Reprogrammable Untethered Actuator for Soft Bio-Inspired Robots

Runhuai Yang,* Miao Jin, Minmin Jin, Haisheng Qian, Qian Gao, Guoqing Jin, and Shiwu Zhang*

1. Introduction

In contrast to rigid systems, the compliance of soft robots allows them to safely and adaptively interact with living organisms and fragile objects.[1–6] Therefore, soft robots have significant potential for use in various complex situations, including biomedical applications, home service tasks, and harsh industrial environments.[7,8] Moreover, soft robots can readily exhibit muscle-like motion and better mimic the locomotion of soft biological systems as compared with conventional rigid-bodied robots.[9–14] In most cases, soft robots are driven using techniques based on pneumatics.[15,16] However, the structure of pneumatically actuated robots, including their power supply, pistons, and valves, is bulky, and this limits their functionality in situations where they have to be untethered.

With the aim of developing untethered soft robots, various active materials that respond to external and internal stimuli such as heat, magnetic and electric fields, light, and chemical reactions have been developed for use in soft robots and actuators.[17–26] However, the limitations of active materials restrict their applicability. For example, electroactive actuators require a rigid and bulky power source, whereas most hydrogel-based actuators can only be used in a liquid environment. Moreover, most previous studies on shape-memory polymers (SMPs) have focused on the heating of the materials through conduction, which is inconvenient from a practical viewpoint.[27]

Harvesting energy from light may allow for versatile untethered actuation strategies for soft robots.[28–32] When triggered by a photochemical reaction or photothermal heating,[33–35] the use of active materials for the actuators in soft robots allows for safe interactions and adaptable locomotion. However, stimulating the behavior of soft robots as well as reprogramming them and implementing reversible control in them remains a challenge. Herein, a method for fabricating soft, bio-inspired, untethered, infrared-driven actuators that exhibit reversible and reprogrammable behaviors is proposed. By exploiting the mismatch in the infrared-driven thermomechanical responses of the two active materials constituting the bilayered soft actuator, reversible actuation can be realized. More importantly, the shape of robots based on this actuator can be reprogrammed such that they exhibit bio-inspired locomotion specific to the environment. For instance, a soft robot based on the actuator can be made to exhibit several bio-inspired functions, such as crawling, rolling, and object manipulation, in a complex environment. Furthermore, reversible and reprogrammable actuation can be achieved in untethered soft bio-inspired robots using the bilayer of infrared-actuated thermoresponsive materials, suggesting that other materials with similar properties can also be adopted in a similar method. The proposed method allows for the integration of different active materials toward the realization of infrared-driven, untethered reversible, and reprogrammable actuators for soft bio-inspired robots.
light-active materials undergo shape deformation to produce untethered bio-inspired actuation or movement.\textsuperscript{36–40} Most light-active materials are driven by ultraviolet/visible lasers, which are both harmful and inconvenient to use. In contrast, infrared light has the advantages of high penetration and not causing damage to living tissue. Thus, a biocompatible infrared-active material that can be driven by an ordinary infrared lamp would be ideal for actuating untethered soft robots for use in the home or biomedical environment. It is reported that the combination of polyactic acid (PLA) and carbon nanotubes (CNTs) was able to be used as an infrared-response material.\textsuperscript{41} Based on the recent light-active materials, it is promising to design actuators with untethered and harmless driving method.

Reversibility and reprogrammability are also highly desirable features for the active materials to be used in soft robots.\textsuperscript{42–44} Although SMPs can be programmed, conventional SMPs cannot be used as soft actuators because they undergo one-way, irreversible transformations. For example, most heat-shrinkable materials contract at a certain temperature but do not revert to their original expanded state when cooled.\textsuperscript{45} Similar limitations exist with respect to the actuation of shape-memory alloys as well. This type of irreversible behavior is not conducive for achieving reversible motion in actuators. Moreover, most active materials only deform along a single trajectory, which is preprogrammed during the fabrication process.\textsuperscript{46} After fabrication, the orientation of the polymer chain segments and the macroscopic shape-shifting geometry of these materials remain unchanged.\textsuperscript{47} However, if the “preprogrammability” of these materials were to be improved to “reprogrammability,” it would become possible to repeatedly reconfigure the shape of the actuators after fabrication. This would allow for modulation of the locomotion and functionality of soft robots based on the environment.\textsuperscript{48}

Although several active materials have been developed for use in soft actuators, only a few exhibit characteristics that make them suitable for use in bio-inspired, untethered, infrared-driven, reversible, and reprogrammable soft actuators.\textsuperscript{45,49} In this article, we propose a method for the fabrication of an infrared-driven, untethered, reversible, and reprogrammable actuator for soft bio-inspired robots. A polymer bilayer structure comprising an infrared response layer (IR-layer) and a transparent elastic layer (T-layer) was fabricated. The bilayer was designed such that the glass transition and melting temperatures of its two layers were mismatched. The mismatch in the glass transition temperatures (\(T_g\)) of the two layers meant that there was a temperature range over which reversible actuation can be realized, whereas the difference in their melting points (\(T_m\)) meant that the same was true for reprogrammability. The actuator does not require a high-energy intensity laser and can be readily powered and controlled using a low-cost commercially available medical infrared lamp. Moreover, the initial shape and motion of the actuator can be reprogrammed with ease. A soft robot based on the actuator exhibited multiple functionalities, such as transitions in its movement mode from crawling to rolling, as well as the ability to grab and manipulate objects under various situations.

2. Results and Discussion

To allow for reprogrammability, the layers of the bilayered actuator must be composed of materials with significant differences in their melting points. At the same time, ensuring that their glass transition temperatures are also significantly different is essential for guaranteeing infrared actuation. Guided by these constraints, PLA and CNTs (used as the photothermal component) were used to fabricate the IR-layer (glass transition temperature of IR-layer, \(T_g\text{IR} = 50^\circ\text{C}\) and melting temperature of IR-layer, \(T_m\text{IR} = 115^\circ\text{C}\)), whereas a film of polyethylene terephthalate (PET) was used as the T-layer (glass transition temperature of T-layer, \(T_g\text{T} = 81^\circ\text{C}\) and melting temperature, \(T_m\text{T} = 255^\circ\text{C}\)). The fabrication, infrared-driven actuation, and reprogramming processes are shown in Figure 1A. The IR-layer was fabricated by the melting blend method using a PLA powder and CNTs. The temperature range for the infrared-driven actuation was \(T_g\text{IR} \rightarrow T_g\text{T}\), whereas the temperature range for the fabrication and reprogramming processes was \(T_m\text{IR} \rightarrow T_m\text{T}\). The premelting powder was prepared by mixing the PLA powder and CNTs for 30 min in a vortex mixer. Next, the IR-layer was fabricated by melting the PLA/CNT mixture on the plasma-activated T-layer, and the temperature of the bilayer structure was decreased, ensuring that the IR-layer became fixed on the T-layer. A typical SEM image of the cross section showing the IR-layer and T-layer is shown in Figure S1, Supporting Information. When the temperature of the IR-layer was reduced to less than \(T_g\text{IR}\), the layer shrank, and the bilayer structure became bent because of the increase in the modulus of the IR-layer.

The infrared-driven actuation process is shown in Figure 1A2. Before the irradiation of the bilayer structure, the temperatures of the IR-layer and PET layer are lower than \(T_g\text{IR}\) and \(T_g\text{T}\), respectively. When the infrared lamp is turned on, the infrared-driven thermal component of the IR-layer absorbs energy from the infrared light, and the temperature of the layer increases quickly. During this increase in the temperature, there exists a range of temperatures higher than \(T_g\text{IR}\) and lower than \(T_g\text{T}\) within which the modulus of the IR-layer decreases significantly, and the bilayer structure becomes flat due to the residual elasticity of the T-layer and the mismatch in thermal expansion. Subsequently, when the infrared lamp is turned off, the bilayer structure cools to room temperature (RT). When the temperatures of the two layers become lower than their respective \(T_g\) temperatures, the bilayer structure bends because of the glass transition of the IR-layer. A schematic of the reprogramming process is shown in Figure 1A3. To change the initial shape of the bilayer structure, it must be heated to the temperatures between \(T_m\text{IR}\) and \(T_m\text{T}\). In this case, the shape of the bilayer structure is manually determined by an external force, which is applied when the IR-layer is in the molten state and the T-layer is flexible. Subsequently, the bilayer structure is allowed to cool to RT, and the external force is removed. In this manner, the shape of the bilayer structure can be set, and reprogramming can be performed. Photographs of the actuator during the fabrication, infrared-driven actuation, and reprogramming processes are shown in Figure 1B.

The differential scanning calorimetry (DSC) curves of the two layers are shown in Figure 1C. The critical temperatures

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and temperature range for reversible actuation and reprogramming are marked in the curves. In other words, the temperature ranges for reversible actuation and fabrication/programming are indicated. \( T_{g-IR} \) and \( T_{m-IR} \) were obtained from the curve for the PLA/CNT layer, whereas \( T_{g-T} \) and \( T_{m-T} \) were obtained from the curve for the PET film. The temperatures can be arranged in the following order: \( T_{g-IR} < T_{g-T} < T_{m-IR} < T_{m-T} \). As mentioned previously, \( T_{g-IR} \) should be lower than \( T_{g-T} \), so that the temperature range of \( T_{g-IR} \) to \( T_{g-T} \) can be used for reversible actuation by infrared irradiation. The condition \( T_{g-T} < T_{m-IR} < T_{m-T} \) is also critical with respect to the fabrication, actuation, and reprogramming processes, in that the temperature of the bilayer structure should be 1) higher than \( T_{g-T} \) to soften the T-layer; 2) higher than \( T_{m-IR} \) to melt the IR-layer and change its shape; and 3) lower than \( T_{m-T} \) to prevent the base layer, that is, the T-layer, from melting and breaking. Therefore, the bilayer structure can be controlled by ensuring that its infrared-induced temperature lies between \( T_{g-IR} \) and \( T_{g-T} \). Furthermore, the structure can be reshaped and reprogrammed by ensuring that its temperature lies between \( T_{m-IR} \) and \( T_{m-T} \).

Figure 2 shows the mechanical behavior and the critical factors of the bilayer actuator. An infrared camera was used to capture images during the actuation process, and therefore, record its temperature distribution. After a few seconds of uniform irradiation of the actuator and its surroundings with the infrared lamp, the temperature of the actuator reached 57 °C (the initial temperature of the actuator before irradiation is 21 °C, same as RT), as shown in Figure 2A, whereas that of the irradiated surroundings remained at 21 °C. In the absence of a photothermal component, the surrounding environment was not significantly affected when irradiated with the infrared lamp. The mechanical characteristics of the bilayer structure were evaluated using the three-point bending test (Figure S2, Supporting Information), and the force–distance curves were recorded both with the infrared lamp turned on and off, as shown in Figure S3, Supporting Information. The modulus of the
bilayer structure was calculated from the force–distance curves and analyzed statistically, as shown in Figure 2B. As the temperature increases, the IR-layer undergoes a glass–rubber transition, resulting in a decrease in the modulus of the bilayer structure by 36%\(^{\circ}/C_6\), as shown in Figure S4, Supporting Information. In contrast, the modulus of a standalone IR-layer decreases by as much as 51%\(^{\circ}/C_6\), as shown in Figure S4, Supporting Information. The change in the modulus of the bilayer structure was smaller than that in the modulus of the standalone IR-layer because the temperature of the former was lower than \(T_{g-T}\) and the T-layer remained in the glassy state.

When \(T < T_{g-IR}\), a moment \(M_{IR-layer}\) is balanced by an equal and opposite moment \(M_{T-layer}\) at the ends of the bilayer structure, as shown in Figure S6, Supporting Information. As \(M_{IR-layer}\) is proportional to the Young’s modulus of IR-layer,\(^{[50]}\) \(M_{IR-layer}\) is smaller than \(M_{T-layer}\) after glass transition of IR-layer and the actuator is inverted by \(M_{T-layer}\) when \(T_{g-T} > T_{g-IR}\). As a result, the T-layer had a straightening effect on the bent structure during the irradiation of the actuator with the infrared lamp.

The difference of the thermal expansion of the bilayer between \(T_{g-IR}\) and \(T_{g-T}\) may also contribute to the actuated transformation speed. The thermomechanical analysis (TMA) curve of IR-layer and T-layer is shown in Figure S5, Supporting Information. According to the TMA results, the coefficient value ranges for the IR-layer is 1.2\(^{\circ}/C_2\) to 8.0\(^{\circ}/C_4\) K\(^{-1}\), and for the T-layer is 0.1\(^{\circ}/C_2\) to 1.0\(^{\circ}/C_4\) K\(^{-1}\). It was observed that the coefficient of thermal expansion of the IR-layer increased quickly, especially after glass transition temperature, and the difference of the thermal expansion of the IR-layer and the T-layer became larger when \(T > T_{g-IR}\). According to the model of thermal deformation of bilayer structure,\(^{[51]}\) the deformation induced by thermal can be calculated as

\[
\frac{1}{\rho} = \frac{6(T_{flatten} - T)(\alpha_{IR-layer} - \alpha_{T-layer})(1 + \delta^2)}{h(3(1 + \delta)^2 + (1 + \delta n)(\delta^2 + \frac{1}{n^2}))}
\]

where \(1/\rho\) denotes the curvature of actuator, \(h\) denotes the thickness of the actuator, and \(n\) denotes the ratio of elastic modulus of the IR-layer to the elastic modulus of the T-layer. \(\alpha_{IR-layer}\) and \(\alpha_{T-layer}\) denote the coefficient of thermal expansion of the IR-layer and the T-layer, respectively. \(T\) denotes temperature, and \(T_{flatten}\) denotes the temperature at which the actuator is only flattened by thermal expansion \((T_{flatten}\) of the actuator can be considered as constant but cannot be measured, because in our experiment, the temperature of the actuator was higher than \(T_{m-IR}\) when the actuator was flattened, whereas Equation (1) can only be used when the temperature is lower than melting point.\(^{[2]}\))

Based on this mechanism, the bilayer actuator is driven by the change of temperature, and the curvature increases as \(T\) increases \((T < T_{flatten})\). According to Equation (1), although \((\alpha_{IR-layer} - \alpha_{T-layer})\) becomes larger after \(T > T_{g-IR}\), the curvature decreases rapidly as \(T\) increases.

In this article, we defined the chord length of the actuator as the distance between its two ends, and defined the chord ratio of the actuator as the chord length against the actuator length.
quickly balanced by the Equation (1), the increase of actuator exhibited a high degree of deformation. According to In contrast, when the two layers had similar thicknesses, the extent of deformation of the actuator was not signi

whereas a δ value of 0.1 means that the thickness of T-layer was 0.12 mm and there was no IR-layer for the sample. There was no obvious extent of deformation of the sample when single T-layer or IR-layer (δ = 0:1 and δ = 1:0) was illuminated. In addition, the extent of deformation of the actuator was not significant when one of the layers was much thicker than the other one. In contrast, when the two layers had similar thicknesses, the actuator exhibited a high degree of deformation. According to Equation (1), the increase of δ and h reduces the change of curvature caused by the increase of temperature. Another probable cause is the moment value is significantly influenced by the thickness, and the inversion by T-layer after glass transition is quickly balanced by the MIR-layer. Therefore, although the actuator with 3:1 was easier to be reprogrammed in our experiment because of the IR-layer was thicker than T-layer, it was not recommended to use it because of the limited extent of deformation.

The actuator with δ of 1:3 also had a limited extent of deformation. Because the thickness of the T-layer was much thicker than the IR-layer, the actuator with a δ of 1:3 was hard to reprogram and shrank to a shape with low curvature. Therefore, the optimal ratio between the thickness of IR-layer and T-layer was 1:1, whereas the change of chord ratio of the actuator with other thickness ratio was much smaller. Although the actuator with a thickness ratio >1:1 can be easily programmed, it was not recommended that to use them since the actuation ability was limited. The temperature response of the actuator (δ = 1:1) to prescribed values of heating conditions is shown in Figure S7, Supporting Information.

The transformation speed of the actuator was influenced by the illumination of different infrared power intensities, as shown in Figure 2D. The transformation speed can be defined as the rate of change in the chord ratio of the actuator. The infrared power intensity illuminated on the actuator was controlled by change the distance, and the relationship between the distance and the infrared power intensity is shown in Figure S8, Supporting Information. It was observed that the transformation speed increased with an increase in the infrared power intensity, especially when the infrared power intensity >0.1 W cm⁻². We also tested the actuator illuminated from either side, and it was observed that there was no difference while the actuator was driven from both sides, as shown in Figure S9, Supporting Information. The performance of actuator was tested for 100 cycles, and the result shows that there was no fatigue after 100 actuation cycle, as shown in Figure S10, Supporting Information.

Based on the observed performance of the actuator, a bio-inspired crawling robot was designed and fabricated, as shown in Figure 3. The δ value of the actuator used was 1:1, whereas its length was 20 mm; these values were used to ensure that the

**Figure 3.** Infrared-driven bio-inspired soft robot climbing zigzag-textured sloped structure with crawling gait. A) Snapshots of climbing process. T = 0 s: robot is at starting position; T = 2 s: robot is irradiated with infrared lamp and its body expands; T = 8 s: robot moves to next step, and its body shrinks; and T = 45 s: after being irradiated a few times, robot successfully climbs slope. B) Infrared images of soft robot with infrared lamp turned off and on. C) Changes in head-to-tail distance of robot with infrared lamp turned on and off. D) Heights of head (purple circles) and tail (yellow circles) of robot on slope during climbing process (see Movie S1, Supporting Information).
actuator exhibited the highest rate of the change with respect to its chord ratio. As shown in Figure 3A, the robot can climb a sloped structure with a zigzag texture; the sloped structure was fabricated using PLA without any photothermal component. A conventional medical infrared lamp used widely for physical therapy was used as the infrared source (power of 150 W and an infrared power intensity of 0.2 W cm$^{-2}$). Compared with ultraviolet, infrared provides a safer and biocompatible power source, and the power we used is lower than the permissible exposure intensity (under continuous exposure to 980 nm light, the maximum permissible exposure of skin is 0.726 W cm$^{-2}$[52,53]). Therefore, this infrared lamp is much safer and is also available readily. Thus, the fabricated robot can be controlled with ease using a low-cost light source with an ultralow intensity. It can be seen from the figure that the robot became flat and climbed to the next step when irradiated by the infrared lamp (Figure 3A, $T = 2$ s). Furthermore, it shrank when the infrared lamp was turned off (Figure 3A, $T = 8$ s). In this manner, the robot can be made to steadily climb the steps on the sloped structure by repeatedly turning the infrared lamp on and off. The infrared camera recorded the temperature changes during the irradiation process, and it was observed that the photothermal effect increased the temperature of the robot rapidly (Figure 3B). However, the temperature of the sloped structure did not change significantly, confirming that, in the absence of a photothermal component, the PLA structure was not significantly influenced by the low-power infrared lamp. Therefore, the robot can be driven without causing obvious disturbances to the surroundings. Figure 3C shows the changes in the head-to-tail distance of the robot with the switching of the infrared lamp. It can be seen that the robot can be readily actuated in a reversible manner. Figure 3D shows the heights of the head and tail of the robot on the slope. These data further confirmed that the robot can continuously climb the slope with reversible actuation.

The fabricated actuator can be reprogrammed into geometries with different curvatures. Furthermore, the reprogramming process was repeatable even after the actuator had been subjected to several infrared driving cycles. As shown in Figure 4A.1, the initial geometry of the actuator was arc-like, and the actuator exhibited a large radius of curvature, making it suitable for use in a soft robot with a crawling gait. After several actuation cycles, reprogramming was performed using an external force and a high heating temperature ($>T_m+1\alpha$). After the robot had been allowed to cool, it can be reshaped to exhibit the desired local curvature, as shown in Figure 4A.2. Furthermore, it can also be reversibly driven by the infrared lamp. Moreover, the robot can be reprogrammed once more to resemble a circle, as shown in Figure 4A.3 and used as a soft robot with a rolling gait. The actuator was also reprogrammed in various directions, as shown in Figure S11, Supporting Information. The results show that the reprogramming and actuation was isotropic. Benefiting from the reprogrammability of our actuator, an initial mechanical steady-state was programmed, and the actuator bended around the programmed axis as an anisotropic deformation, driven by an isotropic shrinking.

With reprogrammability, the actuator can be made to exhibit different geometries and curvatures and thus can be used as a multifunctional caterpillar-like robot. To describe the shape of the caterpillar-like robot, we measured the radii of curvature of its head and body; these are denoted as $R_{\text{head}}$ and $R_{\text{body}}$, respectively, as shown in Figure 4b. Robots with different radii of curvature were fabricated and evaluated to explore how the shape of the robot affects its mobility. First, the effects of $R_{\text{head}}$ and $R_{\text{body}}$ on the crawling speed were studied. Four different robots were evaluated, as shown in Figure 4B. For Cases 1 and 2, $R_{\text{body}}$ was 12 mm, whereas $R_{\text{head}}$ was 6 and 2 mm, respectively. Furthermore, for Cases 3 and 4, $R_{\text{body}}$ was 6 mm, whereas $R_{\text{head}}$ was 6 and 2 mm, respectively. The results showed the moving speeds in Cases 1 and 2 were higher than those in Cases 3 and 4, indicating that although the shape of the robot body significantly affected the climbing speed, that of its head, namely, $R_{\text{head}}, did not.

In contrast, although $R_{\text{head}}$ had a marginal effect on the climbing speed, it played an important role in some situations. In nature, some animals can transform to exhibit a wheel-like shape to be able to roll quickly. This ability enables them to quickly escape from dangerous situations.[54] Inspired by this ability, a soft robot was designed such that its initial shape can be changed and its movement mode switched from “crawling” to “rolling” to perform different tasks. Thus, a major advantage of the proposed robot is its reprogrammability in that it can be reshaped and made to switch from a crawling gait to a rolling gait, like a caterpillar. Furthermore, instead of moving only through infrared actuation, the robot can be transformed and made to move under the effect of gravity to ensure a significantly higher speed at a low cost. The transition from “crawling” to “rolling” is shown in Figure 4D. A robot was reprogrammed to exhibit a circular shape. The robot was then made to climb to the top of a sloped structure under infrared-driven “crawling” motion (Figure 4C). Next, the infrared lamp was turned off for a few seconds, during which period the robot reconfigured its body to form a wheel, after which it rolled down the slope under gravity. The rolling ability of robot specimens with different $R_{\text{head}}$ and $R_{\text{body}}$ values was also studied, as shown in Table S1, Supporting Information. The results showed that both $R_{\text{head}}$ and $R_{\text{body}}$ significantly affected the transition from “crawling” to “rolling.” The “rolling” mode cannot be achieved when $R_{\text{body}}$ was too large compared to the length of the robot. For example, when the length of the robot was 20 mm, it cannot be made to transit from “crawling” to “rolling” when $R_{\text{body}}$ was 13 mm. However, when the value of $R_{\text{body}}$ was appropriate (6 mm), $R_{\text{head}}$ had a determining effect on the transition to the “rolling” mode. In contrast, when $R_{\text{head}}$ was too large (6 mm), the robot still cannot make transition to the “rolling” mode. As $R_{\text{head}}$ was decreased, the success rate of the transitioning process increased. When $R_{\text{head}}$ was changed to be 10% of the length of the robot (i.e., 2 mm), the success rate of the transitioning process became 100%. This confirmed that the proposed soft robot can be readily made to exhibit multiple movement modes through reprogramming. As mentioned previously, $R_{\text{head}}$ does not significantly affect the climbing speed. Therefore, $R_{\text{head}}$ can be regulated to switch the movement mode of the robot.

For certain $R_{\text{head}}$ values, the robot can be made to function as a gripper for small objects in a narrow environment, such as a ditch, as shown in Figure 4D. The manipulation and gripping of objects in a narrow and complex space is a difficult task. To be able to grip a small object in a narrow environment, the structure and movement of the robot should be designed with precision. As shown in Figure 4D, the soft robot can grab...
a small object in a corner. It was found that $R_{\text{head}}$ significantly influenced the grabbing success rate. For instance, by reprogramming the shape of the robot such that $R_{\text{head}}$ was 2 mm, the robot can be made to grip small objects in a narrow ditch. However, when the $R_{\text{head}}$ value was not appropriate (4 mm), the robot failed at grabbing objects. Therefore, by reprogramming the curvature of the head, the behavior and functionality of the robot can be controlled with precision.

We have demonstrated a design and fabrication method for integrating soft active materials with different infrared-driven thermomechanical responses into a reversibly actuating and reprogrammable actuator for untethered soft bio-inspired robots. We integrated polymers with mismatched glass transition and melting temperatures ($T_{g-\text{IR}} < T_{g-T} < T_{m-\text{IR}} < T_{m-T}$), such that the fabricated robot can be reversibly actuated at temperatures between $T_{g-\text{IR}}$ and $T_{g-T}$ and reprogrammed at temperatures between $T_{m-\text{IR}}$ and $T_{m-T}$.

The infrared-driven reversible actuation of the soft robot allows for the realization of untethered movement and object manipulation. Compared to conventional heating platform which provides prescribed temperature, IR source provides a convenient and untethered driven method. The power of IR
source used in our experiment was 150 W, whereas the power of our heating platform was 800 W. Compared to the existing light-driven untethered polymer-based actuators, the performance advantages of the proposed actuator are low power and rapid. For example, compared to a typical SMP-based light-driven actuator-based crawling robot, the crawling speed of the proposed actuator-based soft robot was three times faster.\(^\text{[27]}\)

Although most of the light thermal or light reaction materials required an ~1 W cm\(^{-2}\) illuminating power,\(^\text{[55,56]}\) the proposed actuator was driven by a 0.2 W cm\(^{-2}\) illuminating power. Moreover, the reprogrammability makes it adaptable to various tasks and environments. In other words, the geometry of the robot can be reprogrammed depending on the requirements. The soft robot is not only capable of exhibiting animal-like movement but also shows the ability to transform its shape like them.\(^\text{[43]}\) In addition, the reprogrammable actuator can also be used as an untethered manipulator, as shown in Figure S12. Supporting Information. By reprogramming the curvature of the actuator, the track of its end effector can be redesigned and regulated. Thus, the actuator can not only serve as a platform for fabricating bio-inspired soft robotics but also for reprogrammable soft manipulators. The load capacity of the actuator was also measured by a force meter. For a typical actuator (weight = 0.0455 g), the maximum force is 0.005 N, as shown in Figure. S14, Supporting Information. Therefore, the actuator can be used to push or carry loads which is ten times heavier than the actuator.

Another benefit of the proposed method is that the materials used are readily configurable, low cost, and can be 3D printed with ease. PLA is used widely as a biomaterial and biodegrades into CO\(_2\) and H\(_2\)O.\(^\text{[57]}\) The materials used can also be 3D printed into complex structures for specific tasks. This further extends the designability of the proposed soft robot. In addition, this work provides a strategy for designing soft robots from different biomaterials in that the choice of materials that can be used is not limited to PLA, CNTs, and PET. All soft biomaterials that exhibit thermal-based shape-change characteristics, the photothermal effect, and a mismatch in their glass transition and melting temperatures can be used to fabricate the bilayer structure. This includes polycaprolactone, hydrogels, polyurethane, and Fe\(_3\)O\(_4\) nanoparticles. Since the decay in modulus of IR-layer provided the straightening effect of the actuator, a series of SMPs with this property can also be adopted in this structure.

In addition, since the difference of the thermal expansion of bilayer can induce bending,\(^\text{[58]}\) and the coefficient of thermal expansion was significantly increased by the glass transition of PLA\(^\text{[59]}\), the difference of the thermal expansion of bilayer between \(T_{g-1R}\) and \(T_{g-T}\) also contributed to the actuated transformation speed. Therefore, it is possible to further improve the actuation ability by enlarging the difference of the thermal expansion and the difference of modulus between IR-layer and T-layer in the temperature range for the infrared-driven actuation. Moreover, compared to the heating process, our results show that the cooling process spent more time. Therefore, the acceleration of convection with the surroundings or by using the bionic autonomic perspiration\(^\text{[40]}\) can further improve the actuation speed for the actuator. This article also verified the influence of the infrared power intensity to the transformation speed. Other infrared sources (e.g., daylight and infrared irradiation source) which have sufficient power intensity may also be used to drive the actuator. The motion of actuator was performed on a zigzag-textured slope because the motion was intrinsically symmetric. By equipping bionic asymmetric bristles or microneedle structures,\(^\text{[61]}\) the actuator-based soft robots can be adaptable to more environment and situations. Therefore, this study should aid the design and fabrication of personalized biomaterial-based smart soft robots that are tailored for the given environment.

3. Conclusion

In summary, in this article, we present a novel method for designing and fabricating reprogrammable untethered actuators for soft bio-inspired robots. We evaluated the effects of various critical parameters on the performance of the actuator. The mismatch in the infrared-driven thermomechanical responses of the two constituent layers of the bilayer structure allowed for reversible actuation and reprogrammability. Furthermore, we were able to make the actuator exhibit “crawling” and transformation behavior, which confirmed its potential for use in multifunctional bio-inspired soft robots. The integration of reprogrammability and reversibility together with untethered infrared-driven low-powered actuation represents a significant advance in the development of the next generation of soft robots.

4. Experimental Section

Fabrication of Bilayer Actuator: The PLA (4032D, NatureWorks, USA), multiwalled CNTs (MWCNTs) (Guosen technology, China. Purity > 60 wt%; inner diameter = 0.8–1.8 nm; and outer diameter = 1–2 nm), and the PET films (Mingheng Ltd, Hefei, China) used were obtained commercially. To improve the adhesion between the T-layer and the IR-layer, the T-layer was roughened with sandpaper (grit size: 1000), and its surface was activated using a plasma cleaner (Harrick PDC-32 G, USA) for 1 min. The PLA powder was sieved using a 300 mesh screen and blended with the MWCNTS (1 wt% in PLA) for 30 min in a vortex mixer. The mixture process is shown in Figure S15, Supporting Information. After PLA/CNTs mixture was obtained and the T-layer was activated, the PLA/CNTs mixture was spread on the T-layer and oscillated at 50 Hz on a vibration platform. The thickness of IR-layer was controlled by changing the spread time and oscillation time. The fabrication of bilayer actuator was then performed on a heating platform at 120 °C for 10 s and was moved away for cooling in RT.

Measurement of Glass Transition and Melting Temperatures: \(T_{g-1R}\), \(T_{g-T}\), \(T_{m-IR}\), and \(T_{m-T}\) were measured using a DSC system (NETZSCH STA 2500, Germany). The DSC curves for the IR-layer and T-layer were recorded separately. During the DSC process, the test layer was heated from 25 to 270 °C, with the rate of increase in the temperature being 5.0 °C min\(^{-1}\). All the tests were performed in a nitrogen environment. The thermal expansion of IR-layer and T-layer was measured by DMA (Q400, TA instruments).

Measurement of Modulus: The modulus of the bilayer structure was measured by the three-point bending test, which was performed with a universal material testing machine (Reger 6010, Shenzhen, China). Assuming that the cross section of the bilayer structure was rectangular, the flexural modulus was calculated as follows:

\[
E_b = \frac{L^3}{48I} \left(\frac{\Delta F}{\Delta a}\right)
\] (3)
where \( h \) is the thickness, \( b \) is the width, and \( L \) is the length of the bilayer between two supports, as shown in Figure 2. Further, \( I \) is the moment of inertia of the cross-sectional area about the neutral axis, \( \Delta F \) is the load at a given point on the load deflection curve, \( \Delta d \) is the maximum deflection of the center of the material, and \( E_s \) is the flexural modulus.

Analysis of Infrared-Driven Reversible Actuation and Motion: The infrared source was a commercial medical infrared lamp (150 W, Philips, USA). The motion, position, and distance of the soft robots were recorded with a digital camera (EMS II, Olympus, Japan). By calculating the pixel coordinate of the robots and calibrating the image coordinate to real coordinate, the positions and distances were obtained. The thermal images of the robot were recorded with a thermal imaging system (SEEK Thermal, USA).

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.

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