

Supplementary information

Influence of Lithium Salt-Induced Phase Separation on Thermal Behaviors of Poly(vinylidene fluoride)/Ionic liquid Gels and Pore/Void Formation by Competition with Crystallization

Noboru Osaka^{1*}, Yuichi Minematsu¹, Masatoshi Tosaka²

¹Department of Chemistry, Faculty of Science, Okayama University of Science, 1-1

Ridai-cho, Kita-ku, Okayama 700-0005, Japan

²Institute for Chemical Research, Kyoto University, Gokasho, Uji, Kyoto 611-0011,

Japan

* Corresponding author. E-mail: osaka@chem.ous.ac.jp

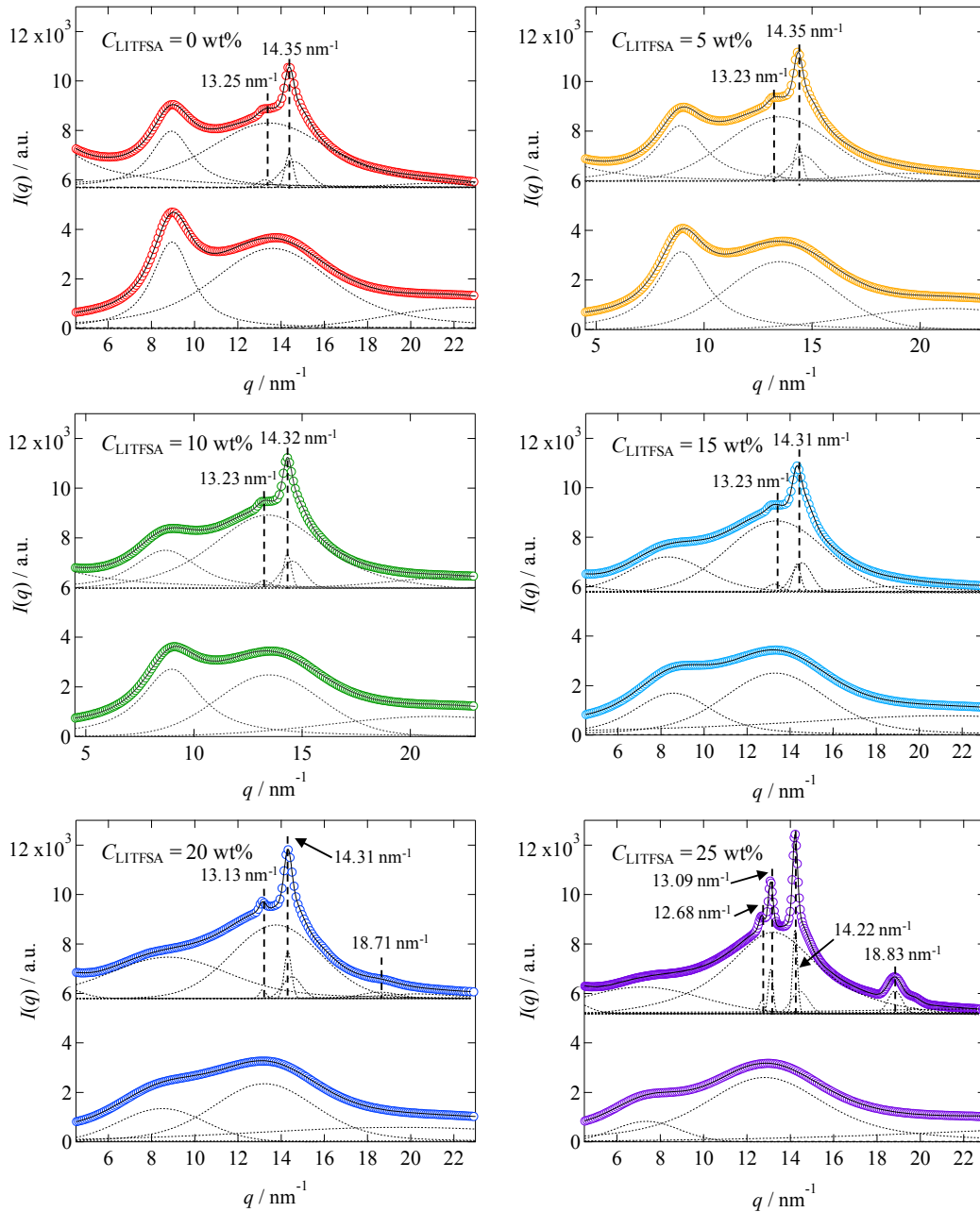


Figure S1. Results of fitting on the WAXS profiles of $[\text{C}_2\text{mim}][\text{TFSA}]/\text{LiTFSA}$ solutions (down) and $\text{PVDF}/[\text{C}_2\text{mim}][\text{TFSA}]/\text{LiTFSA}$ gels (top) depending on C_{LiTFSA} at 20 °C. The dotted curves for fits are Voigt functions for $[\text{C}_2\text{mim}][\text{TFSA}]$ and Gaussian functions for PVDF, respectively.

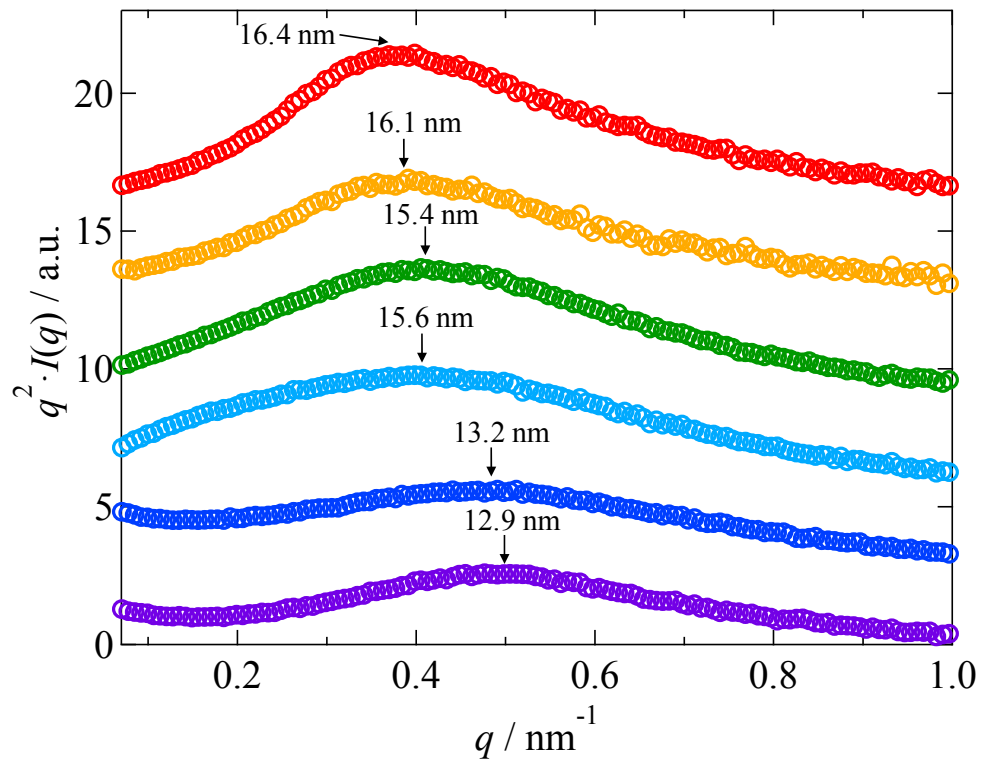


Figure S2. Lorentz-corrected SAXS curves of PVDF/[C₂mim][TFSA]/LiTFSA gels depending on C_{LiTFSA} obtained at 20 °C. The arrows indicate the peak positions and the corresponding periodic long distances obtained by Bragg's equation from the peak positions.

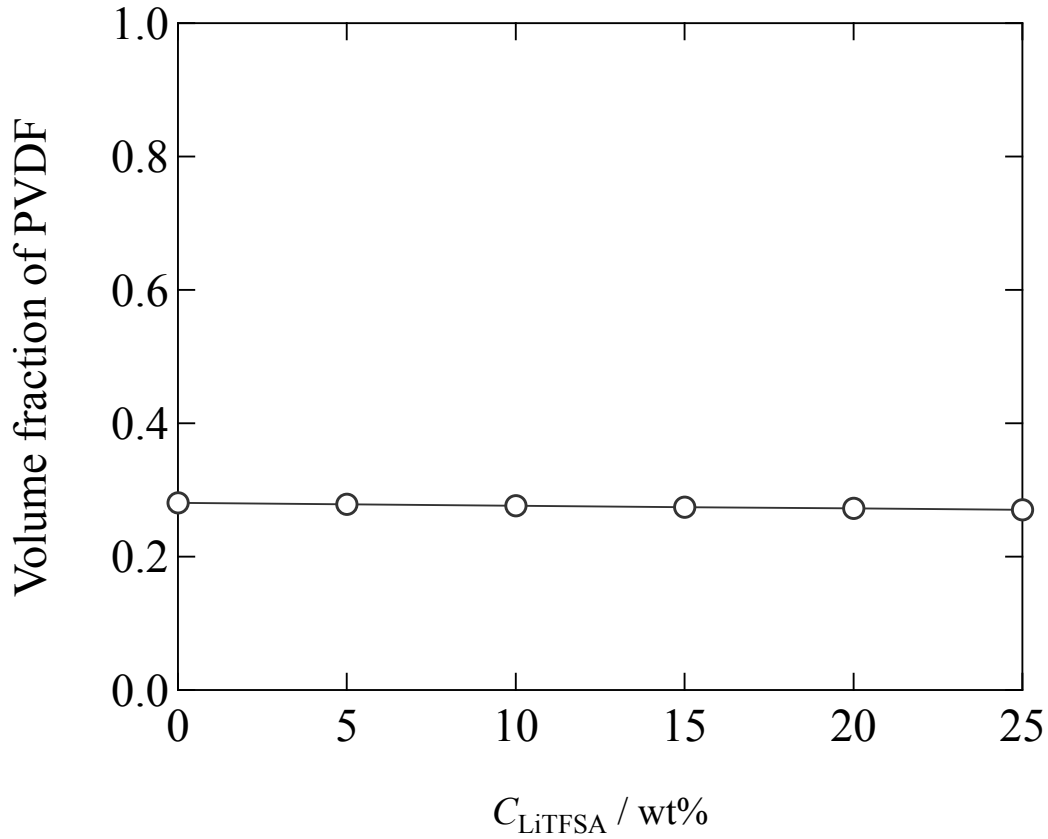


Figure S3. The estimated volume fractions of PVDF in PVDF/[C₂mim][TFSA]/LiTFSA gels depending on C_{LiTFSA} .

Volume fraction of PVDF was estimated by the following equation,

$$\text{volume fraction of PVDF} = \frac{\frac{\varphi_{PVDF}}{d_{PVDF}}}{\frac{\varphi_{PVDF}}{d_{PVDF}} + \frac{\varphi_{[C_2mim][TFSA]}}{d_{[C_2mim][TFSA]}} + \frac{\varphi_{LiTFSA}}{d_{LiTFSA}}}$$

φ_{PVDF} , $\varphi_{[C_2mim][TFSA]}$, and φ_{LiTFSA} are weight fractions of PVDF, [C₂mim][TFSA], and LiTFSA, respectively. d_{PVDF} , $d_{[C_2mim][TFSA]}$, and d_{LiTFSA} are densities of PVDF, [C₂mim][TFSA], and LiTFSA, respectively. d_{PVDF} is 1.68 g/cm³ for amorphous PVDF [S1]. $d_{[C_2mim][TFSA]}$ is 1.52 g/cm³ for [C₂mim][TFSA] [S2]. d_{LiTFSA} is 1.33 g/cm³ for LiTFSA [S3].

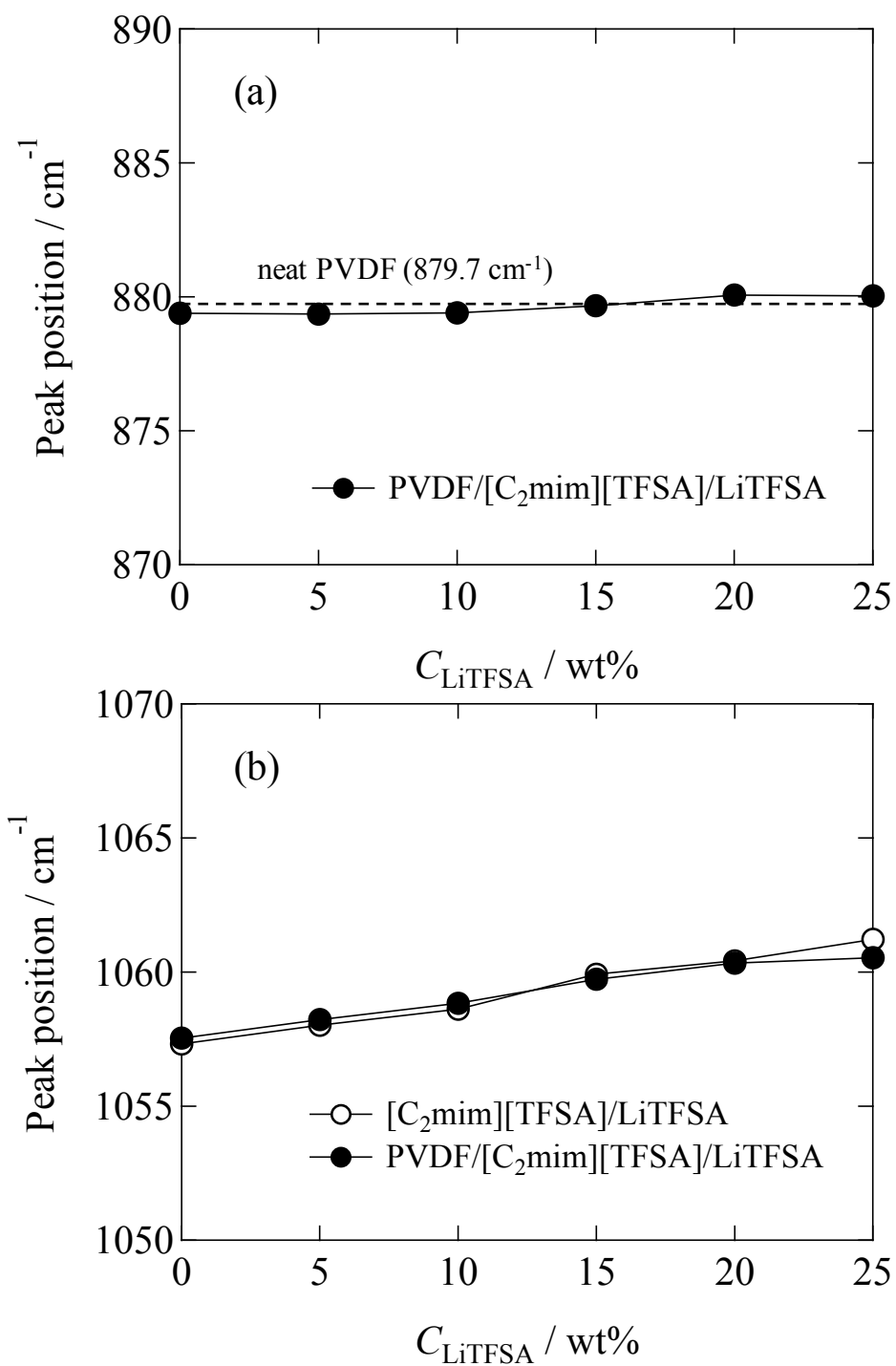


Figure S4. (a) Peak positions of FT-IR spectra depending on C_{LiTFSA} obtained at $185 \text{ }^\circ\text{C}$ in the regions of (a) $\text{CF}_2\text{-CH}_2$ bending vibration of PVDF and (b) SO_2 symmetric stretching vibration of [TFSA^-] anion interacting with cation.

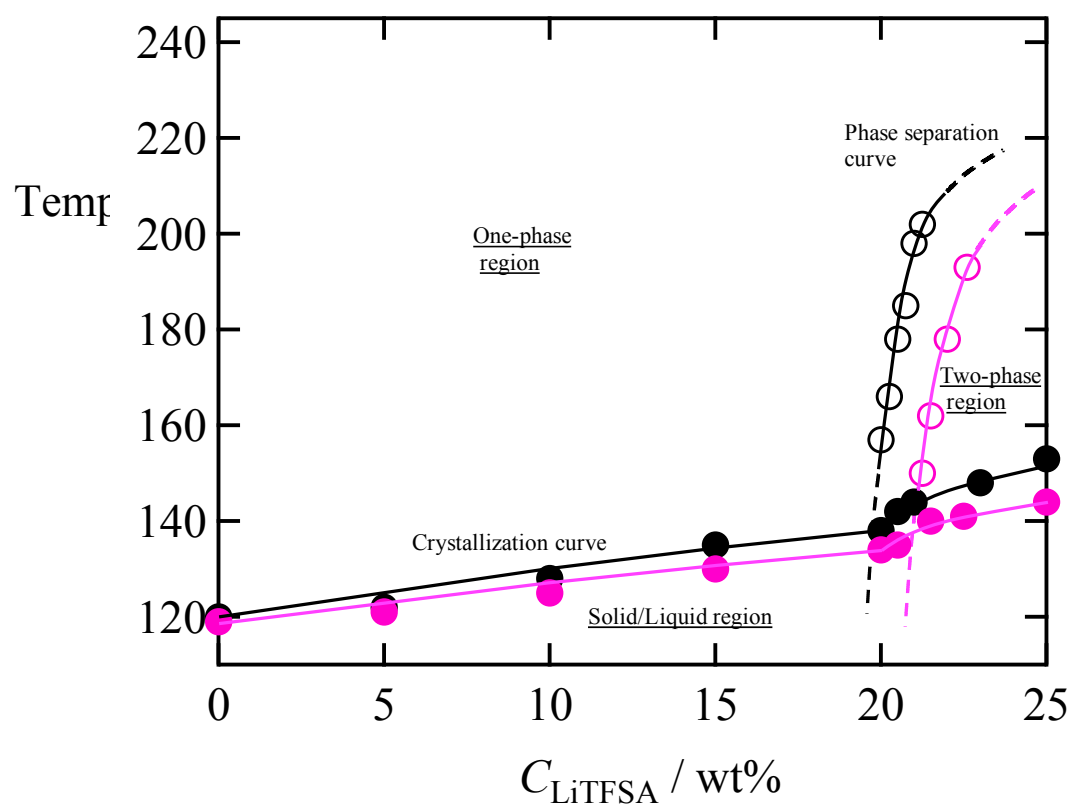


Figure S5. Summary of phase behaviors of the PVDF/[C₂mim][TFSA]/LiTFSA systems (black color) and the PVDF/[C₂dmim][TFSA]/LiTFSA systems (magenta color) obtained by cooling at 1 °C/min. Solid circles (●, ●) represent the crystallization temperatures and opaque circles (○, ○) represent the phase separation temperatures. The onset temperatures of the phase transitions were determined by using polarized optical microscope. [C₂dmim][TFSA] (99%) was purchased from IoLiTec.

References

- S1. Ameduri, B., *Chem Rev*, 2009, **109**, 6632-6686.
- S2. Kadyan, A.; Pandey, S., *J Chem Thermodyn*, 2018, **116**, 159-165.

S3. Lannelongue, P., Bouchal, R., Mourad, E., Bodin, C., Olarte, M., le Vot, S., Favier, F. and Fontaine, O., *J Electrochem Soc*, 2018, **165**, A657-663.