Supporting Information

Wavelength-Selective and Rebound-able bimorph Photoactuator Driven by Dynamic Mass Transport Process

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Experiment Detail

Synthesis of Au Nanocrystals (AuNCs): Au nanoparticles (AuNPs) and nanorods (AuNRs) with different aspect ratio are synthesized via seeding growth methods, which have been widely studied. The product is centrifuged at 8000rpm for 20min and re-dispersed in DI water of same volume.

Modification of AuNCs: The synthesized AuNCs are then modified with sulfhydryl-terminated polyethylene glycol (mPEG-SH) to avoid the aggregation of CTAB stabilized AuNCs when solvent is volatilized. 1mL fresh prepared mPEG-SH (5mg/mL) is mildly added into 10mL AuNCs solution, and the mixture is kept in dark overnight for sufficient replacement. Then the spare CTAB and PEG are removed with centrifugation. The achieved precipitates are re-dispersed in DI water of same volume. The mPEG-SH modified AuNCs can be stored for several months.

Fabrication of the bimorph photoactuators: In each film, the amount of AuNCs is carefully tuned, so that the achieve films present equal peak absorbance. Appropriate amount of mPEG-SH modified AuNCs solution is mixed with chitosan powder (15mg, MW 400kDa), 2.5mL DI water, 0.1mL acetic acid and 1ml ethanol. The mixture is stirred overnight until it turns to uniform solution. Then the solution is casted on a 75mm×25mm glass slide, and heated up to 40°C to desiccate. Existence of ethanol decrease the solution surface tension, thus the solution will obtain better wetting capability on glass substrate. The dry CS films present pink, blue

and grey, respectively. Then PDMS (base agent and cure agent are in 10:1 mass ratio) is casted onto the CS layer via spin coating method (3000rpm×16min, about 10mg) and cured at 100°C for 30min.

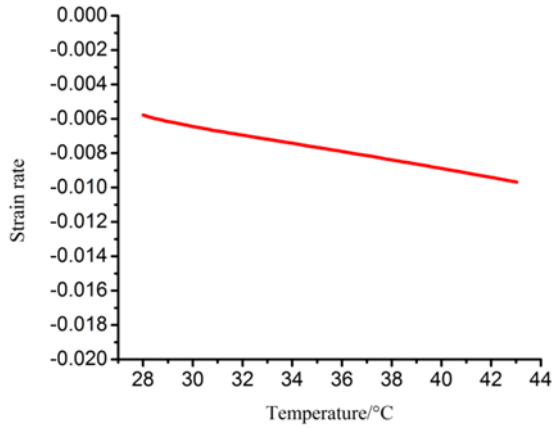

Figure S1. Thermal-mechanical curve of chitosan film among 28-43°C.

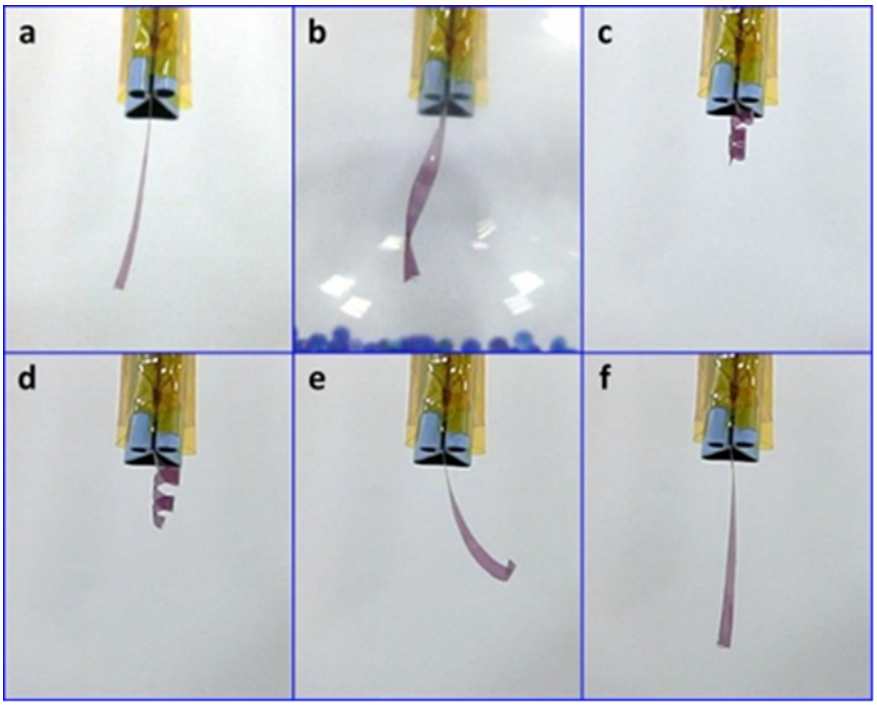

Figure S2. a. Strip **1** is placed in the air. **b.** The strip slightly twists anticlockwise when put into the dry air. **c.** The strip rapidly and strongly twists clockwise within 1s after put into the air. **d.** After 8s. **e.** After 15s. **f.** After 22s, the strip completely recovers its initial state.

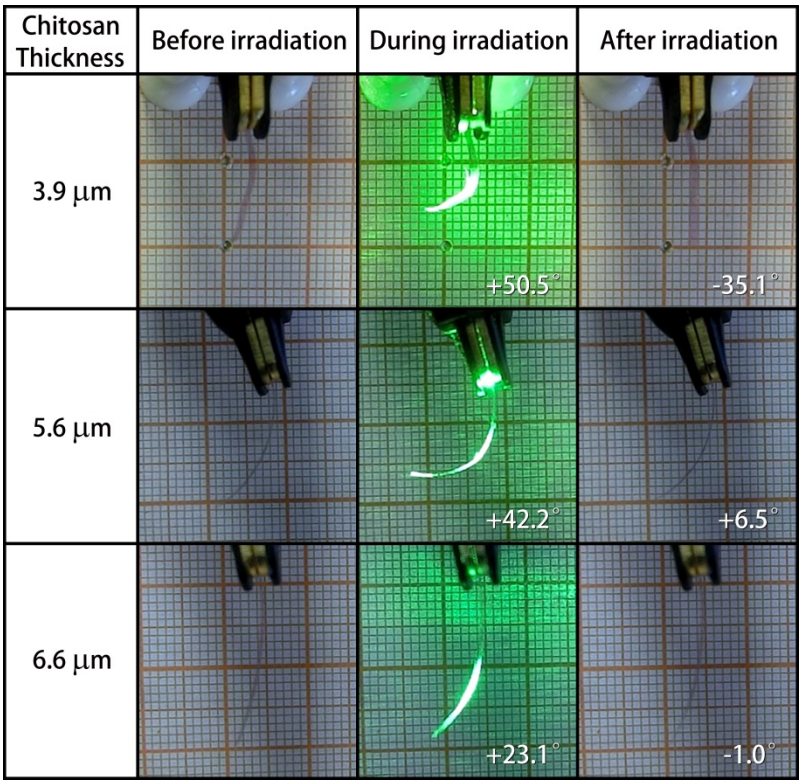

Figure S3. Chitosan thickness influence on actuation performance. When chitosan layer gains thickness, the during-irradiation bending gradually decreases and the after-irradiation rebound sharply disappears.

Numerical simulation. The numeric simulation is obtaind with finite elements method and is based on below reasonable assumptions:

1. Surface water adsorption rate is linearly related to the surface humidity.

2. Polymer size is linearly related to low humidity.

3. CS matrix is governed by the Hooke's law within the deformation range.

The CS layer is divided into 31 layer elements, including a boundary layer (Layer 0) and 30 inner layers (Layer 1~30) of same thickness. The initial and balance humidities are H_{max} and H_{min} respectively. For the boundary layer, released/reabsorbed rate is in proportion to the local humidity H and humidity difference between H and H_{min}, while the diffusion rate from

ajacent layer to boundary layer is in proportion to local humidity H. So the humidity of boundary layer at time *t* is given by

$$H_{0,t} = H_{0,t-\Delta t} - H_{0,t-\Delta t}(H_{0,t-\Delta t} - H_{min})K + H_{1,t-\Delta t}(H_{1,t-\Delta t} - H_{0,t-\Delta t})D$$

(1)

In eq 1, *K* is the sorption rate constant, *D* is Fickian diffusion coefficient. For inner layers 1~29, the recursion formula of humidity is given by

$$H_{i,t} = H_{i,t-\Delta t} - H_{i,t-\Delta t}(H_{i,t-\Delta t} - H_{i-1,t-\Delta t})D + H_{i+1,t-\Delta t}(H_{i+1,t-\Delta t} - H_{i,t-\Delta t})D$$

(2)

In eq 2, *i*=1, 2, 3… 29. While for the layer 30, the humidity is given by

$$H_{30,t} = H_{30,t-\Delta t} - H_{30,t-\Delta t}(H_{30,t-\Delta t} - H_{29,t-\Delta t})D$$

(3)

The simulation of rebound bending can be obtained with similar recursion formulas. The humidity distribution reflects the stress, or strain without restraint (The ajacent layers will hinder the deformation). The real strain-depth curve should be a straight line

$$L_i = L_0\left(1 + \frac{d}{30R}i\right)$$

(4)

In eq 4, L_i is the real length of layer *i* (*i*=0, 1, 2… 30); *R* is the curvature radius of the bending strip; *d* is the thickness of CS layer. The elastic potential energy is given by

$$E = Y\sum_{i=0}^{30}\left[L_d(1 + aH_i) - L_i\right]^2$$

(5)

In eq 5, L_d is the chitosan length at dry state; a is the hydroscopic expansion coefficient. According to the lowest energy principle, only when line L_i is the fitting line of $L_d(1+aH_i)$, the elastic potential energy can be minimized. The overall bending angle is given by

$$\theta = \frac{L_0 30k}{L_d \ d}$$

(6)

In eq 6, k is the fitting slope of humidity distribution curve. Because $L_0/L_d \approx 1$, the bending angle can be simplified as

$$\theta = \frac{30}{d}k$$

(7)

Thus the slope-time curve can represent the angle-time relation. Set K=0.3, D=0.5, H_{max}=1, H_{min}=0.1, then it obtains the curve in figure 4c, which is very similar to experimental results.