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Polymer Gel Electrolytes based on Ionic Liquids for Li-Ion-Batteries

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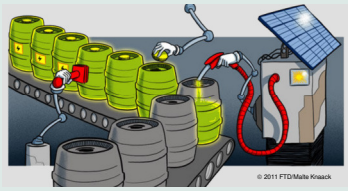
Summary

- Interaction of ionic liquid based electrolytes with graphite electrodes
- Improvement of the Li-ion battery performance with gel polymer electrolytes
- Li^+ conductivity, safety, cycle stability

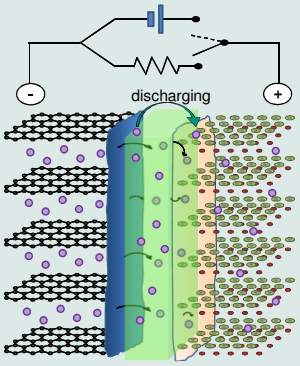


Motivation

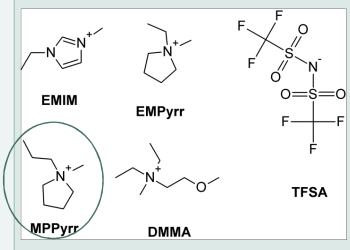
- Intrinsically safe Li-Ion battery electrolytes are not available
- Combination of ionic liquids and gel polymer electrolytes for new feasible gel polymer electrolytes: incombustible and leak proof
- Increase of the Li-ion mobility by the use of organic carbonates
- Functionality of Li-ion batteries:

Li-Ion Cell

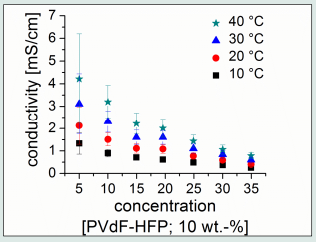
- Negative graphite electrode
- Positive NMC electrode
  (NMC = $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$)
- Conducting salt: LiTFSA*
- Gel features:
  - highly elastic, temperature stable
  - gel as separator
  - accurate ion mobility
- PVdF-HFP based gel electrolyte



Ionic Liquids in PVdF-HFP gel electrolytes



EMIM: ethylmethylimidazolium
EMPyrr: ethylmethylpyrrolidinium
MPPyrr: methylpropylpyrrolidinium
DMMA: diethylmethyl(2-methoxyethyl)ammonium
*TFSA: bis(trifluoromethylsulfonyl)azanide
(= TFSI: bis(trifluoromethylsulfonyl)imide)



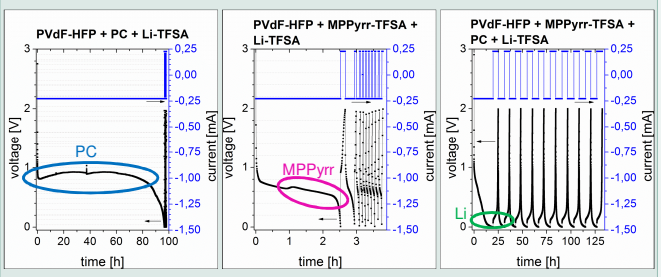
Conductivity of MPPyrr-TFSA based gel electrolytes as a function of the PVdF-HFP concentration at different temperatures. c (LiTFSA) = 0.5 mol/kg

- Mixture of MPPyrrTFSA + LiTFSA + PVdF- HFP: most balanced properties
- Decrease of the ionic conductivity with increasing PVdF-HFP content
- Best gel properties at 10 – 20 wt.-% PVdF-HFP concentration

Ionic liquid (+PVdF-HFP + LiTFSA)	κ [mS/cm] (20 °C)	ECW [V]
EMIM-TFSA	1.7 ± 0.6	5.1
EMPyrr-TFSA	0.07 ± 0.02	6.1
MPPyrr-TFSA	1.0 ± 0.3	5.9
DMMA	0.37 ± 0.18	5.4

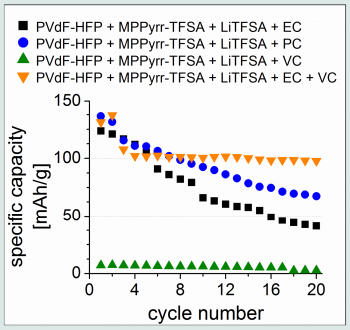
c (PVdF-HFP) = 10 wt.-%
c (LiTFSA) = 0.5 mol/kg electrolyte

Organic carbonates in IL-PVdF-HFP gels, Li|C half cells



- Li|gel polymer electrolyte|C - cells without a separator
- Current: C/20 ⟶ full charge and discharge in 20 hours
- Intercalation of **propylene carbonate (PC)** and **N-methyl-N-propylpyrrolidinium (MPPyrr)** into the graphite layer
- Synergetic effect of PVdF-HFP + MPPyrr-TFSA + PC at the graphite-electrolyte interface enables **lithium** intercalation
  ⟹ Successful cycling of the Li|C - half cell without any additive!

NMC|gel|C - cell performance



- Discharge capacity is shown
- Current: C/20 (charge and discharge without I/U-charging)
- Temperature: 20 – 25 °C
- Without any additional separator
- Only gel polymer discs were used, no wetting of the electrodes
- Swagelok type cell design
- Thickness of the gels: ~ 200 – 300 µm
- c (LiTFSA) = 0.7 mol/kg
- c (organic carbonate) = 25 wt.-%
- c (PVdF-HFP) = 12 wt.-%
- EC = ethylene carbonate
- PC = propylene carbonate
- VC = vinylene carbonate

- Cell with 25 wt.-% vinylene carbonate (VC) not working
- Continuous decrease of the specific capacity without the addition of VC
- High reversibility of the specific capacity with EC and VC in the gel matrix

Conclusions

- Ionic liquids can successfully be applied in PVdF-HFP gel polymer electrolytes
- Best results for the ionic liquid N-methyl-N-propylpyrrolidinium bis(trifluoromethylsulfonyl)azanide
- SEI effect on graphite electrodes based on a synergetic effect of PVdF-HFP, MPPyrr-TFSA, Li-TFSA, and PC
- Successful realization of Li-ion-polymer cells with a cell configuration of [$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$|gel polymer electrolyte|C]

Acknowledgements

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