

Motivation

The Ionic Liquid EMIM-TFSI (1-Ethyl-3-methylimidazolium-bis(trifluoromethylsulfonyl)imide) has been widely studied as electrolyte because of its good thermal stability, promising ionic conductivity and low viscosity. However, previous work indicates that the performance of battery cells with EMIM-TFSI based electrolyte is still disappointing due to high viscosity and low lithium ion conductivity. One way to raise the lithium ion mobility is the addition of organic carbonates.

Experimental and Results

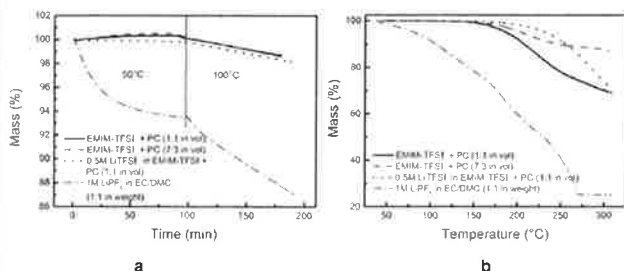
Physical and electrochemical characteristics of mixtures with EMIM-TFSI and propylene carbonate (PC) were studied to optimize the composition. PC was selected due to suited thermal properties. LiTFSI, LiBF₄ and LiPF₆ were used as conductive salt in the electrolyte mixtures. All samples were dried before testing.

Physical data of EMIM-TFSI and PC at 20 °C

	Viscosity (mPa·s)	Flash point (°C)	Melting point (°C)	Boiling point (°C)	Vapor pressure (kPa)
EMIM-TFSI	38,8	> 350	-13	--	--
PC	2.89	132	-49,2	242	0.017

Thermal stability

- no weight loss after 90 min heating at 50 °C
- less than 2% weight loss after 90 min heating at 100 °C
- almost no weight loss up to 150 °C, apparent weight loss at around 160 °C with heating rate 10 K/min
- extreme weight loss of commercial electrolyte

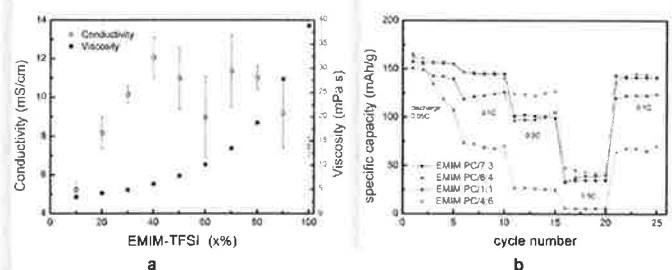


Thermogravimetric Analysis (TGA) of electrolyte mixtures
(a) isothermal mass change at 50 °C and 100 °C. Exposition time : 90 min.
(b) mass change between room temperature and 300 °C, heating :10 K/min.

* NMC: LiNi_{0.33}Mn_{0.33}Co_{0.33}O₂,
LiBF₄: Lithium tetrafluoroborate,
LiPF₆: Lithium hexafluorophosphate

Electrolyte with different EMIM-TFSI content

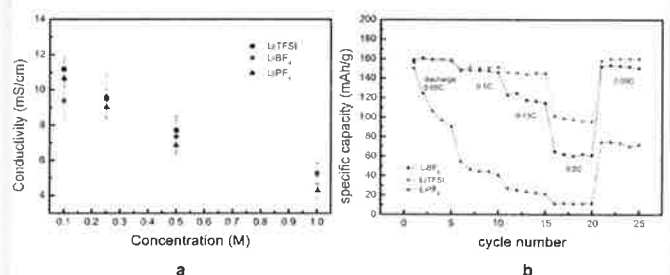
- Addition of EMIM-TFSI causes an increase in viscosity
- Conductivity rises with increasing volume content of EMIM-TFSI from 0 to 40%
- Further increase of EMIM-TFSI content increase causes lower conductivity values
- At 70 vol-% of EMIM-TFSI a second conductivity maximum 11.4 mS cm⁻¹ appears
- mixtures with higher conductivity shows higher specific capacity in the NMC/Li cell



(a) ionic conductivity (at room temperature) and viscosity (at 20 °C) of EMIM-TFSI mixed with PC;
(b) specific capacity of electrolytes with different PC content at room temperature in swagelok cells using NMC/Li⁺ as electrodes, discharged at variable C-rates between 3.0 and 4.3 V.

Electrolyte with different conductive salts

- similar conductivity with different lithium salts
- better compatibility between Li-TFSI and NMC
- high capacity loss of the cell with LiPF₆ based electrolyte



(a) ionic conductivity of mixture EMIM-TFSI+PC (7:3 in vol.) with different lithium salts at room temperature;
(b) specific capacity of electrolyte with different lithium salts at room temperature in swagelok cells using NMC/Li⁺ as electrodes, discharged at variable C-rate between 3.0 and 4.3 V

Conclusion

- Physical and electrochemical properties of electrolyte mixtures with EMIM-TFSI and PC were systematically studied.
- Electrolytes with 30 vol% of PC showed best performance.
- Variation of lithium salts has small effect on the ionic conductivity of the electrolyte.
- LiTFSI based electrolytes confirm best compatibility with NMC electrodes.