

Supporting Information for

**Poly(ethylene glycol) (PEG)-crosslinked poly(vinyl pyridine)-
PEG-poly(vinyl pyridine)-based triblock copolymers prepared by
RAFT polymerization as novel gel polymer electrolytes**

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Experimental.

Synthesis of the PVdF-HFP gel polymer electrolyte films: PVdF-HFP (0.75 g, 1.88×10^{-3} mmol) was dissolved in 10 mL acetone under stirring at 50 °C until a clear homogeneous solution was obtained. After this time, BMIM-TFSI (0.5 g, 1.19 mmol) and LiTFSI (0.264 g, 0.925 mmol) were added to this solution and again stirred at 50 °C for 1 h. The solution was casted directly onto the PP (polypropylene) plate and solvent was allowed to evaporate slowly at room temperature for two days. The film was finally dried under vacuum at 40 °C for 12 h, then heated at 60 °C for 2 h and 80 °C for 2 h to remove any trace of moisture present in the films.

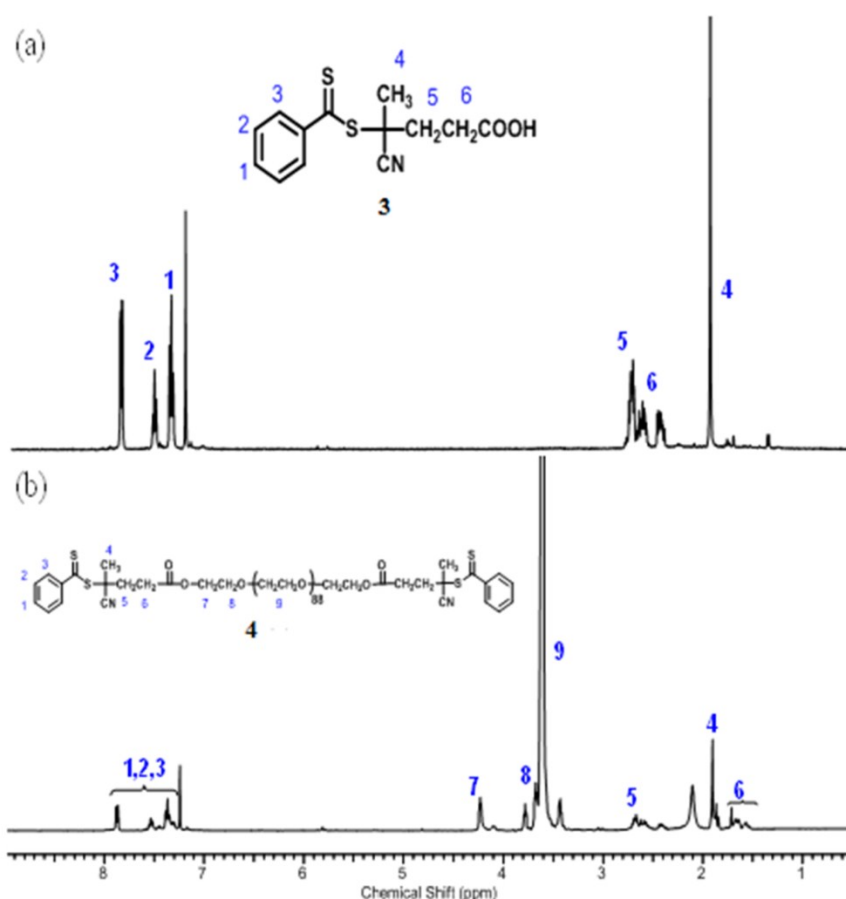


Figure S1. ¹H NMR spectra of the 4-cyano-4-((phenylcarbonothioyl)thio)pentanoic acid **3** (a) and the macro-CTA **4** (b) in CDCl₃.

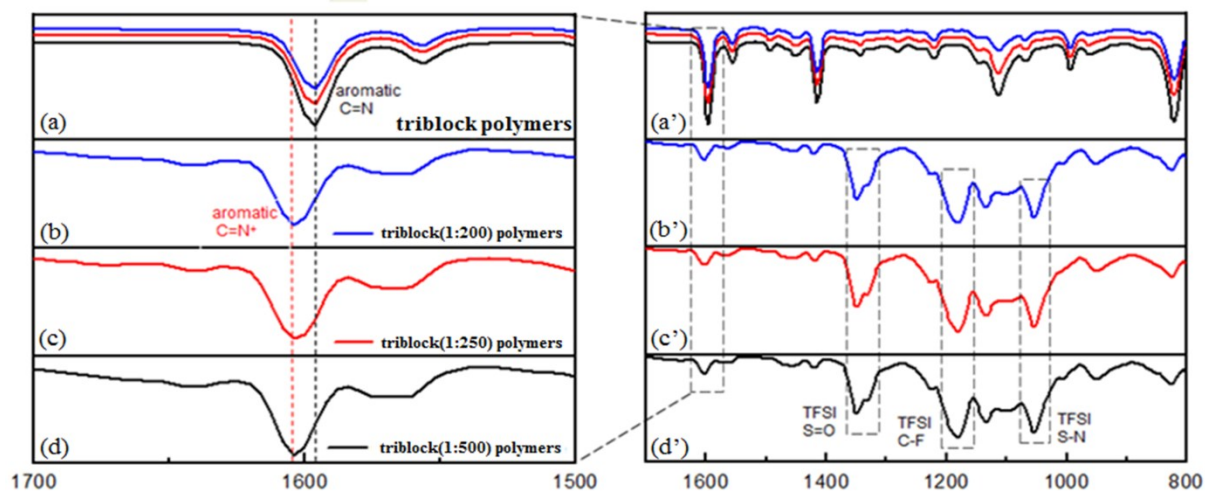


Figure S2. FT-IR spectra of the pristine triblock copolymers (a') and PEG-crosslinked PVP-PEG-PVP triblock copolymer gel polymer electrolytes with Triblock (1:200) (b'), Triblock (1:250) (c'), and Triblock (1:500) (d'), and their expanded spectra (a, b, c, and d)

Table S1. Stress and strain values obtained from the triblock copolymer gel electrolytes.

Membrane	Stress–Strain Curve	
	Stress (MPa)	Strain (%)
Triblock (1:500)	2.37	135
Triblock (1:250)	4.29	578
Triblock (1:200)	5.33	682
PVdF-HFP	2.13	137

Table S2. TGA and DSC data obtained from the PEG-crosslinked PVP-PEG-PVP triblock copolymer and the PVdF-HFP gel polymer electrolytes

Membrane	TGA		DSC
	On set (°C)	decomposition	T_g (°C)
Triblock (1:500)	293		-59.3
Triblock (1:250)	296		-60.1
Triblock (1:200)	304		-63.2
PVdF-HFP	380		-67.8