actuation
67 method that shows many advantages in developing soft actuators.(30) On one hand, controllable
68 and quantifiable electrical inputs allow precise controlling actuators to generate accurate and
69 desired mechanical deformations without external heavy and complicated setups. On the other
70 hand, electric based actuators also offer great compatibility of electronics integration with the
71 control system in the robots. Thermal induced deformations in actuators by inputting electricity
72 to conductive wires has been reported for soft actuators.(24) However, the dimension,
73 especially the thickness of these soft actuators are still way too big, that limits their applications
74 in biomedical areas.(20)(31) How to develop small size, ultrathin bio-inspired soft actuators
75 raises to be one of the key problems in soft robots, and two specific tasks are need to be solved:
76 (1) developing thin-film actuators with ultra-thin flexible electrodes; (2) endowing the
77 biomimetic actuator ability of separated piecewise control to enhance its flexibility and
78 controllability in various application scenarios.

79 In this work, we developed an ultrathin film that are responsible to thermal inputs for
80 soft actuators by combining flexible electronic processing technologies and mechanics designs.
81 A bi-layer thin film with significant difference of thermal expansion coefficient and a 200 nm-
82 thick gold (Au) electrode was developed to serve as the actuator. The actuators exhibit a spiral
83 shape with 900° bending in their original state, but can straighten out with electricity generated
84 heat input through the electrode. To illustrate the advantages of the materials and mechanics in
85 the soft actuators, we developed piecewise controllable soft robots based on these actuators by
86 simply using multiple separated electrodes. These robots show great bionic capabilities, such

87 as mimicking the tongue of a chameleon to hunt a moving ant, and simulating a plant's vine to
88 wind around a tiny pole. The precise programmed fabrication and piecewise control of the thin-
89 film based soft actuators will provide important references for future bionic actuators and robots.

90 **2. Materials and methods**

91 *2.1. Fabrication of thin-film actuator*

92 First, poly(methylmethacrylate) (PMMA) solution ($20\text{mg}\cdot\text{mL}^{-1}$, chlorobenzene solvent)
93 serving as a sacrificial layer was spin-coated on a quartz glass substrate and baked at $200\text{ }^{\circ}\text{C}$
94 for 20 min. Afterward, poly(pyromellitic dianhydride-co-4,4'-oxydianiline), amic acid solution
95 (Sigma-Aldrich) was spin-coated on the PMMA layer at 3000 rpm for 30 s and annealed at
96 $250\text{ }^{\circ}\text{C}$ for 30 min. The formed $2\text{-}\mu\text{m}$ polyimide (PI) film with low heat expansion coefficient
97 was served as passive deformation layer. 200-nm Au was deposited onto the PI film by a
98 sputtering system (Quorum Sputter coater, Q150 T S) and then coated by photoresist (AZ 5214)
99 at 3000 rpm for 30 s with soft-bake of $110\text{ }^{\circ}\text{C}$ for 3 min. After exposure under ultraviolet light
100 with a film mask and development in AZ 300MIF developer, the Au layer was etched in the
101 gold etchant (I_2/KI solution) to form the desired pattern. After removing the PMMA sacrificial
102 layer in acetone for 12 hrs, the anisotropic conductive film (ACF) wire was connected with the
103 acquired ultrathin gold electrode for further heating by external voltage source. Finally, the
104 polydimethylsiloxane (PDMS, Sylgard 184, per-polymer and cross-link agent = 5:1) was spin-
105 coated at 1000 rpm for 60 s on the electrode and cured at $75\text{ }^{\circ}\text{C}$ for 30 min, serving as the
106 positive deformation layer. After cutting and peeling the film off the substrate, the film was
107 annealed on the hotplate at $150\text{ }^{\circ}\text{C}$ for 10 min and cooled to room temperature in air to achieve
108 a stable state. Thus, the helical actuator was acquired.

109 *2.2. Actuation and characterization*

110 After bonding the ACF wire to a designed printed circuit board (PCB), the actuator was
111 connected with a DC power source. When a certain voltage was applied, the actuator was heated

112 and straightened to present a state of elongation. All deformation processed were recorded with
113 a macro-lens camera (SONY). Temperature changes of the heated actuators were recorded by
114 an infrared camera (FLIR-C7200). The cross-section microstructure of the actuator was
115 observed by using a scanning electron microscope (JEOL/JSM-5600). The thickness of
116 actuators with different PDMS layers was measured by the optical surface profiler (OSP,
117 Veeco/Wyko NT9300).

118 *2.3. Thermal simulation*

119 Electrothermal simulation is acquired based on the finite element analysis (FEA). The
120 commercial software ABAQUS was used to study the temperature distribution of the one-
121 electrode device with the heat convection coefficient ($26 \text{ W m}^{-2} \text{ K}^{-1}$) of air and an input power
122 of 0.18 W. The entire device was modeled by hexahedron heat-transfer elements (DC3D8).
123 Mesh convergence of the simulation was ensured for all the cases. The thermal conductivity,
124 heat capacity and mass density used in the simulations were $315 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, $130 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
125 and $19300 \text{ kg} \cdot \text{m}^{-3}$ for Au, $0.15 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, $1460 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ and $965 \text{ kg} \cdot \text{m}^{-3}$ for PDMS, 0.12
126 $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$, $1090 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ and $1420 \text{ kg} \cdot \text{m}^{-3}$ for PI.

127 **3. Results and discussion**

128 Figure 1a illustrates the schematic diagram of the ultrathin soft actuator and highlights
129 the key materials used for it, consisting a $43 \mu\text{m}$ thick polydimethylsiloxane (PDMS) layer as
130 the main body, a 200 nm thick Au electrode supported by a thin polyimide (PI, $2 \mu\text{m}$ thick)
131 layer as thermal actuation part. Figure 1b shows the fabrication process of the soft actuator. The
132 fabrication started on a clean glass substrate, where a thin poly(methylmethacrylate) (PMMA)
133 layer was spin-coated and baked as the sacrificial layer. Then, poly(pyromellitic dianhydride-
134 co-4,4'-oxydianiline), amic acid solution was spin-coated on the PMMA layer and annealed at
135 $250 \text{ }^\circ\text{C}$ to form a $2 \mu\text{m}$ -thick dense PI film, as the passive deformation layer.(32) Next,

136 sputtering deposition and photo lithography defined 200 nm-thick Au thin layer on the PI layer
137 formed the electro-thermal converting electrode.(33)(34) Immersing the sample in acetone for
138 12 hrs allowed fully dissolving PMMA and thus we could easily peel off the thin film from the
139 glass substrate. After bonding ACF wire on the Au electrode pins, the heating electrode could
140 be supplied by external power (Figure 1d). Finally, the PDMS with per-polymer and cross-link
141 agent ratio of 5:1 was spin-coated on the PI and Au electrode at spinning speed of 1000 rpm for
142 60s and cured at 75 °C for 30 min. After cutting the edge of the sample into designed width of
143 4 mm, the actuator was peeled off with a straight shape (Figure 1c). In order to make the actuator
144 own a stable steady state, an annealing process was carried out at 150 °C for 5 min. The high
145 temperature annealing state made the PDMS layer being in excess of expansion, and thus
146 resulted in a large shrinkage after cooling down to room temperature.(35)(36) Figures 1c and e
147 present the state and shape of the actuator before and after annealing or actuation. The dynamic
148 deformation process of the actuator on a hot plate at 150 °C can be seen in Movie S1. Thus, an
149 ultrathin electrically controlled soft actuator with a helical structure is obtain. Cross-section
150 scanning electron microscopy (SEM) image of the actuator shows clear thin layer structure, and
151 the thickness of each layer was also measured and confirmed by an optical surface profiler to
152 be 2 μm thick for PI and 43 μm thick for PDMS.

153 To study the effect of PDMS thickness on the actuator deformation ability, we prepared
154 a group of soft actuators with different thick PDMS layers ranging from 43 μm to 178 μm by
155 simply controlling the spinning durations at a fixed speed of 1000 rpm (Figures S1 and S2).
156 The results showed that the deformation of PDMS is highly relevant with the thickness, where
157 thicker layers induced the smaller deformations. As shown in Figure S1, the annealed 45 μm-
158 thick actuator exhibits a 900° bending angle and the shortest length among all devices,
159 demonstrating great performance of deformation and elongation during the actuating process.
160 The heating area of the electrode is 2 mm in width and 32 mm in length (Figure S3). After

161 cutting into a 4 mm-wide stripe, the entire actuator could realize actual actuation on the 32 mm
162 length, excluding the wire connection area. Figure 2a shows the original state and actuated state
163 of the actuator under different voltage inputs, where we can find the actuator can fully straighten
164 itself out under an voltage input of 10 V that equals to 179 mW since the resistance of the
165 electrode is 558 Ω (Movie S2). The deformation process derives from the significant difference
166 in thermal expansion coefficient between PI and PDMS. The thermal expansion coefficient of
167 positive PDMS layer is around $310 \times 10^{-6} \text{ K}^{-1}$, that is more than 10 times greater than that of the
168 passive PI layer with a value of $20 \times 10^{-6} \text{ K}^{-1}$.⁽³⁷⁾ Thermal energy generates greater expansion
169 in the PDMS than that of PI layer, and thus causes the actuator to bend towards the PI layer
170 (Figure 2a and 2b). The electrode in between PI and PDMS offers the actuators highly efficient
171 thermal induced deformations compared to using external heat source such as a heat plate.
172 Infrared (IR) camera recorded temperature information during the thermal actuating process
173 provides quantitative data of the electric-thermal conversions, as shown in Figure 2d. The
174 temperature reaches 62.3° under 10V voltage input, which is consistent with the finite element
175 analysis (FEA) simulated results (Figure 2c). The actuator also exhibits good durability for
176 long-term and multiple times operations, where the actuation capability still maintains the same
177 after 100 cycles operation at 10 V (Figures 2e and S6). Since the actuation temperature did not
178 exceed the annealing temperature of 150°C , the heating would not cause changes of the
179 material properties and the steady state of the structure. The maximum travel amplitude of the
180 actuator is more than twice its initial body length of 14.5 mm to 32 mm, that lays a foundation
181 for the various multi-functional bionic applications. Moreover, the actuator could elongate to
182 different lengths in the range between 14.5 mm and 32 mm under controlled voltage inputs
183 (Figure S4, Movie S2). The relationship between the elongate length and the voltage is
184 summarized in Figure 2f, where we can find the relationship is stable and directed, which further
185 reflects accuracy and precision of electrically control. Figures 2g and Figure S5 shows the great

186 linear behavior of actuation temperatures as a function of input voltage, which provides the
187 basis for a specific control.

188 In addition, we also analyze the deformation process of the actuator during electrical
189 heating and cooling in air. Figure S7 shows that it takes about 2 s to straighten out and 2.5 s to
190 recover to the initial shape. During the straightening process, the actuator slowly elongates at
191 the beginning but instantly straightens out in a short 0.2 s and then slowly turn to the final stable
192 state. This phenomenon is due to an instantaneous release of the potential energy during the
193 deformation process and thus caused a “hump” during the actuation process. This sudden and
194 large deformation is quite similar to the predation behavior for a short time in the animal
195 kingdom. In nature, many reptiles utilize their tongues to catch their preys. For example, geckos
196 and chameleons crawling on the walls use their tongues to catch mosquitoes; anteaters feed on
197 ants by sticking their long tongues into ants’ nests; frogs rely on their tongues to catch flies.
198 Inspired by these hunting behaviors in nature, we used the soft actuator to mimic the
199 chameleon’s tongue for hunting insects, as demonstrated in Figure 3. When a chameleon
200 discovers a moving prey coming closer, it will stick out its tongue to capture the prey via the
201 sticky part on the upper side of its tongue and then contract the tongue to bring the prey back
202 to its mouth. Here, the soft actuator’s straightening and recovery process perfectly imitates the
203 chameleon tongue during hunting process. So, we added a small amount of semi-cured PDMS
204 on the top of the actuator to serve as the adhesive part of the tongue to the prey. Figure 3b shows
205 the height of the ants during the hunting process, where “0” represents the ground height level.
206 The hunting process can be divided into four parts: approaching, sticking, lifting and catching.
207 As an ant walks nearby, the soft actuator is turned on by input electricity, heated and then
208 stretched out to approach the ant. When the actuator is fully extended, it sticks to the ant’s body,
209 and the entire actuation process is finished. Then, the turn-off of the electricity inputs allows
210 the actuator starting to recovery process, and the ant is lifted up and can't escape from the

211 “tongue” because of sticky adhesion, which proves that the actuator can successfully and
212 accurately capture a moving insect. The entire capture process, shown in detail in Figure 3c and
213 Movie S3, is approximately 7 s while the instantaneous adhesion of the ant only takes less than
214 0.5 s. Benefiting from good flexibility and controllable deformation of the actuators, this work
215 provides a good example for the bionic animal tongue in the future.

216 In the plant kingdom, “soft actuators” on the plants’ body are still very common. Some
217 carnivorous plants, such as Venus flytrap and pitcher plants, are best known for controlling the
218 opening and closing of their organs to hunt preys. In addition, rattan plants around us also can
219 control the deformations of their whiskers or vines during growing. Rattan vines need to climb
220 or twine themselves around some branches. As the plant grows, it constantly looks for new
221 branches to twine around, making the whole body stronger. Interestingly, if another branch is
222 placed nearby, the well twined vine often opens part of its body and continue to twine around
223 the new branch, which reflects the wisdom of plant growth. Considering the same spiral shape
224 and deformable properties, we can also use our soft actuators or robot to mimic the plant's vines.
225 However, a single heating electrode can only control the deformation of the whole body with
226 one actuation mode, while in many cases, the robot with one actuator cannot be completely
227 twined around a branch by this simple actuation mode. Therefore, we introduced a double-
228 electrode system to realize piecewise independent control to make the soft robot more
229 intelligent (Figure 4a). The resistances of thermal actuator 1 and thermal actuator 2 are 275 Ω
230 and 304 Ω , respectively. As shown in Figure 4b, when the actuator 1 is on (5.5 V) and actuator
231 2 (5.5 V) is off, the root of the soft robot will straighten out, but the tip is still curved. Similarly,
232 when the actuator 1 is off and actuator 2 is on, the tip of the soft robot will straighten out, but
233 the boot is still curved (Movie S4). If both actuators are on and off at the same time, this soft
234 robot can be treated as a single-electrode version. The temperature distributions during the
235 piecewise control were also recorded, as shown in Figure 4d.

236 As shown in Figure 4e and Movie S5, a thin soldering wire is placed next to the soft
237 robot to simulate a branch that plants twine around. When both actuator 1 and 2 are turned on,
238 the device looks like a straight plant's vine. Firstly, we run the device in single-electrode mode
239 that means turning off both electrodes at the same time, and the actuator gradually curls up into
240 a spiral, trying to twine around the solder. At about 2 s, the root of the soft robot has been
241 successfully twined. However, the top of the soft robot can't finish twining when it is fully
242 recovered. The reason is that the top of the device recovers to spiral firstly, and the tip has
243 already finished the deformation when the root begins to twine around the solder. To realize
244 the successful performance as the true plant's behavior, the double-electrode mode is adopted.
245 As demonstrated in Figure 4f, the end of the device is under the branch. After turning actuator
246 2 on, the tip of the device opens slowly. Then, when the actuator 2 is turned off again, the tip
247 of the device begins to recover and place above the branch. Eventually, the soft robots
248 successfully twined itself around the artificial branch. These results also provide ample
249 evidences that electrically actuation can achieve precise control, regardless of the area of
250 radiation that light or heat-driven actuators requires.(29)

251 This work provides only biomimetic examples of individual organs. In the future,
252 depending on the principle of electronic control, more effective bionic actuators or robot could
253 be achieved by introducing new thermal-response or electrical-response deformable materials
254 and advanced structural or mechanical design.(38) In order to realize the real intelligent robots,
255 complex intelligent feedback and control system is needed,(39) that requires the cooperation of
256 more scientists in electrical engineering, computer science and control engineering.

257 **4. Conclusion**

258 In summary, we developed an ultrathin electrically controlled film for multifunctional
259 bionic soft actuators. The 45 μm -thick actuator, containing passive invariant layer, nanoscale
260 heating electrode and thermal expansion layer, exists in the initial state of a spiral structure.

261 Under different actuation voltages, unbending angle of the actuator can be precisely controlled.
262 Moreover, after 100 cycles, the actuator's behavior is still completely repeatable. Given the
263 similarities in structure and function, the actuators are used to imitate the chameleon's tongue
264 and the vine of the plant. The artificial tongue can quickly straighten out and capture alive active
265 ants. For plant biomimetic applications, the artificial vine with piecewise controllable double
266 electrodes can be twined around a thin wire very easily. The introduction of double electrodes
267 reflects the programmable and rich control ability of the electrical actuation, which will greatly
268 promote the controllability and flexibility of soft robots. Meanwhile, this work also provides a
269 good reference for the future development of bionic intelligent robot.

270 **Conflict of interest**

271 The authors declare that they have no conflict of interest.

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276 **Author contributions**

277 Dengfeng Li and Xinge Yu conceived the idea and designed the experiments. Xinge Yu
278 supervised the project. Dengfeng Li, Song Wang, Jiahui He performed the experiments and
279 characterizations. Zhaoqian Xie helped to perform the theoretical stimulation. All authors
280 discussed the results and commented on the manuscript.

281 **Appendix A. Supplementary materials**

282 Supplementary materials to this article can be found online at.....

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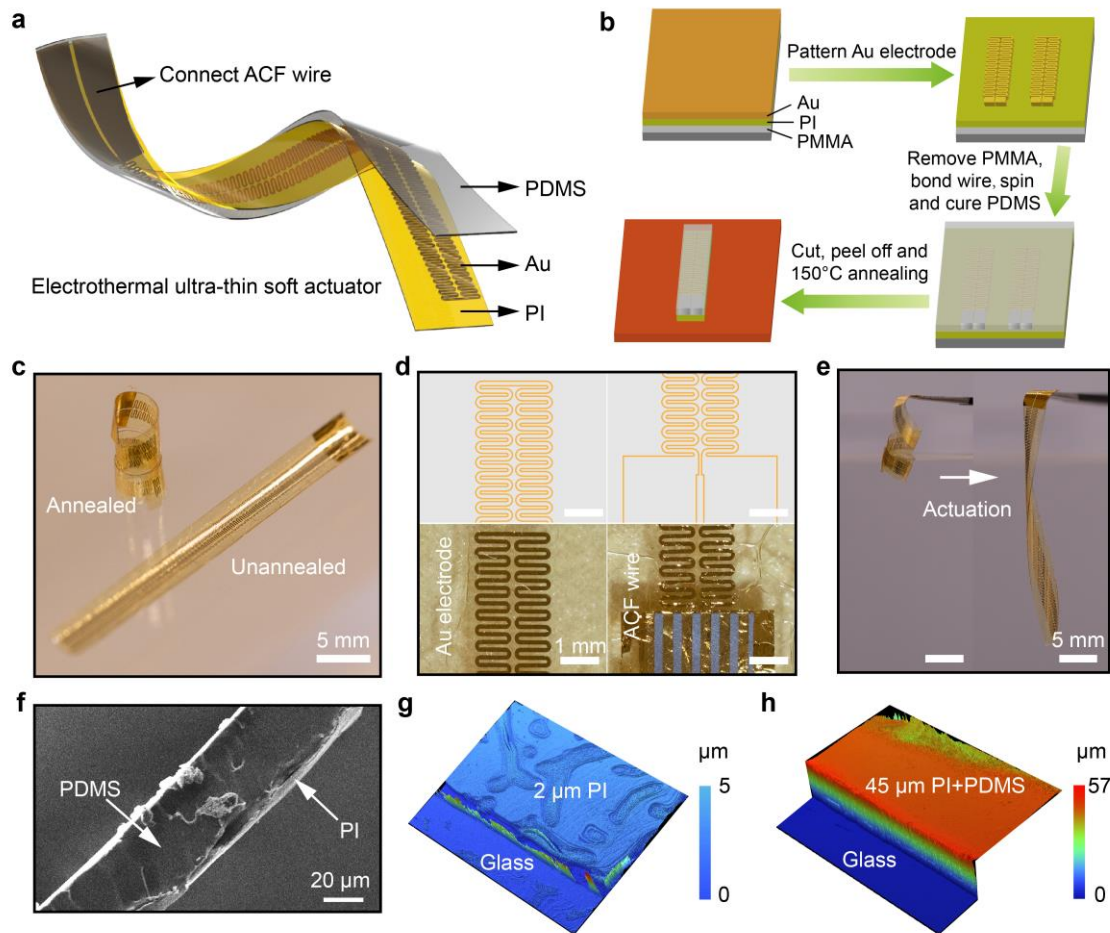
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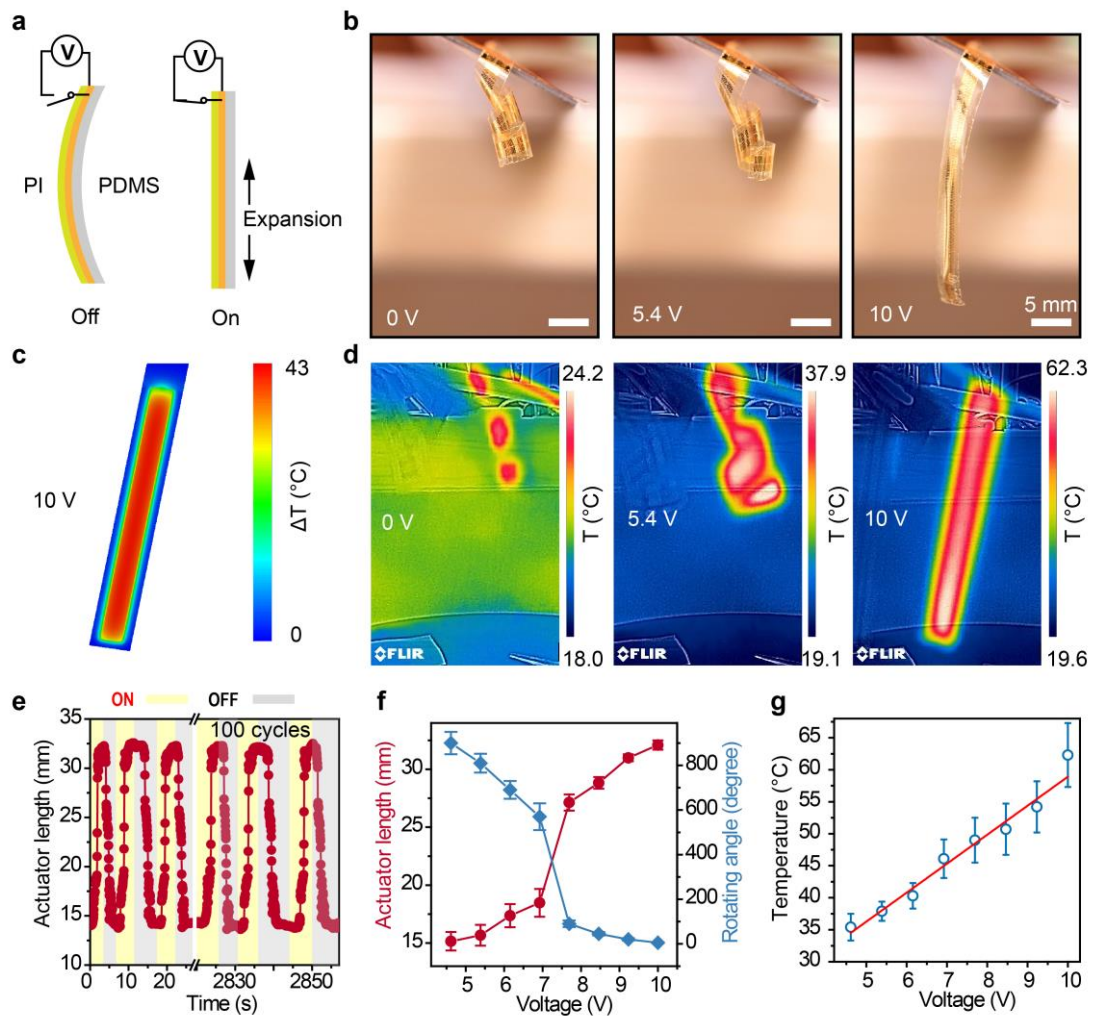
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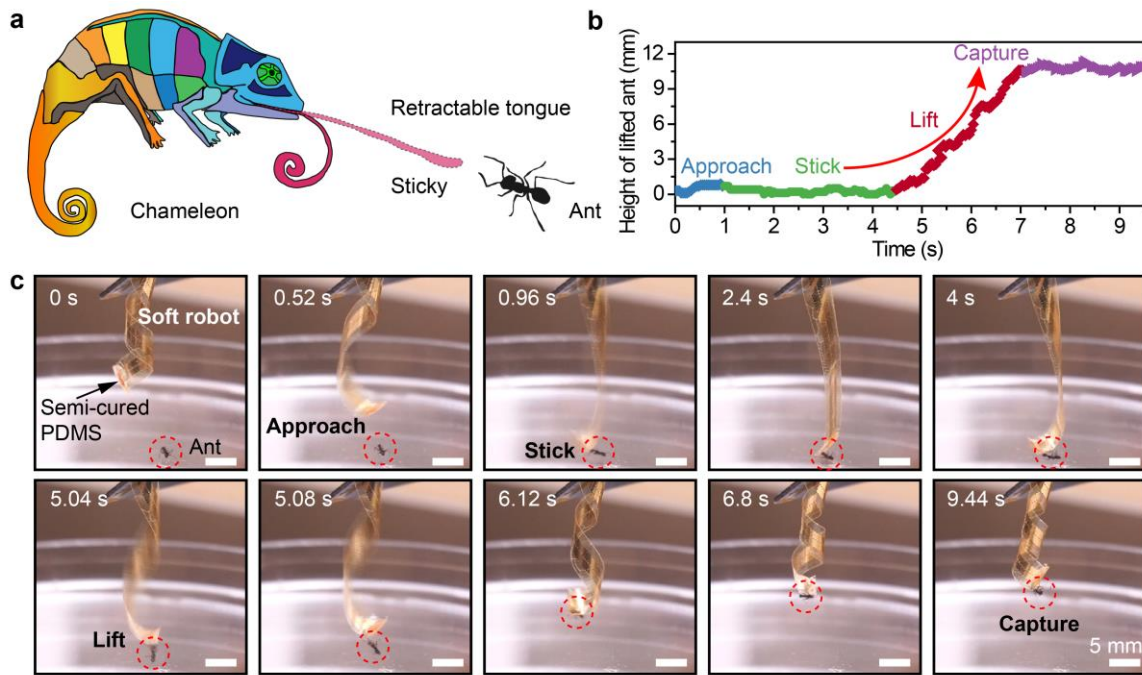
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389 **Figure 1. Electrothermal ultra-thin soft actuator.** (a) Schematic diagram of the soft actuator.
 390 The thin film actuator consists an invariant PI layer, a 200 nm thick gold electrode, and a
 391 thermally expandable PDMS layer. (b) Flow chart of the fabrication process. Through
 392 lithography and wet etching for the patterned gold electrode, spin-coating of the flexible
 393 polymer PI and PDMS layers, the actuator is finally annealed to form a spiral structure. (c)
 394 Optical images of the annealed and unannealed film. (d) Structural design and optical images
 395 of the gold electrode, and the ACF wire connection for external voltage supply. (e) Optical
 396 images of a pensile actuator undergoing the annealing process. (f) Cross-section SEM images
 397 of the film. (g) Surface morphology and thickness of PI layer. (h) Surface morphology and
 398 thickness of film actuator.



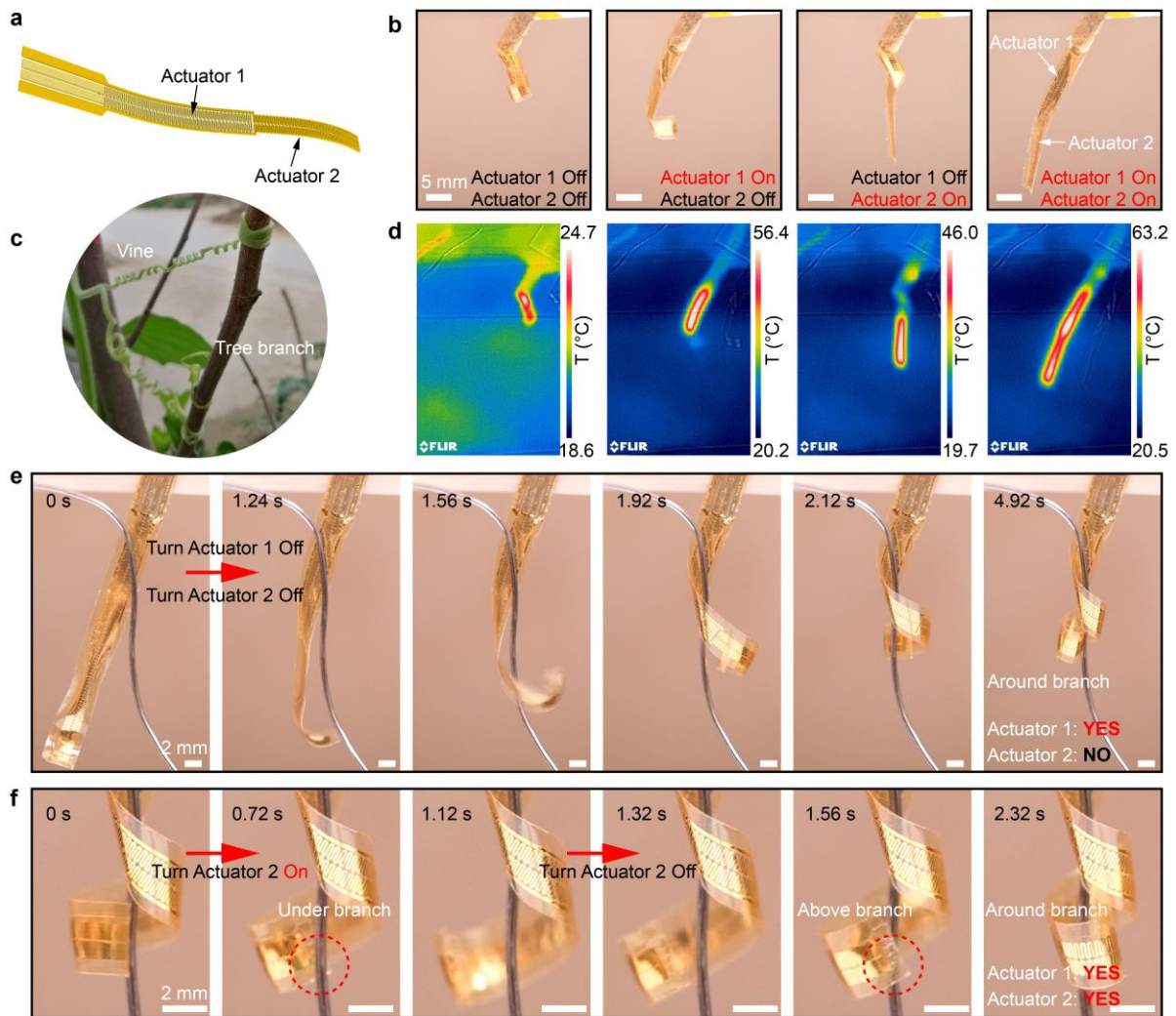
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400 **Figure 2. Deformation performance under different actuating voltage.** (a) Schematic
 401 illustration of the actuating principle. (b) Optical images of actuator under different voltage
 402 inputs. (c) Temperature distribution in an actuator under 10 V simulated based on the finite
 403 element analysis (FEA). (d) IR camera recorded temperature distributions in the actuator under
 404 different voltage inputs. (e) Changes of actuator length during actuating and recovery for 100
 405 cycles. (f) Relationships between actuator length/rotating angle and actuating voltages. (g)
 406 Relationship between the actuator temperature and actuating voltage.



407

408 **Figure 3. Capturing an ant to mimic the tongue of a chameleon.** (a) Schematic illustration
 409 of a chameleon hunting process, where the tongue is straightening out to capture an ant. (b) The
 410 height of the ant prey varies with the hunting process: approaching, sticking, lifting and catching.
 411 (c) The hunting process of the artificial chameleon tongue. Semi-cured PDMS are used to mimic
 412 the viscosity of the tongue. The moving ant is successfully hunted within 10s.



413

414 **Figure 4. Intelligent piecewise control of the soft robot for artificial vine.** (a) Design of the
 415 soft robot with double electrodes: actuator 1 and actuator 2. (b) Piecewise control by turning on
 416 each actuator separately. (c) A photo shows a vine of a gourd is twined around a tree branch.
 417 (d) Temperature distributions of the soft robot during piecewise control process. (e) Mimicking
 418 the twining process by the single-electrode mode that is turning on and off both actuators at the
 419 same time. (f) Achieving successful twining by piecewise control. Opening the top of the soft
 420 robot that fails to twine around the branch. The re-opening and closing allows the top to
 421 successfully twine around the branch as well.