Programmable Self-Locking Micromachines with Tunable Couplings

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Intelligent micromachines that perform tasks in complex microenvironments by adjusting structural configurations and physical properties offer substantial potential for use in many fields, including precision medicine, microfluidic channels, micromanipulations, and bioengineering. Recent progress in smart materials and 4D microprinting provides a feasible strategy for fabricating compound micromachines by designing and assembling predictable 3D-to-3D shape-morphing components. However, shape transformations of these components can only be triggered simultaneously by external stimuli, and orderly triggering among them cannot be achieved yet for an intended purpose of performing more efficient actions. Herein, a novel sequentially triggered strategy is proposed for developing intelligent micromachines that consist of multiple reconfigurable components that work interactively with each other. Furthermore, self-locking micromachines with tunable coupling mechanisms that coordinate accurate time-sequence deformations among components are achieved. Three types of locking couplings are modulated based on finite-element analysis (FEA)-aided design. The proposed method provides an effective strategy to create next-generation intelligent micromachines that can change their shapes to adapt to complex situations and complete multiple tasks.

Intelligent machines can perform multitarget or multistep tasks by adjusting their structural configurations\textsuperscript{[1,2]} and physical properties\textsuperscript{[3]} autonomously in response to external stimuli and, thus, have potential applications in various fields, including soft robotics\textsuperscript{[2,4,5]} medical treatments,\textsuperscript{[6]} and smart electronics.\textsuperscript{[7]} The realization of complex functions in such machines requires precise and sequential deformation coordination among multiple components. However, the difficulty of deformation coordination increases, as the size decreases to the microscale. Currently, advanced 4D microprinting techniques\textsuperscript{[6–11]} have been used to directly write multifunctional, 3D-to-3D shape-morphing, compound micromachines with dozens of micrometer-range feature sizes in a single-material-single-step mode. However, these micromachines can only carry out simple and single deformations, such as overall swelling, shrinking, bending, and folding, which impede the realization of further applications. Furthermore, programmable modular design strategies based on the assembly of 4D microbuilding blocks have been proposed in recent years\textsuperscript{[12–14]} to customize complex 3D shape-changeable micromachines, and these engineered micromachines typically consist of a large number of basic shape-morphing blocks that can achieve large, precise, and predictable deformations, yet that function independently of each other without effective deformation couplings among individuals.

An intelligent micromachine should be a complex, orderly, and coordinated system, rather than merely a collection of functional components. The realization of complex functions requires controllable shape-morphing coordination through sequential and cooperative triggering among the multiple components of the machine.\textsuperscript{[15,16]} Typically, the self-locking mechanism\textsuperscript{[17]} is one of the key functionalities of intelligent machines. This mechanism can generate a controllable, steady, and firm locking configuration by precise coordination couplings between the lock lasso and the lock core. However, at the microscale, it is still quite challenging to achieve independent and sequential coordination operation in different regions of a tiny machine under common actuation fields, such as the chemical field,\textsuperscript{[18,19]} magnetic field,\textsuperscript{[20,21]} heat,\textsuperscript{[22,23]} or ultrasound.\textsuperscript{[24]} This makes it difficult to achieve complex functions such as self-locking. Therefore, one of the crucial steps toward intelligent micromachines is to develop an effective microscale actuation strategy for sequential and coordinated couplings of different microparts.

In this work, we propose a novel shape-morphing strategy for the construction of sequentially triggered micromachines based
on bilayered microparts. Using this strategy, a self-locking micromachine shaped like a plastic tie strap can be designed and fabricated with a 4D direct laser writing (4D DLW) technique. The micromachine can form into a loop to provide a steady and firm locking connection under accurate deformation coordination in timed sequences among the different components. Thus, the method achieves effective interactions among the different components of a microindividual. Moreover, with finite-element analysis (FEA)-aided design, three locking levels with different degrees of locking couplings can be modulated by changing the exposure dosage of femtosecond laser pulses of DLW. Self-locking function of micromachines with tunable couplings offers various potential applications at the macroscale, such as cell grasping and binding. From a broader perspective, this strategy of deformation coordination control will facilitate the development of next-generation intelligent micromachines.

We developed a pH-sensitive gel material that was suitable for fabricating smart micromachines based on 3D DLW. The acidic environment can inhibit carboxyl dissociation in the gel networks, while the alkaline solution promotes it. This causes osmotic pressure differences between the gel networks and external solution to drive directional migration of water molecules and, thus, induce shrinking or swelling. Our previous work\textsuperscript{[25]} revealed the mechanism that the critical pH triggered gel swelling is directly related to the degrees of the gel-network polymerization, which can be regulated by the fabrication laser power. Therefore, when the external pH changes from acid to alkal, a bilayer microbeam (\(\approx 100 \mu m\) length) consisting of a high-laser-power (HLP) layer and a low-laser-power (LLP) layer shows a layer-by-layer sequential swelling effect due to the time difference \(\Delta t\) between the initial swelling moments of two monolayers, resulting in the time-related nonmonotonic bending deformations. Because of the nonnegligible \(\Delta t\), the layer-by-layer sequential swelling effect can be used to drive the dynamic shape-morphing process of microstructures.

In mechanics, the swelling kinetics of a monolayer beam can be quantitatively characterized by the logistic sigmoidal equation\textsuperscript{[26]}

\[
\varepsilon(t) = \frac{l(t) - l_{\text{design}}}{l_{\text{design}}} = \varepsilon_{\infty} + \frac{\varepsilon_0 - \varepsilon_{\infty}}{1 + (t/t_0)^q}
\]

(1)

where \(\varepsilon(t)\) and \(l(t)\) are the strain and length at time \(t\), respectively. \(l_{\text{design}}\) is the design length for the fabrication. \(\varepsilon_0\) and \(\varepsilon_{\infty}\) represent the initial and final strains, respectively. \(t_0\) is the characteristic time depending on the material properties, and \(q\) is the swelling index.\textsuperscript{[25]} The curve in Figure 1a shows the swelling kinetics of monolayer beams in HLP and LLP during the ambient pH changing from acid to alkal, which illustrates the definition of \(\Delta t\).

Because of the time-related sequential deformation, multilayered microstructures consisting of several monolayers with shape-morphing time difference can exhibit diverse shape-morphing phases. These can be used as shape-morphing components to design the structure of smart micromachines. In the following, we discuss the sequentially triggered deformation characteristics of two basic geometric configurations (i.e., the microbilayer and the microjoint) during dynamic changes in the ambient environment. The microbilayer is composed of an HLP layer and an LLP layer (Figure 1b). The bending deformation driven by the changes in external pH is quantified by Timoshenko’s beam theory.\textsuperscript{[27]} The curvature of bending \(\kappa_B\) shown in Figure 1b is described as follows.

\[
\kappa_B = \frac{6(\varepsilon_{\text{high}} - \varepsilon_{\text{low}})(1 + m)^2}{[3(1 + m)^2 + (1 + mn)(m^2 + m n^{-1})](a_{\text{low}} + a_{\text{high}})}
\]

(2)

where \(m = a_{\text{low}}/a_{\text{high}}\) and \(n = E_{\text{low}}/E_{\text{high}}\) are the ratio of thicknesses and Young’s moduli between two layers, respectively, and \(\varepsilon_{\text{high}}\) and \(\varepsilon_{\text{low}}\) represent the strains of two layers. The subscripts “low” and “high” denote LLP and HLP, respectively. The bending distance \(d_B\) is defined as the length between two ends of the midplane, given by

\[
d_B = \frac{1}{\kappa_B} \sqrt{2(1 - \cos \theta_B)}
\]

(3)

where \(\theta_B\) is the angle corresponding to the bending arc (see Figure 1b and the Supporting Information). The sequential swelling induces three dynamic bending states: 1) the initial state (in acid solution), in which the microbilayer entirely shrinks and shows slight bending; 2) the enhanced state, in which \(\kappa_B\) increases to the maximum and \(d_B\) to the minimum, owing to the sequential swelling of two layers in response to the change in external pH from acid to alkal; and 3) the final state, in which the microbilayer unbends to a flat beam and the surrounding solution becomes alkal.

The folding properties of microjoints are described by the folding angle \(\theta_F\), which is the complementary angle of \(\theta_B\), i.e., \(\theta_F = \pi - \theta_B\), and the folding distance \(d_F\), which can be deduced as

\[
d_F = (\varepsilon_{\text{high}} + 1)l_0 \sqrt{\frac{1 + \cos \theta_B}{2}}
\]

(4)

where \(l_0\) is the initial length of the beam. For simplicity, the bending part is regarded as a zero-length point. Three dynamic bending states also occur in the microjoints, i.e., the initial, enhanced, and final states, as shown in Figure 1c.

Utilizing the sequential deformation characteristics of the basic geometric configurations discussed earlier, we designed and fabricated a micromachine shaped-like plastic tie strap with \(\approx 100 \mu m\) length, \(\approx 20 \mu m\) width, and \(\approx 5 \mu m\) height, as shown in Figure 2a (see detailed size in Figure S6, Supporting Information). It consists of a bendable microbilayer body with two microjoints at both ends, a head with two foldable microjoints, and a homogeneous lasso-shaped tail. Figure 2bi–viii shows the entire shape-morphing cycle of this micromachine. After fabricating, the micromachine was transferred to an acidic solution where it shrunk to an arc shape (step i). Then, an alkaline solution was injected slowly to trigger a series of sequential deformations at the double-layer parts of the body and the head. If the transverse size of the head was smaller than the inner size of the tail when the tip of the head reached the tail (step ii) during the enhanced bending process, the head came through the lasso of the tail (step iii). Then, the body began to unbend while the head unfolded. Because the transverse size of the head became larger than the inner size of the tail, the tail was hooked by the head, indicating a locking state (step iv). When the solution was changed back to the acidic environment, the micromachine
returned to a smaller size, but remained locked (step v), because the head was still larger than the tail. As the solution becomes more alkaline, finally, the sequential deformation happened again at the double layer of the head and the body. The transverse size of the head became smaller than the tail during the enhanced process, leading to an unlocking transformation of the micromachine (steps vi–viii).

In general, the locking and unlocking process of the micromachines can be expressed by the inserting degree and inserting ability versus stimulus time. The inserting degree represents the relative position of the body along with the head and the tail lasso, which is expressed quantitatively as \( \frac{l_{H-T}}{d_{body}} \), i.e., the distance between the tip of the head and the tail \( l_{H-T} \) (when the head is in the tail, \( l_{H-T} \geq 0 \), and conversely, \( l_{H-T} < 0 \)) divided by the diameter of the bending body \( d_{body} \). Utilizing the geometrical relationships, both \( l_{H-T} \) and \( d_{body} \) can be expressed by the curvature of bending \( \kappa_{b-b} \), the bending distance \( d_{b-b} \), and the folding angle \( \theta_{F-b} \) given in Figure 1. Therefore, the inserting degree yields

\[
\frac{l_{H-T}}{d_{body}} = \frac{\frac{d_{b-b}}{\kappa_{b-b}}}{2 \sin \left( \frac{\theta_{F-b}}{2} \right)} - \frac{\pi}{\kappa_{b-b}} = \frac{\kappa_{b-b}}{2} \left[ L_{H} - \frac{d_{b-b}}{2} \csc \left( \frac{\theta_{F-b}}{2} \right) \frac{1}{2} \right]
\]

where \( L_{H} \) is the total length of the head. Figure 2c presents the profile of the inserting degree in the locking process. Four important points are highlighted. \( H_{10} \) is the initial state before locking, \( H_{12} \) is the moment that the tip of the head is just at the lasso of the tail \( \left( l_{H-T} = 0 \right) \), \( H_{13} \) is when the head reaches the deepest inserting distance, and \( H_{13} \) denotes the end of the deformation. First, the head is out of the arrow. Therefore, the inserting degree is a negative value. When the external pH changes from acid to alkali, the body experiences enhanced bending caused by the sequential deformation, such that the tip of the head approaches and enters the tail. Therefore, the inserting degree graphically increases from negative to positive through the point \( H_{12} \) and reaches its maximum at \( H_{13} \). Subsequently, the body begins
to unbend, resulting in a downward trend of the curve. However, because the transverse size of the head is still larger than the inner size of the tail, the head is not able to escape the tail, and the inserting degree remains positive at $H_{L3}$.

The unlocking process also has four critical points (Figure 2d). $H_{U0}$ is the initial state before unlocking, $H_{U1}$ is when the head reaches the deepest inserting distance, $H_{U2}$ is the moment that the tip of the head is just at the lasso of the tail, whereas the light-blue area represents the head is into the tail’s lasso. i–viii) in (c,d) denote the same states as those in (b). e) Trend of the inserting ability, i.e., $s_T/s_H$ with the time varying during the change in external solution from acid to alkali. The light-gray area denotes that the transverse size of the head is smaller than the tail’s, whereas the dark-gray area denotes that the head is larger than the tail.

Figure 2. Shape-changing process of the self-locking micromachine. a) Optical microscopic images of the micromachine in unlocking and locking states. Scale bar: 20 μm. b) Complete shape-morphing cycle. The gold arrow denotes the external solution changing from acid to alkali, whereas the gray one means the transition from alkali to acid. The colors refer to different fabrication parameters, as shown in the color bar. c) Profile of the inserting degree $l_{H+T}/d_{body}$ changing during the locking process. d) Profile of the inserting degree during the unlocking process. The sky-blue area in the background indicates that the head is out of the lasso of the tail, whereas the light-blue area represents the head is into the tail’s lasso. i–viii) in (c,d) denote the same states as those in (b). e) Trend of the inserting ability, i.e., $s_T/s_H$ with the time varying during the change in external solution from acid to alkali. The light-gray area denotes that the transverse size of the head is smaller than the tail’s, whereas the dark-gray area denotes that the head is larger than the tail.

The inserting degree starts with a positive inserting degree, because the head is fixed in the tail. When the external pH changes from acid to alkali, the body again undergoes enhanced bending. The inserting degree increases to its maximum and then decreases with the unbending of the body. The head escapes the lasso of the tail at $H_{U2}$. The inserting degree continues to decline until it reaches the value of $-1$, which means that the distance of the head’s tip out of the tail is equal to the diameter of the bending body. Finally, it increases toward zero, i.e., $l_{H+T}/d_{body}$ equals the flat length of the micromachine, while $d_{body}$ is positive infinity (the body unbends to a straight line).

The inserting ability is expressed as $\Delta s_T/s_H$, that is, the difference between the transverse size of the head $s_H$ and the inner size of the tail $s_T$, i.e., $\Delta s_T = s_T - s_H$, divided by $s_T$. Here, $s_H$ can be deduced as the function of the folding angle $\theta_F$ and folding distance $d_{F-h}$, as shown in Figure 1c:

$$s_H = d_{F-h} + s_{h-pillar} + s_{h-arrow} = l_{h-arrow} \sin \frac{\theta_F}{2}$$

$$+ s_{h-pillar} + s_{h-arrow}$$

where $s_{h-pillar}$, $s_{h-arrow}$, and $l_{h-arrow}$ are the pillar’s width, the arrow’s width, and the arrow’s length of the head, respectively.

When $\Delta s_T/s_T < 0$, the head is not able to enter or escape the tail, because it is too large. When $\Delta s_T/s_T \geq 0$, the head is
capable of entering or escaping the tail. Figure 2e shows the profile of inserting ability, where $T_1$ and $T_3$ indicate that the transverse size of the head is equal to the inner size of the tail, i.e., $\Delta s_{T-H} = 0$, and where $T_2$ indicates that the head is at its smallest size. The inserting ability increases from a negative to its maximum positive value $T_2$, with the critical point $T_1$ as the separation. Then, it decreases and tends to reach a steady negative value through the critical zero point $T_3$. This implies that the head is larger than the tail at first. When the sequential deformation happens, however, the head first folds to a smaller size and is able to enter or escape the tail and then unfolds back to its original size. Notably, the head may be fixed by the tail after unfolding if locking occurs.

Three tunable locking levels are proposed based on the configurations of the locking states. These intuitively exhibit the relative positions between the head and the tail (Movie S1, Supporting Information). The locking levels present the depth degree of the head entering the tail, which depends on the DLW parameters. Level-1 (Lv1) locking has the smallest inserting depth. Although the head is not completely in the lasso of the tail yet, locking can still be achieved owing to the elastic contact between the head and the lasso. In the Level-2 (Lv2) locking.
the whole head is inserted into the tail during sequential deformation, and it is hooked with the end of its tail. Level-3 (Lv3) locking has the deepest insertion depth. Hooking occurs based on the adhesion interaction between the bilayer and the inside part of the tail. Figure 3a–f shows the locking processes of the three levels with typical-moment experimental images and corresponding FEA results of the initial and final moments, with the corresponding profiles of the inserting degree and ability. Micromachines with different locking levels were fabricated under different laser powers in the body for tunable locking, but with the same laser powers in the head and tail. For convenience and accuracy, we captured the data of the inserting ability from the average of several micromachines and used it as a general profile for comparing the inserting degrees of the three locking levels. The shadow area is the locking phase (ranging from \( H_{11} \) to \( H_{11} \)). In Lv1, the locking phase happens at \( H_{11} \), which has already passed the maximum point \( T_2 \). It ends as soon as point \( H_{11} \) is reached—i.e., point \( H_{11} \) is located between point \( T_2 \) and \( T_3 \), leading to a short transforming period during which the head is larger than the tail before it is entirely inserted into the tail. The locking transformation happens when the head reaches its maximum folding point, \( T_2 \), in Lv2. Therefore, the whole head has enough time to insert into the tail and hook the tail by its end. Lv3 has the most time for the head to insert into the tail with the locking transforming starting point \( H_{11} \) located between \( T_1 \) and \( T_2 \). Consequently, the whole head and a piece of body enter the tail and are finally fixed by the adhesion of the body and the inside of the tail lasso.

In addition, the self-locking micromachines can realize unlocking transforming reversibly in the next external changes in ambient pH from acid to alkali. In general, the different locking levels of the micromachine have similar unlocking transforming processes. Therefore, we only show one case here. Five typical moments are marked, corresponding to the experiment images in Figure 3g: 1) the initial state of locking, in which the head is inserted and hooked to the tail; 2) the enhanced state, in which the body part bends into the maximum curvature and the head unfolds to a smaller size to escape the tail; 3) the moment when the head escapes from the tail; 4) the steady unbending period; and 5) the final state with the micromachine flattening back to its original configuration (Movie S2, Supporting Information). The analysis of the inserting degree and inserting ability profiles is also used for the unlocking process (Figure 3h). The varying trend of the inserting ability describes whether and when the head can escape the tail, which can be seen as the same as the locking process. Nevertheless, the inserting degree shows an opposite trend in comparison with that of the locking, indicating that the unlocking motion in the micromachine is the inverse process of the locking.

In general, several conditions should be satisfied for locking and unlocking. 1) In the locking process, for the head to smoothly enter the tail, the transverse size of the head must be smaller than the inner size of the tail when the tip of the head reaches the tail; i.e., \( H_{11} \) must be located between \( T_1 \) and \( T_3 \). 2) For timely hooking, the head must unfold to a larger size than the tail to forbid the head to escape from the tail; i.e., at the time of \( T_3 \), the inserting degree must be a positive value. 3) To unlock—that is, for the head to escape the tail successfully—the head must be smaller than the tail; i.e., the inserting degree corresponding to \( T_3 \) must be negative.

In this work, we developed intelligent shape-morphing micromachines capable of achieving complex self-locking and unlocking behaviors by encoding their functional components. In combination with the FEA-aided design of structures and parameter optimization, we achieved a one-step self-locking micromachine under simple pH alternate stimuli, instead of complex operations. This one-step shape-morphing originated from a series of accurate linkage motions, including body bending, head folding, aiming at and inserting into the tail, and, eventually, head-tail fixing. Such motions required indispensable and correct selections of structural configurations, fabrication parameters, and actuation methods. Moreover, we proposed control strategies for the locking modes and showed three tunable locking coupling levels depending on the relative position of the head and tail, offering adjustable tightness to the micromachines. The self-locking micromachine realized accurate shape-morphing control at the microscale, with multiple parts, steps, and levels for the first time. It is advantageous in terms of ease of operation, fast response, good reversibility, and controllability. It should be pointed out that the sequentially pH triggered strategies have limitations, because it requires an external microenvironment with periodic changes in pH over a wide range. However, the design frameworks based on the sequentially triggered structural deformations offer inspirations for constructing smart microstructures in response to other external cues such as temperature and humidity. Taken together, this work provides a novel strategy for developing next-generation intelligent machines at the microscale.

Experimental Section

Preparations of Materials: The precursor to pH-responsive materials used in this work was a mixture reported by Jin et al. [9] which contained acrylic acid (AAc; Aladdin, China) and N-isopropyl acrylamide (NIPAAm; Aladdin, China) as the monomers, dipentaerythritol pentaacrylate (DPEPA; Aladdin, China) as the crosslinker, 4,4′-Bis(diethylamino)benzophenone (EMK; Red Chemical, China), N,N-dimethylformamide (DMF; Aladdin, China), and triethanolamine (TEOA; Aladdin, China) dissolved in ethyl lactate (EL; Aladdin, China) as the photoinitiator, and polyvinylpyrrolidone (PVP; Aladdin, China) as the additive.

Fabrication of Micromachines: The self-locking micromachines were fabricated using a DLP system (Photon Professional GT, Nanoscribe GmbH, Germany) with “Galvo” scanning mode and a 63 × oil-immersion objective (numerical aperture = 1.4). Sample preparation and the development procedure followed previous studies. [11,12] The laser powers varied in different parts of the microstructures, and the scanning speed was fixed at 5 mm s⁻¹.

Characterizations of Locking and Unlocking: After developing the micromachines, they underwent shrinking in a hydrochloric acidic solution with pH = 4.5 (hydrochloric acid, Aladdin, China). To increase the environmental pH, we slowly injected a sodium hydroxide solution with pH ≈ 12.5 (sodium hydroxide, Aladdin, China) using a syringe pump (LSP02-18, Longer Precision Pump, China) with the flow rate of 3.5 × 10⁻⁴ L s⁻¹. During the change in external pH, the micromachines underwent self-locking shape morphing. After that, the hydrochloric acidic solution was injected with the same flow rate to decrease the pH, whereas the micromachines remained in the locking state. Subsequently, the sodium hydroxide solution was added into the system once again and the micromachines unlocked. The entire shape-morphing process of the microstructures was observed and recorded using a digital microscope (RH-2000, Hirox, Japan).
Finite Elements Analysis: Commercial FEA software, ABAQUS (Dassault Systèmes S.A., France), was used to predict and validate the configurations of the micromachines in locking and unlocking states. The 3D models of the micromachines were meshed using the element type C3D8H (8-node linear brick, hybrid). Proper mesh sizes were chosen to ensure computational convergence and accuracy. The nonlinear mechanical response of the pH-responsive micromachines was described with a hyperelastic mechanical constitutive model with dimensionless crosslinking densities $N_v$ and Flory interaction parameters $x_i^{(2)}$ which depended on the material properties (Figure S4, Supporting Information). Then, the mechanical model was imported into ABAQUS with a user-defined subroutine, UHYPER, for simulations. Shape-morphing was triggered by the change in chemical potential $\mu$ defining $\mu_{\text{bulk}} = 0$ in all cases, and $\mu_{\text{bulk}}(<0)$ depending on the fabrication parameters (Figure S5, Supporting Information).

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

Acknowledgements
This work was supported by the National Natural Science Foundation of China (NSFC) under grants nos. 91848201, 11988102, 11521202, 11872004, and 11802004 and Beijing Natural Science Foundation under grant no. L172002.

Conflict of Interest
The authors declare no conflict of interest.

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

Keywords
intelligent micromachines, self-locking, sequential deformation, tunable couplings

Received: October 14, 2020
Revised: January 18, 2021
Published online:

[1] V. Cacucciolo, J. Shintake, Y. Kuwajima, S. Maeda, D. Floreano, H. Shea, Nature 2019, 572, 1.