

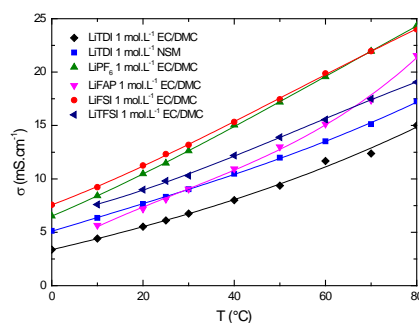
LiTDI and LiFSI as substitutes for LiPF₆ in Li-ion batteries electrolytes

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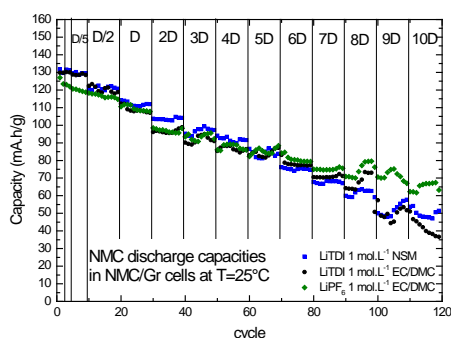
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Most of the lithium-ion batteries (LiBs) used in electrical devices currently available contain lithium hexafluorophosphate (LiPF₆) dissolved in a mixture of alkylcarbonates as the electrolyte mainly because of [1] the high conductivity of LiPF₆ solutions, its help in the formation of a protective SEI on graphite and its ability to passivate aluminum, a low-cost metal used as current collector. Other important electrolyte properties include low viscosity and a large life-span without safety risks, especially when damaged. However, the thermodynamic instability and moisture sensitivity of LiPF₆ induce high risks of releasing HF and/or PF₅ via its thermal decomposition/hydrolysis in the presence of water. These drawbacks have been highly criticized justifying the importance in finding a replacement. Owing to (i) its high thermal stability, (ii) low sensitivity to the presence of water and (iii) its electrochemical stability [2], the lithium 4,5-dicyano-2-(trifluoromethyl)imidazol-1-ide (LiTDI) is seen as a promising substitute for LiPF₆ in LiBs.



In a previous study [3] the poor ion pair (IP) dissociation of LiTDI was discussed and found to be around 30 % in an ethylene carbonate/dimethyl carbonate (EC/DMC) mixture. This poor IP dissociation is not only responsible for the lower conductivity of LiTDI in EC/DMC as compared to LiPF₆ but also for the important capacity drops at higher C rates. In



order to address these defects, a new thermally stable solvent mixture [4] (denoted NSM) allowing a better dissociation of the salt (37%), a higher conductivity (8.5 mS.cm⁻¹ instead of 6.1 mS.cm⁻¹ in EC/DMC at 25°C), a lower viscosity (2.4 mPa.s instead of 2.8 mPa.s in EC/DMC at 25°C) and good capacity retention at high C rates (up to 10C) in half-cells (graphite/Li and NMC/Li) as well as NMC/graphite cells has been found.

Another promising substitute for LiPF₆ is the lithium bis(fluorosulfonyl)imide (LiFSI) for its high conductivity, high thermal stability and its resistance to hydrolysis. In this work, transport properties (conductivity, viscosity, transference number and ion-pair dissociation) of LiTDI and LiFSI, their passivating properties (SEI and aluminum) and the cell capacities obtained while using them will be compared to that of several salts including LiPF₆.

1. Eshetu, G.G., et al., *Electrochimica Acta*, 2013. **102**(0): p. 133-141.
2. Niedzicki, L., et al., *Journal of Power Sources*, 2011. **196**(20): p. 8696-8700.
3. Berhaut, C.L., et al., *Electrochimica Acta*, 2015. **180**: p. 778-787.
4. Patent pending.