

Self-Helical Fiber for Glucose-Responsive Artificial Muscle

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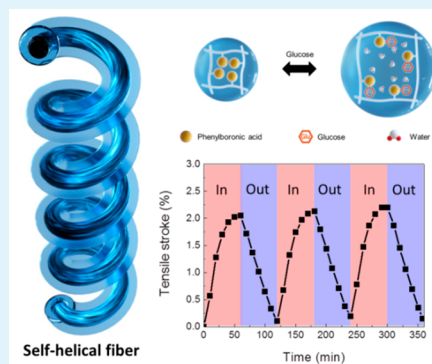
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Supporting Information

ABSTRACT: A helical configuration confers a great advantage in artificial muscle due to great movement potential. However, most helical fibers are exposed to a high temperature to produce the coiled helical structure. Hence, thermoset polymer-composed hydrogels are difficult to fabricate as helical fibers due to their thermal degeneration. Here, we describe a self-helical hydrogel fiber that is produced without thermal exposure as a glucose-responsive artificial muscle. The sheath–core fiber was spontaneously transformed into the helical structure during the swelling state by balancing the forces between the untwisting force of the twisted nylon fiber core and the recovery force of the hydrogel sheath. To induce controllable actuation, we also applied a reversible interaction between phenylboronic acid and glucose to the self-helical hydrogel. Consequently, the maximum tensile stroke was 2.3%, and the performance was six times greater than that of the nonhelical fiber. The fiber also exhibited tensile stroke with load and a maximum work density of 130 kJ/m³. Furthermore, we showed a reversible tensile stroke in response to the change in glucose level. Therefore, these results indicate that the self-helical hydrogel fiber has a high potential for use in artificial muscles, glucose sensors, and drug delivery systems.

KEYWORDS: actuator, fiber, helical, glucose, artificial muscle



1. INTRODUCTION

Artificial muscle is currently needed for application in biological systems that require actuation, such as humanoid robots, prosthetic limbs, exoskeletons, and microfluidic actuators.^{1–12} Especially, fiber-shaped artificial muscles have great potential for applications due to multiple motions such as bending, twisting, and tensile movement.^{13–21}

To enhance tensile actuation, the helical structure has been attempted in artificial muscle because its helical structure has a high potential movement. In fact, several studies demonstrated that the tensile action is amplified by mandrel structure in which straight fiber structure is transformed into a helical structure like tendril structure.^{12–14,16} However, the helical configuration should be inevitably exposed to high temperature to form its structure. This thermal process can change the inherent properties of the polymer, which limits the use of helical structure in thermoset polymers such as hydrogels.

In the present study, we report self-helical fibers that are spontaneously formed by the force balance between the untwisting force of the core and the recovery force of the sheath without thermal treatment. Furthermore, we enable the reversible actuation of self-helical fibers in response to glucose levels as a bioartificial muscle.

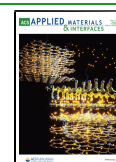
To fabricate self-helical fibers, a hydrogel was first coated onto a twisted nylon. During the swelling process in an aqueous environment, the hydrogel-coated nylon fiber was spontaneously transformed into the helical structure. The self-

helical formation is based on the balance between the untwisting force of the core (nylon) and the recovery force of the sheath (hydrogel). To induce controllable actuation, we further attempted to apply a reversible interaction between phenylboronic acid and glucose to this self-helical hydrogel. As a result, the self-helical fiber exhibited a reversible tensile stroke in direct proportion to glucose concentration, and the performance was dependent on the spring index, that is, the ratio of mean coil diameter to wire diameter in the helical structure. The maximum tensile stroke was 2.3%, and the performance was six times greater than that of the nonhelical fiber. Moreover, self-helical fibers exhibited tensile stroke with load and a maximum work density of 130 kJ/m³. The tensile stroke was dependent on the changes in glucose concentration. Therefore, these results indicate that self-helical fibers offer excellent potential to be used in artificial muscles, glucose sensors, and drug delivery systems.

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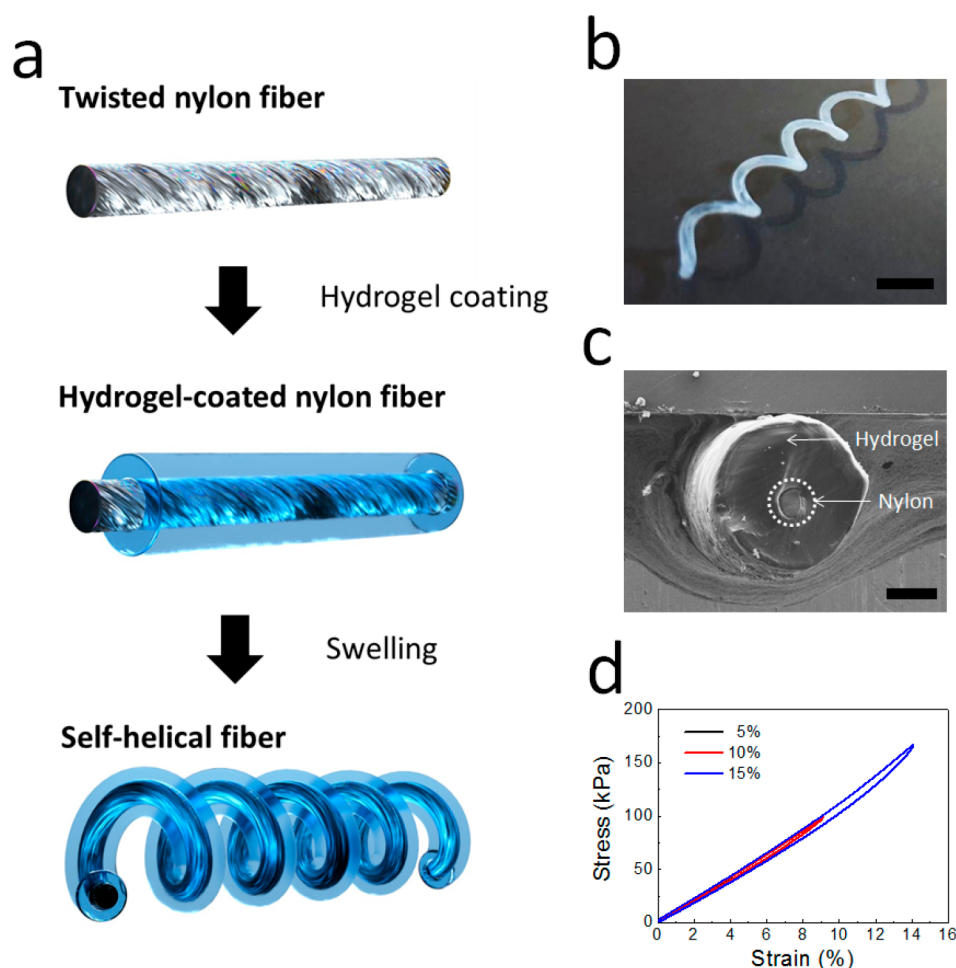


Figure 1. Fabrication and structure of the self-helical artificial muscle. (a) The fabrication process for a glucose-responsive artificial muscle using the self-helical fiber. (b) Photographic image of the final fabricated self-helical fiber (scale bar: 5 mm). The diameters of the nylon fiber core and dried hydrogel sheath were 130 and 760 μm , respectively. (c) Scanning electron microscope (SEM) image showing a cross-sectional image of the self-helical fiber (scale bar: 200 μm). (d) The stress–strain curve of the self-helical fiber.

2. RESULT AND DISCUSSION

The fabrication process of the self-helical fiber is shown in Figure 1a. The nylon fiber was twisted with 300 turns/m in clockwise direction. The two ends of the twisted nylon fiber were tethered to prevent untwisting during the fabrication process. Thereafter, the hydrogel was coated onto the twisted nylon fiber. Consequently, the twisted nylon fiber was used as the core, and the hydrogel was used as the sheath for the sheath–core structure. The sheath–core fiber was formed without untwisting. When the sheath–core fiber was swelled in the phosphate-buffered saline (PBS) solution, the swelling ratio of 1.8 was nearly saturated after 60 min (Supporting Figure S1). During swelling, the hydrogel-coated nylon fiber was spontaneously transformed from a straight line into a self-helical fiber in counterclockwise direction and the length decreased. (Supporting Movie 1).

As swelling of the hydrogel sheath proceeded, the modulus of the hydrogel decreased. The core untwisted and the sheath twisted as the equilibrium between the core and sheath forces was broken. After such untwisting of the core, the untwisting force of the core and the recovery force of the sheath reached equilibrium. Therefore, twisting of the sheath resulted in spontaneous transformation into a helical structure to minimize the strain energy. As shown in Figure 1b, the self-

helical fiber was formed in free-standing fashion without two-end tethering.

The cross-sectional morphology of the self-helical fiber is shown in the SEM image in Figure 1c. The sheath of hydrogel (diameter of 760 μm) was a circular structure, and the nylon fiber (diameter of 130 μm) was located in the center of the self-helical fiber. In addition, it seems that the core nylon and sheath hydrogel tightly adhered sufficiently to transfer the torsional movement of the nylon fiber to the sheath. The mechanical property of the self-helical fiber with swelling state is shown in Figure 1d. The strain–stress curve shows that the stress linearly increased to 170 kPa with strain at 15% and returned to its initial state when the strain decreased. Residual deformation is less than 0.07%, and the hysteresis of the cycle showed 8.63% energy loss. Therefore, these findings indicate that the structure of the self-helical fiber is stable. The balance between the two forces of the core and sheath of the self-helical fiber can be continuously maintained like a spring.

Because a self-helical structure is controlled by the untwisting and recovery forces of the core and sheath, there are two factors that determine the coiling property.^{13,16} First, it is possible that the relative diameter of a coil has a critical role in determining the tensile stroke and stress of the fiber, as shown in Figure 2a. The coil spring index (C), which is the average coil diameter (D) divided by the fiber diameter (d),

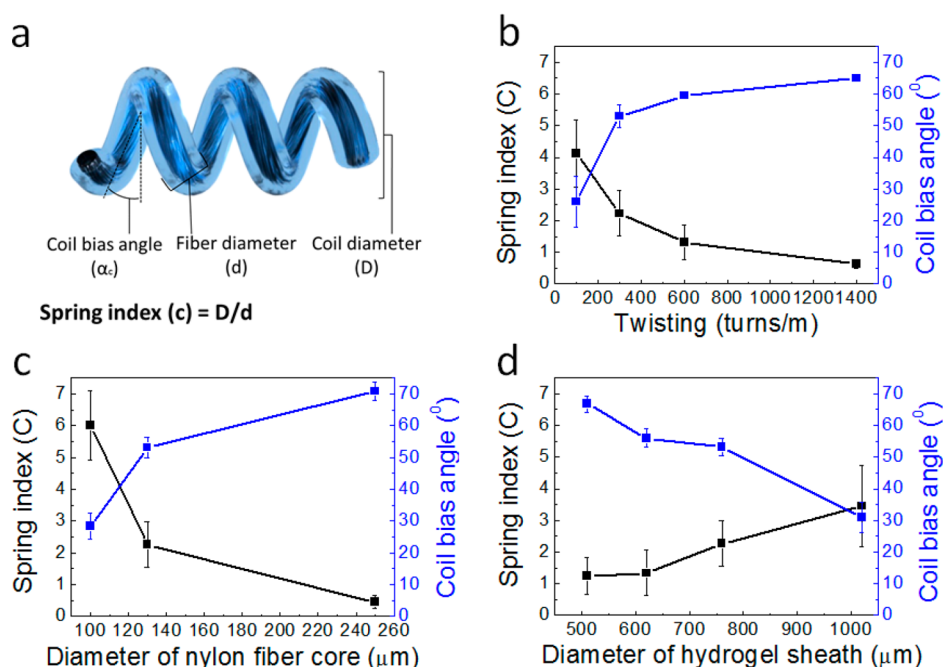


Figure 2. Self-helical fiber structure was controlled by the untwisting force of the nylon fiber core and recovery force of the hydrogel sheath. (a) Schematic illustration of the self-helical fiber structure. (b) The relationship between the spring index as well as the coil bias angle and the twisting of the nylon fiber core. (c) The relationship between the spring index as well as the coil bias angle and the diameter of the nylon fiber core. (d) The relationship between the spring index as well as the coil bias angle and the diameter of the hydrogel sheath.

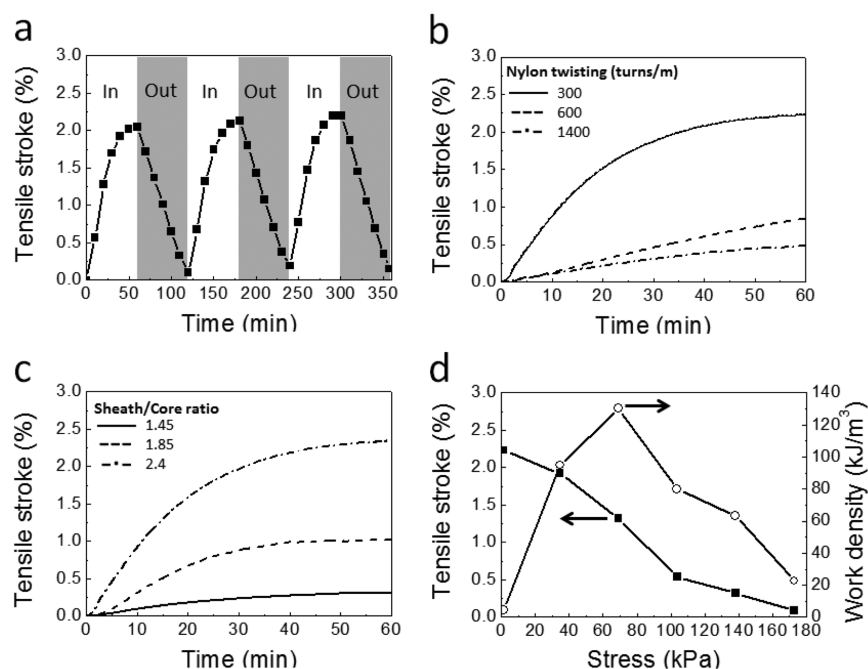


Figure 3. Tensile stroke of the self-helical fiber. (a) Reversible tensile stroke of the glucose-responsive self-helical fiber with the change in glucose concentration. (b,c) Comparison of the tensile stroke of the self-helical fiber structure: the structure was controlled by (b) twisting of the nylon fiber core and (c) the sheath/core ratio (the ratio of sheath thickness to interior core diameter). (d) The relationship between the tensile stroke (■, solid square) as well as the contractile work density (○, empty circle) versus the glucose concentration when it was reversibly changed. The 130 μm diameter nylon fiber core and 760 μm diameter hydrogel sheath were used.

was used to determine the scale-independent quantity. Second, the coiling direction could be significant for tensile actuation. The coil bias angle (α_c), which defines the degree of coil fiber, determines the coil direction.

As shown in Figure 2b, the spring index and coil bias angle can be controlled by the number of turns in nylon core fiber.

To verify the influence on the untwisting force of nylon fiber, we compared the spring index and bias angle according to twisting turns of the nylon core. With increasing the number of twisting turns of the nylon core from 100 to 1400 turns/m at a core diameter of 130 μm and dried sheath diameter of 760 μm, the spring index decreased from 4.1 to 0.6 and the bias angle

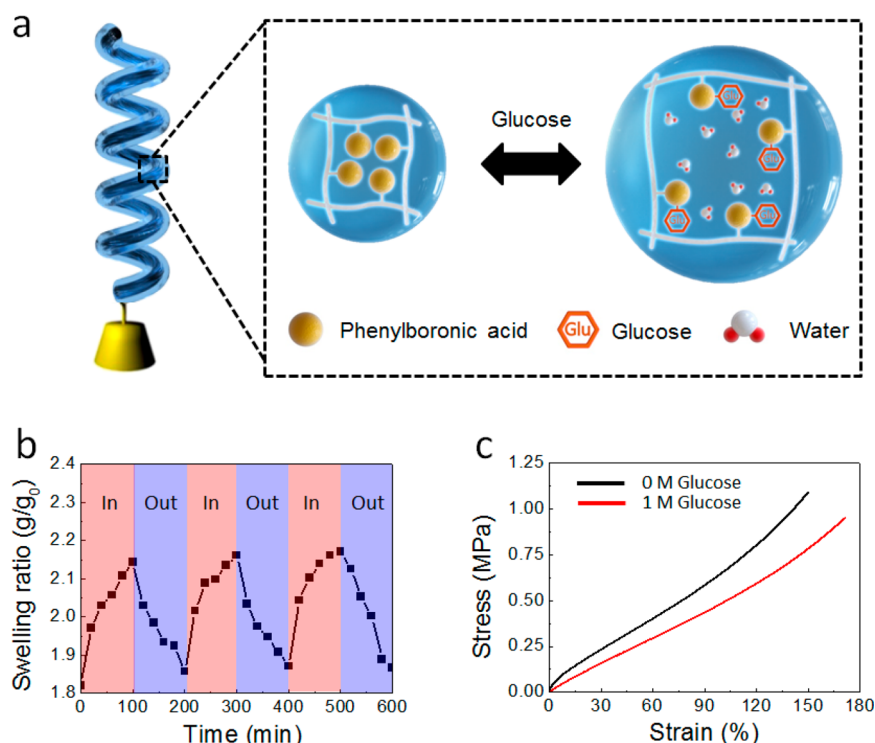


Figure 4. Actuation mechanism of the glucose-responsive self-helical actuator. (a) Schematic illustration of the tensile actuation mechanism dependent on glucose concentration. The swelling ratio of the hydrogel fiber was dependent on glucose concentration because phenylboronic acid was bonded to glucose. (b) Reversible swelling ratio change of the phenylboronic acid-based hydrogel with glucose concentration. (c) Strain–stress curve of the hydrogel fiber with 0 and 1 M glucose concentrations.

conversely increased from 26° to 65° . Next, we compared the spring index and bias angle according to the diameter of the nylon core. When the diameter of the core increased from 100 to $250\ \mu\text{m}$ with core twisting of 300 turns/m and sheath diameter $760\ \mu\text{m}$, the spring index decreased from 6 to 0.4 and the coil bias angle conversely increased from 28° to 71° . These findings indicate that the untwisting force of the core fiber can be regulated by the number of twists and the diameter of the core fiber. Therefore, the increase in untwisting force of the core fiber leads to more twisting in the sheath and an increasing normalized coil number, that is, the number of coils divided by the initial fiber length (Supporting Figures S2 and S3). As a result, increasing the untwisting force of the core fiber gives rise to a decreased spring index and an increased bias angle due to a large number of coils relative to length.

Moreover, the recovery force of the hydrogel sheath can influence the morphology of the self-helical fiber. To verify this, the spring index and bias angle were measured according to the diameter of the sheath with 300 turns/m twisting and a core diameter of $130\ \mu\text{m}$ (Figure 2d). Consequently, the sheath diameter's increment leads to the increase of the spring index from 1.2 to 3.4 and a decrease of the bias angle from 67° to 31° . Therefore, the increased recovery force of the hydrogel sheath caused a decreased normalized coil number (Supporting Figure S4). As a result, increasing the recovery force of the sheath fiber gives rise to an increased spring index and a decreased bias angle due to preventing the untwisting of the core. Also, the strain energy, by twisting, increased with sheath thickness, and it shows that the coil gets closer to linear as the sheath thickness decreases.

To provide biological applications of the self-helical hydrogel to artificial muscle, we further fabricated the

glucose-responsive artificial muscle. Because phenylboronic acid has been known to reversibly interact with glucose, we integrated phenylboronic acid into the acrylamide hydrogel sheath for glucose-responsive actuation. FT-IR spectra of the acrylamide, vinylphenylboronic acid, and acrylamide/vinylphenylboronic acid composite of the hydrogel sheath determined that vinylphenylboronic acid was well integrated into the acrylamide hydrogel (Supporting Figure S5). In the composite polymer, there were several peaks as follows: the peak at $3434\ \text{cm}^{-1}$ (assigned to the $-\text{NH}_2$ group²²), at $1419\ \text{cm}^{-1}$ (assigned to the C–H bending vibration on the benzene ring²²), and at $1337\ \text{cm}^{-1}$ (assigned to the B–O stretching vibration²³).

As shown in Figure 3a, the glucose-responsive fiber showed contracted actuation of approximately 2.3% when the glucose concentration increased from 0 to 1 M. Conversely, the fiber recovered to the initial state when the concentration decreased from 1 to 0 M (Supporting Movie 2). This reversibility of the actuation was stable during three cycles. Importantly, the self-helical fiber exhibited a contraction six times greater than that of the nonhelical fiber (tensile stroke of 0.4%) (Supporting Figure S6). Therefore, it is possible to induce a high tensile stroke with low force compared to straight fiber.

Moreover, we compared the tensile stroke for the self-helical fiber according to twisting of the nylon fiber core (Figure 3b). The diameters of the core and sheath were constant during this experiment. As a result, the 2.3% tensile stroke of the self-helical fiber with 300 turns/m was approximately 2.9 times greater than the twisting fiber with 600 turns/m (tensile stroke of 0.8%), and it was also approximately 4.6 times greater than the twisting fiber with 1400 turns/m (tensile stroke of 0.5%). Furthermore, we tested the tensile stroke of self-helical fiber

according to the spring index, which was previously known to regulate the tensile stroke.^{13,16} Consistently, the spring index decreased with increased untwisting force, as shown in Figure 2, indicating that the tensile stroke decreased with increased twisting of the core.

Next, Figure 3c indicates the tensile stroke for a self-helical fiber according to the sheath/core ratio (the ratio of the sheath thickness to the interior core diameter). When the core fiber's turning number was constant, the maximum tensile stroke of 2.3% was observed with a sheath/core ratio of 2.4. The tensile stroke was 4.6 times greater than the ratio of 1.85 (tensile stroke of 1.0%) and 5.8 times greater than the ratio of 1.45 (tensile stroke of 0.4%). With an increasing sheath/core ratio, the recovery force of the sheath was higher than the untwisting force of the core. Therefore, the spring index and the tensile stroke are dependent on the sheath/core ratio. To verify the effect of the self-helical fiber as artificial muscle, we further tested the contractile work density with respect to the applied load. The maximum work density of 130 kJ/m³ was observed at 90 kPa stress for a self-helical fiber (Figure 3d). In addition, the tensile stroke of the self-helical fiber was dependent on the low glucose concentration from 2 to 10 mM, as shown in Supporting Figure S7, and the results support the above mechanism of tensile actuation based on glucose.

In the self-helical fiber, it is likely that glucose causes the change of the modulus of the hydrogel sheath (Figure 4a). In fact, the swelling of the hydrogel sheath is positively proportional to the glucose concentration. As previously reported,^{18,24,25} glucose interacts with the phenylboronic acid in the hydrogel sheath, and such interaction causes the neutral phenylboronic acid to be charged, which increases the hydrophilicity of the hydrogel's network, leading to swelling by water absorption. Consequently, the swelling ratio of the hydrogel increased from 1.8 to 2.1 when the glucose concentration was increased from 0 to 1 M (Figure 4b). Conversely, the swelling ratio returned to the initial value of 1.8 when the glucose concentration decreased from 1 to 0 M, indicating that the swelling ratio is dependent on glucose concentration. Meanwhile, the reversible change of swelling ratio responding to glucose concentration can also affect the sheath modulus. Thus, we measured the strain–stress curves, showing the mechanical property of the hydrogel with glucose concentration (Figure 4c). The average Young's modulus was 0.73 MPa with 0 M glucose and 0.56 MPa with 1 M glucose. When the modulus of the hydrogel is decreased by glucose, the untwisting force of the core is greater than the recovery force of the sheath, which can ultimately induce twisting of the hydrogel.¹⁴ In contrast, reducing the glucose concentration causes the modulus of the hydrogel to increase, which returns the self-helical fiber to its initial state. Therefore, these findings indicate that glucose-mediated tensile actuation is regulated by the change of the hydrogel's modulus through water influx or efflux based on the hydrophilicity in the sheath hydrogel.

3. CONCLUSION

The present study shows that a self-helical fiber is spontaneously formed by the equilibrium that is established between the untwisting force of the twisted core fiber and the recovery force of the hydrogel sheath. The structure was reversibly controllable by the untwisting and recovery forces. The maximum tensile stroke was 2.3% and was six times higher than that of the nonhelical fiber. The fiber also showed tensile stroke with load and maximum work density of 130 kJ/m³,

which was dependent on the change of glucose concentration. Overall, these results indicate that the self-helical fiber has a high potential to be applicable to artificial muscles, glucose sensors, and drug delivery systems.

4. EXPERIMENTAL METHODS

Fabrication Process of Self-Helical Fiber. A self-helical artificial muscle was fabricated using a commercial nylon fiber (Coats & Clark, U.S.A.) as the core and a phenylboronic acid-containing polymer network (PA-BA) as the sheath. Acrylamide, 4-vinylphenylboronic acid, *N,N'*-methylenebis(acrylamide), DMSO, Irgacure 2959 photoinitiator, and D-(+)-glucose were purchased from Sigma-Aldrich. The sheath was formed on the nylon fiber core by ultraviolet polymerization where Irgacure 2959 serves as the photoinitiator and *N,N'*-methylenebis(acrylamide) serves as the cross-linker. The monomer solution contained acrylamide (78 mol %), 4-vinylphenylboronic acid (20 mol %), and *N,N'*-methylenebis(acrylamide) (2 mol %) in DMSO. Irgacure 2959 was added at a concentration of 2 wt % of the total monomer content. The DMSO concentration in the final solution was 44 wt %. This monomer solution was coated on the nylon fiber using a silicon tube, and photopolymerization was initiated by irradiating the solution for 20 min with 365 nm ultraviolet light (Dymax, Bluewave 200). The temperature during fabrication was 23 °C.

Characterization. The morphology of the self-helical fiber was determined by SEM (SEM-S4700 microscope, Hitachi, Japan). The actuation movie of the glucose responsive actuator was determined by digital microscope (VHX_950F, KEYENCE, Korea). The elastic properties were measured using a universal testing machine (UTM, INSTRON 5966, INSTRON, Massachusetts, U.S.A.).

The Calculation of the Swelling Ratio. The swelling ratio was calculated by the following equation:

$$\text{Swelling ratio} = \frac{W_w - W_d}{W_d}$$

where W_w is the weight of the wet hydrogel and W_d is the weight of the dry hydrogel.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsami.0c03120>.

Swelling ratio with time, normalized number of coils, FT-IR, tensile stroke (PDF)

Self-helical fiber swollen in 20 mM PBS at 28 °C (MP4)

Tensile actuation of glucose responsive artificial muscle fully swollen in 20 mM at 28 °C about 24 h (MP4)

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Author Contributions

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Notes

The authors declare no competing financial interest.

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