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Supporting information

Tuning electromechanical properties of dielectric elastomers via dynamic ionic crosslinks

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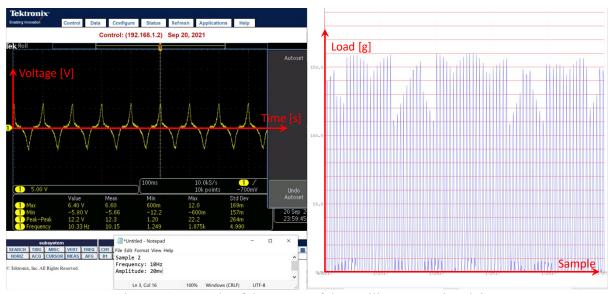


Figure S1 Example of the output of the oscilloscope and Arduino

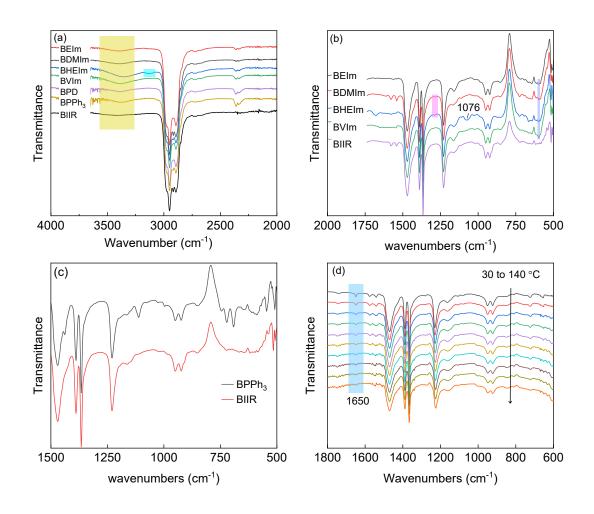


Figure S2 (a) FTIR spectra (measured at 30 °C) of BIIR and BEIm, BDMIm, BHEIm, BVIm, BPD and BPPh₃ gels after dissolving in toluene for 96 h and drying in 50 °C vacuum for 96h at 4000 ~2000 cm⁻¹, (b) FTIR spectra (measured at 30 °C) of BIIR and BEIm, BDMIm, BHEIm and BVIm gels after

dissolving in toluene for 96h and drying in 50 °C vacuum for 96h at $2000 \sim 500 \text{ cm}^{-1}$, (c) FTIR spectra (measured at 30 °C) of BIIR and BPPh₃ gel after dissolving in toluene for 96h and drying in 50 °C vacuum for 96 h at $1500 \sim 500 \text{ cm}^{-1}$, (d) the temperature dependent FTIR spectra of BVIm mixture at $1800 \sim 600 \text{ cm}^{-1}$ from 30 to 140 °C

As shown in Figure S2 (a) to (c), the alkylation of nucleophiles on the molecule of BIIR can be confirmed by following peaks:

- (1) BEIm: the peaks at 3580 to 3250 cm⁻¹ (assigned to the N-H vibration), the peak at 662 cm⁻¹ (assigned to out-of-plane deformation of the imidazole ring);
- (2) BDMIm: the peaks at 3580 to 3250 cm⁻¹ and 662 cm⁻¹, and the weak peak at 1286 cm⁻¹ (assigned to the combination of C-H stretching and out-of-plane bending of C-N on the imidazole ring).
- (3) BHEIm: the peaks at 3580 to 3250 cm⁻¹, 1286 cm⁻¹ and 662 cm⁻¹, and the peak 3132 cm⁻¹ (attributed to C-H vibration of the imidazole ring).
- (4) BVIm: the peak at 3580 to 3250 cm⁻¹ and 662 cm⁻¹
- (5) BPD: the peak at 3580 to 3250 cm⁻¹
- (6) BPPh₃: the peak at 3580 to 3250 cm⁻1, the peaks at 1439, 1111 and 692cm⁻1 (assigned to C-C out-of-plane vibration on the benzene ring), and the peak at 746 cm⁻1 (attributed to out-of-plane C-H vibration on the benzene ring).

As shown in Figure S3 (d), the peak at 1650 cm⁻¹ of BVIm is assigned to C=C vibration of BIIR molecules, which becomes weak due to the free radical polymerization triggered during the heat process, indicating the decrease of C=C caused by covalent crosslinking.

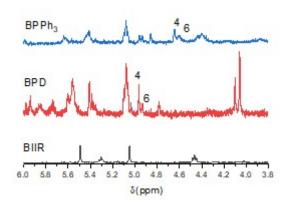


Figure S3. The olefin region of ¹HNMR spectra of pure BIIR, BPD and BPPh₃.

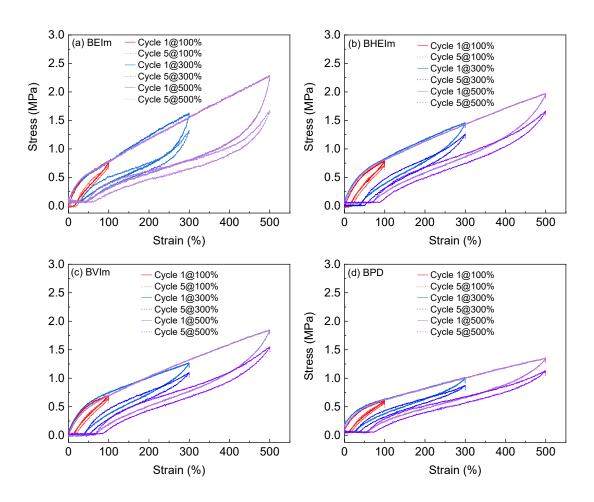


Figure S4 Cyclic tensile curve of (a) BEIm, (b) BHEIm, (c) BVIm, and (d) BPD with the maximum strain at 100%, 300% and 500%

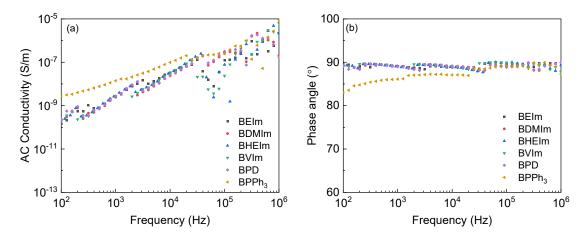


Figure S5 (a) Conductivity and (b) phase angle vs frequency curves of BIIR ionomers.