Untethered Soft Actuators by Liquid–Vapor Phase Transition: Remote and Programmable Actuation

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Bioinspiredsoftroboticshaveuniqueadvantagesinsuperioradaptabilityandcomplexmotionsforfieldexplorationandinteractionwithhumans. Themobilityandoutputforce, however, are still the critical challenges for many promising applications. It is attractive to develop “untethered” robotics to improve the mobility by getting rid of the external electrical or pneumatic tethers while achieving massive output stroke and force. Inspired by the creatures’ movements induced by the multiplication of cells and asymmetric volume changes of the tissue, an untethered soft actuator composed of self-contained liquids and superelastic chambers is proposed, and by remote stimuli (e.g., near-infrared light), the encapsulated liquids transit from liquid to vapor, giving rise to volume change in the corresponding chambers. Programmable motions, i.e., photophobia of artificial sunflower, can be realized, indicating a massive and linear driving stroke (up to 160% in elongation, 0.5 mm °C⁻¹) and output force (14.5 N with 6 g self-weight, 0.33 N °C⁻¹). The untethered soft actuator suggests a feasible approach to develop smart, soft, and autonomous robotics and holds promise in fields ranging from surgery to rehabilitation and rescue.

1. Introduction

Nature offers a lot of meaningful samples for the development of soft robotics. The remarkable flexibility and dexterity of frogs tongues, elephants trunks, and other biological muscular systems always inspire the new design and driving strategies for soft actuators and robotics.[1] Unlike traditional robotics, which are made of rigid materials and components to fulfill the requirements of high precision, rapid movement, and high force, soft robotics are primarily made of soft materials for the purposes of safety, adaptability, and complex motions, which can be very difficult and hardly achievable for traditional counterparts.[2–4] To date, a variety of biologically inspired solutions for soft actuators and robotics have been proposed. Among these representatives, electroactive polymers,[5–10] shape memory alloys,[11–14] shape memory polymers,[15–20] and multilayer composites are paid more attention.[21–28] The balance between output force and considerable strain, however, is still one of the critical challenges for soft actuators: low strain for shape memory alloys (<10%) and electroactive polymers (<10%), and inadequate load capacity (<1 N) of multilayer composites limited their range of applications and potential for further developments.[5, 29–32]

In nature, volume changes, induced by localized multiplication of cells or external stimuli of the tissues, are widely observed in the movements of living bodies, such as the phototropism of plants, the opening and closing of pinecones, and the leaf motion of flytrap.[33–35] Inspired by these phenomena, the soft pneumatic robotics have been developed with advantages of significant driving force, high robustness, and exceptional mechanical compliance.[3, 4, 36–43] Although the use of pneumatics in soft robotics grows, the most commonly utilized energy source remains to be a tether or a stationary compressor.[44] The necessity of external compressors and pressure-regulating components for hydraulic or pneumatic fluid elastomer actuators limits the size of robots and their applications in highly integrated and mobile systems. In addition, in recent studies, significant trade-offs are revealed to implement a self-contained, untethered, and programmable soft actuator: entirely soft systems often require a tethered connection to support pneumatic or electrical hardware, whereas untethered systems typically depend on bulky on-board components such as batteries, microprocessors, pumps, or motors.[45] Researchers have made some progress in freeing the soft robotics from the “umbilical cord” and integrating the control units, for example, using the gas generated from fuel decomposition or explosion to achieve the action.[46, 47] Due to the limited size of the energy source, logic control unit, and the constant consumption, the endurance and driving ability

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are relatively weak, which indicates that they cannot support long-term and complex movements. It is still challenging but promising to develop an untethered soft actuator with large deformation, large driving force, and programmable complex motions.

To explore novel methodologies in self-contained and untethered soft actuators, a type of soft robotics has been recently designed based on liquid–vapor phase transition mechanism, which follows the fundamental thermophysics and provides a vast volume expansion.\cite{48-50} As trapped inside the elastomer matrix, microbubble of ethanol demonstrated heat-triggered liquid–vapor phase change, which gave rise to volume expansion and contraction in the soft actuator itself.\cite{51,52} The fabrication and further applications for programming motions, however, are limited to the accurate adjustments of ethanol content and the bubble size. In addition, the requirement of heating wire and external power supply eventually hindered the actuation capabilities and application prospects.

As shown in Figure 1a, inspired by the stem bending of sunflowers due to the volume changes induced by the asymmetric multiplication of cells,\cite{33} we proposed a self-contained, untethered, and programmable soft actuator based on liquid–vapor phase transition, which can realize heat-triggered elongation, bending, twisting, and programmable complex motions, as the scheme shown in Movie S1, Supporting Information. The soft actuator is a capsule structure with a multichamber, as shown in Figure 1b,c, in which multiple types of liquids can be capsuled in each super-elastic chamber. As the capsuled liquid in each chamber is evaporated, the pressure induced high strain (160%, and can be up to 900%, determined by the material property of silicone rubber) in the super-elastic shells and can produce desired deformation and force with high resolution. The liquid–vapor phase transition process can provide a stable pressure in the chamber determined by the temperature we set. The soft actuator can display an elongation rate higher than 160% and output force as large as $\approx 250$ times of its mass (14.5 N output force by 6 g self-weight). Due to its self-contained structure and the capability of being remotely controlled, the proposed soft actuator is free from air pumps, wires, and electric boards to achieve controllable actuation, which makes it succinct, entirely soft, self-contained, untethered, and programmable.

![Figure 1](image-url)

**Figure 1.** Scheme of an untethered soft actuator which can be remotely manipulated by nIR irradiation (808 nm in wavelength and light density of 0.8). a) Sunflower shows positive phototaxis, which can be attributed to the perception of light and the volume changes induced by the asymmetric multiplication of cells. b) Scheme of the soft actuator driven by heat, based on the liquid–vapor phase transition of the capsuled liquid. c) The soft actuator with graphene is bending under the irradiation of nIR.
2. Results and Discussions

2.1. Driving Mechanism and Performance of Liquid–Vapor Phase Transition Soft Actuator

The actuation mechanism of the proposed soft actuator relies on the liquid–vapor phase transition of the capsuled liquids, as well as the volume change of the super-elastic shell induced by the internal pressure in each chamber. Given the effect that a wide variety of liquids can be vaporized as long as the temperature stays over their boiling point, multiple types of liquids with boiling points lie within the working temperature range of the adopted silicone (Ecoflex 00-50) super-elastic shell (i.e., from −53 to 230 °C) and can be selected as the driving mediums to meet different actuation needs.

As the temperature rises over the boiling point of the adopted liquid, the capsuled liquid gradually changes state from liquid to vapor through the liquid–vapor coexisting state, and the phase transition process will get to dynamical balance at each specific temperature. Thus, there should be three stages of the soft actuator in the process of driving, which correspond to the three states of the capsuled liquid, i.e., pre-elongation stage at the liquid state, steady-elongation stage at the liquid–vapor coexisting state, and balance stage at the vapor state, as shown in Figure 2a. Here, ethanol is selected to reveal the actuation mechanism of the proposed soft actuator. The capsuled ethanol is 0.5 mL in volume, and the super-elastic capsule structure is initially set to be 11.5 mm in outer diameter, 3 mm in shell thickness, and 58 mm in length.

In the pre-elongation stage (stage I in Figure 2a), as the temperature increased from room temperature (20 °C) to the boiling point (78.4 °C), most of the capsuled ethanol remained at the liquid state, and no apparent elongation occurred. As the temperature increased above the boiling point, the steady-elongation stage (stage II, from 78.4 to 128 °C) started. The internal pressure in the capsuled cell gradually increases along with the liquid–vapor transition proceeding, which gives rise to continuous elongation during the temperature elevation. When the capsuled liquid completely transits to vapor (stage III, from 128 to 200 °C), the elongation is dependent on the internal pressure and the temperature and shows much slower updating along with the temperature elevation in our experiments shown in Figure 2a, which might be attributed to the intense strain constraint by the chamber shell during elongation.

For the steady-elongation stage (stage II), liquid–vapor is dynamically coexisting, whose balance plays a vital role in

Figure 2. Elongation of a soft actuator cell in different stages when heating. a) Temperature-dependent elongation of the proposed soft actuator. The maximum elongation reaches about 160%. The simulation result has a consistent tendency with experimental data, and they are well matched after 10 °C offset of the simulation, which could be attributed to the temperature difference of the inner and outer chamber. Three stages during temperature elevation are clearly revealed, corresponding to the three states of the capsuled liquid (shown by the inset figures), which indicates the controllability of the proposed soft actuator. b) The typical expansions of the proposed soft actuator at different temperatures, with 0.5 mL ethanol capsuled inside. Insets show the finite element analysis results of the deformation at different temperatures. The scale bar is 1 cm.
actuation. The phase-transition process is dependent on the temperature and the stress of the super-elastic shell, which can be mainly depicted by the saturated vapor pressure theory. Clausius–Clapeyron relation (1) reveals the thermodynamic equilibrium of a single-component system and can be applied to estimate the vapor pressures at each specific temperature. Under the three fundamental assumptions,[53] the Antoine equation (Equation (2)) can be derived from Equation (1) to describe the relation between vapor pressure and temperature. The empirical Antoine constants can be obtained by the vapor–pressure plot fitting.[54,55] During stage II, the temperature elevation promotes the liquid–vapor transition, leading to the rise of internal pressure in the chamber. Therefore, the capsule expands, and the internal pressure reduces, accompanying the increase in the stress of the chamber shell. At each specific temperature, the internal pressure in the chamber, volume expansion of the capsule, and the stress of the shell should meet the dynamic balance, which determines the relation of the elongation versus liquid–vapor phase transition. Thus, as the volume of the chamber expands to its maxima and becomes stable at the set temperature, the pressure inside the chamber matches the saturated vapor pressure of the capsuled liquid. This particular value of pressure is used as the input in the calculation of the finite element analysis, to explain and analyze the elongation induced by liquid–vapor phase transition in stage II.

\[
dPdT = \frac{\Delta H}{T\Delta V} = \frac{\Delta H}{T(V_g - V_l)}
\]

(1)

where \(dp/dT\) is the slope of the tangent to the coexistence curve on the pressure–temperature diagram at each point, \(\Delta H\) is the heat of vaporization, \(T\) is the absolute temperature, \(\Delta V\) is the specific volume change of the phase transition, \(V_g\) is the molecular volume of the gaseous phase, and \(V_l\) is the molecular volume of the liquid phase.

\[
P = 10^A + \frac{1}{C}
\]

(2)

where \(P\) is the vapor pressure (mmHg) and \(A, B,\) and \(C\) are component-specific constants. For ethanol, the constants are 7.68117, 1332.04, and 199.2 for the temperature above the boiling point.[56,57]

In stage II, the liquid–vapor coexisting state exists in the temperature range from 78.4 to 128 °C, which is determined in our experiments. As the temperature rises over 128 °C, the capsuled ethanol is mostly transited to vapor phase (stage III), and the volume change of the actuator can be demonstrated by the classical ideal gas equation (Equation (3)), assuming that the capsuled liquid is completely converted to a vapor state. For super-elastic material, the relation of stress versus strain follows the strain energy density function, and it is usually nonlinear for large deformation.[58–60] It is found that the elongation of the soft actuator is little changed in stage III (range from 128 to 200 °C) in our experiments, which indicates that, in this stage, only by temperature elevation, the increase in the internal pressure almost cannot further overcome the constraint of the super-elastic shell.

\[
V = \frac{nRT}{P}
\]

(3)

where \(P, V,\) and \(T\) are the pressure, volume, and absolute temperature, respectively. \(n\) is the number of moles of gas, and \(R\) is the ideal gas constant.

During actuation, as the elongation (as the output in our model) is determined by the liquid–vapor transition, the internal pressure, and the stress of the deformed chamber shell, which are all related to the temperature (as the input), it is crucial to reveal the relationship of elongation versus temperature for depicting the driving diagram of the proposed actuator. A finite element model for deformation is established (Abaqus/CAE). Considering a static situation at a specific temperature, the internal pressure in the chamber could be obtained by the Antoine equation (Equation (2), during stage II) or the classical ideal gas equation (Equation (3), during stage III) and is then taken as an input parameter in the model, to predict the elongation.

The parameters of the adopted material for the chamber shell, Ecoflex 00-50, are the density of Yeoh model, Young’s modulus of 43 kPa, and a Poisson’s ratio of 0.457. The model and simulation details are shown in Supporting Information 6.2.

From Figure 3a and b, it is found that the maximum elongation can reach about 160% at 140 °C, and the tendency can be predicted by the simulation. It is noted that there is an offset of the simulation result compared with the experiment in Figure 3a, which may be attributed to the error made in practical temperature measurement. The temperature was measured by a thermal couple fixed at the outer surface of the capsule, and due to the poor thermal conductivity of the super-elastic material (Ecoflex thermal conductivity is 0.2 W m \(^{-1}\) K \(^{-1}\))[61] there is a temperature difference between the inner chamber and the outer surface. It reveals that, after the 10 °C offset of the simulation curve, the simulation plot fits the experimental results quite well, which indicates the validity of our simulation model. Therefore, the model we proposed can be applied to the design and deformation prediction of the soft actuator, as shown in Figure 2a and Movie S2, Supporting Information. The proposed soft actuator shows a promising driving ability at stage II, which is the liquid–vapor coexisting state. In the following context, the driving ability at stage II will be discussed in detail.

To demonstrate the actuation capabilities of our proposed soft actuators, the relationship of actuation stroke (the elongation) versus temperature is measured and analyzed. As shown in Figure 3a, the actuation stroke stepped grows along the temperature elevation (with a step of 10 °C) and reaches the maximum stroke of 35.4 mm with an uncertainty of ±0.15 mm at 140 °C. This performance can nearly match the origami-based pneumatic soft actuator (200%)[40] and is larger than that of liquid crystal elastomers (130%) and multimaterial soft actuator (<110%).[62,63] The proposed actuator also shows promising linearity of stroke versus temperature during driving, as Figure 3b shows, with a slope of 0.5 mm °C\(^{-1}\) and Pearson’s R of 0.999. In addition, multicycle unidirectional actuation has also been tested, as shown in Figure 3c, to reveal the repeatability during actuation, and the maximum driving rate is 3.28 mm s\(^{-1}\), whereas the maximum recovery rate is 2.02 mm s\(^{-1}\). As shown in Figure 3d, the unidirectional repeatability of the soft actuator is ±0.5 mm, the accuracy of maximum stroke is within 1%, and the recovery accuracy within 2%. Figure 3e shows the response time of around 55 s, as the soft actuator is driven by a 220 °C heating source (a heat gun, with a distance of 5 cm
Figure 3. The driving performance of the untethered soft actuator. a) Gradient-increased heating test of the soft actuator, from 60 to 140 °C. The soft actuator elongated 35.4 mm (which is 160% of its initial length), and the response time of each step is within 50 s. b) The temperature dependence of the elongation of the soft actuator. It is clear that the driving elongation is linear dependent on the temperature from 75 to 128 °C, with a slope of 0.5 mm °C⁻¹, and the uncertainty of the measured elongation is ±0.15 mm. c) Repeatability of the driving property. The temperature of the soft actuator is set at 140 °C, corresponding to the elongation of 160%, and the unidirectional repeatability is within ±0.5 mm. d) The repeatability of the elongation and the recovery value during the cycled heating–cooling process of the soft actuator. The accuracy of elongation is within 1%, and the recovery accuracy is within 2%. e) The time-response property of the soft actuator. There is a lag of about 50 s as the elongation responded to the temperature, and the elongation needs around 55 s to get to a steady value. f) The output force of the soft actuator. The maximum output force is 14.5 N at 140 °C, and the output force shows a linear dependence on the temperature in the range from 75 to 110 °C, with a slope of 0.33 N °C⁻¹ and Pearson’s R of 0.998.
to the actuator, inducing heat loss), which can be improved by optimizing the heating method. We noticed that there is a 50 s time lag between elongation starting and the temperature rising, and it could be attributed to the dynamical process of the liquid–vapor phase transition, during which it took time for the internal pressure to overcome the deformation stress of the capsule shell. The high control accuracy and excellent linear correlation with the temperature of the proposed soft actuator provide a new way to support high-precision control in the development of soft robotics and supply an alternative method to develop reversible precision actuators for soft robots.

The driving force is also one of the critical roles in soft actuators, which dramatically affects their potential applications. To characterize the mechanical properties of the proposed soft actuator, the actuator was positioned and radially constrained through a glass tube. As shown in Figure 3f and Figure S1, Supporting Information, the temperature dependence of the output force exhibits a similar profile to the elongation (Figure 2a). Apparently, the force outputs mainly in the steady-elongation stage (stage II) with a maximum output force of 14.5 N, which is about 250 times its weight (6 g). During stage II, it is also noted that the temperature dependence of the output force is approximately linear, with a slope of 0.33 N°C⁻¹ and Pearson’s R of 0.998. The linear properties in the driving zone, i.e., the actuated elongation and the output force, bring great convenience to the subsequent designs of the soft actuator.

2.2. Driving Strategy of the Untethered Soft Actuator

The ability for complex 3D movements is greatly desired for soft robotics. Thanks to its self-contained capsule structure of the proposed soft actuator, it is convenient and promising for complex movements. As mentioned earlier, for a single cell (a specific liquid encapsulated in a single super-elastic chamber), the final deformation is determined by the internal pressure that is induced by the liquid–vapor phase transition and the stress of the super-elastic shell. So homogeneous deformation for pneumatic actuators, e.g., elongation along z-axis while expanding along x- and y-axis, coexists, which can be attributed to the isotropic constrain of the super-elastic shell. To achieve anisotropic motion basically represented by elongation, bending, and twisting, the shell structures need to be carefully designed. Braided structures are widely used to supply anisotropic constraints in soft actuators, similar to those used in the McKibben actuators.[4,6] Inspired by this design method, polyimide (PI) thread, which has a high modulus of elasticity (about 70 GPa for PI fiber) and is resistant to high temperature, is used to coil the soft actuator to enhance the actuation performances of the elongation and twisting, so that it is possible to obtain anisotropic deformation. Using the cross-coiling method, as shown in Figure 4a, the soft actuator can elongate straightly under symmetrical constraints. By constructing asymmetrical constraints, such as single-direction coiling shown in Figure 4c, the soft actuator will twist counterclockwise if the coiling is clockwise. In this case, the twisting angle is determined by the temperature and the coiling structure, which could be precisely regulated from 0° to 180°. For bending, it can be well regulated by the shell thickness of the chamber (d₁ and d₂ in Figure 4b), and the soft actuator will bend to the thicker side in a uniform thermal field in virtue of the asymmetric constraint.

For soft robotics, stepped complex movements can be achieved by the combination of several actuators, which is similar to the multijoint movement, even though the structures and actuating strategies are usually complicated. In this study, we further explore the potential applications of the proposed soft actuator in achieving programmable complex motions by one single cell rather than conventional reported multijoint systems, which would supply an alternative for compact and integrated actuators. Three different designs have been adopted, i.e., multichamber design, multiliquid design, and hybrid design, by combining the multichamber with multiliquid. Figure 4d shows the multichamber design of the proposed soft actuator. There are several independent chambers in one single cell, and each chamber is filled with the same liquid (the liquid volume can be different, which is essential for the actuation, determined by the movement design). We can heat one or more chambers programmatically at different periods by remote near-infrared (nIR) irradiation to control the liquid–vapor transition state in each chamber, thereby driving the actuator cell to perform a compound movement. To further improve the programmable driving abilities, multiliquid design, in which several liquids (i.e., ethanol, water, and n-hexane) with different boiling points are filled in the chamber of the actuator cell, is proposed to achieve complex driving movement by regulating the temperature, as shown in Figure 4e. For a single liquid in an actuator cell, the relationship of driving movement versus temperature can be described as shown in Figure 2a, which would be saturated when the adopted liquid is almost entirely transited to vapor. For multiliquid design, a stepped temperature response can be achieved. When the temperature gradually rises, one liquid with a low boiling point will first be transited to vapor, and the soft actuator achieves the first-step actuation. As the temperature rises and reaches the boiling point of another type of liquid, the second-step actuation will be triggered, and so forth. By carefully selecting the matches of different liquids, considering material properties such as mutual solubility and density, multistep actuation by regulating the temperature can be performed as design. The hybrid design by the combination of multichamber and multiliquid, as shown in Figure 4f, can support more complex movements, such as the flexible swing of the elephant trunk. These novel and useful design methods provide an alternative approach for soft robotics to achieve dexterous bionic movements.

Although the anisotropic design of the shell with multichamber and multiliquid design supplies promising routes for compound movements, it also introduces difficulties to fabrication. The multinozzle in situ 3D printing provides an alternative and promising way to fabricate complicated structures, which is compatible with the fabrication of the proposed soft actuators.[10,65–70] In further studies, we have already developed an integrated fabrication process using 3D printing for the proposed soft actuators, which is addressed in previous studies.[71,72]

2.3. Remote Control and Programmable Actuations

The untethered and programmable control is promising and desired for soft robotics, which will significantly broaden their
In this study, the self-contained multichamber or multiliquid design brought unique convenience to the untethered and programmable control. A set of nIR lasers (808 nm in wavelength and light density of 0.8 W cm\(^{-2}\) for each of the nIR lasers, whereas the solar mediation intensity is 0.1 W cm\(^{-2}\)) were used to provide remote heating of the multichamber soft actuator. By the preprogrammed localized heating strategy, each chamber of the soft actuator could be heated independently in a predesigned sequence to achieve controllable and dexterous movements.

Figure 5a shows an artificial adaptable phototaxis using the proposed soft actuator with three chambers. Similar to the phototaxis growth of sunflower, when the laser set is turned on programmatically, an actively adjustable motion can be triggered, and the photophobic movements can be achieved by sequentially activating each chamber. As shown in Figure 5a,b, the soft actuator is centered, surrounded by three sets of nIR heating sources matching the three chambers. When each chamber is activated by nIR irradiation, the soft actuator will show negative phototaxis and bend toward the opposite direction of the heating source. Synthetic motions can be achieved by combining the activation of different chambers, e.g., the actuator will bend to the inactive direction when two adjacent chambers are active simultaneously and elongate upward when all three chambers are active, as shown in Movie S3, Supporting Information. The remotely controllable bionic movement of this soft actuator provides new inspirations on the design of the soft robotics.

Similar to the combination of muscles and bones in live creatures, the integration of soft actuators and rigid structures brings together both of the advantages, such as the robustness and safety of soft actuator and the capacity and high precision of rigid mechanisms, providing great promising applications. Here, we suggest a biology-inspired design in which...
the proposed soft actuators act as the driving muscles to actuate rigid structures. As shown in Figure 6a,b, with the heat from a set of nIR lasers, the soft actuator extends and pushes a raised arm down by inducing a bending at the elbow. As the soft actuator can push the arm from the bent state to the fully extended state (the arm is driven to rotate 43° and can be more than 90°), it exhibits excellent bonding ability with the rigid parts, due to its untethered design. We can also implement the actuator at the joint of the leg, which drives the calf to move forward around the knee to kick a ball. The actuators are driven by the noncontact photothermal driving, which moves smoothly and stably, as shown in Figure 6c–e and Movie S4 and S5, Supporting Information.

The soft actuator exhibits excellent compatibility with the rigid components in the experiments, which indicates that it can be applied to hybrid robots and mechanical exoskeletons for lightweight and flexible design.

3. Conclusion

In this study, we propose a novel self-contained soft actuator based on the liquid–vapor phase transition, which is untethered and programmable. It is revealed that large stroke (35.4 mm, 160% elongation concerning its initial length) and prodigious output force (14.5 N provided by a 6 g self-weight) can be achieved during the actuation stage. Experiments also indicate that the proposed actuator shows linear actuation properties, such as a driving stroke of 0.5 mm °C⁻¹ with Pearson’s R of 0.999 and an output force of 0.33 N °C⁻¹ with Pearson’s R of 0.998. The driving stroke has an uncertainty of ±0.15 mm and repeatability of ± 0.7 mm. The self-contained structure of the proposed soft actuator can be broadened to multichamber, multiliquid, and hybrid designs for complex motions in programable and remote controls. The soft actuator presented in this study has dimensions in centimeters due to the manufacturing process, but it can be shrunk to millimeters and micrometers with more advanced soft materials 3D printing technologies.[77–79] This self-contained, untethered soft actuator establishes a foundation in addressing the critical challenges in emulating the movement of creatures in nature and suggests a feasible approach to develop smart, soft, and autonomous robotics. The entirely soft robotics of this kind holds promise in many fields, ranging from surgery to rehabilitation and rescue.

4. Experimental Section

Materials Prepared for the Fabrication of Untethered Soft Actuator: We used a fused deposition modeling-based 3D printer to make the casting mold with complicated structures. The material we chose was polylactic acid (PLA). Oily release agent (LR-11, YinJing, Inc) was introduced to help mold release. Two-part silicone elastomer (Ecoflex series 00-50, Smooth-on, Inc.) was utilized to fabricate soft structure with soft cavity.
using soft lithographic molding. The molds were reusable, and the soft 
structures were easy to be produced in bulk at a relatively low cost. 
Absolute ethanol (EtOH, 99.5%) was used as the liquid–vapor phase
change material.

Fabrication of Untethered Soft Actuator by Liquid–Vapor Phase Transition:
The soft actuator we proposed had a capsule structure with liquid–vapor 
phase-changeable materials inside, and its internal space was divided 
into several chambers as needed. For molding, premixed Ecoflex was 
poured into the predesigned sectional molds and heated to 60 °C for 
30 min, which was followed by pouring the liquid–vapor phase-
changeable material into the cavity and sealing the capsule with 
Ecoflex prepolymer. The soft actuator was wound around by PI thread 
if needed. When using the nIR laser to drive the soft actuator, 
0.6 wt% graphene was needed to be added into the capsule structure 
to improve the photothermal conversion efficiency. We also used carbon 
nanotubes or even graphite powder to improve the thermal conductivity 
of Ecoflex. However, we found that the efficiency of photothermal driving 
was the highest when doping graphene in the shell material in our experi-
ments, which significantly reduced the time taken of the driving stage 
of the soft actuator. Details of fabrication are shown in Supporting Information 6.1.

Characterization of Untethered Soft Actuator by Liquid–Vapor Phase Transition: We used an optical microscope (Nikon) to observe and mea-
sure the dimensions of the elastic capsule and its cross section. Ordinary 
optical photos and videos were recorded by an SLR camera (Nikon D750). 
A universal tension and compression testing machine (TY8000, Tianyuan, 
Inc., China) was used to investigate the output force.

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

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L.Y. conducted the mechanical analysis. W.J., D.N. guided the material 
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