

Supporting Information

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Bioinspired Multi-Stimuli Responsive Actuators with Synergistic Color- and Morphing-Change Abilities

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Figure S1. SEM cross-section images of the glucose-assisted assembled CNCs film. The value of p/2 for the CNCs/glucose composite membrane is 201±7 nm.



Figure S2. Digital pictures of the cholesteric LC film as the viewing angle increases from 90° to 120° .



Figure S3. The alterable structure color of the cholesteric LC can be characterized by colorimeter at different view angles (90°, 100° , 110° , 120°).



Figure S4. Digital pictures of an actuator reversibly bending in the vertical orientation when humidity is switched on and off. The vertical bending angle of the actuator increases from 0° to 144.2° as the moisture absorption time increases from 0 to 14 s, and then it takes 71 s to revert to its original position without humidifying.



Figure S5. Humidity-actuating performance of the actuator under different original ambient relative humidity (RH) (70%, 50%, 30%). Temperature and the applied additional moisture remain constant.



Figure S6. Digital pictures of a) AgNPs aqueous dispersion and b) PU/AgNPs mixture.



Figure S7. SEM images of the sliver nanoparticles. The value of diameter for the AgNPs is 221±25 nm.



Figure S8. Schematic depiction of the experimental setup used for testing the actuation force of the actuating film.

To confirm photo-induced mechanical stress, we firstly utilize electromagnetic gauge transducer to measure the light-generated force of the actuator films by applying NIR light (808 nm) to a strip-shaped sample $(20 \times 5 \times 0.153 \text{ mm}^3)$ at room temperature. The mechanical force generated by the sample film can be monitored from the following formula: F = mg, where *m* is the real-time mass on indicator of the balance, *g* is the acceleration of gravity. Then, the mechanical forces are converted into stress followed by the following formula: P = F/S, where *S* is cross sectional area of the strip-shaped film.



Figure S9. a) Humidity actuation angle and b) actuation stress as a function of time. The cycle of force up and down with humidity on and off, respectively, can be repeated for 3 times without damping.



Figure S10. a) Humidity- and b) NIR-actuating performance of the actuator before and after subjected to 5,000 bending cycles.



Figure S11. The temperature variation during the NIR-actuation process.



Figure S12. Digital images for structural color change of the CNCs/glucose composite under relative humidity 90%.

 Table S1. Comparison of color changing performance of the recently reported materials.

Ref.	Materials	Conditions	Time	Colors	Other functions
[1]	Eu ³⁺ -poly(N-isopropylacrylamide) - potassium 6-acrylamidopicolinate	Tb ³⁺	1800s	Red – Green	Actuation
[2]	zinc-ion-intercalated layered polydiacetylene- cellulose paper (T-paper)	40 °C to 140 °C	20 s	Blue – Purple	Actuation
[2]	PI tape-T-paper-MXene/ graphene	40 °C to 140 °C	20 s	Green – Orange	Actuation, Self-sensing
[3]	Sr ₂ P ₂ O ₇ :Eu,Y	Strain: 20 %-100 %	/	Blue – Purple	/
[4]	Au nanoparticles- poly(N-isopropylacrylamide)	24 °C to 50 °C	/	Red - Grayish violet	/

[5]	single-wall carbon nanotube-line elastomer	NIR irradiation	15 s	Cyan – Blue	Actuation
[6]	liquid crystalline networks-PA	Humidity	6 s	Dark green – Green	Actuation
[7]	P(AAm-co-AAc) - poly(N-isopropylacrylamide)	0 °C to 50 °C	80 s	Green – Red	Actuation
		NIR irradiation	30 - 240 s	Green – Red	Actuation
[8]	Fe ₃ O ₄ @PVP-CNCs- poly(N-isopropylacrylamide)	10 °C to 35 °C	/	Green – Yellow – Red	/
This work	CNCs@glucose-PU/AgNPs	Humidity	9 s	Blue – Green – Brown	Actuation
		NIR irradiation	16 s	Brown – Green – Blue	Actuation

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