

Supplimentary Data

**Microphase-separated structures of ion gels consisting of ABA-type block copolymers and an ionic liquid: A key to escape from the trade-off between mechanical and transport properties**

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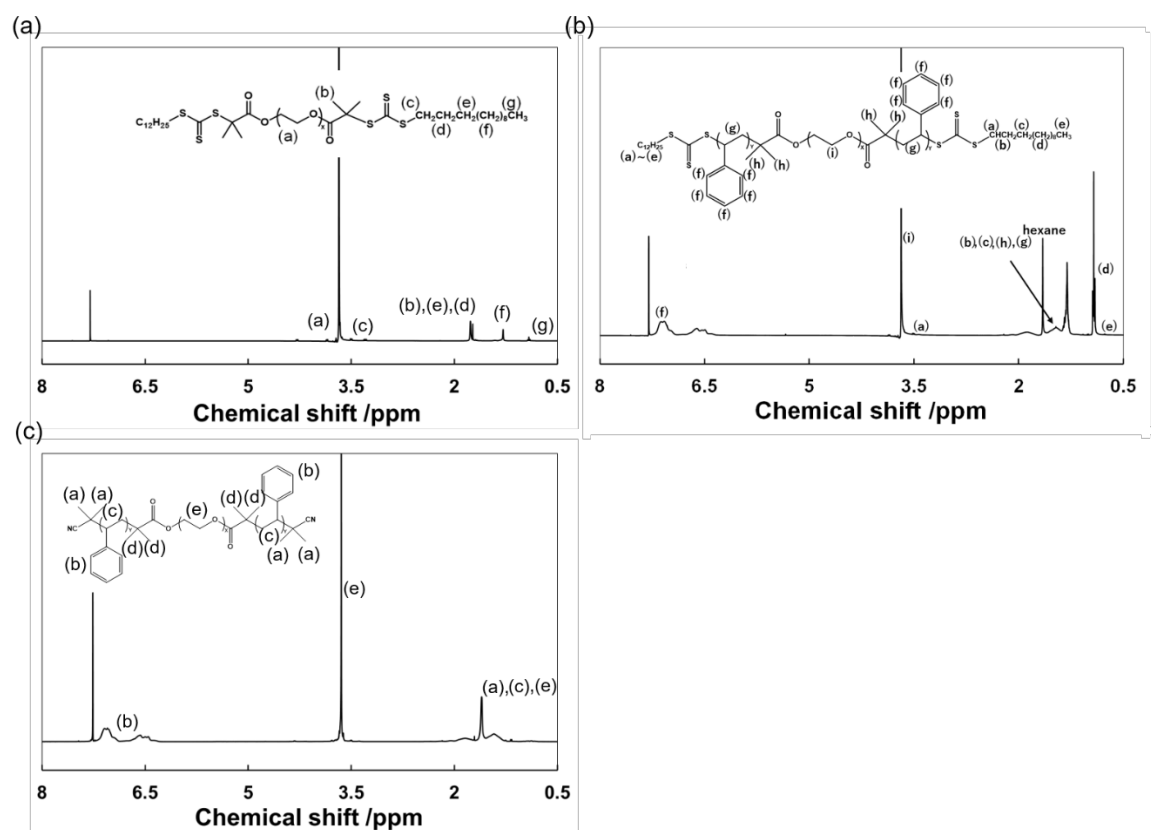
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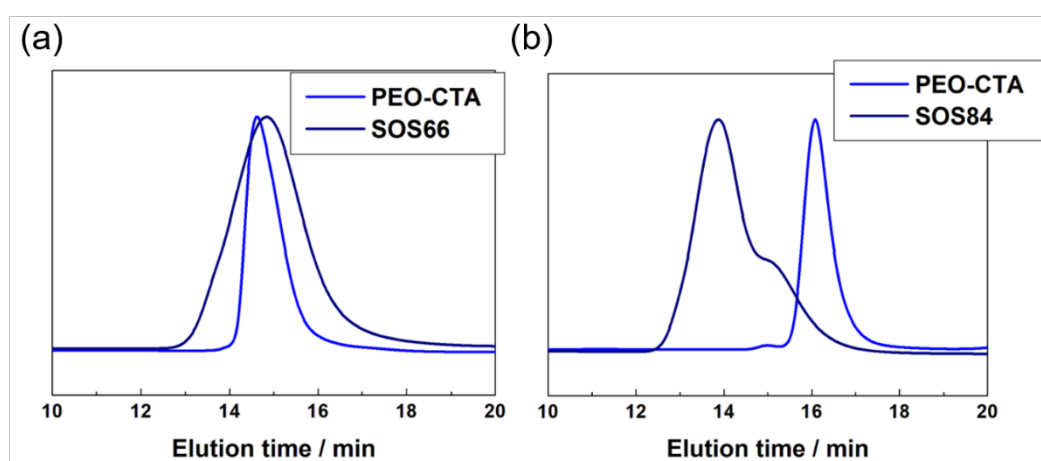
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**Figure S1**  $^1\text{H}$  NMR spectra of (a) PEO-CTA, (b) CTA-PSt-*b*-PEO-*b*-PSt-CTA, and (c) PSt-*b*-PEO-*b*-PSt in  $\text{CDCl}_3$ .



**Figure S2.** GPC curves of (a) SOS-66 and (b) SOS-84 (eluent: THF, detector: RID, standard: PSt).

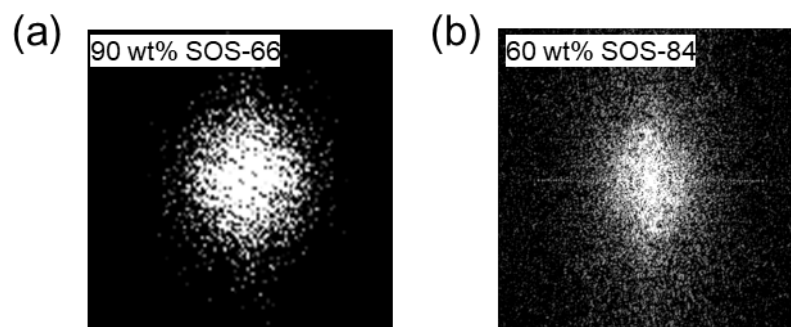
**Table S1.** Composition,  $\phi_{\text{soft}}$ , and microstructure of ion gels.

<b>Polymer</b>	<b>Polymer content (wt%)</b>	<b><math>\phi_{\text{soft}}</math> (%)</b>	<b>Microstructure <sup>1)</sup></b>
SOS-66	40	68.5	S <sup>3)</sup>
	80	44.1	H <sup>R</sup> <sup>2,3)</sup>
	90	38.9	G <sup>2,3)</sup>
SOS-84	40	59.2	L <sup>2,3)</sup>
	60	42.9	G + L <sup>2,3)</sup>
	70	35.6	H <sup>2)</sup>
	80	28.7	S <sup>R</sup> <sup>2)</sup>

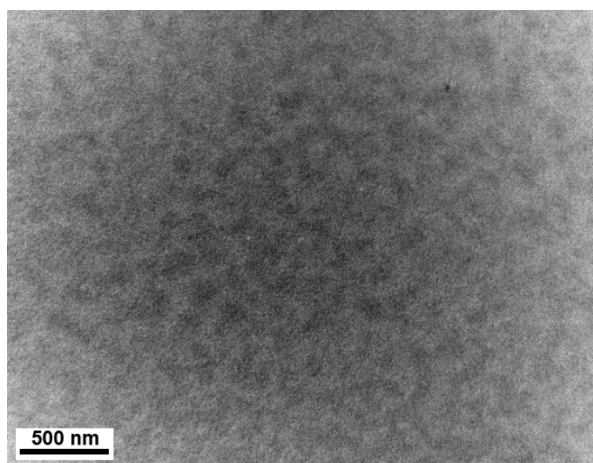
1) S: spheres; S<sup>R</sup>: reverse S; H: hexagonally-packed cylinders; H<sup>R</sup>: reverse H; G: gyroids; L: lamellas.

2) Assigned by AFM measurements.

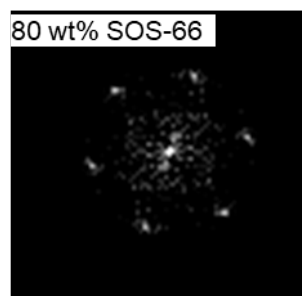
3) Assigned by SAXS measurements.



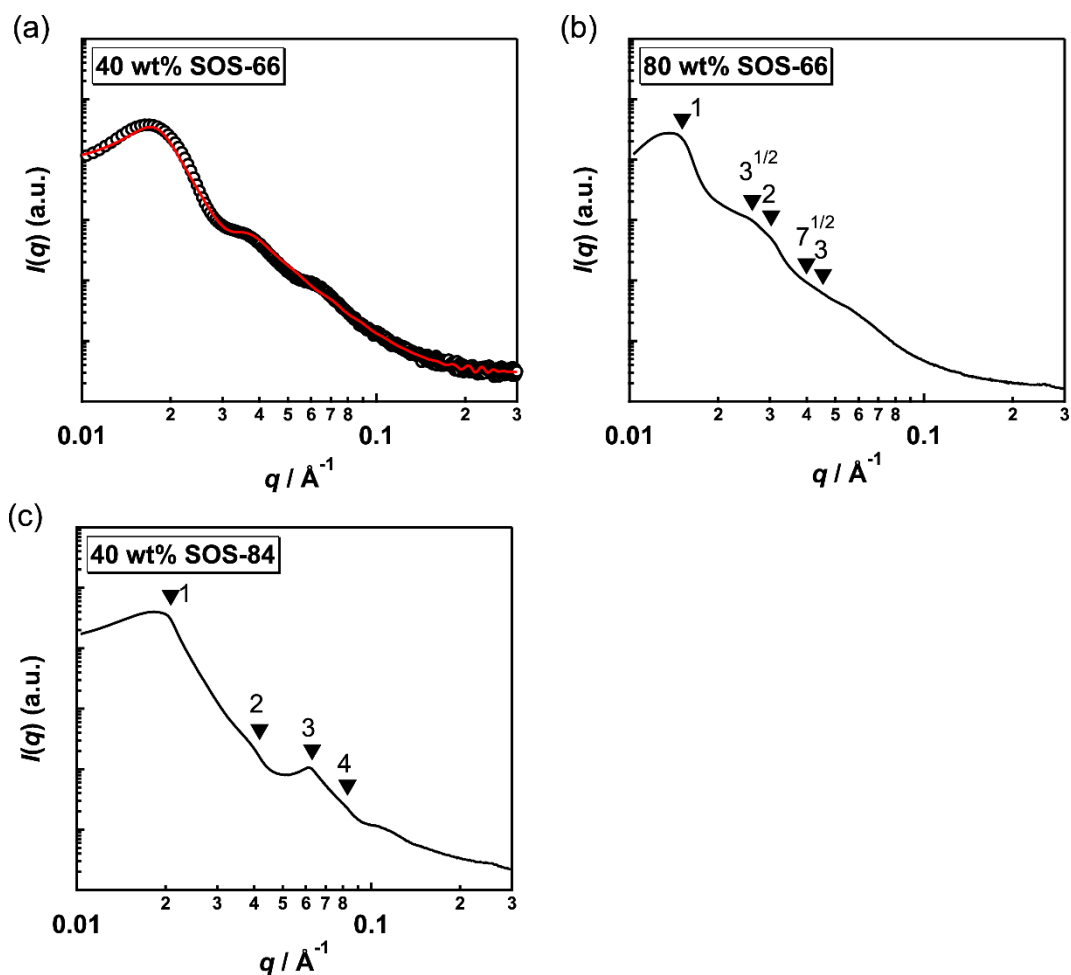
**Figure S3.** FFT images (500 nm×500nm) of (a) 90 wt% SOS-66/[C<sub>2</sub>mim][NTf<sub>2</sub>] and (b) 60 wt% SOS-84/[C<sub>2</sub>mim][NTf<sub>2</sub>] ion gels.



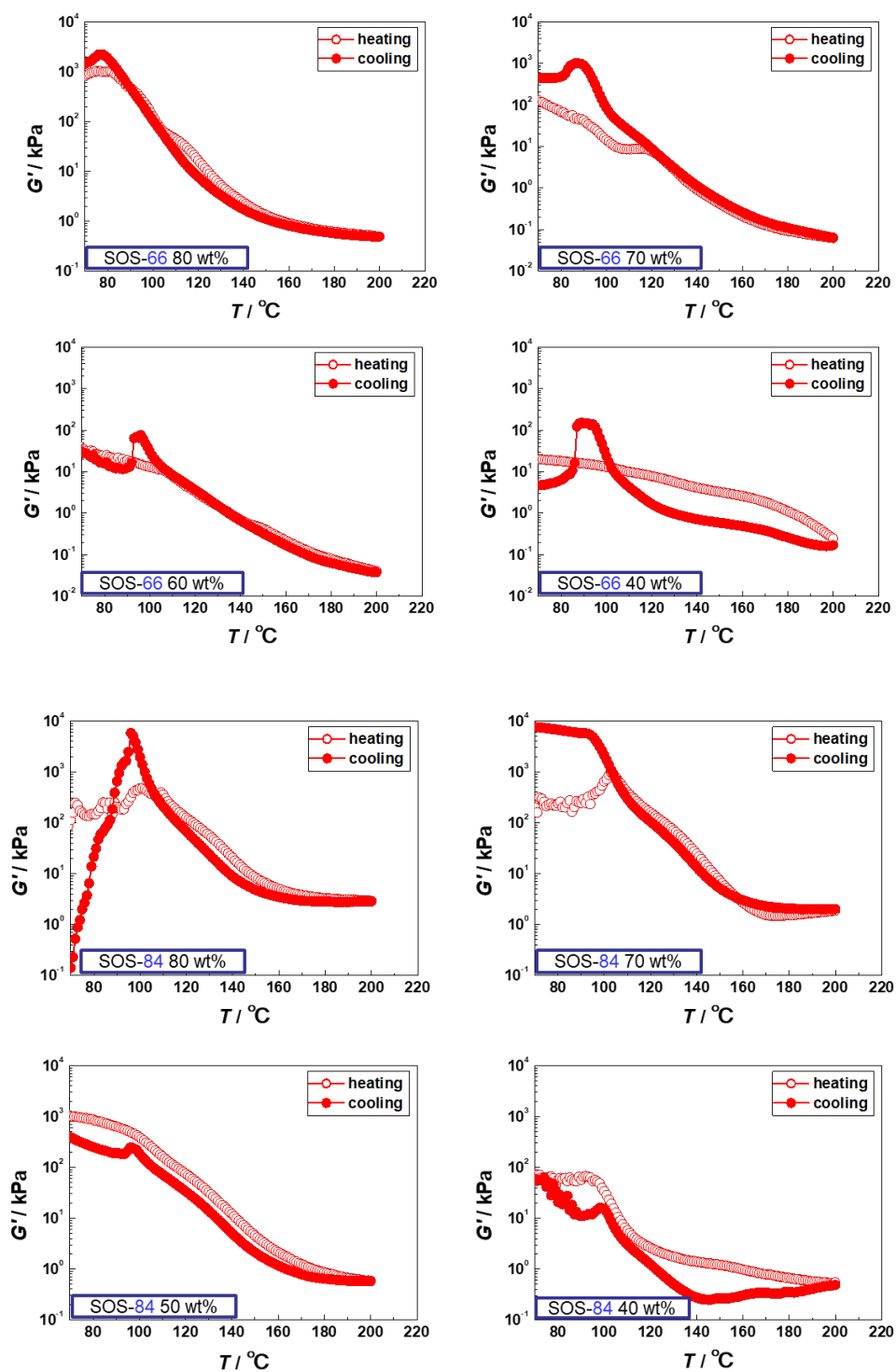
**Figure S4.** TEM image of 60 wt% SOS-84/[C<sub>2</sub>mim][NTf<sub>2</sub>] ion gel, obtained by JEM-1010 (JEOL, Japan) using a single-tilt sample holder. The ion gel sample was dissolved in THF to prepare the 0.5 wt % SOS ion gel/THF solution. The solution was cast onto the carboncoated copper substrate and evaporated at room temperature. The THF was completely dried under vacuum to prepare a thin ion gel sample (~several hundred nm). The measurements were performed at room temperature.



**Figure S5.** FFT images (500 nm×500nm) of 80 wt% SOS-66/[C<sub>2</sub>mim][NTf<sub>2</sub>].



**Figure S6.** SAXS profile of SOS/[C<sub>2</sub>mim][NTf<sub>2</sub>] ion gel with various polymer content. The profiles were assigned to (a) S, (b) H, and (c) L. A fitting line for S structure corresponds to the Percus-Yevick model, which assumes spherical cores without a highly-ordered packing structure. [S1]



**Figure S7.** Temperature sweep measurements of  $G'$  for SOS-66/[C<sub>2</sub>mim][NTf<sub>2</sub>] and SOS-84/[C<sub>2</sub>mim][NTf<sub>2</sub>].

**Table S2.** Structure,  $\sigma$ , and  $\sigma_0$  at 100 °C of ion gels with various polymer content.

<b>Polymer / wt%</b>	<b>Structure</b>	<b><math>\sigma</math><sup>1)</sup> / mS cm<sup>-1</sup></b>	<b><math>\sigma_0</math><sup>2)</sup> / mS cm<sup>-1</sup></b>
SOS-66 / 40 wt%	S	5.73	9.73
SOS-66 / 80 wt%	H <sup>R</sup>	0.014	2.56
SOS-84 / 40 wt%	L	2.51	13.1
SOS-84 / 60 wt%	G + L	0.37	9.25
SOS-84 / 70 wt%	H	0.023	8.29
SOS-84 / 80 wt%	S <sup>R</sup>	0.0066	5.45

- 1) Ionic conductivity of ion gels consisting of SOS and [C<sub>2</sub>mim][NTf<sub>2</sub>].
- 2) Ionic conductivity of [C<sub>2</sub>mim][NTf<sub>2</sub>] solutions of PEO containing the same volume fraction of PEO as the corresponding ion gels.

#### REFERENCES

[S1] K. Hashimoto, M. Hirasawa, H. Kokubo, R. Tamate, X. Li, M. Shibayama, M. Watanabe, Transport and Mechanical Properties of ABA-type Triblock Copolymer Ion Gels Correlated with Their Microstructures, *Macromolecules*, **52**, 8430-8439 (2019). doi: 10.1021/acs.macromol.9b01907.