

Wireless Autonomous Soft Crawlers for Adjustable Climbing Actuation

Lei Tian^{†*}, Ji-Ji Tan[†], Wei-Liang Dong, Bo Yang, Cui-Hua Li, Dai Wang, Hai-Yu Huang, Xin-Tong Li, Cai-Zhen Zhu^{*}, and Jian Xu

Institute of Low-Dimensional Materials Genome Initiative, College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518060, China

Electronic Supplementary Information

Abstract Artificial soft actuators, featured with non-equilibrium internal circumstance and fast, programmable shape transformations, have attracted strong research interest recently due to their flexibility, highly controllable, and designability. However, wireless soft actuators, achieving the locomotion on different large slopes with multiple energy conversion, have been rarely reported. Herein, we create a asymmetric bilayer strategy to construct autonomous soft crawler *via* “breathing” moisture to motivate the mechanical deformation. The soft crawlers present conspicuous performances including periodic tumbler locomotion predicted *via* improved Timoshenko’s equation, multiple reversible shape-morphing (circle, helix, despiralization, etc.) determined by their fiber orientation, controlled drive mode (front drive and rear drive) and rapid climb speed (4.76 cm/min) at wide slope angles. Through architecture design, they can be series-wound or shunt-wound to construct multi-joint complex actuators. Besides climbing, a intelligent soft ring-pull with admirable cycle performance for preventing overheating or something untouchable, has been proposed. The soft crawlers also realize multiple energy conversion to be actuated by light irradiation. We envision that this soft crawler system has an enormous potential in intelligent machine, microscopic diagnosis and treatment, biosensing, energy harvesting and conversion.

Keywords Soft crawler; Tumbler locomotion; Reversible shape-morphing; Autonomous; Energy conversion

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INTRODUCTION

The controllable autonomous action of living organisms provides us with numerous inspirations in intelligent materials that can create a sequence of motions in response to an external stimuli such as magnetic/electric fields,^[1–3] temperature,^[4] solvent,^[5] humidity^[3,6] and light.^[7,8] Mimicking these intelligent responses, artificial soft actuators, featured with non-equilibrium internal circumstance and fast, programmable shape transformations, have attracted strong research interest recently due to their flexibility, highly controllable, and designability.^[9] Following this concept, the actuator family is booming, and original actuators including pneumatic actuator,^[10] flapping-wing robot,^[11,12] microscopic robotic,^[13] lionfish-inspired robot,^[14] terrestrial-aquatic microrobot,^[15] octobot,^[16] also artificial flytrap^[17] have been created. Praiseworthy, as a typical example, a deep-sea vehicle gathered artificial muscle drive control and autonomous swimming of the soft robot, has been realized,^[18] which motivates the

development of the actuator to a broader space containing microswimmers,^[19,20] electronic skins,^[21] target capture/release^[7,22] and biomimetic actuations.^[23]

Notably, as an illustrative example of gastroscop capsule robot, soft actuators have been proven enormous potential for real-world applications.^[9] However, at this stage, most of soft actuators possess only the ability of unsustainable single motion on a flat surface, finitely converting external stimuli into mechanical deformation.^[24] For example, spiropyran polymers as photoswitches demonstrate a unidirectional crawling movement on a ratcheted PDMS surface.^[25,26] Also under light irradiation, one can realize the wireless control including shape morphing,^[27–29] phototropic moving^[30] and self-oscillation.^[31] And even more interesting, inchworm-inspired triboelectric soft robot has been created by rotary freestanding triboelectric nanogenerator converted from mechanical energy.^[32] The difference is that these wired soft robots have the ability to climb slopes with different angles. As a research hotspot, some recent studies have focused on the design of soft crawlers that are capable of directional crawling on a concrete ramp. For example, stretchable kirigami-skinned crawlers have been fabricated to efficiently crawl on a 15° ramp by the buckling-induced directional frictional properties, which are actuated pneumatically using air.^[33] However, few researchers have developed wireless soft actuators for multiple energy conversion converting external

* Corresponding authors, E-mail: leitian@szu.edu.cn (L.T.)
E-mail: czzhu@szu.edu.cn (C.Z.Z.)

[†] These authors contributed equally to this work.

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energy into potential energy and mechanical energy, achieving the locomotion on different large slopes.

Inspired by the caterpillars in nature tramping over hill and dale, a asymmetric bilayer strategy armed with the ability to independently climb *via* reversible “exhales and inhales” is proposed. Following this, we here report a wireless artificial soft crawler based on a bilayer film architecture composed of polyimide (PI) and ionic polymer (PSO) fibers. The PI film as the supporting layer provides the skeleton structure of soft crawlers, while humidity-sensitive PSO fibers perform the actuator operation *via* “breathing” moisture to motivate the mechanical deformation. Under temperature control, the PSO/PI bilayer film presents a periodic tumbler phenomenon, opening the beginning of autonomous soft crawler. The major innovation of this study is that we take full advantage of reversible deformation-recovery to endow PSO/PI film with the capacity of autonomous climbing. Additionally, these soft crawlers created with clear division of work possess the distinctive feature, which can alternately proceed front drive and rear drive inspired by four-wheel-drive vehicles. Through architecture design, they can be series-wound or shunt-wound to construct multi-joint complex actuators. Furthermore, they also possess admirable cycle performance of actuating motion used as a soft ring-pull. In consideration of multiple energy conversion, we also demonstrate that the soft crawlers can be actuated by illumination of simulated solar, indicating their enormous potential in multi-scene driven mode and intelligent energy-harvesting system.

EXPERIMENTAL

Materials

2-Acrylamido-2-methylpropane sulfonic acid (AMPS, 98%, Shanghai Macklin Biochemical Co., Ltd., China), acrylonitrile (AN, 99%, Shanghai Macklin Biochemical Co., Ltd., China), azobisisobutyronitrile (AIBN, Shanghai shanpu Chemical Co., Ltd., China) was purified by recrystallization in methanol solution. Dimethyl formamide (DMF) and methanol were purchased from Shanghai Macklin Biochemical Co., Ltd., China used directly without further purification. Deionized water was produced by an apparatus for pharmaceutical purified water (Aquapro Co., Ltd., China). polyimide film (PI, Wuxi Encyclopedia Electronic Materials Co., Ltd., China).

Preparation of PSO Copolymer

In a typical process, AMPS and AN with different ratios were dissolved in a mixture of DMF/methanol in a 250 mL three-neck round-bottomed flask. Then, 0.1 g of AIBN dissolved in 1.0 g of DMF was added to the above system. The polymerization was performed at 70 °C and maintained for 12 h. The resulting polymer was dried by rotatory evaporation, and stored in a 60 °C oven.

Preparation of PSO/PI Bilayer Film

PSO with different content ratios was dissolved in methanol and DMF solutions of equal mass, and then stirred at 65 °C for 12 h to obtain a solution with 40% spinnability. After magnetic stirring at 65 °C for 12 h, the homogeneous solution was obtained. Attach the PI membrane to the collector. A single-capillary spinneret was loaded into a syringe pump (YSP-101, YMC) with a 22-G metallic needle with a feed rate of 1.0 mL/h

and applied a voltage of 15.6 kV. The collector with PI film is 9 cm away from the spinneret. All the experiments were carried out at about 45 °C and relative humidity of 30%–50%.

Tumbler Locomotion

The bilayer film was cut into a rectangular spline with a length of 73.41 mm and A width of 6.23 mm. The spline was placed in a relatively closed thermal platform (PI film direction downward). By adjusting different temperatures, the bending angle of the spline was collected, and its bending curvature was calculated by formula conversion.

Climbing Experiment of Soft Crawlers

The stepped slope consists of two parts. The bottom is a 3 mm thick wooden board with a length and width of 200 and 110 mm, respectively. The top is a cardboard board. The bilayer films were cut into 8 mm wide splines and 90 mm long splines and placed on slopes respectively. The ramp is placed on a heated platform with a glass cover, by adjusting the angle of the slope, the climbing data of soft crawler under two exchange modes of heating and cooling (hot air/cold air blown by blower) were collected.

Characterization

Scanning electron microscopy (SEM)

The fiber morphology was captured by SEM (JSM-7800F & TEAM Octane Plus). The samples were coated on the conductive adhesive and sputtered with platinum by a JEC-3000FC auto fine coater at a current of 30 mA for 90 s before examination.

Dynamic stability control

Differential scanning calorimetry (DSC) is measured using the MODEL DSC-200F3 produced by Netzsch-Geratebau GmbH. Data were collected between –50 and 400 °C at a heating rate of 10 °C/min and two cycles were tested.

Mechanical tensile tests: the measured data of CMT-1203 produced by Zhuhai Sansi-Taijie Electrical Equipment Co., Ltd. are used in Mechanical tensile tests, and the tensile rate is 2 mm/min.

Xenon lamp light source

The Solar-500 xenon lamp produced by Beijing Newbit Technology Co., Ltd. is adopted, and the light intensity is 4.3 kW/m² when the distance between the light source and the object is 300 mm by adjusting the light source.

Infrared thermal imager

Ti480 PRO infrared thermal image produced by FLUKE Company was used to collect experimental data.

RESULTS AND DISCUSSION

Design of Soft Crawlers

Most current actuators can be got arbitrary play on the flat surface, such as rolling,^[34] bending,^[35] spiralling,^[31] jumping,^[36] and so on. The sustainable actuating movement mainly depends on interfacial thermal stress,^[24] elastic stress (swelling mismatch),^[37] or molecular transformation.^[28,38] Therefore, driving soft actuators to directional controlled climbing in a mild form is rarely reported, but is the original intention of design in this work.

The representative climbing phenomenon in nature comes from the insect larva, like a caterpillar showing in Fig. 1(a), which completes a crawl undergoing shrinking and stretch-

ing steps to move forward. The climbing procedure is inundated with the accumulation and release of energy. Following this, ionic polymer fibers with orientation structure are formed on PI substrate *via* electrospinning, obtaining a flexible bilayer film (Figs. S1–S3 in the electronic supplementary information, ESI). The ionic polymer layer possesses abundant sulfonic acid (SO_3^-) group, indicating strong humidity sensitive property. By means of adjusting the ambient humidity *via* temperature, the bilayer film can realize the repeated bending and recovering as shown in Fig. 1(b). To simulate the crawling movement of caterpillars, the bilayer film is armed with polypropylene (PP) feet at both ends (Fig. 1c). It should be noted that the bending procedure relies on the orientation structure of PSO ionic fibers. As SEM image shown in Fig. 1(d), horizontal orientation along the long axis promotes its bending action, otherwise it is stationary.

The driving mechanism, called as a reversible “exhalation and inhalation” of moisture, has been proposed in Fig. 1(e). The “breathing” process causes the shrinking and stretching of polymer chains under the direct influence of temperature, accompanied by the asymmetric actuating of PI layer. Hence, the periodic bending motion of PSO/PI bilayer film ultimately ascribes to the deformation mismatch. Meanwhile, the tunable fiber orientation leads to multiple reversible shape-morphing of soft crawlers (Fig. 1f, Fig. S4 in ESI). A helical bilayer film is generated when the orientation is 45° from the direction of extension, while a circle is formed with the horizontal orientation. More interestingly, the helical bilayer film can present the rotation in place to complete the despiralization (Movie S1 in ESI). Combined with the above analysis, both the biological behavior and bilayer film features encourage us to establish a actuator platform to realize directional

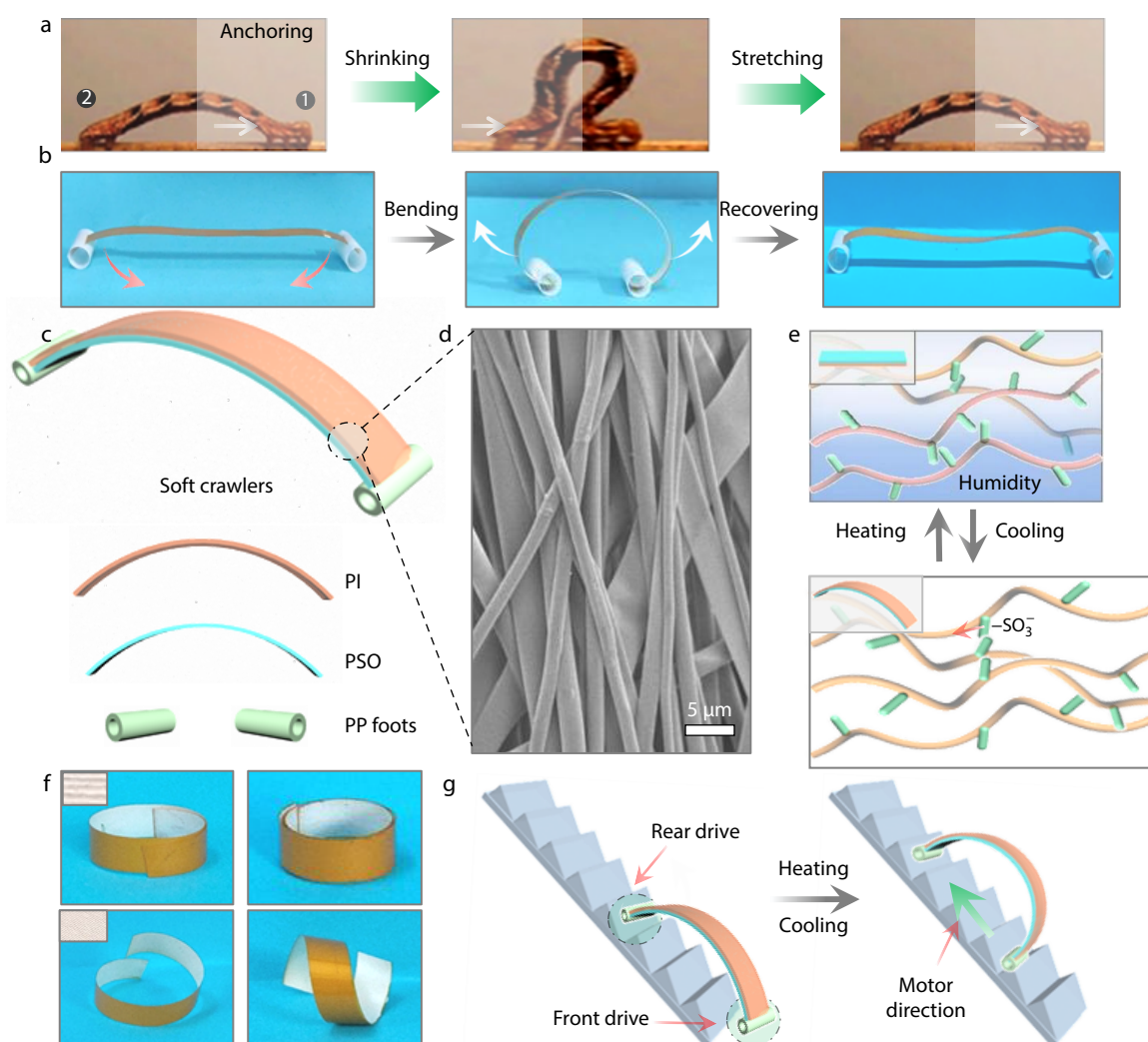


Fig. 1 The design concept and actuating principle of soft crawlers. (a) The photographs showing a locomotion cycle of a caterpillar including shrinking and stretching with a fixed side; (b) Images of designed soft crawlers showing the bending and recovering procedures; (c) The architecture of soft crawlers prepared from bilayer films composed of PI and PSO fibers, and PP tubes served as their feet; (d) SEM image of PSO fibers showing a horizontal orientation along the long axis; (e) Schematics of the working mechanism demonstrating the gradient change in humidity caused by temperature; (f) Orientation induced reversible deformation. Horizontal orientation: 1-2-turn circle. Oblique orientation: helix. (g) Schematic illustration of the locomotion mechanism of soft crawlers, showing a locomotion pattern of “front-rear drive”.

and autonomous crawling on different slopes as shown in Fig. 1(g).

Soft Tumblers from Reversible Shape-morphing

Based on the above findings, PSO/PI bilayer films possess excellent thermal responsiveness. Fig. 2(a) shows the original and flat PSO/PI film with a distinct double membrane structure. To better control their actuating, a hot table, with temperature gradient that has been confirmed by infrared imaging in Fig. 2(b), is used. When the bottom temperature reaches 50 °C, the PSO/PI bilayer film starts at both ends and bends towards the middle (Fig. 2d), quickly forms a half ellipse. The both ends keep bending, but do not meet. As shown in Fig. 2(e), due to the anisotropy, when one end is higher than another one, the PSO/PI bilayer film is going to swing towards the latter. Subsequently, the latter end becomes higher once it touches

the bottom, while the PSO/PI bilayer film would swing back. The bending and swinging of these soft tumblers we named (see Fig. S5 and Movie S2 in ESI) are detailedly explained in Fig. 2(c) using the mechanism of repetitively transformed barycenter. Moreover, we further develop a theoretical model as shown in Eq. (1) based on Timoshenko's equation to investigate tumbler movement, where the difference is taking temperature as the main parameter.

$$\kappa = \frac{F(T)}{h_2} \cdot \frac{mn(1+m)}{1+4mn+6m^2n+4m^3n+m^4n^2} \quad (1)$$

where κ is the curvature of bilayer actuators; h_2 is the thickness of PI substrate; $F(T)$ is a function of temperature; m is the thickness ratio of two layers; n is the ratio of Young's modulus. To simplify the model, the correlation between actuation strain and mechanical properties to humidity is negligible.

As a result, a three-dimensional (3D) graph of the curvature

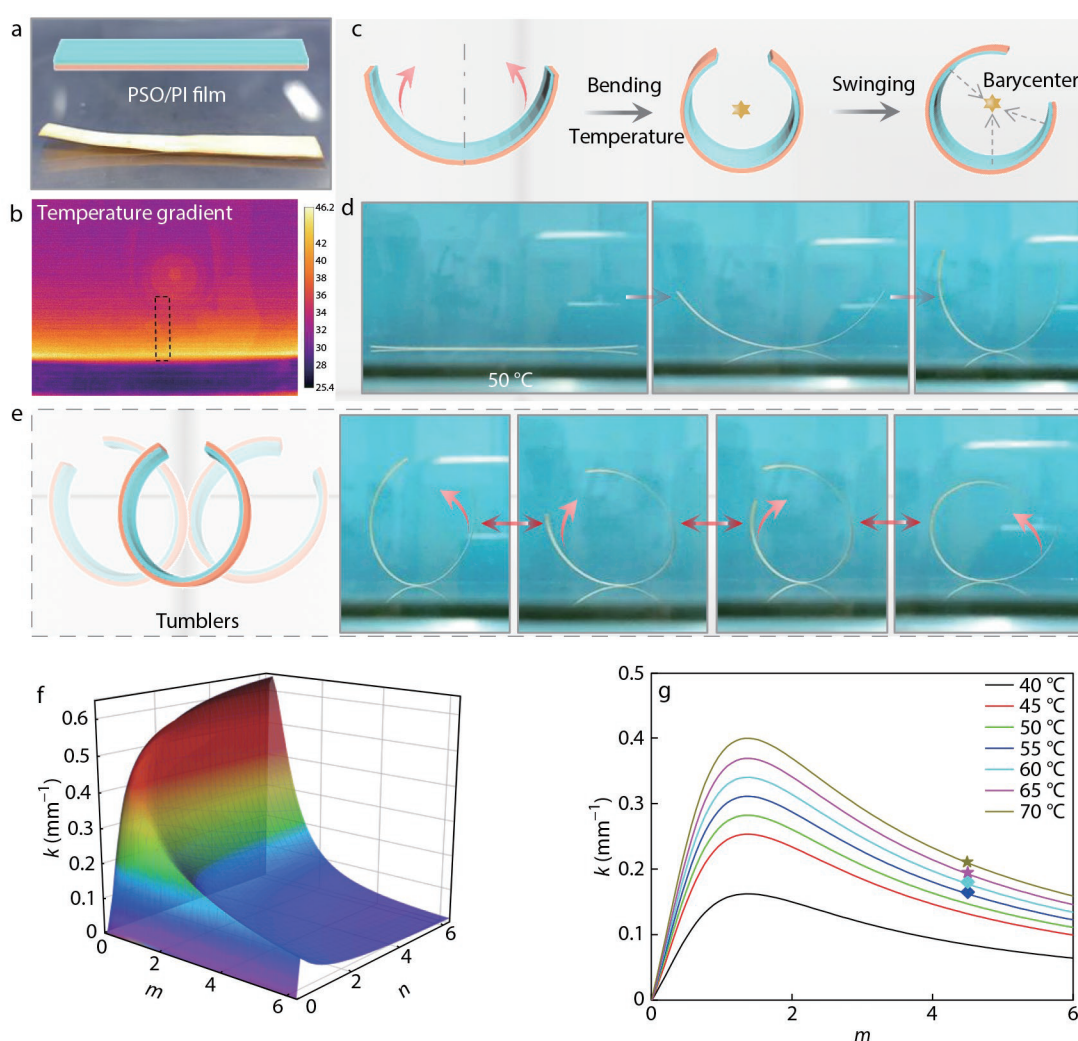


Fig. 2 Soft tumblers. (a) Image of primitive shaped PSO/PI bilayer film; (b) Infrared imaging photograph of PSO/PI film standing on the hot table, demonstrating a temperature gradient change in the vertical orientation which induces the asymmetrical deformation; (c) Schematic illustration indicating the principle of swing during the temperature-induced deformation; (d) Images showing the bending procedure from original planar film in 40 s; (e) Furthermore, bended PSO/PI film presents tumbler nature with changing center of gravity; (f) 3D plot of the curvature κ as a function of the thickness ratio m and the ratio of Young's modulus n at 50 °C according to the Timoshenko Eq. (1), where κ with vertical orientation is positive; (g) Corresponding κ predicted by the Timoshenko equation as a function of $n=0.0928$ at different temperatures.

κ as a function of two influencing parameters (m and n) at 50 °C is shown in Fig. 2(f). On this basis, we can obtain complete distribution map of curvature κ . Therefore, the curvature of a certain bilayer film can be accurately predicted according to their parameters m and n . In this work, n of PSO/PI bilayer film can be measured which is a fixed value. Then, the 2D curves of PSO/PI bilayer film at various temperatures are generated as shown in Fig. 2(g). There is an optimal m for the largest curvature κ . Additionally, one conclusion we can draw is that curvature κ is increased with the increasing of temperature in the same m case. To verify the accuracy of the curve, we measure four experimental values as the points showing in Fig. 2(g), which is close to the theoretical analog value. It indicates that this 3D model can excellently predict the loco-

motion of soft tumblers.

Temperature-triggered Soft Crawlers

Combining the periodic tumbler locomotion, a soft crawler with the length of l is prepared as shown in Fig. 3(a). The bending structure indicates that PSO/PI bilayer film not only possesses excellent flexibility, but also presents skeleton support performance. To enhance the support force and definite crawling distance, a stepped slope with different angles is designed. The climbing process of soft crawlers is shown in Figs. 3(b) and 3(c) and Movie S3 (in ESI). At the starting position, a 7 cm-long soft crawler can go up three stairs before heating (Fig. 3b, 0 s). Fig. 3(c) shows the infrared images of the temperature changes during the climbing. When heating the actuator position, the around temperature as shown in Fig. 3(c) is rising

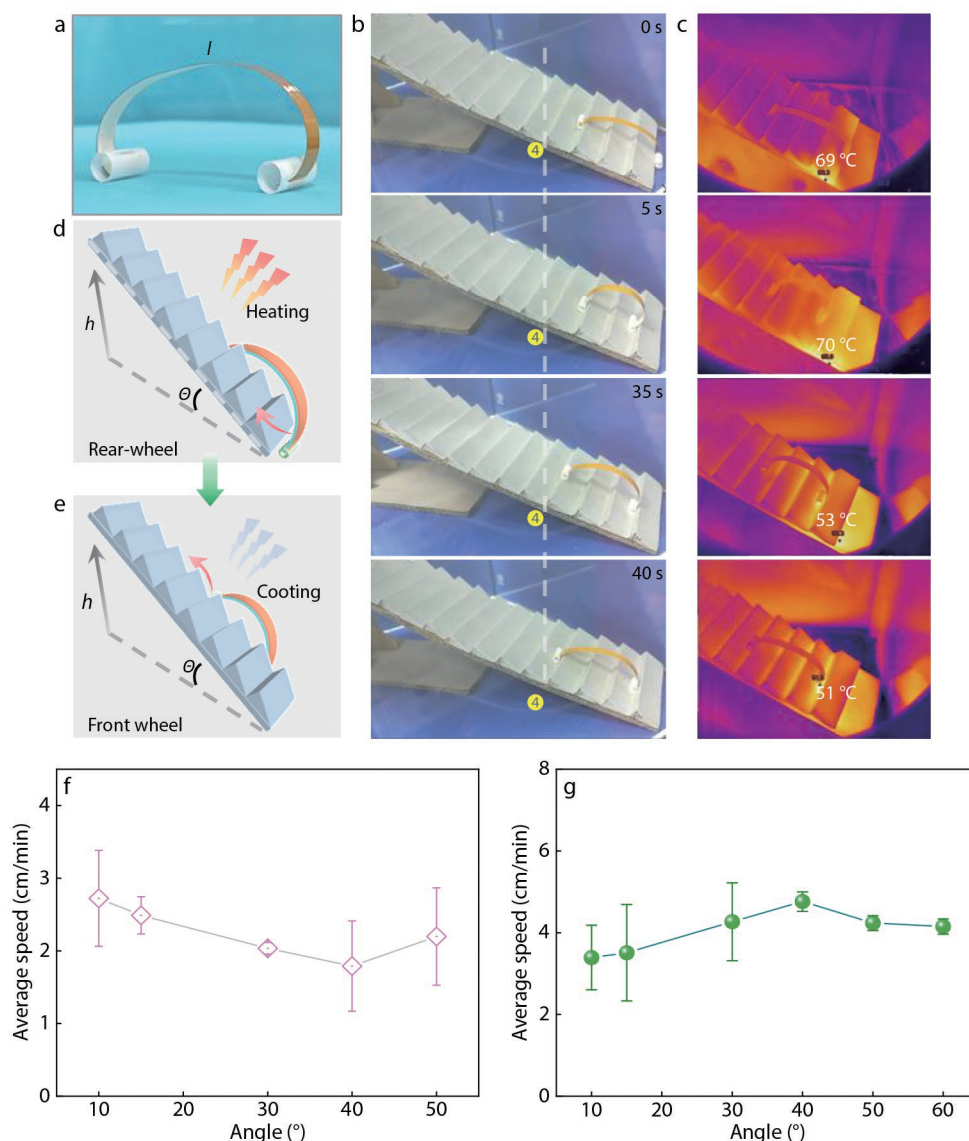


Fig. 3 Temperature-triggered wireless soft crawlers. (a) A photograph of prepared soft crawlers with l cm in length; (b) Images of temperature-triggered autonomous soft crawlers under periodic heating and cooling; (c) The corresponding infrared images showing the temperature changes induced the climbing. The inserts label the real-time temperature. (d, e) Schematic illustration showing the wirelessly actuated climbing process on the slope with a tunable angle. The soft crawlers first perform rear-wheel via heating, and then front-wheel during cooling; (f, g) The average speeds of soft crawlers with 7 cm and 9 cm in length at different angles θ .

rapidly to 70 °C. The rear foot of soft crawlers, regarded as “rear wheel”, first start to bend and then climb up one stair after 5 s (s). Fig. 3(d) demonstrates the climbing mechanism that, we called “rear drive”, the heating triggers the actuating of rear wheel. Then, the soft crawler is fast cooled down to 53 °C after 35 s. Meanwhile, the “front wheel” of bended soft crawler recovers rapidly, while the “rear wheel” is fixed. The “front drive” has been shown in detail in Fig. 3(e). Thus, the soft crawler accomplishes one cycle of crawling, and climbs one stair within 40 s.

In order to further investigate the climbing locomotion, the climb speed has been detailedly investigated in Figs. 3(f) and 3(g). The length of soft crawlers has great influence on their span on the slope. From Figs. 3(f) and 3(g), the 9 cm-long soft

crawler, whose climb speed can reach to 4.76 cm/min, is generally faster than the 7 cm-long (1.58 times). Furthermore, one advantage is that the soft crawler is basically not restricted by slope angle. The climbing locomotion can maintain high speed at a 60° slope (Fig. 3g). This finding will further expand the application prospect of these soft crawlers.

Thermo-responsive Complex Soft Actuators

According to the actuating mechanism, we can take advantage of PP foot to connect PSO/PI film, expanding the tailorable actuators. As shown in Fig. 4(a), a series-wound crawler is fabricated by adding the articulation point. Hence, there is more than two reversible deformations during a actuating period. However, we find that the climb speed has a certain reduction

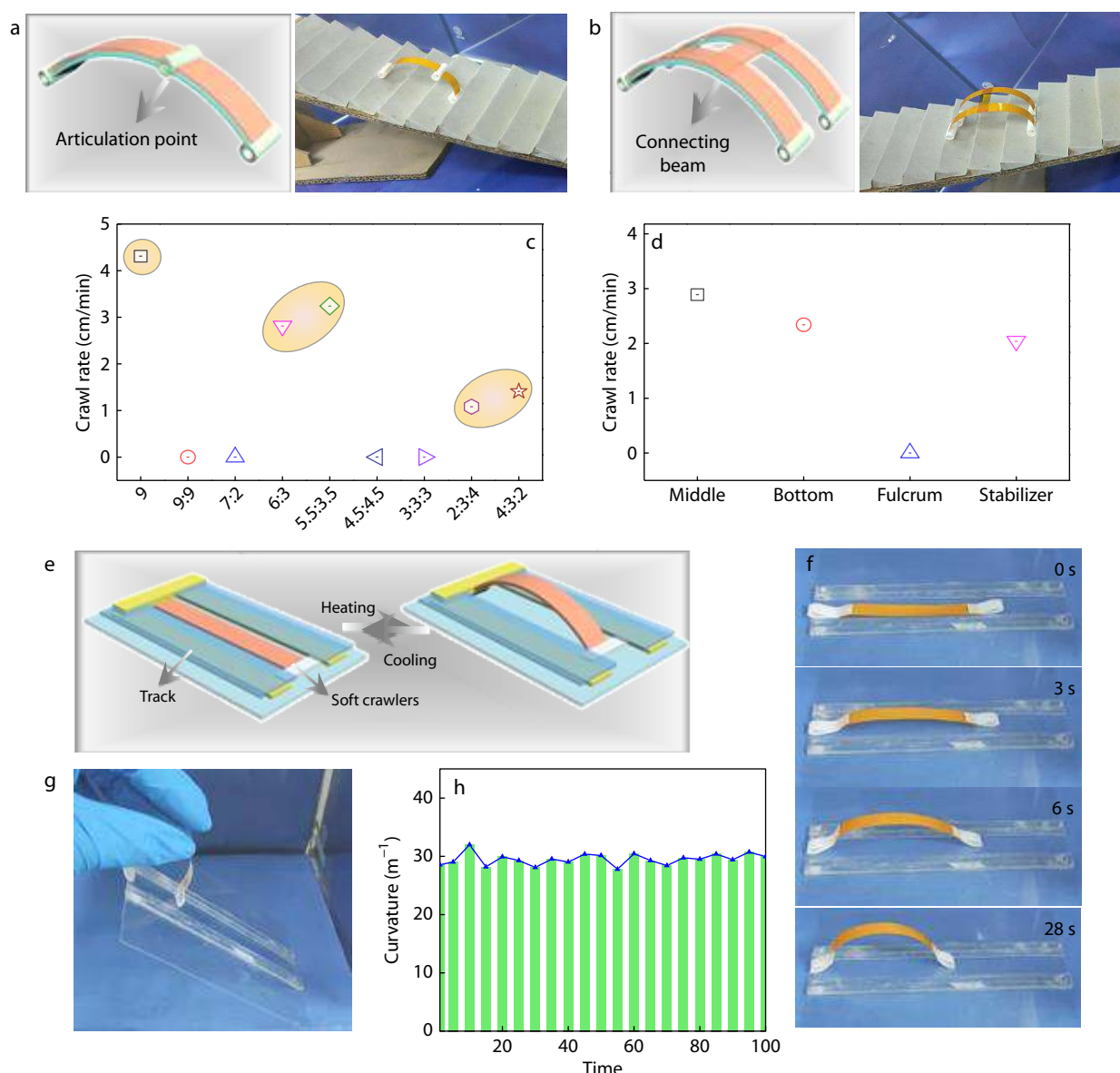


Fig. 4 Thermo-responsive complex soft actuators. (a) Image of series-wound crawlers with articulation points and b, shunt-wound crawlers with different connecting beams; (c, d) Climb speeds of soft crawlers with various articulation points and connection points; (e) Schematic illustration showing the thermo-responsive soft ring-pull; (f) Images of autonomous locomotion of soft actuators under heating; (g) photograph of thermo-responsive soft actuators which is convenient for lifting; (h) Cycle performance of thermo-responsive soft ring-pull.

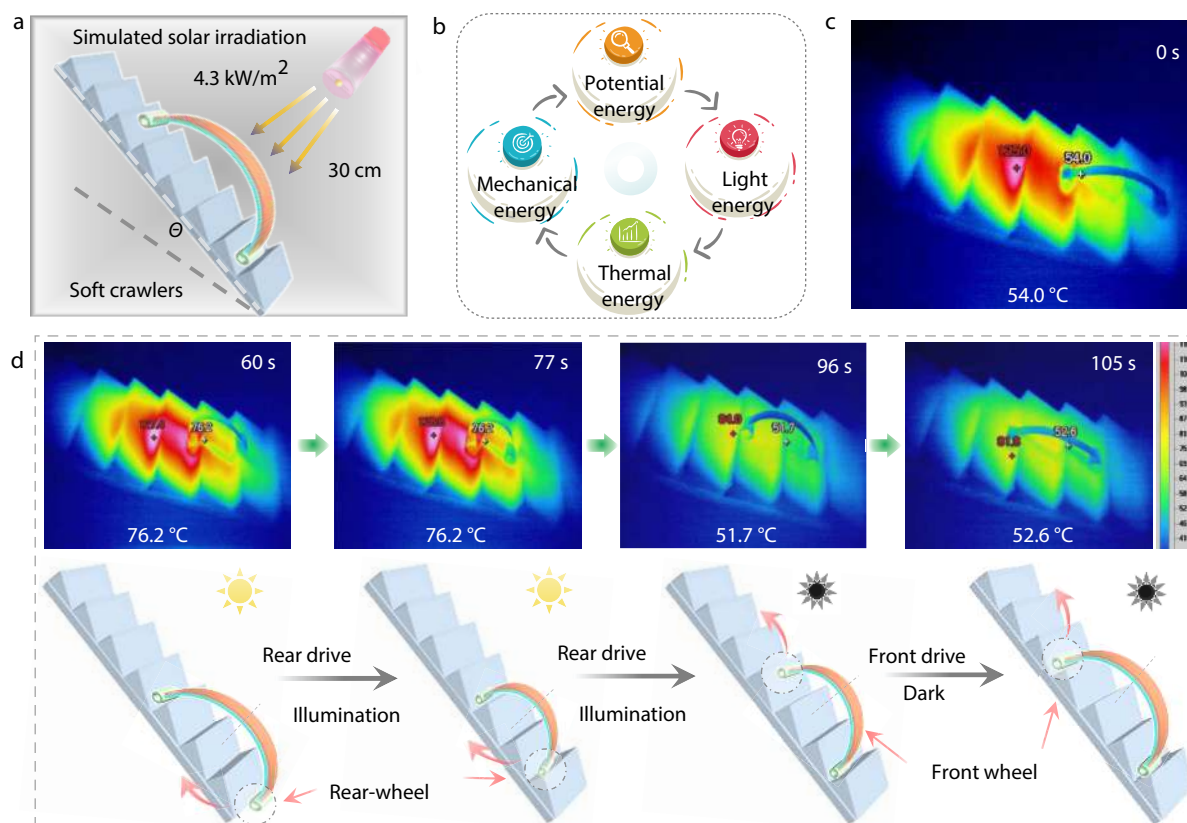


Fig. 5 Light-driven soft crawlers. (a) Schematic illustration demonstrating the simulated solar-driven climbing of soft crawlers; (b) Multiple energy conversion in soft crawlers during the climbing; (c) Initial photo of light-driven soft crawlers; (d) Images and schematic diagram of continuous locomotion driven by simulated solar, and corresponding climbing mechanism.

due to the existence of articulation points which could prolong the actuating time. In Fig. 4(c), a series of soft crawlers with different number and location of articulation points have been investigated. As a result, the fewer the number of points, the higher the speed. Moreover, some soft crawlers cannot be actuated with a improper point. Similarly, the shunt-wound crawlers formed by different connecting beams have also been prepared in Fig. 4(b). The connecting beams are designed to appear at middle, bottom and foot, which the highest climb speed reaches to 2.89 cm/min (Fig. 4d). Furthermore, the shunt-wound crawlers are able to share the foot. Although the speed is reduced, the articulation points and connecting beams are supposed to carry the cargo and provide a medium for other actuation behaviors and functions.

Furthermore, the intelligent soft ring-pull has been proposed as shown in Fig. 4(e). Poly(ethylene terephthalate) (PET) is used as the track, and PP is added at the end of PSO/PI film to form the soft ring-pull. When heating it, the soft ring-pull rapidly slides up on the track as shown in Fig. 4(f). The formed arc is used as a ring-pull for preventing overheating or something untouchable (Fig. 4g), even if we can apply it in frictional electricity, control connection, and so on. Moreover, the soft ring-pull presents admirable cycle performance, which can still achieve the curvature of about 30 m^{-1} after one hundred cycles (Fig. 4h). Combined with the above performance, the soft crawlers have greatly expanded their application potential.

Light-driven Soft Crawlers

Considering the driven mode, the thermal energy can be converted from luminous energy, like solar energy. Therefore, we further demonstrate simulated solar-driven climbing of soft crawlers as shown in Fig. 5(a) and Movie S4 (in ESI). The intensity of simulated solar is 4.3 kW/m^2 , which is 30 cm from soft crawlers. One should be noted that there is a multiple energy conversion, including mechanical, thermal, light and potential energy, existed during the actuating. Fig. 5(c) shows the initial state of light-driven soft crawlers. When the light illumination, the temperature rises rapidly to from $54\text{ }^{\circ}\text{C}$ to $76.2\text{ }^{\circ}\text{C}$ (Fig. 5d). During illumination, the soft crawlers also execute rear drive first. And then, the temperature quickly drops to $51.7\text{ }^{\circ}\text{C}$ to drive the front drive via covering the illumination. The results further demonstrate that light/temperature-driven soft crawler is an efficient form of wireless actuating for the practical application scenarios.

CONCLUSIONS

In conclusion, an autonomous soft crawler based on asymmetric bilayer film architecture, driven by the deformation mismatch from reversible “exhalation and inhalation” of moisture, has been proposed. The humidity-sensitive bilayer actuator exhibits periodic tumbler locomotion with repetitively transformed barycenter, which

has been explained via improved Timoshenko's equation. Inspired by the crawling creatures, the innovation of this work is that the soft crawlers are created with clear division of work, which can alternately proceed front drive and rear drive. Additionally, multi-joint complex actuators are constructed by series-wound or shunt-wound of soft crawlers to provide a medium for abundant actuation behaviors and functions. Moreover, a intelligent soft ring-pull for preventing overheating or something untouchable is fabricated, demonstrating its admirable cycle performance of actuating motion. In addition, soft crawlers can also be actuated by illumination of simulated solar, indicating their enormous potential in multi-scene driven mode and intelligent energy-harvesting system.

NOTES

The authors declare no competing financial interest.



Electronic Supplementary Information

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