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ITUN
Institute of Thermal-, Environmental- and Resources' Process Engineering
Prof. Dr.-Ing. habil. Andreas Braeuer

Quantification of Electrolyte Solvent Composition from Lithium-ion Battery Recycling using Raman Spectroscopy

Tom Goldberg, *Andreas S. Braeuer*

Motivation & Introduction

650.000 t (proj. 2025) [2]

Spent lithium-ion battery (LIB)

10-15 wt.% of total LIB content [1]

Black mass

Electrolyte solvent mixture (ESM)

5 main electrolyte solvents in LIBs production

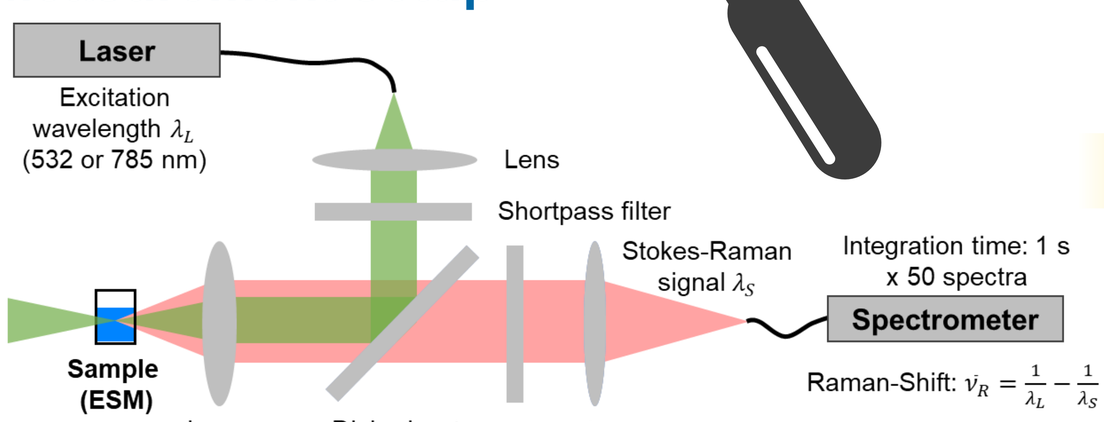
New EU regulation concerning batteries and waste-batteries (Recycling rate: 70 wt.% of total LIB by 2030) shows **recycling necessity of the electrolyte solvent mixture (ESM)**

Different LIB types, generations and manufacturers result in different and changing ESM composition

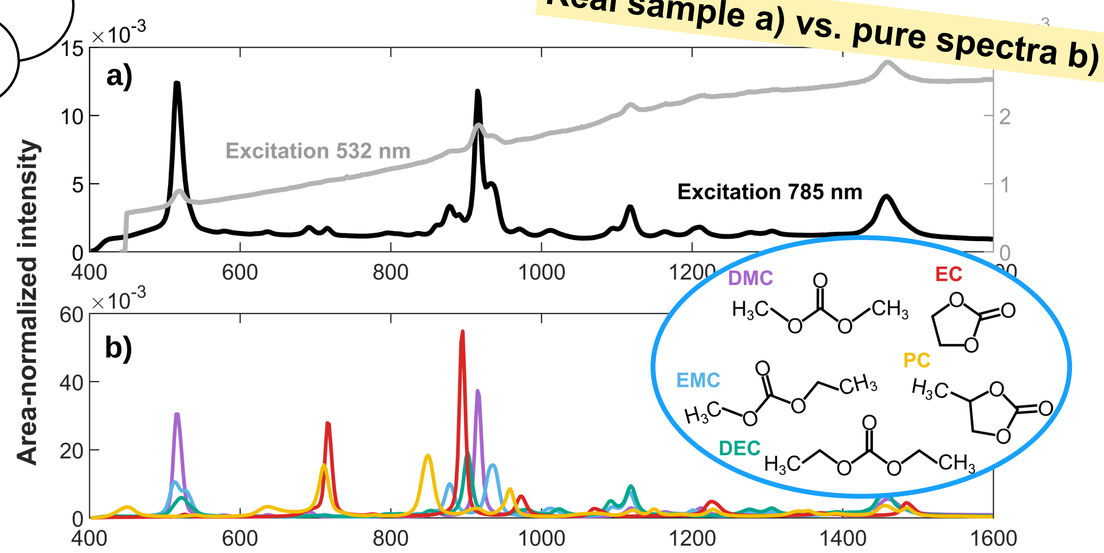
Composition analysis as starting point for conceptual design (thermodynamic data), dynamic simulations and process control

➔Raman spectroscopy with high temporal and spatial resolution for **rapid online composition analysis**

Measurement setup



Real sample a) vs. pure spectra b)



➔Longer excitation wavelength (785 nm) for lower fluorescence interference
➔Substantial peak overlap from structural similarity of the solvent compounds

Composition evaluation (CLS vs. PLS)

Classical Least-Square (CLS) regression

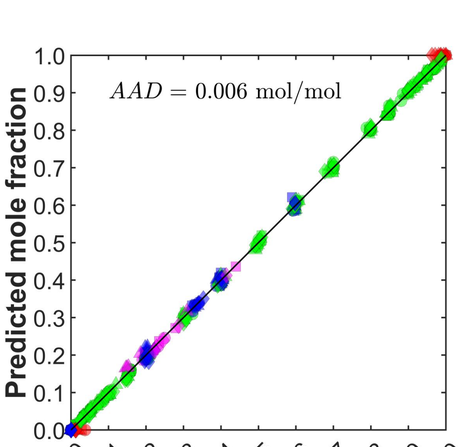
= **Physically-based** regression model

- **Spectra reconstruction** as a linear combination of pure compound spectra (linear unmixing)
- **Simultaneous background fit** during regression
- Composition evaluation based on **binary calibration sets**

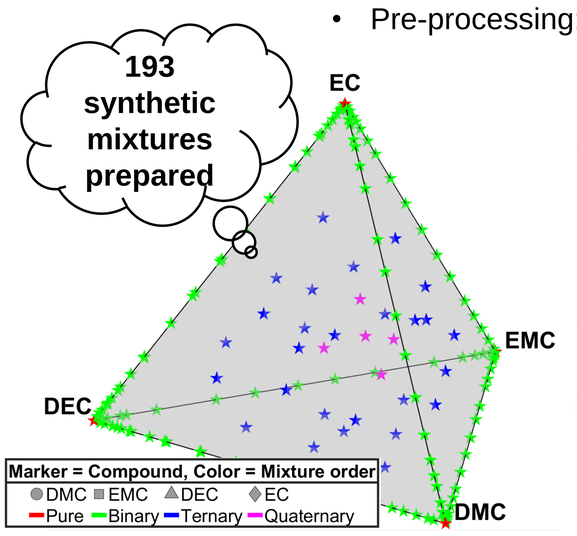
Partial-Least-Square (PLS) regression

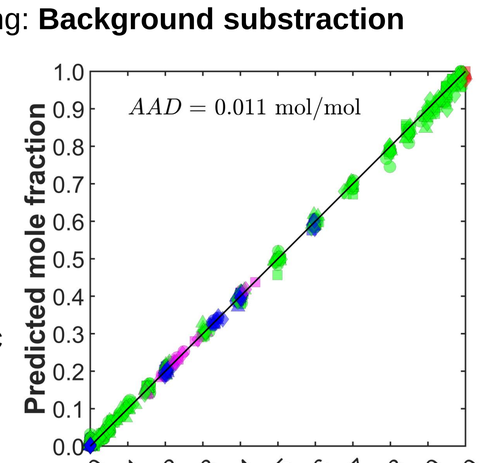
= **Chemometric** regression model

- Synthetic mixtures (up to multicompound) serve as **training data for global calibration model**
- Both spectral and composition data are projected onto **latent variables** maximizing their covariance
- Optimal number of latent variables determined by cross-validation
- Pre-processing: **Background substraction**

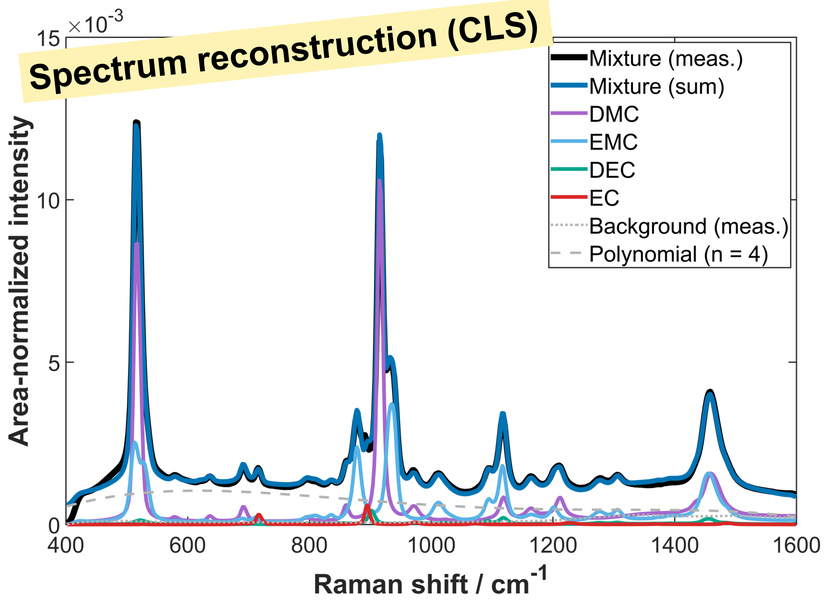


193 synthetic mixtures prepared





Application on real sample from LIB shredder



	DMC	EMC	DEC	EC	PC
GC-MS	0.50	0.45	0.02	0.03	n.d.
Raman (CLS)	0.49	0.46	0.03	0.02	n.d.
Raman (PLS)	0.54	0.45	n.d.	0.02	n.d.

No PC content

Summary & Conclusion

➔Evaluation of ESM composition from LIB recycling demonstrated; further optimisation planned
➔PLS with higher calibration effort allows better composition evaluation for higher-order synthetic mixtures, but is less robust for application on real samples from LIB shredder

[1] Xiao J., Zhou T., Shen R., Xu Z., *ACS Sustainable Chemistry & Engineering*, 2023, 11(12), 4707–4715. DOI: 10.1021/acssuschemeng.2c07116
[2] Lei, S., Sun, W., Yang, Y., *J. Hazard. Mater.*, 2022, 424 (Pt. D), 127654, DOI: 10.1016/j.jhazmat.2021.127654

TU Bergakademie Freiberg **| Institute of Thermal-, Environmental- and Resources' Process Engineering**
Leipziger Straße 28 | 09599 Freiberg | +49 3731 39-2534 | Tom.Goldberg@tun.tu.-freiberg.de | http://tu-freiberg.de/fakult4/itun