STIMULI-RESPONSIVE HYDROGEL MICROSPRINGS FOR MULTIPLE AND COMPLEX ACTUATION

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ABSTRACT

This paper describes stimuli-responsive hydrogel microsprings (SR springs) for realizing multiple and complex actuation. SR springs, made of double network hydrogel of p(NIPAM-co-AAc) and calcium alginate, were continuously formed by using a bevel-tip capillary. The size, pitch and cross-sectional pattern of SR springs were variable by controlling flow rate, buoyancy force and laminar flow patterns during the spring formation process. By heating single-layered or double-layered SR springs, we achieved five different types of complex spring movements: cross-sectional compression, axial compression, axial expansion, winding up and winding down. We believe that our SR springs could pave the way to various microscale chemomechanical applications such as autonomous soft machines/robots and drug release systems.

INTRODUCTION

Stimuli-responsive (SR) hydrogel is an attractive material for biocompatible soft-actuators driven by physical [1] or chemical stimuli [2]. NIPAM-based SR hydrogel has been the most commonly used one as soft-actuators for autonomous micro-robots [3], micro-valves [4] and drug release [5]. A huge variety of movements driven by shrinking/swelling of those SR hydrogel is critical to design such autonomous systems, however, the movement of SR hydrogel is limited: Only isotropic shrinkage/swell of bulk hydrogel [6] or bending movement with a bilayer hydrogel film [7] have been adopted to actuate those systems (Figure1a). Thus for realizing complex movements, a complex patterning of SR materials is necessary [8].

To achieve complex movement, we here propose microspring-shaped stimuli-responsive (SR) hydrogels with pitch variation and simple cross-sectional patterns. Our spring-shaped actuator enable us to realize various actuating movements (Figure 1b). Because a spring shape can attain large deformation according its geometry, spring shaped components are widely used in engineering such as a shape memory alloy coil actuator [9]. SR springs was fabricated by microfluidic gelation of p(NIPAM-co-AAC) and calcium alginate [10] with bevel-tip capillary [11][12]. Cross-sectional patterns of the spring can be designed by forming laminar flow patterns inside the capillary (Figure 1c). For controlling cross-sectional pattern of SR hydrogel in springs, we adjusted the direction of laminar flow pattern to the bevel-tip. For forming variable pitch springs, buoyancy force was used for adjusting the pitch.

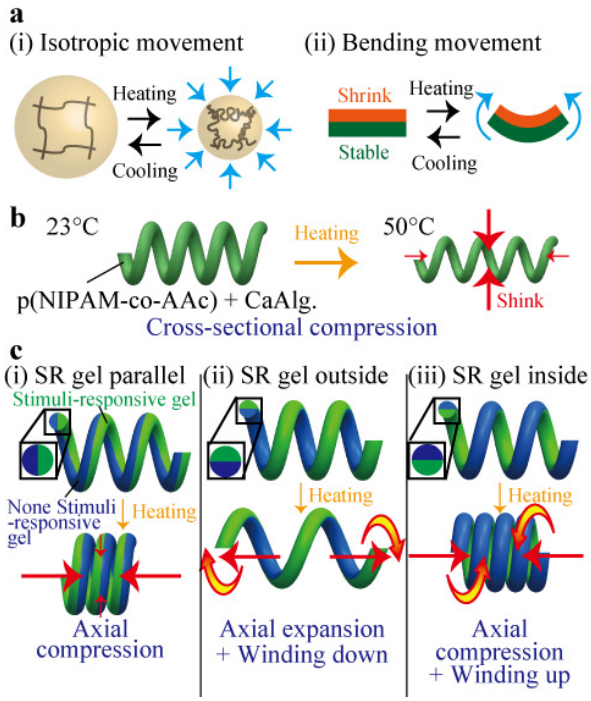

Figure 1: a) Previous stimuli-responsive (SR) hydrogel actuators: i) isotropic movement and ii) bending movement. b) Our proposal method: We proposed a simple spring-shaped actuator which was compressed in cross-sectional direction by heating. c) Three types of double-layered SR springs: i) SR gel parallel was compressed in axial direction, ii) SR gel outside deformed with combination of axial expansion and winding down, iii) SR gel inside deformed with combination of axial compression and winding up.

METHOD

Fabrication of hydrogel microspring

We prepared a bevel-tip capillary by cutting a perfluoroalkoxy (PFA) microtube (ARAM, inner diameter: 200 μm or 300 μm) The tip angle was adjusted to 20˚. The tube was bent at right angle and set horizontally to a liquid surface (Figure 2a). For forming SR springs, we prepared poly (*N*-isopropylacrylamide-*co*-acrylic acid) (p(NIPAM-co-AAC), SIGMA-ALDRICH, 741930) solution as a thermo-responsive hydrogel and sodium alginate solution (NaAlg, WAKO, 194-13321) as a rapidly gelation material. Mixing solution of p(NIPAM-co-AAC) and NaAlg rapidly gelated by forming double-network with interacting Ca^{2+} ions. By using a syringe pump (KD Scientific, LEGATO 180), 2.57% w/w p(NIPAM-co-AAc) + 0.43% w/w NaAlg solution was extruded into $CaCl_2$ solution (1 M or 2 M; WAKO, 039-00475) at constant flow rate (20 or 100 μL/min) with the bevel-tip capillary, and then SR springs were continuously fabricated upward

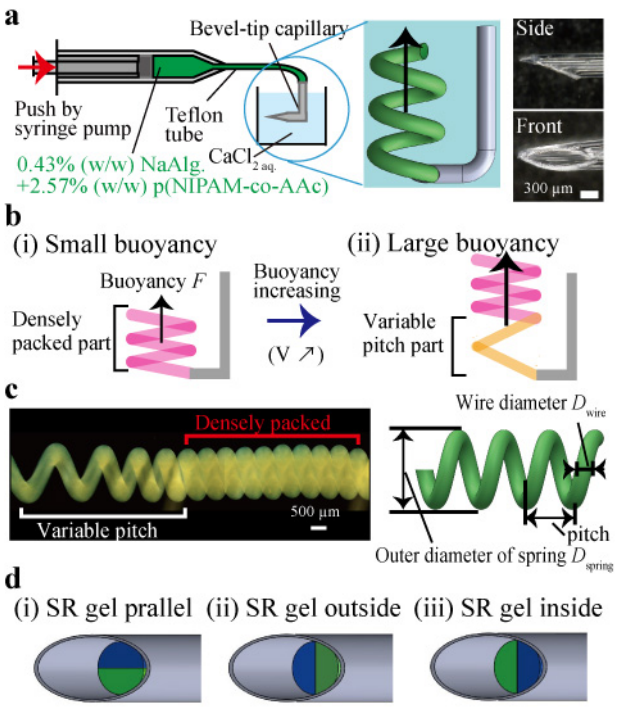

Figure 2: a) Fabrication setup: By using a syringe pump, mixture solution of p(NIPAM-co-AAc) (a stimuli-responsive material) and sodium alginate (NaAlg) solution (a rapidly gelation material) was extruded into $CaCl_2$ solution at constant flow rate with the bevel-tip capillary. b) Pitch change of springs: i) At small buoyancy, densely packed spring was formed. ii) At large buoyancy, pitch of spring was variable. c) Parameters of spring: Wire diameter D_{wire}, Outer diameter of spring D_{spring} and pitch p. d) Direction of laminar flow: i) SR gel parallel; ii) SR gel outside; iii) SR gel inside.

dragged by buoyancy force (Figure 2a, b). Size variety of hydrogel microsprings were measured by an inverted fluorescence microscope (OLYMPUS, IX73P1-22FL/PH) equipped with an image software (OLYMPUS, cellSens). A wire diameter D_{wire}, an outer diameter of spring D_{spring}, and a pitch p were evaluated (Figure 2c).

Double-layered SR springs

For forming double-layered SR springs, we used 1.5% w/w NaAlg + 1.5% w/w propylene glycol alginate (PGAlg, WAKO, 165-17415) as a non stimuli-responsive layer. The PGAlg was used for adjusting the viscosity. By using double-layered laminar flow created in the bevel-tip capillary with a Y-connector (ISIS, VFY106Y), we fabricated double-layered SR springs. By changing the direction of laminar flow pattern, three different types of double-layered SR springs (SR gel parallel spring, SR gel outside spring and SR gel inside spring) were fabricated (Figure 2d).

Stimuli-responsiveness

We confirmed the responsiveness of single-layered SR springs and three types of double-layered SR springs. Those fabricated springs were heated from 23˚C to 50˚C in 1 M $CaCl_2$ solution with a hotplate. The responsiveness of the spring was observed by a digital microscope (Keyence, VH-5500) and actuations were measured every 30 seconds for 5 minutes.

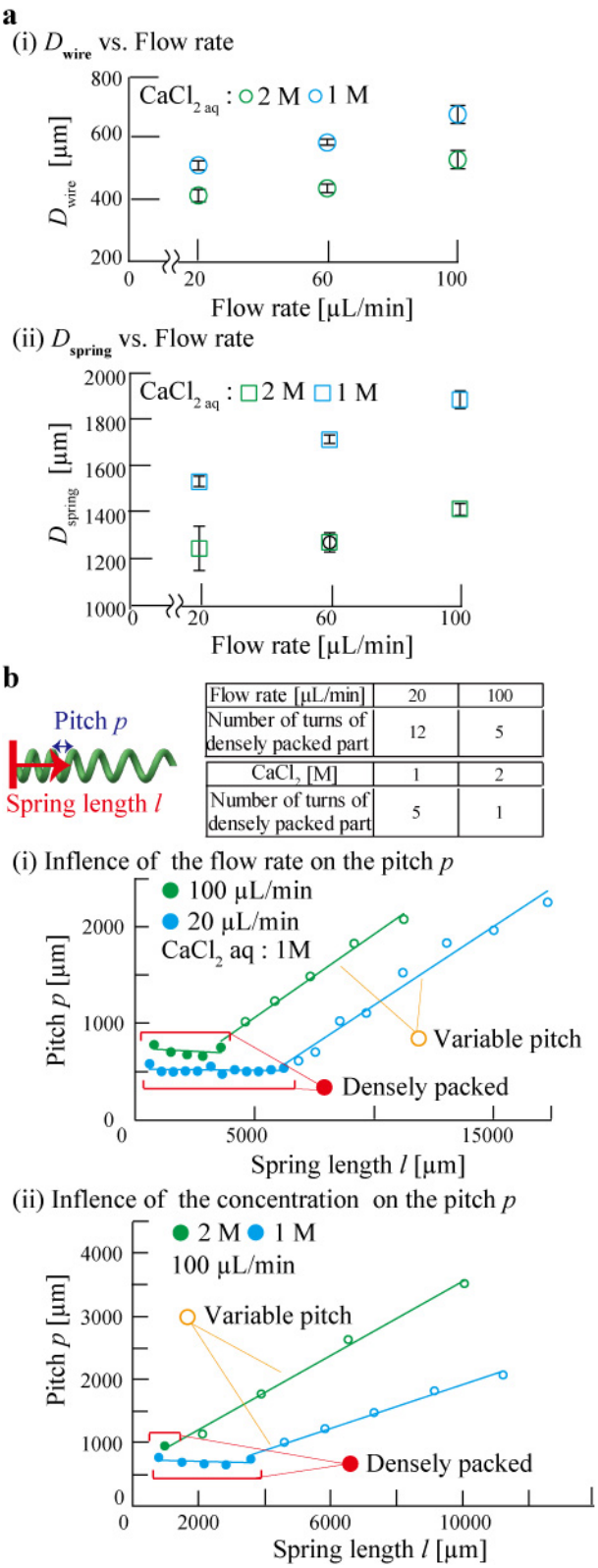

Figure 3: a) Size dependency on the flow rate and the concentration of $CaCl_2$ solution: i) D_{wire}; ii) D_{spring}. b) Degree of variable pitch of the fabricated SR springs. Filled circles show densely packed part and white circle show variable pitch part: i) influence of the flow rate, ii) influence of the concentration of $CaCl_2$ solution.

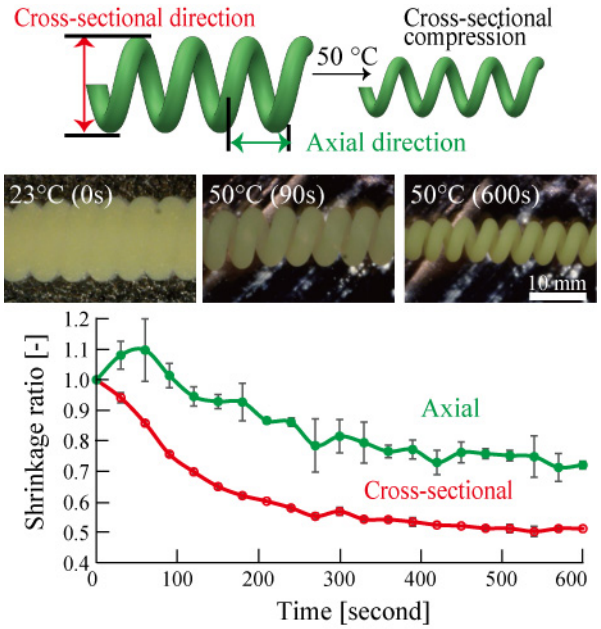

Figure 4: Actuation of a single–layered SR springs: The single-layered SR spring was mainly compressed in cross-sectional direction by heating at 50˚C.

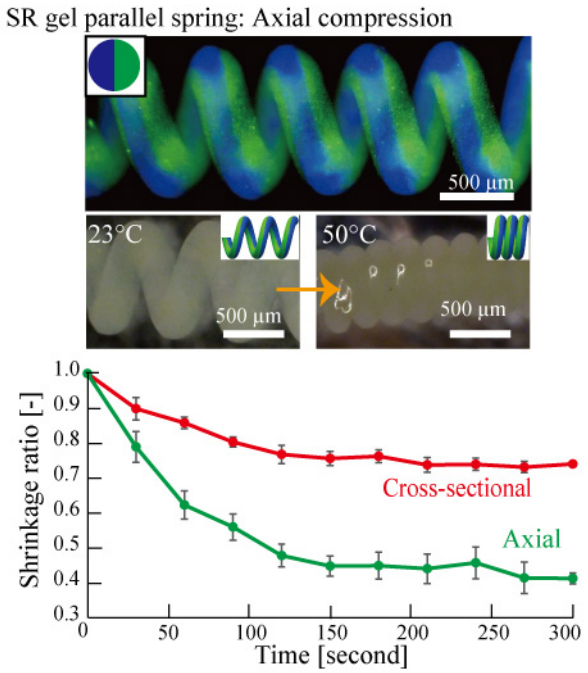

Figure 5: Actuation of a SR gel parallel spring: Fluorescent image shows the SR gel parallel spring was divided into two parts; blue part was a non-responsive layer and green part was a SR gel. The SR gel parallel spring was mainly compressed in axial direction by heating at 50˚C.

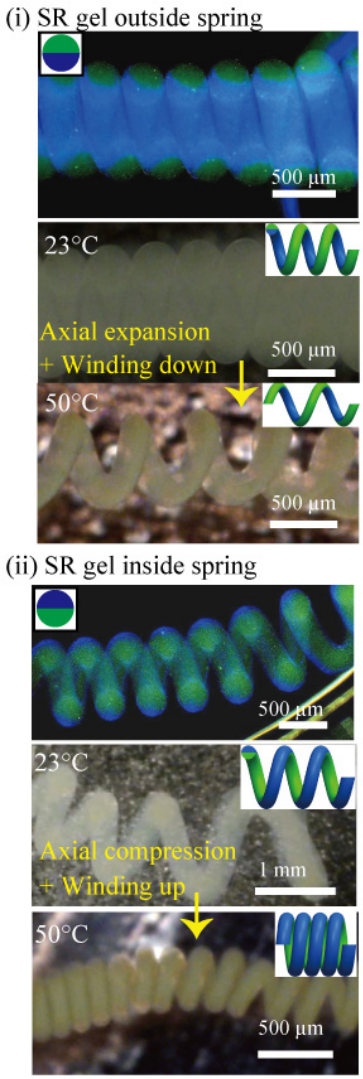

Figure 6: Actuation of a SR gel outside spring and a SR gel inside spring: (i) the SR gel outside spring was axial expansion and winding down by responding to high temperature (50˚C). (ii) the SR gel inside spring was axial compression and winding up by heating at 50˚C.

RESULTS

Size variety of hydrogel microsprings

The wire diameter D_{wire} was proportionally correlated to the flow rate and inversely correlated to the concentration of $CaCl_2$ solution (Figure 3a (i)). The outer diameter of spring D_{spring} was similar tendency (Figure 3a (ii)).

Fabricated single-layered SR springs had two different parts: densely packed part and variable pitch part (Figure 2c). It is estimated that the pitch of SR spring was firstly densely packed and when the buoyancy force exceeded a deforming force of spring, the densely packed part was transited a variable pitch part (Figure 2b). Length of the densely packed part (Figure 3b filled circles) decreased by increasing the flow rate and $CaCl_2$ concentration, because the buoyancy force of the springs also increased during the fabrication process (Figure 3b). Gradient of variable pitch was approximately same at each condition (Figure 3b). On the other hand, by comparing the length of densely packed part, it is estimated that the influence of concentration of $CaCl_2$ solution on the length of densely packed part was larger than the influence of flow rate.

Stimuli-responsiveness

By heating at 50 ˚C, the single-layered SR spring was mainly compressed in cross-sectional direction with shrinkage ratio 0.5 (ratio of axial direction compression: 0.7) (Figure 4). The shrinkage of the single-layered SR spring converged in approximately 300 seconds.

A fluorescent image of the double-layered SR springs

shows that spring was clearly divided into two parts (green part was SR gel layer; blue part is non stimuli-responsive layer; Figure 5 top, Figure 6 (i), (ii) top). In contrast to the result of the single-layered SR spring, the SR gel parallel spring was mainly compressed in axial direction with shrinkage ratio 0.4 (ratio of cross-sectional direction: 0.8) (Figure 5). These two results indicate that the direction of shrinkage can be controlled by patterning our SR springs.

Furthermore, although the SR gel outside spring behaved as axial expansion with winding down (the number of turns:19→14) (Figure 6 (i)), the SR gel inside spring behaved as axial compression with winding up (the number of turns:12→19) (Figure 6 (ii)). These results indicate that torsion motions in both direction can be realized by patterning our SR springs.

CONLCUSION

We successfully fabricated SR springs with two different parts: densely packed part and variable pitch part. The length of densely packed part was controlled by mainly concentration of $CaCl_2$ solution and by slightly the flow rate. Above these results, by cutting out a specific part from the hydrogel microsprings, any pitch spring could be obtained. Shape parameters of the springs, D_{wire} and D_{spring}, were also controlled by the concentration of $CaCl_2$ solution and the flow rate.

We succeeded in the single-layered SR gel spring and three types double-layered SR gel springs. By heating those springs, we achieved five different types of complex spring movements: cross-sectional compression, axial compression, axial expansion, winding up and winding down.

It is expected that various multiple complex actuation could be realized by other complex compartmentalization [13] and encapsulating functional materials [14] such as other stimulus responsive hydrogel [15][16] or cells [17][18]. The success of multiple complex movements would open new avenues to various microscale biochemical applications such as autonomous soft robots and drug release systems.

ACKNOWLEDGEMENTS

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