Investigation of New Guanidinium-Based Ionic Liquids as Possible Electrolytes in Lithium-Ion Batteries

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Ionic Liquids (IL) are substances that consist only of ions and exist in the liquid state at temperatures below 100 °C. Various features such as their very high flash point, low vapour pressure and the extended electrochemical potential window make them quite attractive as electrolytes for lithium-ion batteries. On the other hand their mostly high viscosities often lead to low ionic conductivities.

Two new guanidinium-based ionic liquids (Me_5cHexGua-TFSI and Me_4(2-EtHex)$_2$Gua-TFSI, Fig. 1) were investigated. Their conductivity, viscosity, potential window at platinum and compatibility with the anode materials graphite and $Li_4Ti_5O_{12}$ (LTO) and the cathode material $LiFePO_4$ (LFP) were determined. The results of the guanidinium-based electrolytes were compared with the data of a carbonate-based standard electrolyte [1 M $LiPF_6$ in EC:DMC (1:1)(wt)] and two other IL-based electrolytes [0.3 mol/kg $LiPF_6$ in BMIM-PF_6 and 0.3 mol/kg LiTFSI in BMPL-TFSI].

The guanidinium-based electrolytes – 0.3 mol/kg LiTFSI in Me_5cHexGua-TFSI and 0.3 mol/kg LiTFSI in Me_4(2-EtHex)$_2$Gua-TFSI – show high viscosities (501 mPas and 751 mPas at 23 °C, Fig. 2) and low conductivities (0.3 mS/cm and 0.1 mS/cm at 25°C, Fig. 3). On the other hand an increased anodic stability of 4.8 V vs. Li/Li^+ on platinum is detected.
The new guanidinium electrolytes are not compatible with graphite electrodes as no adequate solid electrolyte interphase (SEI) is being formed and therefore intercalation of the cation of the IL can occur.
An application of LTO and LFP electrodes with the two guanidinium-based electrolytes is generally possible. Due to the high viscosities and low conductivities the application in lithium-ion batteries is however limited.

Abbreviations:
BMIM-PF_6: 1-butyl-3-methylimidazolium hexafluorophosphate
BMPL-TFSI: 1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl)imide
$LiPF_6$: Lithium hexafluorophosphate
LiTFSI: Lithium bis(trifluoromethylsulfonyl)limide

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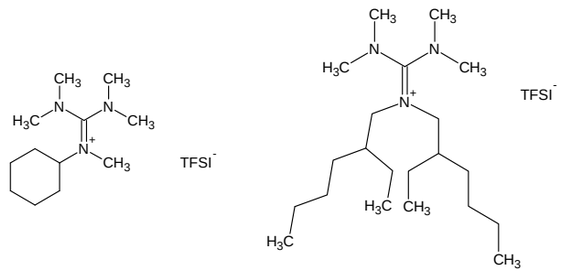

Fig. 1: Chemical structures of (left) Me_5cHexGua-TFSI and (right) Me_4(2-EtHex)$_2$Gua-TFSI.

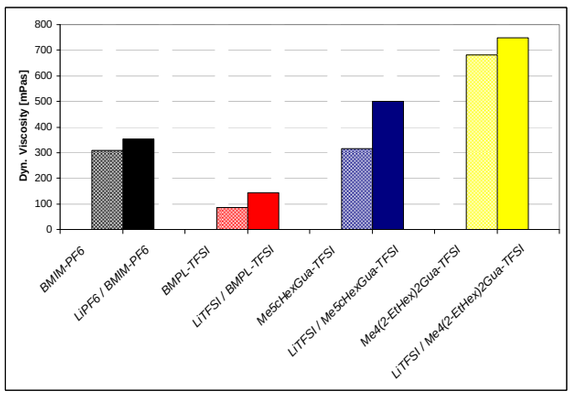

Fig. 2: Comparison of viscosities of different ionic liquids (with and without salt) at 23°C.

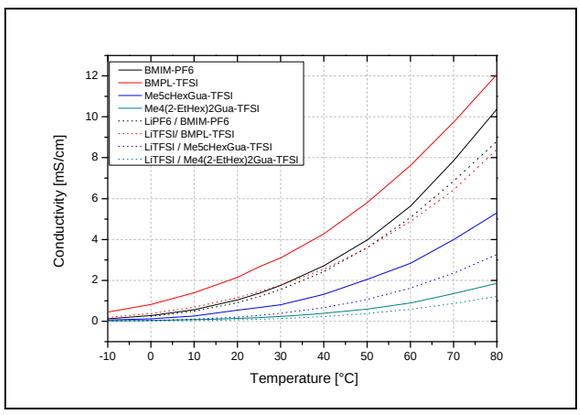

Fig. 3: Comparison of conductivities of different ionic liquids (with and without salt) at 25°C.