Optimizing the performance of the near-infrared (NIR) photothermal conversion via modulating the domain size of chiral nematic phase in the co-assembled cellulose nanocrystals composite films

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Fig. S1 The vis-NIR transmittance spectra (a) and CD spectra (b) of pure CNCs and \mbox{CP}_{x}

composite films.



Fig. S2 Cross-sectional SEM images of (a) CP_1 ; (b) CP_2 ; (c) CP_3 ; (d) CP_4 , (e) CP_5 and (f)

CP₆ composite film.



Fig. S3 The magnified cross-sectional SEM images of (a) CP_5 and (b) CP_6 composite

film.



Fig. S4 UV-vis-NIR absorption spectrum of pure PEDOT: PSS film.



Fig. S5 The surface temperature of CP_x composite films induced by NIR light irradiation (750 nm) under different power of light.



Fig. S6 The surface temperature of CP_x film increases and decreases through alternatively turn-on and turn-off light irradiation under different power. (a) CP_1 ; (b) CP_2 ; (c) CP_3 ; (d) CP_4 ; (e) CP_5 ; (f) CP_6 composite film.



Fig. S7 A linear fitting correlation between cooling period (*t*) and $-ln\theta$ with an *R*-squared value of over 0.99. θ refers to the ratio of ΔT to ΔT_{max} .



Fig. S8 Characterizations of HPC/PEDOT: PSS/OS/TA (a, b) and PVP/PEDOT: PSS/OS/TA (c, d). Both samples contained 3.43 wt.% PEDOT. (a, c) The cross-sectional SEM images. (b, d) POM images. These images confirmed that there was no chiral nematic nanostructure in these composite films.



Fig. S9 The increase and decrease of the surface temperature of pure PEDOT: PSS film through alternative turn-on and turn-off light irradiation under different power.



Fig. S10 POM images of (a) CNCs/PEDOT: PSS/OS and (b) CNCs/PEDOT: PSS/TA film.



Fig. S11 POM image of pure CNCs film.



Fig. S12 POM image of CP₆ composite film.



Fig. S13 POM images of CNCs/PEDOT: PSS composite films with the mass ratio of (a)

CNCs : (PEDOT: PSS) = 95:5; (b) CNCs : (PEDOT: PSS) = 80:20.



Fig. S14 (a) POM image of pure CNCs (b.d.) film. (b) The cross-sectional SEM image of

pure CNCs (b.d.) film.



Fig. S15 The comparison of temperature changes of CP_x composite films (the red line) and CP_x (b.d.) films (the black line) with different PEDOT: PSS contents, under the 1.5 W light radiation (750 nm laser).

Sample codes	Weight ratio
	CNCs : (PEDOT:PSS) : OS : (TA + zonyl) ^a
CP1	75.19 : 3.96 : 8.35 : 12.50
CP ₂	71.60 : 7.95 : 7.95 : 12.50
CP ₃	67.96 : 11.99 : 7.55 : 12.50
CP ₄	64.29 : 16.07 : 7.14 : 12.50
CP ₅	60.58 : 20.19 : 6.73 : 12.50
CP ₆	56.83 : 24.36 : 6.31 : 12.50

Table S1 Sample codes of CP_x composite films.

 $^{\rm a}$ The ratios of TA and zonyl were kept at 10 % and 2.5 %, respectively.

Table S2 The compositions of CNCs/PEDOT: PSS, CNCs/PEDOT: PSS/OS, CNCs/PEDOT:PSS/TA, PEDOT: PSS/OS/TA, HPC/PEDOT: PSS/OS/TA, PVP/PEDOT: PSS/OS/TA andCNCs(b.d.)/PEDOT: PSS/OS/TA composite films.

Sample	Weight ratio
CNCs/PEDOT: PSS	CNCs : (PEDOT: PSS)
	93.54 : 3.96
	89.55 : 7.95
	85.51 : 11.99
	81.43 : 16.07
	77.31 : 20.19
CNCs/PEDOT: PSS/OS	CNCs : (PEDOT: PSS) : OS ^a
	84.19 : 3.96 : 9.35
	80.60 : 7.95 : 8.95
	76.96 : 11.99 : 8.55
	73.29 : 16.07 : 8.14
	69.58 : 20.19 : 7.73
CNCs/PEDOT: PSS/TA	CNCs : (PEDOT: PSS) : TAª
	83.54 : 3.96 : 10
	79.55 : 7.95 : 10
	75.51 : 11.99 : 10

71.43 : 16.07 : 10

67.31:20.19:10

PEDOT: PSS/OS/TA (PEDOT: PSS) : OS : TA : Zonyl 15.96 : 33.66 : 40.31 : 10.07 27.99 : 27.99 : 35.21 : 8.81 37.42 : 23.56 : 31.21 : 7.81 45.00 : 19.99 : 28.00 : 7.01 51.22 : 17.07 : 25.37 : 6.34 HPC (or PVP, CNCs(b.d))/PEDOT: PSS/OS/TA HPC (or PVP, CNC(b.d)) : (PEDOT: PSS) : OS : TA^a 75.19 : 3.96 : 8.35 : 10 71.60 : 7.95 : 7.95 : 10 67.96 : 11.99 : 7.55 : 10 64.29 : 16.07 : 7.14 : 10 60.58 : 20.19 : 6.73 : 10

^a: The zonyl content was kept at 2.5% if not specified.

Samples	PEDOT content	η _{ΡΤ}
	(wt.%)	(%)
(Pure PEDOT: PSS)	28.57	68.7
CP1	1.13	71.2
CP ₂	2.27	73.8
CP ₃	3.43	77.0
CP ₄	4.59	77.0
CP ₅	5.77	77.6
CP ₆	6.96	64.2

Table S3 NIR photothermal conversion efficiency (η_{PT}) of pure PEDOT: PSS and CP_x

composite films.