

Self-healable poly(ionic liquid) ion gel via vacuum polymerization

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(Introduction) Ionic liquids (ILs) are molten salts with properties such as non-volatility, non-flammability, and high ionic conductivity. We recently developed ion gels composed of physically entangled polymers swollen with ILs, exhibiting stretchability and self-healing^[1,2]. In this study, we report a novel ion gel (IG) based on a poly(ionic liquid) (PIL) with physical network. The gel is synthesized via one-step *in situ* polymerization of an ionic monomer in an IL under vacuum, achieving nearly 100% monomer conversion. We discuss the polymerization process and mechanical properties of the resulting entangled PIL ion gel.

(Experimental Section) We selected *N*-[2-(methacryloyloxy)ethyl]-*N,N*-dimethylbutan-1-aminium trifluoromethylsulfonylimide ([N1142MA][TFSI]) and 1-ethyl-3-methylimidazolium trifluoromethylsulfonylimide ([C₂mim][TFSI]) as monomer and solvent for the IG, respectively. For the self-supporting IGs, their mechanical properties were evaluated by dynamic viscoelasticity measurements and tensile tests.

(Results & Discussion) Fig. 1(a) shows the effect of AIBN concentration on the monomer conversion during polymerization of [N1142MA][TFSI] in [C₂mim][TFSI]. Under vacuum (~0.67 Pa), the conversion remained nearly 100% even at 0.67 mM AIBN, in contrast to the lower conversion under ambient pressure with Ar. *M_w* reached 237 kDa(0.67 mM), forming the IG without any chemical crosslinkers was self-standing, indicating physical entanglements of PIL chains. This was enabled by the non-volatility of both the monomer and solvent. As shown in Fig. 1(b), the IG exhibited excellent elasticity with over 1000% elongation and fully self-healed within 30 minutes—faster than previously reported highly entangled neutral polymer-based IG^[1,2].

(References)

[1] Y. Kamiyama *et al.*, *Sci. Adv.*, **2022**, 8, eadd0226. [2] Y. Kamiyama *et al.*, *Soft Matter*, **2022**, 18, 8582.



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I have been engaged in the development of thermoresponsive polymer in IL systems since my master's studies(Mai K-U, *et.al. Chem.comn.*, **2025** Web on release), and currently focus on Highly entangled poly(ionic liquid) ion gels as a Ph.D. candidate at Hokkaido University and National Institute Materials of Science.

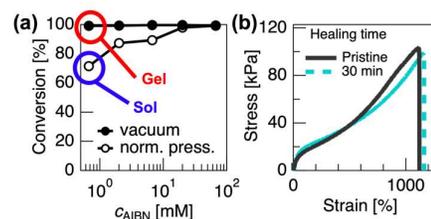


Fig. 1(a) Relationship between C_{AIBN} and [N1142MA][TFSI] monomer conversion polymerized in [C₂mim][TFSI] at 80 °C under vacuum (closed circles) and under normal pressure filled with Ar (opened circles). **(b)** Stress–strain curves for the ion gels. (Black: UHMW PIL pristine gel, and Green: UHMW PIL self-healed gel; healing time is 30 min.)