Soft mechanical metamaterials with unusual swelling behavior and tunable stress-strain curves

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Soft adaptable materials that change their shapes, volumes, and properties in response to changes under ambient conditions have important applications in tissue engineering, soft robotics, biosensing, and flexible displays. Upon water absorption, most existing soft materials, such as hydrogels, show a positive volume change, corresponding to a positive swelling. By contrast, the negative swelling represents a relatively unusual phenomenon that does not exist in most natural materials. The development of material systems capable of large or anisotropic negative swelling remains a challenge. We combine analytic modeling, finite element analyses, and experiments to design a type of soft mechanical metamaterials that can achieve large effective negative swelling ratios and tunable stress-strain curves, with desired isotropic/anisotropic features. This material system exploits horseshoe-shaped composite microstructures of hydrogel and passive materials as the building blocks, which extend into a periodic network, following the lattice constructions. The building block structure leverages a sandwiched configuration to convert the hydraulic swelling deformations of hydrogel into bending deformations, thereby resulting in an effective shrinkage (up to around -47% linear strain) of the entire network. By introducing spatially heterogeneous designs, we demonstrated a range of unusual, anisotropic swelling responses, including those with expansion in one direction and, simultaneously, shrinkage along the perpendicular direction. The design approach, as validated by experiments, allows the determination of tailored microstructure geometries to yield desired length/area changes. These design concepts expand the capabilities of existing soft materials and hold promising potential for applications in a diverse range of areas.

INTRODUCTION

Development of soft adaptable materials that change their properties and performances in response to changes under ambient conditions is of growing interest in recent years, due to their applications in tissue engineering (1, 2), soft robotics (3–6), biosensing (7, 8), and flexible displays (9–11). To design these soft responsive materials, various active materials have been exploited, including hydrogels (12–16), shape memory polymers (17–20), and liquid crystal elastomers (21, 22). The resulting composite systems are capable of responding to well-defined external stimuli such as ionic concentration, heat, pH, and light (23–29). In particular, the material systems that incorporate hydrogels as the active components are very attractive due to the superior biocompatibility and high level of swellability [for example, up to 1000-fold when immersed in certain solvents (30, 31)]. Their programmable large swellings can be used in a diversity of environmentally responsive devices [for example, microfluidic valves (32, 33) and artificial muscles (34, 35)]. The existing studies mainly focused on the positive swelling behavior (that is, corresponding to a positive volume change during water absorption) as a means to achieve desired shape programming or other functionalities. By contrast, the negative swelling behavior (that is, corresponding to a negative volume change during water absorption) represents a relatively unusual phenomenon that does not exist in most natural materials. This unusual behavior, similar to negative thermal expansion (36–44), can be used to systematically control the changes in length, area, and volume, in combination with the traditional positive swelling behavior. Capabilities of hydrogel-based systems for large reversible expansion/shrinkage are also attractive for a variety of potential applications, such as drug delivery, biocompatible stents, molecular sieves, and moisture/pH sensors. Despite its potential, the development of material systems that can achieve a large negative swelling remains a challenge, and very limited designs can be found in literature (45). In particular, no strategy has been reported to achieve large, anisotropic, negative swelling on demand.

Here, we introduced a class of soft mechanical metamaterials that can achieve large effective negative swelling ratios, with desired isotropic/anisotropic features. The design concepts incorporate horseshoe-shaped sandwich microstructures originally developed for stretchable electronics (46, 47) into periodic networks related to those proposed for lightweight, loading-bearing structures (48, 49). Inspired by Lakes and colleagues’ seminar works on the designs that rely on bending bilayer bars to achieve negative thermal expansion (37, 43), the horseshoe-shaped sandwich microstructure designed herein can effectively convert a hydraulic swelling deformation of hydrogel into a bending deformation and, thereby, a shrinkage or an amplified expansion of the entire network. Since this network design does not involve any instabilities, it is different from the design adopted by Liu et al. (45) that leverages buckling to enable tunable negative swelling ratios. During the shrinkage of the network metamaterial, the horseshoe microstructure pattern is also able to make more efficient use of the space without interacting with the neighboring microstructures, as compared to the arc pattern (similar to half of a sinusoidal wave) that may appear during buckling. By using validated design tools based on the computational mechanics, these network composites can yield a wide range of desired swelling responses, including some unusual behavior, such as a concurrent expansion along a direction and shrinkage along the perpendicular direction. A systematic comparison to the existing soft materials illustrates the powerful capability of the proposed design concepts. Furthermore, these network composites can be designed to offer a variety of tunable stress-strain curves, which has important utilities in deployable antennas and soft robotics to actively control their configurations and/or motions.

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RESULTS

Figure 1 presents a schematic illustration of the design strategy and fabricated samples. Here, the material system is constructed in a periodic two-dimensional (2D) triangular lattice configuration, in which the filamentary, horseshoe composite microstructures of hydrogel and passive materials serve as the building blocks. This type of reconfigurable, network composite material can be fabricated using the techniques of multimaterial 3D printing (see Materials and Methods for details), similar to the concepts of 4D printing (50–52). The horseshoe-shaped microstructure consists of two identical circular arcs, with an arc angle of \( \theta \) and a radius of \( r \), as shown in Fig. 1B. Each arc segment has a sandwich structure (see fig. S1A for more details) of a supporting layer (a digital polymeric material, \(~65\) MPa; RGD8530, Stratasys; in blue; width \( W_1 \)), an active layer (a hydrogel, \(~0.2\) MPa; SUP705, Stratasys; in red; width \( W_3 \)), and an encapsulation layer (an elastomer, \(~0.5\) MPa; TangoBlackPlus, Stratasys; in green; width \( W_2 \)). Here, a thin encapsulation layer (90 \( \mu \m) in width) serves to maintain a well-bonded interface of the hydrogel layer and the supporting layer during the swelling and deswelling processes. Small holes are arranged on top of the encapsulation layer to allow water (or other solutions) to flow in and out, similar to the design adopted by Mao et al. (19, 20).

The horseshoe-shaped sandwich microstructure can be represented mainly by four nondimensional parameters, including the arc angle \( \theta \), width ratio \( W_2/W_1 \), normalized width \( W_1/r \), and length ratio (\( s/r \)) of the active layer to the supporting layer, as long as the width of the encapsulation layer is fixed in the design. By tuning these design parameters, the middle of the hydrogel layer can be well separated from the neutral mechanical plane of the sandwich structure such that the hydraulic swelling deformations of hydrogel can be converted into the bending deformations. Additional details of a specific example that illustrate this mechanism appear in fig. S2. Both an effective shrinkage and expansion of the entire network material are possible with this mechanism, depending on the magnitude of the arc angle and the placement of the active layer relative to the supporting layer (fig. S1B).

Figure 1 (C and D) demonstrates a representative network composite material that can achieve a reversible negative swelling, with a fractional area change up to \(~70\%\) (and a relative length change up to \(~45\%)\). This design adopts a straight microstructure (\( \theta = 0^\circ \)) such that the bending deformations can induce a relatively large change of the end-to-end distance. When the network material is immersed in water, the hydration process begins to expand the hydrogel layer and the supporting layer. As an example, Fig. 1 (E and F) presents the stress-strain curves of the above design after water absorption for different times. Without any water absorption, the network material, consisting of straight microstructures, undergoes stretching-dominated deformations under a uniaxial stretching, leading to a relatively linear stress-strain response. After water absorption (for example, for 45 min), the network material with reshaped horseshoe microstructures undergoes bending-dominated deformations at the initial stage of uniaxial stretching, as shown in Fig. 1G and fig. S4. The stress thereby increases slowly with increasing strain during this stage. As the horseshoe shapes begin to reach full extension (~65% strain), the slope of the stress-strain curve increases rapidly due to the transition into a stretching-dominated deformation mode. This leads to a J-shaped, nonlinear stress-strain response that widely exists in biological materials (for example, human skins and heart valves) (53, 54). The complete extension of the horseshoe microstructures typically defines a critical strain (\( \epsilon_c\)) that marks the transition point of the J-shaped stress-strain curves. As shown by Fig. 1E, the J-shaped stress-strain curves can be effectively tuned by controlling the time of water absorption. As the saturated network material experiences the dehydration process, the stress-strain curve moves leftward gradually until the J shape vanishes, indicating a good recovery. FEA can capture the tunability of the stress-strain response in a quantitative manner, as evidenced by the reasonably good correspondence with experiments (Fig. 1F).

Because of the design flexibility enabled by tailoring the microstructure geometries, the above network composite design provides an ideal platform to offer desired swelling ratios in a wide range. Figure 2 summarizes a collection of theoretical and experimental results that elucidate the effects of three dominant design parameters, including the arc angle \( \theta \), width ratio \( W_2/W_1 \), and length ratio (\( s/r \)). For relatively slender horseshoe microstructures, we develop a mechanics model that simplifies the sandwich microstructures as planar beams consisting of only the supporting layer (\( W_1 \)) and the active layer (\( W_2 \)), with a negligible stiffness contribution from the soft, thin encapsulation layer. Assuming planar cross-sections during deformation, the theory of composite beam (see Supplementary Methods and fig. S5 for details) shows that the curved beam (with curvature radius \( r \)) maintains an arc shape, as the active layer undergoes a swelling ratio (\( \epsilon_{\text{hydrogel}} \)). Because of the strain mismatch between the active layer and the supporting layer, the arc radius changes to \( r_{\text{deformed}} \) and is solved as...
Fig. 1. Design concepts of soft mechanical metamaterials with large negative swelling ratios and tunable stress-strain curves. (A) Schematic illustration of soft network composite design, with inset denoting a representative unit cell. (B) Cross-sectional view and exploded view of sandwiched horseshoe microstructure consisting of supporting layer (RGD8530; in blue), active layer (SUP705; in red), and encapsulation layer (TangoBlackPlus; in green). The two images at the bottom illustrate a cartoon of the designed microstructure and corresponding image of a 3D-printed sample. (C) Experimental (top) and computational (bottom) results on the evolving configurations of a representative network material during the hydration and dehydration processes. The design parameters are the arc angle $\theta = 0^\circ$, width ratio $W_2/W_1 = 2.5$, normalized width $W_1/r = 0$, and length ratio $s/r = 0.67$. (D) Swelling-induced strain components as a function of processing time for the network material in (C). (E) Measured stress-strain curves of the network materials in (C) at different stages of hydration and dehydration. Square and triangular symbols denote the results during hydration and dehydration, respectively. (F) Calculated stress-strain curves of the network materials at different stages of hydration, in comparison to experimental results. (G) Experimental (top) and computational (bottom) results on the deformation sequences of a hydrated (~45 min) network material under a uniaxial stretching. Scale bars, 5 mm (B) and 40 mm (C and G).
The isotropic strain response arises from the homogeneous microstructure design. Figure 2A and fig. S5 show that the analytic solution (Eq. 2) corresponds reasonably closely to the results of FEA and experiment on the effective strain of the network material for a wide range of arc angle \( \theta \) and length ratio \( s/r \). In Fig. 2A, the dimensionless swelling strain \( \varepsilon_{\text{swelling}} \) of the network material normalized by the value \( \varepsilon_{\text{hydrogel}} \) at a nearly saturated state of the hydrogel. The negative arc angle represents the placement of the active layer to the inner side of the supporting layer, as opposed to the case of positive arc angle discussed above (fig. S1B). A distinct swelling behavior is evident for different arc
angles, as shown in Fig. 2 (D to F). Specifically, for $\theta = -220^\circ$, a large positive swelling ratio (linear strain of up to $-98\%$) can be achieved by expanding the horseshoe microstructures with water absorption, as evidenced by the deformed configurations (Fig. 2D) from both the experiment and the FEA. For $\theta = -90^\circ$, the network material undergoes an expansion at the initial stage of hydration, and the associated swelling ratio ($e_{x,swelling}$) increases to approach a peak value ($10.8\%$) at the dimensionless swelling ratio of $e_{hydrogel} = 0.42$. Beyond this critical point, the swelling ratio ($e_{x,swelling}$) decreases with further water absorption and becomes negative at $e_{hydrogel} = 0.74$. At the state of $e_{hydrogel} = 1$, the shrinkage reaches its maximum, corresponding to a negative swelling ratio of $e_{x,swelling} = -4.3\%$ based on FEA (and $-5.1\%$ based on the experiment). This represents an unusual type of swelling behavior—expanding first and then shrinking during hydration. For $\theta = 0^\circ$, as discussed in Fig. 1, the straight microstructures enable a large negative swelling ($e_{x,swelling} = -47.8\%$ and $-47.1\%$ based on FEA and the experiment, respectively). Further increase of the arc angle (for example, to $\theta = 90^\circ$ or $180^\circ$) leads to a reduced negative swellability, as shown in Fig. 2A, due to the self-contact of microstructures that occurs at an earlier stage of hydration (Fig. S6). In the examples studied here, the network designs with $\theta = -220^\circ$ and $0^\circ$ offer the largest expansion and shrinkage, respectively, and both show a monotonous dependence on the swelling ratio ($e_{hydrogel}$) of hydrogel.

Figure 2 (B and C) highlights the effect of length ratio ($s/r\theta$) and width ratio ($W_2/W_1$) on the swellability of two representative network designs ($\theta = 0^\circ$ and $-220^\circ$), as characterized by the effective strain at $e_{hydrogel} = 1$. Both figures show an increased magnitude of effective strain (that is, $|e_{x,swelling}|$) at a larger length ratio ($s/r\theta$). For a given length ratio (for example, $s/r\theta = 0.67$), the magnitude of effective strain usually increases first with increasing width ratio ($W_2/W_1$) because the actuation force increases, as shown in Fig. 2B for the design with $\theta = 0^\circ$. At a critical width ratio ($W_2/W_1 \approx 3.2$ in Fig. 2B), the magnitude of effective strain $e_{x,swelling}$ reaches a peak, after which $|e_{x,swelling}|$ decreases. This can be attributed to the reduced offset of the neutral mechanical plane relative to the middle of the hydrogel layer, with the further increase of the width ratio beyond $W_2/W_1 \approx 3.2$. Because of this mechanism, we observed an optimal value of width ratio ($W_2/W_1$) independent of the length ratio ($s/r\theta$) (Fig. 2B), which can maximize the shrinkage of the network material. A similar dependence can be also found for the design with $\theta = -220^\circ$ (Fig. 2C). According to these dependences, we can expect a reduced swellability with an increasing filling ratio of the cellular network material or equivalently decreasing the porosity. For the design with straight microstructures (that is, $\theta = 0^\circ$) and fixed length ratio ($s/s_{\text{total}} = 0.75$), the magnitude of swelling-induced strain decreases gradually from $-47.2\%$ to $11.0\%$ (and to $3.5\%$), as the filling ratio (defined on the basis of the undeformed configuration) increases from $5\%$ to $12.5\%$ (and to $22.5\%$) (Fig. S7).

Excluding the isotropic expansion/shrinkage during water absorption, anisotropic swelling/desorption is also achievable by exploiting heterogeneous horseshoe microstructures in the representative unit cell of network materials. By simply tailoring the selections of length ratio ($s/r\theta$) and arc angle ($\theta$) for the differently oriented horseshoe microstructures, a variety of usual swelling responses can be accessed. Figure 3 and figs. S8 to S10 demonstrate a representative collection of examples designed for this purpose. In particular, Fig. 3A presents the swelling path in the space of strain components ($e_{x,swelling}$ and $e_{y,swelling}$) for our designed network materials, in comparison to the traditional hydrogels (31), polymers (55), and elastomers (56), as well as a buckling-guided metamaterial design (45). The traditional soft materials (for example, elastomers, polymers, and hydrogels) usually offer an isotropic positive swelling, with the linear expansion strain that can exceed 150% for specially designed hydrogels (31). In terms of the isotropic negative swelling, the materials presented here can reach a linear strain of $-48\%$, exceeding that ($-16\%$) reported previously (45). In terms of the anisotropic swelling, the traditional composite materials are typically constrained by the swellability of the constituent materials, which give limited levels of responses. By contrast, the current network design, because of the versatile flexibility, is able to offer a diversity of anisotropic swelling responses, including some unusual behavior (for example, expanding along one direction while shrinking along the perpendicular direction). Figure 3B shows an example that shrinks only along the horizontal direction ($x$) during hydration while staying undeformed along the perpendicular direction ($y$). This is achieved by using hydrogel layers only at the two diagonally oriented horseshoe microstructures and thickening the vertically oriented microstructures to suppress the deformations along this direction. More details of this design are in fig. S9A. Anisotropic negative swelling is also possible (Fig. 3C and fig. S9B) by exploiting different length ratios ($s/r\theta = 0.67$ versus 0.25) for the differently oriented microstructures. In this example, the ratio ($K_{xy} = e_{x,swelling}/e_{y,swelling}$) is maintained at approximately 0.27 during the entire process. Figure 3D illustrates a network material designed to respond to water absorption in the form of expansion along the $y$ direction and shrinkage along the $x$ direction. Here, the hydrogel layers are adopted only at the horizontally oriented microstructures, such that a lateral expansion can be achieved through a rotation of the passive components oriented diagonally (see fig. S9C for details). Figure 3E presents another type of design to offer a similar response through distinct placements of the hydrogel layer relative to the supporting layer at the different microstructures (see fig. S9D for details). In this example, the ratio ($K_{yy}$ of swelling-induced strains varies considerably during the deformation process. Additional examples that offer anisotropic positive swelling ratios are provided in figs. S8 and S10. In all of the above cases, FEA results correspond well with the experimental measurements (figs. S9 and S10). Excluding the above experimental demonstrations as summarized in Fig. 3A, the proposed network design is also able to cover most of the blank spaces between the plotted red curves, thereby substantially expanding the accessible regime of existing soft materials in the space of swelling ratios $e_{x,swelling}$ and $e_{y,swelling}$.

Because of the versatile control of the microstructure geometries through water absorption, a systematic tunability of the stress-strain curve is achievable, taking into account the responses along two principal directions simultaneously. Figure 4A shows a network material that becomes softer as the hydration proceeds. The J-shaped stress-strain curves shift rightward nearly at the same pace for the uniaxial stretching along the $x$ and $y$ directions (Fig. 4, B and C, and fig. S11). The elastic modulus at the infinitesimal deformations decreases by nearly three orders of magnitude from the dry state to the hydrated state (Fig. S12A). The critical strains ($e_{x,cr}$ and $e_{y,cr}$) that mark the transition point of the J-shaped stress-strain curves can be tuned well by controlling the time of hydration (fig. S12B). In contrast to the case of $x$-directional stretching, the diagonally oriented horseshoe microstructures serve as the main load-bearing components for the $y$-directional stretching, as shown by the deformation sequences in Fig. 4 (B and C). Figure 4D presents a network material that expands along the $x$ direction and shrinks along the $y$ direction during hydration, similar to the design in Fig. 3E. As a result, the stress-strain curve along the $x$ direction moves rightward, while the one along the $y$ direction moves leftward, as
hydration proceeds, as shown by the FEA calculations and experiments (Fig. 4D, middle and right). This indicates a decrease of elastic modulus $E_x$ and critical strain $\varepsilon_{cr,y}$ as well as an increase of $E_y$ and $\varepsilon_{cr,x}$ (Fig. S12, C and D). Figure 4E demonstrates another type of mechanical tunability by exploiting the network designs (as in Fig. 3C) with an anisotropic negative swelling. The resulting stress-strain curves move rightward for both the $x$ and $y$ directions, but at different rates (Fig. S12, E and F).

**DISCUSSION**

In summary, the design concepts, fabrication techniques, and quantitative design methods reported here provide access to soft mechanical metamaterials that can achieve large effective negative swelling ratios and tunable stress-strain curves, with desired isotropic/anisotropic features. The demonstrated unusual swelling behaviors encompass a transition from expansion to shrinkage during hydration and a concurrent expansion along a direction and shrinkage along perpendicular
direction. The validated design methods allow the determination of tailored microstructure geometries to yield desired length/area changes.

The proposed network design is basically scale-free, but fabrication capabilities set the size limit of the network materials. The commercial multimaterial PolyJet 3D printer, which we used in the current work, offers a minimum layer thickness of 30 μm during 3D printing, which sets the limit of width ($W_3 = 90 \mu m$) for the encapsulation layer, since a few layers are required to form a relatively reliable structure. A good negative swelling can be observed when the design in Fig. 2F is scaled down by a factor of 0.6 (fig. S16). To fabricate even smaller network materials with similar swelling performances, other advanced manufacturing techniques [for example, multimaterial projection microstereolithography]
should be exploited. Considering the bottom-up feature of the proposed metamaterial design that builds on the horseshoe-shaped composite microstructures, it can also be extended to other 2D (for example, square, Kagome, honeycomb, etc.) or 3D (for example, tetrahedral, octahedral, etc.) networks. The tunable stress-strain curves can be used to achieve structures with varying stiffness, which can potentially be exploited in deployable antennas and soft robotics to actively control their configurations and/or motions. Collectively, the findings reported here open opportunities for the design of structures that can achieve targeted length/area/volume changes, as required for applications in aerospace (57), optical (58), and microelectronic areas (59).

**MATERIALS AND METHODS**

**Fabrication of the network materials**

All of the network materials in this work were fabricated using the multimaterial PolyJet 3D printing technique (Objet350, Stratasys). The three different materials (RGD8530, TangoBlackPlus, and SUP705) used in the fabrication were all available from the multimaterial 3D printing technique (Object 350, Stratasys). Of these materials, TangoBlackPlus is a soft elastomer (~0.5 MPa; see the stress-strain curve in fig. S13A) that mainly consists of exo-1,7,7-trimethylbicyclo[2.2.1]hept-2-yl hept-2-yl acrylate and photoinitiators. RGD8530 is a kind of digital polymeric material (~65 MPa; see the stress-strain curve in fig. S13B) mixed by TangoBlackPlus and VeroWhite, a rigid polymer mainly consisting of exo-1,7,7-trimethylbicyclo[2.2.1]hept-2-yl acrylate, tricyclosedecane dimethanol diacrylate, titanium dioxide, and photoinitiators. Both TangoBlackPlus and RGD8530 can be cured by ultraviolet light at room temperature. SUP705 is a hydrogel material (~0.2 MPa; saturated linear swelling strain of ~21%) that can swell when immersed in water (19). It is composed of poly(oxy-1,2-ethanediyl)-α-(1-oxo-2-propenyl)-ω-hydroxy-1,2-propylene glycol, polyethylene glycol, glycerin, phosphine oxide, phenylbis (2,4,6-trimethylbenzoyl)-, and acrylic acid ester.

**Measurements of stress-strain curves and swelling ratios**

The hydration process was carried out at room temperature, with the hydration level controlled by the time of water absorption. The dehydration process was carried out in a drying oven (with air environment) maintained at 75°C. The photographs of the specimens at different stages of hydration and dehydration were recorded using a digital camera (760D, Canon), from which the effective swelling ratios of the network materials can be determined. The stress-strain curves were measured in the air environment using a commercial mechanical testing machine, after the samples were taken from the water. A low loading rate (2 mm/min) was adopted to ensure that the deformations were nearly quasi-static and that the viscoelastic effect could be neglected. Since most of the mechanical testing was finished within an hour, the deswelling deformations induced by water evaporation were negligible in such a short time (fig. S14). Most of the experiments were performed on approximately three different samples for each design, and we obtained consistent results.

**Finite element analyses**

The commercial software ABAQUS (SIMULIA) was used to carry out the FEA, in which the implicit solver was used to calculate the deformations and stress-strain curves. The geometrical nonlinearities were taken into account in the FEA. Eight-node linear hybrid brick elements with reduced integration were adopted, with refined meshes to ensure computational accuracy. Because of the relatively small thickness of the supporting layer as compared to the entire sandwich structures, the strains were typically quite small (for example, <3%) in the supporting layer (RGD8530) such that a linear elastic constitutive relation can be used in the FEA for simplicity. The Young’s modulus ($E_{\text{RGD8530}} = 65$ MPa) and Poisson’s ratio ($\nu_{\text{RGD8530}} = 0.4$) were determined by mechanical testing. For the encapsulation layer (TangoBlackPlus), a Mooney-Rivlin hyperelastic constitutive relation with a Poisson’s ratio of ~0.5 was used, and the uniaxial stress-strain curve measured in the experiment (fig. S13B) was imported in the simulations. The hydrogel layer (SUP705) was simplified as a linear elastic solid with an isotropic swelling, and the material constants included the Young’s modulus $E_{\text{SUP705}} = 0.2$ MPa and Poisson’s ratio $\nu_{\text{SUP705}} = 0.5$. To simulate the hydration process of the network material, an equal triaxial swelling strain was applied to the hydrogel layer to calculate the deformed configurations. The time-evolving swelling ratio of the hydrogel material after water absorption was determined according to the experimental and FEA results of deformation history in a straight sandwich microstructure (fig. S2). On the basis of the deformed configurations at different stages of hydration, a uniaxial stretching was then applied to the rectangular-shaped specimens of network material with the same geometry as that in the experiment to calculate the stress-strain curve. Note that a hyperelastic constitutive model can capture the mechanical responses of the hydrogel layer more accurately. A set of FEA that adopted the Mooney-Rivlin law (with parameters $C_{10} = 0.031$, $C_{01} = 0.0077$, and $D_{1} = 0$ in ABAQUS) to model the hydrogel layer served as a reference for comparison. The results (fig. S15) showed that the magnitudes of swelling-induced strains based on the hyperelastic model were slightly lower than the results based on the linear-elastic model for two representative designs. These slight differences can be attributed to the relatively low levels of strain in most regions of hydrogels during deformation.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/6/eaar8535/DC1

**Methods: Mechanics model of the swelling-induced deformations**

fig. 51. Illustration of the sandwich configuration and two different placements of the hydrogel layer relative to the supporting layer.

fig. 52. Deformations of a straight sandwich structure during the hydration and dehydration processes.

fig. 53. Distribution of the maximum principal strain for a representative unit cell of the network material at the different stages of hydration and dehydration processes, corresponding to the different states shown in Fig. 1C.

fig. 54. Distribution of the maximum principal strain for the network structure at the different stages of uniaxial stretching, corresponding to the different states shown in Fig. 1G.

fig. 55. Illustration of the mechanics model of the swelling-induced deformations and results on the swelling-induced strains.

fig. 56. Deformations of the network materials during hydration.

fig. 57. Effect of the filling ratio on the swelling-induced strains of the network materials with straight microstructures (that is, arc angle $\theta = 0^\circ$).

fig. 58. Soft network materials with anisotropic positive swelling ratios.

fig. 59. FEA and experiment results on the swelling-induced deformations for the four designs in Fig. 3 (B to E).

fig. 60. FEA and experiment results on the swelling-induced deformations for the two designs in fig. 5B.

fig. 61. Experimental (left) and computational (right) results on the deformation sequences of a hydrated (~45 min) network material under a uniaxial stretching along the y direction, as in Fig. 4C.

fig. 62. Tunable elastic modulus and critical strain of the soft network materials.

fig. 63. Measured mechanical properties of the constituent materials.

fig. 64. Measured configurations of a representative network sample (that is, the initial state of Fig. 1G) after it was taken from the water and put in the air environment under a natural convection condition for 0, 30, and 60 min.
fig. S15. Computed swelling-induced strain (\(\varepsilon_{\text{swelling}}\) or \(\varepsilon_{\text{swelling}}^\text{d}\)) versus the dimensionless swelling ratio of the hydrogel.

fig. S16. Demonstration of the network materials with two different scaling factors, with the sample in Fig. 2F to serve as a reference.

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