Supporting Information

Halloysite Nanotubes Enhance the Mechanical Properties and

Thermal Stability of Iridescent Cellulose Nanocrystal Films

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Fig. S1 TEM images of CNC1 (A) and CNC2 (B).



Fig. S2 TEM image of HNTs.



Fig. S3 Photographs (from left to right) of HNTs, **CNC1**, **CNC1** mixed with 1 wt. %, 3 wt. %, 5 wt. % and 10 wt. % HNTs suspensions before (A) and after (B) standing for one day. Photographs (from left to right) of HNTs, **CNC2**, and **CNC2** mixed with 1 wt. %, 3 wt. %, 5 wt. %, and 10 wt. % HNTs suspensions before (C) and after (D) standing for one day.



Fig. S4 Zeta potentials of HNTs, **CNC1**, **CNC1**/HNTs, **CNC2** and **CNC2**/HNTs suspensions. Error bars are the standard deviation of measurements on three samples.



Fig. S5 (A, B) SEM images and corresponding SEM-EDS mapping of **CNC1** mixed with 5 wt. % HNTs composite film. A and B correspond to two different regions of the film.



Fig. S6 (A, B) SEM images and corresponding SEM-EDS mapping of **CNC1** mixed with 10 wt. % HNTs composite film. A and B correspond to two different regions of the film.



Fig. S7 (A) FTIR spectra and (B) PXRD patterns of films prepared from HNTs and **CNC2**. The spectral traces correspond to: film made from **CNC2** (B) and **CNC2** mixed with 1 wt. %, 3 wt. %, 5 wt. % and 10 wt. % HNTs. Standard cards of HNTs (01-081-9524) and CNCs (00-056-1718) are from the JCPD database.



Fig. S8 (A) Photographs of prepared films, (B) SEM images, and (C) POM images of the series of composite films made using CNC2. Panels a to e: CNC2 film (a), composite films made from CNC2 mixed with 1 wt. % (b), 3 wt. % (c), 5 wt. % (d) and 10 wt. % (e) HNTs.



Fig. S9 (A) Normalized CD spectra of pristine **CNC1** film (a) and composite films of **CNC1** mixed with 1 wt. % HNTs (b), 3 wt. % HNTs (c), 5 wt. % HNTs (d) and 10 wt. % HNTs (e). (B) Normalized reflectance spectra of pristine **CNC1** film (a) and composite films prepared from **CNC1** mixed with 1 wt. % HNTs (b), 3 wt. % HNTs (c), 5 wt. % HNTs (d) and 10 wt. % HNTs (e).