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All-Solid-State Rechargeable Batteries: Manufacturing of Composite Cathodes

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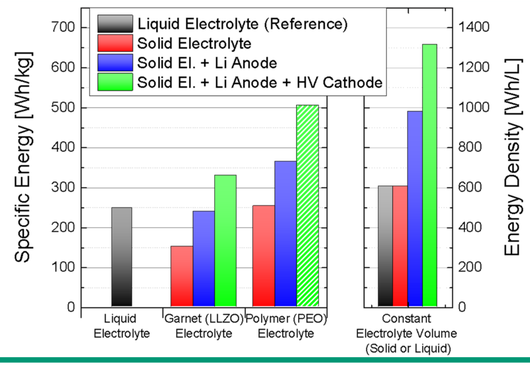
Motivation

- All-solid-state batteries have great potential for both improvement of cell safety and increase of energy density
- Calculations reveal specific energies beyond 400 Wh/kg and energy densities of more than 1200 Wh/L
- Safety improvements by removing the flammable liquid electrolyte
- Great challenges remain in manufacturing of the components and the cell stack
- Especially the composite cathode requires a complex structure to ensure both good electric and ionic conductivity

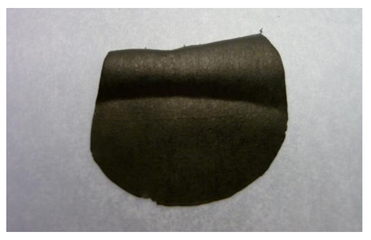


Manufacturing of solid-state electrolytes and composite cathodes

- Solid precursors
- All steps carried out in inert Ar atmosphere.
- Electrolyte PEO : $LiClO_4$ (Li: O ratio: 1:16)
- Processing
  - solvent-based (wet)
  - solvent-free (dry)
- Additional steps for homogenizing and shaping

Powder	Mixing / Drying	Intermediate	Extrusion	Semi-manufactured	Shaping	Electrolyte / Cathode
Polymer, Salt for Cathode: Active Material, Carbon Black	solvent-based or solvent-free (dry)	Polymer with dissolved salt rigid at RT	Twin-screw extruder (heated)	Rectangular shape	e.g. rolling, pressing	Thin foils ~10-500µm

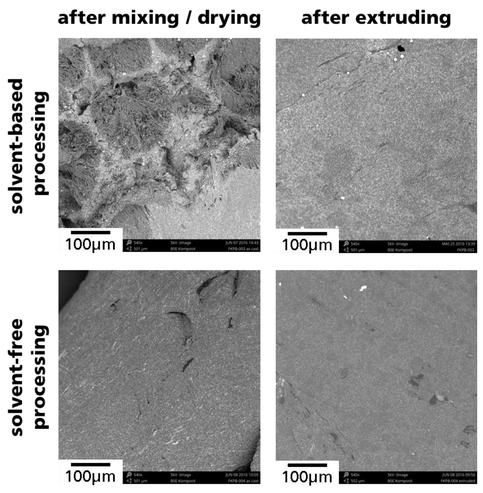
	PEO (M_w=100k)	Salt $LiClO_4$	Active Material LFP	Additive Carbon Black
Electrolyte	87%	13%	-	-
Composite Cathode	52,2%	7,8%	32%	8%



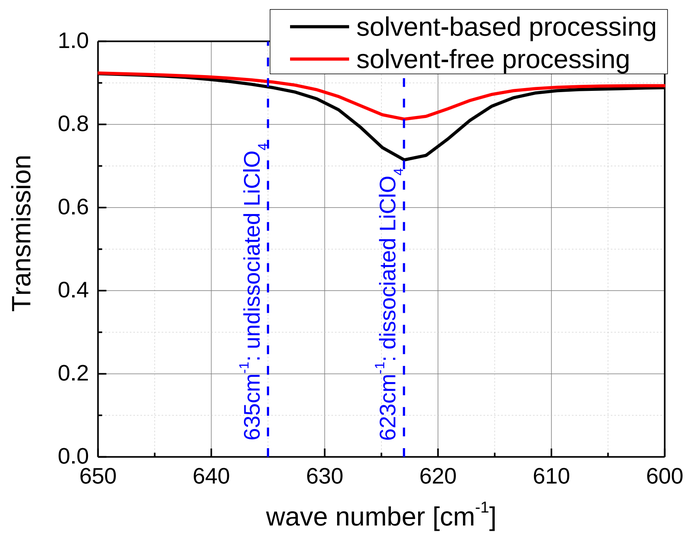
Results

Material Analysis

Cathodes: SEM top-view

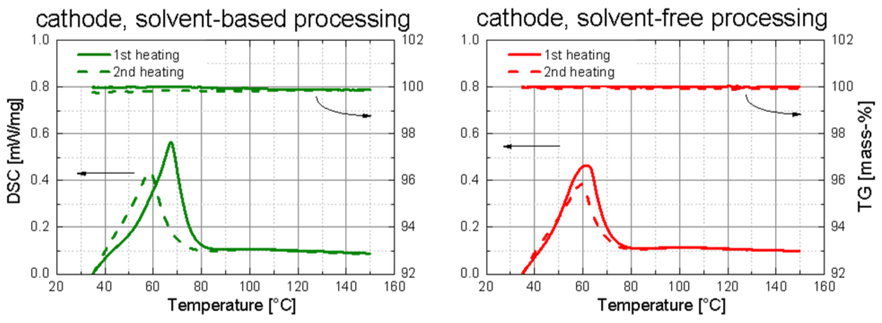


IR-spectra of processed electrolytes



- Solvent-free processed electrodes show more homogeneous structure after mixing
- IR-spectra show that $LiClO_4$ is fully dissociated in polymer for both wet and dry processing

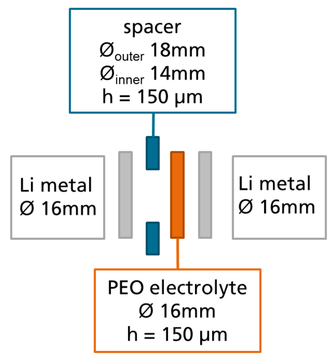
Cathodes: TG-DSC



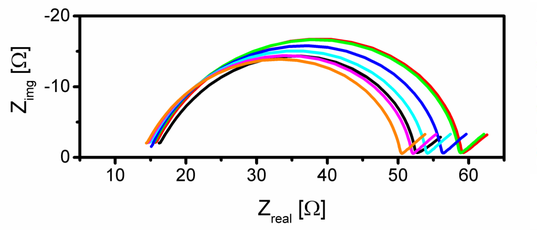
- similar behavior for both processing types
- sample history is important (e.g. prior heating during mixing)

Electrochemical Characterization

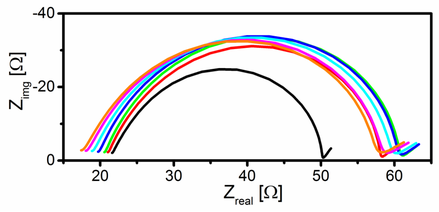
- 30 cycles in plating/stripping experiment
- EIS measurement after each cycle
- 70°C, 0.1 mA/cm²
- Spacer prevents short circuiting and fixes the thickness h = 150µm of the electrolyte layer

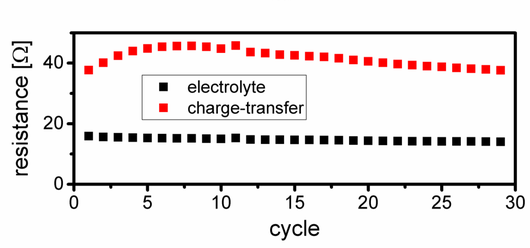


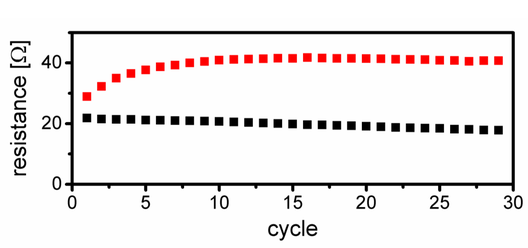
solvent-based



solvent-free







- Both electrolytes show same good conductivity
- Stable charge-transfer in solvent free electrolyte shows stable PEO / Li interface

Conclusion

- All-solid-state electrolytes and composite cathodes can be produced in a dry, solvent-free process
- SEM, IR, TG-DSC and CV/EIS measurements confirm that $LiClO_4$ salt is fully dissociated in PEO polymer for both wet and dry processing
- Dry processing eliminates the need for hazardous solvent (e.g. acetonitrile) in the production of solid-state electrodes and electrolytes
- Solvent-free processing may lead to more cost-effective, safe production in the large scale
- Further investigations will focus on improving methods to characterize quality parameters of the product, e.g. porosity and homogeneity