

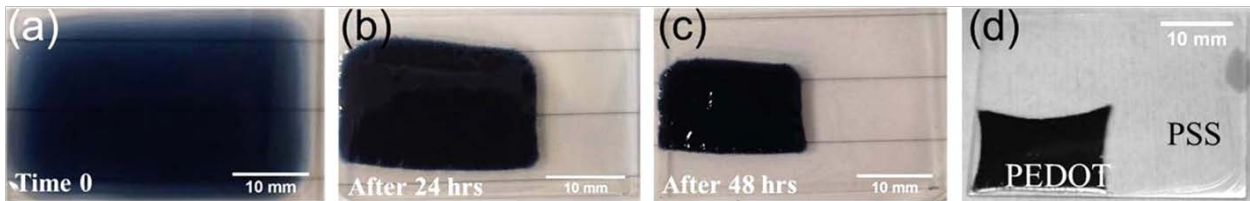
## Supplementary information

### A continuum theory of organic mixed ionic-electronic conductors of phase separation

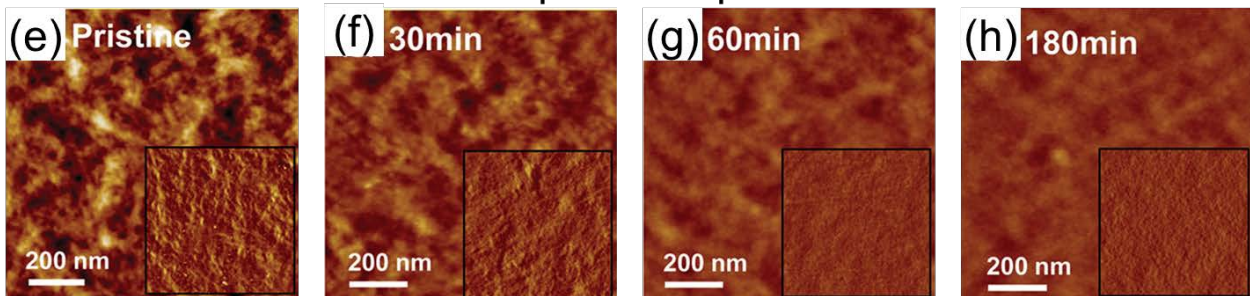
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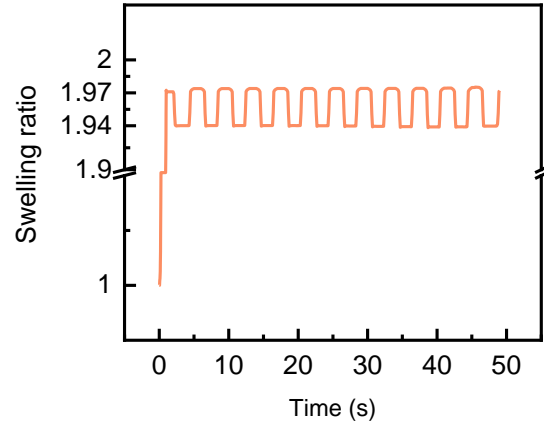
#### Horizontal phase separation



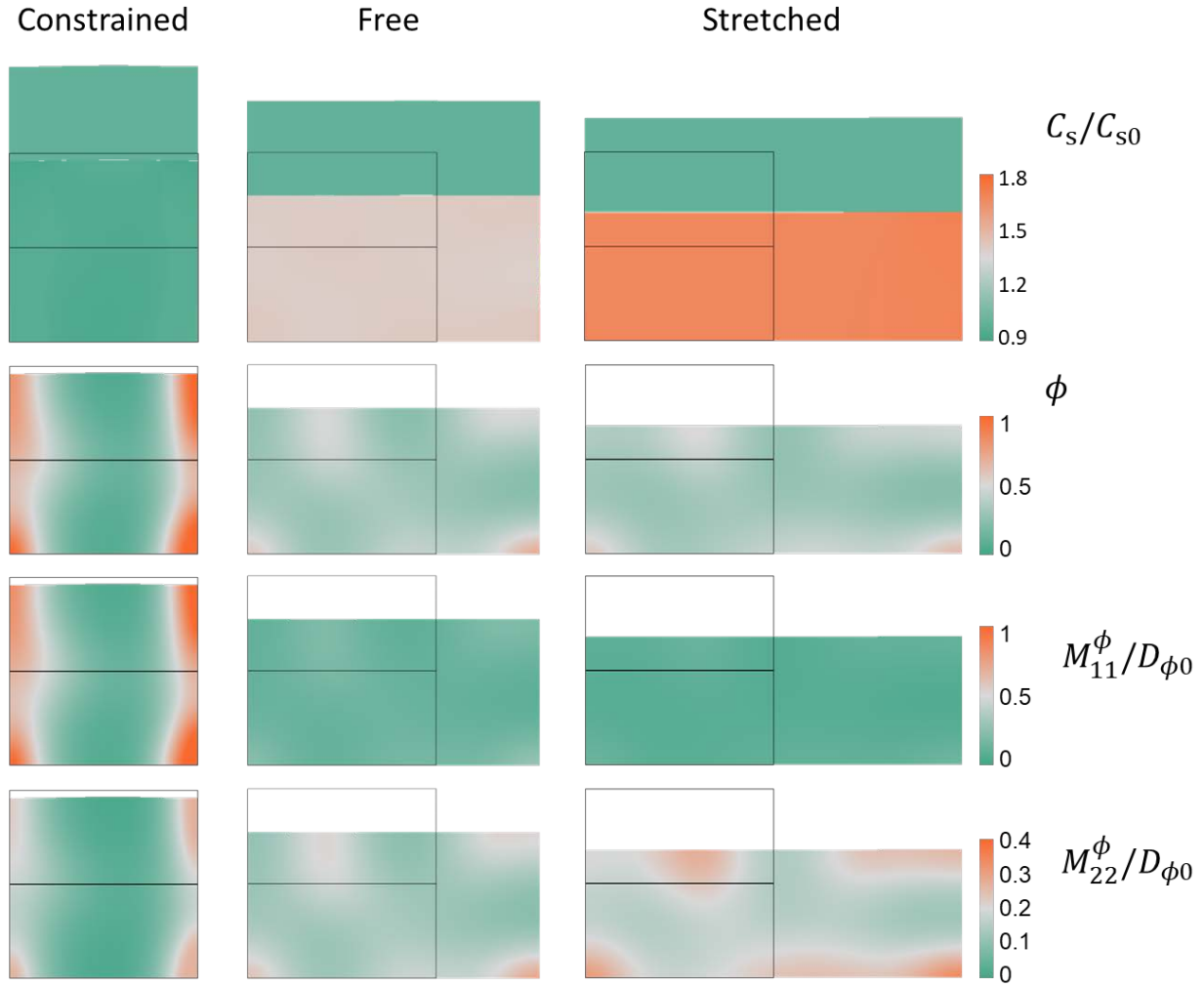
#### Vertical phase separation



**Figure S1.** Experimental observations of phase separation in PEDOT:PSS. Optical images of PEDOT:PSS film undergoing phase separation in the in-plane directions at (a) time zero, (b) 24 hours, (c) 48 hours, and (d) completely dried after processing. Scale bar is 10 mm. Topography and the phase images (inset) of PEDOT:PSS films undergoing phase separation along the vertical direction measured by AFM at (e) initial state, (f) 30 mins, (g) 60 mins, and (h) 180 mins after treatment. The scale bar is 20 nm. (a)-(d) Reprinted (adapted) with permission from Ouyang et al. (Ouyang et al., 2015). Copyright 2022 American Chemical Society. (e)-(h) Reprinted (adapted) with permission from Yeo et al. (Yeo et al., 2012). Copyright 2022 American Chemical Society.



**Figure S2.** Swelling ratio of the PEDOT:PSS channel at the source electrode during the first 12 cycles.



**Figure S3.** Contour plots in the deformed configuration of the solvent concentration, local volume fraction of PEDOT, the normalized phase mobility  $M_{11}$  (in-plane) and  $M_{22}$  (out of plane) after 12 cycles.

## References

- Ouyang, L., Musumeci, C., Jafari, M.J., Ederth, T., Inganäs, O., 2015. Imaging the phase separation between PEDOT and polyelectrolytes during processing of highly conductive PEDOT:PSS films. *ACS Appl. Mater. Interfaces* 7, 19764–19773.
- Yeo, J.-S., Yun, J.-M., Kim, D.-Y., Park, S., Kim, S.-S., Yoon, M.-H., Kim, T.-W., Na, S.-I., 2012. Significant vertical phase separation in solvent-vapor-annealed poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) composite films leading to better conductivity and work function for high-performance indium tin oxide-free optoelectronics. *ACS Appl. Mater. Interfaces* 4, 2551–2560.