Thin Film Technology
Lukas Lödige
lukas.loedige@kit.edu
+49 721 608 46990

Thin Film Technology
Thilo Heckmann
thilo.heckmann@kit.edu
+49 721 608 41426

Karlsruhe Institute of Technology

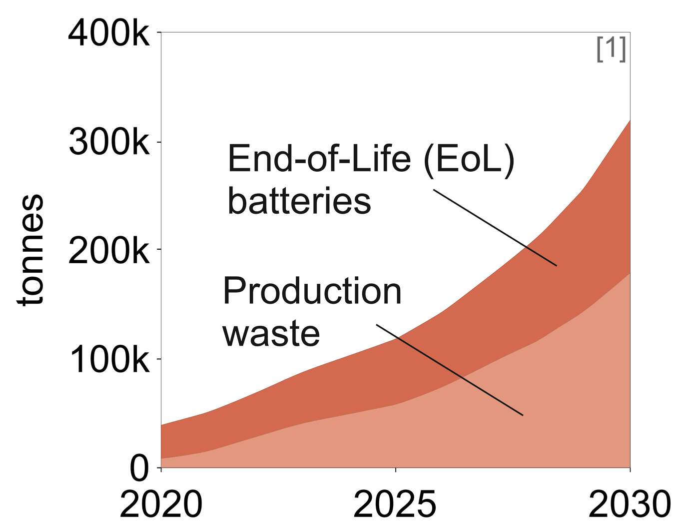
About Mass Transfer of Multicomponent Electrolyte Solvent Mixtures during Recycling of Lithium-ion Batteries

Lukas Lödige[1,2], **Thilo Heckmann**[1,2], **Philip Scharfer**[1,2], **Wilhelm Schabel**[1,2]
[1] Thin Film Technology (TFT), Karlsruhe Institute of Technology (KIT), Karlsruhe
[2] Material Research Center for Energy Systems (MZE), Karlsruhe

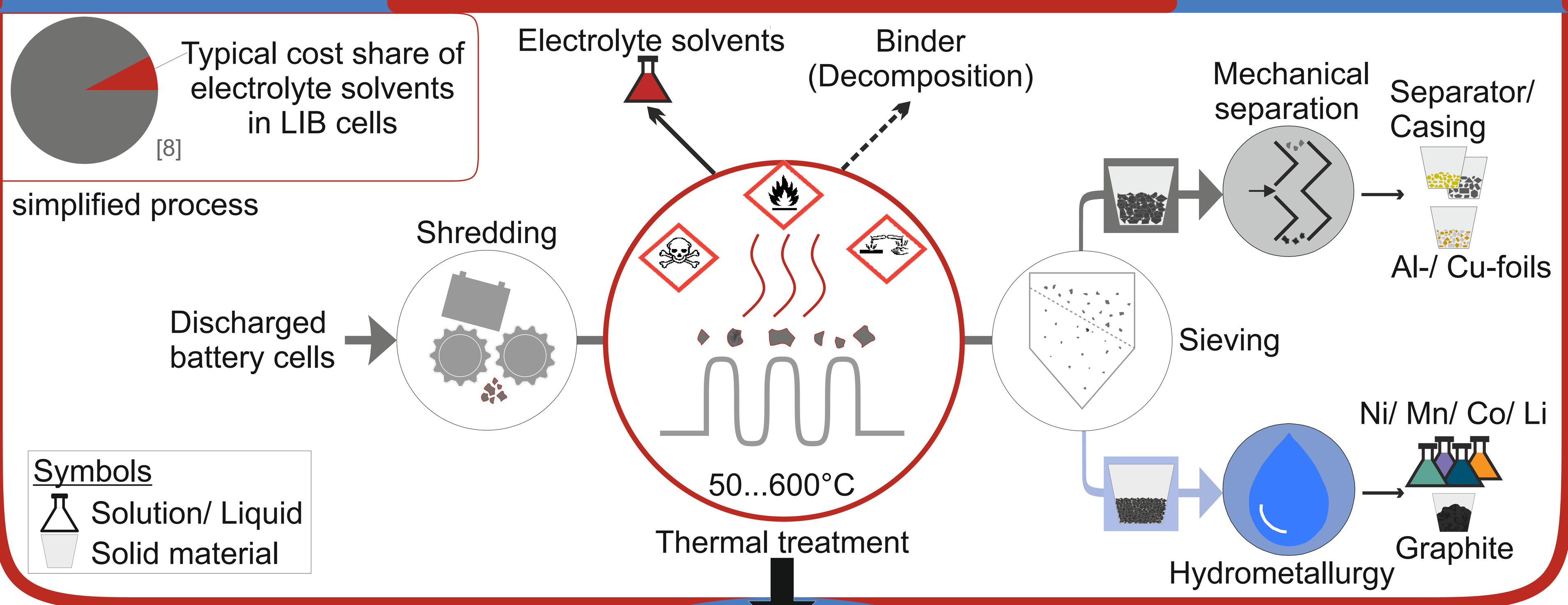
Motivation

Improved recycling is inevitable for sustainable LIB market and future material supply security

Rapidly increasing amount of recyclable Lithium-ion batteries (LIBs) in Europe



Process flow chart

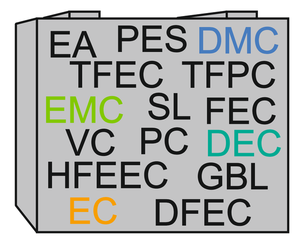


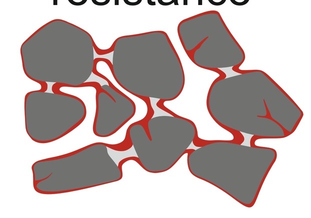
Challenges

Predict optimum residual electrolyte solvent loading in terms of ecology and economy

Complexity

Multicomponent mixtures | Structure-dependent mass transport resistance



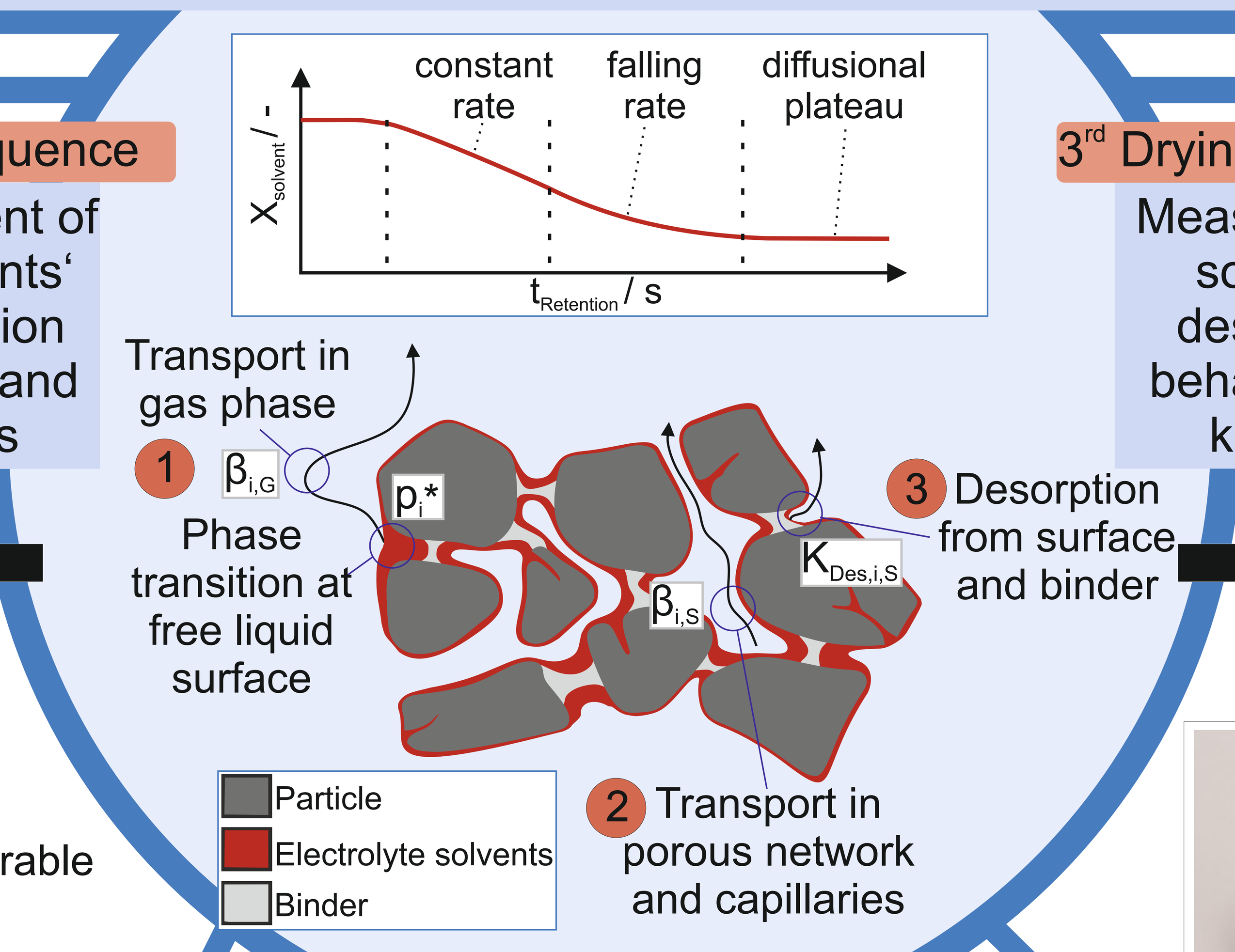


Representative electrolyte solvent mixture:
DMC - Dimethyl carbonate
EMC - Ethyl methyl carbonate
DEC - Diethyl carbonate
EC - Ethyl carbonate

Aim of the Investigations

Understanding the microscale phenomena in drying and desorption of multicomponent solvent mixtures in complex structures

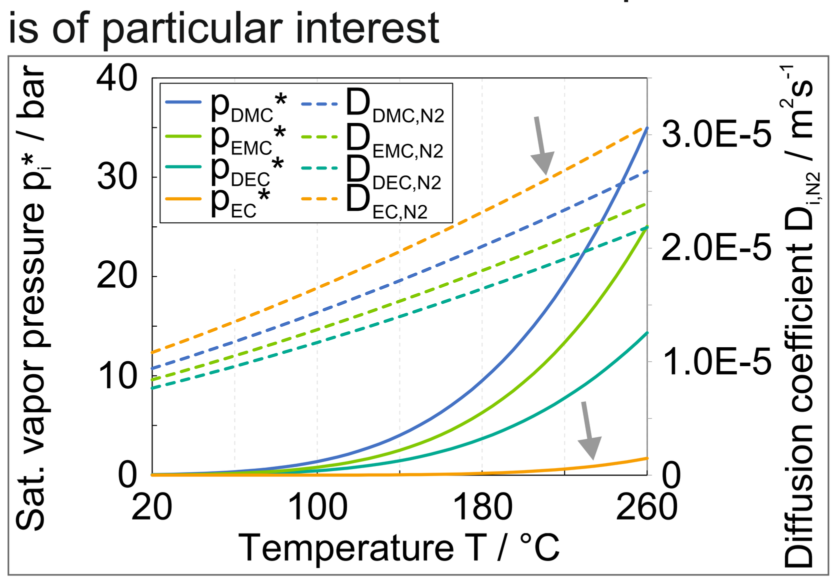
Methods and Results



1st Drying sequence

Assessment of the solvents' evaporation behavior and kinetics

Full removal of low volatile component EC is of particular interest



➡ EC shows unfavorable thermodynamic ($\alpha_{i,j}$) but favorable gas kinetic (K_G) behavior compared to the other solvents

Are these considerations confirmed by the experiments?

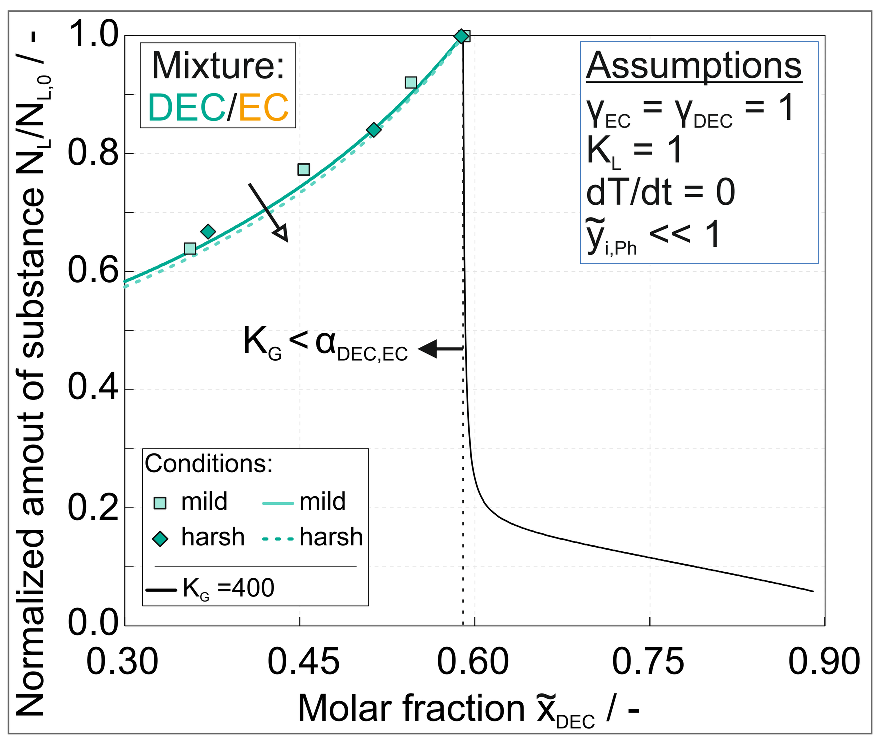
➡ Predictive calculation of the evaporation for the binary mixture EC/DEC into a N_2-flow

Governing equations:

$$NTU_{i,N2} = \frac{A_{Ph}\,\beta_{i,N2}}{\dot{N}_{N2} / \tilde{\rho}_{N2}} \qquad \alpha_{DEC,EC} = \frac{\gamma_{DEC,mix}\, p_{DEC}^*}{\gamma_{EC,mix}\, p_{EC}^*}$$

$$K_G = \frac{1+NTU_{DEC,N2}}{NTU_{EC,N2}} \cdot \frac{1+NTU_{EC,N2}}{NTU_{DEC,N2}} \qquad K_L = \exp\left(\frac{\dot{N}_i / \tilde{\rho}_L}{A_{Ph} / \beta_{i,L}}\right)$$

[2]



➡ Simulative prediction of the evaporation (with assumptions) of a DEC/EC mixture matches the experimental results

» Validation of assumptions and simplifications | Extension of the calculation to more components

2nd Drying sequence

Understanding the mass transport of solvent in the porous network of the particulate system

Defined drying of particulate samples with varying porosity ε and tortuosity τ

[3] Convective drying | Vacuum drying [4]

Important aspects

Diffusion coefficient in a particulate system [5]	Mass flow with resistance in the particulate system [5]	Diffusion regime depending on Kn number [6]
$D_{i,S} = D_{i,G}(1-\sqrt{1-\varepsilon})$	$\dot{m}_i = K_S\,\tilde{M}_i \frac{1}{\frac{1}{\beta_{i,G}} + \frac{s}{D_{i,S}}} \tilde{\rho}_G\,(\tilde{y}_{i,Ph} - \tilde{y}_{i,\infty})$	$D_{ij,eff} = \frac{\varepsilon}{\tau} D_{ij,St}$ 0.1<Kn; $D_{ij,eff} = \frac{\varepsilon}{\tau} D_{ij,Kn}$ 10>Kn

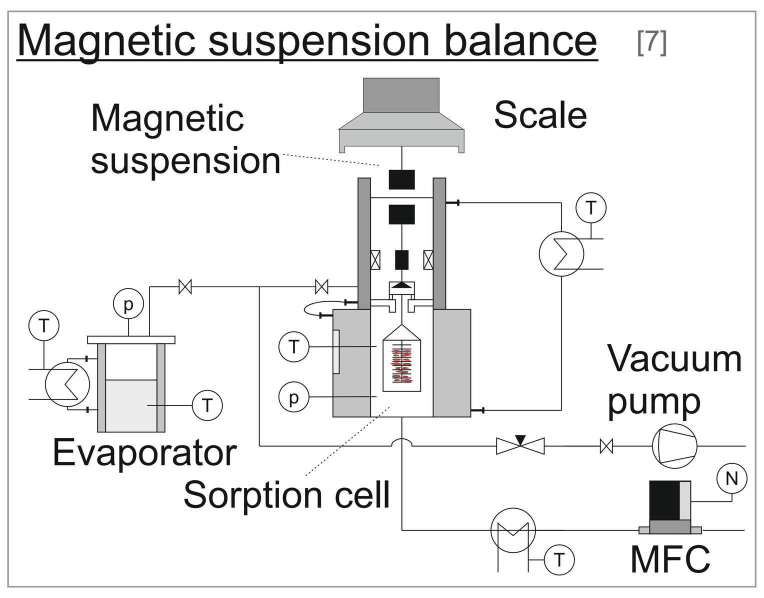
➡ Evaluate the dominant mass transport resistances during drying

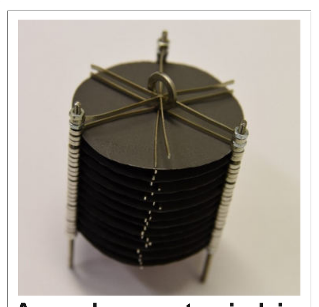
» Extension of the model to include structural influences

3rd Drying sequence

Measuring the solvents' desorption behavior and kinetics

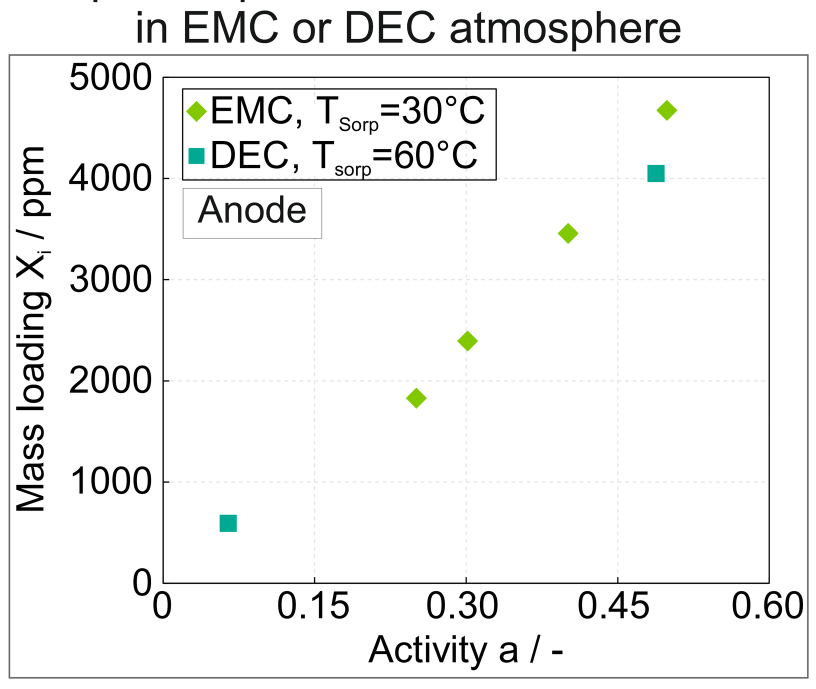
Magnetic suspension balance [7]





Anode material in a sample holder

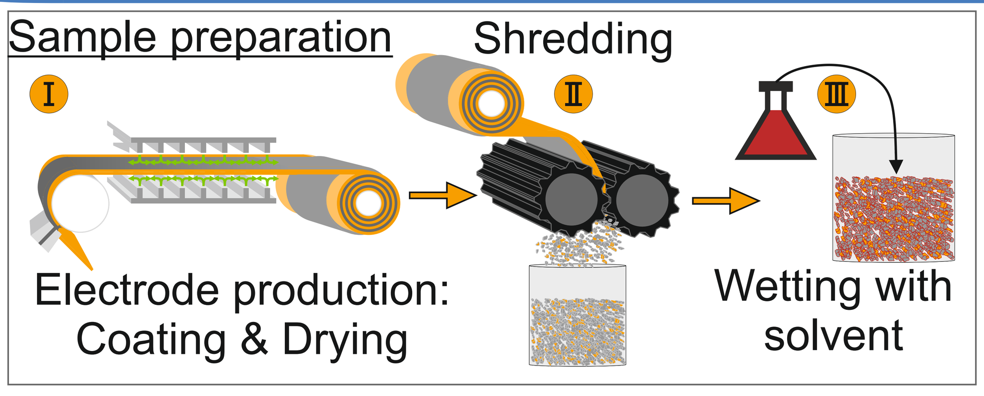
Gravimetric measurement: Sorption equilibria for a standard anode in EMC or DEC atmosphere



➡ Information on residual solvent loadings under certain process conditions

» Integration of the desorption into the model

Sample preparation



Outlook

Experimental investigation of the mass transport resistances ➡ Holistic description of the drying process in a simulative model ➡ Process optimization: residual solvent content, time, energy consumption

➡ Development of efficient recycling processes requires deep understanding of heat and mass transfer

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