

Supporting Information

Interfacial Ion Transport in Cellulose Nanocrystal-Based Ionogels with Low Ionic Liquid Content

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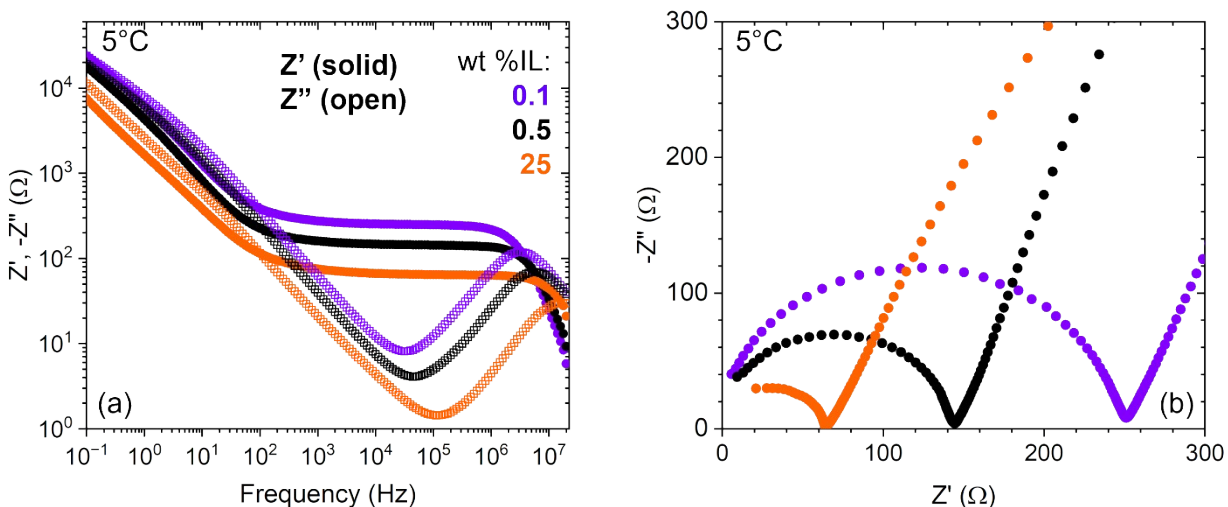


Figure S1. (a) Representative Bode Plot (Z' and $-Z''$ vs. Frequency) for 0.1, 0.5, and 25 wt% IL systems at 5 °C and (b) corresponding Nyquist plots ($-Z''$ vs. Z') of these same systems. These were selected to illustrate the presence of a frequency-independent plateau in (a), and equivalent circuit model behavior in (b) for the entire range of IL concentrations. Consistent with the equivalent circuit model, the resistance, R corresponds to the value of Z' when $-Z''$ is at a minimum, as seen in both (a) and (b).

Water-Mediated Stability of CNC-DMF mixture:

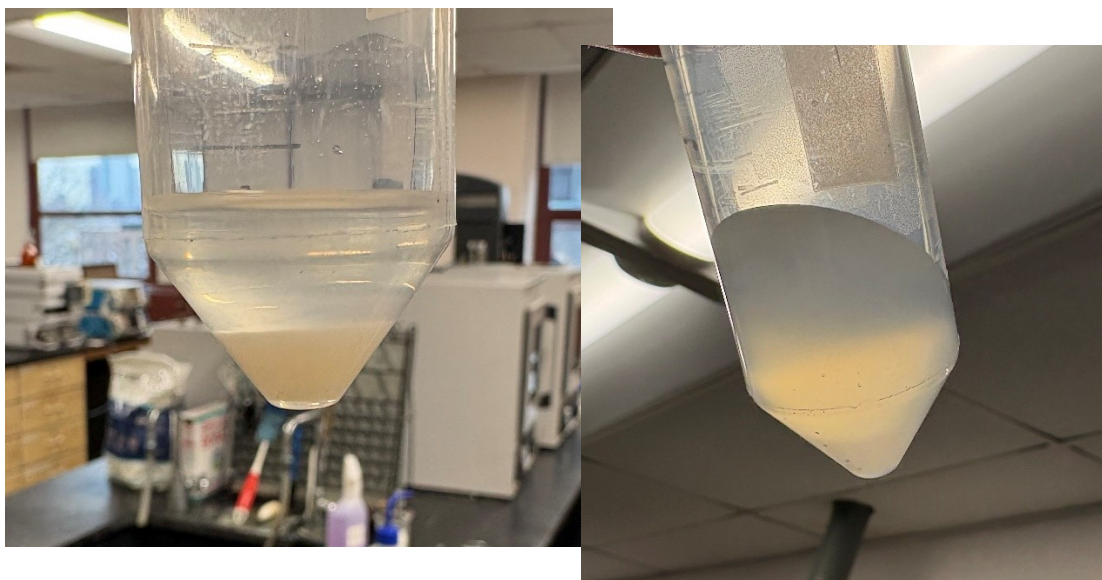


Figure S2. CNC-DMF mixture (left) and CNC-DMF-water mixture (right) after storing for 24 h.

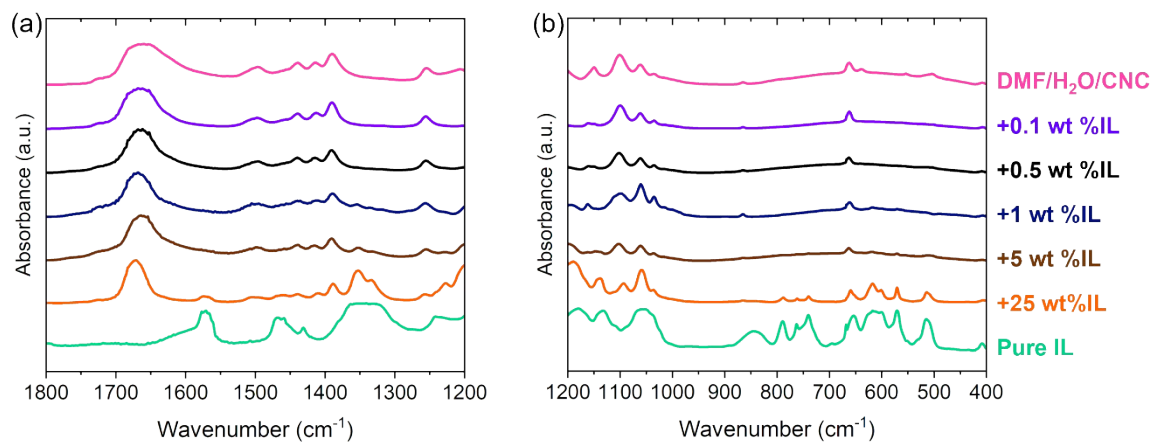


Figure S3. FTIR absorbance spectra for DMF-water-CNC-HMIm-TFSI systems at (a) 1800-1200 cm^{-1} and (b) 1200-400 cm^{-1} .

Table S1. Characteristic DMF band at $\sim 1511\text{ cm}^{-1}$ for the N-CH₃ stretching vibration shifts toward lower wavenumbers with the increasing IL amount.

Sample name	Peak wavenumber (cm^{-1})
DMF	1502
DMF+CNC	1501
DMF+CNC+H ₂ O	1496
DMF+CNC+H ₂ O+0.1 wt% IL	1495
DMF+CNC+H ₂ O+0.5 wt% IL	1496
DMF+CNC+H ₂ O+1 wt% IL	1497
DMF+CNC+H ₂ O+5 wt% IL	1497
DMF+CNC+H ₂ O+25 wt% IL	1505

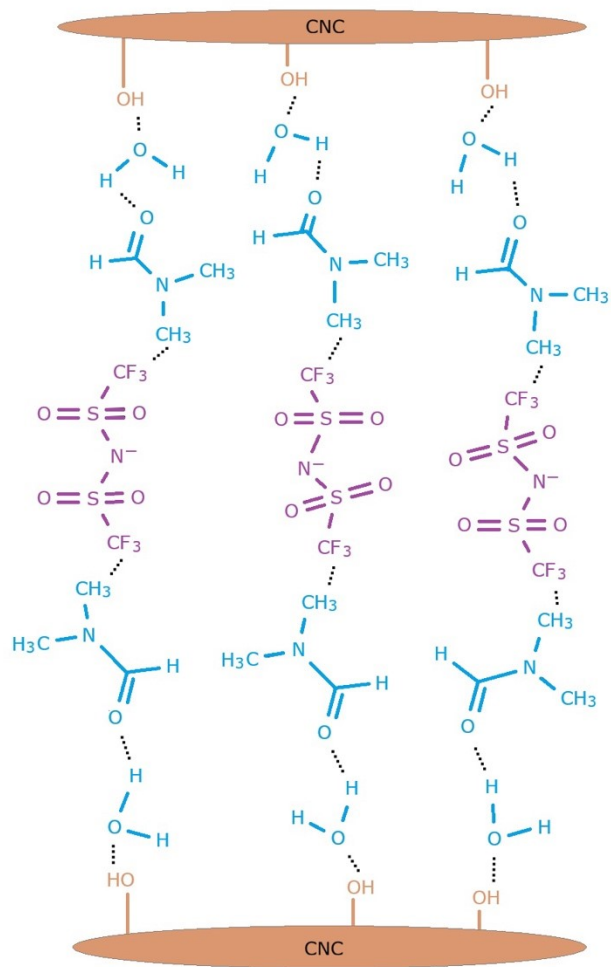


Figure S4. Schematic diagram of hydrogen bonds between water-DMF and ion-dipole interactions between HMIIm-TFSI-DMF.

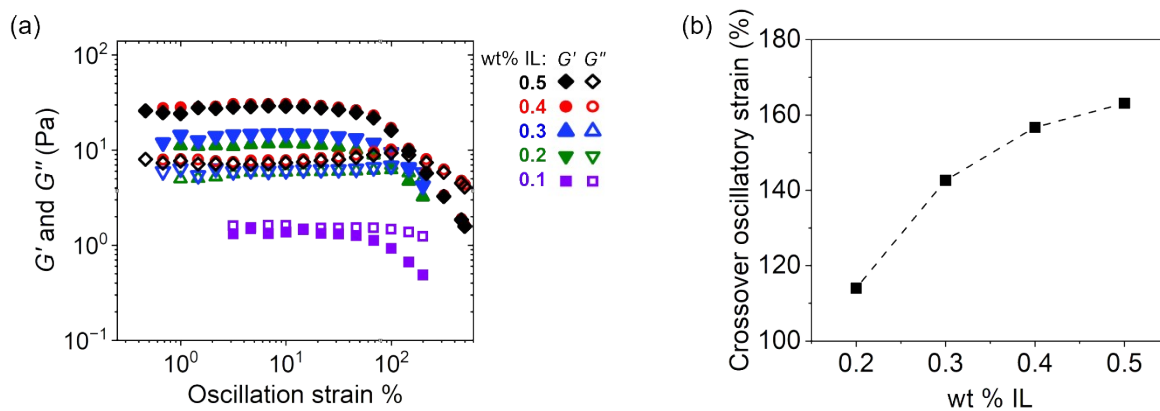


Figure S5. (a) Strain sweep data of CNC-HMIm-TFSI gels with DMF-water measured at room temperature at 1 rad/s frequency at 25°C, G' (closed symbols) and loss G'' (open symbols). **(b)** Crossover strain increases with varying HMIm-TFSI wt %.

1wt% IL 5wt% IL 10wt% IL 25wt% IL

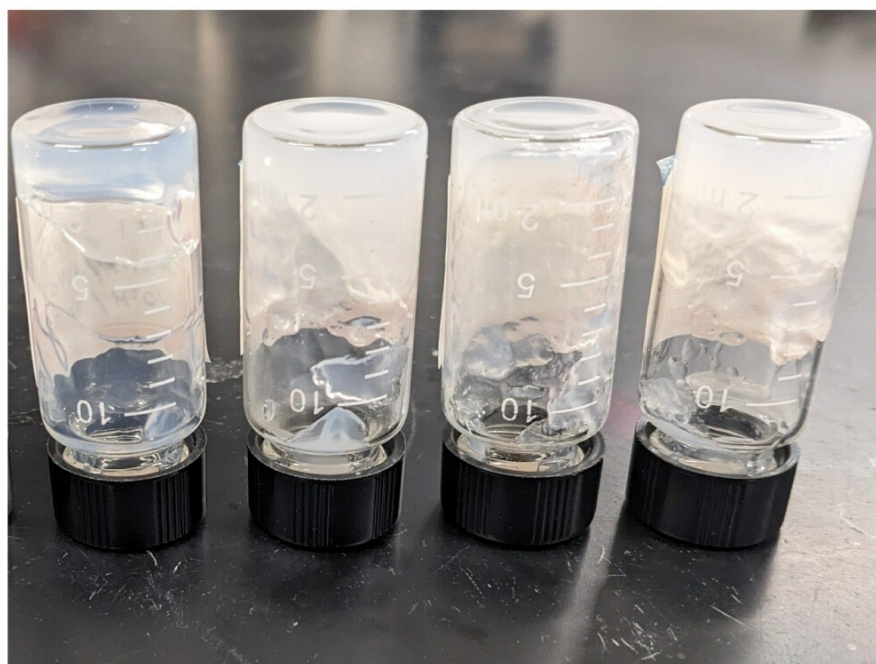


Figure S6. Photograph of CNC/IL gels with 1-25 wt% HMIm-TFSI amounts.

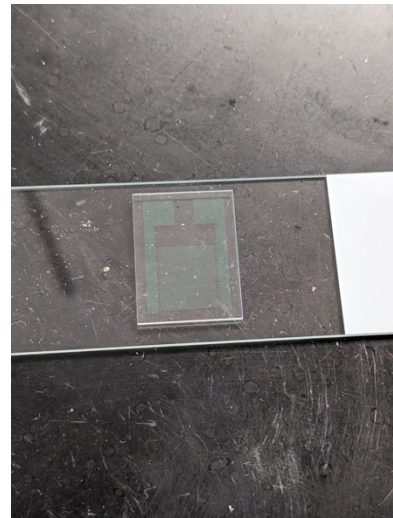


Figure S7. Photograph of an apparatus used to apply electric fields on sample droplets placed on an interdigitated electrode (on the right).

0 V

25 V

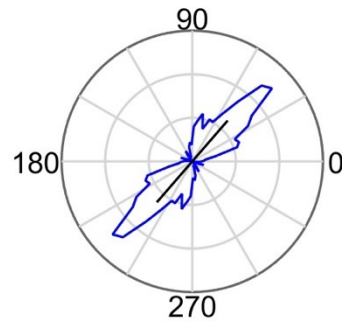
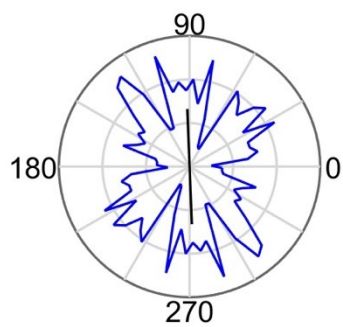
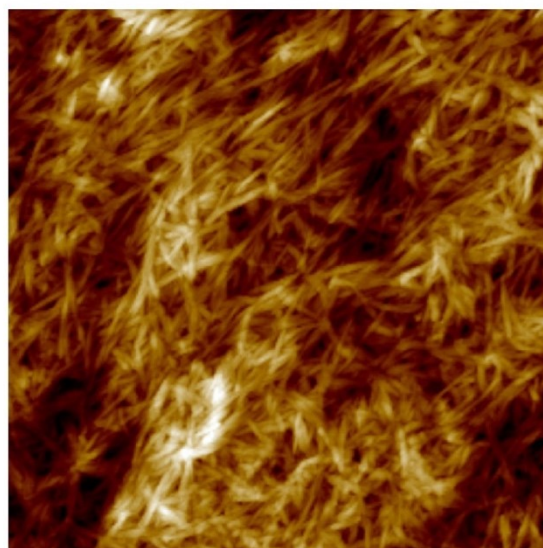
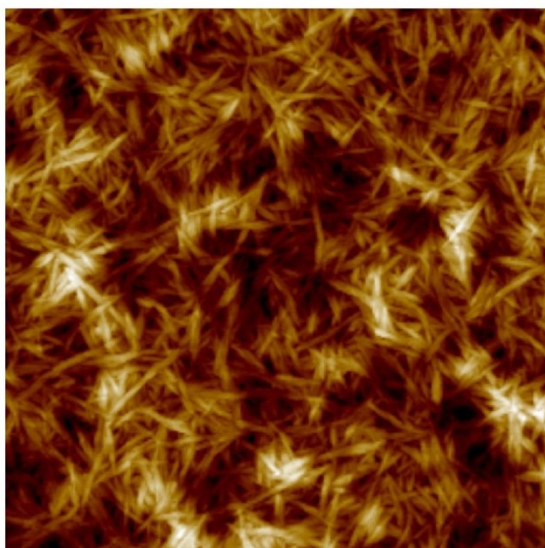


Figure S8. GTFiber analysis obtained from AFM topography of CNC gel with 0.1 wt% HMIIm-TFSI before (0 V) and after (25 V) electric field application. The orientation map across the selected area yields the nanocrystal orientation under applied fields.