

Supporting Information

**Tailoring intermolecular interactions in ion gels with rationally designed
phosphonic acid polymers**

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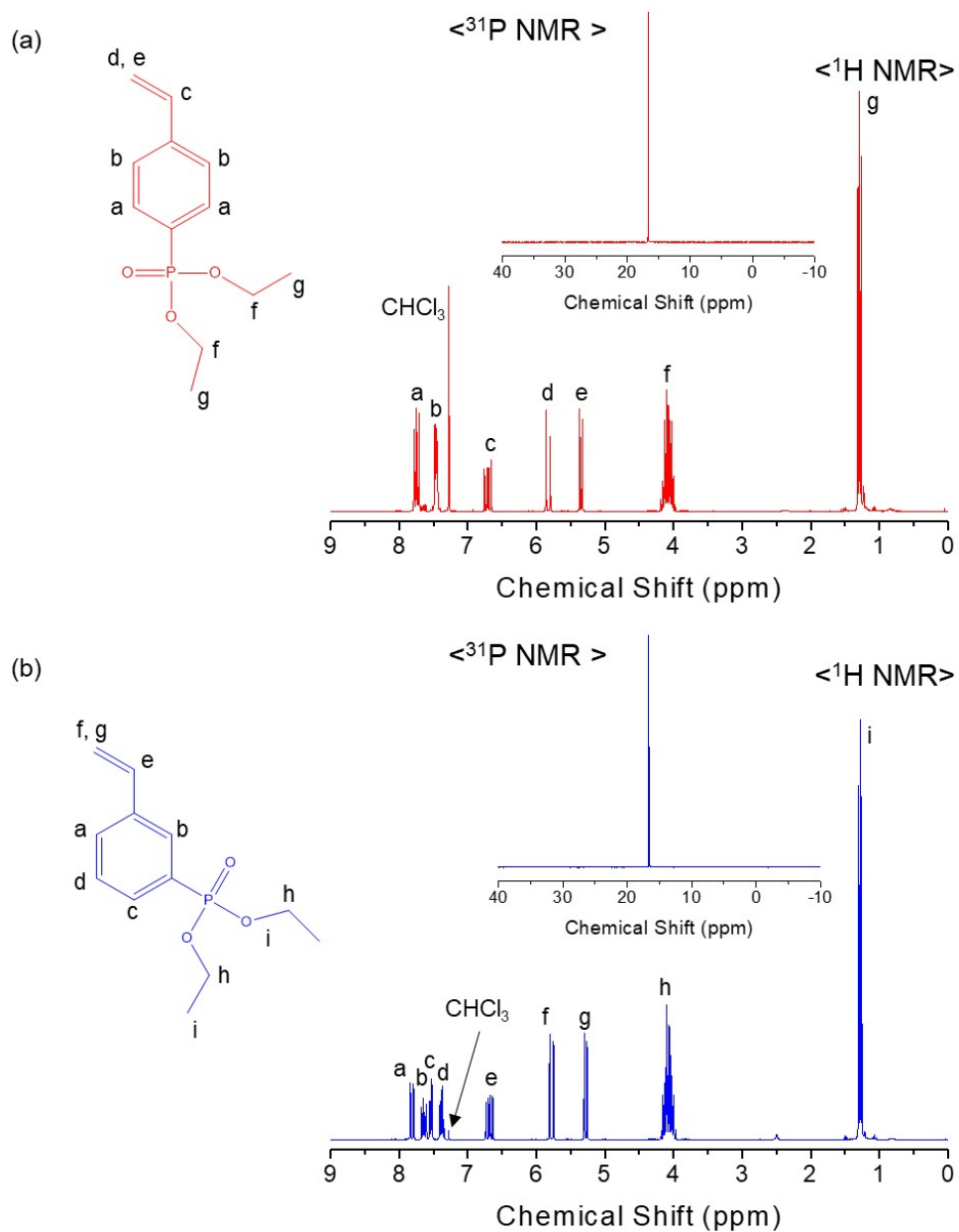


Fig. S1 ^1H and ^{31}P NMR spectra of (a) diethyl 4-vinylbenzylphosphonate and (b) diethyl 3-vinylbenzylphosphonate in CDCl_3 .

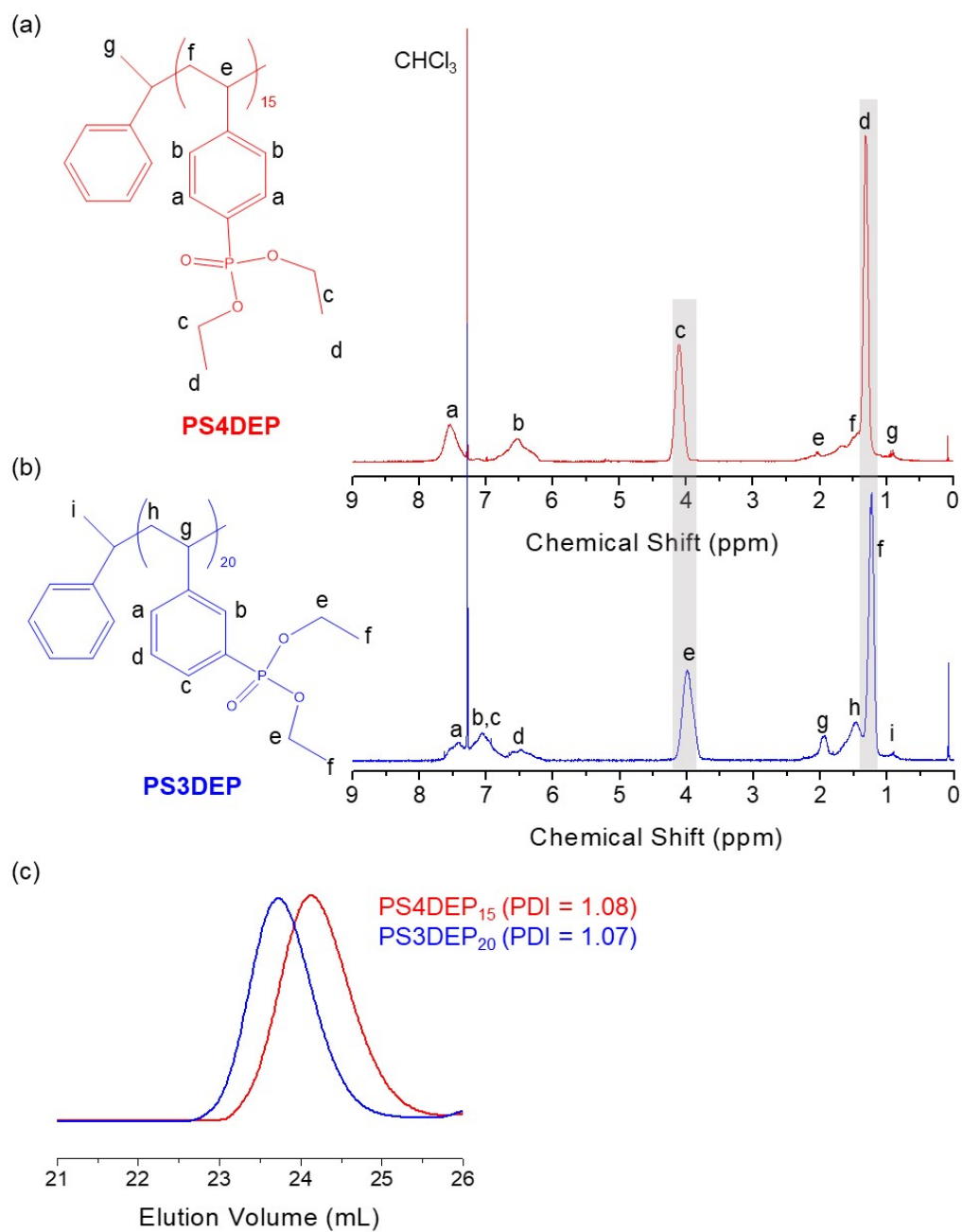


Fig. S2 ^1H NMR spectra of (a) PS4DEP and (b) PS3DEP in CDCl_3 . (c) SEC traces and PDI values of PS4DEP and PS3DEP.

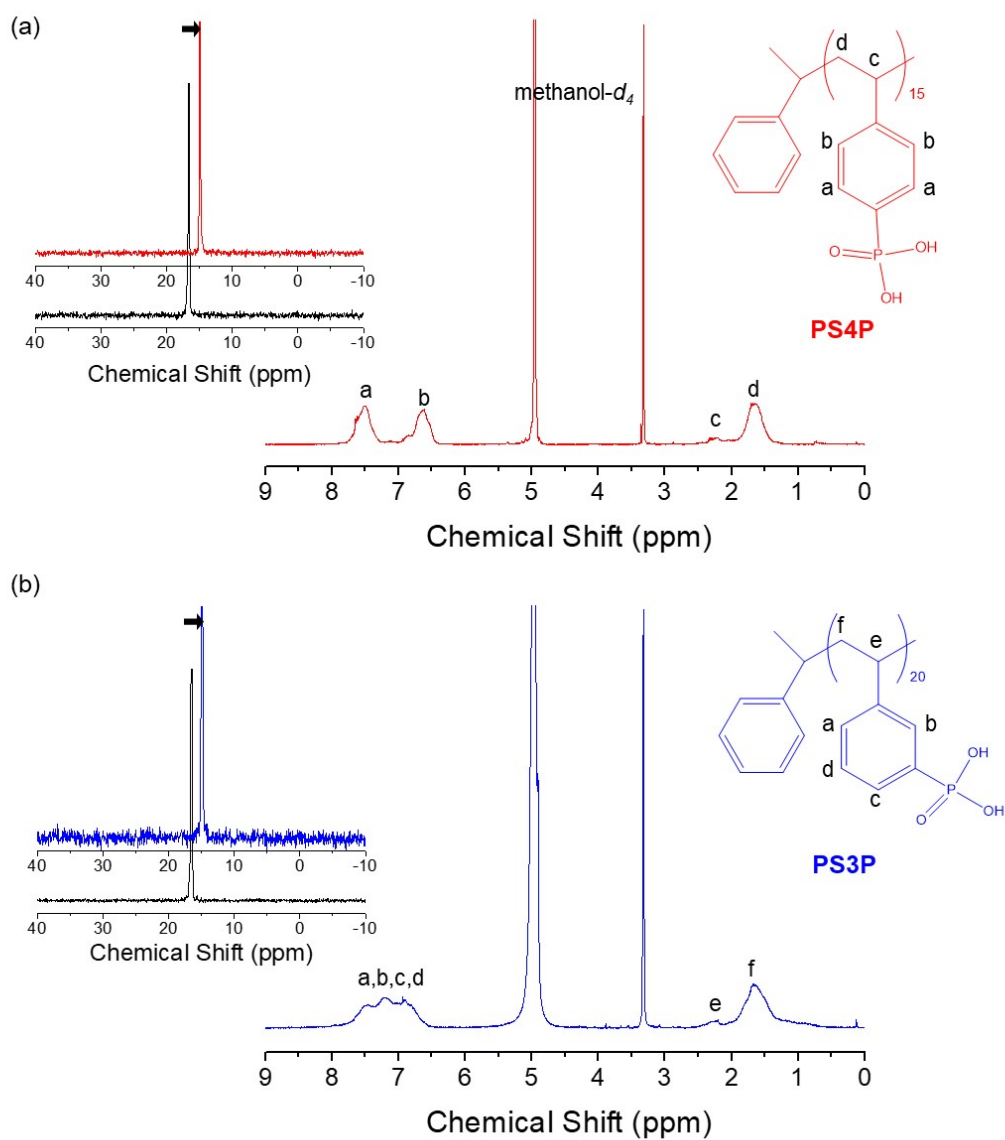


Fig. S3 ¹H and ³¹P NMR spectra of (a) PS4P and (b) PS3P in methanol-*d*₄. Compared with ³¹P NMR peaks of PS4DEP and PS3DEP in CDCl₃ (black line), those of PS4P and PS3P in methanol-*d*₄ shifted to upfield.

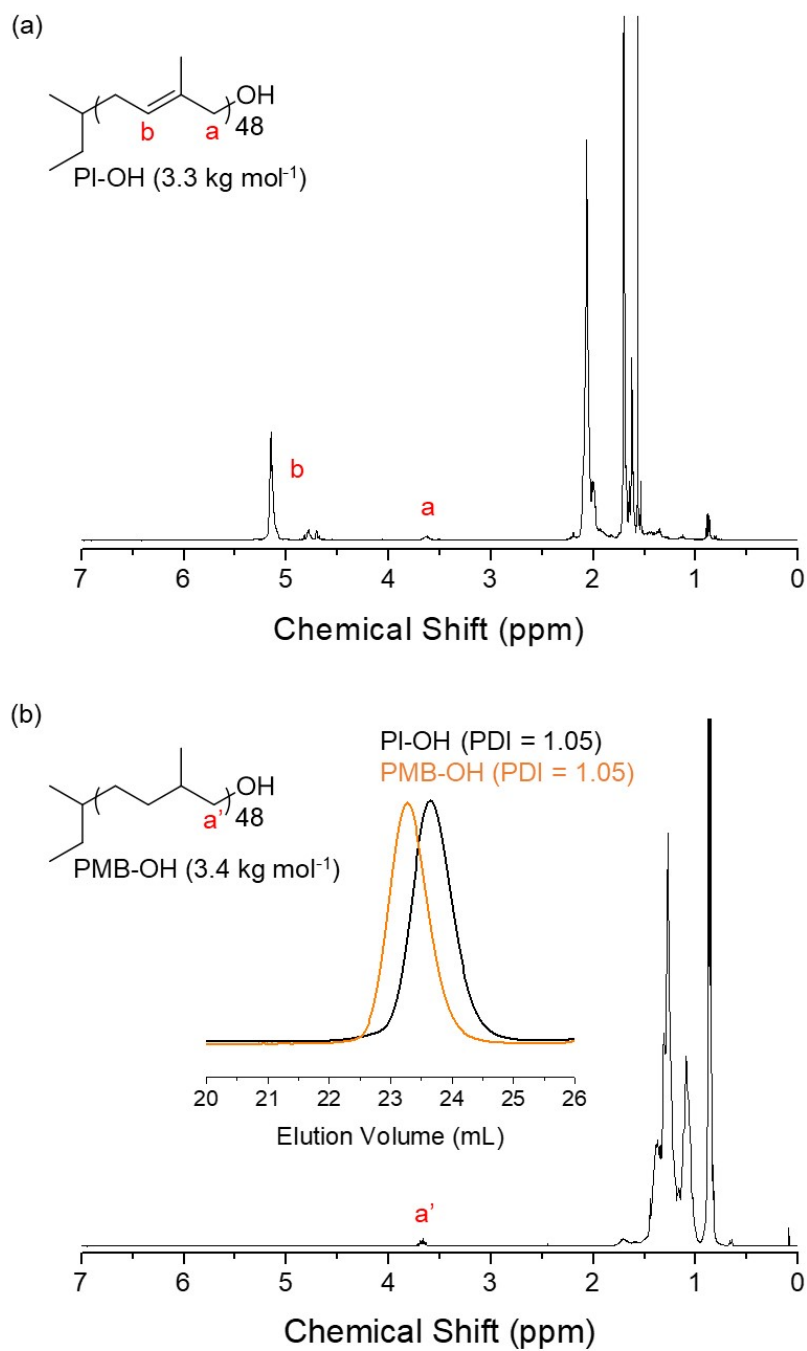


Fig. S4 ¹H NMR spectra of (a) PI-OH and (b) PMB-OH in CDCl₃. SEC traces of PI-OH and PMB-OH are shown in the inset of (b).

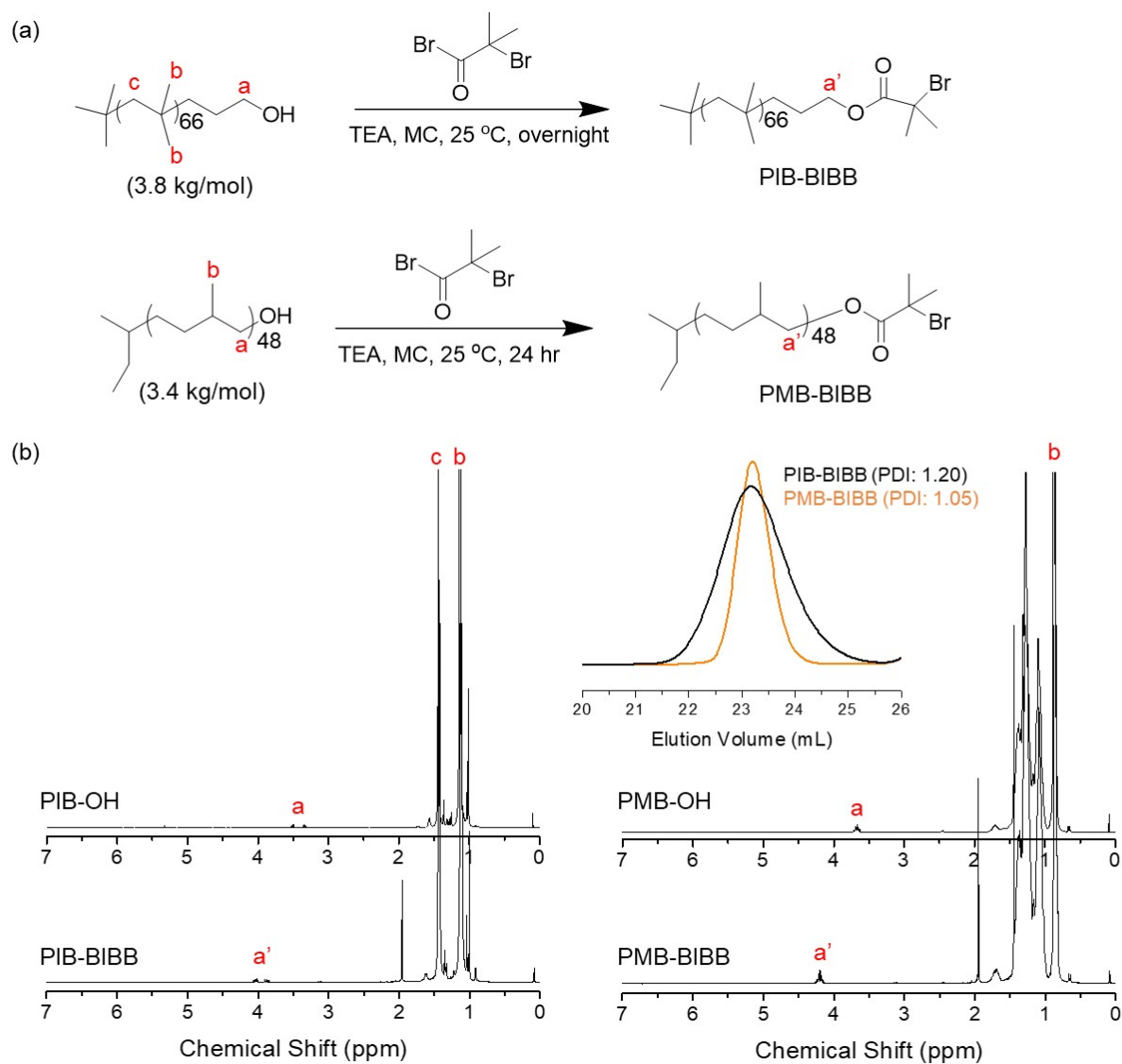


Fig. S5 (a) Synthetic routes and (b) ^1H NMR spectra of PIB-BIBB and PMB-BIBB macroinitiators. SEC traces and PDI values of PIB-BIBB and PMB-BIBB are shown in the inset of (b).

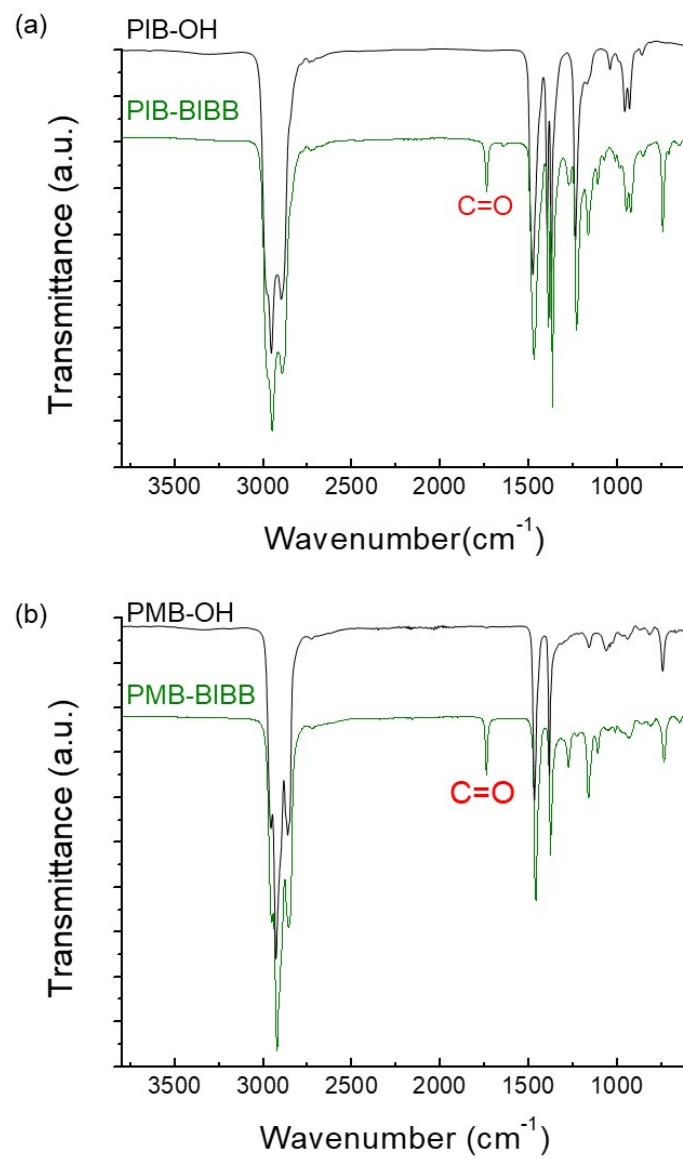


Fig. S6 FT-IR spectra of (a) PIB-OH/PIB-BIBB and (b) PMB-OH/PMB-BIBB.

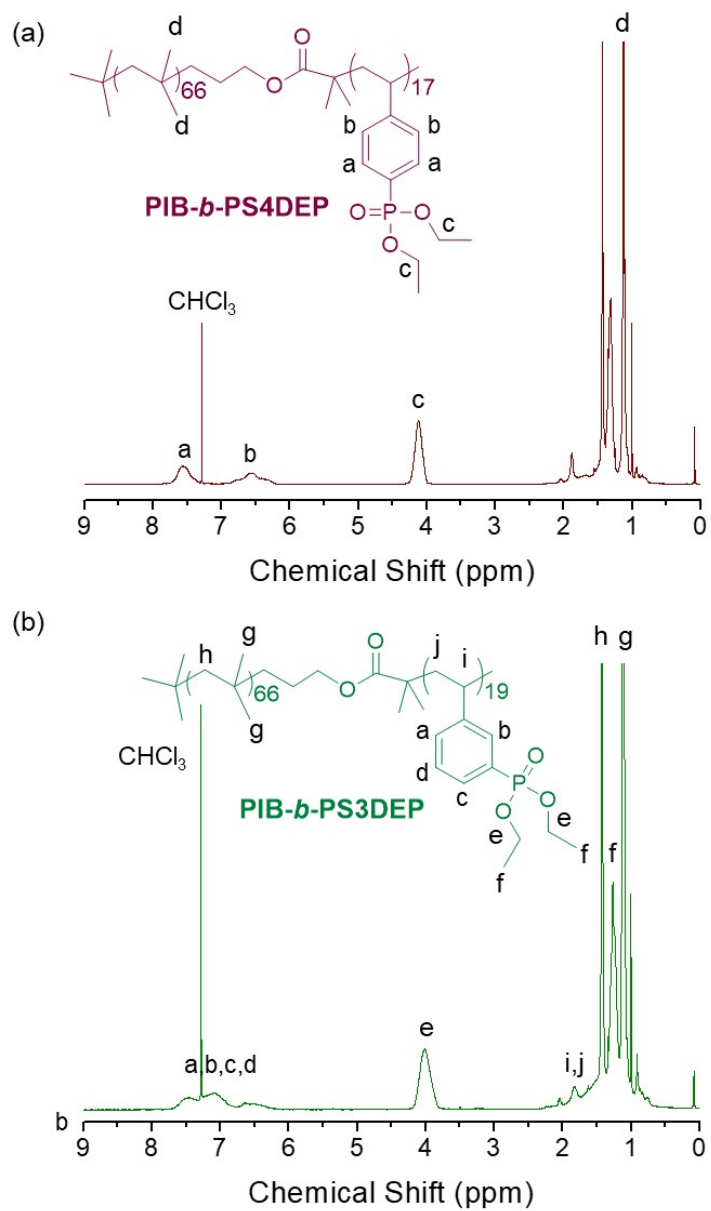


Fig. S7 Representative ¹H-NMR spectra of (a) PIB-*b*-PS4DEP and (b) PIB-*b*-PS3DEP in CDCl₃.

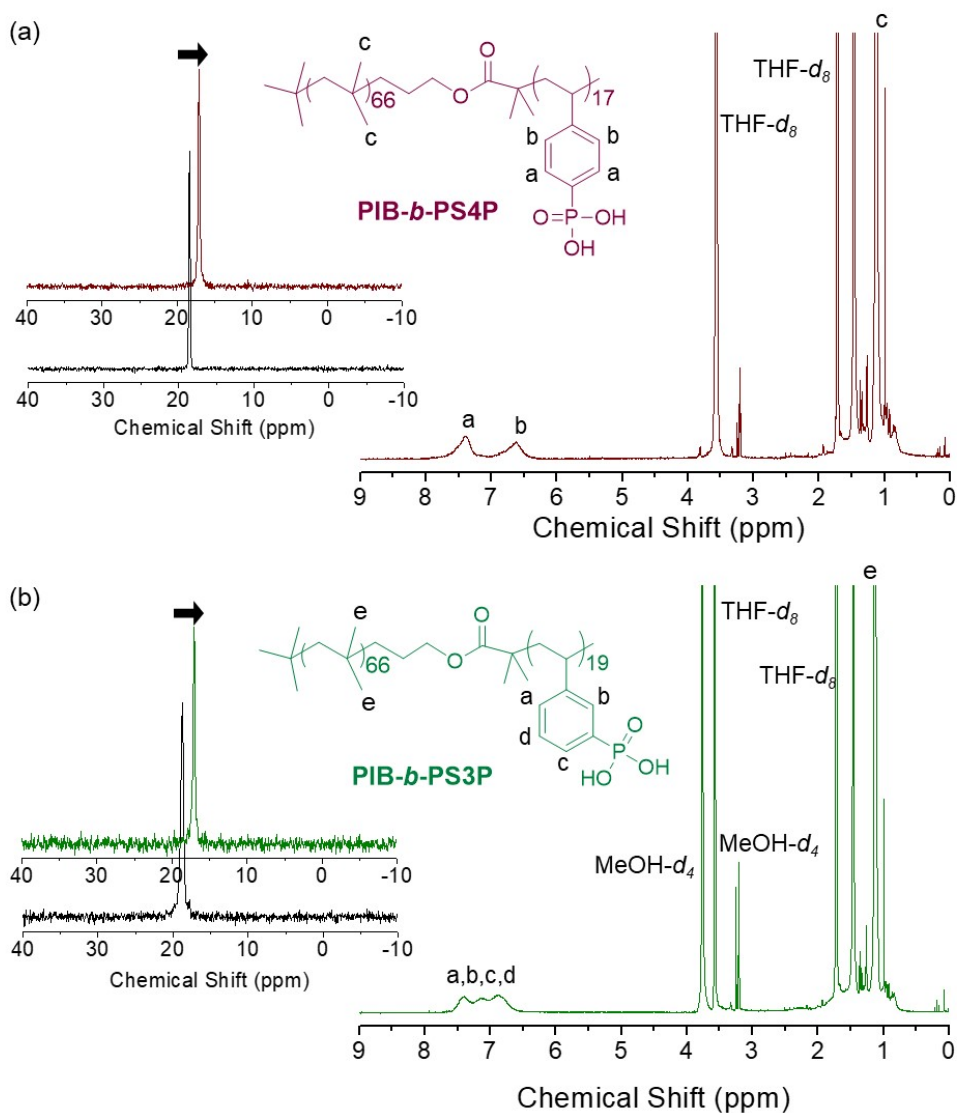


Fig. S8 ¹H-NMR and ³¹P-NMR spectra of (a) PIB-*b*-PSP and (b) PIB-*b*-PS3P in methanol-*d*₄/THF-*d*₈. Compared with ³¹P NMR peaks of PIB-*b*-PS4DEP (black) and PIB-*b*-PS3DEP (black), those of PIB-*b*-PS4P (brown) and PIB-*b*-PS3P (green) shifted to upfield.

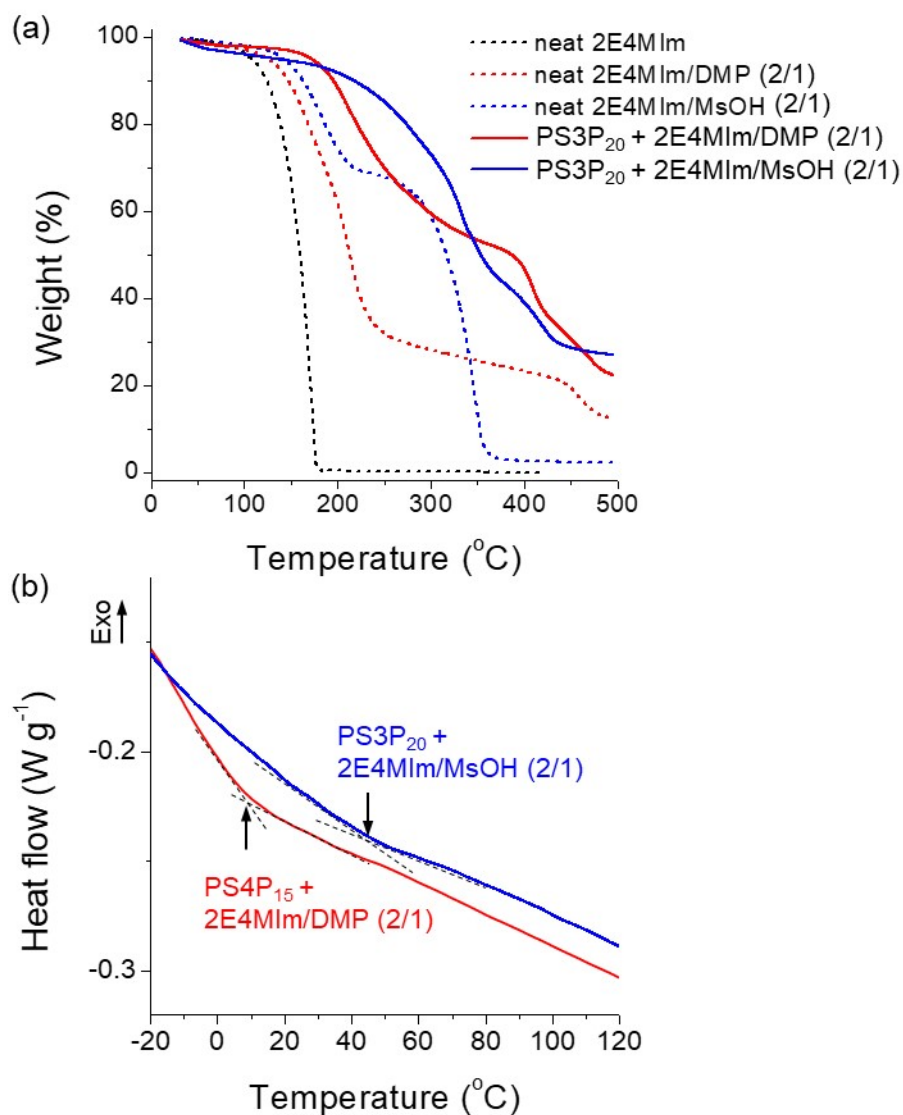


Fig. S9 (a) TGA thermograms of neat 2E4MIm, neat 2E4MIm/MsOH (2/1), neat 2E4MIm/DMP (2/1) (dotted lines), and those of PS3P ion gels comprising 2E4MIm/MsOH (2/1) and 2E4MIm/DMP (2/1) (solid lines). (b) Representative DSC thermograms of PS3P₂₀ doped with 2E4MIm/MsOH (2/1) and PS4P₁₅ doped with 2E4MIm/DMP (2/1). The heating rate was 10 °C/min.

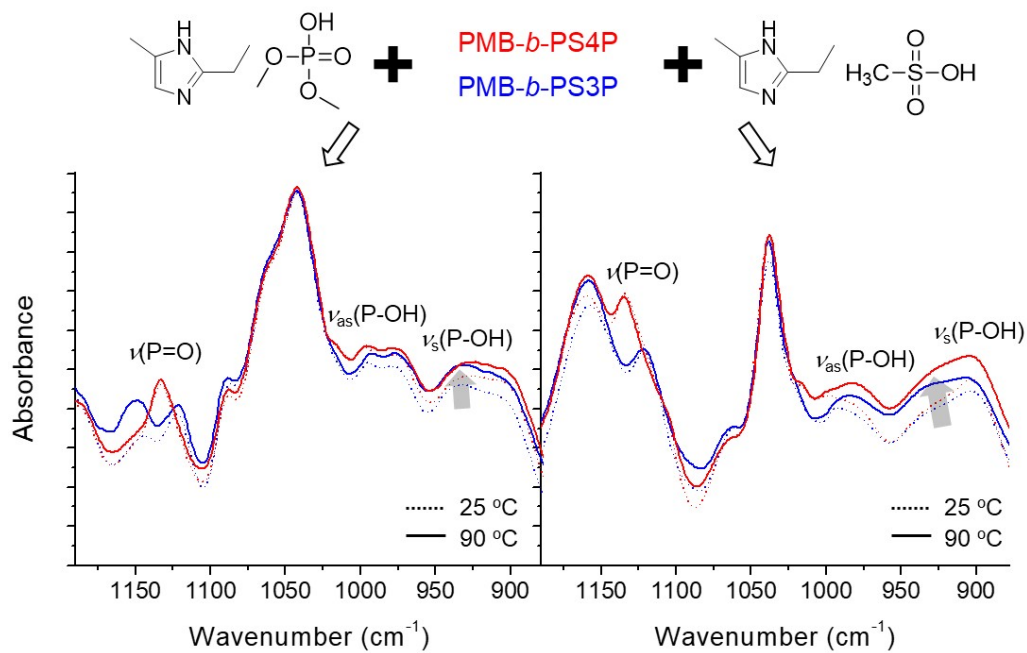


Fig. S10 FT-IR spectra of PMB-*b*-PS4P (red) and PMB-*b*-PS3P (blue) comprising different ionic liquids, measured at 25 °C (dotted lines) and at 90 °C (solid line) in the wavenumber range of 1190-850 cm⁻¹.

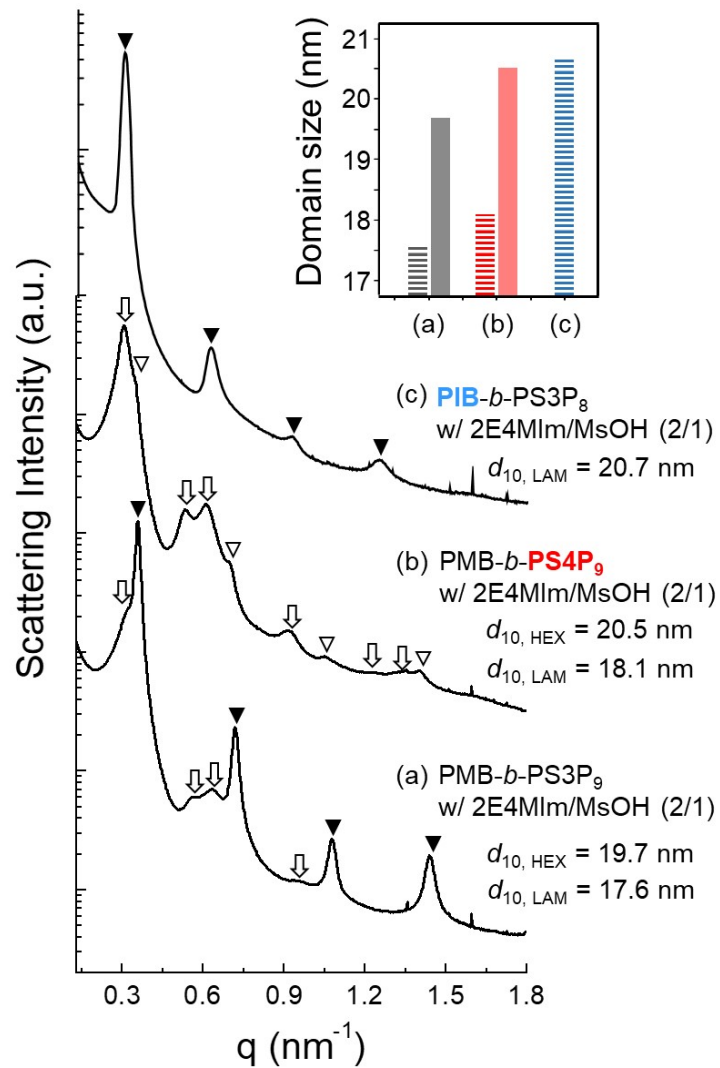


Fig. S11 SAXS profiles of (a) PMB-*b*-PS3P₉, (b) PMB-*b*-PS4P₉, and (c) PIB-*b*-PS3P₈ block copolymers comprising 2E4MIm/MsOH. The domain size of each ion gel is shown in the inset plot.

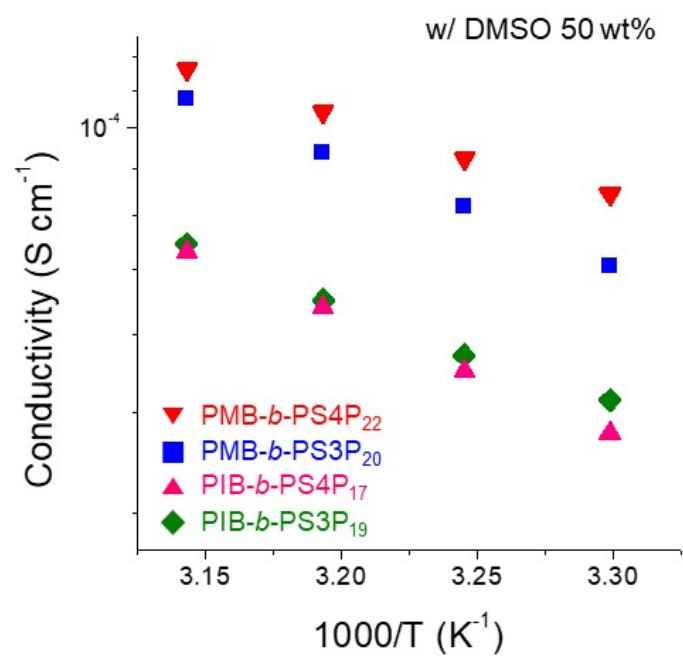


Fig. S12 Temperature-dependent ionic conductivities of four sets of block copolymers of PMB-*b*-PS4P₂₂, PMB-*b*-PS3P₂₀, PIB-*b*-PS4P₁₇, and PIB-*b*-PS3P₁₉ containing DMSO (50 wt%).