Supplementary Information

## Self-Alignment Sequence of Colloidal Cellulose Nanofibers Induced by Evaporation from Aqueous Suspensions

Kojiro Uetani<sup>1,\*</sup>, Shogo Izakura<sup>2</sup>, Takaaki Kasuga<sup>2</sup>, Hirotaka Koga<sup>1</sup> and Masaya Nogi<sup>1</sup>

- 1 Institute of Scientific and Industrial Research (ISIR), Osaka University, Mihogaoka 8-1, Ibaraki-shi, Osaka 567-0047, Japan; uetani@eco.sanken.osaka-u.ac.jp (K.U.); hkoga@eco.sanken.osaka-u.ac.jp (H.K.); nogi@eco.sanken.osaka-u.ac.jp (M.N.)
- 2 Graduate School of Engineering, Osaka University, Mihogaoka 8-1, Ibaraki-shi, Osaka 567-0047, Japan; izakura.shogo@chem.eng.osaka-u.ac.jp (S.I.), tkasuga@eco.sanken.osaka-u.ac.jp (T.K.)
- \* Correspondence: uetani@eco.sanken.osaka-u.ac.jp; Tel.: +81-6-6879-8441



Figure S1. AFM image of the CNFs used in this study.



**Figure S2.** Birefringence images of the dried nanopapers in the *y* direction. (**a**) Glass cell without hydrophobization treatment showing CNFs adhered to the wall. (**b**) Hydrophobized cell without CNFs adhered to the walls properly dried to form a nanopaper.



**Figure S3.** Estimation of the apparent concentration at which birefringence starts to occur by linear fitting. (**a**, **b**) Suspensions with initial concentrations of 0.087 and 0.52 wt% show birefringence in the *z* direction at 1.26 and 1.33 wt%, respectively. (**c**, **d**) Suspensions with initial concentrations of 0.087 and 0.52 wt% show birefringence in the *y* direction at 0.57 and 0.55 wt%, respectively.



**Figure S4.** (**a**, **b**) Appearance, (**c**, **d**) thermogravimetric profiles, and (**e**, **f**) AFM images of nanopapers dried from (**a**, **c**, **e**)  $\phi_i = 0.087$  wt% and (**b**, **d**, **f**)  $\phi_i = 0.52$  wt% suspensions.