Title: Untethered unidirectionally crawling gels driven by asymmetry in contact forces

Authors: Aishwarya Pantula, [1] Bibekananda Datta, [2] Yupin Shi, [3] Margaret Wang, [1] Jiayu Liu, [2] Siming Deng, [2] Noah J. Cowan, [2,3] Thao D. Nguyen, [2,4,5] David H. Gracias [1,3,5-9*]

Affiliations:

[1] Department of Chemical and Biomolecular Engineering, Johns Hopkins University, Baltimore, MD 21218, USA.

[2] Department of Mechanical Engineering, Johns Hopkins University, Baltimore, MD 21218, USA.

[3] Laboratory for Computational Sensing and Robotics (LCSR), Johns Hopkins University, Baltimore, MD 21218, USA.

[4] Hopkins Extreme Materials Institute (HEMI), Johns Hopkins University, Baltimore, MD 21218, USA.

[5] Department of Materials Science and Engineering, Johns Hopkins University, Baltimore, MD 21218, USA.

[6] Department of Chemistry, Johns Hopkins University, Baltimore, MD 21218, USA.

[7] Department of Oncology, Johns Hopkins School of Medicine, Baltimore, MD 21205, USA.

[8] Sidney Kimmel Comprehensive Cancer Center (SKCCC), Johns Hopkins School of Medicine, Baltimore, MD 21205, USA.

[9] Center for MicroPhysiological Systems (MPS), Johns Hopkins School of Medicine, Baltimore, Baltimore, MD 21205, USA.

*Corresponding author email: dgracias@jhu.edu

Abstract: Reversible thermoresponsive hydrogels, which swell and shrink in the temperature range of 30-60 °C, provide an attractive material class for operating untethered soft robots in human physiological and ambient conditions. Crawling has been demonstrated previously with thermoresponsive hydrogels but required patterned or constrained gel substrates to break symmetry for unidirectional motion. Here, we demonstrate a locomotion mechanism for unidirectionally crawling gels driven by spontaneous asymmetries in contact forces during swelling and deswelling of segmented active thermoresponsive poly (N-isopropyl acrylamide) (pNIPAM) and passive polyacrylamide (pAAM) bilayers with suspended linkers. Actuation studies demonstrate the consistent unidirectional movement of these gel crawlers across multiple thermal cycles on flat, unpatterned substrates. We explain the mechanism using finite element simulations and by varying experimental parameters such as the number of segments, linker stiffness, and design. We elucidate design criteria and validate experiments using image analysis and finite element models. We anticipate that this mechanism could potentially be applied to other shape-changing locomotors.

One-Sentence Summary: We manipulate geometry to break symmetry in a multisegmented thermoresponsive locomotive gel robot.

Main Text:

INTRODUCTION

Stimuli-responsive, untethered gels that can traverse aqueous environments can have biomedical, microfluidic, and soft robotic applications. (*1–3*) Compared to tethered robots that utilize fluidic or pneumatic signals to control their movement, untethered hydrogel robots can be mass-produced, are maneuverable in tight spaces, and can operate in the absence of wiring or external power sources. (*4*) Particularly, stimuli-responsive hydrogels, which can undergo volumetric phase transitions (such as swelling and deswelling) in response to environmental stimuli such as heat, light, pH, chemicals, biochemicals, and electromagnetic fields, are attractive materials for building untethered robots. (*3–8*) Previously, researchers have structured these materials to create a range of shape-changing and functional transformer hydrogels such as grippers, fluidic actuators, and valves. (*3, 9–11*)

A relevant class of stimuli-responsive hydrogels is based on reversibly-thermoresponsive poly (N-isopropylacrylamide) (pNIPAM). These hydrogels undergo lower critical solution temperature (LCST) transitions in the human physiological and ambient temperature range. Moreover, the LCST can be tuned by varying the side-chain length, crosslinking with compatible copolymers, or mixing with ionic liquids. (*12, 13*) Prior studies have utilized molding, 3D printing, and stereolithography to create micro and mesostructured pNIPAM structures capable of locomotion. For example, Maeda et al. used the Belousov–Zhabotinsky oscillatory reaction to locomote a catalyst modified pNIPAM strip at a constant temperature. (*14*) Elsewhere, researchers have used inherently simple single-segment pNIPAM bilayers to create a variety of crawling robots. (*15–19*) However, it has been challenging to break morphological symmetry (a fundamental requirement of directed locomotion) using just swelling and deswelling of a gel bilayer. Consequently, attempts at creating bilayer hydrogel crawlers have necessitated ratcheted substrates or constrained channels to break symmetry and direct motion, limiting the applicability of these robots. (*19*) While it has been possible to break symmetry using cardiac cell-based robots, this scheme requires the patterned deposition of proteins and live cells on some parts of the robot, not others. (*20, 21*)

Here, we describe the use of systematic and asynchronous changes in robot morphology, driven by the transient swelling and deswelling of connected gel segments, to break the spatial (fore-aft) symmetry of contact forces and direct motion on flat surfaces. We show that symmetry is spontaneously broken simply by asymmetry in contact surface forces caused by the structural design of the robot without the need for additional patterning on the gel surface or substrate. We used gel bilayers with different thickness ratios and lateral dimensions to induce spatiotemporal asymmetries in swelling and deswelling. We also used a suspended linker with different stiffness and varied the number of bilayer segments and linker pattern to manipulate contact force asymmetry and tune robot displacement. We fabricated our robots by direct ink writing (DIW), a 3D printing technique that uses pressure-based extrusion for layer-by-layer assembly of multiple materials. We characterized the robots over multiple heating and cooling half-cycles and simulated their thermo-chemo-mechanical behavior using finite element analysis. We developed a conceptual framework for designing tunable, scalable, and versatile untethered stimuli-responsive locomotors that move unidirectionally. (*10, 22–29*)

RESULTS

Design and fabrication of multisegmented bilayer gel robots

Crawling via cycles of musculoskeletal traveling waves provides a common and effective means of locomotion for organisms across taxa, including snakes, salamanders, and inchworms. (*30–35*) Usually, such traveling waves emerge due to a spatiotemporal pattern of descending neural commands (i.e., from a central pattern generator (*36*)) that drives muscles to create kinematic phase differences along a fore-aft body axis.

Here, we propose a locomotion strategy that exploits the geometry of actuators by engineering morphological asymmetries in lateral dimensions and thickness of bilayers to generate the requisite fore-aft phase differences during swelling and deswelling without needing to use any independent stimuli. We designed morphological gradients along the fore-aft axis of the soft robotic chassis, which lead, in turn, to gradients in actuator dynamics, ultimately instantiating a wave-like locomotor gait that only requires a single input.

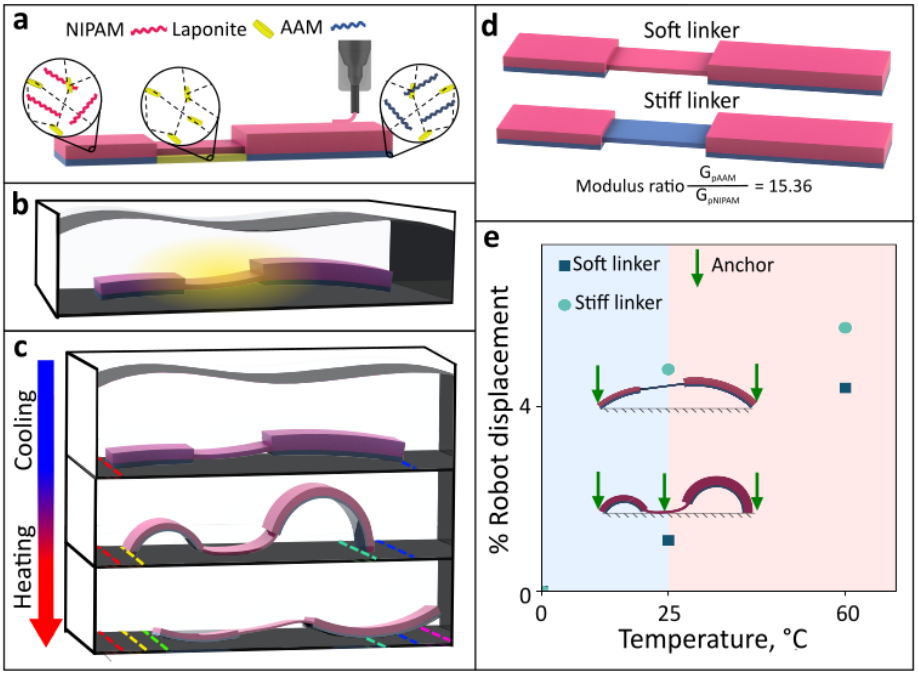

Figure 1. Design and fabrication of multisegmented gel robots with a suspended linker. (a) 3D printing process. **(b)** Sacrificial layer dissolution. **(c)** Thermal actuation. One thermal cycle comprised a cooling half-cycle at 25 °C and a heating half-cycle at 60 °C. **(d)** Varying linker stiffness. **(e)** Varying the contact force distribution using linker stiffness to increase robot actuation distance during thermal cycling. The percentage robot displacement is the distance crawled normalized by its body length in one thermal cycle. The area shaded in blue describes the cooling half-cycle, and the area in red describes the heating half-cycle.

We designed multisegmented robots with two bilayers of different morphology and bilayer ratios connected by a suspended linker (**Figure 1a**). The bilayers were composed of a thermally responsive swelling gel (active material) and a non-swelling gel (passive material). We used DIW, an extrusion-based 3D printing technique, to fabricate our bilayers and robots. We used NIPAM as our active ink and acrylamide (AAM) as our passive ink. We made the inks by mixing the monomers with a shear-thinning additive Laponite nano-clay, in water. After printing, we crosslinked the structures by UV exposure. (*10, 29*) From our differential scanning calorimetric

(DSC) measurements (**Fig. S15**), we observed that the lower critical solution temperature (LCST) of pNIPAM was 35.7 °C.

We introduced a suspended central linker to couple the asymmetric bilayer segments. As opposed to a non-suspended linker that maintains continual contact with the substrate, we could tune the contact area between the suspended linker and the substrate based on its stiffness (described later). We developed a sacrificial ink composed of an aqueous Laponite solution that supported a second gel layer during fabrication. Since Laponite did not crosslink during UV curing, it dissolved readily when the structure swelled in water to create the suspended linker. (**Figure 1b**).

Following the sacrificial layer dissolution, we conducted thermal cycling of our robots in water by setting the temperature to 25 °C for the cooling half-cycle and 60 °C for the heating half-cycle (**Figure 1c**). We heated the actuation setup well above the LCST to ensure that the temperature of the gel itself was above the LCST since there were thermal gradients in the water. We modified linker stiffness, morphology, and the number of bilayer segments to investigate their influence on the asymmetry and crawling displacement of the robots (**Figure 1d,e**).

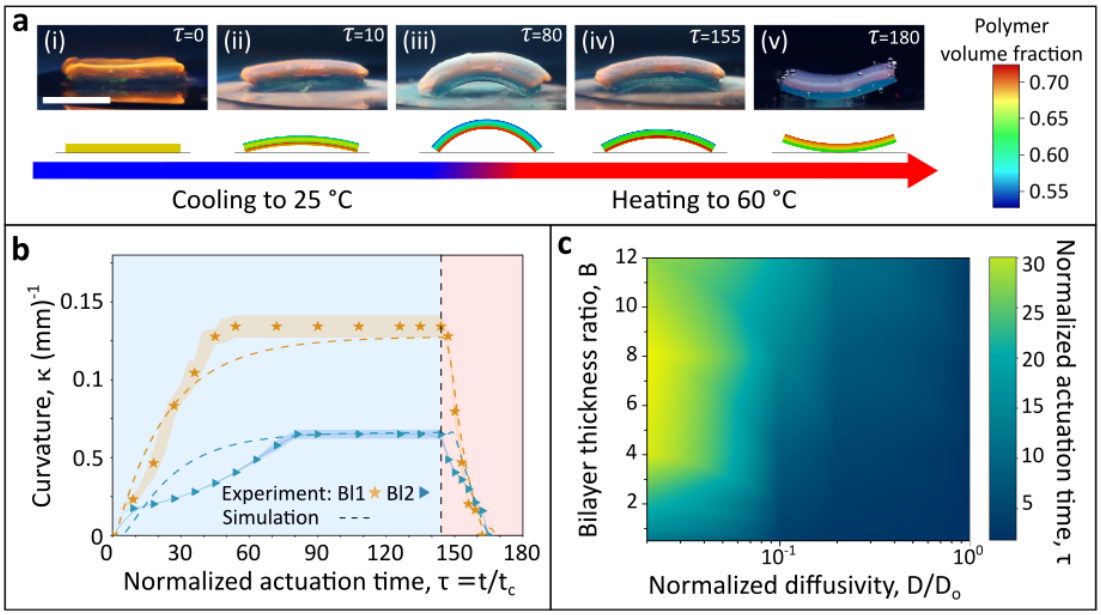

Figure 2. Tuning actuation time and curvature of the bilayers by varying thickness ratio during thermal cycling. (a) Experimental images and simulation snapshots of the bending of the 0.8 mm pNIPAM-0.4 mm pAAM bilayer versus normalized actuation time defined as $\tau = t/t_c$, (where $t_c = l_c^2/D_o$, $l_c = 1$ mm, $D_o = 5\times10^{-9}$ m^2/s). The images were taken during one thermal cycle by first cooling to 25 °C **(i-iii)** and then heating to 60 °C **(iv-v)** over ten hours. D_o is the self-diffusivity of water at 60 °C. (*37*) The bilayer deswelled beyond its cured state and achieved negative curvature at the end of heating **(v)** $\tau = 180$. The scale bar in **(i)** indicates a length of 10 mm. The color scaling gradient indicates the polymer volume fraction from the finite element simulations with lower numbers indicating larger swelling. The polymer volume fractions at cured state, fully swollen at 25 °C, and fully deswollen state were 0.67, 0.59, and 0.72, respectively. **(b)** Plot of the curvature of the 0.8 mm pNIPAM-0.4 mm pAAM bilayer (Bl1) and 0.8 mm pNIPAM-0.8 mm pAAM bilayer (Bl2) observed over normalized actuation time during the cooling (blue area) and heating (red area) half-cycles. Both bilayers fully swelled by $\tau = 80$ and deswelled by $\tau = 165$. The data points are from experiments with the shading representing the standard deviation with a sample size of three. The dashed lines represent the fitted model from simulations. **(c)** Plot of the normalized actuation time vs bilayer thickness ratio, $B = h_{\text{pNIPAM}}/h_{\text{pAAM}}$, and normalized diffusivity, D/D_o.

The morphology of the bilayers can be tuned during swelling and deswelling

The relative transient swelling characteristics of the two bilayers were important to achieve the desired morphological asymmetry of the crawling robot. We measured the transient curvature of rectangular bilayers with varying dimensions over a thermal cycle (cooling and heating half-cycles). We used 10 mm × 15 mm rectangular bilayers with two thicknesses of the pAAM layer (0.4 mm, 0.8 mm) and four thicknesses of the pNIPAM layer (0.4 mm, 0.8 mm, 1.2 mm, and 1.6 mm). We plotted the radius of curvature of the bilayers during the heating and cooling half-cycle against the time *t,* normalized by the characteristic diffusion time t_c of the bilayer: $\tau = t/t_c$. We defined the characteristic diffusion time as $t_c = l_c^2/D_o$, where $l_c = 1$ mm is the characteristic diffusion length of the bilayer and $D_o = 5\times10^{-9}$ m^2/s is the self-diffusivity of water at 60 °C. (*37*) During the cooling half-cycle, the thinnest bilayer reached its equilibrium curvature fastest because of its smaller characteristic diffusion length. It also had the highest curvature of all the bilayers because it had the lowest bending stiffness. (**Figure 2 a, b, and Fig. S2**). We observed that during the cooling half-cycle, the normalized swelling time varied widely (τ = 50-110) because of the large difference in the pNIPAM thickness across the bilayers. A thicker pNIPAM layer results in a larger characteristic diffusion length and bending modulus. During the heating half-cycle, the bilayers deswelled twice as fast (τ = 150-180). *(38)*

We applied finite element analysis (FEA) of the swelling- and deswelling-induced transient bending of the bilayers to determine the swelling and deswelling properties of the hydrogels (**Table S2**). We described the rapid deswelling of pNIPAM by modifying the constitutive model to include two different diffusivities for the swelling and deswelling process. We applied FEA to investigate the effects of material and geometric properties of the bilayer on the actuation time, defined as the time required to reach 63.2% of the equilibrium curvature (**Figure 2c**). The actuation time was most sensitive to the diffusivity of the pNIPAM and the bilayer thickness ratio. Bilayers with a smaller thickness ratio and a larger diffusivity for the active material produced shorter actuation times.

The flexible linker and morphology of the bilayers break symmetry

We hypothesized that two dissimilar gel bilayer segments connected by a suspended linker would generate asymmetry in contact forces leading to unidirectional robot displacement during thermal cycling. We utilized the bilayer thermal cycling data to choose two bilayers with the greatest curvature difference. We picked a small (0.8 mm pNIPAM-0.4 mm pAAM) bilayer and a large (1.2 mm pNIPAM-0.4 mm pAAM) bilayer and connected them with a suspended flexible pNIPAM linker. The specific lateral and thickness dimensions of the robot are in **Fig. S4a**. We thermally stimulated the robot in water by switching the temperature between 25 °C and 60 °C.

During the cooling half-cycle, the robot was allowed to swell from its cured state at 60 °C to room temperature (25 °C), and we observed swelling-induced bending in both bilayer segments. Simultaneously, the suspended linker swelled and sagged to touch the underlying surface along with the outer edge of both bilayers. This sagging caused the linker to act as an anchor and move the bilayers towards it by the end of the half-cycle (**Figure 3a(i-iii)**). Once we switched to the heating half-cycle, the robot transitioned from its swollen-bent state to a deswollen-flattened state (**Figure 3a(iii-v)**). The linker straightened and achieved its original suspended state while the two bilayers deswelled asynchronously. During deswelling, due to size and curvature asymmetry between the larger and smaller bilayer, the anchor was transferred from the linker to the larger bilayer. As a result, the robot moved unidirectionally as the small bilayer and linker were drawn toward the larger bilayer.

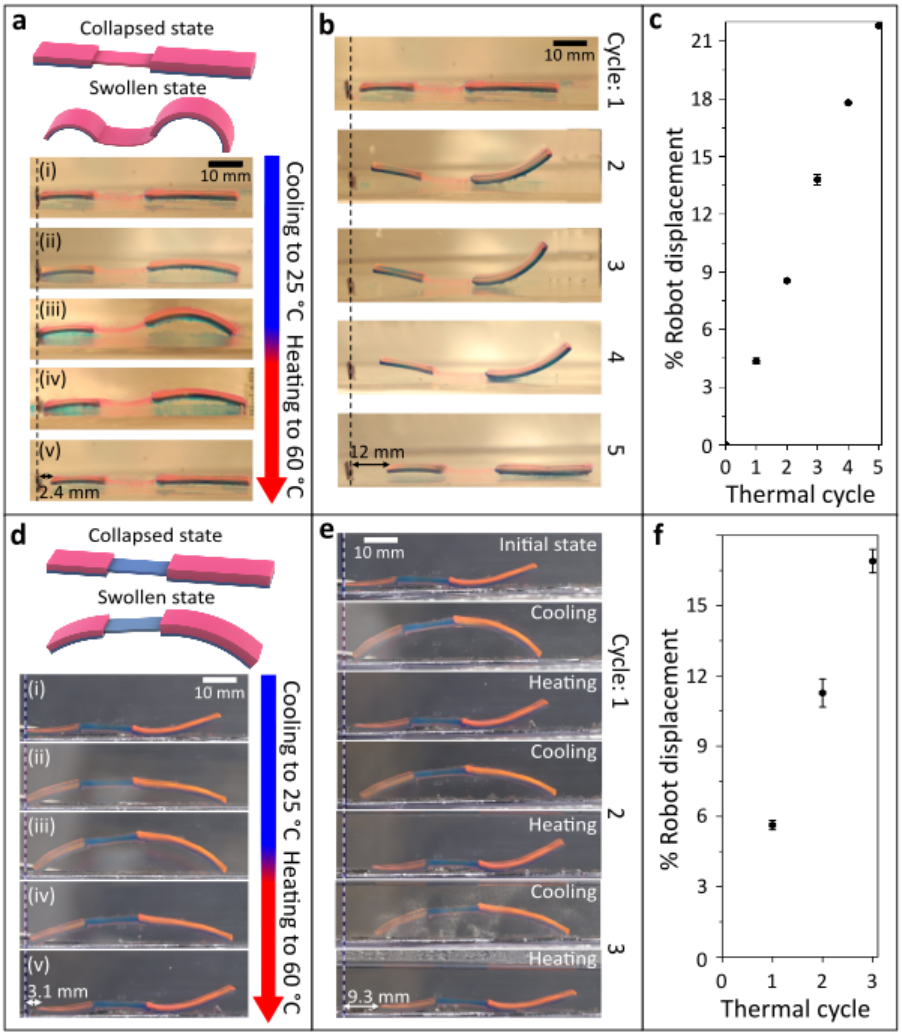

Figure 3. Thermal cycling of two-segment robots with a flexible or a stiff linker. Robot with **(a-c)** a flexible pNIPAM linker. **(a)** Experimental images of the robot across one thermal cycle consisting of one cooling/heating half-cycle. **(i- iii)** Cooling half-cycle: **(i)** Cured state, **(ii)** intermediate state, and **(iii)** swollen state. **(iv-v).** Heating half-cycle: **(iv)** Intermediate state, and **(v)** deswollen state of robot. **(b)** Experimental images of the robot observed at the end of each of five thermal cycles. **(c)** Plot of the percentage robot displacement at the end of each thermal cycle. This robot with a flexible linker crawled 12 mm after five cycles which is equivalent to 4.4% body length/cycle. **(d-f)** Robot with a stiff pAAM linker. **(d)** Experimental images of the robot across one thermal cycle consisting of one cooling/heating half-cycle. Cooling half-cycle: **(i)** Cured state, **(ii**) intermediate state, and **(iii)** swollen state. Heating half-cycle: **(iv)** Intermediate state, and **(v)** deswollen state of robot. **(e)** Experimental images of the robot observed at the end of each of three thermal cycles. **(e)** Plot of the percentage robot displacement at the end of each thermal cycle. This robot with the stiff linker crawled 9.3 mm in three cycles which is equivalent to 5.6% body length/cycle. Error bars in **(c)** and **(f)** indicate a standard deviation with a sample size of three.

We subjected this robot to five thermal cycles and measured the displacement of the robot after each cycle (**Figure 3b, c**). We observed that our robot crawled consistently in the direction of the larger bilayer, moving approximately 4.4% body length/cycle. The final curvature of the robot segments varied slightly with each cycle, likely due to differences in the reorganization of the molecular chains of the crosslinked polymer and hydrogen bonding that occur between heating and cooling half-cycles. Also, the pNIPAM layer deswelled beyond its cured state length and caused the bilayers to bend in the opposite direction as compared to its swollen state. (*39*)

Varying the linker stiffness speeds up the gel crawler

We hypothesized that the stiffness of the linker would strongly influence the morphology and, consequently, the gait and contact forces of the robot. To study this effect, we printed the same two-segment robot with identical dimensions but replaced the flexible pNIPAM linker with a stiff pAAM linker (**Figure 3d-f**). Notably, during the cooling half-cycle, the stiff linker held its flat shape. The stiff linker remained suspended between the bilayers and did not play any role in anchoring. As a result, there was an increase in the net contact force asymmetry between the larger and smaller bilayer, and the unidirectional displacement increased from 4.4 to 5.6 % body length/cycle) (**Movie S1**), proving our hypothesis.

We also confirmed that the locomotion direction of the robots depends on the relative orientation of the small and large bilayers. We thermally stimulated both the flexible and stiff linker robots over 20 thermal cycles and observed consistent unidirectional motion in the direction of the larger bilayer (**Fig. S4b,d**).

Asymmetry in contact forces generates unidirectional motion

We obtained insight into our experimental findings by performing finite element analysis of the thermo-chemo-mechanical behavior of the robot during thermal cycling. The constitutive model by Chester et al. assumed that the hydrogel was in a dry polymer state at the beginning of the simulation. (*40*) In contrast, the hydrogels that we fabricated by DIW were in a partially swollen state. To achieve this initial swollen configuration, we simulated the free swelling of both the pNIPAM and pAAM materials at a reference temperature of 50 °C as described in the Supplementary Materials**.** The effects of gravity and buoyancy were prescribed by applying a body force, $b_y = \Delta\rho g$, where $\Delta\rho = 100$ kg/m^3 is the difference in the density between the polymer and water and g is the gravitational acceleration. We discretized the bottom surfaces of all three segments of the robot (**Fig. S11**) using contact surface elements. We assumed a rigid surface underneath the hydrogel segments to prevent interpenetration. The surface was assumed to have a frictional coefficient, $f_k = 0.1$, to describe the frictional sliding of the robot during the thermal cycle. For low sliding velocities, most literature reported the coefficient of friction of pAAM hydrogel to be below 0.1. (*41*) We chose $f_k = 0.1$ to ensure a sufficient difference in the friction force between the segments to produce locomotion.

We lowered the temperature from 50 °C to 25 °C at a rate of 1.67 °C/min and held it at 25 °C for 2.45 hours, then increased the temperature to 60 °C at 0.58 °C/min and held it at 60 °C for 2 hours, to simulate the actuation experiments. This temperature ramping caused the thermally responsive pNIPAM to swell during the cooling half-cycle and deswell during heating half-cycle. The swelling strain mismatch between the active pNIPAM and passive pAAM hydrogel caused the bilayer segments to curve downward during cooling, pushing the pNIPAM linker into contact with the surface.

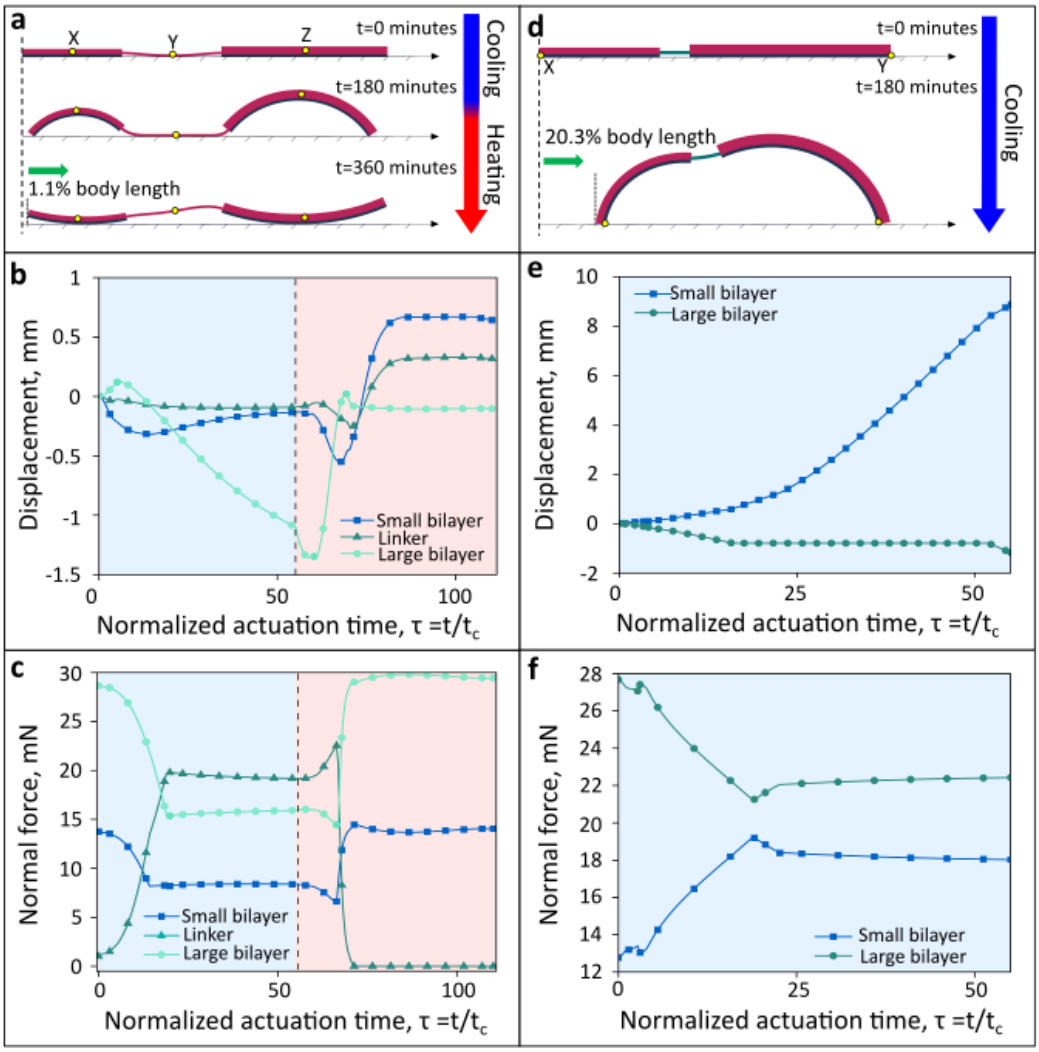

Figure 4. Finite element analysis of a two-segment robot with different linkers. (a-c) Robot with a flexible linker. **(a)** Temporal snapshots of the robot crawling upon cooling ($t = 180$ minutes) and heating ($t = 360$ minutes). **(b)** A plot of the lateral displacement of the center of the bilayers and the linker versus normalized time for cooling (blue area) and heating (red area) half-cycles. **(c)** A plot of the normal force (the force perpendicular to the contact surface) exerted by different segments on the contact surface versus normalized time. **(d-f)** Robot with a short and stiff linker. **(d)** Temporal snapshots of smaller bilayer moving towards the larger bilayer upon cooling. **(e)** A plot of lateral displacement of the outer edges of the bilayers versus normalized time. **(f)** A plot of the normal force on the contact surface exerted by the bilayers versus normalized time.

The left edge of the smaller bilayer moved to the right, and the right edge of the larger bilayer moved to the left (**Figure 4a**). The opposing motion of the bilayer segment resulted in a minimal 0.1% translation of the center of the linker of the robot. The mismatch in the deswelling strain between pNIPAM and pAAM caused the robot to curve upward during heating. The curving/bending of the larger bilayer and deswelling of the linker pulled the robot to the right and caused a net displacement of 1.1% body length over the thermal cycle. (**Figure 4b**).

We plotted the normal force (the force perpendicular to the contact surface) acting on the two bilayer segments and linker to understand the mechanism of forward translation of the hydrogel robot (**Figure 4c**). We calculated this by summing the nodal normal forces on the contact surface of an individual segment. The normal force scales with the magnitude of the friction force as the inverse of the Coulomb friction coefficient.

Initially, the normal force was highest for the larger bilayer because it was the largest segment in the robot. During cooling, the bending of the bilayers pushed the linker down on the surface, which caused it to experience the largest normal force and, thus, the largest friction force. The normal force acting on the bilayers decreased approximately by 50%. At the same time, the normal force increased by more than four times than for the linker. The higher normal force caused the linker to act as an anchor and the center of both bilayers to move toward the linker. Increasing the temperature to 60 ºC caused both bilayers first to straighten and then curl upward, lifting the linker from the contact surface. The larger bilayer was displaced by -0.1 mm during heating, whereas the smaller bilayer was displaced by 0.63 mm (**Figure 4a**). Once again, the larger bilayer experienced the largest friction force of the three segments and acted as an anchor, allowing the smaller bilayer and linker to slide to the right, producing a net forward motion (**Movie S2**).

We performed three additional finite element simulations to validate the hypothesis for the mechanism of the robot locomotion. First, we repeated the finite element simulations for a frictionless contact surface ($f_k = 0.0$). The robot experienced no net translation (**Fig. S13**). We next changed the frictional anchoring of the robot by switching the positions of the smaller and larger bilayers to produce net motion in the opposite direction. Having the larger bilayer on the left relocated the anchoring point during heating to the left and caused the smaller bilayer to slide to the left, producing a net motion to the left. The stroke (net motion) of the robot was the same in magnitude but opposite in direction as the baseline case, and the friction forces acting on the larger and smaller bilayers and the linker were the same as for the baseline case. We observed the same behavior in our experiments when we turned our flexible linker robot around, thermally stimulated it, and observed it crawl in the opposite direction (**Movie S3)**. We elaborate more on these studies in the SI (**Fig. S14**).

Finally, we changed the design of the robot by replacing the pNIPAM linker with a stiffer passive hydrogel linker. We simulated the motion of a robot with a linker that was four times shorter and approximately 46 times stiffer during the cooling half-cycle (**Figure 4d**). The bending of the bilayers during cooling lifted the linker from the contact surface, creating an asymmetric arch. We observed that the point of contact of the larger bilayer, point Y, was displaced by 2.1%. In contrast, the point of contact of the smaller bilayer, point X, was displaced by a substantially larger amount, 20.3%, in the opposite direction (**Figure 4e**). The normal force was larger at point Y for the larger bilayer than at the contact point X for the smaller bilayer. This asymmetry in contact forces caused point Y to act as an anchor, and the robot translated to the right during the cooling half-cycle (**Figure 4f**). The trend observed in our simulations that a robot with a stiffer linker shows higher unidirectional displacement agrees with the experimental observations (**Figure 3**).

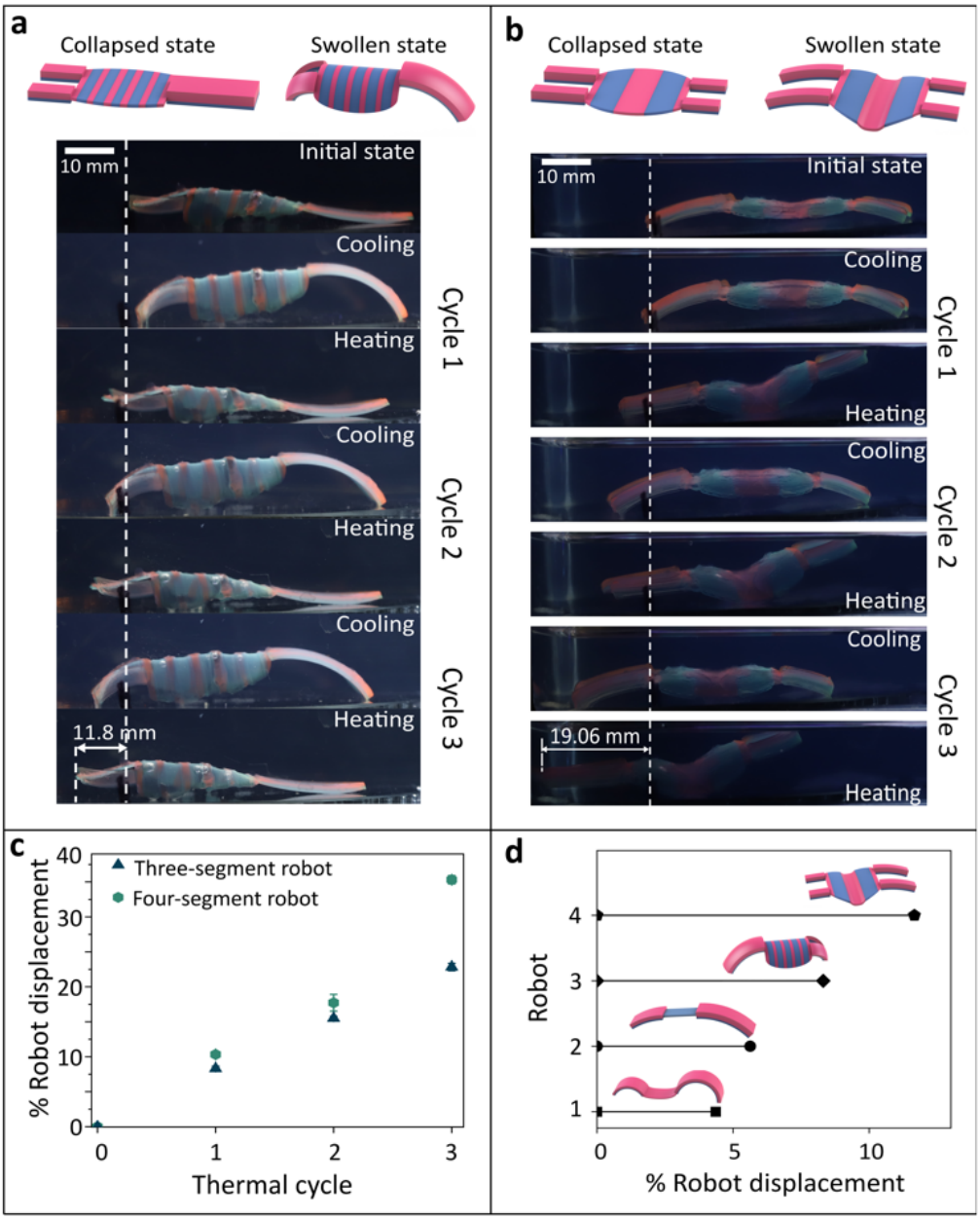

Figure 5. Displacement of gel robots with varying bilayer segments and linker morphology. (a) Experimental images of a three-segment robot at the end of each heating and cooling half-cycle over three consecutive cycles. **(b)** Experimental images of a four-segment robot at the end of each heating and cooling half-cycle over three consecutive cycles. **(c)** Plot of the percentage robot displacement of the three-segment and four-segment robots over three thermal cycles. The error bars indicate the standard deviation with a sample size of three. The three-segment robot moved 7.63% body length/cycle, whereas the four-segment robot moved 12.2% body length/cycle. **(d)** Comparison of the average percentage displacement of all the experimentally demonstrated robots.

Alternating linker patterns and adding bilayer segments increases displacement

We studied the dependence of the number of bilayer segments and the linker pattern on the morphology and displacement of gel crawlers. We hypothesized that these factors would induce a larger asymmetry between the bilayer segments, increasing the displacement of the robot. We varied the number of bilayer segments of the robots from two to four (**Figure 5a, b; Fig. S5,6**). Taking inspiration from an accordion, we also redesigned the linker from the fully pNIPAM or pAAM composition (**Figure 3)** to a combination of alternating pAAM and pNIPAM strips, as shown in **Figure 5**. We hypothesized that the alternating geometry would allow the linker to elongate while maintaining its shape. We stimulated all the robots across multiple thermal cycles, and the results are discussed below.

First, we 3D printed a three-segment robot with an alternating linker (**Fig. S5a)**. This gel robot consisted of two small and one large bilayer segment connected by a linker composed of five stripes of pAAM and six stripes of pNIPAM. During the cooling half-cycle, the linker elongated and had minimal anchoring to the contact surface, effectively behaving like a stiff linker (**Figure 5a**). During the heating half-cycle, an asymmetry was induced both by the morphological differences in the bilayer curvature and the uneven distribution of the two small bilayer segments. We observed a stick-slip motion and a large displacement of 7.63% body length/cycle **(Movie S4)**. Also, the robot moved unidirectionally towards the two smaller bilayers due to the increase in contact area on the side of the smaller bilayers. We also performed a 20-thermal cycle study on this gel robot to confirm consistent unidirectional motion (**Fig. S5b**). We also compared its displacement to a similar two small and one large bilayer segment robot but with a flexible pNIPAM linker (**Fig. S6**). Since this linker sags and has high contact area with the underlying surface during the cooling half-cycle, the displacement is reduced to 3.9% body length/cycle (**Movie S5**).

Next, we 3D printed a four-segment robot with an alternating linker (**Fig. S5c)**. This gel robot consisted of two small and two large bilayer segments connected by a linker composed of two stripes of pAAM and three stripes of pNIPAM. During the cooling half-cycle, the linker again elongated and had minimal anchoring to the contact surface, also effectively behaving like a stiff linker. Since the contact area was greater on the side of the two larger bilayers, the robot consistently moved in that direction and displacement was high, measured at 12.2% body length/cycle (**Figure 5c-d, Fig. S5d**, **Movie S6)**. These studies confirm

that the manipulation of nomal force asymmetry can be manipulated by varying the morphology and relative contact area of the bilayers and linker.

DISCUSSION

We have described and validated a mechanism for untethered unidirectional crawling of multisegmented gel robots on flat unpatterned substrates. The mechanism relies on a spatial morphological gradient along the fore-aft axis, which in turn leads to an asymmetry in contact forces of gel segments and linkers, leading to unidirectional locomotion. Using thermal cycling of a model gel robot consisting of two bilayer segments with either a stiff or flexible linker, we characterize the mechanism experimentally and compare the results to FEA simulations with good agreement. We observed consistently that the magnitude and asymmetry of contact forces between different parts of the robot and the underlying surface are important factors that determine the extent of unidirectional displacement. Also, these factors can be tuned by varying the dimensions and number of the small and large bilayers. We observe an increase in the displacement on increasing the number of bilayers, and robots move consistently in the direction of the bilayers with the larger contact forces.

We have also observed that the robot displacement can be controlled by varying the linker stiffness. Stiff or elongating linkers have lower contact with the underlying surface and consequently anchor less, resulting in greater contact force asymmetry between the bilayer segments and leading to larger displacement.

In terms of speed of locomotion, the time scales of swelling of gel robots are limited by diffusion and are generally slow as compared to, for example, piezoelectric or electromagnetic actuators. But they have advantages, including being powered and operating in aqueous environments without the need for tethers or wires, which is important for locomotion in small and tortuous spaces. We also note that many soft-bodied organisms, such as worms, move slowly. Moreover, many dynamic processes in nature, such as growth, morphogenesis, disease progression, digestion, and decay, are slow processes.

While we heated and cooled the entire setup in our study, the robots have the potential to function autonomously in environments where the temperature oscillates or varies. Also, in the future, it is conceivable that localized heating or cooling modalities could be integrated within the robot body to facilitate programmable or autonomous operation in small aqueous spaces independent of the temperature of the environment. Of note, there are several battery-operated resistive heaters and Peltier elements as well as magnetic, acoustic, or optically excitable particles that could potentially be incorporated within the gels. In terms of the operating temperature range, we note that the LCST in pNIPAM and other gels, such as blends of biocompatible poly(oligo(ethylene glycol) methyl ether methacrylate (pOEGMA) / di(ethylene glycol) methyl ether methacrylate (pDEGMA), are widely tunable across the biological range of temperatures. (*13*)

Also, in addition to thermally responsive hydrogels, the manipulation of morphology and contact force asymmetry to tune distributed spatiotemporal dynamics of soft robots, as demonstrated in this work, could potentially be utilized with other stimuli responsive materials. For example, hydrogels can swell or deswell in response to multiple stimuli, including light, pH, or DNA hairpins, and locomotion in response to these stimuli can enhance autonomy, programmability, and applicability. (*3, 42*)

MATERIALS AND METHODS

Composition of the hydrogel inks

We formulated shear-thinning hydrogel inks compatible with the DIW printing process mentioned in **Figure 1a** by modifying the rheological properties of aqueous mixtures of monomers, a crosslinker, and a photoinitiator. (*10*) We used N-isopropylacrylamide (NIPAM) for the active ink and acrylamide (AAM) monomer for the passive ink. The thermoresponsive pNIPAM undergoes a coil-to-globule transition above its LCST, which was measured to be 35.7 ºC (**Fig. S15**), while pAAM shows no thermoresponsivity. We used Laponite nano clay as the rheology modifier, Irgacure 2959 as the photoinitiator, and N, N′-methylenebisacrylamide (BIS) as a crosslinker (used only for the passive AAM ink). We also added two dyes to the mixtures to increase contrast during imaging: a fluorescent rhodamine dye (emission 568 nm) in the active ink and methylene blue in the passive ink.

Preparation of the hydrogel inks

NIPAM active ink: We blended Laponite XLG® (BYK USA Inc.) with pure deionized (DI) water, and a 0.12 mg/mL aqueous dye solution of methacryloxyethyl thiocarbamoyl rhodamine B (Polysciences, Inc.). We homogenized the mixture using a planetary mixer (Mazerustar KK-250S, Kurabo Industry Ltd.) for approximately 90 seconds at 2100 rpm. We then added the NIPAM monomer (Scientific Polymer Irgacure-Products Inc.) and the UV photoinitiator 2-Hydroxy-4´-(2-hydroxyethoxy)-2-methylpropiophenone or Irgacure 2959 (BASF). We homogenized the solution again for approximately 150 seconds at 2500 rpm. The relative final concentration of the active ink in (w/w) was: 76% pure DI water; 8.5% NIPAM; 8.5% aqueous rhodamine dye solution; 6.8% Laponite; 0.2 % Irgacure.

AAM passive Ink: Like the active ink, we again blended Laponite with pure DI water and a 0.5 mg/mL aqueous dye solution of methylene blue (Sigma Aldrich). We homogenized the mixture for approximately 90 seconds at 2100 rpm. We then added AAM monomer (Sigma-Aldrich), Irgacure 2959, and the crosslinker N, N′-methylenebisacrylamide (BIS, Sigma-Aldrich). We homogenized the solution again for approximately 150 seconds at 2500 rpm. The relative final concentration of the passive ink in (w/w) was: 74.7 % DI water; 8.3% aqueous methlylene blue dye solution; 7.9% AAM; 6.6% Laponite; 1.7% BIS; 0.8 % Irgacure.

Sacrificial Ink: We blended Laponite with pure DI water to make the sacrificial ink to print the supporting layer for the suspended linker of the robot. We homogenized the mixture for approximately 90 seconds at 2100 rpm. The relative final concentration of the sacrificial ink in (w/w) was: 92.6 pure DI water; 7.4 % Laponite.

After ink homogenization, we kept the inks at room temperature to age for at least a day prior to use to achieve the desired rheological properties. The ink schematics and additional details are in the Supplementary Materials.

3D printing and robot curing

We first 3D printed the bilayers or robots with the inks and later cured them after printing using UV light. For 3D printing, we generated CAD and STL files using Solidworks (Dassault Systèmes) and used the Slic3r software to generate our G-code files. We used a pneumatic extrusion-based printer (Inkredible+ 3-D Bioprinter, Cellink) to print our bilayers and robots. After calibrating the XYZ axes, we manually estimated the nozzle offset of the two printer heads by

printing a calibration structure of two superimposed cylinders also incorporating it into the G-code. Since the robots were printed with three materials (NIPAM, AAM, and sacrificial ink) using a two-nozzle printer, it was necessary to modify the G-code to control the sequence in which the different segments were printed. This sequential printing allowed us to exchange the three inks in the two nozzles during the printing process.

After 3D printing the entire bilayers or robots, we cured them using two UV LED probes (365 nm wavelength, 12 mm lens diameter) and the OmniCure UV (LX 500, Lumen Dynamics) system at 100% exposure intensity for 200 seconds. Since, the robots were wider than the lens diameter, we cured them in sections to achieve a relatively uniform curing. More details on the printer specifications and fabrication setup are included in the Supplementary Materials.

Study design for experimental characterization of the bilayers and robots

We designed our study to systematically investigate the effect of tunable engineering parameters such as bilayer thickness ratio, number of bilayers, and linker pattern on bilayer curvature and robot displacement. We carried out thermal cycling experiments using bilayers and robots and conducted all experiments on silicon wafers in water in the same actuation setup. The goal of each time-lapse experiment was to understand and quantify the behavior of the bilayers and the robot over thermal cycling to tune parameters for the model and establish a comparison between experiments and simulations. We have included more details of the thermal actuation of bilayers and robots below and in the Supplementary Materials.

We did robot characterization experiments over multiple (3, 5, and 20) thermal cycles, beginning with the cooling half-cycle and ending with the heating half-cycle. We performed all experiments in triplicate, and we used the data to generate error margins, as shown and mentioned in each figure legend. We used a combination of MATLAB and ImageJ to estimate parameters like the radius of curvature and robot displacement. We generated timelapse videos of the robot actuation using the Windows Movie Maker (Microsoft). We have included additional details regarding imaging and time-lapse experiments in the Supplementary Materials.

Thermal actuation of bilayers

After printing the bilayers and curing them, we placed three samples of each bilayer design in a 20 cm × 10 cm clear plastic box. We added 750 mL of DI water at room temperature to the box and used a broad-spectrum UV light source to excite the Rhodamine dye in pNIPAM and image the bilayers with a DSLR camera (Canon). After the bilayers equilibrated at 25 °C, we thermally cycled the bilayers in water on a hot plate (Thermo Fisher Scientific) by switching the temperature to 60 °C at 1.75 °C/min for the heating half-cycle and back to 25 °C at 0.8°C/min for the cooling half-cycle. We took images of the swelling and deswelling of the bilayers over 8 hours in 5-minute intervals.

Thermal actuation of robots

We used the same experimental setup and conditions used for the bilayers to characterize the thermal actuation of the gel robots. We generated all the figures in the main text using three or five thermal cycles. For the statistical study of robot displacement shown in **Fig. S4-6**, we stimulated the robots over ten thermal cycles, turned them around, and stimulated them for ten additional thermal cycles.

Finite element analysis

We used finite element analysis to investigate the crawling mechanism of the two-segment robot with a flexible linker. We applied the thermo-chemo-mechanical model of Chester et al. (*40*) to describe the coupled stress response and swelling behavior of the pNIPAM and pAAM hydrogels. The model was implemented as a user element subroutine (UEL) in ABAQUS. (*43*) A detailed description of the thermo-chemo-mechanical model and its finite element implementation can be found in the Supplementary Materials.

Briefly, we assumed in the model that the free energy density of the hydrogel can be decomposed additively into a mechanical part for the entropic behavior of the polymer network and a chemical part for the mixing of the solvent and polymer network. We used a compressible Neo-Hookean potential to describe the mechanical part, and the Flory-Huggins potential to describe the chemical part. (*44*) To model the LCST, we assumed that the Flory-Huggins parameter increased with temperature from a low value for the swollen hydrophilic state to a high value for the deswollen state according to a sigmoidal function. The stress response and chemical potential for the solvent are defined as the partial derivative of the free energy density with respect to the deformation and solvent concentration. We assumed a linear relationship between the solvent flux and the gradient of chemical potential to describe the stress-coupled solvent transport into the hydrogel. The stress response was determined from the mechanical equilibrium equation. The model was implemented into ABAQUS and applied to solve for the swelling and stress response of the hydrogel bilayers and robots.

The model contains ten parameters: the shear modulus, the bulk modulus, the Flory-Huggins interaction parameters at a high and a low temperature, the LCST transition temperatures, the temperature range of the LCST transition, the swelling and deswelling diffusivities, the atomic volume of the solvent, and the reference chemical potential of the solvent. We obtained the values for the shear modulus of the hydrogels from dynamic mechanical characterizations performed in a previous study. (*10*) We simulated the time-dependent swelling-induced bending of the bilayers with different thicknesses ratios to calibrate the Flory-Huggins interaction parameters at the low and the high temperature, the transition temperature, the temperature range of the transition, and the swelling deswelling diffusivity (**Table S2**) as described in the Supplementary Materials.

We used the obtained model parameters (**Table S4**) to simulate the two-segment robot with either a flexible linker or a short stiff linker lying on a frictional rigid surface as described in the Supplementary Materials. The simulations also included the effects gravity and buoyancy on the robot. We applied a temperature profile like the experiment as shown in **Fig. S10**. The simulations calculated the displacements at the centroid of the linker and bilayers and the normal contact force for each specimen.

Supplementary materials and methods

Fig. S1. 3D printing process and ink composition.
Fig. S2. Bilayer timelapse study.
Fig. S3. Swelling study of the two-segment robot with a flexible linker.
Fig. S4. Two-segment robot with varying linker stiffness.
Fig. S5. Varying linker morphology and bilayer segments.
Fig. S6. Three-segment robot with a flexible linker.
Fig. S7. Details of the finite element model of the pNIPAM-pAAM bilayer.
Fig. S8. Temperature profile applied to the pNIPAM-pAAM bilayers.
Fig. S9. Meshing of the two-segment robot with a flexible linker.
Fig. S10. Temperature profile applied to the two-segment robot with a flexible linker.
Fig. S11. Boundary conditions, body force, and contact conditions applied to the two-segment robot with a flexible linker.
Fig. S12. Effect of the added spring at the center of the linker.
Fig S13. Effect of Coulomb friction on the locomotion.
Fig. S14. Displacement and normal force of the two-segment robot with interchanged bilayers and a flexible linker.
Fig. S15. DSC study of pNIPAM-Laponite hydrogel ink.
Table S1. The experimentally measured material properties used for finite element simulations of pNIPAM-pAAM bilayers.
Table S2. The material parameters obtained from fitting to the bilayer swelling experiments
Table S3. Swelling and deswelling strain of the pNIPAM hydrogels from the cured state
Table S4. The material parameters used in the simulation of the two-segment robot
Movie S1. Experimental video of the crawling of the two-segment robot with a stiff linker over three thermal cycles.
Movie S2. Simulation of the two-segment robot crawling with a flexible linker over one thermal cycle.
Movie S3. Simulation of the two-segment robot crawling with a flexible linker and interchanged bilayers over one thermal cycle.
Movie S4. Experimental video of the crawling of the three-segment robot with an alternating linker over three thermal cycles.
Movie S5. Experimental video of the crawling of the three-segment robot with a flexible linker over three thermal cycles.
Movie S6. Experimental video of the crawling of the four-segment robot with an alternating linker over three thermal cycles.

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