Development of Mixed Organic Ionic Plastic Crystal Electrolytes for Energy Storage

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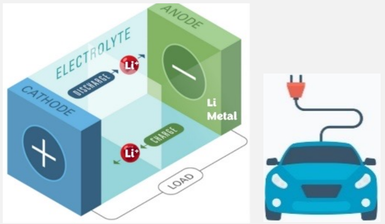
Research Aim

Understanding the effect of combining OIPCs with two different cations has on the physicochemical and electrochemical properties and their potential use as solid-state electrolyte materials.

Introduction



OIPCs (Organic Ionic Plastic Crystals)

Consist completely of cations and anions with a long range ordered crystalline lattice with short range disorder that occurs through rotational motions of molecules.[1,2]

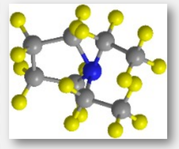
- Beneficial Features:

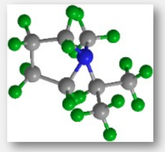
Good ionic conductivity — Negligible flammability

Negligible volatility — Thermally stable

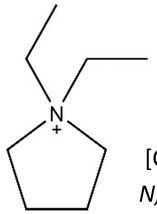
Goal:

Increase defects/ vacancies to tune physicochemical properties.



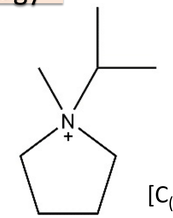


Synthetic Methodology



$[C_2epyr][FSI]$

N,N-diethylpyrrolidinium bis(fluorosulfonyl)imide

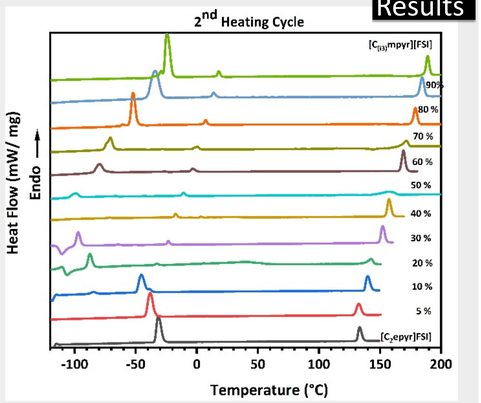


$[C_{(i3)}mpyr][FSI]$

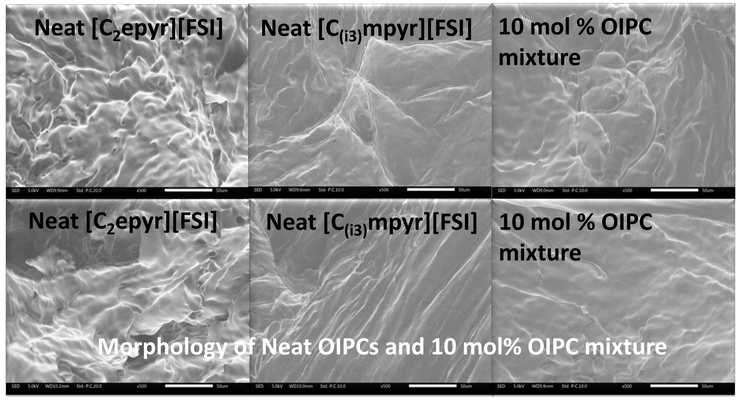
N-isopropyl,N-methylpyrrolidinium bis(fluorosulfonyl)imide

- Varying mol % of $[C_{(i3)}mpyr][FSI]$ (5 – 90 mol %) were mixed with $[C_2epyr][FSI]$ to produce OIPC mixtures.

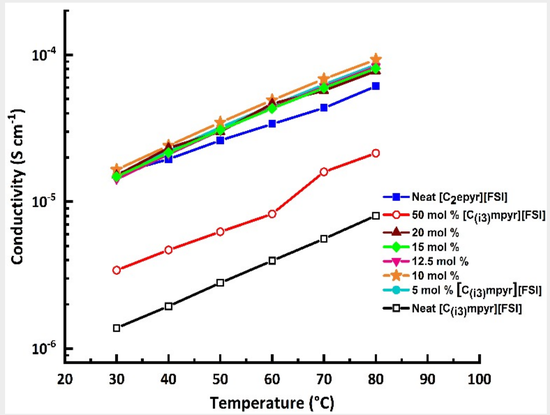
Results



Phase behaviour of OIPC mixtures



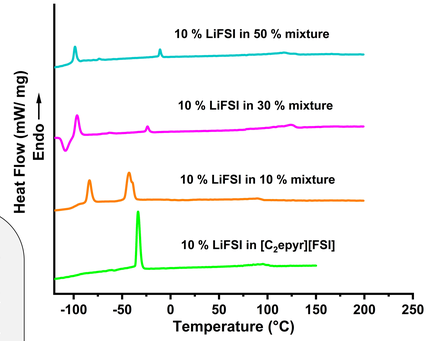
Morphology of Neat OIPCs and 10 mol% OIPC mixture



Ionic conductivity properties of OIPC mixtures

- When a small amount of slightly larger $[C_{(i3)}mpyr]^+$ is introduced, ionic conductivity increased compared to neat $[C_2epyr][FSI]$.
- We assume this is due to increase in defects/ disorder in the OIPC mixture.

Investigation of OIPC Mixtures as Potential Solid-state Electrolytes



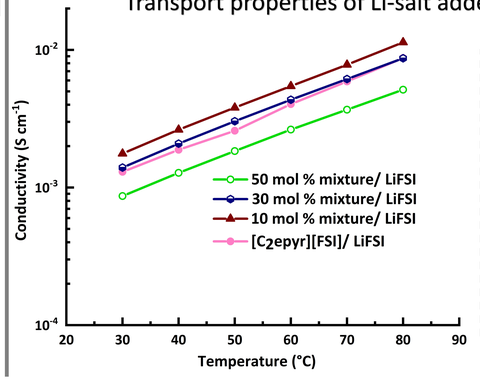
Phase behaviour of Li-salt added OIPC mixtures

- Li-salt added mixtures were quasi-solids at room temperature.
- With 10 mol % LiFSI, melting transition decreases and broadens.

With increasing $[C_{(i3)}mpyr][FSI]$ mol %,

- Melting transitions move to higher temperatures. Melting peaks remain sharp.
- Solid-solid phase transitions move to lower temperatures.
- Presence of solid solutions.

Transport properties of Li-salt added OIPC mixtures



- Li-salt added 10 mol % $[C_{(i3)}mpyr][FSI]$ binary mixture displayed slightly higher ionic conductivity than other mixtures.
- This correlates with the ionic conductivity trends of the binary OIPC mixtures.

Conclusions

- 10 mol % $[C_{(i3)}mpyr][FSI]$ added $[C_2epyr][FSI]$ mixture, showed high ionic conductivity compared to neat $[C_2epyr][FSI]$. This hints to the increase in defects/ disorder when introducing a small of slightly larger cation ($[C_{(i3)}mpyr]^+$) to $[C_2epyr][FSI]$ OIPC system.
- Li-salt added OIPC mixtures show promising results for potential use as solid-state electrolyte materials.

References

1. R. Yunis, T. W. Newbegin, A. F. Hollenkamp and J. M. Pringle, *Mater. Chem. Front.*, 2018, **2**, 1207–1214.
2. D. Al-Masri, R. Yunis, A. F. Hollenkamp, C. M. Doherty and J. M. Pringle, *Phys. Chem. Chem. Phys.*, 2020, **22**, 18102–18113.

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