In nature, animals or plants often use soft organs to move and hunt. Research works on bioinspired materials and devices have attracted more and more interest as which show the potential for future intelligent robots. As key components of soft robots, biomimetic soft actuators are adapted to greater requirements for convenient, accurate, and programmable controlling robots. Here, a class of materials and processing routes of ultrathin actuators are reported for bioinspired piecewise controllable soft robots, where the actuators associate with thermal-responsible soft silicone thin film with thickness as thin as 45 µm and electrically driven by well mechanical designed metallic thin film electrodes. Multiple electrodes in the robots in charge of individual segments control allow the soft robots exhibiting similar functionalities of animals or plants (for example, imitating the tongue of a reptile, such as chameleon to hunt moving preys, and mimicking vines to tightly wind around objects). These bionic results in the soft robots demonstrate their advantages in precise and flexible operation, which provides a good reference for the future research of intelligent soft actuators and robots.
other hand, electric-based actuators also offer great compatibility of electronics integration with the control system in the robots. Thermal induced deformations in actuators by inputting electricity to conductive wires has been reported for soft actuators.[23] However, the dimension, especially the thickness of these soft actuators are still way too big, that limits their applications in biomedical areas.[19,30] How to develop small size, ultrathin bioinspired soft actuators raises to be one of the key problems in soft robots, and two specific tasks are need to be solved: [1] developing thin-film actuators with ultrathin flexible electrodes; [2] endowing the biomimetic actuator ability of segmented piecewise control to enhance its flexibility and controllability in various application scenarios.

In this work, we developed an ultrathin film that are responsible to thermal inputs for soft actuators by combining flexible electronic processing technologies and mechanics designs. A bilayer thin film with significant difference of thermal expansion coefficient and a 200 nm-thick gold (Au) electrode was developed to serve as the actuator. The actuators exhibit a spiral shape with 900° bending in their original state, but can straighten out with electricity generated heat input through the electrode. To illustrate the advantages of the materials and mechanics in the soft actuators, we developed piecewise controllable soft robots based on these actuators by simply using multiple separated electrodes. These robots show great bionic capabilities, such as mimicking the tongue of a chameleon to hunt a moving ant, and simulating a plant’s vine to wind around a tiny pole. The precise programmed fabrication and piecewise control of the thin-film based soft actuators will provide important references for future bionic actuators and robots.

Figure 1a illustrates the schematic diagram of the ultrathin soft actuator and highlights the key materials used for it, consisting a 43 µm thick polydimethylsiloxane (PDMS) layer as the main body, a 200 nm thick Au electrode supported by a thin polyimide (PI, 2 µm thick) layer as thermal actuation part. Figure 1b shows the fabrication process of the soft actuator. The fabrication started on a clean glass substrate, where a thin poly(methylmethacrylate) (PMMA) layer was spin-coated and baked as the sacrificial layer. Then, poly(pyromellitic dianhydride-co-4,4’-oxydianiline), amic acid solution was spin-coated on the PMMA layer and annealed at 250 °C to form a 2 µm-thick dense PI film, as the passive deformation layer (Figure 1bii).[31] Next, sputtering deposition and photolithography defined 200 nm-thick Au thin layer on the PI layer formed the electro-thermal converting electrode (Figure 1biii).[32,33] Immersing the sample in acetone for 12 h allowed fully dissolving PMMA and thus we could easily peel off the thin film from the glass substrate. After bonding ACF wire on the Au electrode pins, the heating electrode could be supplied by external power (Figure 1d). Finally, the PDMS with per-polymer and cross-link agent ratio of 5:1 was spin-coated on the PI and Au electrode at spinning speed of 1000 rpm for 60 s and cured at 75 °C for 30 min (Figure 1biii). After cutting the edge of the sample into designed width of 4 mm, the actuator was peeled off with a straight shape (Figure 1c). In order to make the actuator own a stable steady state, an annealing process was carried out at 150 °C for 5 min (Figure 1biv). The high temperature annealing state made the PDMS layer being in excess of expansion, and thus resulted in a large shrinkage after cooling down to room temperature.[34,35] Figure 1c,e presents the state and shape of the actuator before and after annealing or actuation. The dynamic deformation process of the actuator on a hot plate at 150 °C can be seen in Movie S1 in the Supporting Information. Thus, an ultrathin electrically actuated soft actuator with a helical structure is obtain. Cross-section scanning electron microscopy (SEM) image of the actuator shows clear thin layer structure (Figure 1j), and the thickness of each layer was also measured and confirmed by an optical surface profiler to be 2 µm thick for PI (Figure 1k) and 43 µm thick for PDMS (Figure 1l).

To study the effect of PDMS thickness on the actuator deformation ability, we prepared a group of soft actuators with different thick PDMS layers ranging from 43 to 178 µm by simply controlling the spinning durations at a fixed speed of 1000 rpm. Their thicknesses were acquired and measured from SEM images (Figure S1, Supporting Information) and optical surface profiler (Figure S2, Supporting Information). The results showed that the deformation of PDMS is highly relevant with the thickness, where thicker layers induced the smaller deformations (Figure 1f,g). As shown in Figure 1f, the annealed 45 µm-thick actuator exhibits a 900° bending angle and the shortest length among all devices, demonstrating great performance of deformation and elongation during the actuating process. Else, the shape of the actuator is related to its width. As demonstrated in Figure 1h, the 45 µm-thick actuators are with widths from 2 to 8 mm. The wider actuators exhibit smaller spiral pitches per width (Figure 1j). The 8 mm-wide actuator tends to form a cylinder instead of a spiral. So, we select the 45 µm-thick and 4 mm-wide as parameters for the actuators. The heating area of the electrode is 2 mm in width and 32 mm in length (Figure S3, Supporting Information). After cutting into a 4 mm-wide stripe, the entire actuator could realize actual actuation on the 32 mm length, excluding the wire connection area. Figure 2a shows the original state and actuated state of the actuator under different voltage inputs, where we can find the actuator can fully straighten itself out under a voltage input of 10 V that equals to 179 mW since the resistance of the electrode is 558 Ω (Movie S2, Supporting Information). The deformation process derives from the significant difference in thermal expansion coefficient between PI and PDMS. The thermal expansion coefficient of positive PDMS layer is around 310 × 10⁻⁶ K⁻¹, that is more than 10 times greater than that of the passive PI layer with a value of 20 × 10⁻⁶ K⁻¹.[36] Thermal energy generates greater expansion in the PDMS than that of PI layer, and thus causes the actuator to bend towards the PI layer (Figure 2a,b). Actually, the electric-heat actuating based flexible electrodes is suitable for any thermal-response typed actuators, since the deformation layer can also be replaced by other materials, such as liquid crystal elastomer films, laminated graphene, etc.[32,30] Here, we choose PI and PDMS due to their excellent properties, including low lost, easy to obtain, compatible processing routes, and high expansion difference. The electrode in between PI and PDMS offers the actuators highly efficient thermal induced deformations compared to using external heat source such as a heat plate. Infrared (IR) camera recorded temperature information during the thermal actuating process provides quantitative data of the electric-thermal conversions, as shown in Figure 2d. The temperature reaches 62.3 °C under...
Figure 1. Electrothermal ultrathin soft actuator. a) Schematic diagram of the soft actuator, which consists of a passive layer (Pi), a layer of electrode (Au, 200 nm) and a thermally active layer (expandable PDMS). b) Flow chart of the fabrication process. c) Optical images of the actuator film before and after annealing. d) Design (up) and optical images (bottom) of the electrode and the ACF wire connection for external voltage supply. e) Optical images of a pensile actuator before and after actuation. f) Optical images of annealed actuators with different thickness and deformation angle. g) Relationship between thickness and spin time under 1000 rpm-speed for the actuators in (f). h) Optical images of 45 µm-thick actuators with different width from 2 to 8 mm. i) Relationship between the spiral pitch per width and actuator width. j) SEM image of the film cross-section. Surface morphology of the k) Pi layer and l) actuator film.
10 V voltage input, which is consistent with the finite element analysis (FEA) simulated results (Figure 2c). All the temperatures measurements were done in the air at 23 °C when the heat production and heat dissipation reach a balance. The detailed temperature changes of the actuator during the deformation process were recorded in Movie S3 in the Supporting Information. When the electricity inputs turn on, the actuators’ temperature rises to around 70 °C within 4 s and after a rapid decline due to the instant straightening out of the actuator, the temperature stabilizes to around 60 °C, which indicates that the electrode heating and thermal diffusion are already in equilibrium. After turning off the electricity inputs, the corresponding cooling down during the recovery process takes longer, about 8 s (Figure S6, Supporting Information). Doping nanomaterials with high thermal conductivity such as graphene and AgNWs into the polymer, would shorten the thermal diffusion time and thus accelerate the deformation of the actuator.36,37 Else, the actuator also exhibits good durability for long-term and multiple times operations, where the actuation capability still maintains the same after 100 cycles operation at 10 V (Figure 2e; Figure S7, Supporting Information). Since the actuation temperature did not exceed the annealing temperature of 150 °C, the heating would not cause changes of the material properties and the steady state of the structure. The maximum travel amplitude of the actuator is more than twice its initial body length of 14.5–32 mm, that lays a foundation for the various multi-functional bionic applications. Moreover, the actuator could elongate to different lengths in the range between 14.5 and 32 mm under controlled voltage inputs (Figure S4 and Movie S2, Supporting Information). The relationship between the elongate length and the voltage is summarized in Figure 2f, where we can find the relationship is stable and directed, which further reflects accuracy and precision of electrically actuation.

Figure 2g and Figure S5 (Supporting Information) show the great linear behavior of actuation temperatures as a function of input voltage, which provides the basis for a specific control. In addition, we also analyze the deformation process of the actuator during electrical heating and cooling in air. Figure S8 in the Supporting Information shows that it takes about 2 s to straighten out and 2.5 s to recover to the initial shape. During the straightening process, the actuator slowly elongates at the beginning but instantly straightens out in a short 0.2 s and then slowly turn to the final stable state. This phenomenon is due to an instantaneous release of the potential energy...
during the deformation process and then the actuator starts to oscillate from side to side due to inertia, which thus caused a “hump” during the actuation process. For the actuator, the sudden and large deformation, straightening and recovery process coincides with that of the chameleon tongue during prey capturing process. Inspired by nature, we used the ultrathin soft actuator as an artificial chameleon’s tongue to mimic the process of hunting insects, as demonstrated in Figure 3. We added a small amount of semi-cured PDMS on the top of the actuator to serve as the adhesive part of the artificial tongue to the prey. Figure 3b shows the height of the ants during the hunting process by the artificial actuator, where “0” represents the ground height level. The hunting process of the artificial actuator can be divided into four parts: approaching, sticking, lifting, and catching. As an ant walks nearby, the soft actuator is turned on by input electricity, heated and then stretched out to approach the ant. When the actuator is fully extended, it sticks to the ant’s body, and the entire actuation process is finished. Then, the turn-off of the electricity inputs allows the actuator starting to recovery process, and the ant is lifted up and can’t escape from the “tongue” because of sticky adhesion, which proves that the actuator can successfully and accurately capture a moving insect. The entire capture process, shown in detail in Figure 3c and Movie S4 (Supporting Information), is ≈7 s while the instantaneous adhesion of the ant only takes less than 0.5 s. The pulling force of the actuator during the recovery process was measured by contacting the tip of the actuator with the force sensor. (Figure S9, Supporting Information). The maximum force is 82.3 µN, which indicates that the actuator could lift an object with weight of 8.4 mg. Benefiting from good flexibility and controllable deformation of the actuators, this work provides a good example for the bionic animal tongue in the future. Developing a real artificial chameleon tongue would be a complex project. During hunting process, the chameleon makes coiling by retraction of the muscle and folding its tongue and the tip of the tongue also needs particular force for grasping the prey or object.[38] In addition to the deformation of the tongue, more efforts can be made to achieve the specific grasp by the tongue tip relying on both suction and adhesion force. Moreover, compared with true chameleon tongue, there is still a huge gap in the performance of this actuator from projection length percentage to peak projection velocity and then to force (Table S1, Supporting Information).[39,40] This gap provides more space for the future development of the actuators.

In the plant kingdom, “soft actuators” on the plants’ body are still very common. Some carnivorous plants, such as Venus flytrap and pitcher plants, are best known for controlling the opening and closing of their organs to hunt preys. In addition, rattan plants around us also can control the deformations of their whiskers or vines during growing. Rattan vines need to climb or twine themselves around some branches. As the plant grows, it constantly looks for new branches to twine around, making the whole body stronger. Interestingly, if another branch is placed nearby, the well twined vine often opens part of its body and continue to twine around the new branch, which reflects the wisdom of plant growth. Considering the same spiral shape and deformable properties, we can also use our soft actuators or robot to mimic the plant’s

Figure 3. Object capturing for the mimic of a chameleon’s tongue. a) Schematic illustration of chameleon’s hunting process: An initially cured tongue is straightening out to capture an ant. b) The height evolution of the ant prey during the hunting process. c) Photoshots of the artificial chameleon tongue capturing an ant. Semi-cured PDMS are attached to the tip of the actuator and functioned as the adhesive of chameleon’s tongue.
vines. However, a single heating electrode can only control the deformation of the whole body with one actuation mode, while in many cases, the robot with one actuator cannot be completely twined around a branch by this simple actuation mode. Therefore, we introduced a double-electrode system to realize piecewise independent control to make the soft robot more intelligent (Figure 4a). The resistances of thermal actuator 1 and thermal actuator 2 are 275 and 304 Ω, respectively. As shown in Figure 4b, when the actuator 1 is on (5.5 V) and actuator 2 (5.5 V) is off, the root of the soft robot will straighten out, but the tip is still curved. Similarly, when the actuator 1 is off and actuator 2 is on, the tip of the soft robot will straighten out, but the boot is still curved (Movie S5, Supporting Information). If both actuators are on and off at the same time, this soft robot can be treated as a single-electrode version. The temperature distributions during the piecewise control were also recorded, as shown in Figure 4d.

As shown in Figure 4e and Movie S6 (Supporting Information), a thin soldering wire is placed next to the soft robot to simulate a branch that plants twine around. When both actuator 1 and 2 are turned on, the device looks like a straight plant’s vine. Firstly, we run the device in single-electrode mode that means turning on/off actuators simultaneously. Only actuator 1 succeed in twining. f) Piecewise control of the actuators for achieving a successful twining. The bottom part of the robot fails to twine around the branch; Control of actuator 2 allows to re-open-and-close the actuator to successfully twine around the branch.

Figure 4. Intelligent piecewise control for an artificial vine. a) Design of the soft robotic arm with piecewise control via double electrodes indicated as Actuator 1 and 2. b) Piecewise control by controlling the input signal between two electrodes. c) A photograph of a gourd vine twining around a tree branch. d) Temperature distribution of the robotic arm upon electric stimuli. e) A mimic of the twining process by the robotic arm with single-electrode mode by turning on/off actuators simultaneously. Only actuator 1 succeed in twining. f) Piecewise control of the actuators for achieving a successful twining: The bottom part of the robot fails to twine around the branch; Control of actuator 2 allows to re-open-and-close the actuator to successfully twine around the branch.
has already finished the deformation when the root begins to twine around the solder. To realize the successful performance as the true plant’s behavior, the double-electrode mode is adopted. As demonstrated in Figure 4f, the end of the device is under the branch. After turning actuator 2 on, the tip of the device opens slowly. Then, when the actuator 2 is turned off again, the tip of the device begins to recover and place above the branch. Eventually, the soft robots successfully twined itself around the artificial branch. For this experiment, we need control two power supplies relying on our vision as feedback to adjust accordingly and achieve the function of piecewise control. These results also provide ample evidences that electrically actuation can achieve piecewise control, regardless of the area of radiation that light or heat-driven actuators requires.

Compared to the electrothermal actuators that reported in previous works (Table 1), this work presents advantages from three aspects, ultrathin thickness, low working temperature (below 70 °C) and the ability of piecewise control. The thin film based actuators are easier to form greater deformations with a relatively small power input. At the same time, piecewise control provides more flexibility to the actuators, allowing it to better perform its biomimetic functions. This work provides only biomimetic examples of individual organs. In the future, depending on the principle of electronic actuation, more effective bionic actuators or robot could be achieved by introducing new thermal-response or electrical-response deformable materials and advanced structural or mechanical design. In order to realize the real intelligent robots, complex intelligent feedback and control system is needed, that requires the cooperation of more scientists in electrical engineering, computer science and control engineering.

In summary, we developed an ultrathin electrically controlled film for multifunctional bionic soft actuators. The 45 µm-thick actuator, containing passive invariant layer, nanoscale heating electrode and thermal expansion layer, exists in the initial state of a spiral structure. Under different actuation voltages, unbending angle of the actuator can be precisely controlled. Moreover, after 100 cycles, the actuator’s behavior is still completely repeatable. Given the similarities in structure and function, the actuators are used to imitate the chameleon’s tongue and the vine of the plant. The artificial tongue can quickly straighten out and capture alive active ants. For plant biomimetic applications, the artificial vine with piecewise controllable double electrodes can be twined around a thin wire very easily. The introduction of double electrodes reflects the programmable and rich control ability of the electrical actuation, which will greatly promote the controllability and flexibility of soft robots. Meanwhile, this work also provides a good reference for the future development of bionic intelligent robot.

Table 1. Comparison of different electrothermal soft actuators in materials and performances.

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<tr>
<td>LCE-B/Au/PI</td>
<td>500</td>
<td>8.2</td>
<td>95</td>
<td>Yes</td>
<td>NA</td>
<td>[29,41]</td>
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<td>PDMS/AgNW/PI</td>
<td>200</td>
<td>2</td>
<td>160</td>
<td>NO</td>
<td>NA</td>
<td>[36]</td>
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<td>150</td>
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<tr>
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<td>20</td>
<td>112</td>
<td>NO</td>
<td>0.32</td>
<td>[43]</td>
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<tr>
<td>PDMS/Au/PI</td>
<td>45</td>
<td>10</td>
<td>62</td>
<td>Yes</td>
<td>0.082</td>
<td>This work</td>
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Experimental Section

Fabrication of Thin-Film Actuator: First, poly(methylmethacrylate) (PMMA) solution (20 mg mL⁻¹, chlorobenzene solvent) serving as a sacrificial layer was spin-coated on a quartz glass substrate and baked at 200 °C for 20 min. Afterward, poly(pyromellitic dianhydride-co-4,4'-oxydianiline), amic acid solution (Sigma-Aldrich) was spin-coated on the PMMA layer at 3000 rpm for 30 s and annealed at 250 °C for 30 min. The formed 2-µm polyimide (PI) film with low heat expansion coefficient was served as passive deformation layer. 200-nm Au was deposited onto the PI film by a sputtering system (Quorum Sputter coater, Q150 T S) and then coated by photoresist (AZ 3214) at 3000 rpm for 30 s with soft-bake of 110 °C for 3 min. After exposure under ultraviolet light with a film mask and development in AZ 300MIF developer, the Au layer was etched in the gold etchant (I₂/KI solution) to form the desired pattern. After removing the PMMA sacrificial layer in acetone for 12 h, the anisotropic conductive film (ACF) wire was connected with the acquired ultrathin gold electrode for further heating by external voltage source. Finally, the polydimethylsiloxane (PDMS, Sylgard 184, per-polymer and cross-link agent = 5:1) was spin-coated at 1000 rpm for 60 s on the electrode and cured at 75 °C for 30 min, serving as the positive deformation layer. After cutting and peeling the film off the substrate, the film was annealed on the hotplate at 150 °C for 10 min and cooled to room temperature in air to achieve a stable state. Thus, the helical actuator was acquired.

Actuation: After bonding the ACF wire to a designed printed circuit board (PCB), the actuator was connected with a DC power source. When a certain voltage was applied, the actuator was heated and straightened to present a state of elongation. All deformation processed were recorded with a macro-lens camera (SONY). Temperature changes of the heated actuators were recorded by an infrared camera (FLIR-C7200).

Characterization: The cross-section microstructure of the actuator was observed by using a scanning electron microscope (JEO/L/JSM-5600). The thickness of actuators with different PDMS layers was measured by using the optical surface profiler (Olympus, Veeco/Wyko NT9300).

Thermal Simulation: Electrothermal simulation was acquired based on the finite element analysis (FEA). The commercial software ABAQUS was used to study the temperature distribution of the one-electrode device with the heat convection coefficient (26 W m⁻² K⁻¹) of air and an input power of 0.18 W. The entire device was electrothermal modeled by hexahedron heat-transfer elements (DC3D8). Mesh convergence of the simulation was ensured for all the cases. The thermal conductivity, heat capacity and mass density used in the simulations were 315 W m⁻¹ K⁻¹, 130 J kg⁻¹ K⁻¹, and 19 300 kg m⁻³ for Au, 0.15 W m⁻¹ K⁻¹, 1460 J kg⁻¹ K⁻¹, and 965 kg m⁻³ for PDMS, 0.12 W m⁻¹ K⁻¹, 1000 J kg⁻¹ K⁻¹, and 1420 kg m⁻³ for PI.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
bioinspired electronics, flexible electronics, piecewise control, soft robotics, thermal actuators

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