SUPPLEMENTARY INFORMATION

1. Synthesis of ionic liquid

a) Synthesis of ionic liquid precursor 1-ethyl-3-methylimidazolium bromide(EMIM-Br)

The 1-methylimidazole (1.62 mL) was first added into ethyl acetate (2.73 mL) (ratio 2:3) and stirred at 45°C for one hour. Bromoethane(1.56mL) was added dropwise into the stirred solution and refluxed for another 5 hours. High purity of [EMIm] Br was obtained using extraction (water: ethyl acetate=1:1). The obtained EMIm Br was concentrated in a rotary evaporator at 60°C for one hour. The yellow viscous liquid was dried in a vacuum oven at 60^0 C overnight. (All the reaction was carried out in a nitrogen atmosphere).

b) Synthesis of ionic liquid 1-ethyl-3-methyl imidazolium bis(trifluoromethyl sulfonyl imide) (EMIM-TFSI)

EMIM-TFSI was prepared by the anion exchange between EMIM-Br(ionic liquid precursor) and LiTFSI.(Lithium bis(trifluoromethylsulfonyl imide). At first, 0.01 mol of 1-ethyl-3-methyl imidazolium bromide dissolved in 0.02 mol of acetonitrile and added dropwise 0.01 mol of LiTFSI dissolved in acetonitrile and stirred for 24 hours at 25°C in the presence of a nitrogen atmosphere. After the completion of the anion exchange reaction, the viscous liquid was washed with hexane several times to remove LiBr. The resultant ionic liquid was concentrated in a vacuum rotary evaporator (1.5 h at 65°C), followed by drying in a vacuum oven at 60°C for another 48 hours. According to the calculations, for EMIM-TFSI the theoretical yield based on 0.01 mol of EMIM-Br was found to be 3.91 g , and the resultant product mass 3g, giving the experimental yield of ~76%.

c) Synthesis of ionic liquid 1-ethyl-3-methyl imidazolium (EMIM-Ac)

EMIM-Ac was prepared by the anion exchange between EMIM-Br(ionic liquid precursor) and KAc (Potassium acetate). At first, 0.01 mol of 1-ethyl-3-methyl imidazolium bromide and 0.01 mol of potassium acetate added in 10 mL of methanol and stirred for 24 hours at 25°C in the presence of a nitrogen atmosphere. After the completion of the anion exchange reaction, KBr sediment was decanted off from EMIM-Ac. The resultant ionic liquid was concentrated in a vacuum rotary evaporator (1.5 h at 45°C), followed by drying in a vacuum oven at 60 °C for another 48 hours. For EMIM-*Ac* the theoretical yield based on 0.01 mol of EMIM-Br was found to be 1.7g and the resultant product was 1.43 g , giving the experimental yield of ~84%.

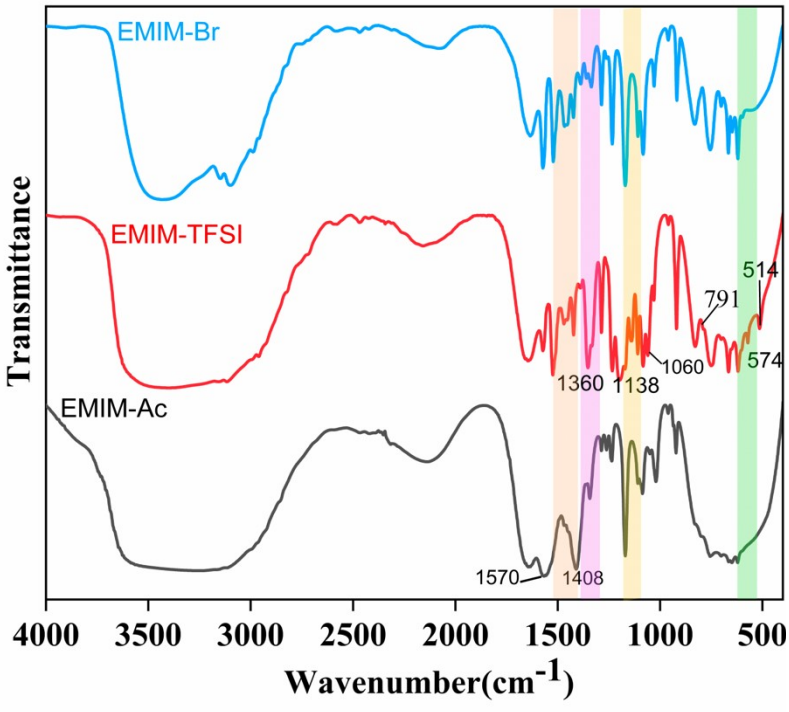

Figure- S1 FTIR Graph of the prepared three ionic liquids. a) EMIM-Br b) EMIM-TFSI C) EMIIM-Ac

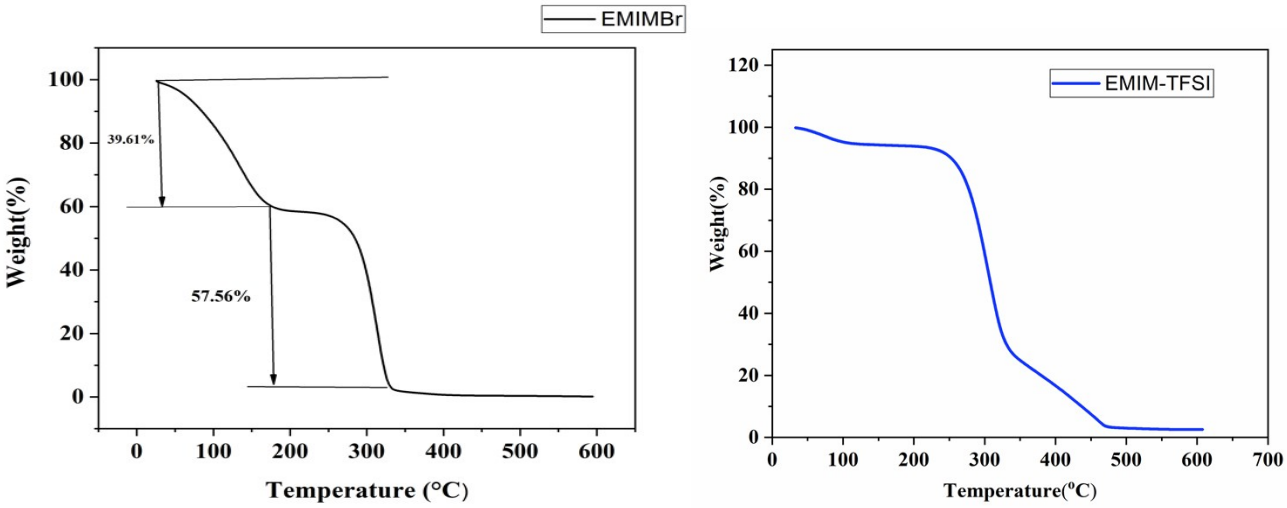

Figure-S2 TGA Graph of the prepared ionic liquids a) EMIM-Br, b) EMIM-TFSI

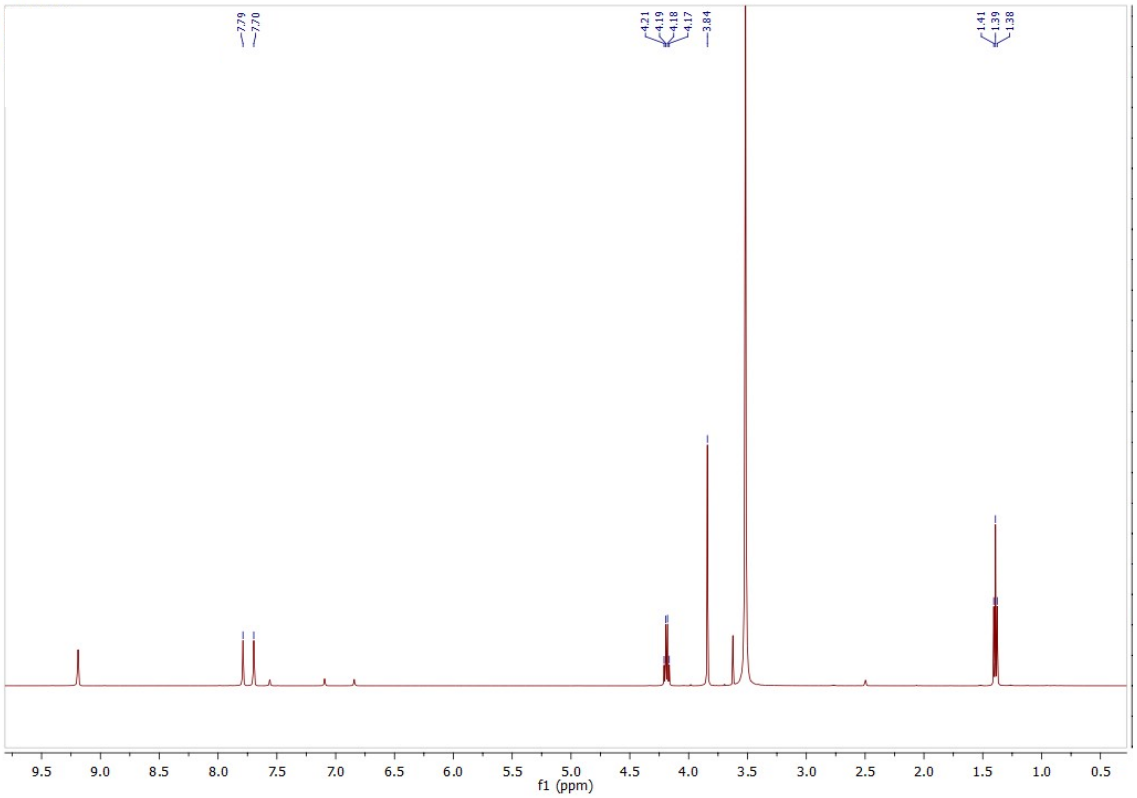

Figure- S3 a) NMR spectra of ionic liquid 1-ethyl-3-methyl imidazolium bis(trifluoro)methane sulfonyl imide (EMIM-TFSI).

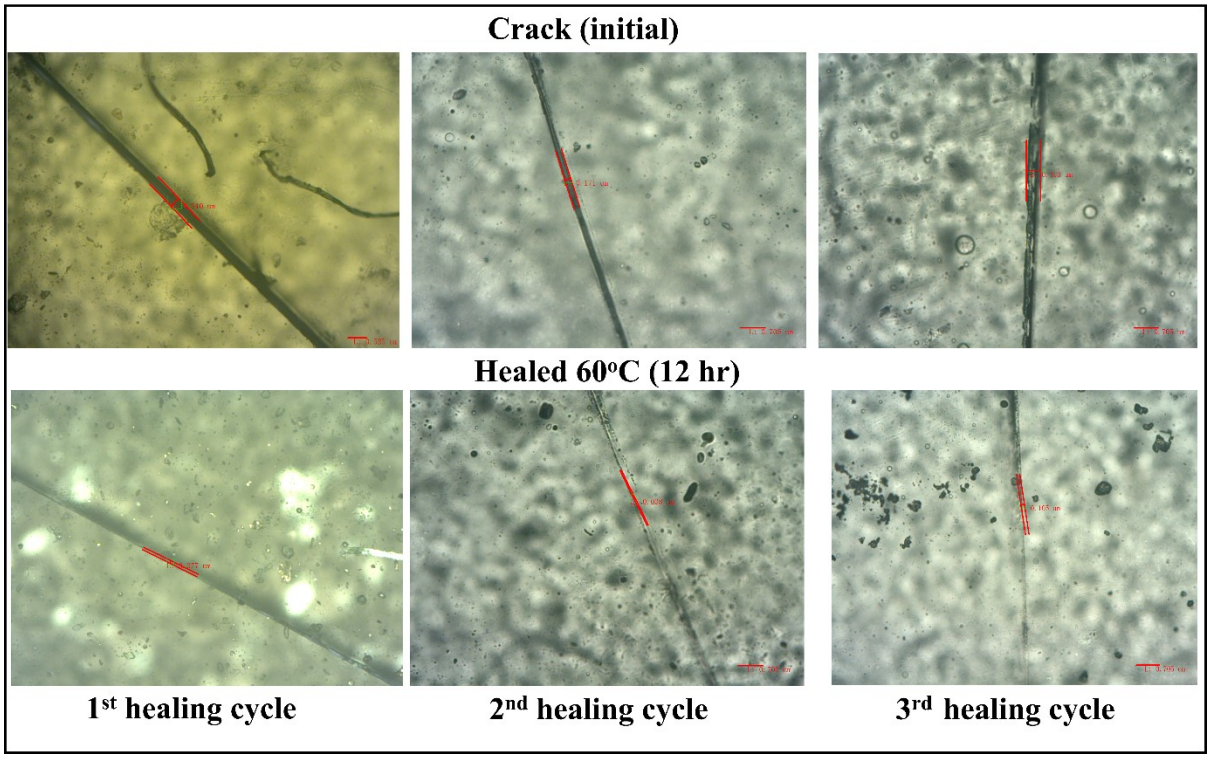

Figure-S4 Optical microscopy images showing multiple self-healing cycles of the polymer electrolyte after repeated cutting and healing.

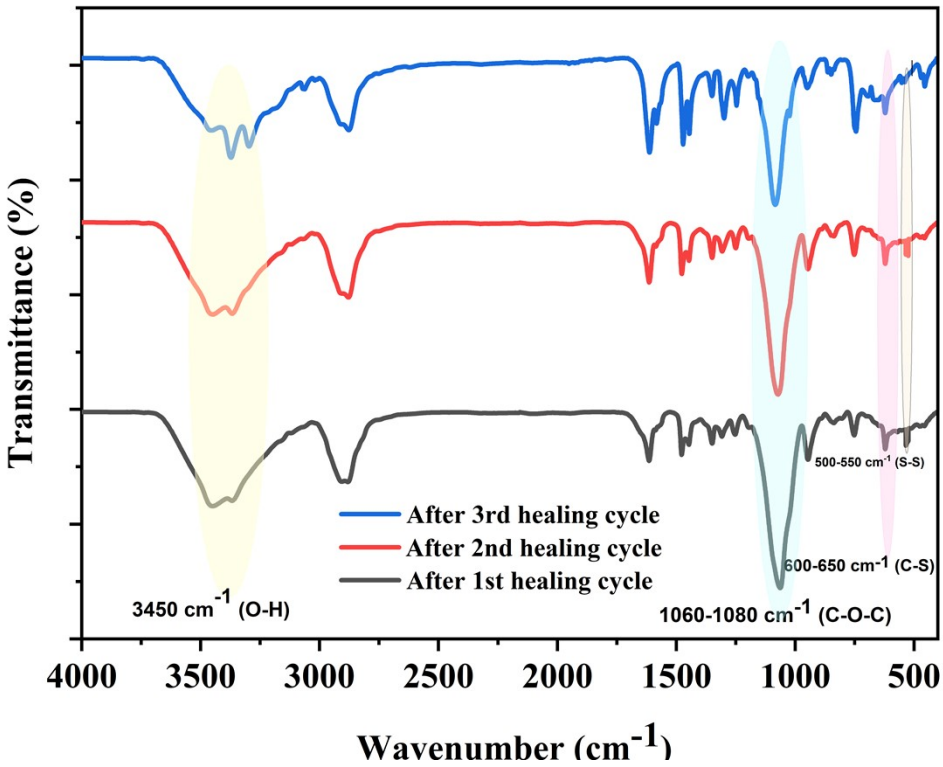

Figure -S5 FTIR spectroscopy of PMALT-0.5 after multiple healing cycles

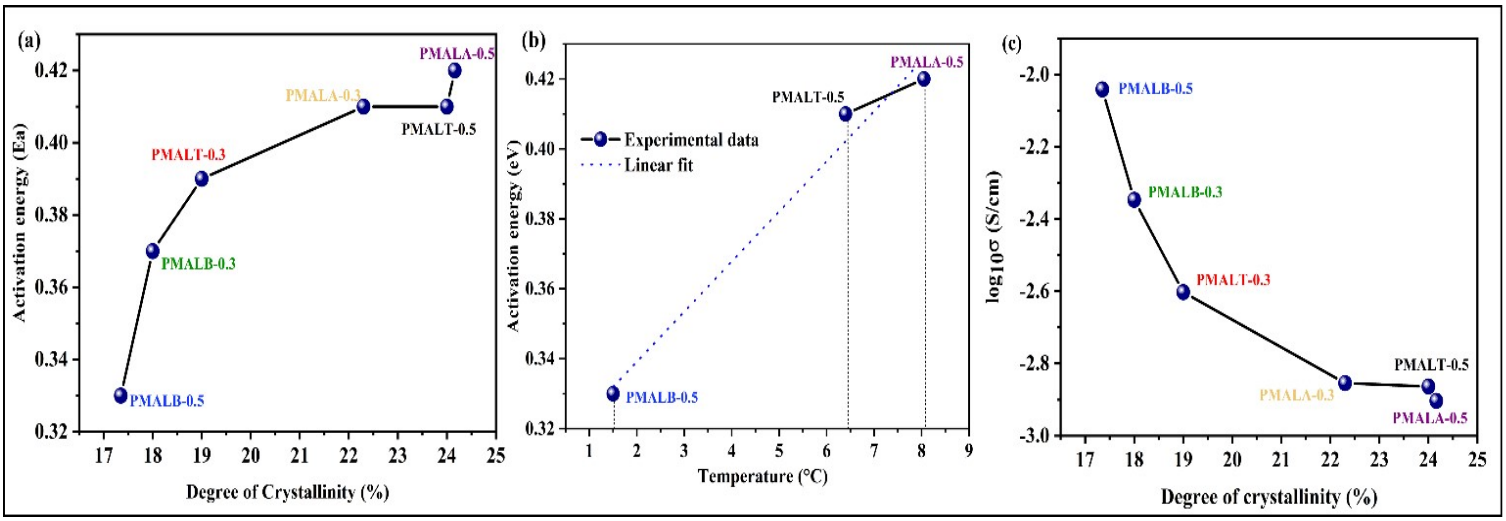

Figure- S6 Relationship between (a)Activation energy (Ea) and Degree of crystallinity (b) Activation energy (Ea) and Temperature (Glass transition temperature)(Tg) (c) logarithm of conductivity (log10 σ) and degree of crystallinity.

Table-S1 Values of crystallinity, activation energy, glass transition temperature and ionic conductivity for all the prepared samples.

PROPERTY	PMALA-0.5	PMALT-0.5	PMALA-0.3	PMALT-0.3	PMALB-0.3	PMALB-0.5
Degree of crystallinity	24.16%	24%	22.3%	19%	18%	17.35%
Activation energy (eV)	0.42	0.41	0.41	0.39	0.37	0.33
Glass-transition temperature	8.05°C	6.4°C	-	-	-	1.5°C
Ionic-conductivity (S/cm)	$1.25 * 10^{-3}$	$1.37 * 10^{-3}$	$1.4 * 10^{-3}$	$2.5 * 10^{-3}$	$4.5 * 10^{-3}$	$9.13 * 10^{-3}$

Equation used for calculating the crystallinity value-

the values of crystallinity for all the samples has been calculated using **the equation ES-**1 below:

$$Crystallinity(\%) = \frac{Area\ of\ peak}{Area\ of\ the\ whole\ curve} * 100 \text{.....(1)}$$

The values calculated follows the order PMALA 0.5 (24.26%) > PMALT 0.5 (24%) > PMALA 0.3 (22.3) > PMALT 0.3 (19%) > PMALB 0.3 (18%) > PMALB 0.5 (17.35).