Dual-gradient enabled ultrafast biomimetic snapping of hydrogel materials

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The design of materials that can mimic the complex yet fast actuation phenomena in nature is important but challenging. Herein, we present a new paradigm for designing responsive hydrogel sheets that can exhibit ultrafast inverse snapping deformation. Dual-gradient structures of hydrogel sheets enable the accumulation of elastic energy in hydrogels by converting prestored energy and rapid reverse snapping (<1 s) to release the energy. By controlling the magnitude and location of energy prestored within the hydrogels, the snapping of hydrogel sheets can be programmed to achieve different structures and actuation behaviors. We have developed theoretical model to elucidate the crucial role of dual gradients and predict the snapping motion of various hydrogel materials. This new design principle provides guidance for fabricating actuation materials with applications in tissue engineering, soft robotics, and active medical implants.

INTRODUCTION

Shape transformation is ubiquitous in living systems, such as prey capture action by carnivorous plants (1, 2). These natural phenomena have been sources of inspiration for engineering functional shape-transforming materials (3, 4). Among others, responsive hydrogels capable of shape transformation under various stimuli have attracted tremendous interests due to their promising applications in soft robotics (5, 6), drug delivery (7), tissue engineering (8), microfluidics (9), and so on (10–12). Typical responsive polymers used for designing shape-transforming materials include poly(N,N-dimethylaminoethyl methacrylate) (PDMAEMA) and poly(N-isopropylacrylamide) (PNIPAM), which are thermo-responsive (i.e., the gels swell/deswell reversibly in response to temperature variation around the critical phase transition temperature) (12–14). The shape transformation of hydrogels largely relies on the differential swelling of hydrogels in different regions of the materials. The in-plane and/or out-of-plane mismatch in volume change drives the gradual shape evolution of hydrogels to take various shapes (13–21). Currently, the majority of efforts at this frontier are centered on enhancing the complexity of shapes that can be achieved by hydrogels and on diversifying the responsiveness of hydrogels to external stimuli (22–26).

In nature, the leaves of the Venus flytrap can rapidly close up to capture insects in one-tenth of a second, a behavior distinct from that of the typically gradual and relatively slow shape transformation of hydrogels (Fig. 1A) (2). The extremely rapid motion of this plant is believed to associate with the accumulation and quick release of energy (27). Mimicking this sudden yet discontinuous motion is essential to the development of ultrafast actuators with broad applications (e.g., in soft robots). The few existing approaches to achieve this type of motion are usually based on reversible switching between concave and convex structures of bistable polymeric sheets, which only allows for limited complexity in structures and actuation behaviors (28–30). Nevertheless, there is still a great need for new design principles of snapping motion of responsive materials.

Here, we report a nature-inspired design of responsive hydrogel sheets with the capability of accumulating elastic energy and rapidly releasing the energy through ultrafast snapping deformation. Our experimental results and theoretical model suggest that the snapping motion of hydrogel materials originate from the dual-gradient structural design of the hydrogels. When reduced graphene oxide (rGO)/PDMAEMA composite hydrogel sheets with dual-gradient structures are used as a model system, we demonstrate that the sheets can accumulate elastic energy by converting prestored thermal or chemical energy and rapidly snap (<1 s) in a reverse direction to release the elastic energy in response to external stimuli (Fig. 1). The snapping velocity, angle, and location of the sheet can be tuned by modulating the magnitude and location of prestored energy within the hydrogel. As a result, the hydrogel sheets can be programmed and snapped to achieve different structures and actuation behaviors. The new design principle of snapping deformation is general and applicable to other materials, such as neat hydrogels or elastomers. This work provides guidance for fabricating actuation materials with applications in tissue engineering, soft robotics, and active implants.

RESULTS

We used an rGO/PDMAEMA composite hydrogel sheet with dual gradients (i.e., chain and cross-linking density gradients) along the thickness direction as an exemplary system for demonstration of our concept (Fig. 2A). The composite hydrogel was fabricated by ultraviolet (UV)–induced free radical generation of GO to initiate polymerization of DMAEMA (monomer) and N,N′-methylene-bis-acrylamide (MBA; cross-linking agent) (Fig. S1). A mixture of GO, DMAEMA, and MBA (in the absence of conventional photoinitiator) was filled in-between a sealed space and irradiated by UV light. The strong UV absorption by GO led to the gradient formation of light intensity along the thickness direction of the mixture (fig. S2A) (22, 31, 32). The higher UV intensity at the UV-exposed side causes the generation of a higher concentration of free radicals on the surface of GOs (33), thus a faster polymerization of both the DMAEMA and MBA at this side (22, 31). As a result, the UV–exposed side (denoted as the HD side) has the higher chain density and cross-linking density of PDMAEMA than the other side (denoted

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showing the cross section of a dual-gradient hydrogel. Venus flytrap. (A) Snapping of the dual-gradient hydrogel sheet. (C) A cartoon showing the cross section of a dual-gradient hydrogel.

as the LD side) (Fig. 2A). The existence of chain density gradient and cross-linking density of the hydrogel was confirmed by a combination of tools including scanning electron microscopy (SEM), confocal laser scanning microscopy (CLSM), x-ray photoelectron spectroscopy (XPS), and Raman spectra (see more detailed discussion in section S3).

When submerged from 20°C water into water with 60°C higher than its volume phase transition temperature (fig. S3A), the originally flat composite sheet curved up toward the HD side along the longitudinal axis into a tubular structure in ~2 min due to the high shrinkage rate of the HD side [Fig. 2C (c2) and fig. S3, B and C]. It is notable that when placed back into the 20°C water, instead of following an exactly reverse pathway of original shape transformation, the hydrogel sheet took a completely different route in which a new intermediate state appeared. It first unrolled until bending angle decreased to ~43° in 30 s and then rapidly snapped (as fast as below 1 s) in a reverse direction along the transverse axis, associated with a sharp increase in bending angle from 38° to 540° [Fig. 2C (c3) and fig. S3D, and movie S1]. The process was followed by a period (~320 s) of continuous rolling to further increase the bending angle to a maximal value of 1089° [Fig. 2C (c5) and fig. S3D]. Last, this inversely snapped structure gradually unrolled to be flat in ~60 min (fig. S3E). The snapping axis direction of the sheets can be regulated by patterning dual-gradient regions into the sheets (fig. S4, A and B). As an example, patterning the hydrogels with left- and right-titled stripes using a mask during UV irradiation enabled the snapping of hydrogels along predefined axial direction to form chiral structures with left- and right-handedness (Fig. 2, D and E, and movie S2). Notably, the inverse snapping of hydrogels can occur for dual-gradient hydrogels with arbitrary initial shapes (fig. S4, C and D). Tuning the initial shape of hydrogels allows for controlling the initial curling configurations of hydrogels and hence the manner of subsequent inverse snapping of hydrogels.

The inverse snapping of hydrogel sheets constitutes a new mechanism of energy transformation, as illustrated in Fig. 3A. When placed back into low-temperature (T L) water, the curled sheet obtained at high temperature (T H) unrolls due to the higher swelling rate of the HD side. During this process, the longitudinal expansion of the HD side relative to the LD side first eliminates the axial shrinkage stress at the HD side and then generates cumulative expansion stress (i.e., elastic energy) at the HD side [Fig. 3A (a1 and a2)]. The accumulated axial expansion stress cannot be released because of the constraint caused by curling along the longitudinal axis, until the bending angle of the curled hydrogel decreases to a threshold value. At this moment, the stress at the HD side can be released instantaneously, leading to abruptly inverse snapping of the sheet [Fig. 3A (a3)]. Subsequently, the bending angle increases gradually until the HD side reaches swelling equilibrium [Fig. 3A (a3 and a4)]. Eventually, the inverse-snapped structure unrolls gradually as the LD side of the hydrogel sheet swells. Notably, the instantaneous release of accumulated elastic energy during snapping can induce quick jumping of hydrogels in 20°C water (fig. S4E and movie S3).

The energy conversion involved in the snapping process can be divided into three stages: (i) conversion of part of the prestored thermal/chemical effective energy (E*) into cumulative elastic energy during unrolling of the curled sheet, (ii) instantaneous release of accumulated elastic energy (E′) in the form of snapping, and (iii) gradual release of the rest energy (E″) to further curl the sheet after snapping. These energies can be correlated through a simple equation

\[ E^* = E' + E'' \]  

The variation of \( T_{Hi} \) (i.e., the initial temperature for stimulating the gel) affects not only the original deformation of the hydrogel sheet at this temperature but also its later inverse snapping deformation when the gel was subsequently immersed in water at \( T_L \). This indicates that \( E^* \) can be modulated by controlling the initial \( T_{Hi} \) used to stimulate the curling of hydrogels (Fig. 3, B and C, and S5).
clarify that the pretreatment time used for energy storage for all stimuli in this article is 10 min, unless otherwise noted. Both $E^*$ and $E'$ increase with increasing $T_H$, leading to an increased bending angle and snapping velocity in 20°C water (Fig. 3, B and C, fig. S5, eqs. S11 to S13, and a comparison of snapping velocity between movies S1 and S4). As such, the snapping of sheets can be quantitatively programmed by tuning the magnitude of prestored energy using different $T_H$. This is further confirmed by their self-regulated actuation capability when the snapping of prestimulated hydrogel sheets was used to actuate the weight lifting in 20°C water (fig. S5D). The maximum lifting weight (i.e., output energy) in 20°C water by the same sheets increased with the increase in initial $T_H$ (Fig. 3D). The same trend was observed for the lifting velocity of the hydrogel when the same weight was lifted (Fig. 3E and movies S5 and S6). The concept of modulating the storage and release of energy in hydrogels enables the design of intelligent materials with programmable motion, mass identification, and power regulation.

The snapping behaviors of hydrogels can be modulated by stimulating the gel with pH and ion strength (IS) variation, owing to the weak polyelectrolyte nature of PDMAEMA (fig. S6). Similar to $T_H$ stimulation, the hydrogel sheet rolled up toward the HD side to store energy in response to an increase in IS or addition of base and quickly snapped to release the energy in a reverse direction when placed in 20°C water (fig. S6 and movie S7). It is worth noting that the inverse snapping of hydrogel sheets can occur in a broad range of external stimuli, such as $T_L$ of...
0° to 35°C, weak IS, and strong acid and base (fig. S6 and movie S8 to S10). This overcomes the limitation of narrow operating conditions for conventional responsive hydrogels (e.g., with lower critical solution temperature).

Furthermore, the composite hydrogels are responsive to near-infrared (NIR) light due to the photothermal effect of rGOs (25). In response to localized NIR irradiation in the air, the hydrogel sheet bent exclusively toward the HD sides regardless of the irradiation side due to the original dual gradients [Fig. 4A (a2) and fig. S7]. Submerging the NIR-irradiated hydrogel sheet into 20°C water can also induce its inverse snapping [Fig. 4A (a2, -1)]. Notably, NIR light enabled local energy storage and, hence, controllable inverse snapping of hydrogel sheets to produce various complex structures under constant stimulating conditions (Fig. 4A and movies S11 and S12). As an example, we showed programmable folding of gel sheet into a cube by controlling the location of light exposure and energy storage within the sheet (Fig. 4B). Such programmable materials have great potential in biomedicine (e.g., for minimally invasive surgery) and soft robotics (34).

We quantitatively analyzed the snapping process and established a general criterion for inverse snapping as follows (see detailed explanation in eqs. S1 to S10)

\[
a_H \frac{a_H}{a_L} > \frac{k_{i-H}}{k_{1-L}},
\]

where \(a_H\) and \(a_L\) are the expansion rates of the HD and LD sides for the curled hydrogel sheet in 20°C water, respectively, and \(k_{i-H}\) and \(k_{1-L}\) are the transverse shrinkage ratios of the HD and LD sides after initial curling, respectively.

Equation 2 suggests that the inverse snapping of the hydrogel sheet is attributed to its dual-gradient structure. The polymer chain density gradient imparts the hydrogel sheet with high expansion rate \(k(a_H/a_L)\), while the higher cross-linking density of the HD side inhibits the shrinkage/deswelling of the hydrogel sheet more than the LD side (32), leading to a relative small value of \(k_{i-H}/k_{1-L}\) of the two sides.

Two types of gradient structures together give \(a_H/a_L > k_{i-H}/k_{1-L}\) as confirmed by the presence of all data points of snapping composite hydrogels (calculated from fig. S8, A and B) in regime (I) in Fig. 5A.

The general criterion of inverse snapping we derived was further verified by comparing the deformation behaviors of bilayered PNIPAM hydrogel sheets with dual gradients (in both polymer density and cross-linking density) and with only cross-linking density, respectively. The dual-gradient PNIPAM sheets exhibited inverse snapping motions, whereas all the single-gradient hydrogels only showed simple bending in response to thermal stimulations (Fig. 5, B and C, and movies S13 and S14). The values of \(a_H/a_L\) and \(k_{i-H}/k_{1-L}\) of these hydrogels (calculated from fig. S8, C and D) fell into the corresponding regions of the diagram in Fig. 5A.

**DISCUSSION**

In summary, we have demonstrated a general principle for the design of hydrogel materials with energy transformation ability to trigger programmable snapping deformation. By controlling the magnitude and location of energy prestorage within hydrogels, the inverse snapping deformations of the hydrogels can be programmed to achieve different actuations and structures. The energy transformation–induced snapping of hydrogels is attributed to their dual-gradient structure (i.e., polymer chain density gradient and cross-linking density gradient). A theoretical model was proposed to interpret and predict the snapping of hydrogels, which is in good agreement with our experimental observations. Moreover, the dual-gradient hydrogel can directly work as a self-propelled actuator with the ability of weight identification and power control under constant stimuli. This research provides new insights into the actuation of materials and practical guidance for the design and fabrication of autonomous actuators, soft robotics, and active implants.

**MATERIALS AND METHODS**

**Materials**

DMAEMA (99%, purified through a column of alkaline alumina to remove the inhibitor before use), MBA (99%), NIPAM (>98%), N, N,N',N'-tetramethylethylenediamine (TEMED; 99%), ammonium...
peroxodisulfate, sodium fluorescein (>99%), and rhodamine 6G (99%) were purchased from Aladdin Reagents Co. (Shanghai, China). Pure water was obtained by deionization and filtration using a Millipore purification apparatus (resistivity > 18.2 megohm-cm). GO was prepared from graphite powder by the modified Hummers method. The size and thickness of the GO sheets (fig. S9) used in this study are ~0.6 to 2.6 μm and ~1.1 nm, respectively.

**Fabrication of dual-gradient rGO/PDMAEMA hydrogel sheets**

The rGO/PDMAEMA composite hydrogel was synthesized by using UV-induced free radical generation of GO to initiate the polymerization of a mixture of DMAEMA, GO, and MBA. Briefly, DMAEMA (monomer, 0.214 ml), MBA (photocrosslinker, 20 mg), and 0.8 weight % (wt %) GO (0.4 g) were added into 0.7 ml of distilled water. After ultrasonic dispersion for about 15 min, the solution was rapidly injected into a laboratory-made mold, which consisted of two glass substrates separated by a 0.5-mm spacer. This mold was then exposed to UV light (15 W, 254 nm) for 8 hours (fig. S1). After polymerization, the as-prepared rGO/PDMAEMA hydrogel sheet was immersed in bulk deionized water for about 48 hours to remove impurities or unopolymerized monomers.

**Fabrication of single cross-linking gradient bilayer PNIPAM hydrogel sheets**

The layer with high cross-linking density (HC side) was first prepared. During this process, 0.2 to 0.4 g of NIPAM and 0.03 g of MBA were added into 1.8 ml of distilled water. After the addition of 3 μl of TEMED and 0.2 g of ammonium persulfate solution (1 wt%), the polymerization was carried out at room temperature in a sealed vessel for 4 hours. The LD layer (with low chain density of PNIPAM and low cross-linking density) was then prepared directly on the top of the HD layer using similar procedures. To reduce the chain density and the cross-linking density of PNIPAM, the amounts of NIPAM and MBA in this process were decreased to 0.1 and 0.006 g, respectively. Each layer of the PNIPAM hydrogel sheets has a thickness of 360 μm.

**Experimental procedures for the actuation of hydrogel sheets**

All the experiments of hydrogel deformation were carried out in a petri dish with a diameter of 150 mm. The temperature of the system was controlled by a thermal-magnetic heater (MS-H-Pro, Scilogen, USA). The pH of the solution was measured by a pH meter (FiveEasy Plus, METTLER TOLEDO, USA). An 808-nm laser (MDL-N-808, Changchun New Industries Optoelectronics Technology Co. Ltd, China) was used as the light source, and a power of 2 W/cm² was used in all the experiments. During the deformation, the hydrogel sheet was first pretreated under the stimuli of \( T_{25} \), IS, pH, or NIR for 10 min and then immersed in 20°C water or other stimuli for inverse snapping deformation. Note that when NIR was used for local pretreatment, each pattern was irradiated for 2 min rather than 10 min.

**Characterization**

The cross-sectional morphologies of the as-prepared hydrogel sheet were observed by SEM (JSM-6390LV, JEOL) operated at 15.0 kV. The sample for SEM imaging was prepared by fracturing the freeze-dried hydrogel in liquid nitrogen and gold sputtering on the fracture surface. To confirm the gradient structure, the cross-sectional morphologies of the samples dyed by the dilute solution of sodium fluorescein (~10⁻⁶ M) and rhodamine 6G (~10⁻⁶ M) were respectively imaged by using CLSM (Nikon A1R plus, Tokyo, Japan) with an exciting wavelength of 488 nm in the single-channel mode. Raman scattering measurements were performed using a Raman system (inVia-reflex, Renishaw) with confocal microscopy under an excitation light of 514.5 nm. Atomic force microscopy (AFM) images were taken by an atomic force microscope (Agilent Technologies Inc.) operating in tapping mode using silicon cantilevers with ~300-kHz resonance frequency. The surface chemical constitutions of the hydrogel sheets were characterized by XPS (Thermo Fisher Scientific, ESCALAB 250).

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