

## Supporting Information

# Soft Nanoconfinement of Ionic Liquids in Lyotropic Liquid Crystals

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Table S1. Fitting parameters obtained from fitting the real and imaginary conductivity with equation (5).

| Compositions (wt%)                        | $\sigma_0(S/cm)$ | $\tau_e (s)$          |
|---|------------------|-----------------------|
| Pure [BMPyr][NTF <sub>2</sub> ]           | 0.19             | $1.00 \times 10^{-8}$ |
| Pure [BMIM][PF <sub>6</sub> ]             | 0.16             | $1.00 \times 10^{-8}$ |
| PEO/[BMPyr][NTF <sub>2</sub> ] (47/53)    | 0.031            | $2.0 \times 10^{-7}$  |
| PEO/[BMIM][PF <sub>6</sub> ] (47/53)      | 0.026            | $2.2 \times 10^{-7}$  |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (52/48) | 0.029            | $2.5 \times 10^{-7}$  |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (64/36) | 0.009            | $9.0 \times 10^{-7}$  |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (74/26) | 0.0055           | $1.5 \times 10^{-6}$  |
| Brij58/[BMIM][PF <sub>6</sub> ] (52/48)   | 0.024            | $2.7 \times 10^{-7}$  |
| Brij58/[BMIM][PF <sub>6</sub> ] (64/36)   | 0.003            | $4.5 \times 10^{-6}$  |
| Brij58/[BMIM][PF <sub>6</sub> ] (74/26)   | 0.00082          | $1.8 \times 10^{-5}$  |

Table S2. The number density of free ions obtained from Dyre Model for ion gels and PEO/IL mixtures.

| Compositions (wt%)                        | Fraction of Free Ions ( $n_{\text{free ions}}/n_{\text{total ions}}$ ) |                            |
|---|--|----------------------------|
|   | $\lambda = 0.7 \text{ nm}$   | $\lambda = 0.9 \text{ nm}$ |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (52/48) | 0.608  | 0.367                      |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (64/36) | 0.679  | 0.410                      |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (74/26) | 0.692  | 0.418                      |
| Brij58/[BMIM][PF <sub>6</sub> ] (52/48)   | 0.366  | 0.221                      |
| Brij58/[BMIM][PF <sub>6</sub> ] (64/36)   | 0.762  | 0.418                      |
| Brij58/[BMIM][PF <sub>6</sub> ] (74/26)   | 0.833  | 0.503                      |
| PEO/[BMPyr][NTF <sub>2</sub> ] (47/53)    | 0.521  | 0.315                      |
| PEO/[BMIM][PF <sub>6</sub> ] (47/53)      | 0.322  | 0.195                      |

Table S3. Maximum CO<sub>2</sub> absorption capacity of PEO and Brij58 mixtures with different ILs at 10 bar and 25 °C.

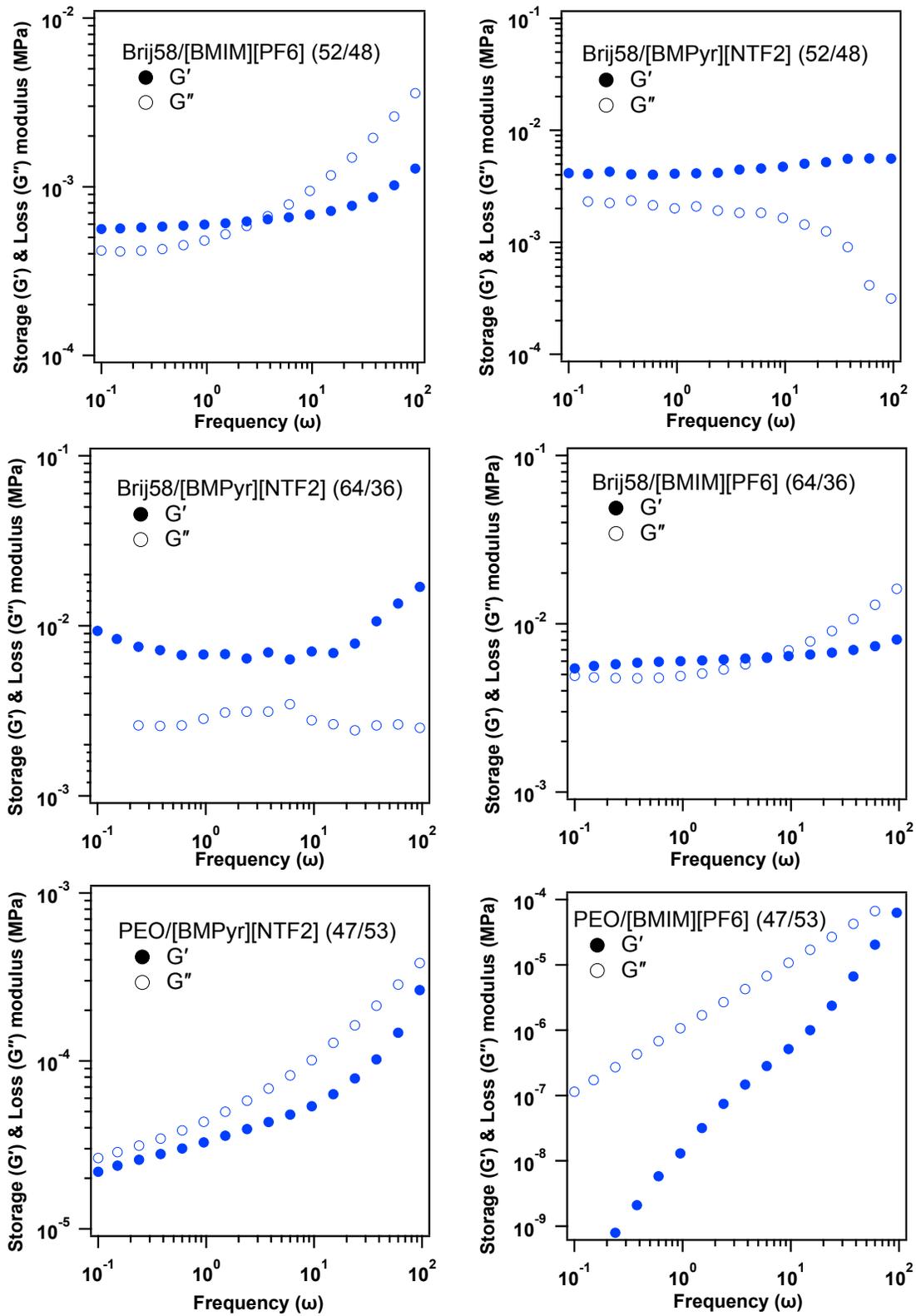
| Compositions (wt%)                        | (mmolCO <sub>2</sub> /g) | wt% | $\Delta\delta^a$ | $\Delta\chi^b$ |
|---|--------------------------|-----|------------------|----------------|
| Pure PEO1000                              | 0.13                     | 0.6 | -                | -              |
| Pure Brij58                               | 0.21                     | 0.9 | -                | -              |
| Pure [BMPyr][NTF <sub>2</sub> ]           | 0.38                     | 1.6 | -                | -              |
| Pure [BMIM][PF <sub>6</sub> ]             | 0.16                     | 0.7 | -                | -              |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (52/48) | 0.51                     | 2.2 | 0.22             | 0.68           |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (64/36) | 0.48                     | 2.1 | 0.21             | 0.95           |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (74/26) | 0.45                     | 1.9 | 0.20             | 1.35           |
| Brij58/[BMIM][PF <sub>6</sub> ] (52/48)   | 0.59                     | 2.6 | 0.41             | 1.07           |
| Brij58/[BMIM][PF <sub>6</sub> ] (64/36)   | 0.42                     | 1.8 | 0.23             | 1.01           |
| Brij58/[BMIM][PF <sub>6</sub> ] (74/26)   | 0.29                     | 1.3 | 0.10             | 0.96           |
| PEO/[BMPyr][NTF <sub>2</sub> ] (47/53)    | 0.29                     | 1.3 | 0.03             | 0.17           |
| PEO/[BMIM][PF <sub>6</sub> ] (47/53)      | 0.26                     | 1.1 | 0.12             | 0.33           |

<sup>a</sup> Enhanced CO<sub>2</sub> absorption (mmol per gram of sample) due to the synergistic effect.

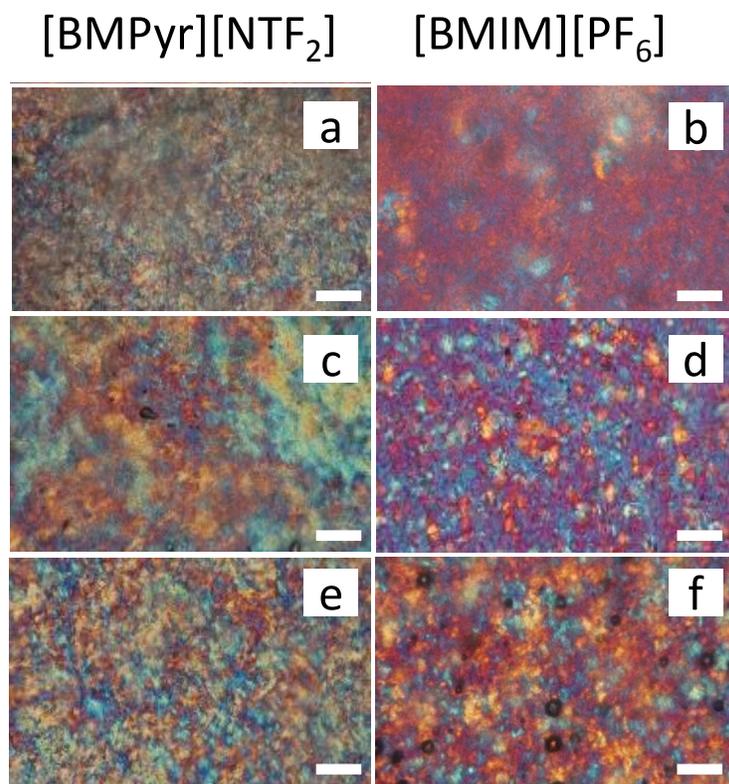
<sup>b</sup> Enhanced CO<sub>2</sub> absorption (mmol per gram of ILs).

*Table S4. Absorption-desorption rate and viscosity (at frequency of 0.1 Hz) of ion gels and PEO/ILs mixtures from 1 to 10 bars at 25 °C.*

| Compositions (wt%)                        | Absorption rate<br>( $\mu\text{mol}/\text{minute}$ ) | Desorption rate<br>( $\mu\text{mol}/\text{minute}$ ) | Viscosity    |
|---|--|--|--------------|
| Pure [BMPyr][NTF <sub>2</sub> ]           | 3.2  | 1.7  | 0.38 (Pa.s)  |
| Pure [BMIM][PF <sub>6</sub> ]             | 1.4  | 0.7  | 0.08 (Pa.s)  |
| PEO/[BMPyr][NTF <sub>2</sub> ] (47/53)    | 1.4  | 0.9  | 400 (Pa.s)   |
| PEO/[BMIM][PF <sub>6</sub> ] (47/53)      | 2.3  | 1.4  | 1.00 (Pa.s)  |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (52/48) | 3.8  | 3.6  | 0.07 (MPa.s) |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (64/36) | 1.8  | 1.5  | 0.10 (MPa.s) |
| Brij58/[BMPyr][NTF <sub>2</sub> ] (74/26) | 2.2  | 1.9  | 0.20 (MPa.s) |
| Brij58/[BMIM][PF <sub>6</sub> ] (52/48)   | 3.2  | 2.9  | 0.01 (MPa.s) |
| Brij58/[BMIM][PF <sub>6</sub> ] (64/36)   | 2.1  | 1.5  | 0.09 (MPa.s) |
| Brij58/[BMIM][PF <sub>6</sub> ] (74/26)   | 1.5  | 0.9  | 0.30 (MPa.s) |



**Figure S1.** The frequency sweep at strain of 0.5% for ion gels at 25 °C.



**Figure S2.** Cross-polarized light micrographs of ion gels prepared with Brij 58, [BMim][PF<sub>6</sub>], and [BMPyr][NTF<sub>2</sub>] with Brij58/ILs wt:wt (a, b) 52/48 , (c, d) 64/36, (e, f) 74/26. The scale bar is 50  $\mu\text{m}$ .

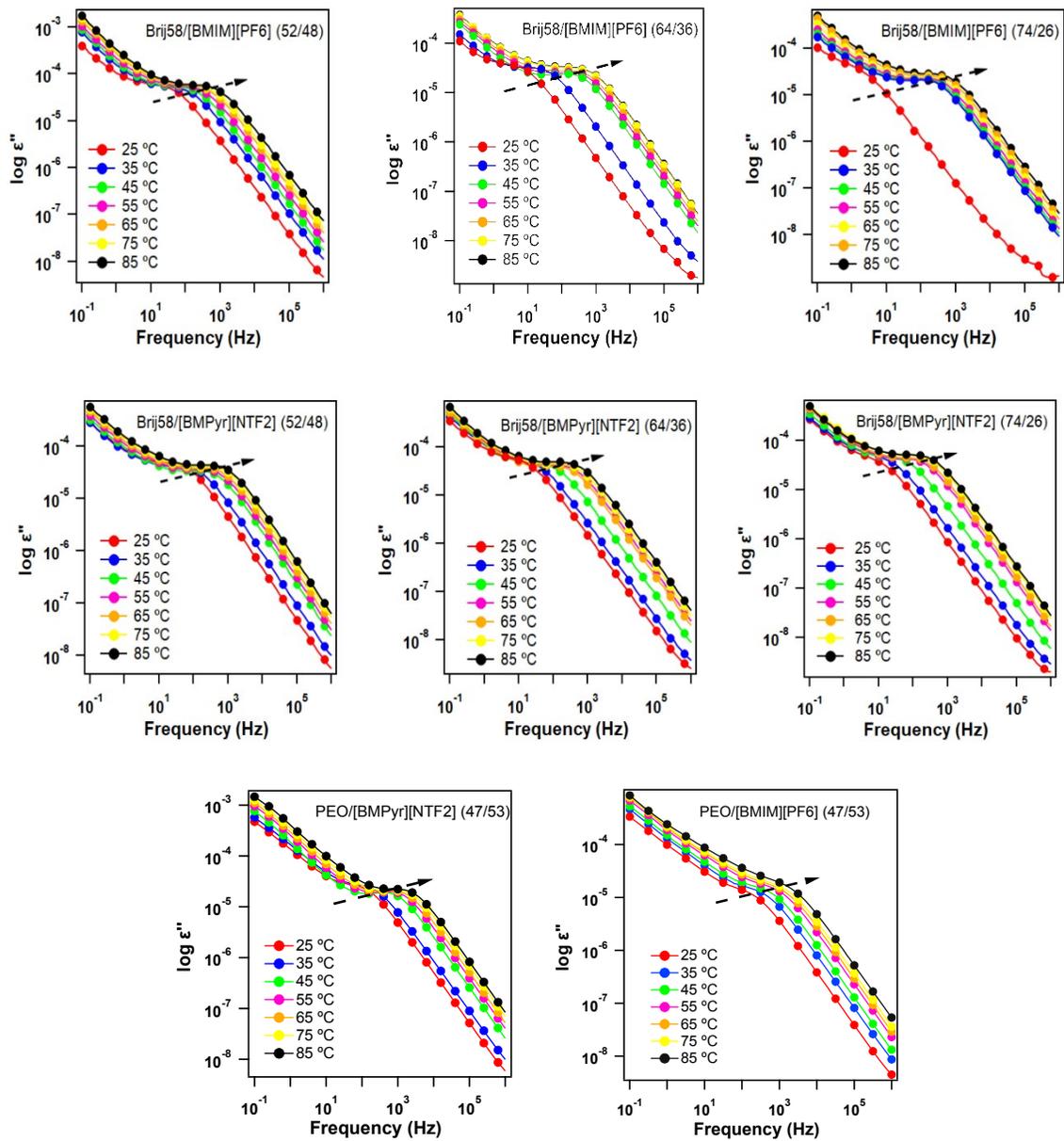


Figure S3. The dependence of dielectric loss spectra  $\epsilon''(\omega)$  on temperature for ion gels.

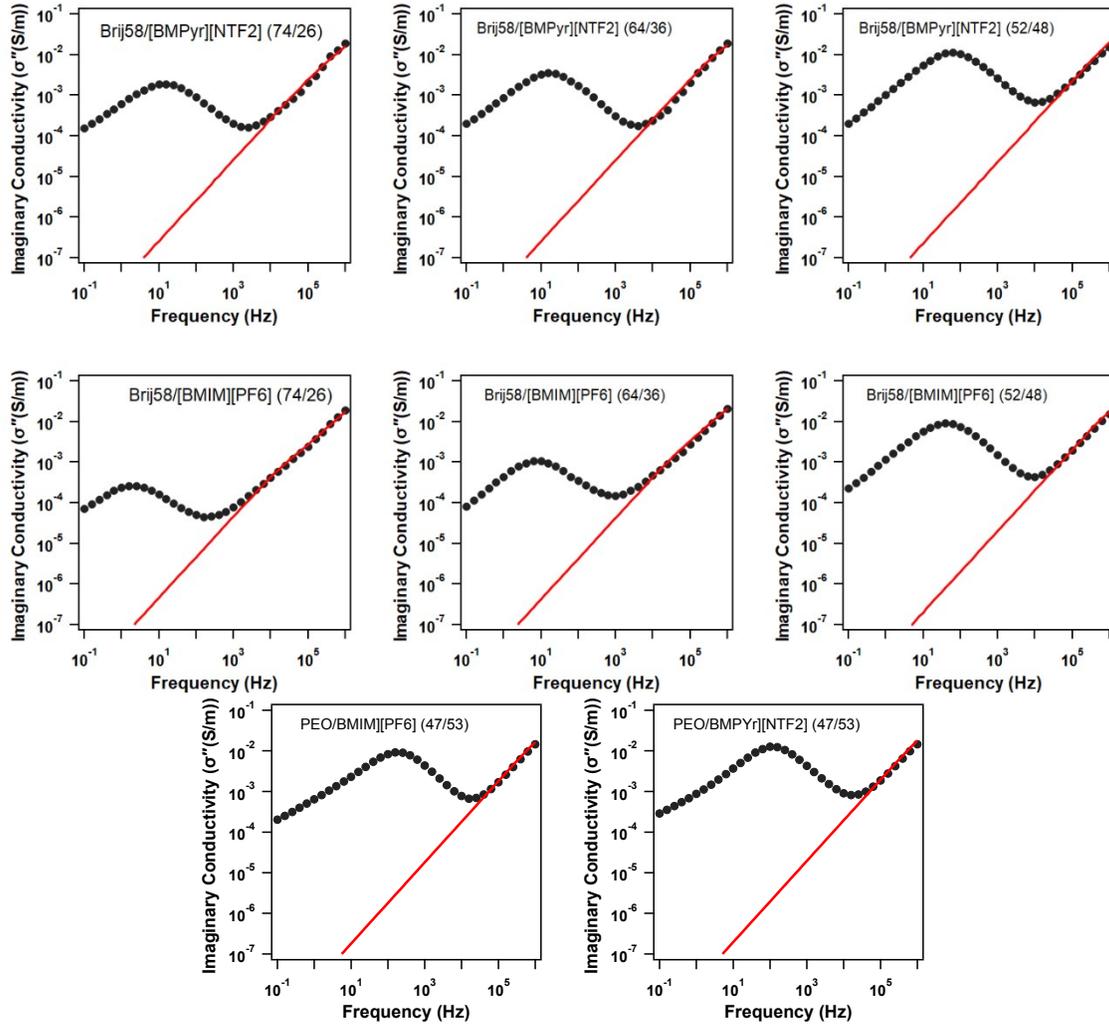


Figure S4. Imaginary parts of the complex conductivity spectrum  $\sigma^*(\omega)$ . The solid red line is the fit to the spectrum using eq (6) and (7).

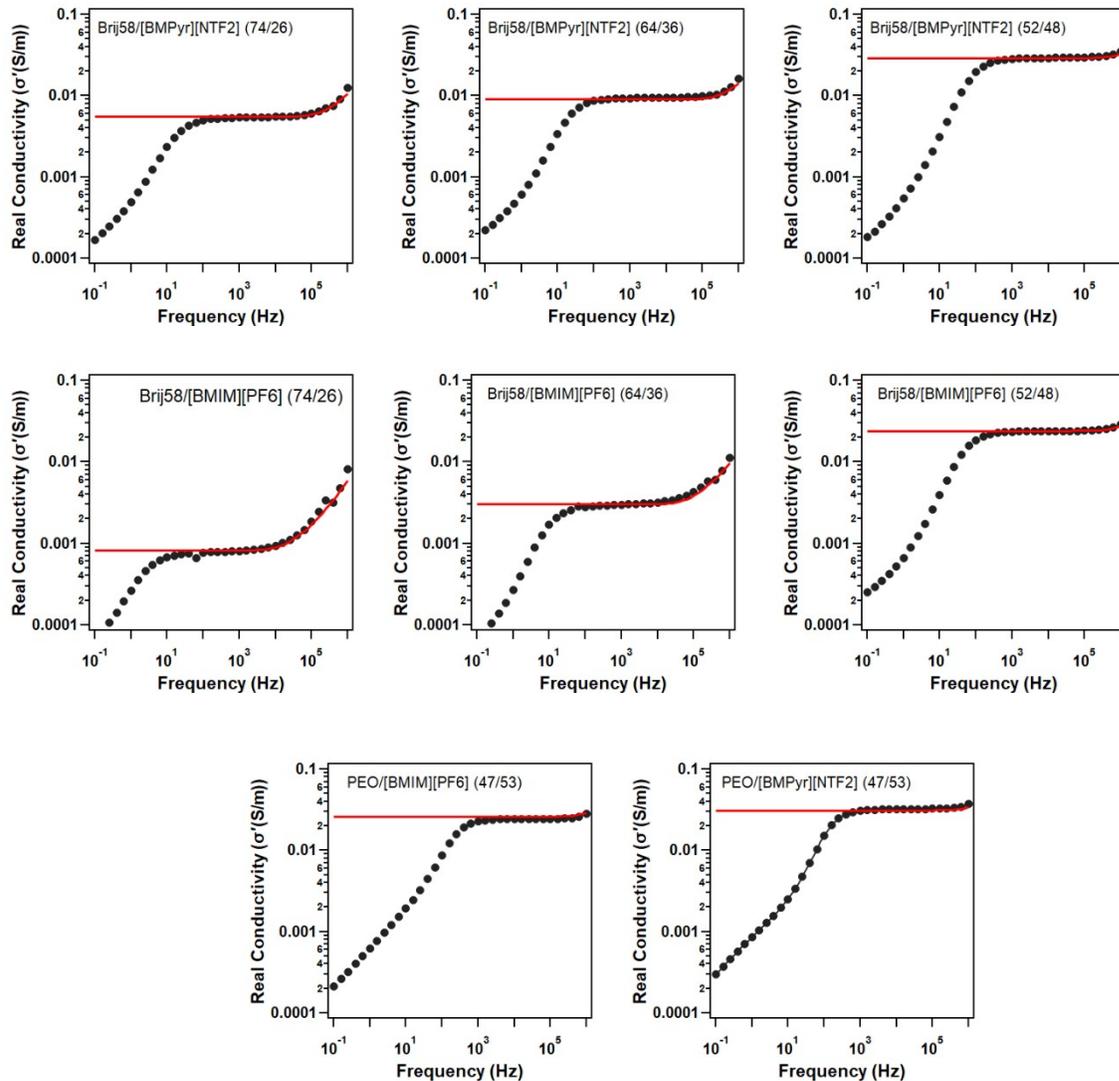


Figure S5. Real parts of the complex conductivity spectrum  $\sigma^*(\omega)$ . The solid red line is the fit to the spectrum using eq (6) and (7).

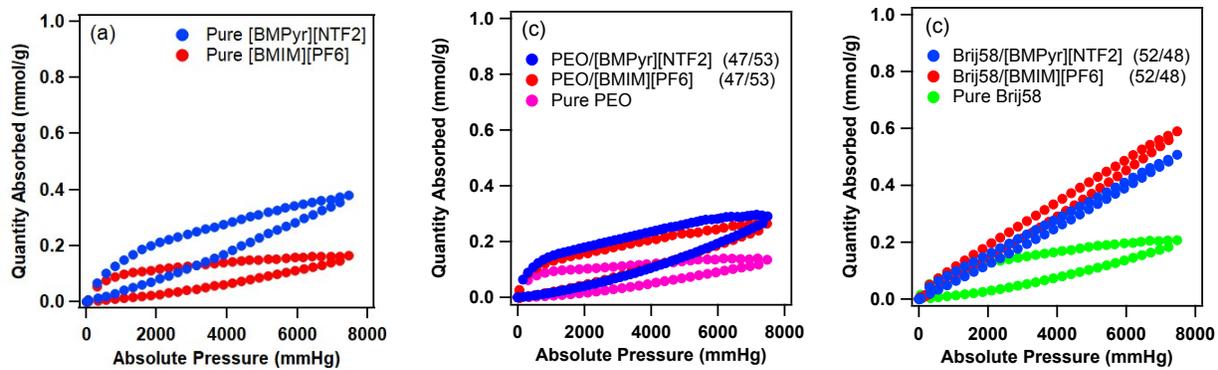


Figure S6. The CO<sub>2</sub> absorption-desorption of (a) pure ILs, (b) PEO/IL mixtures, and (c) ion gels.

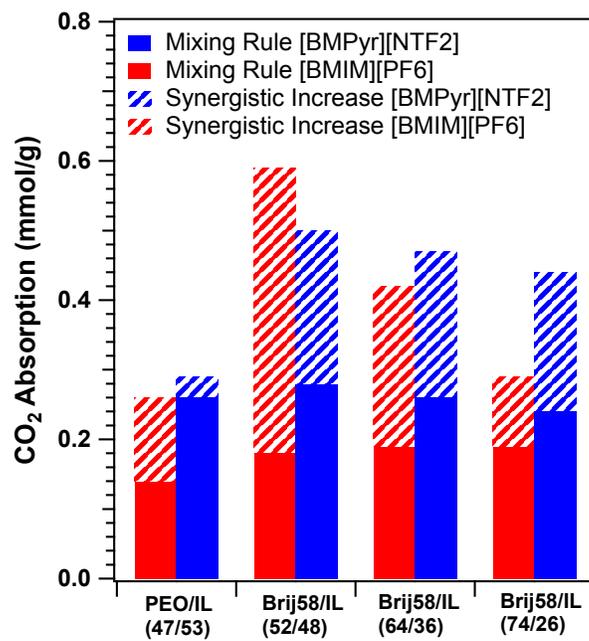


Figure S7. Total CO<sub>2</sub> absorption of LLC ion gels at 10 bar. The contributions from mixing rule and synergistic effect in total absorption are shown by solid fill and diagonal stripes, respectively.

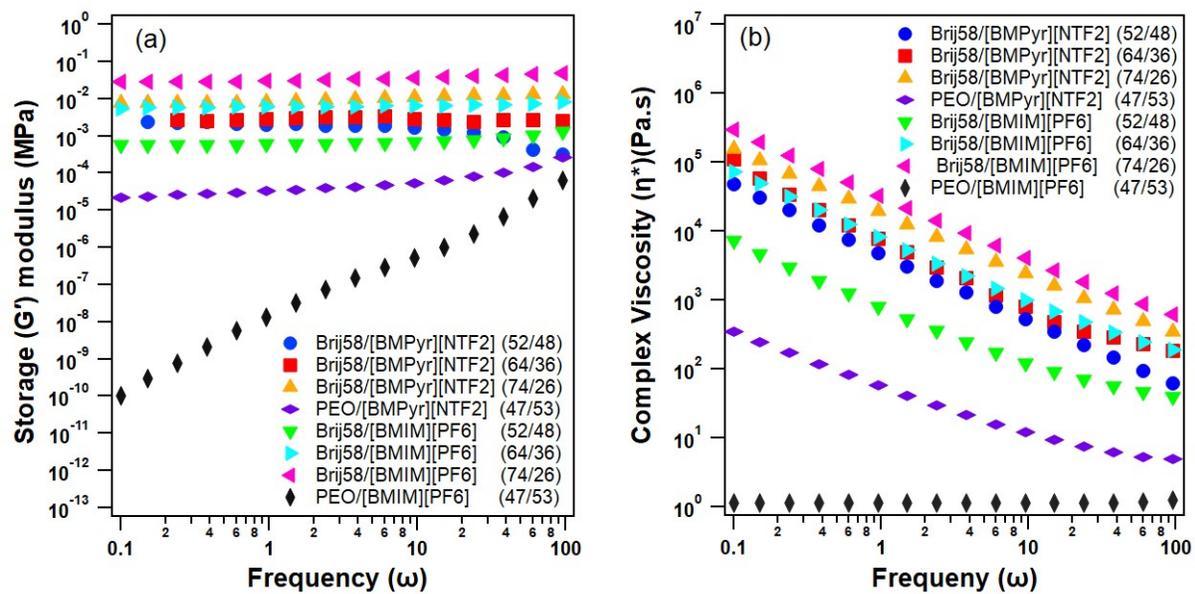


Figure S8. (a) Storage modulus and (b) complex viscosity of LLC ion gels and PEO/IL mixtures at 25 °C.