Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Structural, Energetic and Dynamic Investigation of Poly(ethylene oxide) in Imidazolium-Based Ionic Liquids with Different Cationic Structures

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SUPPORTING

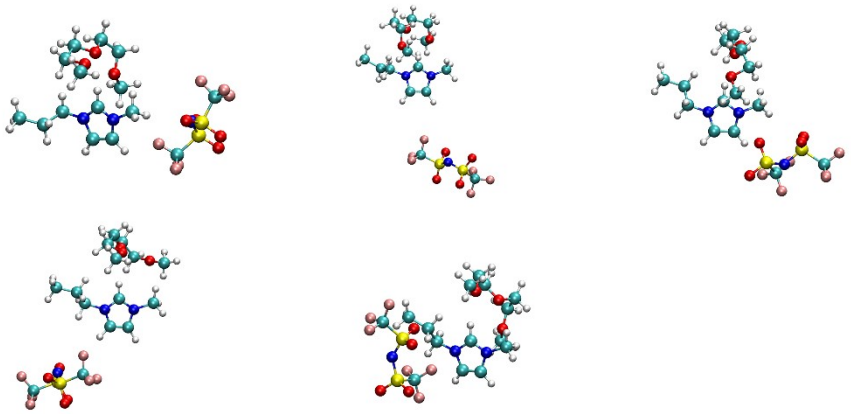
Table S1. Composition of each PEO/IL system for simulated

System	Chains' Number	Cations' Number	Anions' Number
PD_{10}	38	13	26
PD_{20}	36	27	54
PD_{30}	35	45	90
PD_{40}	28	55	110
PD_{50}	25	76	152
PD_{60}	21	96	192
PD_{70}	17	120	240
PD_{80}	12	141	282
PD_{90}	7	185	370
PM_{30}	35	91	91
PM_{50}	25	152	152
PM_{70}	17	241	241

Table S2. Comparison between simulated and experimental densities (ρ) for MIL and DIL at room temperature

configuration	ρ_{Simu}(g/cm^3)	ρ_{Exp}(g/cm^3)[1]	Dev.
[C_3mim] [TFSI]	1.543	1.476	4.54%
[C_6(mim)$_2$][TFSI]$_2$	1.607	1.546	3.95%

(a) Five initial configurations of PEO_3/MIL from MD trajectories:



(b) Five initial configurations of PEO_3/DIL from MD trajectories:

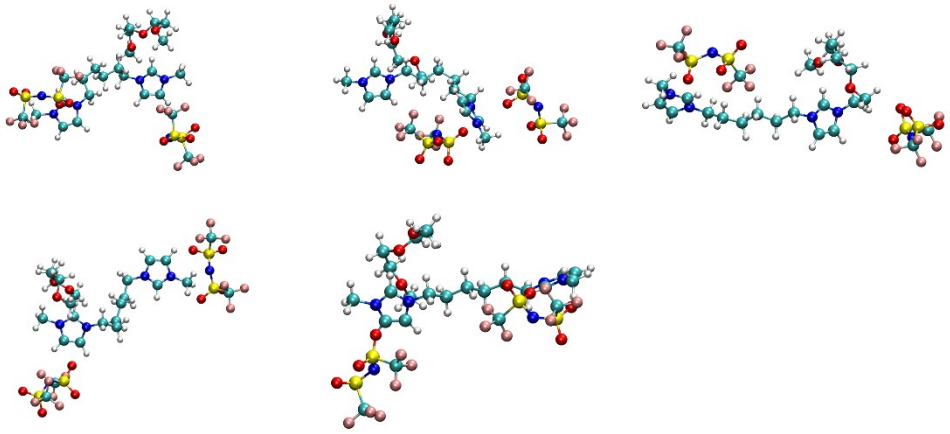


Figure S1. The initial configurations of each PEO/IL pairs derived from our MD simulation trajectories

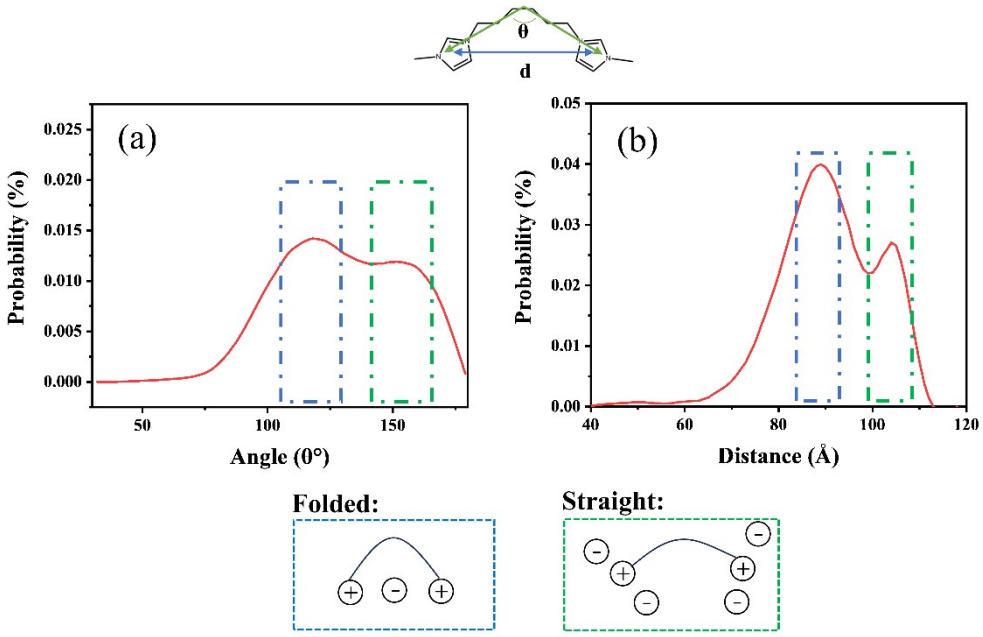


Figure S2. (a) Distribution of the angle formed by two vectors pointing from the center atom of linkage chain to the imidazolium rings and (b) the distance between two imidazolium rings of DIL in PD_{50} at 373K and 1 atm.

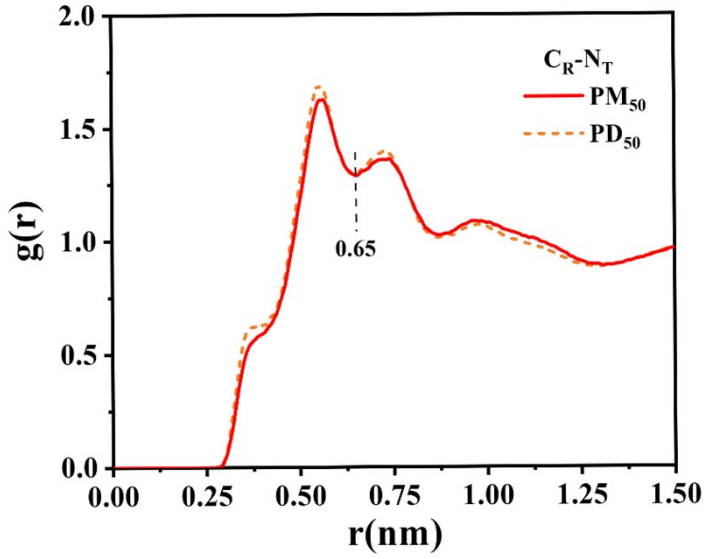


Figure S3. Radial distribution functions for C_R and N_T atoms of different ions in PM_{50} and PD_{50} systems at 373K and 1 atm .

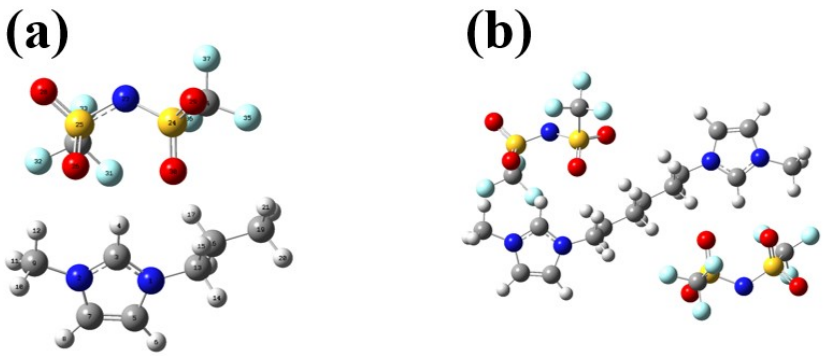


Figure S4. Structures of (a) MIL and (b) DIL optimized at B3LYP/(6-31G* + 6-31+G*)

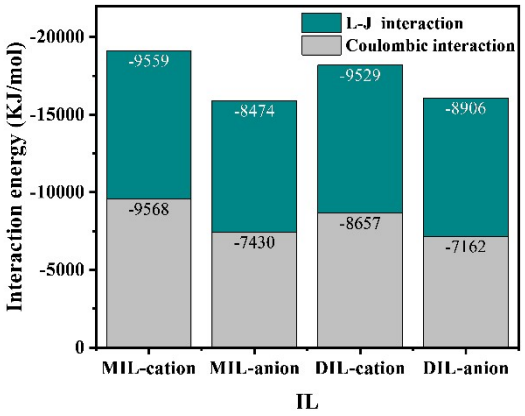


Figure S5. Interaction energies of different ions with PEO in PM_{50} or PD_{50} at 373K and 1 atm

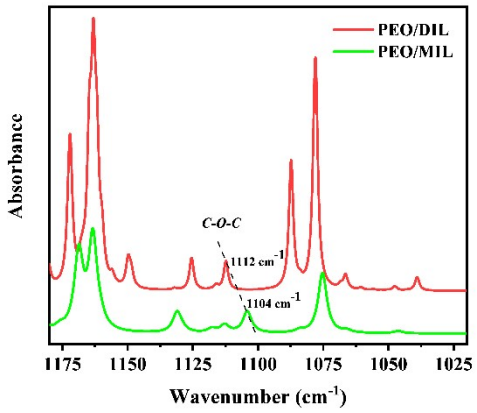


Figure S6. The vibrational spectra of PEO/MIL and PEO/DIL from DFT calculation

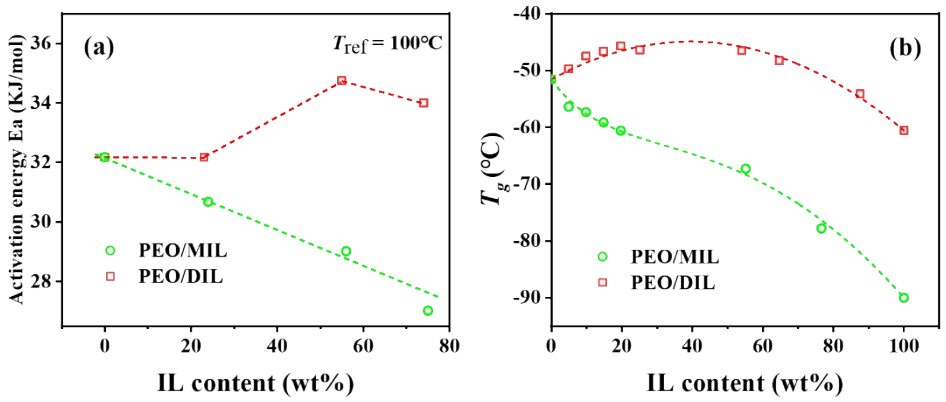
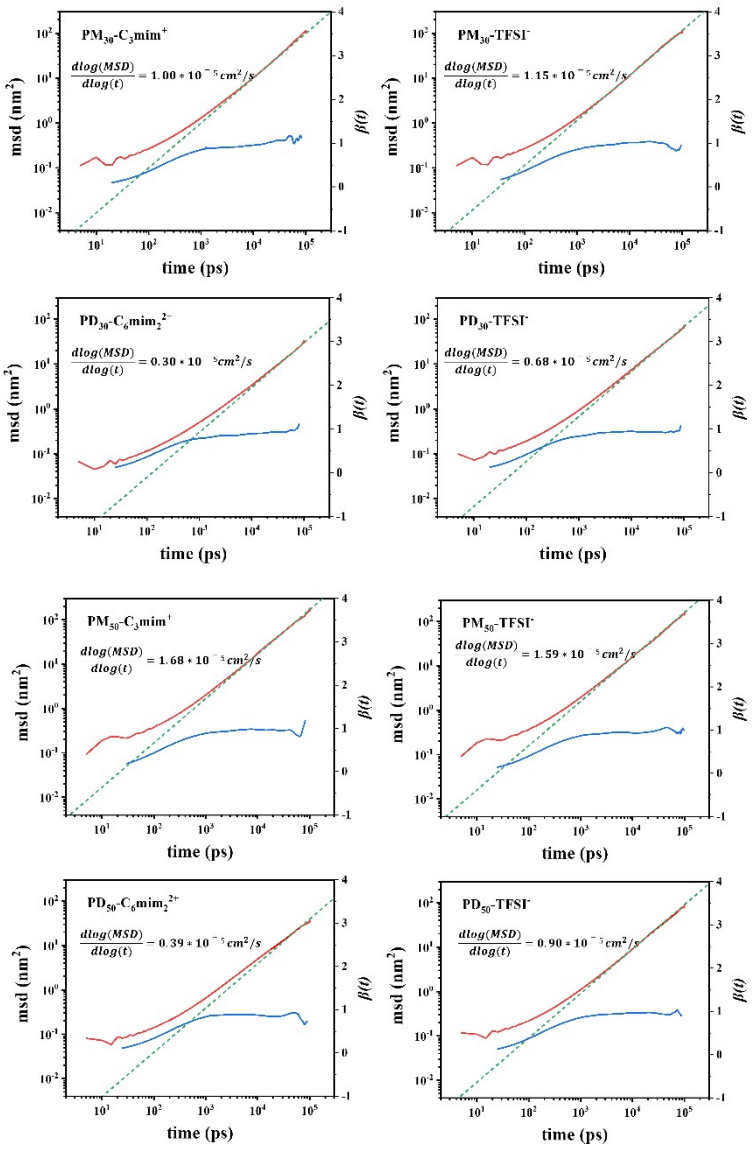


Figure S7. The activation energy E_a value and (b) glass transition temperature of PEO/ILs mixtures at different ILs content[2].



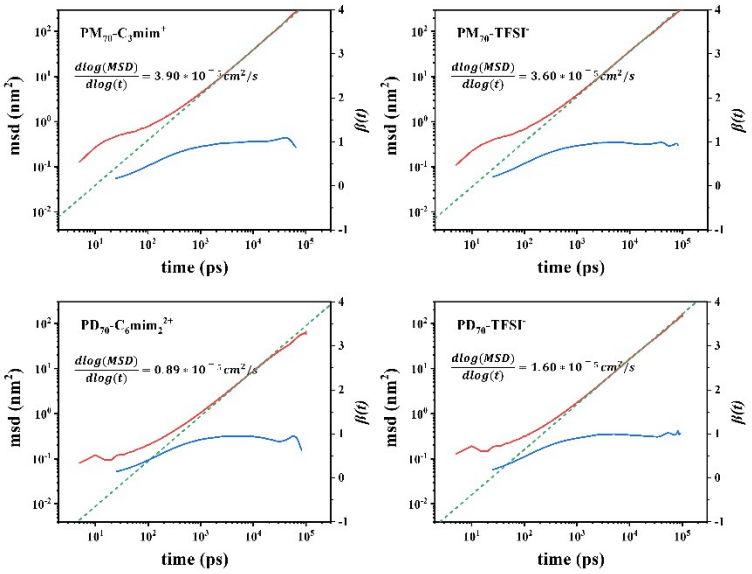


Figure S8. Mean square displacements (MSDs) of cations and anions calculated by 100 ns MD trajectories for PM_{50} and PD_{50} at 373 K and 1 atm. $\beta(t) = \frac{dlog(MSD)}{dlog(t)}$ changes (blue line) versus time were plotted to find the linear region of the MSD curve (red line) with a slope of 1.

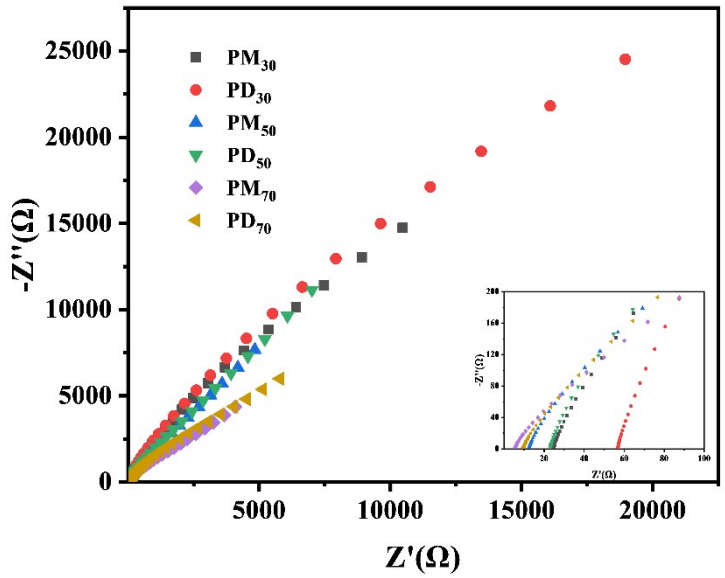


Figure S9. Nyquist plots of PEO/ILs systems with different MIL or DIL concentrations at 373 K

References

1. H. Shirota, T. Mandai, H. Fukazawa and T. Kato, *J. Chem. Eng. Data*, 2011, **56**, 2453-2459.

2. H. Luo, X. He, W. Li, Y. Niu and G. Li, *Macromolecules*, 2022, **55**, 4589-4599.