Supporting Information

Programmable chiroptical multilayer films assembled from self-healing stretchable elastomers for information encryption

Panyi Xi, ^a Yu Cao, ^a Tanyi Tan, ^a Xichong Ye*^{ab} and Feng Liu*^{abc}

^a Shaanxi International Research Center for Soft Matter, State Key Laboratory for Mechanical Behavior of Materials, School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an 710049, P. R. China

^b Xi'an Key Laboratory of Sustainable Polymer Materials, Xi'an Jiaotong University, Xi'an
710049, P. R. China

^e Institute of New Concept Sensors and Molecular Materials, Shaanxi Key Laboratory of New Concept Sensors and Molecular Materials, Xi'an Jiaotong University, Xi'an 710049, P. R. China

*Corresponding Author: xcye917@xjtu.edu.cn (X.-C. Ye); feng.liu@xjtu.edu.cn (F. Liu)



Figure S1. Synthesis route of WPU emulsion.



Figure S2. TEM image of TOCNF, inset: corresponding optical photo of TOCNF solution with 0.1wt% concentration. The length and diameter of TOCNF are approximately 400 nm and 15 nm, respectively.



Figure S3. The optical photographs of the apparent shape changes of WPU and WPU-TOCNF films during wet-stretching and drying.



Figure S4. SEM image of the AgNWs, inset: corresponding optical photo of AgNWs solution with 2 mg/ml concentration. The length and diameter of AgNWs are approximately 20 μm nm and 40 nm, respectively.



Figure S5. Preparation scheme of AgNWs@WPU/TOCNF hybrid film. (scale bar: 1 cm)



Figure S6. Extinction spectra of AgNW solution (a), WPU-TOCNF film and AgNWs@WPU/TOCNF film (b). The extinction spectra perfectly reveal the three components of the AgNWs@WPU/TOCNF film.



Figure S7. Cross-sectional SEM image of stretched AgNWs@WPU/TOCNF film. AgNWs is uniformly distributed in the WPU-TOCNF matrix.



Figure S8. The wet-stretching ratio of WPU-TOCNF films varies with the content of TOCNF.



Figure S9. Left: the azimuthal-integrated intensity distribution curves of the SAXS/WAXS patterns, where 0° represents the perpendicular direction. Right: effect of stretching ratios on Herman's orientation parameter (f_c) in SAXS/WAXS.



Figure S10. Polarized extinction spectra of oriented AgNWs@WPU/TOCNF film under different polarized light angles. The inserted plot shows the direction of linearly polarized light relative to the orientation of AgNWs.



Figure S11. Polarized Optical Microscopy (POM) image of stretched AgNWs@WPU/TOCNF film with 1000% elongation. Because of the anisotropic characters of stretched film, the POM image shows an obvious birefringence pattern under the crossed polarizers: (A) direction of the analyzer; (P) the direction of the polarizer.



Figure S12. The FTIR spectra of the AgNWs@WPU/TOCNF films in their original, wet and healed states. The results reveal that upon exposure to water, the original HBs are disrupted, as indicated by a decrease in absorption peaks in the 1500-1750 cm⁻¹ region (yellow band), accompanied by an increase in the broad absorption band at 3000-3600 cm⁻¹ corresponding to free HBs (gray band). Upon drying and healing, the absorption peaks at 1500-1750 cm⁻¹ recover and intensify, while the signal for free HBs diminishes, confirming the reformation of hydrogen bonding interactions.



Figure S13. The tensile stress-strain curve of the self-healed AgNWs@WPU/TOCNF film.



Figure S14. CD spectra of monolayer oriented AgNWs@WPU/TOCNF film measured at various rotation angles, ranging from 0° to 360°.



Figure S15. CD spectra of stacked AgNWs@WPU/TOCNF film measured at various rotation angles, ranging from 0° to 360°.



Figure S16. CD and g_{abs} spectra of stacked pre-stretched WPU-TOCNF films.



Figure S17. Fluorescence and CD spectra of WPU-R6G film and AgNWs@WPU/TOCNF film (λ_{ex} : 300 nm), inset: the corresponding optical photos of WPU-R6G film under UV-300 nm light. (scale bar: 1 cm)

Twisted angle	CD value (deg)	Decrypted module
-45° / 45°	-12 / 12	/
-40° / 40°	-11 / 11	/
-35° / 45°	-10 / 10	/
-30° / 30°	-9 / 9	/
-25° / 25°	-7.8 / 7.8	/
-20° / 20 °	-6.5 / 6.5	/
-15° / 15°	-5 / 5	/
-10° / 10°	-3.4 / 3.4	/
-5° / 5°	-1.8 / 1.8	
0 °	0	

Figure S18. The table of the ellipticity values and their corresponding encryption module units under more refined angle regulation.



Figure S19. The CD spectra of AgNWs@WPU/TOCNF films under different environmental conditions: (a) before and after repeated mechanical bending cycles; (b) after exposure to high humidity (relative humidity >90%) for 72 hours; and (c) after storage in ambient air for 30 days.