

Self-rolling up micro 3D structures using temperature-responsive hydrogel sheet

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Abstract. This paper proposes a micro self-folding using a self-rolling up deformation. In the fabrication method at micro scale, self-folding is an especially useful method of easily fabricating complex three-dimensional (3D) structures from engineered two-dimensional (2D) sheets. However, most self-folded structures are limited to 3D structures with a hollow region. Therefore, we made 3D structures with a small hollow region by self-rolling up a 2D sheet consisting of SU-8 and a temperature-responsive hybrid hydrogel of poly(N-isopropylacrylamide-co-acrylic acid) (pNIPAM-AAc). The temperature-responsive hydrogel can provide repetitive deformation, which is a good feature for micro soft robots or actuators, using hydrogel shrinking and swelling. Our micro self-rolling up method is a self-folding method for a 3D structure performed by rolling up a 2D flat sheet, like making a croissant, through continuous self-folding. We used our method to fabricate 3D structures with a small hollow region, such as cylindrical, conical, and croissant-like ellipsoidal structures, and 3D structures with a hollow region, such as spiral shapes. All the structures showed repetitive deformation, forward rolling up in 20 °C cold water and backward rolling up in 40 °C hot water. The results demonstrate that self-rolling up deformation can be useful in the field of micro soft devices.

1. Introduction

Several fabrication strategies of three-dimensional (3D) structures at a small scale can be used, including self-assembly of small parts, 3D lithographic patterning, and self-folding [1, 2]. Folding is a useful method of easily fabricating complex 3D structures from engineered two-dimensional (2D) sheets [3]. Therefore, folding has been used in various fields before, such as in small robots [4–7], small 3D structures [8, 9], and medical stents and patches [10–12], at the centimeter or smaller scale. However, a self-folding method that enables automatic folding is necessary because manually folding a 2D sheet to a 3D structure is difficult, particularly at the millimeter or micrometer scale.

The self-folding methods at the centimeter to millimeter scale, for example, use a shape memory

polymer (SMP). Applying heat to an SMP (e.g., polystyrene, poly(vinyl chloride), and polyolefin film) to fabricate a structure leads to SMP shrinking and self-folding [12–17]. An SMP is useful for application in the centimeter to millimeter scale, but is difficult to apply to microscale structures because it is not suitable for microfabrication. On the other hand, the self-folding methods at the micrometer to nanometer scale use force to work strongly at the microscale. That is, surface tension at solder melting [18], magnetic forces [19], contractile forces (e.g., cell traction force) [20], strain caused by emitting ions [21], etc., are used. However, most self-folded microstructures are hollow structures [18–22]. In other words, the folded structure is not a solid 3D structure, but consists only of the surface of the 3D structure (for example, surface of the cubic structure). Few solid microstructures without a hollow region using self-folding have been developed before. In addition, in most of these self-folding methods, the folded shape is fixed and cannot be deployed after folding. Hence, it is not suitable in some applications like soft robots and actuators.

In this work, we propose a micro self-rolling up method as an advanced self-folding (figure 1). This self-rolling up method is a self-folding method like the making procedure of a croissant. That is, the 3D structure is developed by rolling up a 2D flat sheet through continuous self-folding (figure 1). Therefore, it is good at producing rotational symmetry 3D structures without a hollow region, such as cylindrical, conical, and ellipsoidal shapes. Our self-rolling up method can also produce hollow 3D structures (e.g., with a spiral shape). We used the deformation of a temperature-responsive hydrogel [23–29] for self-rolling up. A stimuli-responsive hydrogel, including the temperature-responsive hydrogel, has the advantage of repetitive deformation (i.e., repetition of folding and deployment) being possible after self-folding because it is caused by the swelling and shrinking of the hydrogel. In addition, it can produce two stable structures (i.e., forward rolling in cold water and backward rolling in hot water) using shrinking and swelling of hydrogel. Previous studies reported on self-folding and/or self-bending using a temperature- or pH-response hydrogel by utilizing the strain variation caused by a shrinking and/or swelling layer of the hydrogel and a rigid layer with no deformation [23–25]. A sheet with possible repetitive self-folding and deployment was developed using N-isopropylacrylamide (NIPAM) [26], a type of temperature-responsive hydrogel. Structures driven by a magnetic field, such as that in a swimming micromachine and micro gripper, were also developed by mixing magnetic particles into NIPAM [27–29]. However, most of self-folded structures using stimuli-responsive hydrogel are also limited to a 3D structure with a large hollow region. In this work, we fabricated complex 3D structures with a small hollow region (e.g., croissant-like ellipsoidal structure) using self-rolling up, which is the continuous self-folding of many adjacent hinges. By using an engineered 2D sheet, rotational symmetry 3D structures can be developed using the rolling up deformation of the temperature-responsive hydrogel. For example, a medical patch moving in the body and deploying at a targeted region can be developed by applying the self-rolling up method and magnetic manipulation [27–30]. The 3D structures with a small hollow region can produce a larger area despite their smaller volume. Therefore, the self-rolling up method can be useful in the field of micro soft devices.

2. General Principle and Method

2.1. Materials

Poly(vinyl alcohol) (PVA, Mw 9,000-10,000, 80% hydrolyzed) (360627), 1-butanol, N-isopropylacrylamide (NIPAM) (415324), poly(N-isopropylacrylamide) (pNIPAM) (535311), N,N'-methylenebis(acrylamide) (146072), and acrylic acid (147230) were purchased from Sigma-Aldrich. SU-8 3025 and SU-8 primer (OmniCoat) were purchased from MicroChem. Omnirad 2100 was provided by BASF. Poly(N-isopropylacrylamide-co-acrylic acid) (pNIPAM-AAc) was prepared by following a previously reported method [31].

2.2. General principle

The self-rolling up deformation of a hybrid hydrogel sheet was developed by a simultaneous or continuous self-folding at the hinges. Figure 2(a) shows the general principle of self-folding using the hybrid hydrogel sheet with a temperature-responsive hydrogel. The hybrid hydrogel sheet was composed of pNIPAM-AAc (dot patterned and blue region in figure 2) and SU-8 (white region in figure 2). pNIPAM-AAc is known as a stimuli-responsive hydrogel with responsiveness to temperature and/or pH. SU-8 is an epoxy resin without responsiveness to these stimuli. For example, in water under 30 °C, the pNIPAM-AAc layer swelled by absorbing water. The volume of the pNIPAM-AAc layer at the top surface increased because of the swelling. On the other hand, at the bottom surface, where pNIPAM-AAc and SU-8 made contact, the SU-8 plate prevented pNIPAM-AAc from swelling, and it cannot deform. As a result, a difference in the volume changes at the hinge part led to self-folding at the forward angle (figure 2(b)). The pNIPAM-AAc layer shrunk when the water temperature was increased. The shrinking of the hydrogel layer made the hybrid hydrogel sheet deploy to a flat state at approximately 30 °C. The shrinking was promoted when the water temperature was further increased to over 30 °C. The volume of the pNIPAM-AAc layer at the top surface was decreased by the shrinking, and the SU-8 plate also prevented the pNIPAM-AAc layer from shrinking at the bottom surface. The shrinking and the volume change difference led to the self-folding at the backward angle. In other words, the self-folding of the hybrid hydrogel sheet was caused by the volume change of the pNIPAM-AAc layer at the hinges.

Calculating the self-folding angle θ [deg] was possible considering the previous principle of self-folding. Figure 2(b) shows a cross section of the hybrid hydrogel sheet. The length of the SU-8 plate L [μm], thickness of pNIPAM-AAc layer at a state without swelling and shrinking t [μm], and coefficient of linear expansion of the pNIPAM-AAc layer α [-] were defined. The deformed length of the pNIPAM-AAc layer produced by swelling or shrinking was indicated as $\alpha L/2$ [μm]. The self-folding angle θ at one hinge part was geometrically calculated as follows in equation (1):

$$\theta = 2 \arctan \left(\frac{L \alpha}{2t(1 + \alpha)} \right). \quad (1)$$

2.3. Fabrication Process

Figure 3 shows a fabrication process of the hybrid hydrogel sheet. 1.5 mL of 20 wt.% PVA solution was

spin-coated on a 2 in. Si wafer at 500 rpm for 5 s and at 1000 rpm for 30 s. The wafer was then heated on a hotplate at 70 °C for 90 s and at 100 °C for 90 s to dry the PVA solution (figure 3(a)). Next, 1 mL of SU-8 primer was spin-coated on the Si wafer at 500 rpm for 5 s and at 3000 rpm for 30 s, then dried at 100 °C for 30 s and at 200 °C for 60 s to improve the adhesion between the PVA and SU-8 layers (figure 3(b)). The SU-8 layer with 30 μm thickness was coated and patterned by photolithography with 150 mJ/cm² of UV light on the thin film of the SU-8 primer (figure 3(c)). The UV light was provided using a high-pressure mercury lamp (360–370 nm in wavelength) through a dark field mask. Subsequently, plasma treatment using air was applied to the wafer at 18 W for 5 min (figure 3(d)). The plasma treatment removed the residual thin film of the SU-8 primer and improved the adhesion between the SU-8 and pNIPAM-AAc layers. Polyester spacers with 125 μm thickness were placed on the Si wafer after the plasma treatment, and 0.6 mL of the pNIPAM-AAc solution was immediately dropped on the SU-8 layer. A glass cover (40 × 50 mm) with a spin-coated PVA layer on one side was placed on the spacers such that it remained in contact with the PVA layer and the pNIPAM-AAc solution. Using the spacer can provide a uniform thickness of the pNIPAM-AAc layer. The pNIPAM-AAc solution was then exposed to 150 mJ/cm² of the UV light through a dark field mask to initiate polymerization. Acetone and isopropyl alcohol (IPA) were subsequently applied for developing and rinsed for 10 s each (figure 3(e)). The fabricated hybrid hydrogel sheet on the Si wafer was put into hot water at approximately 30 °C for the lift-off process, which dissolved the PVA layer on the Si wafer (figure 3(f)). The hybrid hydrogel sheet with residual stress and strain was a deployed state at approximately 30 °C. The lifting-off temperature was selected to be 30 °C to prevent peeling off the pNIPAM-AAc layer from the SU-8 layer. After that, the pNIPAM-AAc layer absorbed water and swelled, thereby leading to the self-folding of the hybrid hydrogel sheet (figure 3(g)).

The adhesion between the SU-8 and pNIPAM-AAc layers on the hybrid hydrogel sheet must be carefully treated. The plasma treatment enhanced the adhesion, but was not sufficient. Therefore, we made holes of 25 μm in diameter with 12.5 μm gaps from each other to the SU-8 layer. These holes on the SU-8 layer served as anchor holes to the pNIPAM-AAc layer making the pNIPAM-AAc layer hard to peel off from the SU-8 layer. The hole size was defined as smaller and easily made the pNIPAM-AAc solution get into the holes without a degassing treatment.

2.4. Basic characteristic of the self-folding angle

Controlling the self-folding angle was possible by changing the length of the SU-8 plate, L , considering equation (1). Hybrid hydrogel sheets with one hinge having different SU-8 plate lengths were fabricated. The self-folding angle was then measured using a microscope unit (VHS-2000 and VH-Z20W, KEYENCE). The self-folding angle was measured between the rigid SU-8 plates. SU-8 plates with 30 μm in thickness, 500 μm in width, and different lengths L (from 100 μm to 1000 μm) were fabricated. All the SU-8 plates were designed to have 15 μm interval of creases between each plate. A larger crease width got pNIPAM-AAc into the creases and prevented self-folding by swelling. The minimum width of the SU-8 layer patterning was 15 μm. A pNIPAM-AAc layer with approximately 95 μm in thickness was made on

the SU-8 layers. We used spacers with 125 μm in thicknesses for developing pNIPAM-AAc layers with 95 μm in thicknesses, because it was possible to get clear patterns using the thickness of spacers in the patterning process. The fabricated hybrid hydrogel sheets were put into cold water at 20 $^{\circ}\text{C}$ and hot water at 40 $^{\circ}\text{C}$. The self-folding angles were also measured. Figure 4(a) shows the fabricated hybrid hydrogel samples with 200 μm , 500 μm , and 1000 μm SU-8 plate lengths. Figure 4(b) shows the measurement result of the self-folding angles. The vertical axis in figure 4(b) represents the self-folding angle θ , while the horizontal axis represents the SU-8 length L . The theoretical curves in figure 4(b) were obtained from equation (1). The linear coefficients of expansion α and the approximate pNIPAM-AAc thicknesses were $\alpha = 0$, $t = 95 \mu\text{m}$ at initial state, $\alpha_{\text{Cold}} = 0.23$, $t_{\text{Cold}} = t(1 + \alpha_{\text{Cold}}) = 117 \mu\text{m}$ at 20 $^{\circ}\text{C}$ cold water and $\alpha_{\text{Hot}} = -0.25$, $t_{\text{Hot}} = t(1 + \alpha_{\text{Hot}}) = 71 \mu\text{m}$ at 40 $^{\circ}\text{C}$ hot water, respectively. The coefficient of linear expansion was calculated by measuring the length of the pNIPAM-AAc sheet in 20 $^{\circ}\text{C}$ and 40 $^{\circ}\text{C}$ water. The pNIPAM-AAc thickness was calculated by multiplying the initial thickness of pNIPAM-AAc ($t = 95 \mu\text{m}$) and the coefficient of linear expansion. The theoretical and measured values at an SU-8 plate length ranging from 100 μm to 300 μm were approximately similar. The measured values deviated from the theoretical values when the SU-8 plate length exceeded 300 μm and the self-folding angle converged in both 20 $^{\circ}\text{C}$ and 40 $^{\circ}\text{C}$ water. This deviation might result from an obstruction of the deformation in the pNIPAM-AAc layer caused by the SU-8 layer. An SU-8 plate length of over 300 μm clearly produced a larger volume change of the pNIPAM-AAc layer compared with that under 300 μm . However, the contact of the SU-8 and pNIPAM-AAc layers prevented the deformation of the pNIPAM-AAc layer. Hence, we considered that the amount of the pNIPAM-AAc deformation converged as the SU-8 plate length increased. In addition, the error bars of the self-folding angle at 40 $^{\circ}\text{C}$ hot water were higher than those at 20 $^{\circ}\text{C}$, indicating that pNIPAM-AAc shrinking provided a poorer uniformity of deformation compared to pNIPAM-AAc swelling. Although there was the deviation between the measured and theoretical value, the geometrical modelling was beneficial for considering the trend of the self-folding angle in figure 4(b).

Hybrid hydrogel sheets with multiple SU-8 plates were fabricated using the same process shown in figure 3. A basic cylindrical structure was also assembled using the self-rolling up deformation (figure 5). Figures 5(a)–(c) show the SU-8 plate with 500 μm in width and 200, 500, and 1000 μm in lengths, respectively. Each plate length can produce different self-folding angles; thus, these three rolling structures had different diameters. The diameter of the self-rolling up structure increased as the SU-8 length increased. In addition, the same length of SU-8 plates produced a similar self-folding angle. Hence, the plates over one circle collided with each other, and the structures had no gap. Figure 5(d) shows the structure consisting of the SU-8 plates of different lengths. The 200, 400, 600, 800, and 1000 μm SU-8 plates were connected to each other. The self-folding angles became larger as the length of the SU-8 plates became longer. Hence, providing the self-rolling up structure with a gap in the hybrid hydrogel sheet was possible. These results showed that we could achieve the micro self-rolling up method using the deformation of pNIPAM-AAc and fabricate rolled up structures both with and without gap in the hybrid hydrogel sheet by designing the

SU-8 plate length.

3. Evaluations

3.1. Fabricated 3D structures by self-rolling deformation

Figure 6 shows the fabricated 3D structures using the self-rolling up deformation of pNIPAM-AAc. Structures in the form of a cylindrical, conical (the Tower of Babel), croissant-like ellipsoidal, and spiral shapes were assembled in this study. Figure 6 also shows the 2D designs and images for each 3D structure, self-rolling up structures into the forward direction (20 °C), and self-rolling up structures into the backward direction (40 °C). All the structures were designed to have 15 µm interval of creases between each SU-8 plates. The cylindrical shape shown in figure 6(a) is a basic structure of the self-rolling up deformation. A total of 50 SU-8 plates with 200 µm in length and 3000 µm in width, which were connected at a 15 µm interval of creases, were rolled up. In 20 °C cold water, self-rolling up was caused by pNIPAM-AAc swelling. Hence, the SU-8 plates existed inside the pNIPAM-AAc layer. Meanwhile, in 40 °C hot water, self-rolling up was caused by pNIPAM-AAc shrinking. Therefore, the SU-8 plates existed outside the pNIPAM-AAc layer (Supplementary video). In 40 °C hot water, the cylindrical shape with 1000 µm in diameter (figure 6(b)) had a small internal hollow region with 170 µm in diameter, indicating that our self-rolling up method can provide a “good” solid 3D structure with a small hollow region. However, it was not a “perfect” solid 3D structure with no hollow region. Figure 6(b) shows the conical or the Tower of Babel shape fabricated by changing the SU-8 width stepwise. A total of 40 SU-8 plates with 500 µm in length and 3000 to 200 µm in width, which were connected at a 15 µm interval of creases, were rolled. In this design, the uneven stress caused by the pNIPAM-AAc layer must exist on the hybrid hydrogel sheet in the width direction. However, a uniform rolling up deformation was observed. Therefore, the uneven stress in the width direction can be ignored when designing the rolling up structures. Figure 6(c) shows the croissant-like ellipsoidal shape fabricated by changing the SU-8 plate length and width stepwise. Connecting the SU-8 plates of various lengths continuously led to the development of a structure with various self-folding angles. Figure 6(d) demonstrates the spiral shape with 40 parallelogram plates of SU-8 having 500 µm width and length. A spiral shape using the rolling-up deformation was previously demonstrated [27]. In our study, we achieved both clockwise and anti-clockwise spiral shapes by a bi-directional deformation. Thus, considering the obtained results, the self-rolling up deformation is useful in developing complex 3D structures, such as croissant-like ellipsoidal and rotational symmetry 3D structures with/without a hollow region.

3.2. Time response and repetitive response of self-rolling

The time responses of the self-rolling caused by hydrogel swelling and shrinking were evaluated. A cylindrical hybrid hydrogel (rolling up to the forward direction), which was kept in 20 °C cold water for over 5 min, was dipped into 40 °C hot water. The deployment of the cylindrical hybrid hydrogel and the recurrent self-rolling (rolling up to the backward direction) were observed at this time. Similarly, the

cylindrical hybrid hydrogel (rolling up to the backward direction), which was kept in 40 °C hot water for over 5 min, was dipped into 20 °C cold water. The deformation process was then observed. Figure 7(a) shows the actual images of the self-rolling up process. The entire length of the rolling up structure L_W was also measured during self-rolling up. Figure 7(b) shows the measurement result. The vertical axis represents the entire length of the rolling up structure L_W [mm], while the horizontal axis represents time T [s]. The time response of the deformation converged after 20 s in the case of the rolling up deformation from 20 °C to 40 °C water (using hydrogel shrinking). Furthermore, the time response of the deformation converged after 120 s in the case of the rolling up deformation from 40 °C to 20 °C water (using hydrogel swelling). Therefore, the time response of rolling from the low to high temperature resulted in six times faster rolling speed than that when rolling from the high to low temperature.

The repetitive response of self-rolling up was also evaluated. A hybrid hydrogel sheet with 20 SU-8 plates having 500 μm in width and 500 μm in length, which were connected at 15 μm crease intervals, was used. The hybrid hydrogel sheet was kept in 20 °C cold water before moving to 40 °C hot water and in 40 °C hot water before moving to 20 °C cold water. Each condition was repeated for five times, and the inside and outside diameters of the hybrid hydrogel structure were measured. The inside and outside diameters in this measurement were defined as the longest distance of a perpendicular line drawn from the endmost plate of SU-8 (figure 8(a)). Figure 8(b) shows the measurement results. The vertical line denotes the inside and outside diameters, while the horizontal line denotes the cycles. The diameters of the hybrid hydrogel structure in the case of the repetitive self-rolling up deformation during swelling and shrinking were approximately constant. Therefore, the repetitive response was favourable for at least several times.

3.3. Anisotropy of structure with heating process

In our self-rolling up method, the shape of the hybrid hydrogel sheet was controlled using the design and the water temperature. However, self-rolling up can control the shape by changing the rolling up method (e.g., rolling from one edge or both edges of the hybrid hydrogel sheet). Figure 9 shows the self-rolling up structures formed by changing the rolling up methods. Figure 9(a) shows the case of rolling up from one edge of the hybrid hydrogel sheet (Supplementary video). The deployed hybrid hydrogel sheet in approximately 30 °C water was collected with a syringe and put into hot water from one side. The time for starting the self-rolling up had a lag when the above-mentioned methods were used. Moreover, the rolling shape resulted in one cylindrical structure. Figure 9(b) illustrates the case of simultaneous self-rolling up from both sides of the hybrid hydrogel sheet (Supplementary video). The cylindrical structure kept in 20 °C cold water was also collected with a syringe and dropped into hot water. The moved cylindrical structure was gradually deployed, and self-rolling up on both edges of the hybrid hydrogel sheet simultaneously started. As a result, the self-rolling up structure had two cylindrical parts. The only difference of the structure with one and two cylindrical parts was the method of heat application.

4. Conclusion

We proposed micro self-rolling up method and demonstrated the development of complex 3D structures using the self-rolling up deformation of a hybrid hydrogel sheet consisting of temperature-responsive hydrogel pNIPAM-AAc and SU-8. We fabricated hybrid hydrogel sheets with different lengths and measured the self-folding angle using hydrogel. Complex 3D structures, cylinder, cone, croissant-like ellipsoidal, and spiral shapes were then assembled using the self-rolling up deformation. Our self-rolled up structures can provide 3D structures with a small hollow region in the center of rolling up and no gaps between the rolled-up sheet. In addition, the time response of the self-rolling up was approximately 20 s when increasing the water temperature. We confirmed repetitive deformation for several times. Our self-rolling up method can be used for applications in the field of micro soft devices, such as medical patch.

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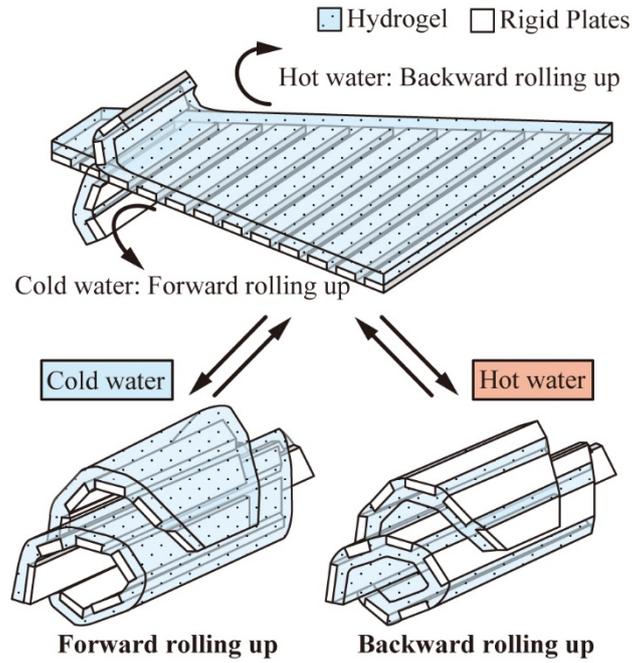


Figure 1. Concept of the self-rolling up deformation used to build micro 3D structures using a temperature-responsive hydrogel with rigid plate array. This self-rolling up method involves rolling up a 2D flat sheet through continuous self-folding, like the making procedure of a croissant. Temperature-responsive hydrogel changes shape by temperature, and produces forward rolling up in cold water and backward rolling up in hot water.

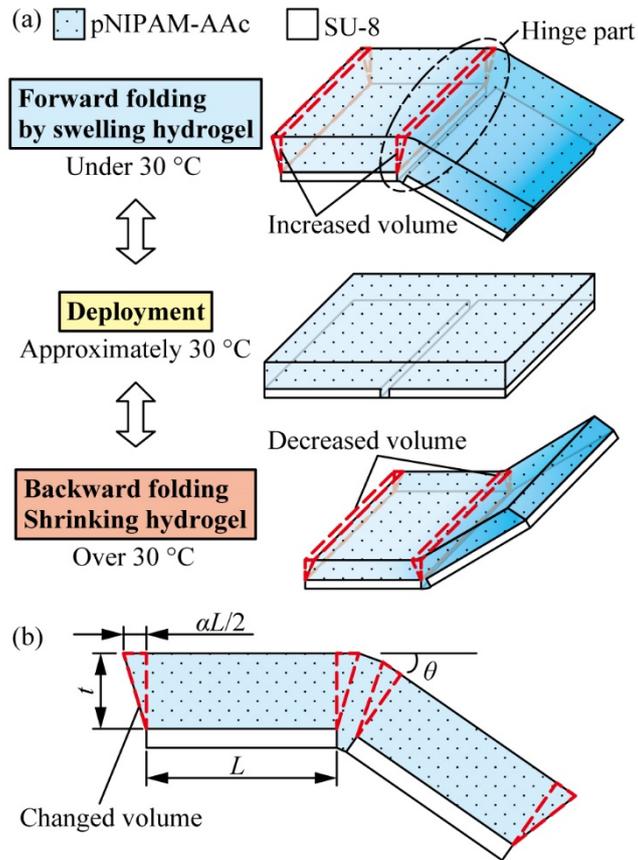


Figure 2. Principle of self-folding using pNIPAM-AAc and SU-8. (a) Schematic images of the self-folding caused by the volume change of the pNIPAM-AAc hydrogel. (b) Model of the cross section in the hybrid hydrogel sheet used to schematically calculate the self-folding angle.

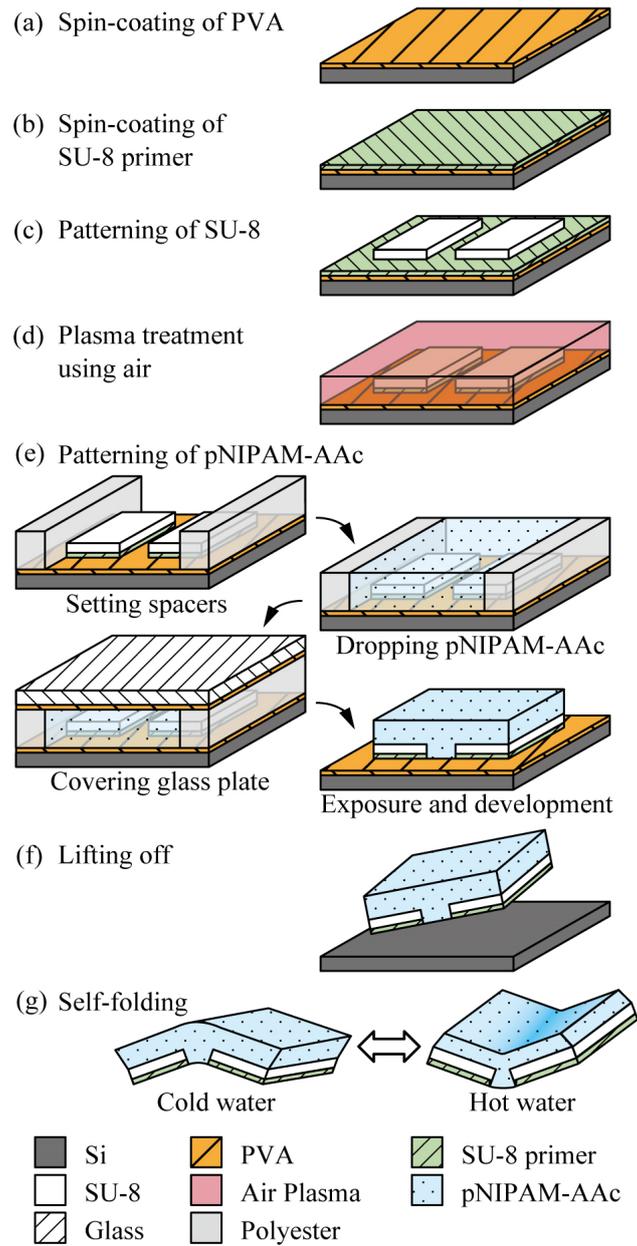


Figure 3. Fabrication process of rolling the 3D structure using the hybrid hydrogel sheet.

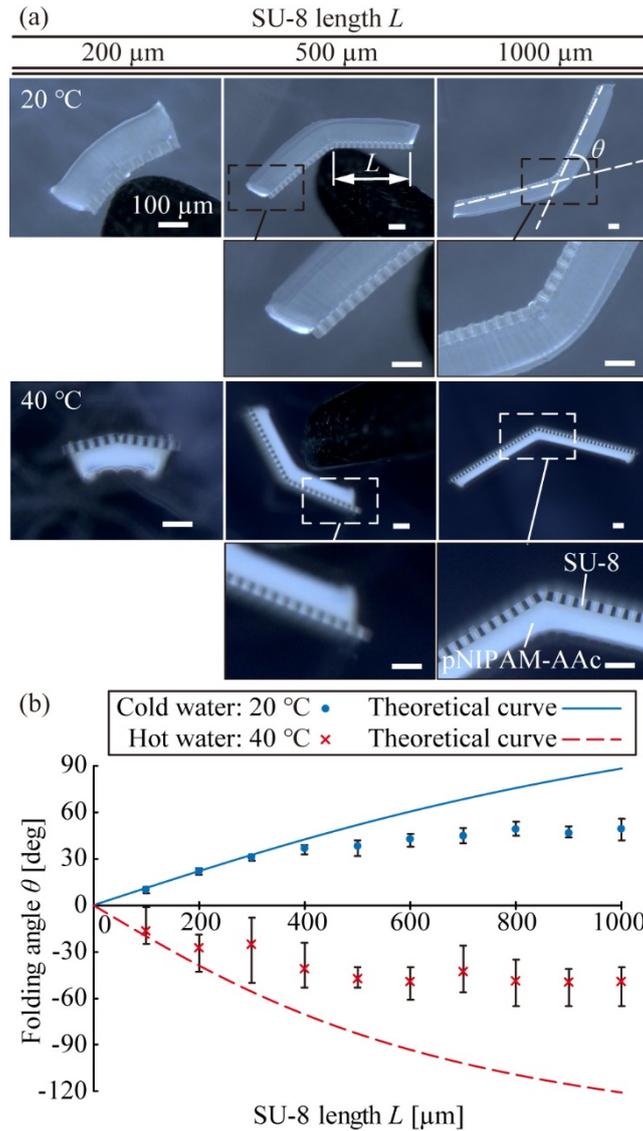


Figure 4. Basic characteristic of the self-folding angle using the hybrid hydrogel sheet. All scale bars are at 100 μm . (a) Actual images of the self-folded hybrid hydrogel sheets during swelling (20 $^{\circ}\text{C}$) and shrinking (40 $^{\circ}\text{C}$) of the hydrogel. The arrays of lines like teeth on the SU-8 sides are holes provided for anchor to the pNIPAM-AAc layer. (b) Characteristic of the self-folding angle with the changing plate length of SU-8 ($N = 5$).

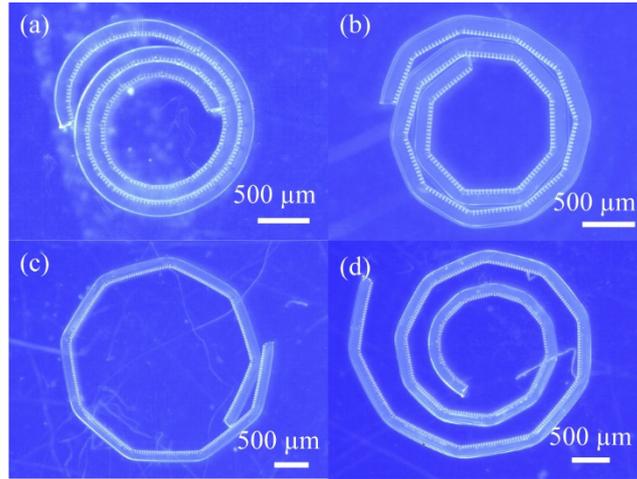


Figure 5. Fabricated self-rolling up structures in 20 °C cold water. (a) 200 μm SU-8 plates. (b) 500 μm SU-8 plates. (c) 1000 μm SU-8 plates. (d) 200 μm to 1000 μm SU-8 plates with increasing 200 μm steps of the SU-8 plates.

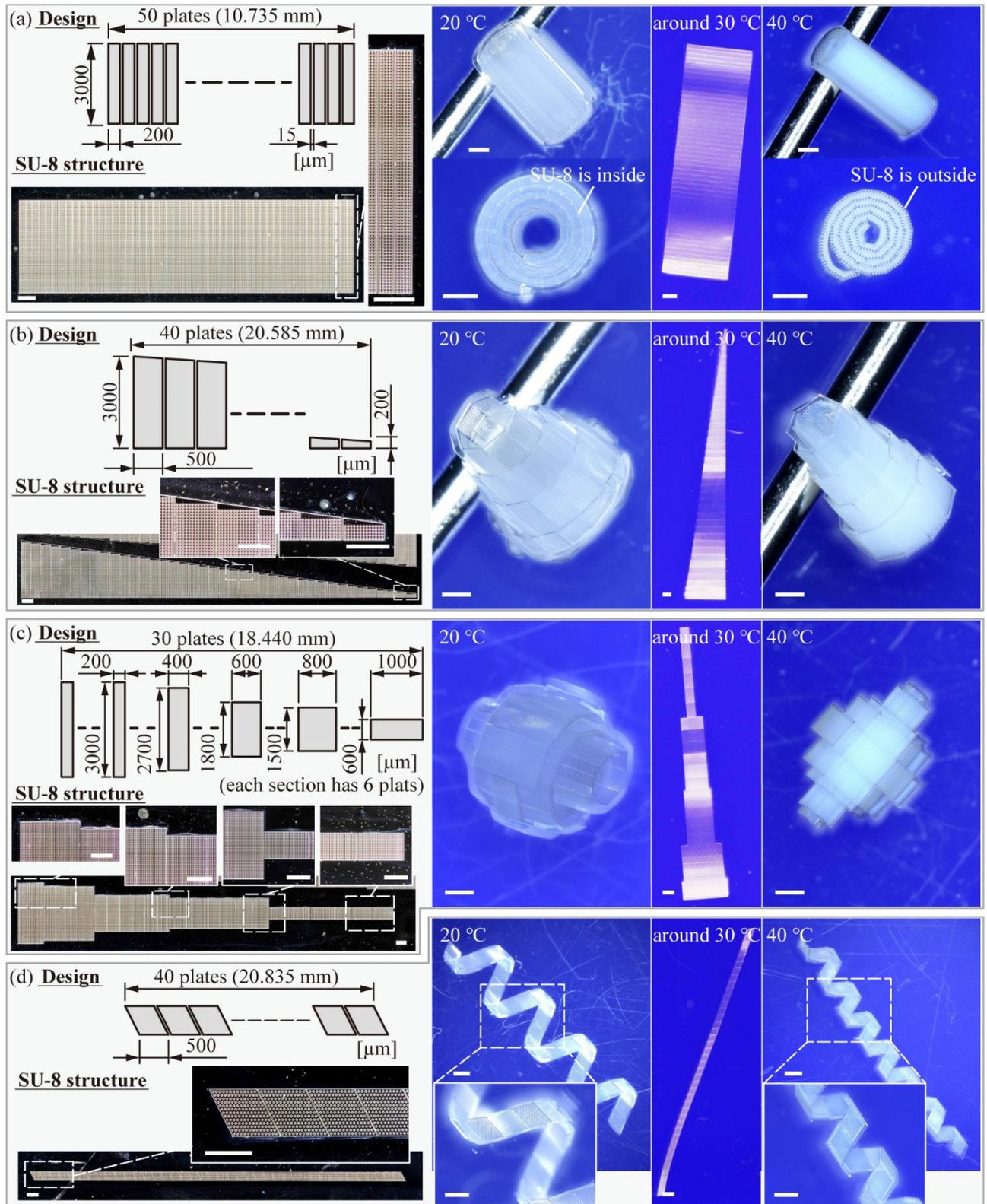


Figure 6. Designs and 3D structures fabricated through self-rolling up at 20 °C, approximately 30 °C, and 40 °C. All scale bars are at 500 μm. (a) Cylinder, which is a basic structure of the self-rolling up deformation. (b) Cone (the Tower of Babel) rolled up from a right triangle sheet. (c) Croissant-like ellipsoid rolled up from a triangle sheet. (d) Spiral, which changes rolling up directions with temperature.

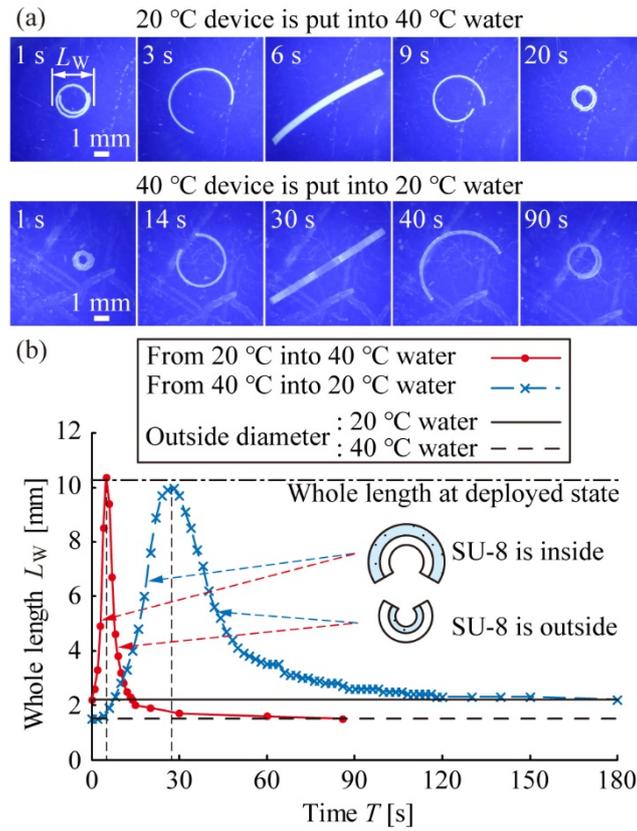


Figure 7. Time response of the hybrid hydrogel sheet. (a) Images of the hydrogel sheet with self-rolling up and deployment. (b) Time responsibility evaluated by measuring the whole length of the hybrid hydrogel sheet.

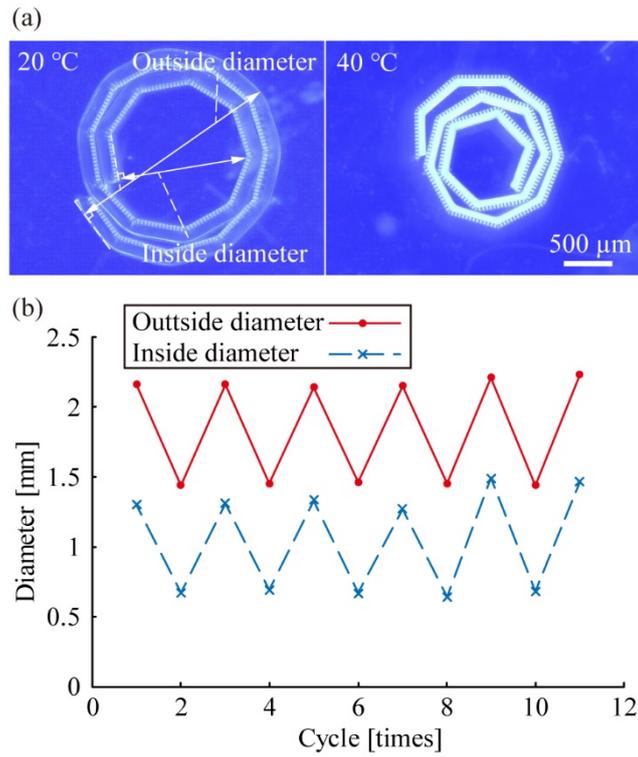


Figure 8. Repetitive response of the hybrid hydrogel sheet. (a) Inside and outside diameters defined in the actual image. (b) Repetitive response evaluated by changing the temperature from 20 °C to 40 °C for five times.

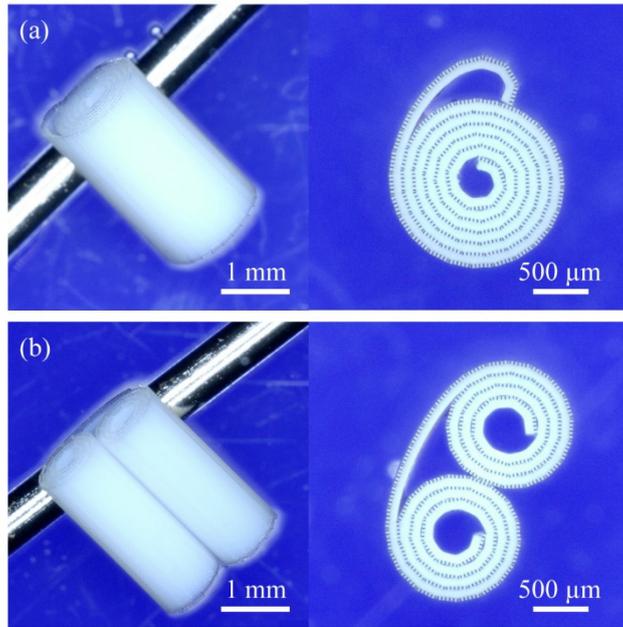


Figure 9. Two stable states of the self-rolling up structure by changing the methods of applying heat. (a) Heat is applied from one edge of the hydrogel sheet to another. (b) Heat is simultaneously applied to the whole region.