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Polypyrrole – decorated cellulose nanocrystal fillers in liquid crystal elastomers for

multi-stimuli response

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Preparation of cellulose nanocrystals (CNCs)

In this procedure, 2 g of microcrystalline cellulose (MCC) was hydrolyzed with 75 wt% of H_2SO_4 at a ratio of 8.75 mL of acid per gram of MCC. Hydrolysis was carried out at 45 °C under continuous mechanical stirring for 3 h. The solution was then diluted with cold deionized water (10 times the volume of the suspension) and left to settle overnight. The clear supernatant was removed, and the remaining residual precipitate was centrifuged 4–5 times (4000 rpm) until the suspension became white (pH= 7). The final suspension was collected after the last centrifugation and freeze-dried for subsequent analysis.

Preparation of LCE

In the first step, RM-257 (0.500 g) was dissolved in toluene (0.155 g) at 80 °C and then cooled to room temperature. The cross-linker; PETMP (0.024 g) and spacer; EDDET (0.120 g) were added at a molar ratio of 13:87, along with the photoinitiator HHMP (0.003 g). The diluted catalyst DPA (0.072 g, in a 1:100 ratio in toluene) was then added to the monomer solution. The mixture was vigorously stirred and placed in a vacuum chamber at 508 mmHg for 1 min to remove air bubbles. The solution was then poured into a Teflon mold, left at room temperature for at least 48 h, and then placed in a vacuum chamber at 60 °C for 24 h to evaporate the toluene.

The film formed in the initial step was stretched to align the mesogens and this orientation was fixed through UV irradiation, inducing a second cross-linking process that resulted in a mono-domain LCE.



Fig. S1 A TEM image of CNCs-PPy dispersion in DMSO.



Fig. S2 XRD spectrum of PPy.



Fig. S3 UV-vis spectrum of CNCs.



Fig. S4 UV-vis spectrum of CNCs-PPy.



Fig. S5 UV-vis spectra of undoped LCE and LCE/CNCs-PPy composite films.



Fig. S6 The extended UV-vis graph of LCE/CNCs-PPy composite films in the range 700 nm to 1000nm.



Fig. S7 FT-IR spectra of CNCs (as KBr pellet) and CNCs-PPy (ATR mode).



Fig. S8 FT-IR spectra of LCE/CNCs-PPy composite films (ATR mode).



Fig. S9 TGA graph of CNCs-PPy.



Fig. S10 DSC curve of a) undoped LCE b) LCE/CNCs-PPy-3 (second cycle).



Fig. S11 POM images of LCE/CNCs-PPy-1 at different temperatures, rate 10 °C/min (a-c: heating; d-f: cooling; scale bar- 500 μm).



Fig. S12 POM images of LCE/CNCs-PPy-2 at different temperatures, rate 10 °C/min (a-c: heating; d-f: cooling; scale bar- 500 μm).



Fig. S13 POM images of LCE/CNCs-PPy-3 at different temperatures, rate 10 °C/min (a-c: heating; d-f: cooling; scale bar- 500 μm).



Fig. S14 POM images of LCE/CNCs-PPy-4 at different temperatures, rate 10 °C/min (a-c: heating; d-f: cooling; scale bar- 500 μm).



Fig. S15 Thermal actuation of LCE/CNCs-PPy-1 composite film.



Fig. S16 Thermal actuation of LCE/CNCs-PPy-2 composite film.



Fig. S17 Thermal actuation of LCE/CNCs-PPy-4 composite film.

Table S1 Thermal response of the films recorded before and after 2 months.

	0 months			After 2 months		
LCE/CNCs-PPy composites	Original length	Contraction at 200 °C	Shrinking percentag	Original length	Contraction at 200 °C	Shrinking percentag
	(cm)	(cm)	e	(cm)	(cm)	e
			(%)			(%)
LCE/CNCs-PPy-1	3.3	2.2	33	2.5	1.7	32
LCE/CNCs-PPy-2	2.2	2.1	5.0	1.5	1.4	7.0
LCE/CNCs-PPy-3	4.0	2.8	30	3.4	2.2	35
LCE/CNCs-PPy-4	3.3	2.4	27	2.6	1.8	31



Fig. S18 Thermal actuation of LCE/CNCs-PPy-3 composite film with a load (0.500 g).



Fig. S19 IR actuation of LCE/CNCs-PPy-1 composite film.



Fig. S20 IR actuation of LCE/CNCs-PPy-2 composite film.



Fig. S21 IR actuation of LCE/CNCs-PPy-3, (i-vi)-IR on; (vii-viii)-IR off (recorded after 2 months).



Fig. S22 IR actuation of LCE/CNCs-PPy-3 composite film placed on a flat surface.



Fig. S23 IR actuation of LCE/CNCs-PPy-3 composite film with a load (0.500 g).

Table S2 Surface temperature of the composite films recorded using thermal imager camera.

	Temperature °C			
LCE/CNCs-PPy composites	Initial	Final		
LCE/CNCs-PPy-1	36.7	58.2		
LCE/CNCs-PPy-2	36.5	60.0		
LCE/CNCs-PPy-3	36.8	70.6		
LCE/CNCs-PPy-4	34.7	77.3		



Fig. S24 Time *versus* temperature curve for LCE/CNCs-PPy (0.01-0.04 wt%) recorded during irradiation with an IR source -808 nm, captured using thermal imaging camera (Fluke Tis60+- 90 Hz).



Fig. S25 IR actuation of LCE/CNCs-PPy-1 composite film with load.



Fig. S26 IR actuation of LCE/CNCs-PPy-4 composite film with load.



Fig. S27 IR actuation of LCE/CNCs-PPy-2 composite film with load (after 2 months).



Fig. S28 Light induced thermal actuation of LCE/CNCs-PPy-4 composite film in response to radiation from an incandescent bulb source.



Fig. S29 Light induced thermal actuation of LCE/CNCs-PPy-2 composite film with load in response to radiation from an incandescent bulb source.