Electronic Supplementary Information (ESI) for

Controlled mechanical properties of poly(ionic liquid)-based hydrophobic ion gels by the introduction of alumina nanoparticles with different shapes

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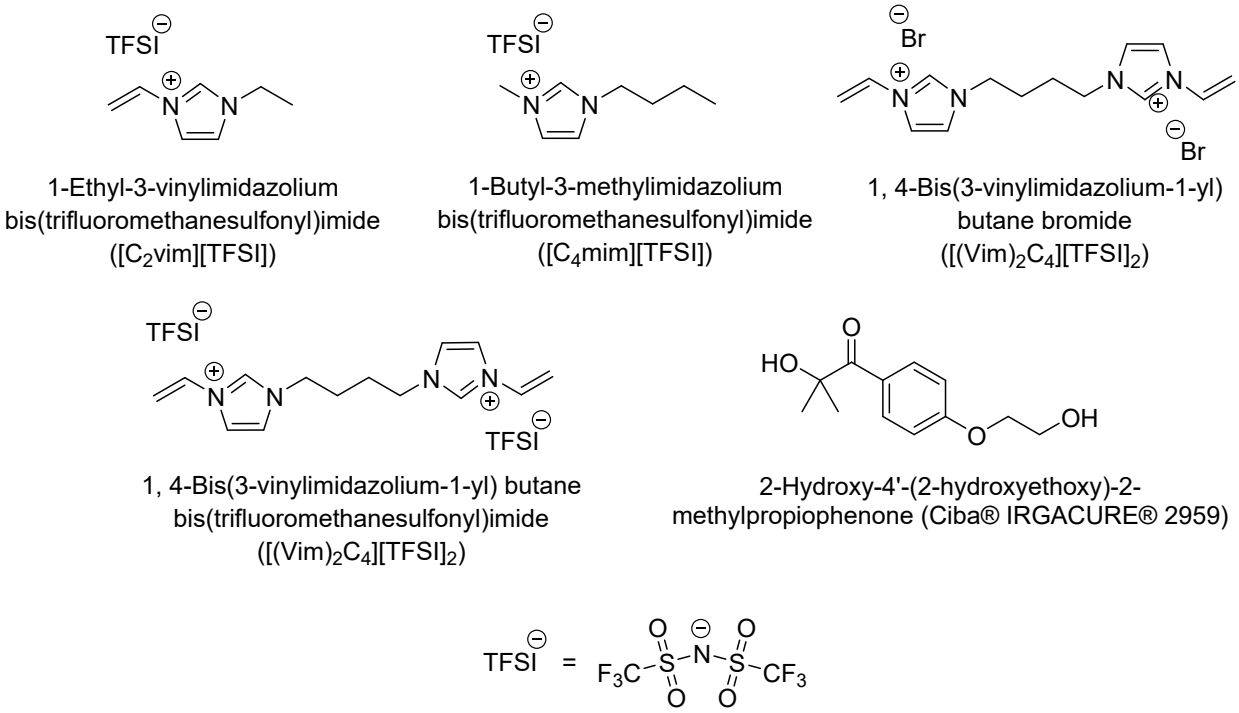
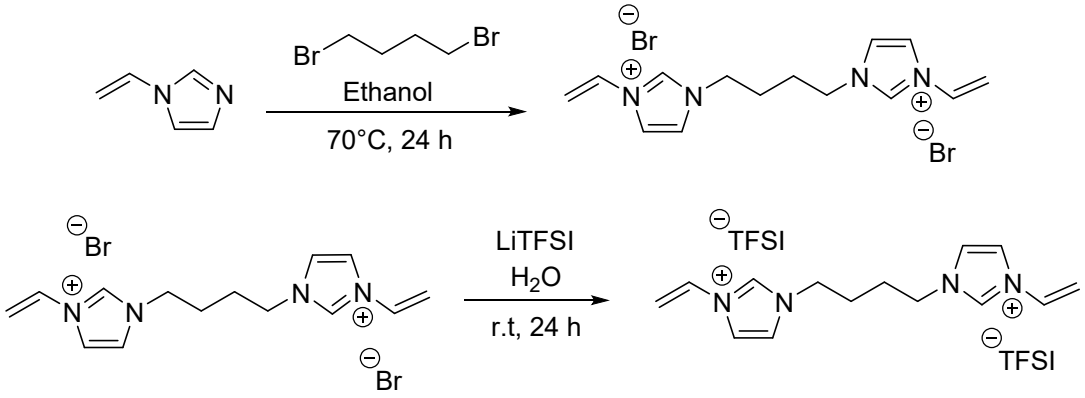


Figure S1 Chemical structures of reagents used for preparing PIL ion gels.

Scheme S1 Synthesis of $[(Vim)_2C_4]Br_2$ and $[(Vim)_2C_4][TFSI]_2$



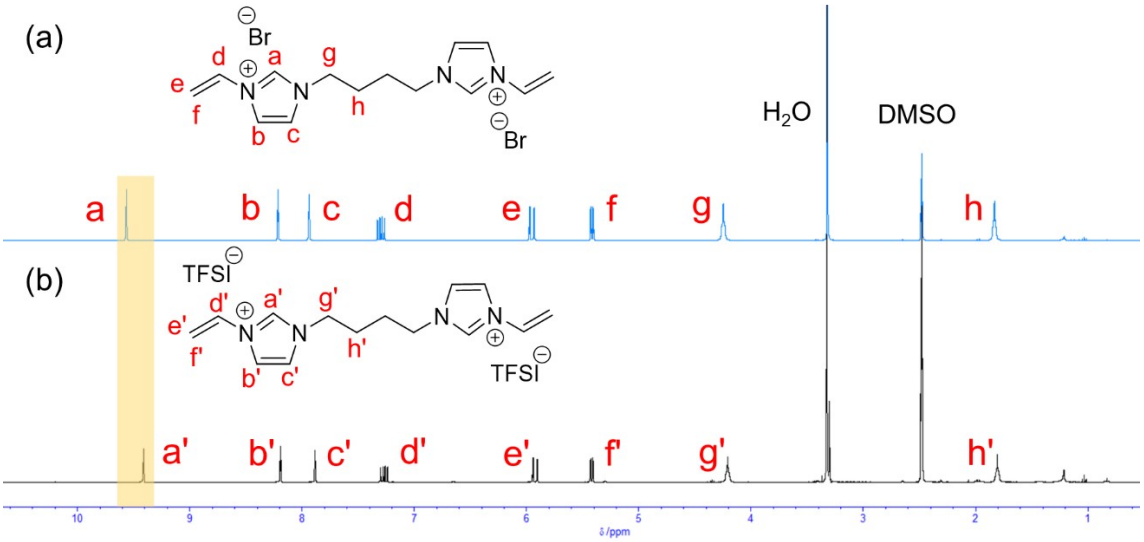


Figure S2 ^{1}H NMR spectra of (a) $[(Vim)_2C_4]Br_2$ and (b) $[(Vim)_2C_4][TFSI]_2$. (Solvent: DMSO-d_6).

Table S1 Amount of reagent used for spherical alumina/[C_4mim][TFSI]/Ethyl acetate (EA) dispersion.

IL [C_4mim][TFSI]	Total EA	Alumina	
[g]	[mL]	[g]	[vol%]*1
3.5	1.2	0.12	0.85
3.5	1.2	0.24	1.7
3.5	1.2	0.36	2.5

*1 Alumina [vol%] is a concentration for the whole solution.

Table S2 Amount of reagent used for rod-shaped alumina/[C_4mim][TFSI]/EA dispersion.

IL [C_4mim][TFSI]	EA dispersed alumina*1	Evaporated EA	Added EA	Total EA	Alumina	
[g]	[g]	[mL]	[mL]	[mL]	[g]	[vol%]*2
3.5	1.2	0.66	0.59	1.2	0.12	0.85
3.5	2.4	1.8	0.57	1.2	0.24	1.7
3.5	3.6	2.7	0.26	1.2	0.36	2.5

*1 Alumina concentration in EA dispersed alumina is 10 wt%.

*2 Alumina [vol%] is a concentration for the whole solution.

Table S3 Amount of reagent used for PIL SN ion gels and spherical or rod-shaped alumina/PIL DN ion gels with different crosslinker concentrations.

Monomer	Cross-linking agent*1		Initiator*2	IL	Total	Alumina*3		
$[C_2vim][TFSI]$	$[(Vim)_2C_4][TFSI]_2$		Irgacure2959	$[C_4mim][TFSI]$	EA			
[g]	[g]	[mol%]	[g]	[g]	[mL]	Shape	[g]	[wt%]
3.0	0.021	0.35	0.0016	3.5	1.2	-	0	0
3.0	0.042	0.70	0.0016	3.5	1.2	-	0	0
3.0	0.060	1.0	0.0016	3.5	1.2	-	0	0
3.0	0.021	0.35	0.0016	3.5	1.2	Spherical	0.24	8.0
3.0	0.042	0.70	0.0016	3.5	1.2	Spherical	0.24	8.0
3.0	0.060	1.0	0.0016	3.5	1.2	Spherical	0.24	8.0
3.0	0.021	0.35	0.0016	3.5	1.2	Rod-shaped	0.24	8.0
3.0	0.042	0.70	0.0016	3.5	1.2	Rod-shaped	0.24	8.0
3.0	0.060	1.0	0.0016	3.5	1.2	Rod-shaped	0.24	8.0

*1 Cross-linking agent [mol%] is monomer basis.

*2 Initiator is 0.1 mol% on monomer basis.

*3 Alumina [wt%] is PIL network (monomer + cross-linking agent + initiator) basis.

Table S4 Amount of reagent used for spherical alumina/PIL DN ion gels with different alumina concentrations.

Monomer	Cross-linking agent*1	Initiator*2	IL	Total	Alumina*3	
$[C_2vim][TFSI]$	$[(Vim)_2C_4][TFSI]_2$	Irgacure2959	$[C_4mim][TFSI]$	EA		
[g]	[g]	[g]	[g]	[mL]	[g]	[wt%]
3.0	0.021	0.0016	3.5	1.2	0	0
3.0	0.021	0.0016	3.5	1.2	0.12	4.0
3.0	0.021	0.0016	3.5	1.2	0.18	6.0
3.0	0.021	0.0016	3.5	1.2	0.24	8.0
3.0	0.021	0.0016	3.5	1.2	0.30	10
3.0	0.021	0.0016	3.5	1.2	0.36	12
3.0	0.021	0.0016	3.5	1.2	0.42	14

*1 Cross-linking agent is 0.35 mol% on monomer basis.

*2 Initiator is 0.1 mol% on monomer basis.

*3 Alumina [wt%] is PIL network (monomer + cross-linking agent + initiator) basis.

Table S5 Amount of reagent used for rod-shaped alumina/PIL DN ion gels with different alumina concentrations.

Monomer	Cross-linking agent*1	Initiator*2	IL	EA dispersed alumina*3	Total	Alumina*4	
$[C_2vim][TFSI]$	$[(Vim)_2C_4][TFSI]_2$	Irgacure2959	$[C_4mim][TFSI]$		EA		
[g]	[g]	[g]	[g]	[g]	[mL]	[g]	[wt%]
3.0	0.021	0.0016	3.5	1.2	1.2	0.12	4.0
3.0	0.021	0.0016	3.5	1.8	1.2	0.18	6.0
3.0	0.021	0.0016	3.5	2.4	1.2	0.24	8.0
3.0	0.021	0.0016	3.5	3.0	1.2	0.30	10
3.0	0.021	0.0016	3.5	3.6	1.2	0.36	12
3.0	0.021	0.0016	3.5	4.2	1.2	0.42	14

*1 Cross-linking agent is 0.35 mol% on monomer basis.

*2 Initiator is 0.1 mol% on monomer basis.

*3 Alumina concentration in EA dispersed alumina is 10 wt%.

*4 Alumina [wt%] is PIL network (monomer + cross-linking agent + initiator) basis.

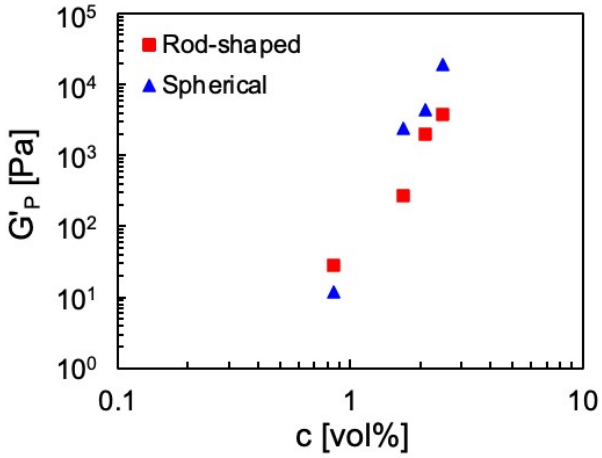
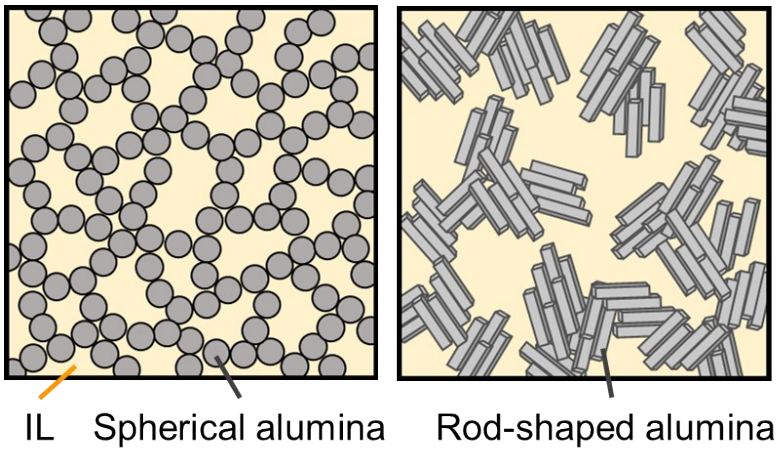


Figure S3 Values of the low frequency plateau of the storage modulus (G'_P) at versus the volume concentration of alumina in alumina/[C_4mim][TFSI]/EA dispersion (strain = 0.1%, frequency = 1 Hz).



Scheme S2 Schematic illustrations of alumina dispersed in [C_4mim][TFSI]/EA solution.

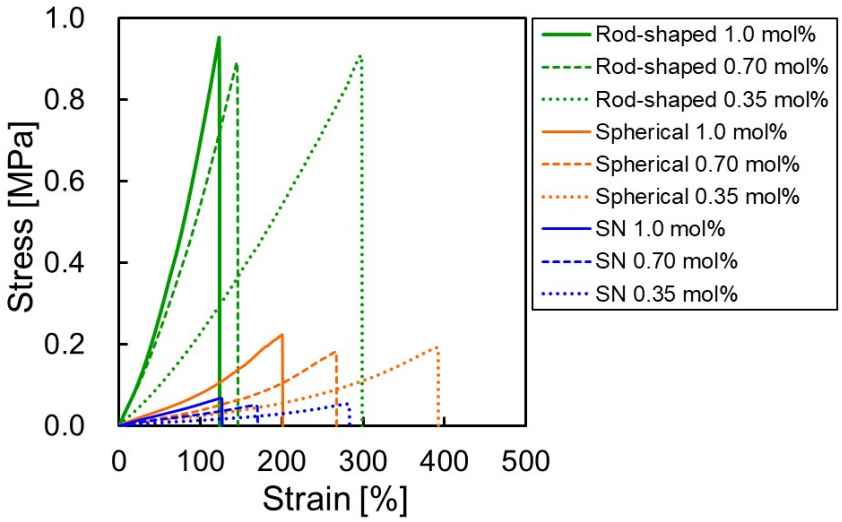


Figure S4 Stress–strain curves of SN ion gels, spherical and rod-shaped alumina/PIL DN ion gels with different crosslinker concentrations. The alumina concentration was fixed at 8 wt% relative to the PIL network.

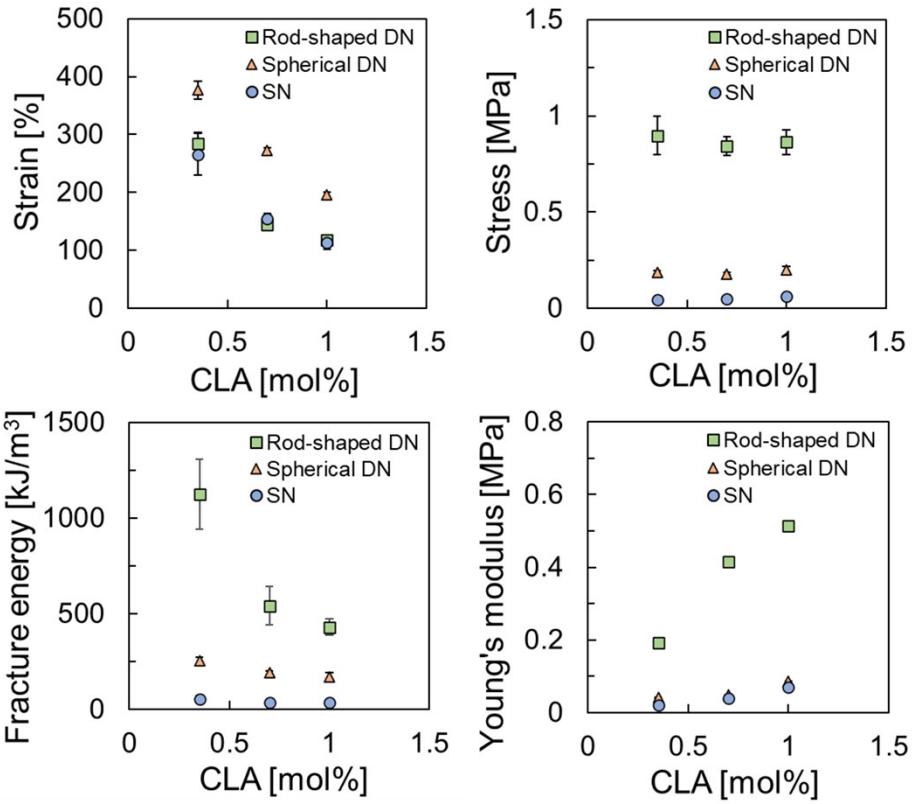


Figure S5 Mechanical properties of SN ion gels, spherical, and rod-shaped alumina/PIL DN ion gels with different crosslinker concentrations. The alumina concentration was fixed at 8 wt% relative to the PIL network.

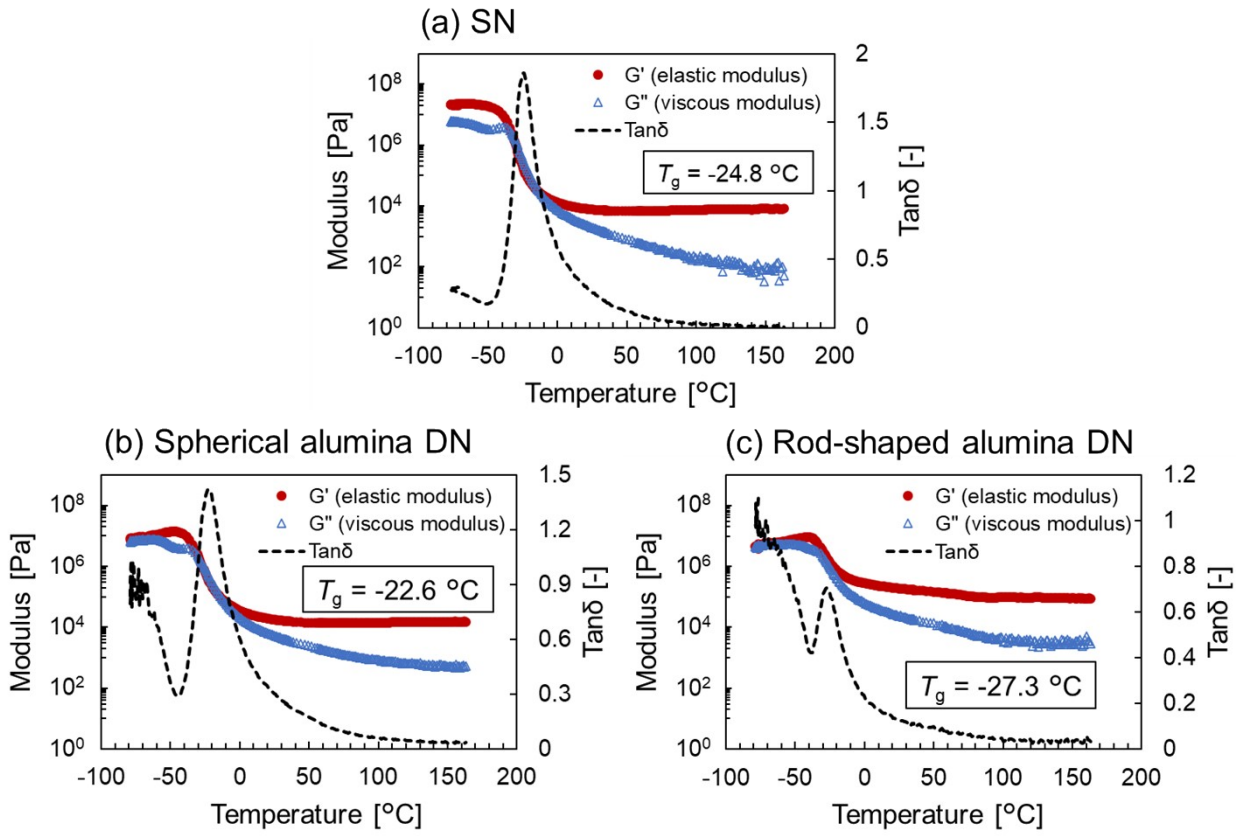


Figure S6 Oscillatory shear temperature-sweep viscoelasticity measurements of (a) PIL SN, (b) spherical alumina/PIL DN, and (c) rod-shaped alumina/PIL DN ion gels.

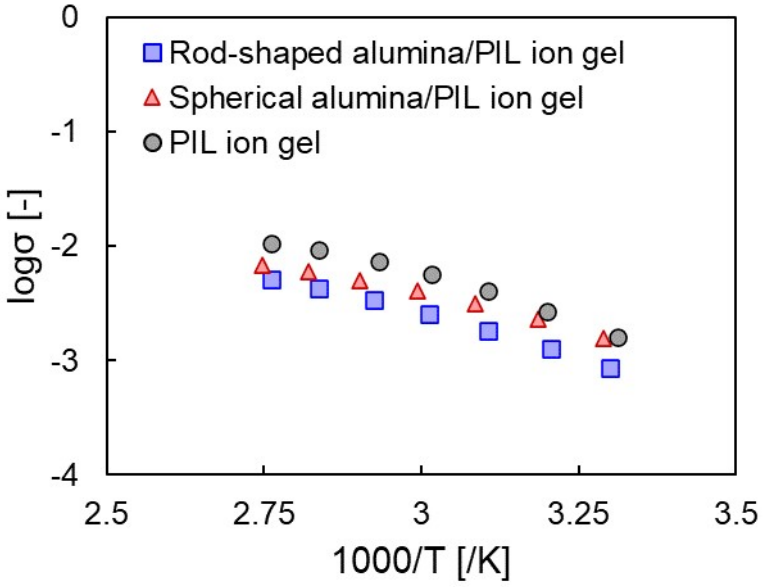


Figure S7 Ionic conductivity (σ) of PIL DN ion gels with different shapes of alumina and PIL SN ion gel.