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Supporting Information

Bio-Inspired Cellulose Nanocrystal-Based Nanocomposite Photonic Film with Hyper-Reflection and Humidity-Responsive Actuator properties

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1 bending behavior analysis of the sandwiched structure nanocomposite film

As shown in Figure S1, we assume that the inner nonswollen layer keep the original film-free length, and the relationship of length change and bending angle of the film can be derived as follows:

$$L = R * \theta \tag{1}$$

$$L + \Delta L_1 = (R + d_1) * \theta \tag{2}$$

$$L + \Delta L_2 = (R + d_2) * \theta \tag{3}$$

From qus. (1), (2) and (3) :

$$\frac{L+\Delta L_1}{L} = \frac{R+d_1}{R} \tag{4}$$

$$\frac{L+\Delta L_2}{L} = \frac{R+d_2}{R} \tag{5}$$

Accordingly, the qus. (4) and (5) can be deduced to:

$$\frac{\Delta L_1}{L} = \frac{d_1}{R} = d_1 * k \tag{6}$$

$$\frac{\Delta L_2}{L} = \frac{d_2}{R} = d_2 * k \tag{7}$$

$\frac{\Delta L}{L}$ is the swelling ratio and k is the curvature of nanocomposite film.

Here we supposed that the inner CNCs/PEGDA layer which backs to the wet air has no curvature change. According to the previous report, the swelling ratio of humidity-driven composites film actuator is too small that cannot be measured precisely by using conventional microscopy methods.[1-3] Moreover, the moisture absorption of PA-6 film has a weak effect on the curvature change of the humidity-driven nanocomposite film.[4] Compared with the great change of the curvature of nanocomposite film driven by humidity source, the own swelling ratio ($\Delta L_2/L$) of

CNCs/PEGDA layer and the swelling ratio ($\Delta L_1/L$) of uniaxially oriented PA-6 layer can be negligible.

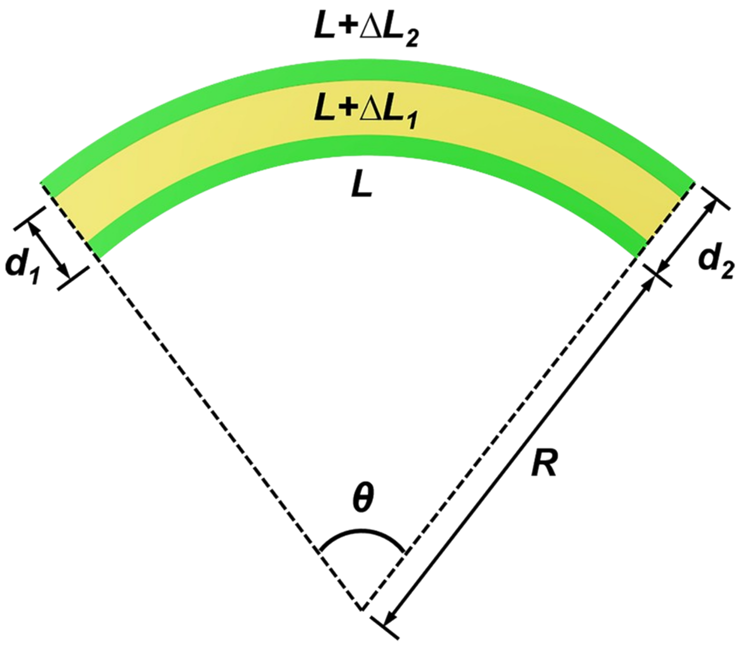

Fig. S1 Scheme representation of bending behavior analysis of sandwiched structure nanocomposite film. θ is the bending angle, d is the thickness of film, L is the length of film, ΔL is the length change of the CNCs/PEGDA layer and uniaxially oriented PA-6 layer.

2 Characterizations of CNCs

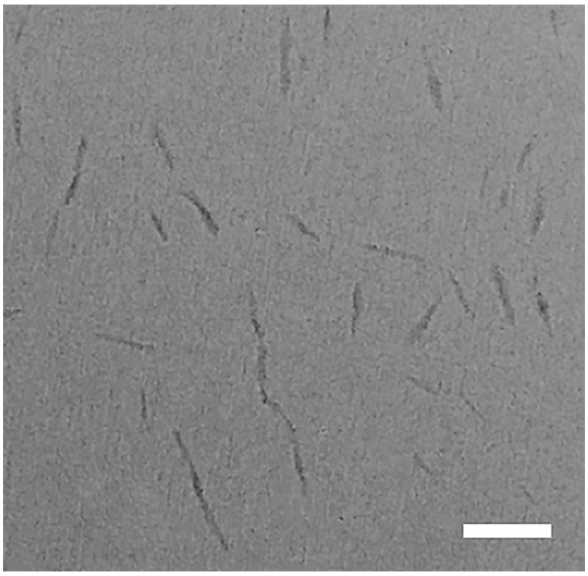

Fig. S2 TEM image of the CNCs suspension prepared from cotton pulp. (Scale bar: 500 nm)

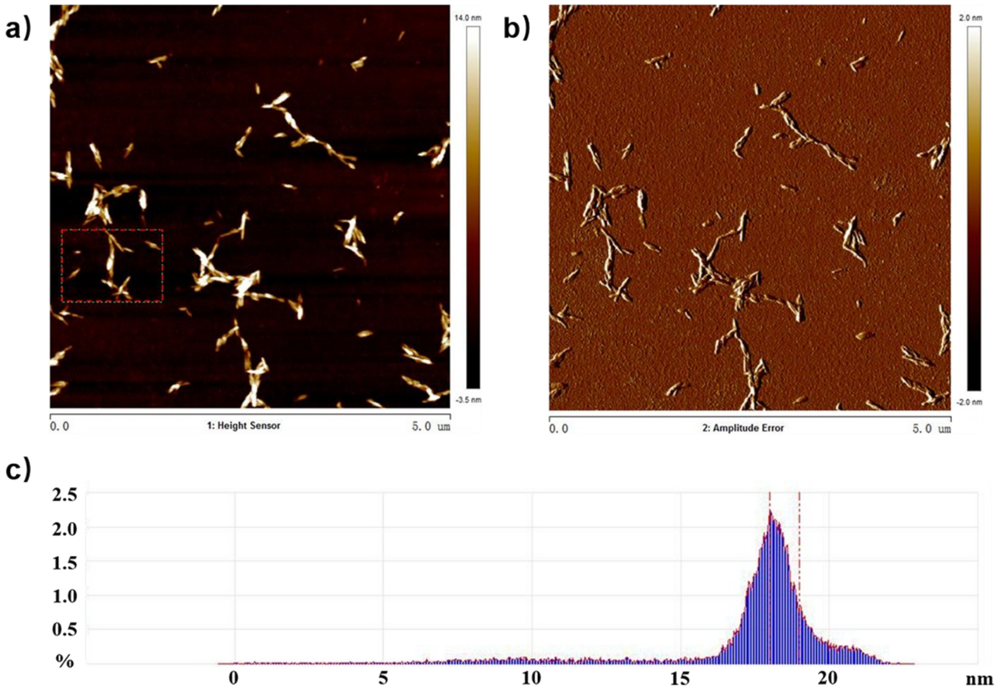

Fig. S3 (a) and (b) AFM image of the CNCs used in this study in height profile mode and amplitude mode, cast onto a freshly silica surface. (c) the height attribution of selected area in (a).

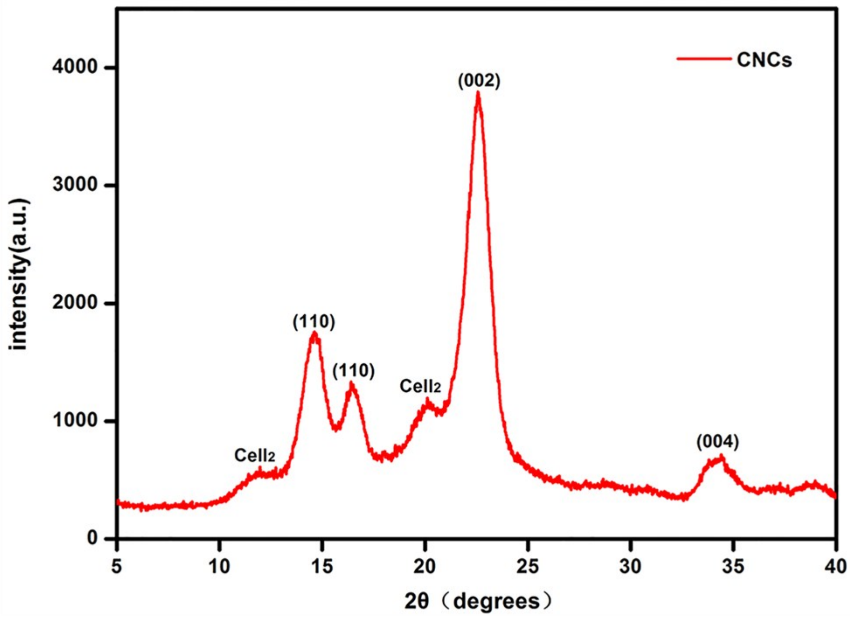

Fig. S4 X-ray diffraction spectra of the CNCs.

3 Characterizations of CNCs self-assembly film with a chiral nematic structure

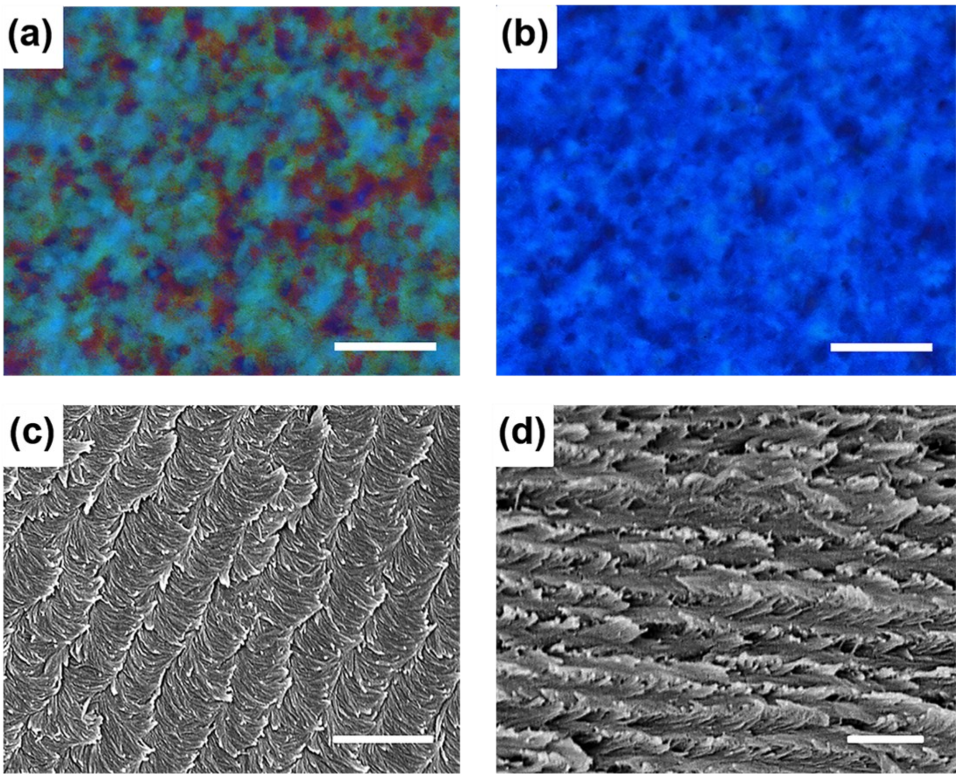

Fig. S5 (a) and (b) POM image of the CNCs self-assembly film observed in 0° and 90°. (Scale bar: 250 μm) (c) Fracture section SEM image of the CNCs self-assembly film. (Scale bar: 1 μm) (d) the cross-sectional SEM image of CNCs self-assembly film showing orderly twisted layer structures. (Scale bar: 250 nm)

4 Effect of CNCs/PEGDA weight ratio on the reflection spectra

Here we studied the influence of weight ratio between CNCs and PEGDA on the reflection wavelength of the bilayer nanocomposite film. 2 mL of 1.5 wt % CNCs suspension was mixed with 300 μL, 500 μL, 700 μL, 900 μL 5 wt % PEGDA solution (the solution contained 96.5 wt % PEGDA and 3.5 wt % 2959), corresponding to the mass fraction ratio of CNCs and PEGDA (66.7/33.3, 54.5/45.5, 46.2/53.8, 40/60 w/w). The mixed CNCs/PEGDA aqueous solution was mild stirred for 15 min, and then dropped on one side of uni-axially oriented PA-6 film assembling in 10 °C, 70 % relative humidity. As shown in Fig. S6, the reflection band and the corresponding reflected color shift to a longer wavelength as the PEGDA content increase.

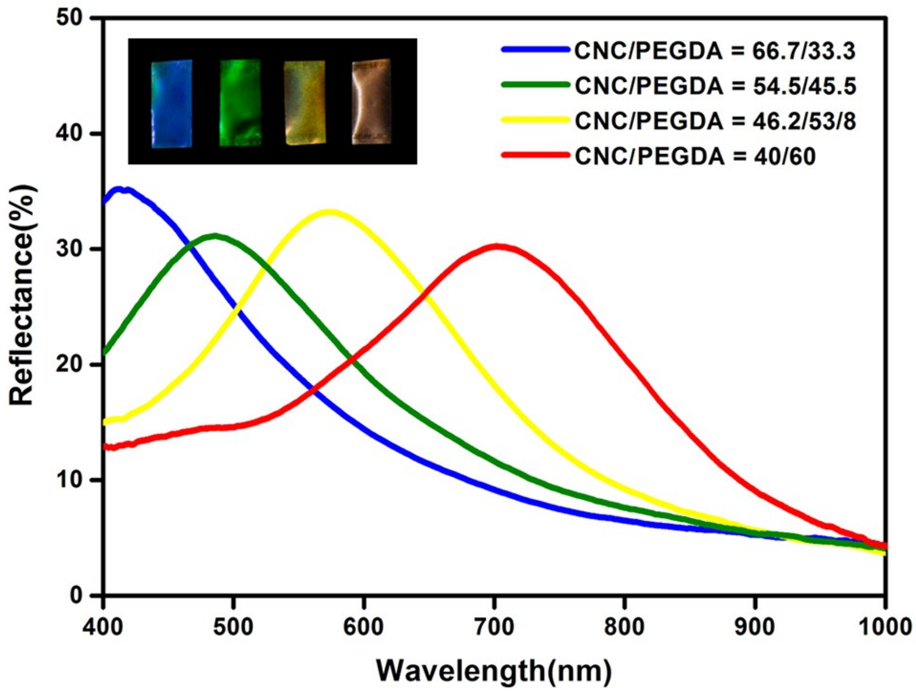

Fig. S6 Normal-incidence optical reflection spectra of the bilayer films with different CNCs/PEGDA weight ratio and the corresponding image of the bilayer films (inside).

5 Influence of the thickness of CNCs/PEGDA layer on humidity responsive behavior

To estimate the influence of the CNCs/PEGDA layer thickness on the dynamic curvature variation of the sandwiched structure nanocomposite films upon the same relative humidity (100 %). Herein, four sandwiched structure nanocomposite films were prepared, in which the CNCs/PEGDA layer thickness varies from 6 μm to 24 μm, the thickness of nanocomposite film was measured by a handheld thickness meter (QuaNix 4500, QuaNix Ltd., Germany). Note that the layer thickness was controlled by changing the assembled volume of CNCs/PEGDA suspensions on unit area of PA-6 film. As shown in Fig. S7, it was found that the maximum curvature becomes small and the bending speed clearly slows down as the CNCs/PEGDA layer thickness was increased. For example, the film with 24 μm-thick CNCs/PEGDA layer thickness has a minimum curvature (only about 0.11 cm^{-1}), and almost no curvature change was observed within about half a minute. The result demonstrates that the thickness of CNCs/PEGDA layer greatly influenced the bending speed and maximum curvature of the sandwiched structure nanocomposite film.

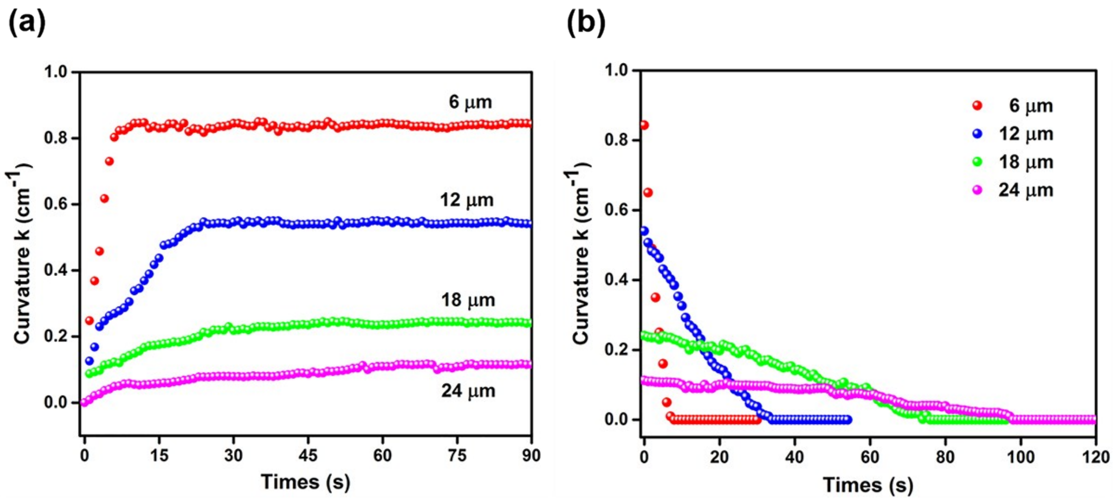

Fig. S7 (a) the bending behaviour of sandwiched structure nanocomposite film, which the thickness of CNCs/PEGDA layer is 6μm, 12μm,18μm and 24μm. (the relative humidity is 100%) (b) Relaxation dynamics of the bended film after removing the humidity exposure.

6 Weight-lifting test of sandwiched structure nanocomposite film

To further exhibit the possible application of the nanocomposite film, we also studied the weight-lifting ability of the sandwiched structure nanocomposite film. As shown in Fig. S8, the film was chipped with a tweeter and one side of the film faced the humidity source. When the wet air is on, the film bended and a light-weight paper crane with a weight of 110 mg was suspended through the film, the lifting distance could reach about 25 mm. After the moisture was evaporated, the bending film returned to the origin flatting state, and the lifting paper crane was put back the substrate.

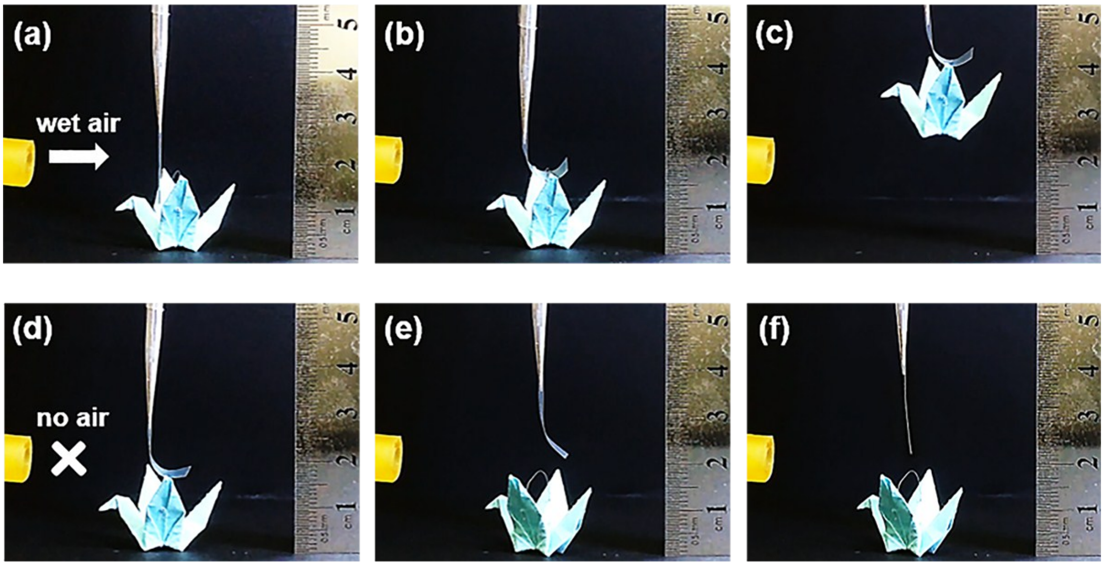

Fig. S8 Weight-lifting test of the sandwiched structure nanocomposite film.

Reference:

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