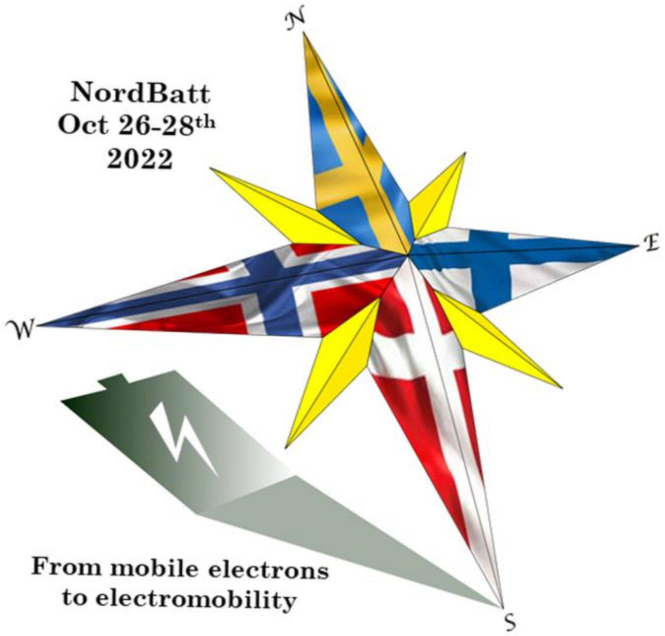

CHALMERS UNIVERSITY
of TECHNOLOGY
SWEDEN

# FINAL PROGRAMME
# INCLUDING ABSTRACTS

# TABLE OF CONTENTS

# WELCOME ADDRESS

## Welcome to NordBatt 2022!

On behalf of the International Scientific Committee and the Local Organization Committee, I am pleased to welcome you all to NordBatt 2022 – taking place as a fully in-person meeting at Chalmers University of Technology, Göteborg, Oct 26th-28th. NordBatt started in Sweden in 2013 and after touring the Nordic countries it is now, after all the struggle the last few years, back in Sweden and larger than ever before – gathering and supporting the Nordic-Baltic battery R&D&I landscape.

The theme of this edition of NordBatt, *"From mobile electrons to electromobility"*, is truly reflected in our programme. While the very usage of batteries to enable electromobility comes natural for a conference organized in "the City of Volvo", we are proud to gather the full eco-system with actors and people that conceptualize, model, create, produce, test, evaluate, install, use, recycle, *etc*… batteries, and not the least those who fund our battery R&D&I activities. Looking at the oral programme you find our 3 truly prolific international Plenary Speakers, some of the best of what the Nordic countries can offer as 8 Invited Speakers, visions from the Nordic battery industry and stakeholders at large via the 7 Special Oral presentations, and not the least 20 more Oral presentations. In total 38 presentations in plenum and to this we add a poster session, and not the least a nice conference dinner for everyone! I hope for engaging and fruitful discussions as well as exchange of ideas.

As the battery community is growing rapidly, we need to make sure that we produce the skilled labor, competencies and excellence needed for the field to innovate and prosper also in the future. Therefore, Early Stage Researchers (ESRs) are especially important. At NordBatt 2022 we emphasize this in several ways; a special, totally free of charge, 1-day pre-conference only for ESRs, to allow them to strengthen their own networks; several oral presentation slots given to ESRs, to enhance their visibility; and poster and oral presentation prizes to ESRs, acknowledging the excellence shown by ESRs and boost their CVs.

As this is the 5th NordBatt we can also celebrate, for example we in collaboration with Wiley will have a Special Collection in the prestigious journal *Batteries & Supercaps* to which all presenters at NordBatt 2022 will be invited to contribute, alongside all previous NordBatt plenary speakers.

Last, but absolutely not the least: our Partners, Sponsors and Exhibitors are those that, together with all of you, make NordBatt 2022 what it is – a meeting place for the entire Nordic battery value-chain. Please make sure to look around the exhibition area to see all new equipment *etc* on display – or perhaps test-drive an electric motorcycle?

Finally, Göteborg can be challenging weather-wise in late October, thus the NordBatt umbrella(!), but I sincerely hope you take some time to enjoy our city relaxing with your colleagues and network.

Patrik Johansson,
NordBatt 2022 Conference Chair

# ACKNOWLEDGEMENTS

We kindly acknowledge our Sponsors:

We are proud to organize NordBatt 2022 together with our Partners:

Our Media Partners are acknowledged for their help in spreading the word and raising our impact:

# GENERAL INFORMATION

**NordBatt 2022** takes place at Chalmers Conference Centre, located at Chalmersplatsen 1, the nearest tram-stop being Chalmers.

**The oral sessions** will take place in Palmstedtsalen**.** Presenters should provide their presentation via a USB stick to the speaker assistant, **at the latest**, in the break before the session in which they present. Usage of own laptops is **not permitted.**

**Plenary Speakers** are given 35 mins + 5 mins for questions.
**Invited Speakers** are given 25 mins + 5 mins for questions.
**Oral Speakers** are given 15 mins + 5 mins for questions.
**Special Orals** (10 mins) are to hear the word of industry, no abstracts available.

**The poster session** takes place in Scaniasalen, Wed 26th of Oct at 17:00. **(Remember to bring your drinks tokens for the bar!)**

**Posters** can be mounted from the morning of Wed 26th of Oct and should not be larger than A0 standing format (84.1 cm x 118.9 cm). Please remove your posters by Fri 28th of Oct 13:00 at the latest.

**Inside the poster session** we have the **Speaker's Corner**, a new feature for NordBatt. The purpose is to facilitate spontaneous short presentations for (very) small audiences. The SC is simply a high-table to place your own laptop; No beamer, no PA-system, etc. – just you, your laptop screen, and the people gathering around you at the table. A designated moderator will arrange the order of presentations, *ad hoc*.

**The NordBatt 2022 exhibition area** is located in the lobby just outside Palmstedtsalen.

**Lunch and Coffee** is included for all attendees and will be served outside Palmstedtsalen.

**Conference Dinner** is included for all attendees and will take place at "Kårrestaurangen" – same building as NordBatt, but in the very South end of the building.

# COMMITTEES

## Nordbatt 2022 Conference Chair

Patrik Johansson

## Nordbatt 2022 International Scientific Committee

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Ulla Lassi
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Fabian Årén

# PROGRAMME

## Day 1: Wed 26th of Oct

| Time | What | Presenter | Title |
|---|---|---|---|
| 09:00 | **Registration** | | |
| 09:50 | Welcome | **Patrik Johansson** (Chalmers University of Technology, Sweden) | Welcome address NordBatt 2022 |
| **Materials/Technologies I**<br>**Session Chair: Patrik Johansson** | | | |
| 10:00 | Plenary | **Jerry Barker** (Jerry Barker Consultants, England) | The Journey to Commercialization of Sodium Ion Batteries |
| 10:40 | Special Oral | **HanHo Lee** (Beyonder, Norway) | Li-ion battery capacitor with high power, extended life and moderate energy for 2030 |
| 10:50 | Invited | **Dorthe Ravnsbæk** (Aarhus University, Denmark) | The role of disorder in intercalation type electrode materials |
| 11:20 | Invited | **Aleksandar Matic** (Chalmers University of Technology, Sweden) | Towards Li-metal batteries – mechanisms and stabilisation strategies |
| 11:50 | **Lunch** | | |
| **Materials/Technologies II**<br>**Session Chair: Dorthe Ravensbæk** | | | |
| 13:00 | Invited | **Maria Hahlin** (Uppsala University, Sweden) | Operando surface analysis of battery electrodes – from model to applied systems |
| 13:30 | Invited | **Pekka Peljo** (University of Turku, Finland) | Pathways towards sustainable stationary energy storage with flow batteries |
| 14:00 | Oral | **Xiuyun Zhao** (University of Eastern Finland, Finland) | Challenges and Recent Progress of Mesoporous Silicon as Lithium-Ion Battery Anode |
| 14:20 | Oral | **Niguse Sahalie** (Uppsala University, Sweden) | Developing Electrolyte Formulations with Zwitterionic Monomers for Better Lithium-Ion Batteries |
| 14:40 | Special Oral | **Eerik Hantsoo** (NOVO Energy, Sweden) | NOVO Energy – a joint venture between Northvolt and Volvo Cars. Sustainability, scale, and a comprehensive approach to volumetric energy density in BEV cells |
| 14:50 | **Coffee** | | |
| **Materials/Technologies III**<br>**Session Chair: Pekka Peljo** | | | |
| 15:30 | Oral | **Jinhua Sun** (Chalmers University of Technology, Sweden) | 3D hollow spheres assembled by 2D nanomaterials for high-performance electrochemical energy storage |
| 15:50 | Oral | **Frode Håskjold Fagerli** (Norwegian University of Science and Technology, Norway) | Surface Termination Control of 2D MXenes for Battery Applications |
| 16:10 | Oral | **Anastasia Mikheenkova** (Uppsala University, Sweden) | Ageing Mapping in Commercial Ni-rich Lithium Ion Cells for Electric Vehicles |
| 16:30 | Oral | **Julia Maibach** (Chalmers University of Technology, Sweden) | How about Glyoxals? Evaluating alternative Electrolytes for Si/Graphite Composite Electrodes in Li-ion batteries |
| 16:50 | Special Oral | **Greger Ledung** (Swedish Energy Agency, Sweden) | Challenges and opportunities along the Swedish battery value chain |
| 17:00 | **Poster Session / Speaker's corner** | | |

## Day 2: Thurs 27th of Oct

| Time | What | Presenter | Title |
|---|---|---|---|
| **Industry / Next Generation Batteries**<br>**Session Chair:  Nataliia Mozhzhukhina** | | | |
| 09:00 | Plenary | **Martin Winter**<br>(Helmholtz Institute Münster, Forschungszentrum Jülich & MEET, University of Münster, Germany) | How smart application of rather simple electrochemical methodology can help to better understand ion transfer and metal anode batteries? |
| 09:40 | Special Oral | **Jessica Olsson & Robin Pettersson**<br>(Business Sweden, Business Finland and Innovasjon Norge) | Nordic Battery Collaboration – competition makes us sharper, collaboration makes us stronger |
| 09:50 | Special Oral | **Martin Kirkengen**<br>(Cenate, Norway) | Innovative Silicon-based Anode Materials for High-capacity Li-ion Batteries |
| 10:00 | **Coffee** | | |
| **Industry / Next Generation Batteries II**<br>**Session Chair: Ulla Lassi** | | | |
| 10:30 | Invited | **Sebastian Pohlmann**<br>(Skeleton Technologies, Estonia) | From Supercapacitors to Superbatteries – high power energy storage and its applications |
| 11:00 | Oral | **Kirill Murashko**<br>(University of Eastern Finland, Finland) | Anchoring Polysulfides and improving the specific capacity of the Lithium-Sulphur batteries |
| 11:20 | Oral | **Muhammad E Abdelhamid**<br>(Institute for Energy Technology, Norway) | Screening of biomass-derived hard carbon anodes for Na-ion batteries |
| 11:40 | Oral | **Anders Brennhagen**<br>(University of Oslo, Norway) | Tracing the (de)sodiation of $Bi_2MoO_6$ through good and bad times with operando XRD |
| 12:00 | **Lunch** | | |
| **Lithium based batteries**<br>**Session Chair:  Aleksander Matic** | | | |
| 13:00 | Invited | **Adriana Navarro-Suarez**<br>(Morrow Batteries, Norway) | Sustainability: Batteries & Gender Equity |
| 13:30 | Invited | **Josh Thomas**<br>(LiFeSiZE AB, Sweden) | Some critical steps in LIB fabrication and their influence on performance. |
| 14:00 | Oral | **Jim Shipley**<br>(Quintus Technologies, Sweden) | The Use of Isostatic Pressing in Research and Development of Scalable Production Technology for All Solid State Batteries |
| 14:20 | Oral | **Marja Vilkman**<br>(VTT Technical research centre of Finland, Finland) | Materials and Processing Methods for Stable Lithium-Metal Batteries |
| 14:40 | Oral | **Ekaterina Fedorovskaya**<br>(Aalto University, Finland) | Aging mechanisms of NMC/graphite Li-ion batteries |
| 15:00 | **Coffee** | | |
| **LCA & Recycling**<br>**Session Chair: Adriana Navarro-Suarez** | | | |
| 15:30 | Oral | **Kerstin Forsberg**<br>(Royal Institute of Technology, Sweden) | The Role of Crystallization in Closing the Loop for Battery Production and Recycling |
| 16:00 | Oral | **Martina Petranikova**<br>(Chalmers University of Technology, Sweden) | Optimized recycling of critical metals from EV Li-ion batteries via combined metallurgy |
| 16:20 | Oral | **Evelina Wikner**<br>(Chalmers University of Technology, Sweden) | A model platform for life cycle assessment of lithium-ion battery production and use |
| 16:40 | Oral | **Ulla Lassi**<br>(University of Oulu, Finland) | Coprecipitation of high-nickel mixed metal hydroxide precursors for LIBs: Improved sustainability in the battery value chain |
| 19:00 | **Conference Dinner** | | |

## Day 3: Fri 28th of Oct

| Time | What | Presenter | Title |
|---|---|---|---|
| **Modelling<br>Session Chair: Fride Vullum-Bruer** | | | |
| 09:00 | Plenary | **Céline Merlet**<br>(Université Paul Sabatier, France) | Multi-scale models of energy storage materials: Always a trade-off between accuracy and speed? |
| 09:40 | Oral | **Yang Li**<br>(Chalmers University of Technology, Sweden) | Electrochemical-Thermal Model-Based Fast Charging for Lithium-Ion Batteries: Nonlinear Inversion-Based Output Tracking Control |
| 10:00 | Special Oral | **Hanna Bryngelsson**<br>(Polestar, Sweden) | Towards more sustainable EV batteries |
| 10:10 | **Coffee** | | |
| **Modelling II<br>Session Chair: Daniel Brandell** | | | |
| 10:30 | Invited | **Simon Clark**<br>(SINTEF Industry, Norway) | Accelerating battery innovation through FAIR model-based design |
| 11:00 | Special Oral | **Henrik Ekström**<br>(COMSOL, Sweden) | Modeling Thermal Runaway in COMSOL Multiphysics |
| 11:10 | Oral | **Ivano E. Castelli**<br>(Technical University of Denmark, Denmark) | Autonomous Workflows for an Accelerated Design of Battery Electrodes |
| 11:30 | Oral | **Melania Kozdra**<br>(Uppsala University, Sweden) | Li+ transport phenomena in a composite solid-state electrolyte |
| 11:50 | Oral | **Øystein Gullbrekken**<br>(Norwegian University of Science and Technology, Norway) | Charge Transport Properties in Concentrated Polymer Electrolytes by Molecular Dynamics Simulations |
| 12:10 | | **Patrik Johansson & Alexey Koposov**<br>(University of Oslo, Norway) | Closure + NordBatt 2024 |

# ORAL ABSTRACTS

# The Journey to Commercialization of Sodium Ion Batteries

**Dr. Jerry Barker**

*Jerry Barker Consultants, Oxfordshire, England, UK*

Jerry Barker is to many of us the face of Faradion Ltd. As the founder and chief scientist, he brought them and the sodium-ion battery technology largely to where it is today. However, his expertise in moving excellent battery material research into products via scale-up, start-ups, and IP has a longer history than that – he was also active in the field of lithum-ion batteries at Valence Technologies for many years.

# The role of disorder in intercalation type electrode materials

## Prof. Dorthe Ravnsbæk

Dorthe Ravnsbæk is a Professor in Materials Chemistry. In 2012, she got into the field of batteries working for two years as a post doc with Prof. Yet-Ming Chiang at MIT. Afterwards she established her own line of research at University of Southern Denmark and since November 2021 at Department of Chemistry at Aarhus University. She has a strong focus on synthesis and structural characterization of electrode materials using operando methods.

# Towards Li-metal batteries – mechanisms and stabilisation strategies

## Prof. Aleksandar Matic

Aleksandar Matic is Professor of Physics and Head of the Division for Materials Physics; Department of Physics, Chalmers University of Technology. His research focuses on materials for energy applications, in particular for next generation batteries, interface engineering of Li-metal surfaces, development of new electrolytes and operando characterization.

# Operando surface analysis of battery electrodes – from model to applied systems

## Assoc. Prof. Maria Hahlin

Maria Hahlin is employed as a senior lecturer at Structural Chemistry Program/X-ray Photon Science program at Uppsala University. She has a strong background in characterizing and understanding functional interfaces in batteries, utilizing traditional UHV based photoelectron spectroscopy for understanding and improving battery interfaces, as well as developing operando possibilities for characterizing battery interfaces using ambient pressure photoelectron spectroscopy.

# Pathways towards sustainable stationary energy storage with flow batteries

## Assoc. Prof. Pekka Peljo

Pekka Peljo heads the Research group of battery materials and technologies at the University of Turku, Finland, since 2020. His team is one of the largest groups in Europe focusing on electrochemistry of flow batteries, focusing on discovery of new redox couples and development of alternative flow battery technologies. He is a coordinator of three European projects and a recipient of the ERC StG.

# Challenges and Recent Progress of Mesoporous Silicon as Lithium-Ion Battery Anode

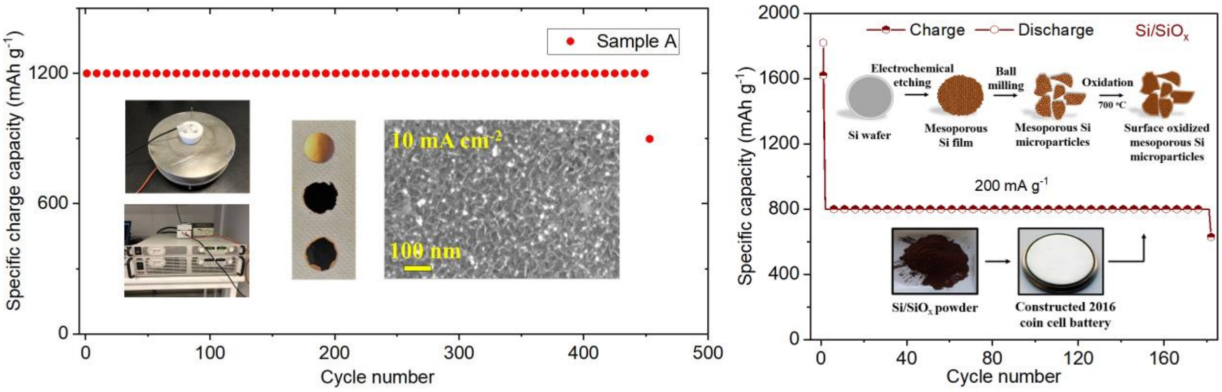
Xiuyun Zhao, Nathiya Kalidas, Manoj Muraleedharan Pillai, Vesa-Pekka Lehto

*Department of Applied Physics, University of Eastern Finland, FI-70210 Kuopio, Finland*

*xiuyun.zhao@uef.fi*

Silicon (Si) is the most promising anode material for achieving high energy density due to its high theoretical capacity (3579 mAh/g, 2194 Ah/L at room temperature) and safe electrochemical potential (0.45 V versus Li+/Li) [1]. However, the huge volume change (~280%) during cycling induces particle fracturing and unstable solid electrolyte interphase, resulting in electrical contact loss and eventually battery failure [2]. Nanostructuring is one of the effective strategies to overcome this issue. Some battery/material companies have claimed their commercial nano Si anodes, in which nanotechnology, such as manufacturing Si nanoparticles, Si nanowire, or amorphous Si, was adopted to reduce the electrode/battery swelling due to repeated expansion and contraction [3].

Mesoporous Si has a nanoscale pore structure, and the void space can give Si some room to expand during lithiation. It makes mesoporous Si could be another choice to realize the practical application of Si anodes. Nevertheless, mesoporous Si has additional characteristics compared to other nano Si materials, such as high specific surface area, highly active surface, and low bulk density, which can deteriorate the performance of Si electrodes. All these challenges need to be considered seriously. The Pharmaceutical Physics Group (PPG) at the University of Eastern Finland (UEF) is developing porous silicon materials with different etching methods and studying their application as lithium-ion anode materials. Self-standing mesoporous Si film and mesoporous Si power were developed respectively to study the effects of pore characteristics on Si anode's performance and how surface modification improves electrode performance. The results (Figure 1) show that reversible capacity and initial Coulombic efficiency of Si film electrodes are strongly related to porosity and surface area, and cycling performance is mainly affected by Si film thickness. Appropriate oxidation of mesoporous Si surface to get optimized surface/oxygen content is a promising way to enhance the electrochemical performance of mesoporous Si anodes essentially.



(a) (b)

***Figure 1.*** *Preparation and electrochemical performance of (a) self-standing mesoporous Si film electrode [4] and (b)surface modified mesoporous Si powder electrode in half cells.*

**References**
[1] M. N. Obrovac, V. L. Chevrier, *Chemical reviews*, **114** (2014) 11444-11502.
[2] M. N. Obrovac, L. J. Krause, *J. Electrochem. Soc.* **154**, (2007) 103-108.
[3] X. Zhao, V. -P. Lehto, *Nanotechnology*, **32** (2021) 042002.
[4] X. Zhao, N. Kalidas, V.-P. Lehto, *J. Power Sources*, **529** (2022) 231269.

# Developing Electrolyte Formulations with Zwitterionic Monomers for Better Lithium-Ion Batteries

Niguse Sahalie[1], Jonas Mindemark[1]

[1]*Department of Chemistry – Ångström Laboratory, Uppsala University, SE-751 21 Uppsala, Sweden*

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High-energy-density lithium-ion batteries (LiBs) have been the focus of extensive research and development efforts throughout the world owing to their role in portable electronic devices and electric vehicles. There are practical requirements for such next generation secondary LiBs to enable electric vehicles with increased range, long battery lifetimes and quick "refuelling". They include higher energy densities and fast charge−discharge rates, which hinge on new high-voltage, high-capacity, and high-power cathode materials and on stable electrolytes with higher ionic conductivities [1] [2].

Current LiBs are nearing their theoretical performance limits. Moreover, they employ expensive raw materials, such as cobalt, that compromise their green profile and limit the cost reduction needed for their widespread uptake. In this regard, several cathode materials have been developed without cobalt. The LNMO ($LiNi_{0.5}Mn_{1.5}O_4$) cathode is one of the best candidates due to its high voltage and high-power capability features. However, most of the electrolytes undergo oxidative decomposition at such high voltage, preventing their real application. Different electrolyte formulations have been reported to mitigate this issue. However, they end up either decreasing the conductivity of the electrolyte or forming a resistive passivation layer, sometimes even both.

Herein, in the BetterLiBs project, we have used novel polymerizable zwitterionic monomers (ZIMONS) as an additive to liquid electrolyte to improve the performance of LiBs. Zwitterionic monomers have equal numbers of positive (ammonium cation) and negatively-charged (sulfonate) functional groups, which can help to easily decouple the two charges of the electrolyte salt. As a result, the ionic conductivity of the electrolyte could be improved. The polymerizable features of ZIMONS are very important for forming a good passivation layer at the electrode/electrolyte interface, leading to stable cycling of the battery. The high thermal stability of ZIMONS is also in line with practical requirements.

The BetterLiBs project partly involves the synthesis, screening, and optimization of more than a dozen ZIMONS for their electrochemical performance. Performing cyclic voltammetry analysis in half cells, ZIMONS additives are found to improve the stability of LP57 electrolytes at both reduction and oxidation voltages. The cycling test with different ZIMONS additives also showed significant improvement in capacity and coulombic efficiency in the high voltage LNMO cathode configured with a $Li_4Ti_5O_{12}$ (LTO) anode as shown in figure 1. Other cells based on different cathode and anode materials are being studied.

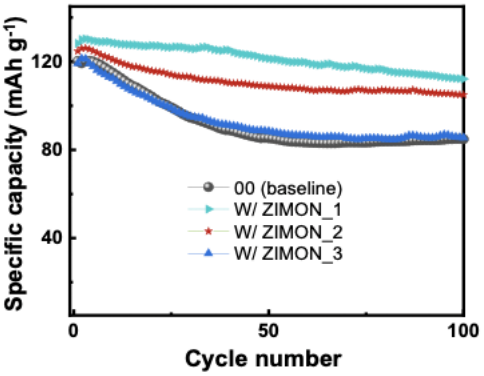

***Figure 1.** The performance of LNMO//LTO cell with and without different ZIMONS additives at 1% (w/w)*

## References

[1] B. Aktekin et al., J. Phys. Chem. C, 122(2018), 11234–11248
[2] S. Park et al., Nature Communications, 12 (2021), 12, 838–849

# 3D hollow spheres assembled by 2D nanomaterials for high-performance electrochemical energy storage

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The assembly structure and morphology of the nanomaterials have significant influence on their properties and the performance of the corresponding devices made by those materials. Creating 3D structures by self-assembling 2D nanomaterials could dramatically improve the overall performance of 2D materials for energy storage and conversion, especially where the high surface area and porous structure are needed. Template-assisted methods are the most popular way to fabricate 3D structure, and freeze-casting using ice as template was considered as an environmental-friendly and simple method. However, it is always challenging to achieve the desirable 3D structures due to the uncontrollable ice crystals growth and the limitation of 2D materials dispersion in the system. I will present an innovative method that we developed recently to self-assemble 2D materials for example graphene and MXene into 3D hollow spheres (**Fig. 1**). Different from the 3D porous network obtained from conventional freeze-drying method,[1] such innovative and simple method allow us to prepare uniform graphene/MXene hollow spheres with controllable size and spherical structure. A series of graphene hollow spheres with different size and 3D structures were prepared. Advanced in situ characterization method was used to understand the formation of the 3D hollow structures.[2] Using as electrode materials, the as prepared 3D hollow spheres exhibit superior electrochemical performance in terms of specific capacitance and rate capability thanks to the spherical structures. Our method is very simple and can be scale up for large scale synthesis with great potential for practical applications.

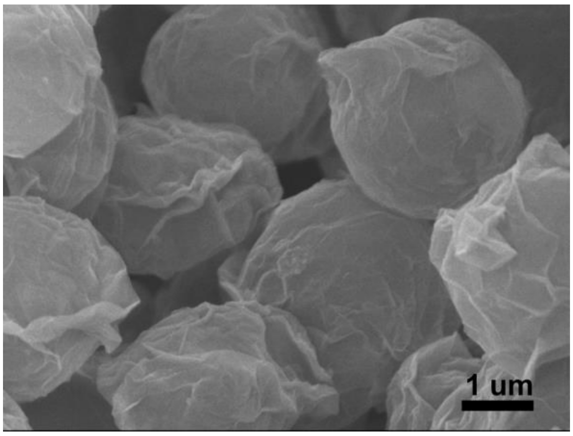

***Figure 1.*** *SEM image of 3D graphene hollow spheres.*

**References**

[1] J. Sun, M. A. Memon, W. Bai, L. Xiao, B. Zhang, Y. Jin, Y. Huang and J. Geng, Controllable Fabrication of Transparent Macroporous Graphene Thin Films and Versatile Applications as a Conducting Platform. *Adv. Funct. Mater.* 2015, 25, 4334–4343.

[2] J. Sun, M. Sadd, P. Edenborg, H. Grönbeck, P. H. Thiesen, Z. Xia, V. Quintano, R. Qiu, A. Matic and V. Palermo, Real-time imaging of Na+ reversible intercalation in “Janus” graphene stacks for battery applications, *Sci. Adv.*, 2021, 7, eabf0812.

# Surface Termination Control of 2D MXenes for Battery Applications

Frode Håskjold Fagerli[1], Zhaohui Wang[12], Tor Grande[1], Sverre M. Selbach[1], Nils Peter Wagner[12], Kjell Wiik[1]

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Due to their high electrical conductivity and ion intercalation capabilities, 2D MXenes have been thoroughly investigated as electrode materials for several different battery and supercapacitor chemistries (Li, Na, Al, Zn, Mg, etc.) since it was first discovered 11 years ago [1,2]. However, in order to control the ion intercalation properties of these MXenes, and thereby allow for optimized electrode performance, it is essential to control the intercalation environment governed by the MXenes surface terminations [3]. From the commonly used HF etching method, a mixture of O-, OH- and F-terminations are formed, and upon various post-etching treatments, the concentration of these terminations can be adjusted [4]. Here, we present gas hydrolysation as an effective method to remove most F-terminations from the MXene surfaces, to achieve improved rate performance of V2CTx and Ti3C2Tx electrodes in LiBs. Due to the many possible applications of MXenes, surface termination control is also of great interest beyond battery applications.

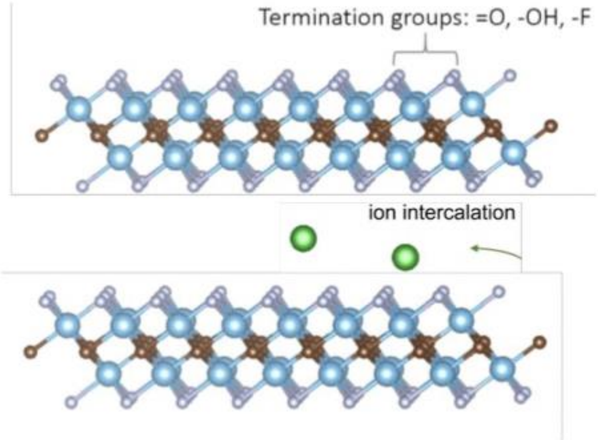

***Figure 1.*** *A general illustration of a 2D MXene and how the surface termination groups interact with the intercalating ions and thus determine the intercalation environment. The blue and brown atoms represent transition metal atoms and C/N atoms.*

**References**

[1] M. Greaves, S. Barg, M. A. Bissett, *Batteries & Supercaps,* **3** (2020) 214-235.
[2] H. Kaland, J. Hadler-Jacobsen, F. H. Fagerli, et al., *Sustainable Energy & Fuels*, **4** (2020) 2956-2966
[3] C. Eames, M. S. Islam, *Journal of the American Chemical Society* **136** (2014) 16270-16276
[4] H. Yu, Y. Wang, et al., *Small* **15** (2019) 1901503

# Ageing Mapping in Commercial Ni-rich Lithium Ion Cells for Electric Vehicles

Anastasiia Mikheenkova[1], Niladri Roy Chowdhury[2], Alexander Smith[3], Yonas Tesfamhret[1], Torbjörn Thiringer[2], Rakel Wreland Lindström[3], Erik Berg[1], Matthew Lacey[4], Maria Hahlin[1]

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[2] *Electrical Machines and Power Electronics – Chalmers University of Technology, SE-412 96 Gothenburg, Sweden*
[3] *Division of applied electrochemistry - KTH Royal Institute of Technology, SE-100 44 Stockholm, Sweden*
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*anastasiia.mikheenkova@kemi.uu.se*

High cost and supply constraints of active materials for lithium-ion batteries (LIBs) motivates continued effort in prolonging usable lifetime. A true understanding of degradation processes behind battery ageing is, therefore, essential. An additional crucial challenge is to mimic relevant ageing as well as to distinguish ageing on different cell elements. If one focuses on a positive electrode LIB ageing contribution, it is common to differentiate ageing effect depending on the consequence. These consequences can be separated into loss of active material (*i.e.* transition metal dissolution, structural deterioration), loss of lithium ion inventory (*i.e.* electrolyte decomposition), and loss of conductivity (*i.e.* current collector corrosion, particle cracking) [1][2]. In the current work we investigate state of charge (SoC) window (0-50%, 50-100%, 0-100%) and temperature (30 and 45 °C) dependent degradation in commercial 2170 cylindrical cells extracted form Tesla Model 3 battery pack. The study was conducted on extracted electrodes with a particular focus on the degradation of the positive electrode. The results have shown that positive and negative electrode ageing is dependent on temperature and SoC range High cost and supply constraints of active materials for lithium-ion batteries (LIBs) motivates continued effort in prolonging usable lifetime: the positive electrode experiences stronger degradation at high temperature and high SoC range resulting in loss of active material and higher resistance (**Fig. 1**). The negative electrode ageing was found to be tightly connected to $SiO_x$ particles degradation. Ageing within the cell geometry is not homogenous and varies at both the negative and positive electrodes [3].

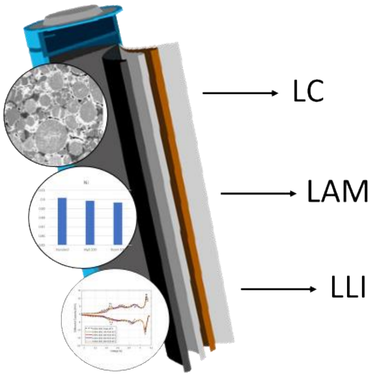

***Figure 1.*** *Degradation sources of lithium ion cell.*

**References**
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# How about Glyoxals? Evaluating alternative Electrolytes for Si/Graphite Composite Electrodes in Li-ion batteries

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Li-ion batteries are a key enabler in the transition from fossil fuels to renewable energy. To further increase their market shares beyond consumer electronics in electric transportation and grid storage, higher energy and power densities are required. One way to achieve higher energy densities is to add silicon to the graphite negative electrodes. In these composite electrodes, the high volume changes of silicon during cycling are buffered to some extent but still put significant strain on the electrode and SEI stability. We therefore investigate alternative, carbonate-free electrolyte compositions and evaluate their impact on the electrode performance. These electrolytes were based on the glyoxylic acetals TMG (1,1,2,2-tetramethoxyethane) and TEG (1,1,2,2-tetraethoxyethane) [1], selected because of their excellent thermal stability.

While TMG performed worse than cells cycled in carbonate-based reference electrolytes, improved performance could be achieved when using TEG. The latter exhibited a much more stable capacity profile with overall higher capacities. The poor performance of TMG could be related to a dramatic drop in both graphite and silicon activity after already 10 cycles. TEG, on the other hand, showed higher retention of silicon activity, with graphite activity remaining unchanged from the 1st to the 100th cycle. XPS showed that TMG formed an SEI with slightly higher carbonate content and earlier and stronger LiTFSI salt decomposition, while -C-O and -$CO_2Li$ concentrations were similar when compared to the TEG SEI. Analysis of the electrode morphology revealed TEG to form a smoother and continuously connected SEI. This surface layer most probably has a higher ability to accommodate the silicon volume changes upon cycling, rendering TEG a very promising electrolyte candidate for realizing silicon-containing anodes with high capacity and high stability. Even without SEI stabilizing electrolyte additive or ionic conductivity enhancing co-solvent, TEG showed enhanced electrochemical performance with Si/Gr anodes compared to a standard electrolyte based on EC:DMC with $LiPF_6$.

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# How smart application of rather simple electrochemical methodology can help to better understand ion transfer and metal anode batteries?

**Prof. Dr. Martin Winter**

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Martin Winter is a true friend of the Nordic battery community. He has been in the field of electrochemical energy storage and conversion for over 30 years, with a focus on development of new materials, components, and designs for lithium-ion and lithium metal cells and beyond.

# From Supercapacitors to Superbatteries – high power energy storage and its applications

## Dr. Sebastian Pohlmann

Sebastian Pohlmann is VP Automotive & Business Development at Skeleton Technologies. After completing his PhD on Supercapacitors in 2014, he joined the company and led the cell and material research and development function for nearly 3 years. Today he focusses on bringing Skeleton's novel products into new applications, with a focus on automotive and on-road applications. Founded in Estonia in 2009, Skeleton Technologies manufactures high power energy storage cells, modules and systems based on Supercapacitors.

# Anchoring Polysulfides and improving the specific capacity of the Lithium-Sulphur batteries.

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Lithium-Sulfur (Li-S) batteries are one of the most promising next-generation energy storage systems. Sulfur is both environmentally friendly and naturally abundant in the earth's crust, and it offers a high theoretical capacity of 1675 mAh $g^{-1}$, which is about an order of magnitude higher than that of the transition-metal oxide cathodes [1]. However, the current Li-S batteries are still plagued by numerous challenges [2]. The sulfur and its final discharge product ($Li_2S$) have an insulating nature that leads to a low material utilization during the electrochemical reactions. Moreover, the high-volume expansion during lithium intercalation and deintercalation process, that pulverizes the cathode and detaches the active mass from the conductive substrates, leads to a decrease in electrical conductivity, and to capacity fading. Another big challenge is the so-called "shuttle-effect" which is caused by the dissolution of the intermediate reaction products (lithium polysulfides) into the electrolyte. The lithium polysulfides can deposit on the metallic lithium electrode, which causes reduction in the coulombic efficiency, and capacity fade [3].

The alleviation of the mentioned problems can be achieved via cathode functionalization, separator modification, and electrolyte tailoring. Moreover, the design and engineering of sulfur host materials have been proven to be a successful approach to improve the capacity and cycling stability of sulfur cathodes [4]. Researchers at Fine Particle and Aerosol Technology Laboratory (FINE) are working on synthesis of the different nanomaterials and carbon-based materials, and composites, including nano $Ti_2O$, graphene, carbon nanoflowers, and carbon nanotubes. These materials are considered promising for utilization as sulfur hosts, as well as for the creation of an additional interlayer between cathode and separator, leading to improved capacity and cycle stability of Li-S batteries [4]. The presented work is an overview of activities related to the utilization, in Li-S batteries, of the materials produced at FINE. The synthesis methods, and the characterizations of the materials, are briefly explained, with a focus on the advantages of produced materials in the Li-S batteries.

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# Screening of biomass-derived hard carbon anodes for Na-ion batteries

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Nowadays, lithium-ion batteries (LIB) are the dominant battery technology and are produced at an increasingly large scale to cope with the growing electrification of the transport sector. This results in large cost reduction and new opportunities for their implementation in stationary energy storage at the grid and household levels. In the next few decades, it is expected that the number of electric vehicles will increase by two to three orders of magnitude and stationary storage may reach up to 1300 GWh. Although LIBs are excellent candidates for electromobility, concerns are rising regarding the long-term sustainability and cost of critical raw materials (e.g. cobalt, nickel, lithium, and copper) used in LIBs. Thus, next-generation batteries, such as sodium-ion batters (SIBs), are considered sustainable, and affordable energy storage solution alternatives [1].

Even though SIBs share a lot of similarities with LIBs, the electrochemical intercalation of $Na^+$ ions into graphite is limited. Instead, hard carbon (HC) is an alternative carbonaceous anode material that has been proven to intercalate $Na^+$ ions at low potentials [2]. Biomass is the most popular type of hard carbon precursor due to its low cost and abundant natural resources compared to petroleum coke or formaldehyde resins. However, HC anodes can suffer from poor rate performance, cycle life, and low initial coulombic efficiency due to limited diffusion kinetics within the particles and undesirable side reactions with the electrolyte leading to unstable SEI layers. This is attributed to the high open surface area and unoptimised internal microstructure of the HC particles [3].

In this work, we explore the effect of different Norwegian abundant biomass sources namely: potato starch, lignosulfonate, Kraft lignin, and microalgae, on the microstructural, chemical, and morphological properties of the resultant hard carbon. The electrochemical performance of these HCs is tested and compared via galvanostatic electrochemical characterisation and EIS while their morphologies and microstructures are examined by SEM, HRTEM, XRD, and SAXS, respectively.

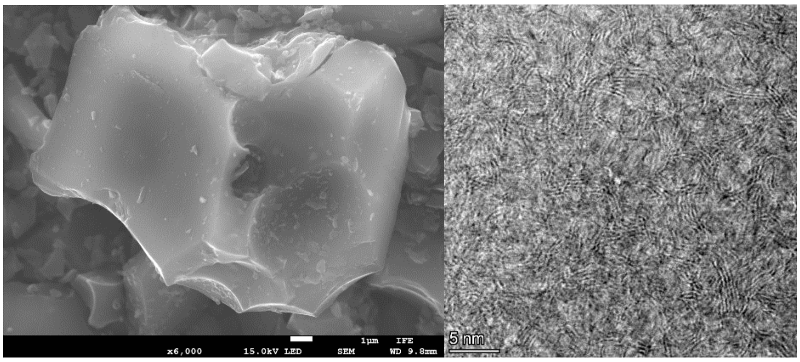

Figure1. SEM and TEM micrographs of lignosulfonate-derived hard carbon

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# Tracing the (de)sodiation of $Bi_2MoO_6$ through good and bad times with *operando* XRD

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Anode materials combining conversion and alloying mechanisms (CAMs) are promising for Na-ion batteries, but their complex cycling mechanisms are challenging to study[1]. Understanding the (de)sodiation mechanism is crucial and, in several cases, requires advanced synchrotron characterization. *Operando* synchrotron studies are generally limited to one or two sodiation cycles, which are not fully comprehensive for CAMs[2]. Therefore, herein, we studied the sodiation and desodiation of $Bi_2MoO_6$-based anodes with laboratory-based *operando* X-ray diffraction exceeding more than 30 cycles to have a complete overview of our CAM's mechanism (Figure 1)[3]. This revealed important aspects of the cycling and degradation mechanisms in the material. During the first sodiation, an irreversible conversion of $Bi_2MoO_6$ occurs, creating Bi nanoparticles embedded in an amorphous Na-Mo-O matrix. The Bi particles then reversibly alloy with Na forming cubic $Na_3Bi$ leading to a specific capacity close to 300 mAh $g^{-1}$ for the 10 first cycles. This is followed by a rapid capacity decay where the sodiation of Bi becomes irreversible leaving several inactive $Na_3Bi$ particles. To the best of our knowledge, this is due to the observed crystal growth of the Bi particles accompanied by structural changes in the insulating Na-Mo-O leading to poor conductivity in the electrodes. The poor electronic conductivity of the matrix deactivates the $Na_xBi$ particles and prevents the formation of the solid electrolyte interface layer as shown by post-mortem scanning electron microscopy studies.

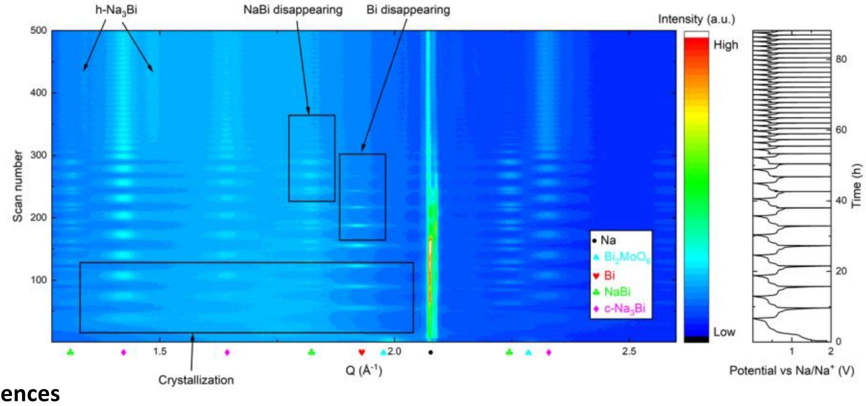

Figure 1: *Operando* XRD of $Bi_2MoO_6$

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## Sustainability: Batteries & Gender Equity

### Dr. Adriana Navarro-Suarez

Adriana Navarro-Suarez is a R&D manager at Morrow Batteries, leading the development of electrolytes for sustainable and cost-effective batteries. She holds a Ph.D. in Materials Science and Technology and a M.Sc. in Materials for Energy Storage and Conversion. Former MSCA fellow at Imperial College London, postdoctoral researcher at Chalmers University of Technology, and visiting graduate student at Drexel University.

# Some critical steps in LIB fabrication and their influence on performance.

## Prof. Em. Josh Thomas

Josh Thomas was Professor of Solid State Electrochemistry at Uppsala University until his retirement in 2009. He founded The Ångström Advanced Battery Centre in 2000 and LiFeSiZE AB (www.lifesize.se) in 2006, as a spin-off from on-going basic battery research at Uppsala University. The role of the company today is to produce full-scale proof-of-concept Li- and Na-ion cell prototypes for subsequent commercial exploitation.

# The Use of Isostatic Pressing in Research and Development of Scalable Production Technology for All Solid State Batteries

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Next generation all solid-state batteries (ASSB) offer increased power and energy density compared to conventional Li-ion batteries (LIBs) with enhanced safety. ASSBs use a dense, bi-functional solid material that acts as both the electrolyte and the separator. Although several materials have been identified for application as solid-state electrolyte, suitable production routes need to be developed for realization of all solid-state batteries. Realization of excellent mechanical contacts and zero porosity give rise to the need for densification of battery components and cells [1][2].

Global industry actors are now investigating the use of extreme pressure to ensure densification of battery components and cells. Isostatic pressing involves the use of a pressure medium to compact components under immense isostatic pressure, a technology that is well known in other industries since the 1960s. While hot isostatic pressing uses a gas pressure medium, Cold and Warm Isostatic presses (CIP, WIP) use liquid media. As the name suggests, CIP operates at room temperature, however WIP can operate at higher temperatures which aids densification. This technology is of great interest to densify batteries, to reduce resistivity and improve battery properties.

Quintus Technologies is the leading producer of isostatic pressing equipment and are engaged in the development and production of HIP, CIP and WIP equipment for future battery systems. This presentation will focus on WIP equipment developed for battery research and scalable production using wire-wound pressure vessels and state of the art technology for temperature uniformity, which has been based on input from major global battery producers and research institutes.

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# Materials and Processing Methods for Stable Lithium-Metal Batteries

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Solid-state Lithium-Metal (Li°) Batteries (LMBs) are foreseen as enablers for *safer-by-design* and higher energy density post-Lithium-ion electrochemical energy storage 2.0 solutions. They are considered as part of the solutions to address the United Nation Sustainable Development Goal N°7 and to reach a carbon neutral European continent by 2050. As LMB-based materials are (at least in part) different from the ones currently used in *state-of-the art* Li-ion Batteries (LiBs), this is creating a technology push to adjust processing methods for manufacturing Generation 4b batteries. Shifting paradigm from LiBs to LMBs is also calling for major improvements of their lifetime as their interfaces are (i) prone to degradation or (ii) can suffer from dendrite growth or contact loss.

Soft-matter-based organic electrolytes offer here an innovative and scalable materials platform to manufacture LMBs and ways to improve their lifetimes through varying their chemical structures and processing. We will present results implementing this strategy from the ongoing Battery $2030^+$ HIDDEN project [1], where Thermotropic Ionic Liquid Crystals (TILCs), the 'materials marriage' of Thermotropic Liquid Crystals (TLCs) with ionically conducting materials, have been developed as a new generation of self-healing electrolytes (see **Figure 1**) aiming specifically at (i) mitigating Li° dendrite growth and (ii) increasing the lifetime of LMBs.

TLCs are fluid-like but dynamically ordered self-assemblies under a certain temperature range. If used as electrolytes, TLCs need to be engineered to encode ionic transport features for generating TILCs specifically designed to assist controlled $Li^+$ transport and to ensure smooth Li deposition onto a Li° electrode, preventing dendrite formation. We will discuss how HIDDEN has leveraged this concept through a series of TILCs enabling a scalable process to coat the electrolytes onto/within the electrodes and to generate data for modelling, thereby implementing the MAP and BIG-MAP concepts at the heart of the Battery $2030^+$ initiative. In closing, we will also disclose a few highlights from two other projects where we are developing materials and processing methods with polymeric electrolytes for LMBs: A national Finnish project NextGenBat [2] and the just-started Horizon Europe project SOLiD.

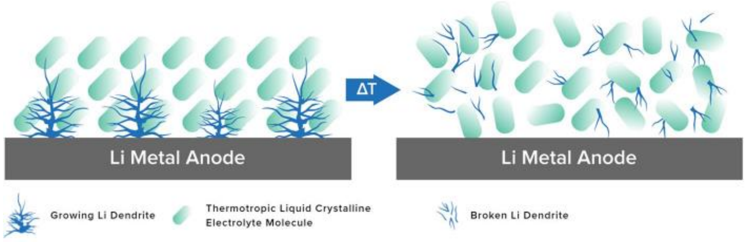

***Figure 1.*** *Schematic of the self-healing method based on a liquid crystalline electrolyte.*

**Acknowledgments:** This research is funded by the European Union's Horizon Europe (SOLiD project) and Horizon 2020 research and innovation programmes (HIDDEN project: Grant agreement N°957202), and by Business Finland's Smart Mobility and Batteries from Finland program.

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# Aging mechanisms of NMC/graphite Li-ion batteries

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Lithium-ion (Li-ion) batteries being the most widely applied technology for energy storage suffer from degradation both over time (calendar aging) and with use (cycling aging), and is related to the battery chemistry, environmental conditions, and use patterns. Degradation investigation has been identified as one of the green principles for responsible battery management, as extending battery lifetime decreases costs and environmental impact. Also, understanding the operating principles and degradation mechanisms of Li-ion batteries elucidate behaviors that can extend battery life.

In this research, we aimed to investigate the electrode degradation processes related to the Li-ion batteries' state of health and environmental conditions (temperature of cycling). For this purpose, Li-ion full batteries with graphite/Si negative and $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NMC) positive electrodes were cycled with recording of electrochemical impedance spectroscopy after each 200 cycles. After end-of-life, spent batteries were opened, and electrodes were investigated to compare difference in structure, morphology and electrochemical behavior. Complexity of the Li-ion batteries makes it difficult to investigate independently all the aging effects as well as to correlate aging processes with battery performance. One of the most important is aging of positive electrodes. Active lithiated NMC layered oxide is cracked during charge and discharge process, suffers from rock salt formation, transition metal dissolution, cathode electrolyte interphase growth and cation mixing. Graphite/Si negative electrode material is also affected by crack formation. Li dendrite growth, surface electrolyte interphase decomposition, transition metal contamination, Si dissolution are other adverse effects lead to capacity fade of full battery. Supporting components like current collectors, conductive electrode additives, electrolyte, and separator can also be damaged during cycling. Degradation of components leads to increasing of the contact resistance, $Li^+$ diffusion limitations, reducing of active materials participating in energy storage, and, as a result, capacity fade destroys the battery. Degradation processes can be detected by capacity fade and impedance growth of the full battery, but it is not clear how to correlate individual aging processes with the full battery behavior. In our work we have investigated performance of the full batteries, changes of electrode structures and degradation mechanisms occurring during cycling as well as the correlations between these.

Acknowledgement

This project (17IND10-LiBforSecUse) has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme.

# The Role of Crystallization in Closing the Loop for Battery Production and Recycling

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Several processes have been developed for the recycling of batteries, in particular lithium-ion batteries. Crystallization and precipitation are important unit operations in many of these processes. Precipitation can be applied to remove impurities such as iron while evaporative crystallization is often applied to recover nickel, manganese, and cobalt as sulphate hydrate salts [1, 2]. These salts can be used in the production of cathode precursor materials provided that the purity is sufficiently high. In battery material precursor production crystallization also plays an important role for the quality of the active cathode materials [2].

This talk will highlight challenges in established crystallization processes in battery recycling and production, and present and discuss novel approaches. Eutectic freeze crystallization can be an energy saving alternative to evaporative crystallization of nickel, manganese, and cobalt sulphate hydrates [3]. Furthermore, antisolvent crystallization can be used for production of nickel sulphate hydrate of high quality [4]. Antisolvent crystallization can also be applied to obtain nickel, manganese, and cobalt carboxylate salts of relevance in the production of new cathode materials [5, 6].

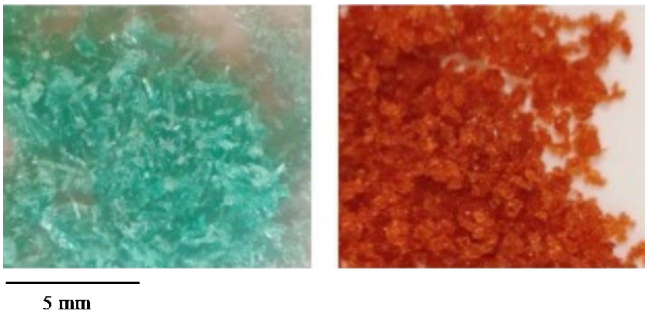

***Figure 1.*** *Crystals of $NiSO_4{\cdot}7H_2O$ (left) and $CoSO_4{\cdot}7H_2O$ (right) obtained by EFC.*

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# Optimized recycling of critical metals from EV Li-ion batteries via combined metallurgy

Martina Petranikova[1], Pol Llorach Naharro[2], Nathália Vieceli[1], Burcak Ebin[1]

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In the upcoming years, increasing e-mobility will demand material recirculation via optimized sustainable recycling. Due to the presence of polymers and organic solvents (binder, electrolyte, separator, casings, etc.), the combination of thermal pre-treatment followed by hydrometallurgical processing will be the most common method for an effective material recovery. Despite the already existing industrial use of such treatment, there is still a vast knowledge gap on how selected parameters affect subsequent hydro-chemical processing. Thus, detailed investigation and optimization of the parameters have been performed in this research. Thermal pre-treatment was tested using oxidative and reductive conditions and the effect of process temperature and process time were investigated. Qualitative and quantitative characterizations of the samples after treatment were performed together with the analysis of the by-products in order to provide of a full picture of such treatment impact on the material and environment. Thermal processing was then followed by leaching with inorganic acid at ambient temperature to determine the leachability of critical metals such as cobalt, manganese, nickel and lithium. Moreover, the effect of treatment on the leachability of the main impurities (copper and aluminum) was determined, which is a significant contribution to the field. Since the presence of carbon affects the degree of active material reduction, it's content after each thermal treatment was followed to predict if the addition of reductive agent is needed. The pyrolysis for thirty minutes was selected as the optimal processing time of the black mass followed by leaching at ambient temperature. Under these conditions, a full recovery of all metals was reached after two minutes for lithium, five minutes for manganese and ten minutes for both cobalt and nickel. The outcome of the research will provide a higher material recovery from EV batteries with lower demand on the energy and reagent consumptions and secondary waste generation.

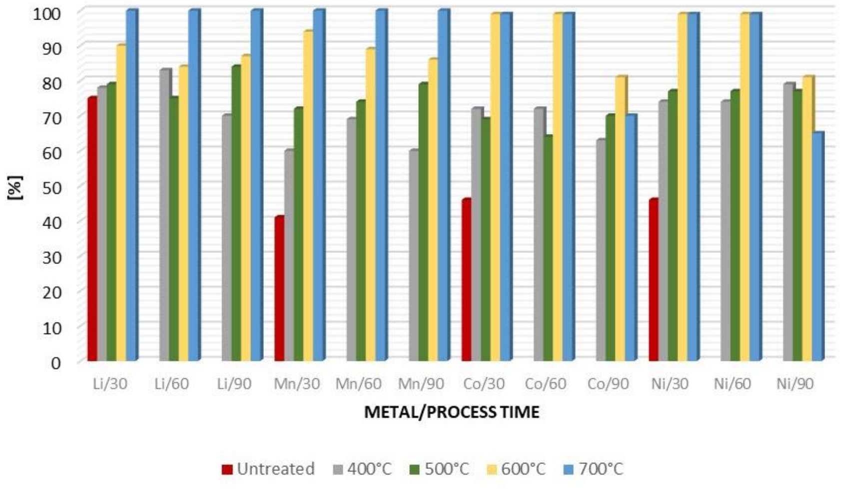

***Figure 1.*** *Recovery efficiency of lithium, manganese, cobalt and nickel from the waste after pyrolysis at 30, 60 and 90 minutes and different temperature via leaching with two molar sulfuric acid at ambient temperature.*

# A model platform for life cycle assessment of lithium-ion battery production and use

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Life cycle assessment (LCA) studies of lithium-ion batteries (LIB) often show a large variation in results. Explanations to this diversity can be found in different modelling approaches and shifting quality of data, e.g. a lack of primary data and use of outdated data, but also a large variety in cell formats, chemistries, and designs analysed [1], [2]. Meanwhile, LCA of LIBs is becoming increasingly important as a tool to guide technology selections towards increased sustainability. To bridge existing data gaps, a multidisciplinary cooperation has been initiated aiming to develop a model platform for conducting LCA of LIBs in vehicle applications, covering both production and the use-phase. This model platform (***Fig. 1***) will comprise a cell design computation model, a cell performance model, a vehicle model, and a production LCA inventory model, which provide input the overall LCA.

In this contribution, the cell design computation model and cell performance model will be presented. The cell design computation model provides input for the production LCA inventory and cell performance models. For a given capacity, the model calculates required masses of constituent materials, electrode areas and thicknesses by assuming common cell design aspects and the volume for the selected cell format. Input data is based on publications and teardown analyses of commercial cells. The cell performance model utilises the physics-based Doyle-Fuller-Newman model implemented in COMSOL Multiphysics. From this, the cell capacity, potential, and impedance can be simulated for any cell design. Both these data models then supply data further to the production LCA inventory model and the vehicle model, in turn enabling the overall LCA.

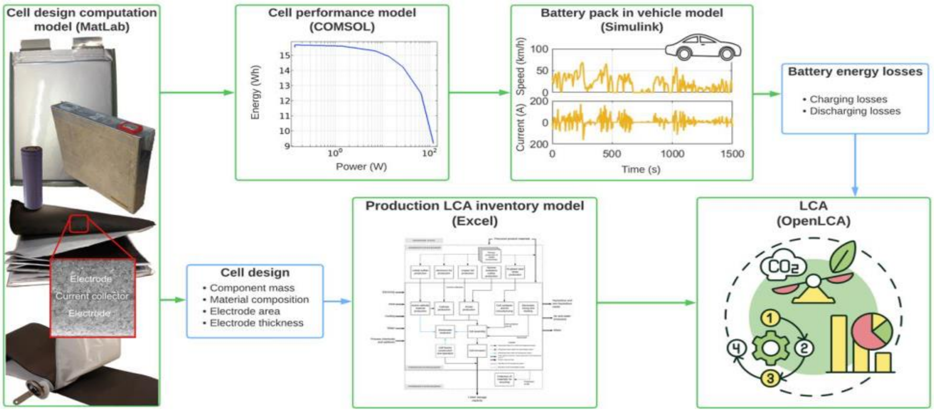

**Fig. 1** Overview of the model platform, from cell design parameters to LCA calculations.

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# Coprecipitation of high-nickel mixed metal hydroxide precursors for LIBs: Improved sustainability in the battery value chain

Ulla Lassi[1,2], Parisa Mehdipour[1,2], Petteri Laine[1,2] ,Yan Lin[1], Tao Hu[1], Juho Valikangas[1,2], Eeki Airola[1,2], Immo Kervinen[1,2], Palanivel Molaiyan[1], Toni Kauppinen[1,2], Pekka Tynjälä[1,2]

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Lithium-ion battery (LIB) precursors, such as NMC811 ($Ni_{0.8}Mn_{0.1}Co_{0.1}(OH)_2$) are typically produced by coprecipitation from metal sulphate solutions. This enables large-scale processing using continuous flow stirred tank reactors (CSTR), and careful control of particle size and morphology, which are critical for LIB cell performance. However, key challenges are the use of cobalt as critical raw material, and the formation of $Na_2SO_4$ (80-120 g/L) without any techno-economic feasible solution [1]. In our recent study [2], we prepared LNO as Co-free cathode active materials by coprecipitation.

During the coprecipitation of metal hydroxide precursors, the following reactions occur:

Metal−ammonia complex formation $Me^{2+} + nNH_3 \rightarrow [Me(NH_3)n]^{2+}$ (1)

Metal−hydroxide precipitation reaction $[Me(NH_3)n]^{2+} + 2OH^{-} \rightarrow Me(OH)_2 + nNH_3$ (2)

The formation of transition metal (TM) hydroxides precursors from a metal sulfate solution involves several physicochemical phenomena. Due to their high solubility product, TM sulfates readily dissolve in water. If the pH of the solution is increased by adding NaOH, TM hydroxides are precipitated and sodium sulphate is formed as secondary product. In the case of impure solutions, impurity phases may form in the presence of TM and group II/III metals. [3]

In this study, we replaced NaOH as precipitating chemical with KOH or $LiOH.nH_20$ (for recycled Li-containing metal sulphate solution). Coprecipitation was done under the molar ratios, pH, temperature and redox conditions, where the solubilities of sulphates were high. This led to the formation high purity potassium and lithium sulphates, respectively, which can be used as fertilizers and lithium salts for batteries. Finally, lithium was successfully recovered as lithium carbonate by $CO_2$ precipitation from the residue of battery recycling solution (see Fig 1).

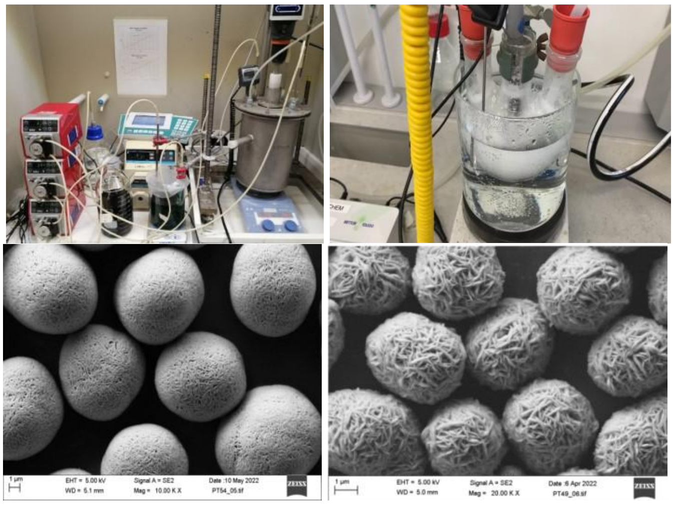

***Figure 1.*** *a) CSTR reactor for coprecipitation, b) reactor for lithium recovery as carbonate, $Ni(OH)_2$ precursor precipitated from pure (c) and impure recycling solution (d).*

**References**

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# Multi-scale models of energy storage materials: Always a trade-off between accuracy and speed?

**Dr Céline Merlet**

*CNRS/CIRIMAT, Université Paul Sabatier, Toulouse, France*

Dr. Céline Merlet is an award-winning researcher, including the Bronze medal of CNRS, in the field of advanced modelling at the molecular level, applied both to supercapacitors and to batteries. Her work is more than often done in conjunction with experimental efforts and has had a special focus on various kinds of carbon structures. She received the special prize Joseph Fourier 2021 as a member of the team developing the Metalwalls software.

# Electrochemical-Thermal Model-Based Fast Charging for Lithium-Ion Batteries: Nonlinear Inversion-Based Output Tracking Control

Yang Li, Torsten Wik, Yicun, Huang, Changfu Zou

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We propose a novel fast-charging control algorithm for lithium-ion (Li-ion) batteries that leverages a high-dimensional electrochemical-thermal pseudo-two-dimensional (P2D) model. The specific control objective is to find the highest battery current while various operating constraints are fulfilled. Conventionally, computationally demanding optimization or simple output feedback control is needed to solve this constrained control problem when an electrochemical-thermal model is used, leading to practical difficulties in achieving low-cost real-time implementation. Instead, this paper provides an optimization-free solution to Li-ion battery fast charging by converting the constrained optimal control problem into an output tracking problem with multiple tracking references, and the required control input -- the charging current, is derived by inverting the high-dimensional nonlinear battery P2D model. As a result, an inversion-based fast control algorithm is obtained for reliable evaluation of Li-ion battery performance under wide constrained control regimes. Results from comparative studies show that the proposed controller can achieve comparable performance to nonlinear and linear model predictive control at much lower computational costs with minimal effort in parameter turning.

# Accelerating battery innovation through FAIR model-based design

### Dr. Simon Clark

Simon Clark is a Research Scientist at SINTEF Industry focused on developing open-source digital tools to accelerate the design of batteries. He earned his Dr. rer. nat. degree in Computational Electrochemistry from Ulm University in cooperation with the German Aerospace Center (DLR) and Helmholtz Institute Ulm (HIU). He currently coordinates the EU H2020 project HYDRA pursuing generation 3b Li-ion batteries for electric mobility applications.

# Autonomous Workflows for an Accelerated Design of Battery Electrodes

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The development of automated computational tools is required to accelerate the discovery of new functional materials, to speed up the transition to a sustainable future. Here, I address this topic by designing new battery electrodes for different intercalation battery chemistries. These workflows are implemented in the framework of Density Functional Theory, using MyQueue and the Atomistic Simulation Environment (ASE). In the first part, I describe a fully autonomous workflow, which identifies materials to be used as intercalation electrodes in batteries, based on thermodynamic and kinetic descriptors like adsorption energies and diffusion barriers [1]. A substantial acceleration for the calculations of the kinetic properties has been obtained due to a recent implementation of the Nudged Elastic Bands (NEB) method, which takes into consideration the symmetries of the system to reduce the number of images to calculate. Moreover, we have established a surrogate model to identify the transition states, which can further reduce the computational cost to at least one order of magnitude [2, 3]. We have applied this workflow to discover new cathode materials for Mg batteries as well as solid state electrolytes for Li, Na, and Mg all-solid-state batteries [1, 3]. In the second part of my talk, I discuss how nanostructured materials can positively impact the Li-ion battery solid/electrolyte interface, to adjust the change in volume during charge/discharge in Si-anodes [4].

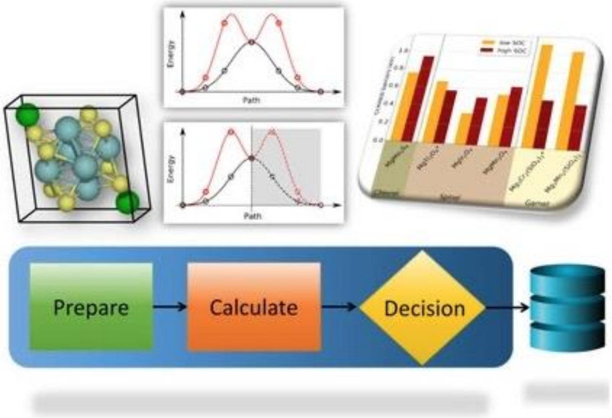

***Figure 1.*** *Schema of the workflow: the automated calculations include the thermodynamic and mechanical stability, Open Circuit Voltages as well as kinetic barriers obtained through the Nudged Elastic Band method. The workflow is chemistry neutral and is able to discover interesting candidates for insertion cathode materials as well as solid state electrolytes without the need of user intervention.*

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# $Li^+$ transport phenomena in a composite solid-state electrolyte

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In the context of all solid-state lithium batteries, we focus on lithium ion transport phenomena at the solid-solid interfaces. Using Molecular Dynamics techniques, we study an interface composed of $Li^+$ conducting ceramic (LLZO) and polymer (PEO or PTMC) materials. Interestingly, although a lot of attention has been dedicated to study lithium-ion transport phenomena in such composite solid-state electrolytes, there are plenty contradictory findings and hypothesis concerning $Li^+$ transport mechanisms in literature. For example, both an increase [1] and a decrease [2] in ionic conductivity has been observed when inserting ionic conductive ceramics to a polymer electrolyte matrix. These phenomena are also related to the type of composite components and the overall composition. Such controversies highlight that some fundamental questions remain unanswered. What is the ion-conducting phase? When is it more beneficial to use a composite electrolyte in comparison to a single-phase system? In this presentation, our strategy to study these complex but fascinating solid-solid interfaces will be discussed. A sensitivity analysis of different relevant simulation parameters provides us with a good base for further exploration of such complex solid-solid interfaces, and the general approach to this sensitivity study is discussed in detail. In particular, the possible impact of some simulation parameters on the outcome of a simulation will be explained together with preliminary results on ionic transport behavior and structure-dynamic properties. Considering that atomistic studies concerning an interface of a $Li^+$ conducting ceramic and a polymer are scarce, the work will hopefully spark more in silico activities to enhance the perspectives on $Li^+$ transport phenomena in composite solid-state materials.

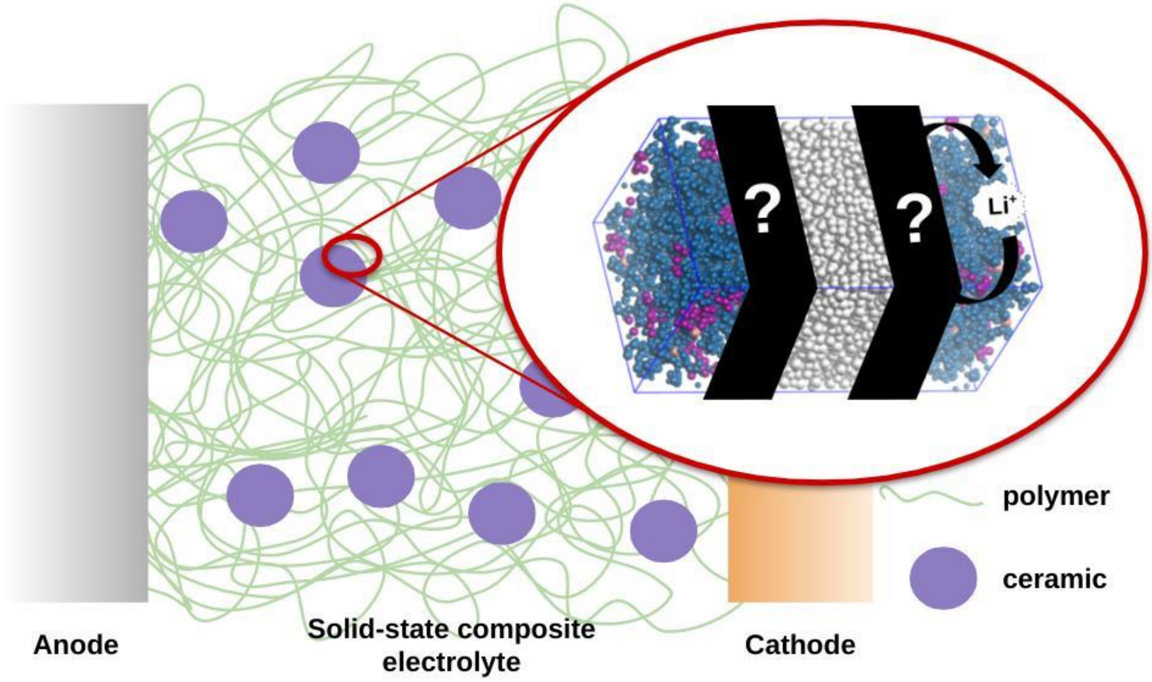

***Figure 1.*** *Schematic representation of an all solid-state battery. The ceramic-polymer interface studied at the atomistic level in this work is enlarged.*

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# Charge Transport Properties in Concentrated Polymer Electrolytes by Molecular Dynamics Simulations

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Understanding charge transport properties in electrolytes is essential for the development of improved electrochemical systems, e.g., lithium-ion batteries with better charge performance and higher energy density [1,2]. A molecular understanding of transport mechanisms is necessary to understand how transport in solid and liquid electrolytes takes place. Molecular Dynamics (MD) simulations allows us to compute transport properties while providing a view of the molecular and ionic motions and correlations that determine them.

In this work, we have studied charge transport properties in poly(ethylene oxide)-lithium bis(trifluoromethanesulfonyl)imide (PEO-LiTFSI) polymer electrolytes from dilute to super-saturated salt concentrations for two polymer chain lengths using MD simulations. We have computed ionic conductivity and transport numbers from equilibrium MD simulations using the Nernst-Einstein (NE) approximation and directly from Onsager coefficients. The NE approximation is for dilute or ideal systems, while the Onsager coefficients take molecular and ionic correlations into account and hence is more suitable for high concentration systems. We observe significant differences between the two methods and discuss these using the self-diffusion coefficients and Onsager coefficients behind the NE approximation and Onsager values, respectively. Surprisingly, the correlations between cations and anions contribute to an increased ionic conductivity, causing superionicity. The average static- and dynamic properties of the coordination environment of Li-ions are analyzed to obtain a deeper understanding of the transport mechanisms in these systems.

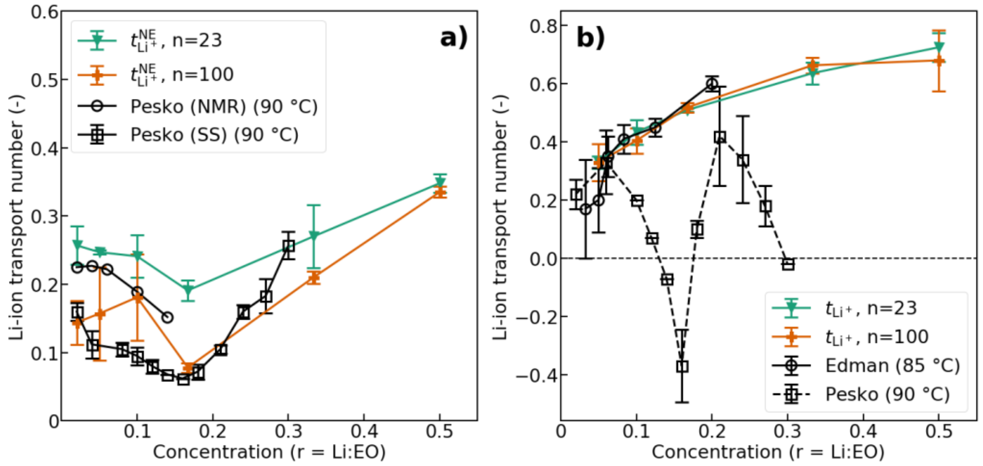

**Figure 1:** *Li-ion transport numbers computed using (a) NE and (b) Onsager methods. The computed values are compared to experimental data by Pesko et al. [3] and Edman et al. [4]*

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| P50 | **Ali Tuna**<br>(University of Turku, Finland) | Electrochemical and Stability Studies of Ferrocene-1,1′-disulfonate Dianion in Aqueous Redox Flow Battery Systems |
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# POSTER ABSTRACTS

# Molecular Design of Film-Forming Electrolyte Additives for Lithium-Ion Batteries and their Evaluation on Cell Level

L.Quach[1], J.-P. Schmiegel,[2] V. Göldner[3], E. Adhitama[2], R. Nölle[2], J. Henschel[2], M. Winter[2],
U. Karst[3*], T. Placke[2*], F. Glorius[1*]

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Owing to their high energy densities and high operating voltages, lithium-ion batteries have dominated the market to date. There remains room for to enable large scale applications. A known reason for poor cycling performance of LIBs is the loss of active lithium on the anode surface because of parasitic side reactions partly owing to the formation of the solid electrolyte interphase (SEI). This SEI can be improved by the addition of suitable additives to hasten the formation of a thin, stable, but flexible SEI leading to longer battery lifetimes, lower internal resistances and higher power capabilities.

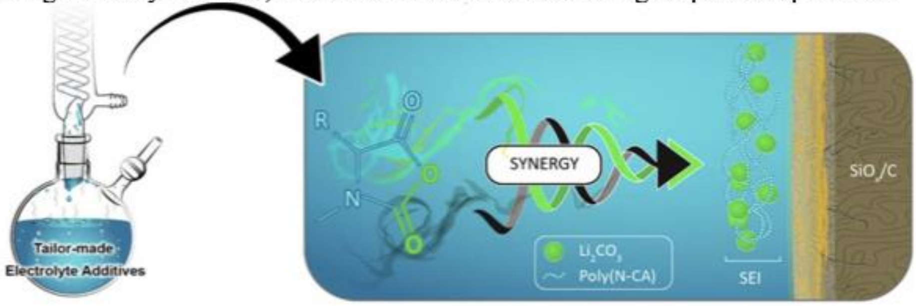

***Figure 1.*** *Tailor-made substrates with a N-carboxyanhydride core as potent electrolyte additives in lithium-ion batteries.*

Fundamental studies showed that the release of $CO_2$ upon decomposition of additives is beneficial to the cell since it produces $Li_2CO_3$, an inorganic part of the SEI. Therefore, ideally additives should be small, light, cyclic and $CO_2$ releasing. *N*-carboxyanhydrides (NCA) turned out to be suitable for the cell through the formation of oligopeptides resulting in an effective and flexible SEI. During the process, differences in oxidative stability or active (irreversible) lithium loss depending on the property of the functional group attached to the NCA core could be distinguished. Various *N*-carboxyanhydrides bearing different functional groups have been investigated, synthesized and evaluated on cell level and analyzed thoroughly utilizing various analytical tools. This study should help to distinguish important factors when considering certain substrates as SEI additive, to recognize molecular patterns significant for film formation, provide further insights into the SEI forming mechanism and will also help to extend the fundamental understanding of significant substrate parameters towards a systematic approach of electrolyte additive design.[1–3]

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# A physics-based assist data-driven SOH prediction model

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Accurate State-of-health (SOH) prediction of the lithium-ion batteries (LIBs) is critical to fine-tune the battery management system. Empirical aging models, physics-based models and pure data-driven methods have been extensively used to predict the remaining life of LIBs [1]. Empirical aging models can usually be very precise for the specific battery chemistry and operational conditions that they were trained with but usually fail when applied to the new test conditions. Equivalent circuit models (ECMs) are one of the most popular semi-empirical approaches for the state-of-charge (SOC) prediction. While ECMs have the privilege to be implemented online, they normally fail to predict the nonlinear aging behavior, what is known as accelerated aging zone or "knee-point" [2]. Physics-based models span over a large length-scale from atomistic to electrode and cell level models. They also cover a broad range of computational burden, ease of implementation and level of accuracy. Physics-based models based on the porous electrode and concentrated solution theories, i.e., Newman model, are not as popular as ECMs for the SOC and SOH predictions. The major drawback of these models is that careful model parametrization of the degradation physics is needed which normally necessitates tedious and expensive post-mortem analysis. Recently, the use of pure data-driven models and machine-learning approaches have emerged as promising ways to predict the SOH. However, due to the lack of publicly available data repositories, the ability of these models to forecast outside of their training criteria has not been evaluated extensively.

Here and in the context of the 'CurrentDirect' project modelling framework, we propose a physics-based assist data-driven approach where the features for the machine-learning approach come from the simulated voltage-time data using the Newman model. The feature selection process was inspired by the concept of aging modes developed by M. Dubarry [3]. In his approach, the aging behavior of a battery can be described by the three categories of losses, i.e., lithium inventory, anode active-material and cathode active-material. The main advantage of this approach is that prior knowledge on physical origin of the degradation (e.g., SEI formation) is not needed. In our methodology, the three losses are emulated by relevant parameters in the Newman model. For example, the loss of anode material is realized by decreasing the volume fraction of the anode in the model. not surprisingly, such a change will automatically exert changes on the relative position of the cathode and anode OCVs, specific surface area of the anode, the C-rate of the cell and the transport properties.

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# *In situ* Crosslinked Gel Polymer Electrolytes for Li-ion Capacitors

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Supercapacitors are the choice when high-power performance is essential, however, for many applications the energy density of supercapacitors is too low to be used. To increase the energy density, it is possible to combine an insertion type anode of a Li-ion battery with a capacitor type double-layer electrode, forming a so-called Li-ion capacitor (LIC) [1]. There are however several issues with LICs that needs to be resolved to enable a more widespread use in applications. The most critical issue is the pre-lithiation of the anode, which can be either electrochemical or mechanical, this adds an extra processing step and increases the cost of assembly. Another issue is the stability at elevated temperatures and flammability of the organic electrolyte most often used in Li-ion capacitors. Recent progress dealing with the issue of lithiation of a hard carbon anode has been made by researchers at CIC energiGUNE, suggesting the use of a cheap and commercial salt dilithium squarate as a sacrificial salt [2]. For battery applications polymer electrolytes (PEs) electrolytes have been suggested as a possible route to improve stability and increase the safety. A similar approach using PEs could be used in Li-ion capacitors but there are only a few publications for such devices. Important properties of electrolytes in general for LICs are wide electrochemical stability window, high conductivity and a low viscosity to ensure a complete infiltration of activated carbon materials [3]. For a solid electrolyte to fulfill these demands the PE content should be low, the conductivity of the liquid component should be high and the electrolyte needs to be able to be crosslinked *in situ*, so that a PE can infiltrate and be formed throughout all the components of the cell. For this we have investigated both covalently crosslinked and hydrogen-bonded polymers as the polymer component in EC:DMC based liquid electrolytes. These electrolytes forms GPEs *in situ* either spontaneously or through the addition of an initiator that enables the breaking of double bonds, similar PEs have been shown in slightly different systems to provide a high conductivity and wide electrochemical stability windows [4][5]. The aim is to adapt the PEs to better suit the demands of the LICs and to study the physical properties as well as the interaction between electrolyte electrode have been investigated.

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# Wet-chemical surface modification of $LiNi_{0.88}Mn_{0.06}Co_{0.06}O_2$ using organo phosphonic acids

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Ni-rich layered oxides ($LiNi_xM_{1-x}O_2$, $x > 0.5$) are the state-of-the-art high-energy cathode materials for Li-ion batteries, combining a high capacity of over 200 $mAh/g_{\text{active material}}$ and a voltage of 4 V versus $Li/Li^+$. $LiNi_xMn_yCo_zO_2$ (NMC, $x + y + z = 1$) is one of the main compositional subclasses within Ni-rich layered oxides. A high Ni-content is desirable because it is the main electrochemically active element, contributing to the high capacity, and the accompanied cost-benefit of a low Co-content. However, a high Ni-content is detrimental to the cycle life and the thermal stability [1]. One of the inherent issues is the reduction of unstable $Ni^{4+}$ to $Ni^{2+}$ and the release of lattice oxygen as charge compensation. $Ni^{2+}$ can migrate to $Li^+$ sites within the anionic framework, blocking the $Li^+$ diffusion layers, and transforming the layered oxide structure into spinel-like and rock-salt phases. These phase transitions occur at the particle surface during cycling and to the bulk material when heated to higher temperatures [2,3]. The oxygen loss upon heating in combination with conventional flammable liquid electrolytes is a serious safety concern. Thus, avoiding release of lattice oxygen is key to stabilising Ni-rich layered oxides with respect to both performance and safety characteristics. Surface modification by coatings and substitutional doping with elements with strong M-O bonds are two widely used strategies to stabilise the material. For instance, polyanionic phosphate-based materials have shown to be effective towards improving electrochemical and thermal characteristics, attributed the strong P-O bonds [4].

This work explores organo phosphonic acid-based surface modifications of $LiNi_{0.88}Mn_{0.06}Co_{0.06}O_2$ (NMC88) applied by wet-chemical processing. The chosen coating materials consist of a phosphonate $-PO(OH)_2$ group bonded to an organic tail, and they have previously been studied as self-assembled monolayer surface modifiers for oxides in organic photodetectors. The aim is to obtain a thin surface coating with a strong affinity to oxygen by attaching the phosphonate functional group to the NMC88 surface. The Ni-rich NMC88 is dispersed in an aprotic solvent with phosphonic acid, reacted, rinsed, and annealed in oxidative or reductive atmospheres. Physical characterisation by TGA is done to track the development during annealing in the various atmosphere and by XPS to investigate species on the modified surface. These analyses are accompanied by electrochemical characterisation in full-cells will be used to evaluate the effect of the surface modification on the cycling stability.

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# Reversible electrochemical solvent co-intercalation for sodium-ion batteries

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In 2014 it was shown that graphite had a far greater capacity to reversibly store sodium-ions than what was previously known. This, however, can only occur through an electrochemical solvent co-intercalation reaction, whereby the sodium-ion is intercalated along with solvent molecules – specifically linear ethers [1]. Since then, several studies have revealed many facets of these reactions, but the subject remains somewhat unexplored, and several basic questions have received quite conflicting answers – Especially conflicting reports on the existence, or non-existence, of the SEI [2][3]. This talk purport to summarize our research activity on electrochemical solvent co-intercalation phenomena for sodium-ion batteries, ranging from fundamental questions such as how to detect solvent co-intercalation, to the nature of the electrochemical reaction – including the stoichiometry of the reaction(s) and the great kinetics of these systems – to our research into the SEI. We also present our efforts to increase the capacity of the system and to mitigate the great electrode volume expansion. Finally, we present our results on full cells, and a new type of battery – the solvent co-intercalation battery (CoIB) [4] – where both the positive and negative electrode operate by a co-intercalation reaction, Figure 1.

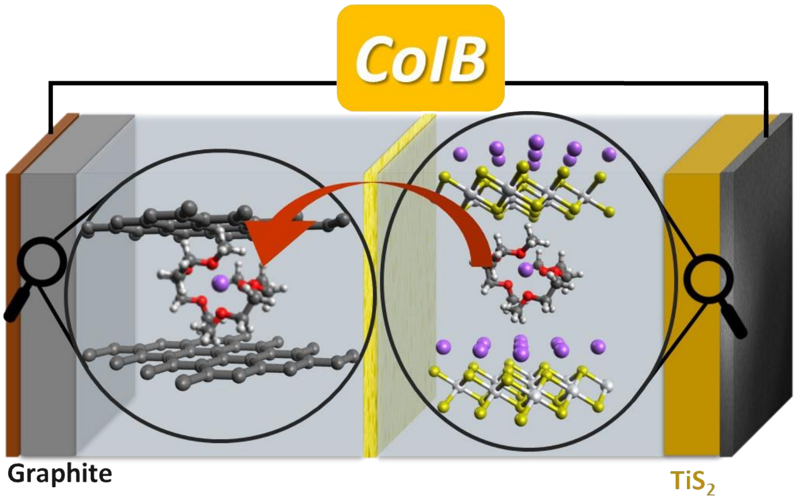

***Figure 1.*** *A Co-Intercalation Battery (CoIB), where both the anode and cathode, here graphite and $TiS_2$, operate by a co-intercalation mechanism.*

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# Insights into circular economy potential of lithium by system dynamic modelling of material streams

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Lithium has several applications in a modern society. Today, the most important application is batteries with 74 % of all lithium in use, followed by ceramics, glass, and lubricating greases [1]. The global lithium production totalled 100k tonnes in 2021 (ibid.). Despite its significance for the electric mobility and electronics industry, the reuse and recycling rates of lithium are rather low. However, the markets for reuse and recycling are evolving with the increase in the number of electric vehicle batteries reaching their end of life. To build a holistic understanding of circular economy potential of lithium flows and storages, this paper presents a system dynamic modelling approach to describe the global lithium streams in the main industrial applications areas. The lithium value chain from extraction to the end of life is determined, and the key value chain stakeholders are identified.

System dynamics approach enables to develop understanding of how dynamic and complex systems behave and, based on that, to design more efficient organisations and policies that influence either a part or the entire system. System dynamic modelling have been utilized to simulate, e.g., closed-loop supply chains, recycling strategies, product life cycles and material flows. We will apply the approach to investigate the systemic lithium flows. Preliminary lithium flow model is created using Vensim (**Fig. 1**).

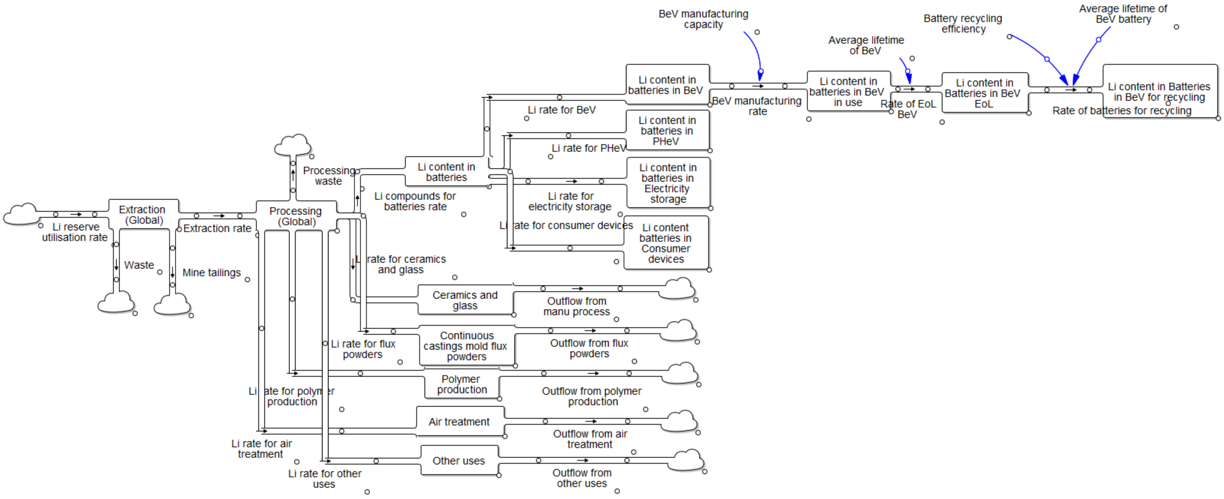

*Figure 1.* *Modelling lithium flows with system dynamics.*

Next step is to collect the relevant data and build the key interdependencies to model the global, European and Finnish lithium flows and storages. Furthermore, the goal is to test the potential of emerging recycling technologies and incentives for the reuse and recycling of lithium. This work is part of Academy of Finland funded GoverMat project, which will develop new insights into circular economy of critical raw materials for electric mobility.

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# Mapping of Lithium in Carbon Fibre Electrodes for Structural Batteries

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Lightweighting of components in electric vehicles helps to increase driving range. Still, the battery systems of today constitutes a large part of the electric vehicle’s weight. “Mass-less” energy storage can be achieved through multifunctional devices such as the structural battery where energy storing capabilities are integrated into load carrying components [1], [2].

The pivotal constituent in these batteries is the carbon fibre, which acts simultaneously as electrode and reinforcement. The carbonaceous microstructure of the carbon fibre can reversibly host and release lithium (Li) ions. During the lithiation and delithiation process the fibre experiences volumetric expansion and contraction together with changes in the mechanical properties [3], [4].

However, little is known of how Li distributes inside the carbon fibre during charge and discharge cycles. With Auger electron spectroscopy (AES), light elements such as Li can be identified at a nanometre scale. With X-ray photoelectron spectroscopy (XPS) the chemical states of elements can be identified. Reliable experiments on lithiated carbon fibres require protection from ambient air and moisture during transfer from glovebox to instrument as to negate side reactions. Here we use AES and XPS compatible with vacuum transfer system to study commercial polyacrylonitrile-based carbon fibres at different states of lithiation. This knowledge increases the understanding of how carbon fibres interact with Li and will potentially inform how future carbon fibres should be designed to improve the multifunctionality of structural batteries.

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# Magnesium doped high-nickel layered oxide cathode enabling safer, high-energy density lithium-ion batteries

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Nowadays lithium-ion batteries (LiBs) are popular electrochemical energy storage devices that are widely used both in everyday commodities and in special duties such as electrical and hybrid vehicles. LiBs have a really wide range of applications because of their high energy density and power, long lifespan, and low self-discharge. High-nickel layered oxides ($Li(Ni_{0.8}Mn_{0.1}Co_{0.1})O_2$ i.e., NMC811)) are considered as a new generation of positive electrode materials due to their excellent performance, high capacity (up to 200 mAh $g^{-1}$) and energy density, low cost, good stability and safety. Manganese can stabilize the crystal structure of the layered oxides during the intercalation/deintercalation of lithium ions and improve the cycling properties of LiBs and in turn, nickel is responsible for the high capacity. Except for the chemical composition, the development of a surface, microstructure and morphology also influence the performance of the intercalation/deintercalation of lithium ions. However, there are several challenges such as fast capacity fade and significant heat/gas release during electrochemical cycling. The possible structure disorder and side reactions can lead to decreasing capacity and hinder the Li ions transportation. Besides, in such materials, the rate of Li ion diffusion is slow for the fast charging at the high-voltage cycling. The doping of NMC811 by electrochemically inactive substituents such as Mg allows to increase the stabilization of the layer structure and to decrease in the Ni ions migration into the Li layer. In the literature, the typical potential window for NMC811 is 2.8 V – 4.3 V, while in this work, the cutoff potential was increased up to 4.6 V to investigate the degradation of such materials and show the ability of NMC811 for application in high-energy LiBs.

In this work, the as-prepared NMC811 precursor was lithiated by the solid-state method and during this process, 0.25, 0.50 and 0.75 wt% of Mg were added. The morphology of the obtained materials was investigated by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) analysis to investigate the overview of the particles, the size of the primary and secondary particles and other structural features. X-ray diffraction (XRD) was used to study the crystal structure and cation mixing. Charge-discharge characteristics and long cycling were studied to investigate the influence of Mg doping on the performance and the specific capacity of the obtained materials. The obtained materials were investigated at two cutoff potentials (4.4 V and 4.6 V) and at two different conditions of the formation cycles (4.2 V and 4.4 V for 0.03C and 0.1C-rates). In addition, galvanostatic intermittent titration technique (GITT) measurements were done to estimate Li-ion diffusion upon cycling. The Mg-doped NMC811 describes much improved capacity and high-rate capability in Li-half cells as compared with the pristine NMC811. All obtained materials have high capacity retention after 100 cycles at the 4.6 V cutoff potential in half cells. This work shows the dependence between structural characteristics and electrochemical performance for the Mg doped NMC811 positive electrode materials. The localization of Mg ions in the NMC811 crystal structure and the reason for increasing cation mixing with increasing the amount of Mg were investigated, as well as how these affect the electrochemical behaviour as the positive electrode material. Moreover, decreasing the cutoff potential of the formation cycles leads to increase the stability of cycling and the specific capacity of the obtained materials.

# Fiber Optical Detection of Lithium and Sodium Plating in Intercalation Batteries

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Plating of metallic lithium or sodium is one of the most critical events causing problems in modern intercalation batteries. The plating on the anode, for example as a result of a too high charging rate, causes detrimental side reactions leading to a reduced lifetime of the battery. The growth of lithium or sodium dendrites can also lead to catastrophic failure as they can penetrate the separator and cause a short-circuit in the cell. This presentation focuses on recent and unique findings from Uppsala University [1], where information from a fiber optical sensor inside a battery was found to reveal early signs of plating [2]. Figure 1 below shows an illustration of the fiber on top of the hard carbon anode in a sodium ion battery, and this type of experiments could eventually provide a battery management system (BMS) with input to optimize the charging rate just below the limit of plating.

Having access to a sensor inside the cell on top of either the anode or cathode thus offers unique possibilities to track detrimental reactions in the battery. We also anticipate that the fiber optical sensor could serve as a research tool to learn more about the intercalation and plating mechanisms, as well as the optical properties of the anode or cathode materials during cycling [3]. Preliminary experiments on a prototype commercial sodium ion battery also indicated that the plating could be detected via the fiber, while it was not clearly seen in the cell voltage even if a true reference electrode was included in the cell. Comments will also be made on future applications of this type of sensor. The small size of the optical fiber allows it to be positioned at specific spots in a large battery cell. This is, for example, advantageous as the plating in commercial cells is known to be distributed heterogeneously.

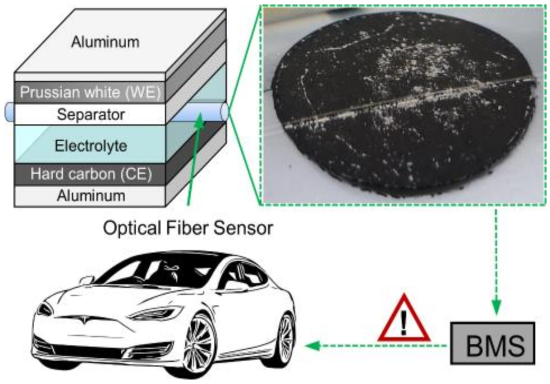

***Figure 1.*** *Schematic picture of the optical fiber on top of the anode, and the possibility to feed information to a BMS.*

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# Investigating Surface Sensitivity of Ni-rich Cathode Material towards $CO_2$ and $H_2O$

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## Abstract

Layered Ni-rich transition metal oxide materials have been considered as the most promising cathode utilized in Li-ion batteries, e.g., $LiNi_{0.8} Mn_{0.1} Co_{0.1} O_2$ (NMC 811). However, one of the drawbacks of NMC 811 is its high air sensitivity, leading to a degradation layer forming on the surface, and a lower cycling performance [1]. Since the degradation mechanism is not fully understood, in this work, we use ambient pressure photoelectron spectroscopy (APPES) [2] to investigate the surface sensitivity of NMC 811 towards $CO_2$ and $H_2O$ *in situ*, aiming to determine the factor triggering the degradation. Before gas exposure, NMC 811 surface was studied in UHV. The changes in surface chemical composition were monitored as a function of time and gas pressure. Results show that carbonate compounds will form on the surface when NMC 811 is exposed to $CO_2$ at around$10^{-3}$ mbar and start to disappear in UHV after $CO_2$ exposure. More interestingly, the photon beam can accelerate the formation of carbonate on NMC particles surface. The same measurements were finished with $H_2O$ exposure as well. Results indicate that lithium hydroxide is formed, where $Li^+/H^+$ exchange on the surface is a possible route.[3] However, this reaction is reversible in UHV as well.

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# Impact of structure on capacity fade during long-term cycling in Prussian White

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A promising cathode material for $Na^+$ ion batteries is the Fe-based vacancy free Na-rich Prussian blue analogue (PBA) known as Prussian White (PW) $Na_{2-x}Fe[Fe(CN)_6]$. PW has a high operating potential (~3.2 V) and theoretical capacity (170 $mAhg^{-1}$) making them competitive with $LiFePO_4$. [1] PBAs have been demonstrated to enable reversible insertion and extraction of sodium for up to 8000 cycles. [2] However, this is only possible if the capacity is limited to about half of the theoretical (~80 $mAhg^{-1}$) corresponding to keeping within $1 \leq x \leq 2$ in $Na_{2-x}M[Fe(CN)_6]$. In all Fe-based PBAs, this composition corresponds to a solid solution reaction where the cubic structure is retained, and minimal volume change occurs during reduction of the carbon bound Fe. [1] If one cycles beyond a critical Na content (which varies depending on the number of $[Fe(CN)_6]^{3-}$ vacancies), a phase transition to a higher density structure (~18% change in volume) is observed. From previous research, we suggested that this phase transition over time will become detrimental to the electrochemical performance due to structural degradation, which will increase the resistance of insertion of a second Na-ion into the structure. [1] As predicted, our recent work demonstrated a capacity fade of 15% in the lower voltage range (1.3 V-2.9 V) after 65 cycles (Figure 1). Interestingly, significantly less capacity fade is experienced from the upper plateau, implying that the structure must be preserved.

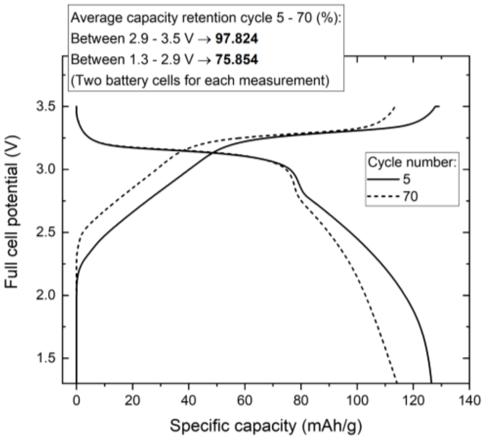

***Figure 1.** Cycling curves showing a loss of capacity from the low voltage region.*

Using *operando* powder X-ray diffraction, the structural changes occurring in PW full cells, when cycled over 100 times within different voltage windows, were monitored to access the role the structural transition plays in the loss of performance over time. Initial analysis suggests that the structure is indeed preserved when cycling in the full voltage range even though the cells have been pre-cycled 100 times in a limited voltage plateau. Furthermore, cells that have been pre-cycled 1000 times in the upper voltage plateau seem to experience structural degradation as the cubic phase does not reoccur during charging. Understanding the role that the structure plays in material performance will allow us to understand the origin of the observed capacity fade and thus develop cycling protocols and strategies for eliminating capacity fade.

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# Mitigating Interfacial Reactivities in Sustainable Aqueous Li-ion Batteries with DMSO as a Co-solvent

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Aqueous Li-ion batteries (ALiBs) are alternative solutions for large-scale grid storage, as water is intrinsically non-flammable, inexpensive, and easy to purify. However, the energy density of ALiBs is limited by the narrow water electrochemical stability window (ESW) of 1.23 V, beyond which water electrolysis will take place. Although using a highly concentrated electrolyte consisting of 21 mol kg$^{-1}$ (21 m) of LiTFSI in water, known as the "water-in-salt" (WiS) electrolyte [1], can widen the electrolyte ESW to ~ 3 V, its high cost and low environmental sustainability impede the successful commercialization of ALiBs. An alternative strategy to decrease the electrolyte activity while maintaining a relatively lower salt concentration is to introduce co-solvents, such as poly(ethylene glycol) (PEG) [2], which will confine water molecules by hydrogen bonds. In this way, aqueous electrolytes containing 2 m of LiTFSI with > 3 V ESW can be achieved.

Herein, we report that dimethyl sulfoxide (DMSO), a green organosulfur solvent with high dielectric constant and full miscibility with water, can be used as an effective co-solvent in dilute ALiBs to mitigate detrimental interfacial reactivities. We observe, via online electrochemical mass spectrometry (OEMS) technique, an over 60% decrease of $H_2$ evolution in model $LiMn_2O_4$ || $LiTi_2(PO_4)_3$/C full cells using a 2 m LiTFSI electrolyte with a DMSO/water molar ratio of 0.33 (**Fig. 1**). We further confirm multiple sources contributing to the $CO_2$ evolution in ALiBs using isotope labeling coupled with OEMS. Our study sheds light on the development of low-cost, robust, and sustainable aqueous batteries for large-scale energy storage applications.

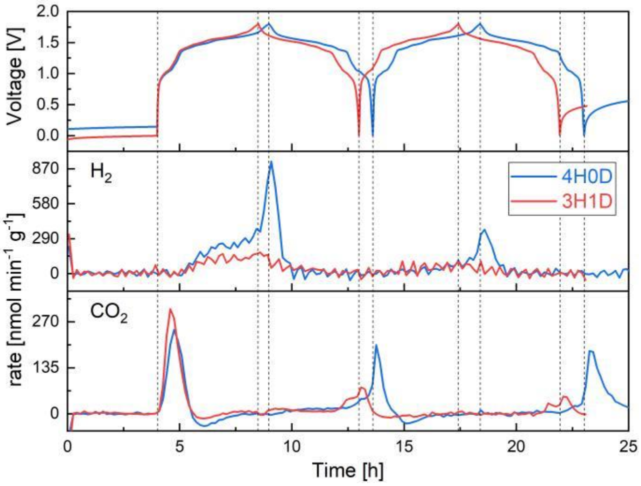

***Figure 1.*** *Quantitative comparison of gas evolution in ALiBs with different DMSO/water ratios. 4H0D denotes a fully $H_2O$-based electrolyte, while 3H1D denotes an electrolyte with a molar ratio of 3:1 for $H_2O$ and DMSO.*

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# Towards the industrialization of solid-state batteries: Impact of dry room exposure on sulfide-based solid electrolyte materials

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All-solid-state battery technology stirs great interest in the battery community and beyond. Sulfide-based solid electrolytes are attracting particular attention, mostly because of their unrivaled ionic conductivity. [1] However, because of their high sensitivity to moisture – they release toxic $H_2S$ when in contact with water – most studies in the literature have, understandably, studied these materials in Argon-filled gloveboxes. If sulfide-based solid-state batteries came to be produced at industrial scale, the use of dry rooms as a production environment would be much more advantageous than a completely inert atmosphere. [2]

At the Battery Processing and Prototyping team of the CEA in Grenoble, we are currently studying how this type of technology could be scaled-up and in which environment. For this, we have exposed sulfide-based solid electrolytes to a typical dry room atmosphere and investigated the impact on operator safety and material characteristics and performance.

During the exposure of sulfide materials $Li_6PS_5Cl$, $Li_7P_3S_{11}$ and $Li_{5.8}PS_{4.8}Cl_{1.2}$ to the dry room atmosphere, the kinetics of $H_2S$ production were followed thanks to accurate laser detection. An array of physico-chemical characterisation techniques (X-ray diffraction, Raman spectroscopy, electrochemical impedance spectroscopy, scanning electron microscopy and X-ray photoelectron spectrometry) was then used to shed light on the degradation mechanisms. Finally, the electrochemical characteristics of solid-state LiIn-NMC cells containing pristine and exposed sulfide material were compared.

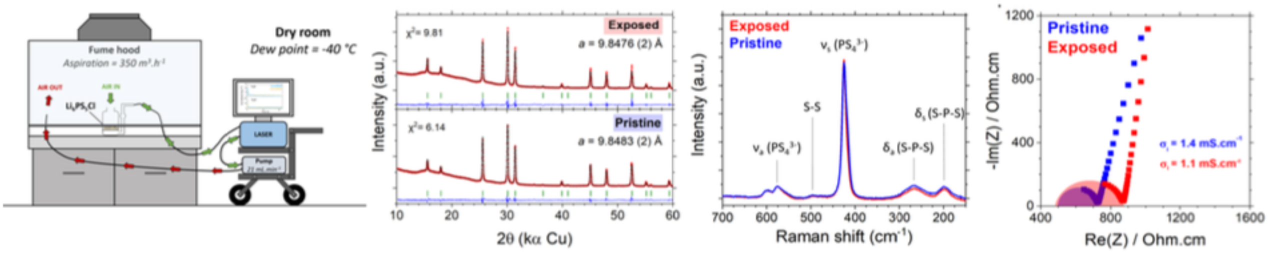

***Figure 1. Exposition of sulfide electrolyte to dry room atmosphere and physico-chemical characterization***

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# Tracking Lithium-Ion Battery Aging with Optimally Designed Experiments

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Electrochemical and mechanical aging-phenomena in lithium-ion batteries have received significant attention from researchers [1]. Physics-based models of these effects support the mechanistic understanding of degradation modes and can thereby help reduce their severity. However, as many aging mechanisms are still poorly understood, direct modelling is often impractical. Instead, the effect is studied by evaluating changes in cyclable lithium and active material contents [2] or changing electrochemical parameters such as diffusion coefficients or reaction rate constants that have a direct impact on model accuracy [3]. Lyu et al. [3] used a simplified electrochemical model and monitored battery degradation by following changes in diffusion time constants, electrode balancing, reaction rate coefficients, and ohmic resistance. However, several of the parameters they attempted to track could not be identified accurately as they use the same dataset to identify all parameters. To overcome these issues and ensure parameter identifiability, we design and use optimal experiments for parameters of a full order Newman-type model [4]. Parameters are estimated and tracked over the course of a batteries lifetime under real-world load-cycles.

In a previous study [5] we demonstrated global optimal experiment design for parametrization of electrochemical battery models. We now extend this work and re-evaluate key parameters over the course of an aging study on commercial, nickel-rich 18650 lithium-ion batteries. We highlight how quantifying changes in physical battery parameters can extend standard performance metrics for a batteries state-of-health by, e.g., including degradation in rate-capability. Additionally, the importance of battery usage conditions such as C-rate or state of charge window on model parameter trajectories is investigated and their relationship with conventional performance metrics such as the bulk cell resistance or rate-capability determined.

Quantifying how specific mechanisms contribute to apparent capacity or power fade is a major step towards battery lifetime optimization. Evaluating parameter changes is essential for electrochemical control strategies relying on accurate model predictions of battery states. A recalibration would make a battery management system aging-sensitive and enable more efficient utilization and a physics-informed state of health.

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# Combining *Operando* XAS and XRD on the Balder Beamline at MAX IV

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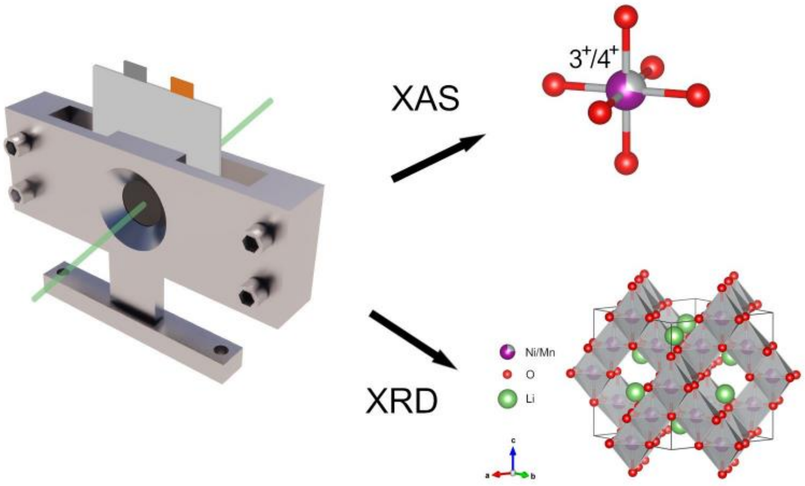
Access to X-ray energies in the 2.4-40 keV makes the Balder beamline suitable for conducting X-ray absorption spectroscopy (XAS) studies on various transition metal containing battery materials. The setup at Balder enables fast scans across quite large energy ranges to capture the full absorption edge, including the XANES and EXAFS regions, in the order of seconds. In combination with the high brilliance available at MAX IV, time-resolved *operando* studies on batteries are as such especially well suited. With the possibility of simultaneous collection of X-ray diffraction data (XRD), Balder provide an excellent opportunity for the combination of tracking changes in oxidation state and local structure from XAS with changes in the long-range crystalline structure from XRD.

In the work presented here, results from combined *operando* XAS and XRD measurements carried out at Balder on the high voltage cathode material $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) will be presented. A cell design optimized mainly for *operando* XRD measurements [1] was employed and proven suitable also for XAS studies. *Operando* XAS and XRD measurements during cycling at 1C were done, covering both the Ni and Mn K-edge, including the XANES and EXAFS regions. Simultaneously, anomalous X-ray scattering was explored by collecting XRD data at the Mn K-edge, as a means to identify the ordering between Ni and Mn in the crystalline structure of LNMO. The full collection of XAS and XRD data across one energy sweep could be done in ~1.5 minutes, showcasing the speed and time-resolution possible at Balder. Finally, practical aspects for conducting a successful battery *operando* experiment on Balder, such as experimental setup, cell design, electrode optimization and beam damage will be discussed.



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# Study of Ga-based Anodes for Li and Na-ion Batteries

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One of the critical points to improve batteries is their energy density. It can be enhanced by modifying electrode active materials. On the anode side, the most common active materials used in Li and Na-ion batteries are carbon-based and have capacities of about 500-800 mAh/cm$^3$ (300-400 mAh/g). They can be substituted with some metals able to form alloys containing a large amount of Li or Na, leading to large energy densities. One example is tin. It is able to reach a capacity of 6969 mAh/cm$^3$ (997 mAh/g) as Li-ion anode and 6213 mAh/cm$^3$ (850 mAh/g) as Na-ion anode.[1,2] However, this kind of alloying electrodes suffer from large volumetric expansion leading to particle cracking and a continuous electrolyte decomposition.[3] These drawbacks could be avoided if these alloys had self-healing properties. There are alloys that are liquid at room temperature and are self-healing. The most common ones are $Ga_{86}In_{14}$, $Ga_{92}Sn_8$ and $Ga_{77}In_{15}Sn_8$ eutectic alloys that have melting temperatures of 16 °C, 25 °C and 11 °C, respectively.

In the recent years, these alloys have been studied in the field of Li-ion batteries as self-healing electrodes by different groups.[4] In this presentation, our study on $Ga_{77}In_{15}Sn_8$ (galinstan) as anode for Li-ion batteries will be presented.[5] It was shown that after delithiation, the liquid galinstan recover its initial shape due to the transition from the lithiated solid phases back to the pure liquid delithiated phase. In the case of Na-ion batteries, there is almost no study on the use of Ga-based alloys as electrode active materials. Our study on the sodiation of Ga will be described in this presentation.[6] This was fundamental to later understand the Ga-based alloys. Then, our study of $Ga_{86}In_{14}$, $Ga_{92}Sn_8$ and $Ga_{77}In_{15}Sn_8$ liquid alloys as electrodes for Na-ion batteries will be presented. The electrochemical reactions of these compounds with Na were studied in details. Electrochemical measurements were coupled with ex-situ X-ray diffraction and scanning electron microscopy analyses to obtain a complete overview of the involved reactions, their potentials, their capacities and the self-healing properties of the alloys. It was shown by ex-situ X-ray diffraction and scanning electron microscopy that only $Ga_{86}In_{14}$ featured completely reversible sodiation/desodiation reactions with a return to fully liquid state after sodiation.

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# Electro-chemo-mechanical failure of interfaces in solid-state batteries

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There is an increasing demand for batteries with even high safety, higher energy density as well as high power density for practical applications in portable electronics market and electric vehicle (EV) industry. However, the energy density of conventional Li-ion batteries (LIBs) will soon reach their limit and they are also suffering from thermal runaway which is related to the flammable nature of liquid electrolyte used. Replacing liquid electrolytes with solid-state electrolytes (SSEs), that are fabricating solid-state batteries (SSBs), shows a great promise to address these issues.

So far, there is still a large gap of available power density in solid-state batteries by a comparison of practical industry standard. This is because an internal short-circuit of battery will be invariably triggered when the operating current density of solid-state batteries is over a critical value. The failure mechanism at micro level is the growth of Li filaments and its penetration of solid electrolyte to connect cathode and anode inside battery. The academic community is intensely arguing where the Li dendrite is formed and how the soft Li can impale a rigid ceramic solid electrolyte.

In this study, electro-chemo-mechanical failure of interfaces in solid-state batteries, which are induced by nucleation, growth, and penetration of lithium (Li) metal inside/through solid electrolytes, is investigated by multiphysics simulation to understand correlation between the properties of solid components and the electrochemical performance of solid-state batteries. Our results show that the stress concentration and preferred deposition of Li metal anode are highly dependent on the ionic conductivity and mechanical strength of the interface between Li anode and electrolyte [1]. Furthermore, the stability and failure of the interface also show strong influence on the preferred location for growth of Li during the electrodeposition process, by visualizing the distribution of Faradic current density and concentration field on electrode [2]. Our understanding will be beneficial for exploring strategies of design of solid electrolytes as well as interfaces for future solid-state batteries [3].

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# Evaluating the passivation layer in non-aqueous aluminium batteries

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Aluminium (Al) is an attractive metal anode for rechargeable batteries due its low density, cost, large abundance in the Earth's crust, and recyclability.[1] Yet, to create Al-batteries the uttermost needed suitable cathodes and non-corrosive electrolytes are elusive. With respect to the latter, up to date, the most widely used non-aqueous electrolyte, enabling reversible plating and stripping of Al, is [EMIm]Cl-$AlCl_3$, where the active ions are $[AlCl_x]^{3-x}$ rather than $Al^{3+}$, lowering the practical capacity and consuming electrolyte in the charge/discharge process.[2] The use of alternative, including aqueous, electrolytes has been hindered in part by the presence of a native passivation layer on the Al metal anode.[3] For non-aqueous electrolytes the latter is intimately related to the formation of an SEI-type layer that lowers the Coulombic efficiency.[4,5] Surprisingly few Al-battery studies have been devoted to this passivation layer, as understanding its reaction(s) with the electrolyte should be vital to improve performance.

Here we use the [EMIm]Cl-$AlCl_3$ electrolyte and study the passivation layer formed by a combination of electrochemical impedance spectroscopy, X-ray photoelectron spectroscopy and scanning electron microscopy applied to symmetric Al cells at OCP (open circuit potential) and after CC-GC (constant current galvanostatic cycling). Both growth of the passivation layer and etching of the Al metal, with formation of pits and secondary pores, are observed (Figure 1). The etching is accompanied by deposition of insoluble decomposition products, leading to an inhomogeneous surface, where the original and freshly formed passivation layer and corroded Al co-exists. Furthermore, both oxide and carbon based passivation layers are observed, the latter possibly similar to an SEI. All this should be invaluable information in order to control and improve the Al plating and stripping.

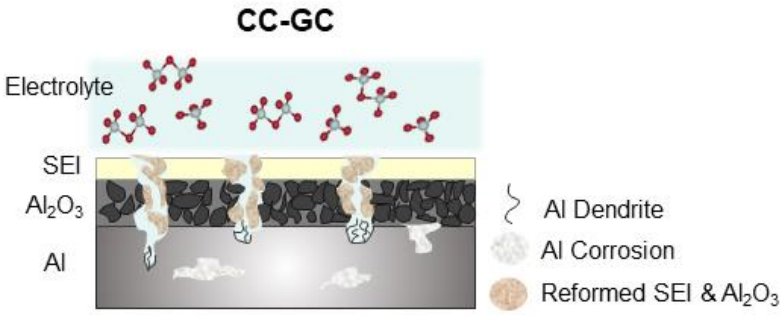

Figure 1: Visualization of the Al metal anode development including corrosion and deposition.

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# Electrochemical Performances of Powder-Impregnated Carbon Fiber-Based Positive Structural Electrodes

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A battery entirely based on carbon fibers, often referred to as the structural battery introduced in 2004 by Wetzel et al. [1], is a multifunctional battery that can carry the load and store the energy simultaneously. An advantage of this battery design is the lower overall weight of the device due to eliminating dead mass by removing current collectors and additives from the entire structure. Instead, introducing carbon fibers into lithium-ion batteries decreases the non-active mass and provides mechanical stability to the system [2].

This work produced positive structural electrodes based on polyacrylonitrile (PAN)-based carbon fibers with a powder impregnation technique in a water-based slurry composition (**Fig.1a.**). This environmentally friendly technique allows reasonable control of impregnation uniformity and active material/fiber ratio. By this method, continuous carbon fibers were impregnated with active material ($LiFePO_4$ /LFP) along with electrode components to obtain self-standing positive electrodes. The as-prepared samples show a good and uniform mixing with the active material through interlayers of carbon fibers in tow, leading to a lower internal resistance (SEM images in **Fig.1b.**). The carbon fiber-based positive electrodes have been tested with different electrochemical methods to obtain their half-cell performances (**Fig.1c.**). The results show good capacity retention and rate capability.

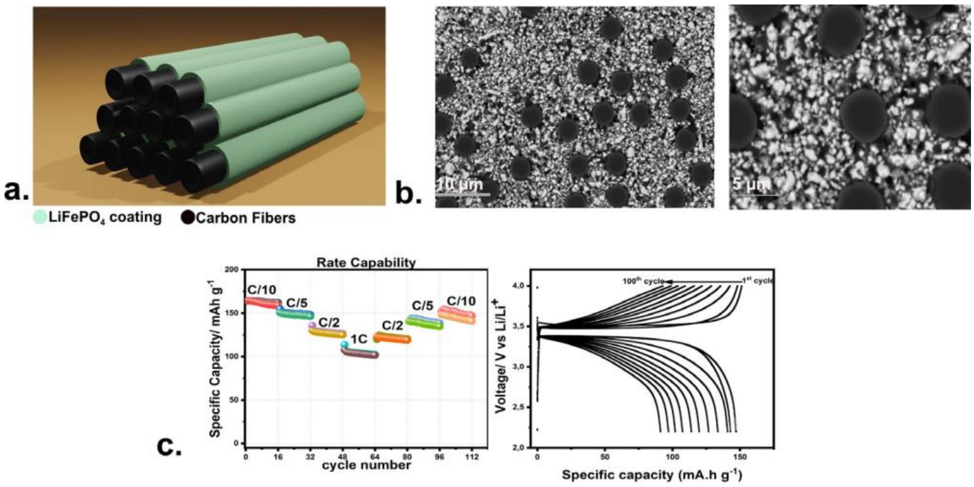

***Figure 1.*** *a. Schematic illustration of coated carbon fibers b. SEM images of as-coated carbon fibers (cross-section) c. Galvanostatic charge-discharge profiles of the coated carbon fibers in terms of rate capability and long-term performances in half cells*

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# Electrolyte Recovery from spent Electric Vehicle Lithium-Ion Batteries using Supercritical Carbon Dioxide Extraction

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The demand for Lithium-Ion batteries (LIBs) steadily increased over the past few years as a result of the excessive production/consumption of electric portable consumer electronics (smartphones, laptops, tables, etc.) and electric vehicles. Inappropriate disposal of spent LIBs potentially triggers a serious threat to the environment due to their hazardous components, such as transition metals (Lithium, Cobalt, Nickel), electrolytes, and other fluorine containing organic components. Reutilization and recycling of spent Lithium-Ion batteries will not only minimize the waste stream containing hazardous materials but are also valuable secondary raw materials for the industry. The state-of-the-art recycling methods use a combination of pyro-and hydro-metallurgy processes to reclaim valuable raw materials from spent LIBs. The recovery of the electrolyte is rarely considered, but rather evaporates and/or decomposes during the pre-treatment and high temperature thermal treatment steps to liberate the valuable active materials. Controlled and safe removal of the electrolyte is inevitable to prevent severe threats produced by the inflammable, toxic and hazardous components of the electrolyte and minimize the environmental impact and safety of the industrial recycling processes.

Supercritical carbon dioxide (Sc-$CO_2$) is a versatile process technology that can be used for organic and metal extraction as well as material production. Sc-$CO_2$ is formed once the pressure and temperature of $CO_2$ exceed its critical point of 31°C and 73.8 bar. In the so-called supercritical state unique properties such as liquid-like densities, gas-like compressibility, viscosity diffusion coefficient and neglectable surface tension will be obtained. Those properties can be fine-tuned to adjust the physicochemical characteristic of the solvent such as solvation, solubility, selectivity, and reactivity for electrolyte recovery.

In this study, a unique Sc$CO_2$ process was developed to recover the electrolyte from spent electric vehicle LIBs. The effects of process temperature and pressure on the recycling efficiency of the electrolyte were investigated. The recovered electrolyte was analyzed by gas chromatography (GC-MS) and the off-gas was characterized by In-Situ Fourier-transform infrared spectroscopy (FTIR) to detect separated compounds and possible reaction products. The results show that the electrolyte solvents, which were dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), and ethylene carbonate (EC) were extracted and recovered from the spent LIBs using supercritical carbon dioxide at low pressure and temperature. The innovative Sc$CO_2$ method eliminates the formation of hydrogen fluoride (HF) and phosphoryl fluoride ($POF_3$) during the battery recycling process.

**Acknowledgements**
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# Ion implantation as a potential strategy for dendrite prevention in solid-state Li batteries

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Li metal would be the desired anode material for future energy storage. It has a tenfold higher theoretical capacity then the commonly used graphite anode and the lowest possible anode potential.[1] So far, the implementation of Li metal has been hindered due to its high reactivity and the formation of dendritic structures during charging posing a severe safety risk. It has been suggested that those hurdles can be overcome by the use of non-flammable ceramic electrolytes like $Li_7La_3Zr_2O_{12}$ (LLZO) instead of flammable liquid ones.[2] Unfortunately, Li dendrites still remain an issue, able to penetrate the ceramic electrolyte even at low current densities. This failure is associated with a significant pressure build up in surface defects, such as flaws and pores. Once those get filled by Li and the Li deposition rate exceeds the Li flux away from the defect, cracks further propagate into the ceramic electrolyte until short circuiting. [3] [4] Delaying or even stopping the propagation of cracks is, therefore, of great interest in order to improve the rate performance of Li metal batteries.

Previous studies have shown that the introduction of surface-near compressive stresses into structural materials can effectively inhibit crack propagation.[5] Taking advantage of this knowledge, we used ion implantation to tune the surface-near mechanical properties of LLZO with the aim to stop Li dendrites by taking away their driving force. To understand, how and to which extent the implanted ions alter surface-near properties, such as composition, microstructure, and mechanical properties, we used a large array of characterization techniques, such as FIB SEM, dark field TEM, cross sectional nano XRD, atom probe tomography, and nanoindentation.

Overall, this study demonstrate that Ag ions can introduce high compressive residual stresses in the near-surface region of LLZO up to -750 MPa at about 600 nm depth. Most importantly, however, that stress peak at 600 nm effectively deflects cracks when a mechanical load is applied with potentially large implications on the ability of LLZO to withstand Li dendrites.

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## Acknowledgement

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# NanoPlasmonic Sensing for *Operando* Monitoring of Internal Processes and Phenomena in Li-ion and Na-ion Batteries

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Insplorion's NanoPlasmonic Sensing (NPS) enables remote and external monitoring of internal processes and phenomena occurring in Li-ion batteries (LIBs) and Na-ion batteries (SIBs). The NPS sensing structure is placed on an optical fiber that is inserted in between the separator and one of the electrodes in the battery cell where it responds to physico-chemical changes in the battery environment close to the probe. The optical NPS data is measured and collected using an Insplorion M8: 8-Channel Analyzer, with which up to eight battery cells can be measured on simultaneously. Insplorion's proprietary NPS technology measures changes in the dielectric function—effective refractive index—of the surrounding medium. The technology can thus be used to measure/monitor various processes occurring in the electrode material such as (de)lithiation, including lithium diffusion into the material, and crystal structure changes. In this presentation we will give an overview of Insplorion's NPS technology, and outline, with several application examples, how it can be used in research and development.

# Quantification of the ion coordination strength in solid polymer electrolytes with lithium and beyond

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Implementing solid polymer electrolytes (SPEs) into batteries have gained great interest owing their inherently advantageous safety properties and the possibility to utilize high-specific-capacity anodes. However, due to the low ionic conductivity compared to liquid electrolytes, the process of implementation as SPEs is halting and a more fundamental understanding of the ion transport is required for full realization in batteries. [1]

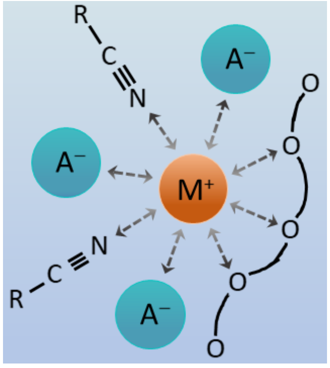

***Figure 1.** The ion coordination strength is the combined interaction strength contribution from the ion–dipole interaction and the coordination number (CN).*

In our earlier studies, important effects of the ion coordination strength, *i.e.* the interaction strength between the cations and the polymer chains (**Fig. 1**), on the cation transport properties in SPEs have been demonstrated, revealing a strong correlation between the ion coordination strength and the transference number ($T_+$) for Li systems. [2]

So far, the ion coordination strength has been reported qualitatively, giving information about the relative coordination strength between different polymer electrolytes. Herein, a novel method based on conventional FTIR measurements is used to quantitatively determine the ion coordination strength for TFSI-based salts of $Li^+$, $Na^+$ and $Mg^{2+}$ in polyethylene oxide (PEO), poly(ε-caprolactone) (PCL) and poly(trimethylene carbonate) (PTMC). Since the investigated systems are solvent-free, the ion coordination strength is resolved by determining the dissociation energies of the salts in each polymer. In **Fig. 2**, the van't Hoff plots of the dissociation of the salts are shown, from which the entropies and enthalpies are extracted, giving the Gibbs free energy for the dissociation of the salts and ultimately the ion coordination strength.

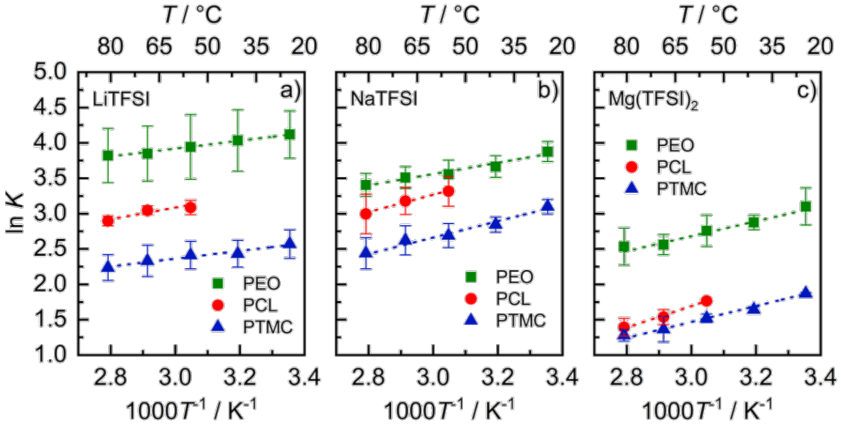

***Figure 2.** Van't Hoff plots of the equilibrium constant K as a function of the inverse temperature for the dissociation of a) LiTFSI, b) NaTFSI and c) $Mg(TFSI)_2$ in PEO, PCL and PTMC.*

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# Revealing the (de)sodiation mechanism of antimony chalcogenides $Sb_2X_3$ with *operando* XRD

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Na-ion batteries (NIBs) are emerging as an environmental-friendly and sustainable alternative to Li-ion batteries (LIBs). [1] There are many challenges associated with the commercialization of NIBs, particularly in the development of high-performing anode materials. A class of materials that are promising anode materials in NIBs base its mechanism on the combined conversion-alloying reaction mechanism. [2] However, conversion-alloying materials (CAMs) typically undergo a set of complex structural and chemical transformations during operation (new phases, amorphization, *etc.*). [3] Investigation of the cycling mechanism using advanced characterization techniques, such as *operando* X-ray diffraction (XRD) is therefore key to opening the Pandora's box of these battery mechanisms.

Among CAMs, Sb-based chalcogenides ($Sb_2X_3$, where X = O, S, Se, Te) have demonstrated superior electrochemical performance in NIBs. $Sb_2X_3$ materials have previously been synthesized to obtain various nanostructures architectures and tested in NIBs. However, worth noting is that the capacity of these materials refers to a special arrangement of the materials in specific combinations of electrolytes, binders, conductive additives, counter electrodes, *etc.* Therefore, there is a need to test these materials under the same conditions to evaluate their performance and operation mechanism in NIBs. Herein, the primary research question is to understand how the size and shape of the $Sb_2X_3$ materials influence the capacity, cycling performance and their lifetime as the active materials in NIB's anodes under the same conditions, along with revealing the (de)sodiation mechanism. The home-lab *operando* XRD of $Sb_2Se_3$ (Figure 1) shows that the (de)sodiation mechanism of this material is indeed quite complex, as it passes through multiple phases (NaSb, $Na_3Sb$, Sb), where some of these phases even are amorphous.

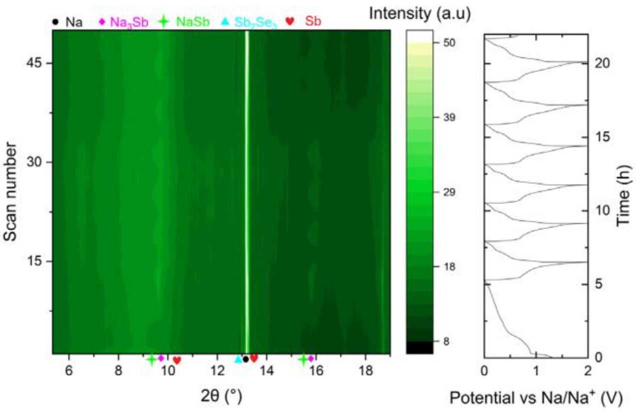

***Figure 1.*** *Operando XRD of $Sb_2Se_3$*

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# High-purity Alumina from Industrial Slag; Material Processing and its Use in Secondary Li-ion Batteries

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Ceramic coatings of separators in conventional Li-ion batteries are implemented to stabilise the system to short-circuiting, which typically results from puncturing of the separator by Li metal dendrite formation. [1, 2] High-purity alumina (HPA) is currently the most widely used choice for separator coatings. HPA is commercially produced by reacting aluminium with alcohol to form alkoxide that is further hydrolysed to obtain the final product, a process which is both highly energy demanding and expensive. [1-3]

This poster will present a possible pathway for obtaining alumina from a Si/Al industrial slag mix developed within a valorisation project funded by EIT RawMaterials (Fig. 1). Further purification steps and calcination of the aluminium-based by-product aims to produce HPA for use in Li-ion batteries, ultimately maximising the utilisation of the slag while also presenting a potentially less energy intensive pathway to obtain HPA.

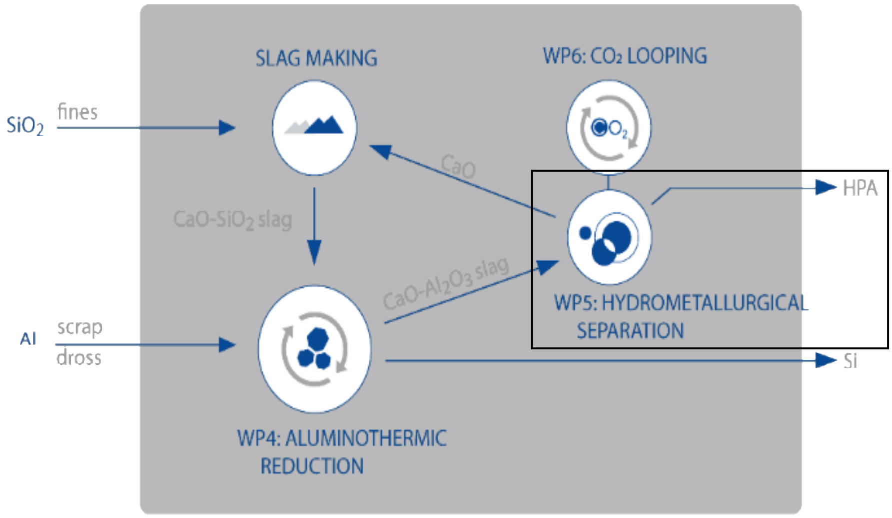

***Figure 1.*** *Schematic presenting the industrial Si/Al slag valorisation project, resulting in Si and high-purity alumina (HPA), where parts of WP5 is what the poster will present.*

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# Iron and Titanium Complexes and their Redox Activity and Stability in Aqueous Solutions for Flow Battery Applications

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A sustainable future is achievable through further utilization of renewable energy sources, such as solar energy. The intermittency of renewable energy sources is a problem that fossil fuels do not have. Cost-efficient energy storage integrated into renewable energy plants can solve this problem. Cost-efficiency can be achieved by using sustainable materials, like abundant metals (Fe and Ti) complexed with cheap organic ligands to form redox-active species to be used in flow batteries as energy storage materials. These materials should also be sustainable and water-soluble to increase the sustainability and cost-efficiency of the final energy storage.

Organic ligands can be used to tune the redox potentials of the metal complexes to obtain a suitable potential. The redox potential of $Fe^{3+}/Fe^{2+}$ can be shifted from +0.77 V *vs.* SHE to +1.03 V *vs.* SHE when complexed with bipyridine [1], and redox potential of $Ti^{4+}/Ti^{3+}$ ($TiO_2$ in acidic conditions) from -0.50 V *vs.* SHE [1] to -1.16 V *vs.* SHE when complexed with 2,3-dihydroxynaphthalene in aqueous solutions. Adding electron-withdrawing groups to ligands increases the formal potential by making the atmosphere of the metal center more positive, whereas introducing electron-donating groups to the ligand structure decreases the redox potential. Ligands and counterions also affect the stability and solubility of the complexes, which affect the performance of the complexes in flow batteries and should be known. Solubility can be increased for example by designing the structure of the complex according to Carnelley´s rule [2], like in a recent study where $[Fe(II)(bpy)_3]^{2+}$ with symmetric and asymmetric structures aiming for flow battery applications were studied [3].

We studied tris(2,2´-bipyridine) derived Fe(II) complexes with different 4,4´-positioned substituents in aqueous solutions and titanium(IV) complexes with catechol derived ligands. (**Fig 1.**) Redox activity was studied via cyclic voltammetry and electrochemical stability with chronoamperometry. The stability of the complexes was observed via color changes and precipitation in the solutions, and values for decomposition rates were obtained via simulations of the cyclic voltammograms, where also chemical reactions taking place to the redox-active species were considered accordingly.

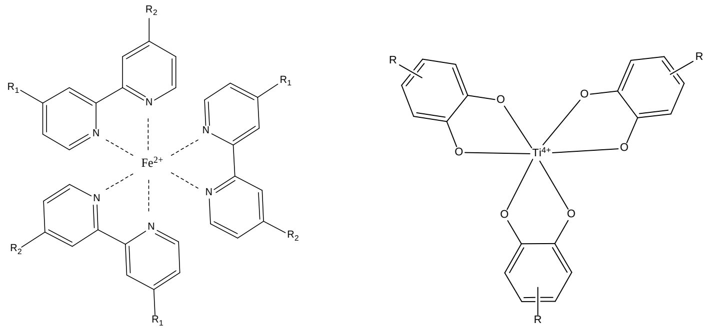

***Figure 1. On the left:** Fe-bipyridine with substituents $R_1$=$R_2$, **on the right:** Ti complex with catechol derivation.*

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# Nordic Battery EDU Belt – Buckle Up

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The lithium-ion battery markets are expanding rapidly both in the fields of electric vehicles and stationary energy storage. The Nordic countries are answering this challenge with the formation of Nordic battery belt including companies such as Northvolt, Umicore, Keliber, and FREYR Batteries. A completely new industry requires new competencies that are not currently widely taught in higher education. The need for skilled employees was already highlighted in projects such as Albatts[1]. We (Vaasa University of Applied Sciences, University of Oulu, Norwegian University of Science and Technology, and Uppsala University) have established a purely Nordic education battery belt (Nordic Battery EDU Belt, **Fig. 1**) that is an inclusive network for higher education.

The forming partner institutions have a long background, expertise, and complementary competencies in the battery value chain from cradle to gate. We consider the battery value chain to include mining and metal refining, precursor and active material synthesis, battery cell manufacturing, electrochemical characterization and testing, logistics, applications, economics, second use, and recycling to the full circular economy. UOulu has a strong background in mining, metal refining, precursor and cathode active material synthesis, and recycling. NTNU has strong expertise beginning from active material synthesis, characterization of materials and cells, recycling, and integration of batteries in the energy system. Recently, activities also include battery manufacturing. UU is established in active material synthesis, battery manufacturing, (electrochemical) characterization, and testing, to applications. While VAMK has long traditions and cooperation with the energy technology industry and in the applications and economics part of the battery value chain.

This project was funded and began in May 2022 and the long-term goal of the network is to ensure skilled employees for the battery industry in Nordic countries but also to create joint specialization courses, facilitate teacher & student mobility, and organize education-focused events for a wider audience. The objective is to have tight connections with the industry and actively listen to the demands of the developing battery industry. The Battery EDU Belt website and community aims to establish itself as a main hub for the Nordic battery news, events, education, community, and career center.

***Figure 1.*** *Logo of the network.*

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# Towards novel polymer-ceramic composite electrolytes for Li-based solid-state batteries

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Deep understanding of the ionic transport mechanism in solid-state electrolytes is vital to enable a market for solid state batteries. Polymer and ceramic electrolytes have been extensively investigated separately, and their ionic conduction mechanisms are already well-defined, respectively.[1] Recently, composite electrolytes have received tremendous attention thanks to the possibility of fusing the advantages of polymer and ceramic compounds in one system.[2] Interestingly, PEO has been favored for the greater part in the studies of composite electrolytes. However, alternative polymers have been proposed, and can be regarded as potential polymer hosts for the fabrication of ultimate hybrid electrolytes.[3] Numerous inorganic materials have been tested as ceramic fillers together with polymers. The research community of solid-state electrolytes has classified them into two categories depending on their $Li^+$ conduction ability.[4] These hybrids are still considered as a novel category of solid-state electrolytes, and they lack a deepened knowledge on their fundamental chemistry, preventing their fast implementation in electrochemical devices.[5] In this work, the effect and the nature of ceramic fillers (active vs inert particles) on the ionic conduction mechanism of polymer-based composite electrolytes are investigated. Fully-amorphous poly(trimethylene carbonate) and semi crystalline poly(ε-caprolactone) are employed as polymer matrices for this study.

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# Using the Novel Tools of 'PDF in the cloud' for Pair Distribution Function Analysis: the Case of $TiO_2$-Bronze Nanocrystals for Li-ion Batteries

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Total scattering and pair distribution function (PDF) analysis allows one to study the material structure even for non-crystalline materials, whether they are nanocrystalline, disordered, or even amorphous. This goes for *ex situ* as well as *operando* studies, where the latter allows one to study material properties during operation. Recently, the website 'PDF in the cloud'[1] (PDFitc, pdfitc.org) has been offered to assist PDF analysis through various apps. In this work, the structureMining[2] app has been used for phase identification of *ex situ* PDF data to obtain starting models for quantitative PDF analysis using the DiffPy-CMI[2] software, the similarityMapping app has been used to study similarity and reversibility for *operando* data through Pearson cross-correlation, and the nmfMapping[4-5] app has been used to study the number of components (phases) needed to describe *operando* data through non-negative matrix factorization (NMF). The science case presented here is about $TiO_2$-bronze nanocrystals that have been synthesized approximately 3, 5, and 7 nm in size[6]. The 3 nm nanocrystals have been incorporated into a Li-ion battery and the material evolution during Galvanostatic cycling is studied in an *operando* total scattering combined with PDF analysis. The analyses of both the *ex situ* and *operando* PDF data are highly assisted by the novel tools of PDFitc to elucidate the structural properties of pristine and chemically lithiated materials as well as the structural evolution during Galvanostatic cycling in a Li-ion battery.

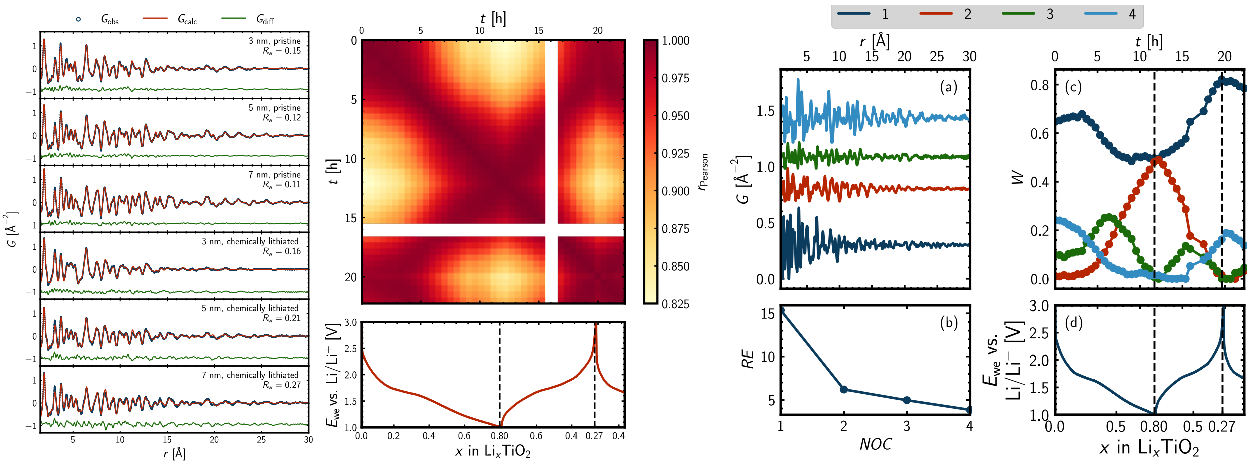

***Figure 1**. **Left**: Fits of ex situ PDF data for pristine and chemically lithiated samples using DiffPy-CMI[2], where the starting models were obtained from the structureMining[3] app at PDFitc[1]. **Middle**: Pearson cross-correlation matrix of operando PDFs during Galvanostatic cycling (below) using the similarityMapping app at PDFitc[1]. **Right**: Non-negative matrix factorization (NMF) analysis for operando PDF data during Galvanostatic cycling using the nmfMapping[4-5] app at PDFitc[1].*

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# Electrochemical Properties of 3D Printed Graphite/Nylon Electrodes

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Additive manufacturing (AM), known as 3D printing, is heavily involved in various industrial sectors due to the design freedom, low-quantity economy, material efficiency, and predictable reproducibility [1]. However, the potential of additive manufacturing for the material design and fabrication of composites, meeting the desired architecture and properties, has not been fully realized [2]. Therefore, the 3D printed graphite/nylon electrodes manufactured by selective laser sintering (**Fig. 1**) have been examined by various electrochemical techniques to launch the initiative and estimate the prospects of this composite material to be used as freestanding electrodes in electrochemical energy storage systems.

The 3D printed electrodes reveal their applicability for electrochemical applications. The electrodes demonstrated the capacitive behaviour in acidic, neutral, and alkaline electrolytes. The outstanding cyclic stability of the 3D printed graphite/nylon electrodes with an average 95% retention of specific capacitance was observed during the experiments. The advantages of the choice of selective laser sintering for electrode printing were confirmed by the material characterization study that represented the retention of the individual properties of the electrode components, providing the conductive network among graphite particles. The present study is a promising step toward the implementation of AM technologies in the production of electrochemically active materials.

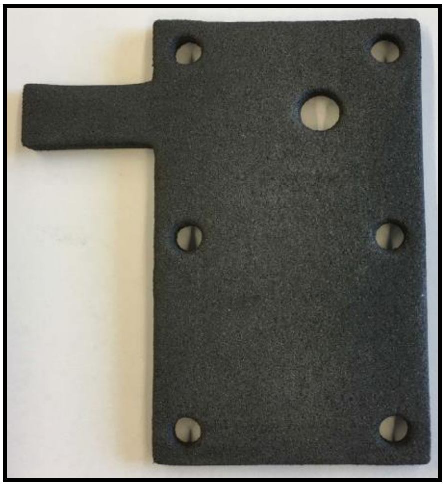

***Figure 1.*** *Digital image of the 3D printed graphite/nylon electrode.*

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# Screen Printed $LiFePO_4$ (LFP) Cathode Development For Solid State Li-ion Batteries

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Li-ion batteries (LIBs) are currently the best-performing technology for energy storage devices but suffer from safety issues due to organic liquid electrolytes. Switching from liquid-state to solid-state electrolytes (i.e., transition towards to Solid-state batteries (SSBs)) is a possible solution to overcome the safety concerns.[1][2] Solid-state batteries are expected to be a game-changing technology for accelerating the popularity of automotive sectors. They offer higher energy density than conventional LIBs, significantly shorter charging time due to superior charge and discharge performance, and lower costs of using less expensive materials. Apart from that, there are still some challenges that arise in selecting appropriate materials and processing methods. However, to pave the way for large-scale application in SSBs, a suitable advanced material needs to be produced and processed in large quantities to fabricate battery cells. Therefore, we have developed a printing-based approach for LFP cathodes (Figure 1) and compatible with SEs for SSBs applications. Manufacturing techniques based on printing are promising for solid-state LIBs fabrication.[3] LFP is a widely known cathode material for its low cost and good thermal stability, which is one of the main cathode materials used in current commercial products of LIBs. Composite solid-state electrolytes (CSE) could be a balanced and feasible approach to high-performance electrolytes thanks to bringing together the advantages of polymer and ceramic electrolytes. Screen printing is a mature technology in other sectors such as printed circuit boards and is easily scalable for developing thicker catholyte for SSBs. The optimization and design of LFP and composite SEs are still underway, of which ink formulation, ion conductivity, and stability are essential. With these results, we hope to improve the availability and usability of screen-printed LFP-based cathodes for large-scale production of SSBs applications.

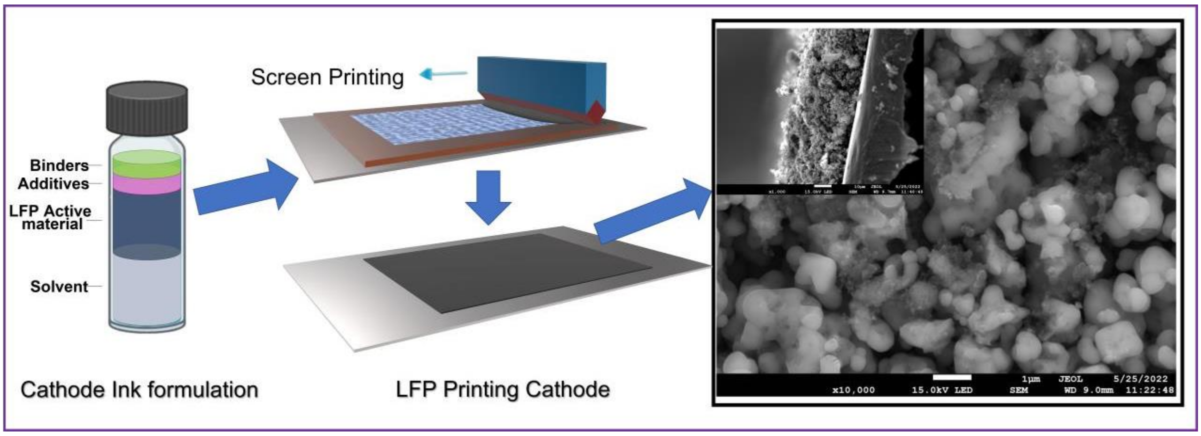

*Figure 1. A schematic of ink formulation for LFP cathode followed by the screen-printing approach to print the cathode layers. The printed cathode layer is shown in the SEM morphology of LFP particles covered with solid electrolytes (LLZO) and a binder (PVDF).*

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# POWER COAST – Nordic Growth Energy

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Two cities on the eastern coast of Finland, Kotka and Hamina, the development company Cursor and the companies and communities in the region are committed to creating and ensuring the success of the area's battery industry and emerging battery cluster. The Power Coast is an area powered by inexhaustible energy and pure willpower. This coast has been shaped by the unique heritage of process industry, a world-class port, and a strong service-minded attitude, and it is now ready to welcome the future players of the battery industry.

In late 2021, Finnish Minerals Group and technology company CNGR Advanced Material announced their plan to establish a new joint venture company to build the pCAM plant that will produce precursor material in Hamina. **Thorsten Lahrs** from Germany has been appointed as the CEO of the joint venture company and will be working mainly from Finland.

The new local plan made by the City of Kotka for the Keltakallio industrial area became legally binding in the winter of 2021. The preliminary agreement to be signed concerns an area of 27.8 hectares situated in Keltakallio. The site can be expanded by 46 hectares later, if necessary.

As the preparations progress, Finnish Minerals Group wanted to hear Kymenlaakso residents' thoughts on the plants. According to a survey commissioned by the company, 95% of the residents in the region are of the opinion that Europe should increase its self-sufficiency in batteries and battery material production.

Kotkan Energia Oy and Nordic Ren-Gas Oy have signed a co-operation agreement regarding the feasibility study of a Power- to-Gas facility producing both renewable methane and green hydrogen.

Welcome to the coast that has the power to make visions come true. [1]

***Figure 1.*** *Power Coast is located in the Southeastern coast on Finland.*

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# Lithium Bis(fluorosulfonyl)imide (LiFSI) as Electrolyte Salt for Electrochemical Capacitors

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The salt LiFSI has shown to exhibit superior electrochemical performance in Li-ion battery chemistries compared to the conventionally used salt $LiPF_6$ [1,2]. Due to its high electrochemical stability window, low susceptibility to hydrolysis, and beneficial safety aspects, LiFSI also is a promising candidate for electric double layer capacitors (EDLCs) and Li-ion hybrid capacitors (LICs). To evaluate the performance for use in capacitors, the interaction of LiFSI with activated carbon, a commonly used active material in capacitive energy storage devices, has been studied. To reveal the long-term cycling behavior, constant voltage hold tests in half-cell configuration have been conducted at selected positive and negative polarizations. Post-mortem analysis via X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM) and electrochemical impedance spectroscopy (EIS) provide complementary information on electrolyte degradation products and evolution of interlayer growth during accelerated aging. While capacitance retention is above 80% for positive potentials up to 4.0 V vs. Li/$Li^+$, electrochemical stability is restricted for negative polarization, possibly due to ion starvation effects and parasitic side reactions such as LiFSI reduction and degradation of the aluminum current collector. The use of carbon-primed aluminum current collectors successfully mitigates the extent of cathodic and anodic corrosion during ageing, in addition to reducing contact resistance between active material and current collector and enhancing the rate capability of activated carbon electrodes.

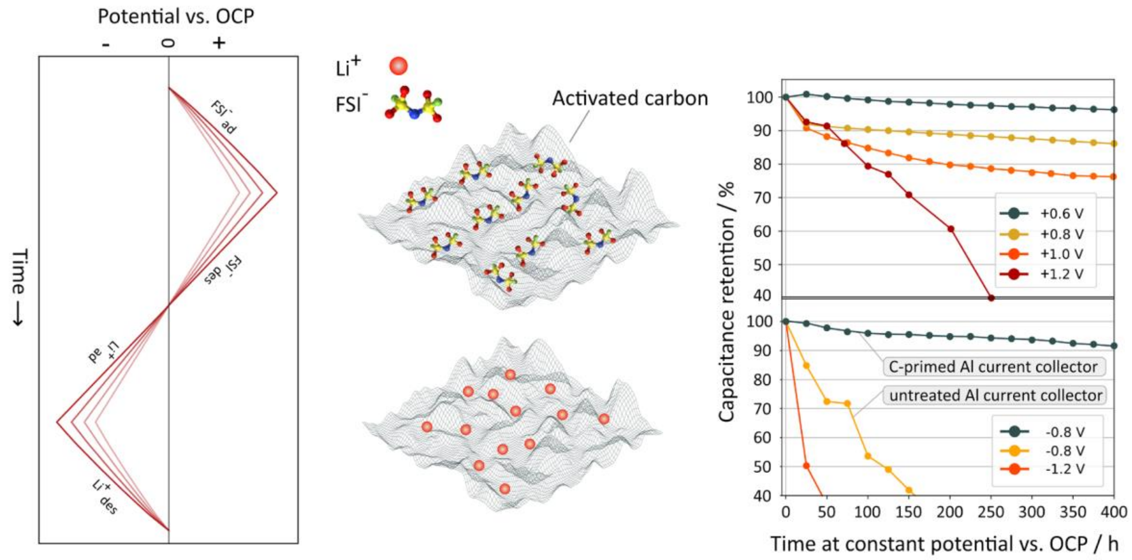

***Figure 1.*** *Voltage profile, illustration of ion adsorption and ageing stability of activated carbon in EC/DMC(1:1) + 1 M LiFSI.*

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# Environmental and resource performance of sodium-ion batteries based on prospective life cycle assessment

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The electrification of society is on-going, and rechargeable batteries are an important part of that transition as they are used in both electric vehicle and stationary energy storage applications. Today, lithium-ion batteries (LIBs) are extensively used much due to *e.g.* their high specific energy density, long cycle life and a continuous cost reduction. However, many LIB chemistries contain rare and/or critical materials, such as nickel and cobalt, in addition to lithium itself. Additionally, producing, using, and disposing LIBs generate environmental impacts in different ways [1, 2]. One promising next generation battery (NGB) is sodium-ion batteries (SIBs). SIB benefits include slide-in manufacturing processes with respect to LIBs and no requirement of any (or few) rare and critical elements [3]. Combined, this might give SIBs a vantage point from a resource and environmental point of view. To evaluate this, the environmental and resource impacts of a SIB have been investigated by prospective life cycle assessment. A flowchart of the studied system is shown in Figure 1. As SIBs are not yet mature, we scale up the production and include scenarios for possible future developments, and furthermore we compare the results to those for LIBs as well as another NGB – the lithium-sulfur battery. With this we hope to identify hotspots to guide SIB development towards reduced impacts.

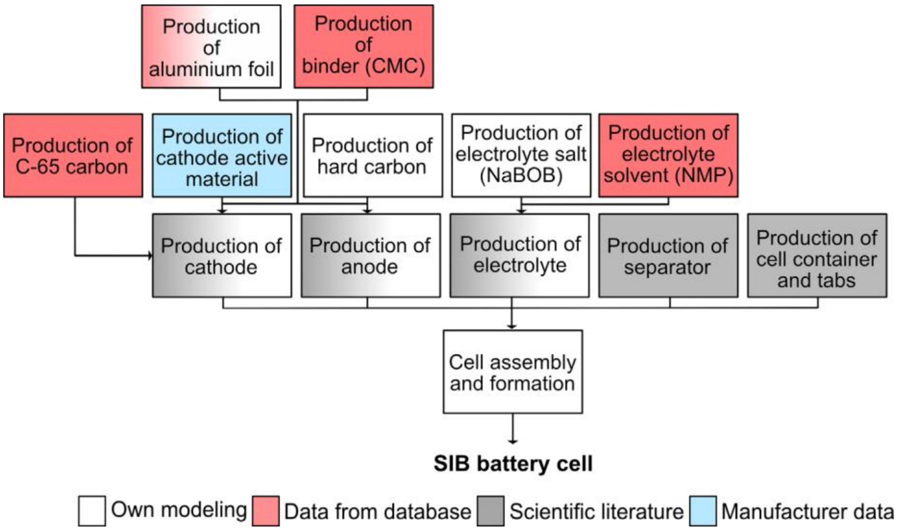

***Figure 1.*** *Initial flowchart of SIB production including data sources.*

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# Prospective life cycle assessment of a promising solid-state battery

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Rechargeable battery technologies have received a lot of attention lately. Much of this interest stems from the ambition to expand electric car fleets and grid-storage capacity to offset the intermediate nature of renewable electricity production and reduce greenhouse gas emission. The dominant battery technology today is the lithium-ion battery (LIB), which is associated with several sustainability and safety issues, such as the content of the critical and/or geochemically scarce metals lithium, nickel and cobalt.

The solid-state battery (SSB) is a novel battery technology utilizing a solid electrolyte instead of a liquid one. SSBs are expected to become a future competitor in the battery area due to the potential for increased energy densities, safer uss and higher cyclability. In this work the SSB literature was first surveyed to identify a promising SSB system that had not been studied from a life-cycle perspective. The battery chosen was an argyrodite-type SSB with an “anode-free” design, containing a silver-carbon nanoparticle layer to promote even lithium plating when charged [1]. Figure 1 shows an illustration and the main components of the studied SSB.

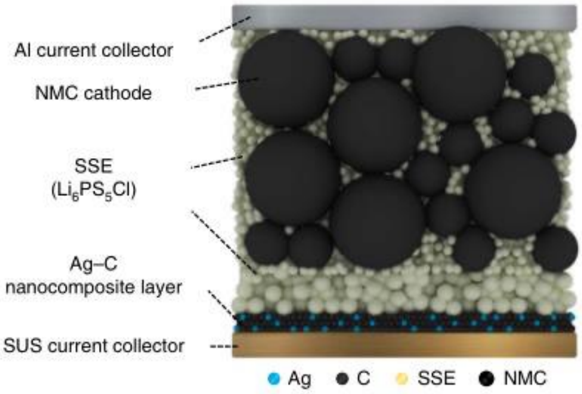

***Figure 1.*** *Cross section of the chosen argyrodite battery. Obtained from [1]*

To identify environmental hotspots and compare the life-cycle environmental impacts of this battery at a point in time where it has reached large-scale manufacturing, a prospective life cycle assessment (LCA) with a cradle-to-gate scope was performed. The functional unit was defined as 1kWh of storage capacity produced and the study focused on greenhouse gas emissions and mineral resource scarcity.

The prospective LCA concluded that at the studied SSB performed comparably to LIBs when it came to greenhouse gas emissions, although this depends on the type of energy supply employed during manufacturing. A hotspot identified was the energy-intensive production route for carbon nanofibres. Furthermore, the study showed that the battery performed better than LIBs when a more short-term indicator of mineral resource scarcity was used, but considerably worse for a more long-term mineral resource scarcity indicator. This indicates that the battery might be unfit for large-scale manufacturing unless the silver, which is the main contributor, could be reliably recycled or substituted. This was tested in a sensitivity analysis where it was shown that replacing the silver with magnesium would reduce the long-term mineral resource scarcity impact with 69%, the more short-term mineral resource impact with 29%, and the greenhouse gas emissions with 14%.

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# Self-Healing Binders for Silicon-Based Electrodes

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Silicon is a promising active material for anodes in lithium-ion batteries. However, it is well known that the compound contends with large volume changes, creating cracks in the material and therefore limiting the lifetime and capacity of the cell. In order to cope with these volume changes, this project aims to develop a self-healing binder that will improve the cycling stability of lithium-ion batteries.

To enable self-healing properties in a material, a variety of bonding types can be used. A notable example is hydrogen bonds that have weak interaction and therefore easily break and form. However, this high reversibility also renders poor mechanical properties. To provide the electrodes with high mechanical stability, dynamic covalent bonds can be used instead. These bonds also allow self-healing properties whilst having higher bonding strength, providing a more mechanically stable electrode [1-3]. Specifically, we have focused on different borate ester bonds [4] and coupled them with the polymeric binder poly(vinyl alcohol) (PVA). These borate ester groups have been synthesized and coupled to crosslink with the PVA, as well as other commercially available borate groups. The electrode slurry preparation with these different binder systems was shown to affect the electrochemical performance of the cells due to the presence of the dynamic covalent bonds. Two preparation techniques (ball milling and centrifugal mixing) were tested and the differences in cell performance, along with the impact on the polymer will be presented.

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# Fluorine-Free “Solvent-in-Salt” Sodium Battery Electrolytes: Solvation Structure and Dynamics

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Superconcentrated electrolytes for monovalent-ion batteries have attracted much attention in the recent decades. In 1985, McKinnon and Dahn [1] have reported on the saturated solution of $LiAsF_6$ in propylene carbonate (PC) as a promising electrolyte. Angell *et al.* [2] then proposed the concept “polymer-in-salt” and this new concept brought researchers out of the confinement of 1 molarity concentrated electrolytes. Superconcentrated electrolytes are extensively explored due to the unusual nature of their solid electrolyte interface (SEI) compared to the one produced by the traditional dilute electrolytes. The solvation structure, dynamics, and transport properties, as well as thermal and electrochemical stabilities of “solvent-in-salt” (SIS) electrolytes are not yet sufficiently understood. Here, we report a new class of non-fluorinated SIS sodium battery electrolytes composed of 50-90 mol% of Na[DEEP] salt in a oligoether functionalized trialkylphoshpate TEOP solvent. An increase in the ionic conductivities is observed as a function of salt concentration (**Figure 1**). In contrast, ion diffusivities decreased with increasing salt concentration. The analysis of the solvation structure and dynamics reveals a solvent-enriched phase (denoted as “the solvent”) with almost an order of magnitude faster ion diffusion than that of the salt-enriched phase (denoted as “the salt”). The higher aggregated ionic structures lead to enhanced ionic conductivities with increasing salt concentration alongside high thermal stability and better anodic stability. These systems provide the very foundation for fluorine-free “solvent-in-salt” sodium battery electrolytes.

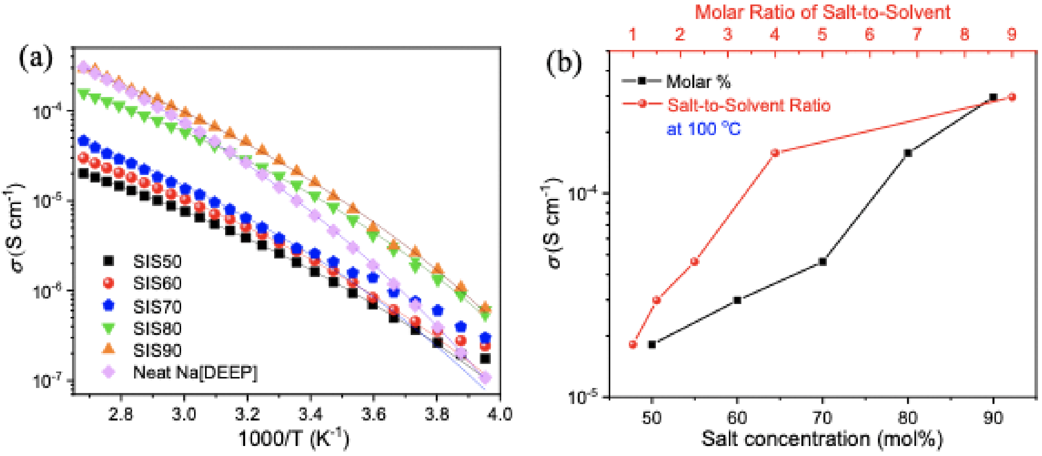

***Figure 1.*** *(a) Ionic conductivity as a function of temperature for the neat salt and the electrolytes. The solid lines represent the best fitting of ionic conductivity to Vogel-Fulcher-Tammann (VFT) equation. (b) ionic conductivity as a function of salt concentration and molar ratio of salt-to-solvent for the electrolytes at 100 °C.*

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# Study of Aluminum Anode Behavior in Water-in-Salt Electrolyte from Various cell Assembly Variables for Aluminum Batteries

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From sustainability perspective, aqueous electrolytes are better alternative than ionic liquid electrolytes for aluminum batteries. However, challenge with inert oxide film on Al anode surface becomes more complex in contact with traditional aqueous electrolytes. A new sub-class of aqueous electrolytes, water-in-salt (WIS) of ($AlCl_3.6H_2O$), has got attention, recently. But Al anode behavior in contact with WIS electrolyte is unclear. In this work, a new polymeric type of coin cell is designed, which easily give a possibility to analysis the behavior of Al during electrochemical performance. An aluminum-graphitic cells using WIS electrolytes were assembled in polymeric coin cell. It was found that modification in cell variable, i.e., electrode distance, resulted to a more stable cycling performance, compared to pervious, author work [1]. However, corrosion product (SEI layer) that was gradually attached on Al surface during electrochemical performance, still cause a capacity decay over cycling. Moreover, metallurgical observation emphasized that $Al_3Fe$ intermetallic phases, which are always precipitated in the Al matrix are the favorite sites for the electrolyte to initiate the corrosion. In the other words, $Al_3Fe$ phases facilitate damaging the oxide barrier, and thus allow the electrolyte to contact the fresh Al surface. In general, the results have led to a deeper insight of Al anode metal behavior in contact with electrolyte for aluminum batteries.

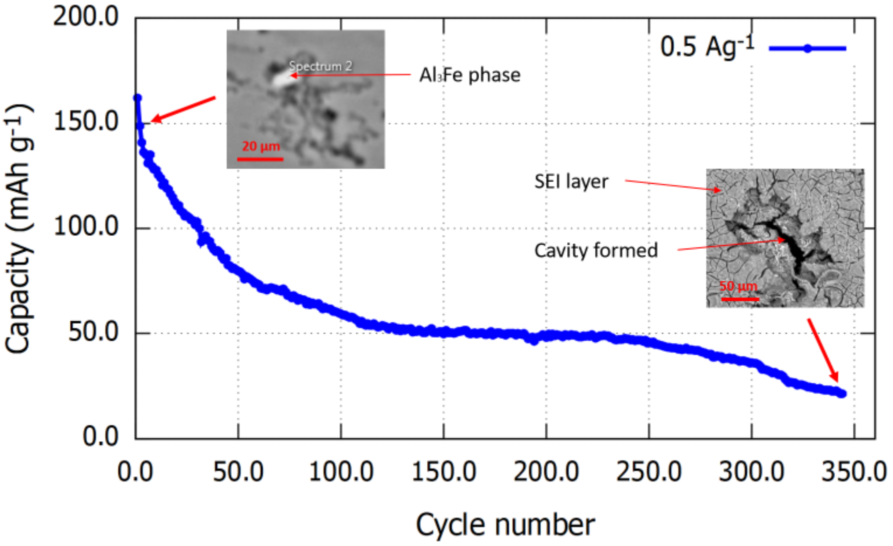

***Figure 1.*** *Cycling performance of aluminum/WIS/nanographite cell at a current density of 0.5* $Ag^{-1}$ *and a voltage range of 0.1-1.6V. Microstructure of Al anode from initial cycling showing corrosion initiate around* $Al_3Fe$ *phases, and from the last cycle revealing Al surface was covered with SIE layer and corrosion produced a cavity in Al.*

**References**

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# State-of-the-art non-flammable liquid electrolytes: performance and interfaces

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Battery safety, particularly regarding flammability, has come under increasing public and scientific scrutiny as lithium-ion batteries become ubiquitous in everyday life. Much effort is being put into formulating nonflammable electrolytes, as electrolytes are usually the most flammable components of a typical battery cell. In the past 20 years, a large number of nonflammable liquid electrolytes have been proposed. However, because they are usually tested under different conditions, it is difficult to rigorously compare them. In this study, a series of electrolyte candidates, consisting of hydrofluorocarbons or phosphates/phosphonates were tested under standardized conditions. To reflect the current state of the art, each electrolyte was used with co-solvents and/or additives that have been previously reported to improve performance. Core performance measures such as cycling data, rate capability and cell resistance were collected. A promising subset of the electrolytes were selected for further study; the compositions of the solid-electrolyte interphases in particular and their influence on the electrochemical properties are discussed.

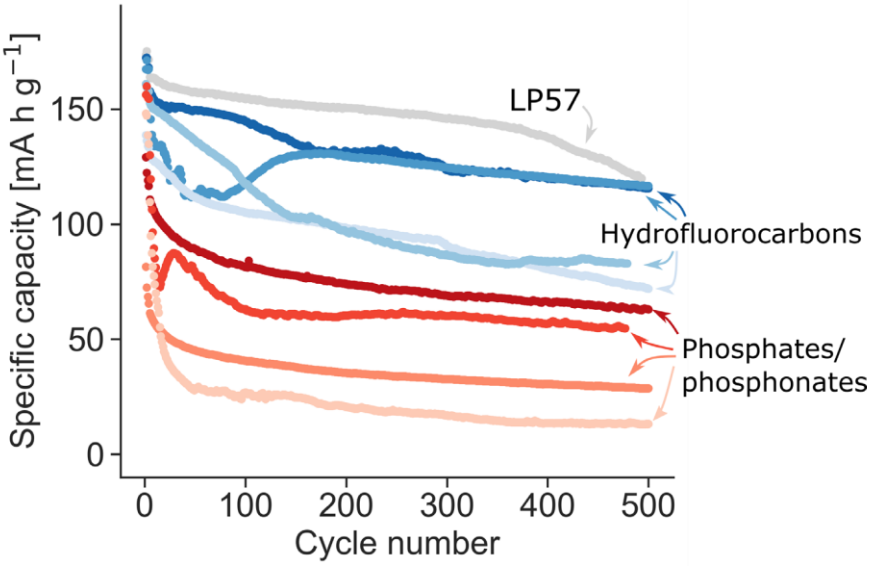

***Figure 1.*** *Cycling performance of selected non-flammable electrolytes, grouped into hydrofluorocarbons phosphates/phosphonates. To reflect the current state of the art, each electrolyte was used with co-solvents and/or additives that have been previously reported to improve performance.*

# Ceramic Interfaces: The Quest Of A Functional All-Solid-State Battery

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The challenges of all-solid-state batteries range from electrochemistry to mechanics. A functional inorganic solid state electrolyte will have to account of a high lithium ion conductivity; an adecuate electrochemical stability window; proper compatibility and contact with the electrodes and desireable dimensions for its use in batteries with proper gravimetric and volumetric capacities. That much struggle is worth several important upgrades, such as increased safety or the possibility of enabling lithium metal anodes.

To tackle all the abovementioned issues, interdisciplinary research must be carried out. At Ångstromlaboratoriet we carry this research focusing both on the bulk of LLZO, the most common oxide electrolyte, its interfacial properties and the assembly of functional cells with such complex electrolyte.

This presentation will present, mostly, the details of our proposed surface treatment to improve the performance of the LLZO electrolyte. Highly densified pellets are immersed in a Boric acid solution, targeting the formation of a boron-rich surface layer that improves both the physical contact and the electrochemical performance. Chemical, structural, electrochemical and morphological characterizations were carried out.

Briefly, other aspects researched by our group will also be introduced.

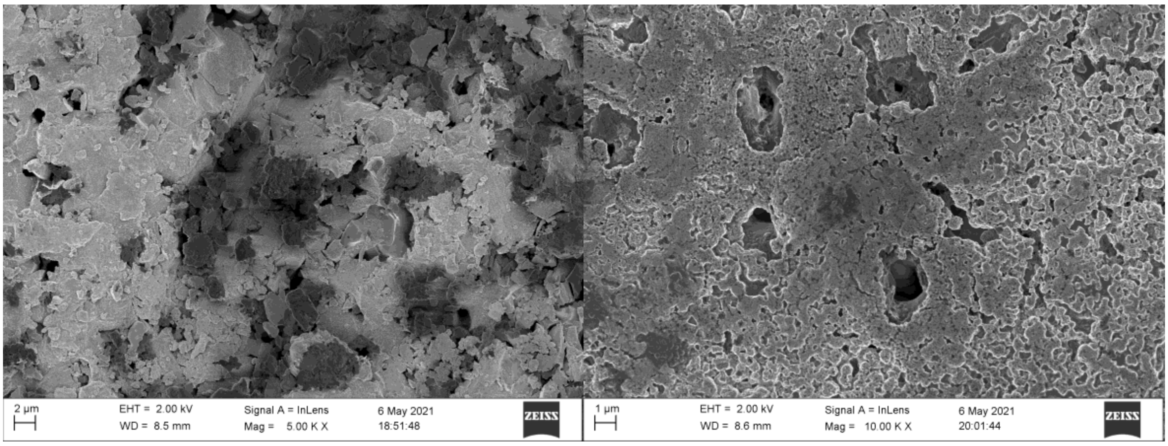

***Figure 1.** LLZO surface prior (left) and after (right) boric acid treatment*

# Spray printing as a suitable method of printing various design NMC cathodes

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Although state-of-the-art Li-ion liquid electrolyte batteries offer satisfactory performance (for now), the producers/customers indicate a need for more adaptable and sustainable methods of fabrication. Printing methods, specifically spray coating (Fig. 1a) belongs to one of the printing methods that allows exceptionally adjustable fabrication.[1] It allows effortless up- and down-scaling of the printing, high resolution (tens of µm) of the features, 3D compatibility, implementation of wide range of inks formulations, ability to introduce large particles (>5 µm), and full fabrication sustainability thanks to negligible material waste during printing.[2] Importantly, spray coating is also suitable for solid-electrolytes deposition.[3] In this study, the spray coating has been used to fabricate NMC cathodes: NMC523 ($LiNi_{0.5}Mn_{0.2}Co_{0.3}O_2$) and NMC88 ($LiNi_{0.88}Mn_{0.03}Co_{0.09}O_2$). For ink formulation, DMF has been used, replacing NMP, that besides being toxic, has a negative impact on the printer components. The fabricated cathodes have further been used to prepare half-cells and pouch cells for electrochemical characterization. The morphological analysis depicted in Figure 1b reveals that the spray coating results in cathodes of relatively low roughness (RMS roughness 3.4 µm, mean roughness 2.7 µm), before calendering. The SEM image (Fig. 1c) indicates that the utilization of spray printing enables the fabrication of high-quality and controllable precision NCM523 and NMC88 cathodes. The initial pouch-batteries cycling demonstrates 2.1% and 3.5% capacity fade after 100 cycles for NMC523 and NMC88, respectively (Fig 1d).

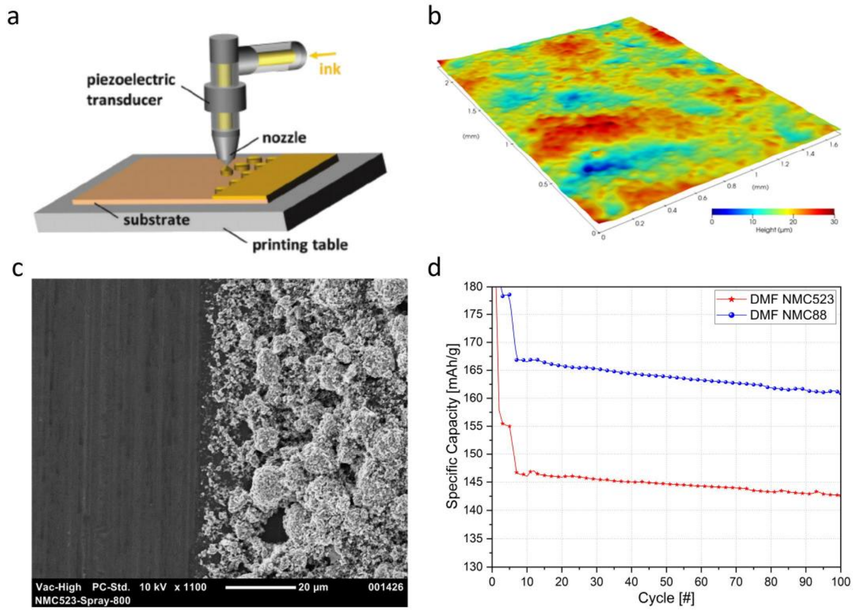

***Figure 1.*** *Spray printing of NMC cathodes. a) Simplified spray printing principles, b) Morphology of printed NMC523 layer, before calendering, c) Edge of the printed layer on the Al electrode substrate, d) Initial electrochemical performance of the pouch cells fabricated based on the spray-printed cathodes.*

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# Aluminum Oxide Coatings on NMC811 Investigated by Synchrotron Photoelectron Spectroscopy

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Aluminum oxide is a commonly used coating material for both battery electrode material and many other applications [1]. The coatings are often very thin to allow for electron conductivity by tunneling, and thus protect the bulk material without significantly deteriorating the electronic properties of the system. For battery applications commonly stated thicknesses are in the range of 0.5-5 nm, i.e. on the limit of yielding fully covering films [1, 2]. If the active material is fully covered, $Li^+$ have to pass through the coating and into the bulk during charge/discharge. The coating can then be expected to affect the $Li^+$ transfer resistance over the interface [2]. It has been debated if the aluminum oxide coating is lithiated into Li-Al-O, and if Al acts as a dopant to the bulk material [3]. Both the coating properties, such as resistivity, and the bulk properties could in those cases be affected during the charge/discharge cycle.

Here we present an initial study on aluminum oxide coatings deposited by atomic layer deposition (ALD) onto $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC811). Samples with different thickness of aluminum oxide is investigated and varied energies at a synchrotron are used to non-destructively gain information from different depths (see **Fig. 1**). These results reveal the initial growth and composition of the aluminum oxide layer on NMC811 and will also serve as a basis for future operando studies of coated battery systems.

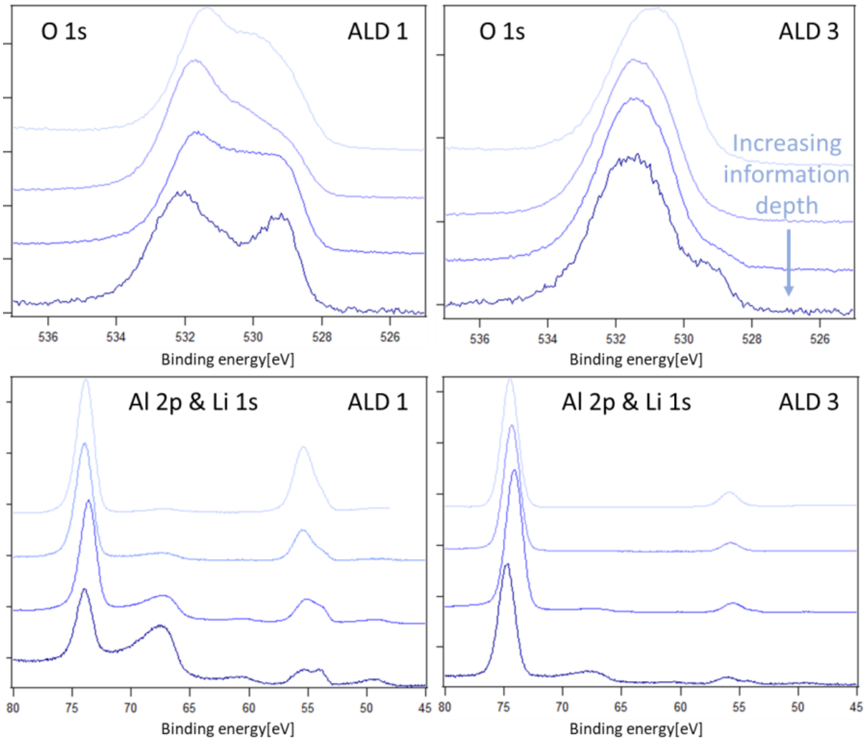

***Figure 1.*** *Spectra from four information depths on NMC811 samples coated with 1 and 3 ALD cycles of Al-O.*

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# Operando Raman Studies of Lithium Nickel Oxide Cathode Material

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$LiNiO_2$ (LNO) is considered a very promising and high energy density alternative to Co-containing Li-ion battery cathodes. It is also the benchmarked cathode material of the European BIGMAP project, which aim is to combine advance operando characterization, computational modelling and artificial intelligence in order to accelerate the research and development of new generation batteries. Unfortunately, the material suffers from severe irreversibility issues and parasitic reactions, related to Ni dissolution and oxygen evolution[1].

Operando Raman spectroscopy is an excellent tool for studying phase transitions in Li-ion battery cathodes.[2] It provides information on both molecular bonds and crystalline phonon modes, and sheds light on short-range order, cation/anion oxidation state and cation ordering. The penetration depth is estimated as top few hundred nanometers of five micrometers sized particle, therefore near-surface area is probed. To the best of our knowledge, no operando Raman data for LNO has previously been reported.

In this contribution we present an operando Raman study of LNO cathodes, whose cycling behaviour is investigated as function of cycle number, C-rate and upper cut-off potential. The results show a complex behavior of band positions and intensities during cycling (figure 1), corresponding to four phases transformations. The $E_g$ and $A_{1g}$ bands evolution is akin to those reported for other Ni-rich materials[3], and is analyzed and compared to the literature results, operando XRD/XAS data and computational studies from the BIGMAP project.

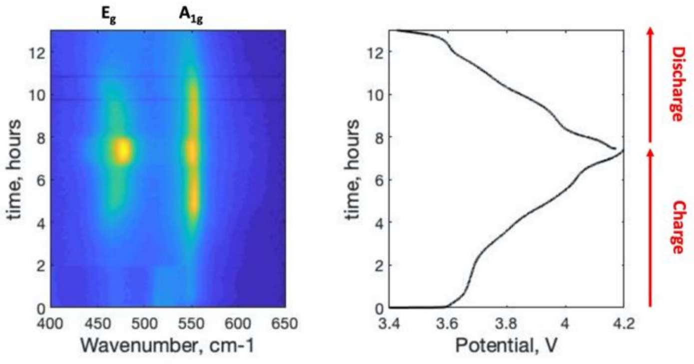

***Figure 1.*** *Operando Raman spectra of $LiNiO_2$ during cycling.*

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# Nonuniform Degradation of Lithium-Ion Batteries – 3D Modeling

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Global trends like electromobility and renewable energies have increased the demand for lithium-ion batteries significantly. Considering their specific requirements, numerical simulations are an important tool to provide optimal designs with shorter development time. The simulation of battery aging is of particular interest as experiments are time consuming and costly. During operation, various aging mechanisms contribute to the capacity loss of lithium-ion batteries [1]. Here, we focus on two of them: First, the solid-electrolyte interphase (SEI) growth caused by a parasitic side reaction that consumes cyclable lithium, and second, mechanical degradation under cyclic stresses caused by volumetric expansion/contraction during lithiation/delithiation. Fatigue cracking of the electrode surface leads to additional SEI formation and growth, further reducing the amount of cyclable lithium. Cracks may also cause a loss of electric contact between the active material and the current collector, leading directly to a loss of capacity.

The celebrated Doyle-Fuller-Newman model [2] serves as the basis of our thermo-electro-chemical modeling approach. We describe the electrode domain as macro-homogeneous continuum, characterized by the volume fractions of the individual components – electrolyte, active particles, binder, conductive aid –, the tortuosity, and the surface-to-volume ratio. The transport of lithium-ions in the binary concentrated electrolyte is a three-dimensional problem, while the transport of lithium in the unresolved active particles effectively reduces to a local one-dimensional problem. Linear elastic effects are considered at the particle scale. Standard Butler-Volmer and Tafel kinetics are used to model electro-chemical reactions. We solve the described problem using the multiphysics simulation software Simcenter Star-CCM+ [3], which we have extended accordingly.

While most studies consider single particle models or are restricted to one-dimensional cases, we perform three-dimensional aging simulations. This allows us to gain insights into the nonuniform effects of the degradation mechanisms as shown below (**Fig. 1**). We believe that our work helps to predict and understand the impact of particular design choices on the aging behavior and, thus, helps to create better designs for today's challenges.

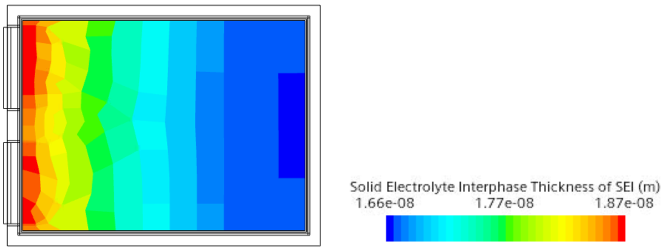

***Figure 1.*** *Spatial distribution of the solid-electrolyte interphase thickness in a simplified battery setup.*

**Acknowledgements** The financial support by the MODALIS[2] project, grant agreement No 875193 of the European Union's Horizon 2020 research and innovation program, is gratefully acknowledged.

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# Critical Evaluation of the Electrochemical Activity of Electrode Materials in Aluminum Batteries: The Crucial and Underestimated Role of the Current Collector

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Rechargeable Aluminum Batteries (RABs) based on $AlCl_3$-EMImCl ionic liquid electrolyte are among the most promising candidate for high energy storage devices/systems, due to the multivalent $Al^{3+}$- ion intercalation property. However, the greatest hamper of the current development of RABs is the many contradictions in the literature. Reported results on active materials for cathodes and metallic current collectors suffer from misinterpretations and make further research difficult. We were able to show that as-synthesized cubic $Co_3O_4$ and Hausmannit $Mn_3O_4$ spinel-type materials are electrochemically inactive in $AlCl_3$/ionic liquid RABs. The whole/entire performance is due to the reaction between the Lewis acidic electrolyte and the molybdenum (Mo) current collector. Since molybdenum is considered in the literature to be stable or rather its activity to be regarded as negligible, we could prove the opposite. Our results show three different oxidation states of the pristine molybdenum metal during the electrochemical process, which correspond to the work on the development of new novel current collectors for RABs. Since such side-reactions can show additional capacities of 150 mAh $g^{-1}$ to 400 mAh $g^{-1}$[1], we come to the result that it is necessary to exclude all possible Mo-sources in RABs.

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# Effects of The Ratio of Bio-based Carbon Nanotubes to Acetylene Black as Conductive Agents on The Properties of Li Ion Batteries

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Li-ion batteries (LIBs) nowadays have been used widely for applications such as the electric vehicles, portable electronic devices, and stationary energy storage industries because of their high energy and power density and reliable lifetime [1]. However, the current energy density and power density of LIBs less meet the performance required by portable electric devices and EVs. To meet these requirements, the loading amount of the active materials is required to be increased. However, an increase in the active material results in an increase in the thickness of the electrode, which in turn increases the internal resistance of the electrode, and thus decrease the battery performance. To overcome these issues, acetylene black is a commonly used conductive agent to reduce the internal resistance of the electrode and provide electronic pathways in the electrodes through "point-to-point" contact between acetylene black and active material particles; however, it is disadvantageous because of 0D material characteristics. On the other hand, carbon nanotubes (CNTs) are promising material as a conductive agent for reducing the electrode's internal resistance due to their high aspect ratio and forming three-dimensional (3D) conductive networks by the "line-to-line" contact with other CNTs and "line-to-point" contact with active material particles [2] [3]. On the other hand, recently, the usage of renewable feedstocks as a carbon source have been developed in the manufacturing of carbon nanotubes. This new development is receiving much attention among the global research communities due to the reduced carbon footprints, positive environmental impacts, and economic benefits [4].

In this study, acetylene black and bio-based carbon nanotubes were used together as conductive agents to increase the electrode performance for high power application. To find the optimum composition, CNT to acetylene black ratio was studied in the range of 15 to 35% for different amount of PVDF in the range of 0.8 to 1.4%, by keeping active material (NMC622) amount constant at 98%. Morphology observation showed that CNTs are well dispersed across the electrode. They are uniformly spread over the entire surface of individual primary NMC particles. The electrical resistivity was measured by four-point probe method, indicate that increasing the amount of CNT resulted in an increasement in the electrical conductivity. By increasing the amount of CNTs up to 30%, higher rate performance, and better cycling performance were achieved, but by reaching to 35% amount of CNT the electrode showed a poor performance, which more likely is because of forming aggregates and non-uniform distribution that can result in a weak conductive network.

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# Phase transformations in layered oxides for Na-ion battery electrodes elucidated through *operando* X-ray scattering

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Na-ion batteries (SiB) are interesting alternatives to Li ion batteries, due the easy access, high abundance and low price of sodium. However, for the SiBs to be truly competitive, development of novel electrode materials with high capacity and good capacity retention is crucial. For this purpose, layered transition metal oxides NaxMO2 (M = transition metal) have received a lot of attention as Na-ion cathodes, since they offer high electrochemical potentials and high capacities.[1] In addition, there is a strong incentive to move towards cheaper and more environmentally friendly transition metals for these materials, such as use of iron and manganese and thereby limit the use of e.g. cobalt.

For the layered NaxMO2, especially two polymorphs are of interest: The so-called P2 and O3.[2] Both polymorphs undergo multiple phase transitions under extraction of sodium, some of which are irreversible. The polymorphs can be co-synthesized by varying the Na-content in the pristine material and thereby the benefits of both structures, such as better structural stability and higher capacity, can be utilized.[3-6].

In this study a series of phase pure and biphasic P2/O3 NaxMO2 (M = Ti, Fe, Mn, Ni) electrodes are investigated by *operando* synchrotron X-ray scattering with the aim to extract relations between their structural behavior and electrochemical properties during charge and discharge in SiBs. *Operando* powder X-ray diffraction (PXRD) reveals that changing the chemical composition of the transition metals, e.g. by Ti or Li substitution, alters the mechanisms behind the phase transitions upon electrochemical extraction of sodium. Furthermore, the *operando* measurements shows that all phases disorder either partially or completely at high voltage, a process which is reversible upon discharge. The voltage at which the disorder occurs depends on the chemical composition of the materials and the P2/O3 ratio. To investigate the nature of the disorder, X-ray total scattering with pair distribution function (PDF) analysis was used.

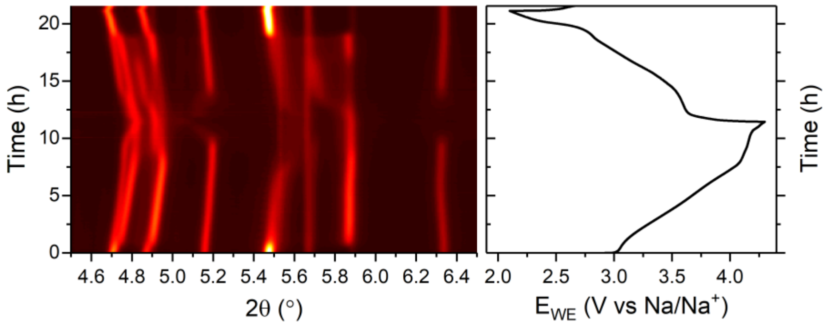

***Figure 1.*** *Operando* PXRD measured for $Na_{0.78}Ni_{0.2}Fe_{0.38}Mn_{0.42}O_2$ with the scattering angle as a function of time (left) and galvanostatic cycling (right).

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# Anionic Gamma-Aminobutyric Acid-functionalized Naphthalene Diimide for Use in Aqueous Organic Flow Batteries

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Aqueous organic flow batteries (AOFBs) are one of emerging electrochemical systems for large scale energy storage systems. One attractive molecule that has recently gained attention towards itself to be used in AOFBs is a family of organic molecules known as 1,4,5,8-naphthalene diimides (NDI). NDI is a molecule similar to quinone with two imide groups attached to a naphthalene core.[1] NDI undergoes two electron reduction reaction that makes it desirable as it can double the charge density of the RFB. NDI molecule has been studied for use in photovoltaics, sensors, semiconductors, electrocatalysts, and lithium-ion batteries, while it is not investigated thoroughly for utilization in AOFBs.[2] In this work, we synthesized GABA-NDI, an NDI molecule bearing negative charges with higher solubility of approximately 250 mM in 1 M ammonium chloride than the previous negitively charged NDI molecules in aqueous solutions and studied its performance in a flow battery. Our battery was built using 10 mL of 200 mM GABANDI as negolyte and 25 mL of 200 mM ammonium ferrocyanide as posolyte in the buffered supporting electrolyte and exhibits an average voltage of 0.76 V. The flow battery consisting of the mentioned couple in a buffered electrolyte is stable for over 200 cycles and gave a coulombic efficiency of 99.96% and a high energy efficiency of 80.9% at 60 mA/cm$^2$. The high energy efficiency in the batteries is an indication of fast kinetics of the molecule and a low ohmic resistance of the cell. Morover, 95% of the theorictal charge capacity of the GABA-NDI was accessed when cycling the battery galvanostatically at 60 mA/cm$^2$. According to the cycling results, GABA-NDI has good cycling stability, but a higher solubility would be needed for it to be feasible in industrial applications. However, the one-step easy synthesis makes this molecule desirable for mass production at a low cost.

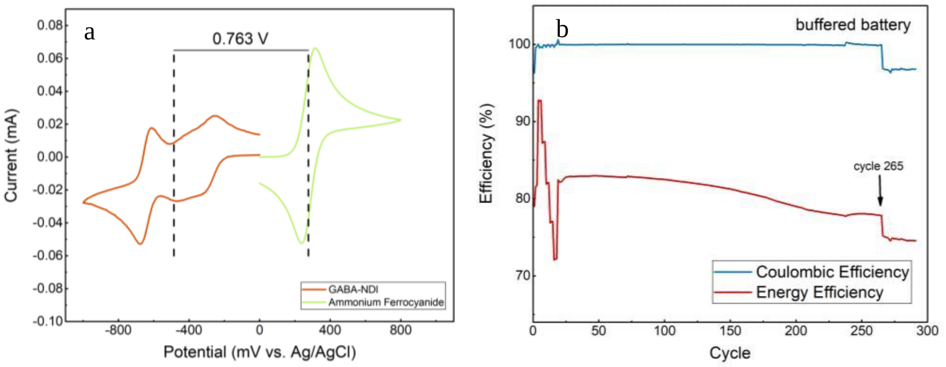

***Figure 1.*** *a) Average voltage between GABA-NDI and ammonium ferrocyanide. b) Coulombic efficiency and energy efficiency of the battery*

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# Lignin-Based Carbon Materials for Energy Storage Applications

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The expanding electrification of transportation and other industrial sectors increase the demand of different chemistries and materials in energy storage devices. Carbon materials are key electrode constituents in electrochemical energy storage (EES) devices, such as supercapacitors and lithium-ion batteries. Carbon is used both as active materials (activated carbons, graphite) and as additives (conductive carbon). However, most of those material streams are currently fossil-based or naturally mined constituting a major sustainability problem, which threaten the future of EES. More options for renewable carbon materials are therefore urgently needed to unsure the perennity and sustainability of the future EES solutions. Meanwhile, the increasing volumes of lignins from paper mills and biorefineries offer an unprecedent opportunity to design future lignin-based renewable carbon materials. The aromatic nature of lignin and its high carbon yield upon thermal conversion make it particularly attractive for these applications. Activated carbons, turbostratic hard carbons and semi-graphitized carbons can be synthesized from lignins following different thermal conversion strategies and used as electrode materials in energy storage devices (e.g. supercapacitors and Na-ion and Li-ion batteries). However, the botanical and process heterogeneity of lignins often translates into varying thermal reactivities producing carbon materials with variable properties and performance.

Design of Experiments (DoE) methodology has only recently become more recognized in the field of energy storage systems. Several different aspects can be studied using this tool, including electrode formulation, charge/discharge conditions and in our case, active material synthesis. [1, 2] Consequently, to better understand and control the variability between lignin-based carbon materials, we devised this systematic approach. This includes 1) the analysis of the thermal reactivity of lignins, 2) the in depth characterization of the lignin-derived carbon properties, such as their morphology, texture, nanostructure, and surface chemistry, 3) the use of chemometric tools for performance optimization and 4) structure-properties-performance relationship analysis. We have applied DoE on the synthesis methods shown in Fig. 1 to produce the suitable types of carbon materials for different energy storage applications.

***Figure 1.*** *Synthesis methods to produce different carbon materials for different energy storage applications from lignin precursor.*

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# Electrochemical and Stability Studies of Ferrocene-1,1′-disulfonate Dianion in Aqueous Redox Flow Battery Systems

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Energy policies are critical for governments to improve the environment and lessen their reliance on foreign energy sources. For this reason, renewable energy systems are receiving more attention than ever before. On the other hand, energy storage devices can ensure the long-term viability of renewable energies [1].

Redox flow batteries (RFBs) are one amongst the promising energy storage technologies for large-scale applications. Both energy and power can be easily adjusted from a few hours to days for storage depending on the application, which is an essential advantage for renewable integration of energy management [2-3].

Aqueous RFBs exhibit faster dynamics and higher ionic conductivity compared with non-aqueous RBFs. The comparatively low cost of aqueous electrolytes for large-scale energy storage applications compared to non-aqueous electrolytes adds additional competitiveness [4].

In this study, Iron (Fe) based Ferrocene-1,1′-disulfonate dianion (FcDS) was used to investigate its electrochemical properties as a posolyte with understanding the stability in different chemical environments. Although FcDS compounds were already reported in the literature, their advanced electrochemical and stability studies were not fully investigated [5].

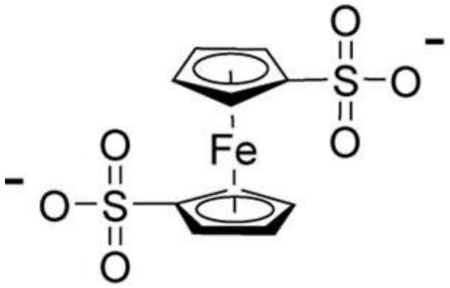

***Figure 1.*** The molecular structure of Ferrocene-1,1′-disulfonate dianion (FcDS)

FcDS is highly water soluble and environmental friendly Fe containing compound. During our electrochemical studies, we realized some solid precipitation formation were occurred and there was a unexpected decrease the redox capacity. Afterwards, we used different chemical environments as using several supporting electrolytes and additives to the solution to stabilize FcDS in longer term applications. Also, we used different analytical techniques (CV, EIS, NMR, UV-vis, etc.) to characterize the species and as well as understand the reason of the decomposition.

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# Comparison of carbon coatings on amorphous Si nanoparticles for lithium-ion battery anodes

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Swelling and surface stabilization of Si nanoparticles (NPs) remains a key priority for the implementation of high silicon content anodes in lithium-ion batteries for all types of applications. Carbon coating, owing to its accessibility, maturity, and suitability, is one of the most common coatings to both mitigate detrimental volume expansion and stabilize solid-electrolyte interphase formation. However, there are many potentially viable routes to carbon coating [1-3], resulting in great variation in the properties of the coating.

In this work, we compare two approaches to carbon coating of amorphous Si nanoparticles synthesized from a silane pyrolysis free space reactor (FSR), which provides mostly spherical Si NPs of a narrow size dispersion. The first approach involves in situ carbon coating using a second-stage hot zone in the FSR to pyrolyze acetylene. This gas-phase carbon coating approach resulted in carbon deposition but without a clearly identifiable coating layer. The second approach used dispersed the Si nanoparticles in a d-glucose based solution and carbonized the powder in an argon-filled furnace. This liquid-phase approach resulted in hard carbon-type of coating with a rougher and distinct morphology. Each of these nanoparticle samples were characterized with electron microscopy, then prepared into coin cells for half cell electrochemical testing. Carbon-coated Si NP contents of 20 wt% and 60 wt% solid mass in the slurry were also compared.

Gas-phase carbon coated Si NPs demonstrated higher rate capability than liquid-phase carbon coated Si NPs at high rates such as 2C or 7.2 $A/g_{active}$ (404 mAh/g vs. 343 mAh/g), but lower capacity at lower rates. For long-term cycling, the gas-phase carbon coated Si NPs showed a stable capacity of about 1,500 mAh/g for 50 cycles while the liquid-phase carbon coated Si NPs had a similar initial capacity but continuously degraded over time. However, this degradation was more gradual while the gas-phase carbon coated Si NPs experienced rapid degradation after its onset. Reducing the carbon-coated Si content to 20 wt% significantly reduced the rate capability and only showed beneficial improvement in stability after at least 100 cycles. These results indicate that the gas-phase carbon coating's morphology and carbon distribution could be tuned to influence particular cycling properties of a Si NP-based anode.

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# *Operando* X-Ray Tomography to Track Li Dendrite Growth on a Graphite Electrode

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**Application for a poster presentation.**

Lithium-ion batteries are currently the dominating energy storage technology, from portable electronics to electric vehicles, due to their tremendous energy density. One on the feature enabling this popularity is the use of graphite at the negative side. With its low potential *vs.* lithium (0.2V *vs.* $Li^+/Li$), high specific capacity of 372 mAh/g and good reversible Li ions intercalation; graphite has rapidly become the state-of-the-art negative electrode material. However, the low intercalation potential is both a bless and a curse because it facilitates the plating of lithium metal at the interface between the electrolyte and the electrode [1]. Lithium plating is a major issue in a battery for two main reasons: firstly, growth of dendritic structures that pierce through the separator can cause a short-circuit and catastrophic failure of the battery. Secondly, a part of the plated lithium might not be possible to strip, leading to capacity loss and poor coulombic efficiency.

Understanding how and when plating occurs is crucial to cope with the above issues. Thus, we have conducted an *operando* X-ray tomography experiment to directly monitor a Li/Graphite cell in terms of electrolyte composition, electrode morphology and current rate. The experiment was conducted at the ID19 beamline (ESRF, France) and we successfully scanned the battery while charging and discharging, obtaining information on kinetics, geometry, and location of Li-metal deposits. Tomography images are linked to the electrochemical data recorded in real time, making possible to gain a strong understanding of the mechanisms behind Li-deposition on graphite electrodes.

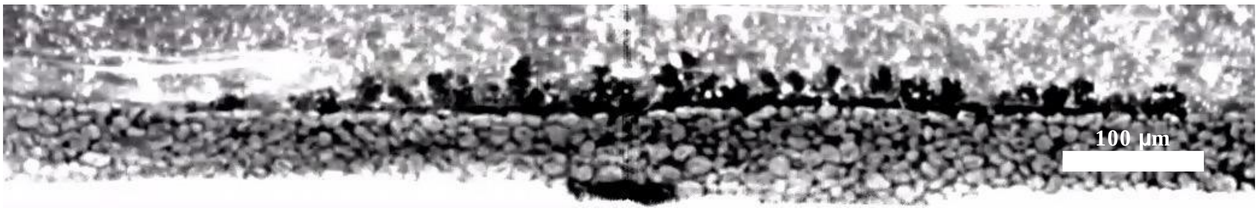

***Figure 1.*** *X-ray tomogram of a cross-sectional slice of a graphite electrode after a 1C lithiation at 100% SOC in LP57 +2%VC. It is possible to identify in light gray the graphite particles at the bottom and at the top, the fibers of the glass fiber separator in white. The black part in between is plated lithium in the shape of dendrites.*

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# Selective recovery of lithium via its carbonation in the thermal treatment of spent EV Li-ion batteries and water leaching

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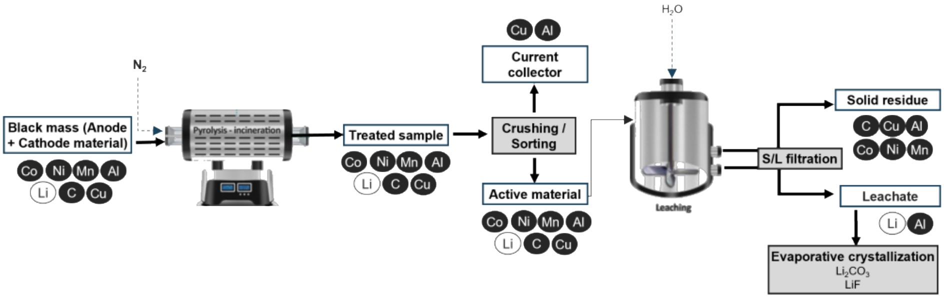

## Abstract

The development of electric vehicle is lead to a surge in the battery demand, as a matter of fact it has doubled in 2021 and the lithium nickel manganese cobalt oxide (NMC) cathode material is dominant in the market [1]. This has caused a significant increasing of the battery key metals demand, which have seen their prices raised, in May 2022, lithium prices were over seven times higher than at the start of 2021. The European Union reacted by including lithium among the critical raw materials in 2020 [2]. The need of efficient recycling technology for lithium-ion batteries remains an essential question in our modern world to recover the critical raw materials needed to support the future demand.

This project is focusing on lithium recovery, the goal is to develop a more effective technology where lithium will be recovered earlier in the recycling process avoiding the significant losses observed in the actual hydrometallurgical processes. The aim is to apply thermal pre-treatment on the black mass material (cathode and anode material). The carbon is utilized as a reductant, indeed the CO produced in the furnace will trigger the carbothermic reduction of the different oxides into Co, CoO, Ni, NiO, Mn, $Mn_3O_4$ under nitrogen atmosphere [3]. The lithium compounds are transformed into more soluble forms, mainly $Li_2CO_3$. This pyrolysis treatment is performed for a range of temperature between 500 - 800°C. Another main advantage of this treatment is the decomposition of the binder (usually PVDF) which allow an easier separation of the active material and the current foils (aluminum and copper). The challenge here is to maximize the lithium transformation before proceeding to its recovery with a water leaching step. The solution is next heated over 95°C, for its evaporation and formation of the $Li_2CO_3$. This technology has already showed promising results and a maximal lithium recovery of 62% was achieved after pyrolysis at 700°C for 60 min and a water leaching at room temperature with a solid/liquid ratio of 1:50 g/mL when the whole battery cell is processed. Under these conditions, the solution purity was 92% and only aluminum is leached with lithium. It is now important to understand that there is a limitation in the recovery; lithium fluoride is a co-product of the electrolyte decomposition and carboreduction. It is very lightly soluble, and its presence can explain the limitation. Moreover, it is also found in the crystallization product. Thus, tracking the behavior of the fluoride in the process is now considered as a key element for the understanding of the process.

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# Towards *in-situ* state of health monitoring of lithium-ion batteries using internal fiber-optic sensors

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We demonstrate a fiberoptic sensor for measuring the lithium-ion concentration inside a lithium-ion battery (LiB), live during charge and discharge. The goal is to monitor the health of the battery based on the dynamic and state-of-charge dependent concentration of lithium ions in the electrolyte.

Batteries lose capacity over time, both due to charging/discharging and to shelf ageing (calendar ageing). Extending the lifetime of a battery directly reduces its environmental footprint. This ageing is caused either by i) unwanted chemical reactions, reducing the number of available lithium-ions or increasing the impedance, or ii) by physical changes that reduces the available lithium storage capacity of the electrodes.

Generally, ageing is studied through monitoring the operational performance (e.g., coulombic capacity, internal resistance), or through post-mortem investigations. Alternatively, advanced equipment such as thermal neutron imaging or X-ray tomography has been used to take snapshots of the internal composition of the battery [1,2]. In this study we propose to monitor and study the health of LiBs through internal measurements using fiber-optic sensors, specifically through live measurements of the internal lithium concentration.

The intrinsic properties of optical fibers (e.g., small, chemically inert, electrically insulating) make them ideal for internal sensing in the chemically harsh interior of a battery. Multiple sensors can also be produced on a single fiber, enabling multi-point or multi-parameter sensing with only one entry point in the cell. Alternatively, the whole fiber can be used as a sensor, giving a continuous measurement along the fiber. In this study we employ a simple probe able to measure the lithium-ion concentration *in operando*.

The fiber is placed in the electrolyte between the electrodes, see Fig. 1a. Because the reflected intensity depends on the concentration of lithium ions (Fig. 1b), we are able to demonstrate and analyze how the concentration changes during cycling (Fig. 1c).

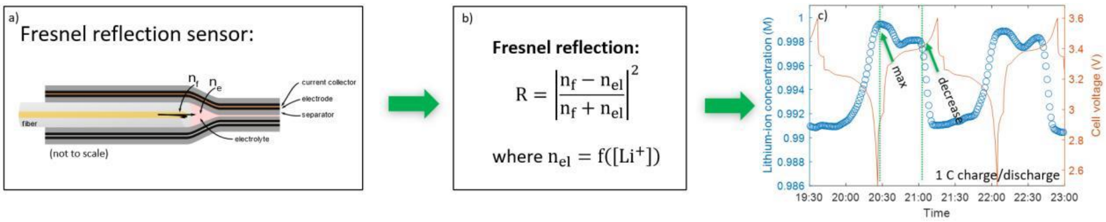

***Figure 1.*** *Schematic of sensor placement between the electrodes, with the reflectivity being a function of the Li-ion concentration. The sensor is in this way able to measure the concentration during charge/discharge cycles.*

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# Robust, High-information Density Data Formats for Battery State-of-Health Forecasting and Transparent Value Chains

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Increased rates of electrification in mobility applications, industrial machinery, and the urgent need to decarbonize electrical grid services, has made the number of lithium-ion battery packs intended for market roll-out soar. With scarce resources (raw materials, production capacity, energy), maximizing the useful life-time of cells and packs is crucial to ensure sustainability. One possible pathway leverages the on-board connectivity and computational resources present in many electrified applications to continuously track the usage and the evolution of both state-of-health (SOH) and state-of-function (SOF). From the collected data, ageing models can be created and used to either take mitigating actions or ensure timely transfer to a 2nd life market.

Here we present a battery data collection strategy that aims to capture the most relevant aspects of battery usage contributing to SOH and SOF evolution. Using high resolution time series of field data collected from two different applications, we evaluate several data aggregation strategies on the merits of relevance to real world scenarios, interpretability, relevance to present and emerging chemistries, data footprint, their availability and not the least their predictive power (PP). For PP we consider both parametric models, Gaussian Process Regression and Neural Network formulations, and compare different subsets of the features space.

Based on the above we finally suggest a homogenized battery information format (BIF). In the BIF information relevant for predicting SOH and SOF is combined with metadata that adds transparency. This mitigates transfer of batteries between different actors within the battery value chain, including potential refurbishers and 2nd life users.

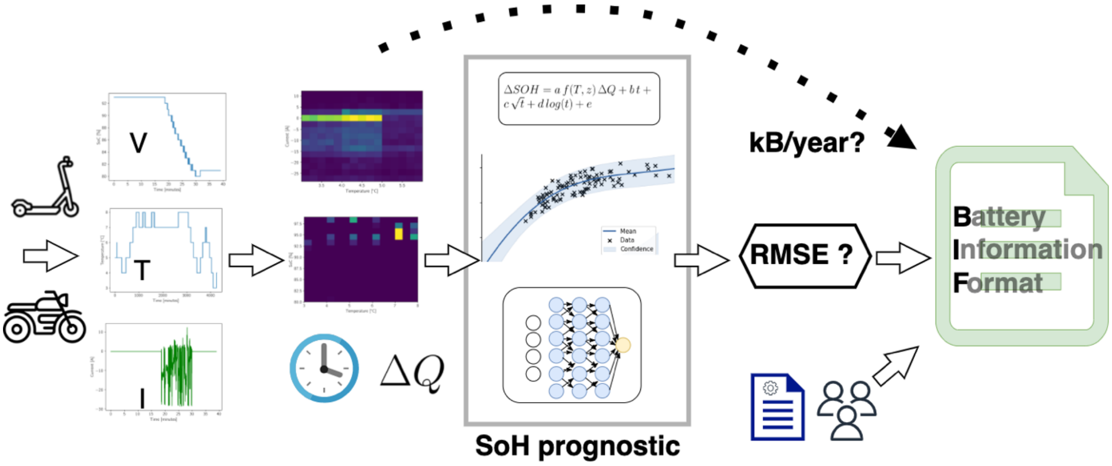

***Figure 1***. *Feature proposals are derived from time-series data to serve as input to SoH prognostics, using three modelling paradigms. Their predictive power is weighed against data foot-print and other requirements to finally suggest a homogenized battery information format.*

# Chemical Modification of PBI and its Performance in a Vanadium Redox Flow Battery

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Polymer membranes based on the benzimidazole (PBI) unit are being employed in various electrochemical energy conversion and storage applications, including vanadium redox flow batteries [1]. Unmodified PBI has shown excellent barrier properties again vanadium ions, while moderate performance in terms of conductivity. Addition of ionic groups in the backbone can increase the conductivity while maintaining the desired barrier properties. Herein dense sulfonated para-PBI (SpPBI), benzylated meta-PBI (BmPBI) and sulfo-benzylated meta-PBI (SBmPBI) were synthesized and casted. Additionally, blends of BmPBI and SpPBI were prepared and tested. No vanadium ions were detected after 1 month permeability test of BmPBI in an H-Cell. Both SpPBI and SBmPBI showed a threefold increase in ion conductivity in 2 M $H_2SO_4$. The area specific resistance (ASR) in a VRFB cell of both sulfonated mPBI was half compared to mPBI, even though the thicknesses were twice as high. Higher energy efficiencies are recorded for SpPBI compared to mPBI at different current densities. Meanwhile, long term cycling, Polarization curves and *in-situ* permeability calculation are to follow for the three modified PBIs.

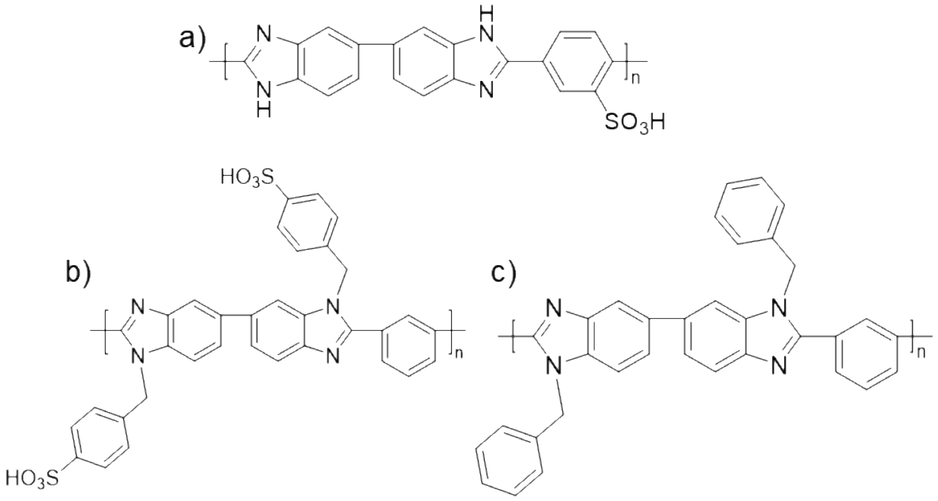

***Figure 1***. *Chemical structures of (a) SpPBI, (b) SBmPBI and (c) BmPBI.*

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# Exploring the Design Space of Solid Boosters for Ferrocyanide-based Flow Batteries

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Aqueous organic redox flow batteries are a promising candidate for the next generation's energy storage technology. While they have the potential to be low-cost and more environmentally friendly than conventional batteries, they suffer from a low energy density. However, the addition of solid boosters to the electrolyte tanks can increase both the system's energy density and energy efficiency significantly if certain criteria are met.[1,2]

A solid booster typically consists of a redox-active material (RM), a binder and a conductive additive. The booster's RM needs to have a reduction potential very closely matching that of the dissolved RM and be insoluble in the electrolyte, regardless of oxidation state.

One popular choice of RM for the positive side is sodium or potassium ferrocyanide. In the present work, using solid boosters based on Prussian White are explored for a symmetric ferrocyanide-based flow battery, and considerations regarding of characterization, efficiency, optimization procedure and stability are presented.

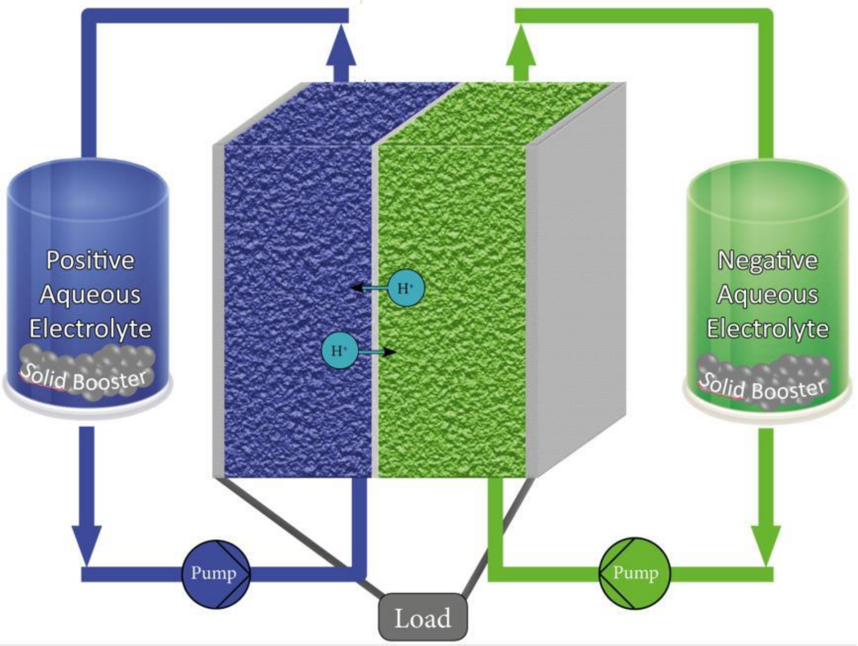

**Figure 1:** A schematic of a flow battery using solid boosters in the electrolyte tanks.

# Electrochemical Driven Disordering of Monoclinic Distorted $VO_2$ as Cathode in Li-ion Batteries.

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Traditional materials for rechargeable Li-ion battery electrodes are based on well-ordered crystalline materials containing scarce elements such as lithium and cobalt [1]. The strong focus on crystalline electrode materials have caused disordered/amorphous electrode materials to be overlooked in the past [2]. Thus, a number of well-performing, cheap and environmentally friendly materials may have been missed as candidates for rechargeable Li-ion battery electrodes.

In this study, we have synthesized a monoclinic vanadium oxide, $VO_2$(M), *via* thermal treatment of a non-distorted rutile $VO_2$ phase and employed it as a cathode against a Li-metal anode. $VO_2$(M) displays an irreversible broadening and loss of scattered intensity (**Fig. 1**) in the Bragg reflections when Li-ions are intercalated into the structure (**Fig. 1**). This behavior is very similar to the disordering behavior of closely related rutile $TiO_2$ electrodes, as reported by Christensen, *et al*. [3]. Similar to the $TiO_2$ structure, $VO_2$(M) has channels for Li migration along one crystallographic axis [4]. To investigate the irreversible order-disorder transformation in $VO_2$(M), we collect *operando* powder x-ray diffraction and total scattering data during charge and discharge, i.e. Li-ion insertion and extraction. The data is analyzed using Rietveld refinement and pair distribution function analysis, respectively. The irreversible phase transformation begins after inserting 0.17 Li/V or approximately two hours into the first discharge at C/12. The broadening of the Bragg reflections increases with continuous intercalation of Li-ions. The subsequent complete charge and discharge look similar from the scattering, which indicate that after the irreversible disordering process, a reversible phase transition occurs in the disordered state.

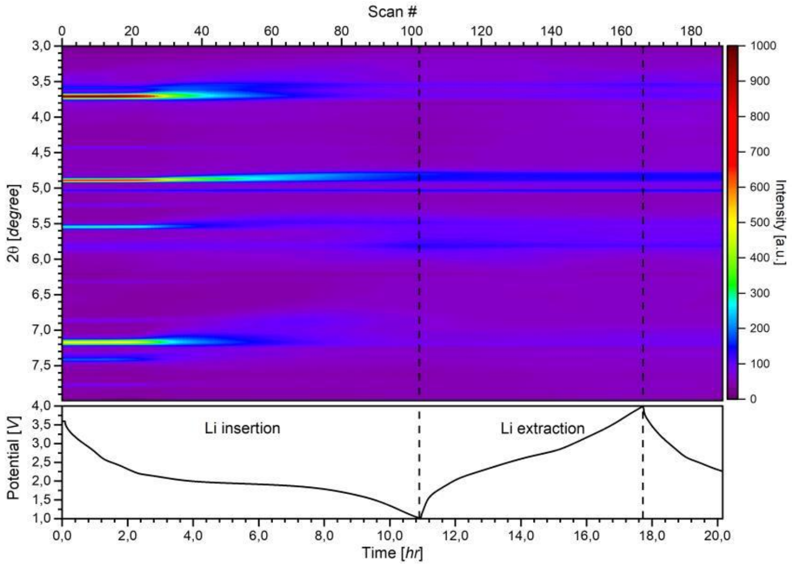

***Figure 1.*** *Overview of diffractograms (scan number, #, as function of diffraction angle, 2θ) and galvanostatic charge/discharge curve (time vs. potential) for $VO_2$(M) cathode vs. a Li-metal anode at a C/12 cycling rate. Diffraction patterns were measured at PETRA-III P02.1 (λ = 0.20734 Å).*

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# $Ti_3C_2T_x$ MXene as an Anode Material in Li- and Na-ion batteries: What is the Electrochemical Capacity Due to?

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The unique properties of the $Ti_3C_2T_x$ MXene continue to attract attention within the energy storage field [1-4]. Many of the previous reports on the use of this material in Li- and Na-ion batteries mainly concentrate on modifying the surface of the MXene or the cycling protocol in attempts to achieve higher specific capacities [5,6]. Very few studies have, on the other hand, addressed the nature of the electrochemical reactions taking place during the electrochemical cycling of the material [7].

This study aims at gaining a deeper understanding of the electrochemical behavior of freestanding $Ti_3C_2T_x$ electrodes without any binder and conductive additive. The results obtained with these electrodes, which were manufactured by vacuum filtration of MXene suspension in deionized water, are used to identify the electrochemical reactions yielding the observed capacities obtained in Li-metal cells using 1 M $LiPF_6$ in EC:DEC (50:50 vol%) as the electrolyte and in Na-metal cells containing 1 M NaFSI in TEG-DME. Constant current and cyclic voltammetry cycling as well as in-house ex-situ XPS and synchrotron-based ex-situ HAXPES and XAS were used to identify the redox processes taking place upon the charging and discharging of the MXene electrodes.

The results, obtained when cycling the freestanding $Ti_3C_2T_x$ MXene electrodes in Li-metal cells, indicate that the reversible capacity can be explained by redox reactions involving Ti species present in the $Ti_3C_2T_x$ surface layer. The charge storage mechanism is hence faradic. The results of XAS measurements carried out in the transmission mode likewise indicate that the redox reactions are surface confined and do not involve the inner C-Ti-C layer of the $Ti_3C_2T_x$ MXene.

When cycling material between 0 and 3 V vs. $Na^+/Na$ in Na-metal cells, the specific capacity of the $Ti_3C_2T_x$ electrodes increases progressively with the cycle number (from 5 mAh/g on the first cycles to 50 mAh/g on the 200th cycle using a cycling rate of 10 mA/g). This effect is suggested to be connected to the deposition of sodium at about 0 V vs. $Na^+/Na$ as this count yield an activation of the electrodes. Comparisons of the cycling behavior of freestanding $Ti_3C_2T_x$ electrodes with those of monolithic $TiO_2$ (amorphous) nanotube electrodes with nanotubes lengths of about 4 µm suggest that the behavior of the $Ti_3C_2T_x$ electrodes cannot be explained by the expected spontaneous formation of $TiO_2$ on the surface of $Ti_3C_2T_x$ electrodes. The capacity for the $TiO_2$ nanotube anode was relatively high, i.e., 160 mAh/g, on the first cycle but was seen to decrease to about 90 mAh/g after 100 cycles when cycling at a rate of 10 mA/g. The reasons for the differences seen between the $Ti_3C_2T_x$ and $TiO_2$ nanotube electrodes will be discussed based on the general electrochemical behavior of the $Ti_3C_2T_x$ electrodes.

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# Exploring the realm between liquid and solid: Oligocarbonates as electrolytes in Li-ion batteries

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In the quest to move to a low carbon society, energy storage is a vital component. Most batteries today, use a liquid electrolyte composed of linear and cyclic carbonate thanks to their high ionic conductivities. However, the linear carbonates are volatile and therefore batteries require extensive thermal management systems, adding cost and weight to the battery modules. Therefore, alternative electrolytes have been explored. Some examples include non-flammable liquids, polymers, and ceramics. [1][1]

Polymer-based electrolytes have often been focused on poly(ethylene oxide) (PEO). However, their low lithium transference number has often been a limitation, leading to research on alternative polymer hosts such as polycarbonates and polyesters which have significantly higher transference numbers [2]. This has been suggested to be due to relatively weak coordination to lithium ions. Although the transition between low molecular weight glymes to high molecular weight PEO is rather established [3], the synthesis and characterization of linear oligocarbonates is still uncharted. In this study, short oligocarbonates have been synthesized and characterized as liquid electrolytes for Li-ion batteries.

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# Influence of sol-gel reaction temperature on morphology and lithium storage performance of $LiNi_{0.5}Mn_{1.5}O_4$ cathode materials

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Lithium-ion batteries (LIBs) for electric vehicles have been increasingly attracted battery researchers during the recent years because of the highly safety and energy density[1]. Considering that cathode materials is responsible for the majority of the weight and cost in state-of-art LIBs, development of high voltage and high-capacity cathode is a significant research direction for next-generation LIBs[2]. Notably, commercial $LiCoO_2$ and its derivatives are widely used for cathode materials, which incurred a significant increase of the Co price. Among Co-free electrode materials, spinel $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) is the most promising cathode due to its high energy density (650 W h $kg^{-1}$), high operating voltage (4.7 V vs. $Li^+/Li^0$), low cost and environmentally friendliness[3]. However, the LNMO spinel materials suffer from the lack of electrolyte compatible owing to such high voltage as well as rapid capacity decay leading to short battery life[4]. To solve these problems, the modification of appropriate LNMO cathode material need to be considered to enhance the electrochemical performance.

In the present work, sol-gel method with different process parameters was carried out attempted to change the morphology and distribution of LNMO particles. Four different LNMO materials, named as LNMO_*x* (*x* = 80/110/140/170), were produced by corresponding oil bath temperature during the sol-gel process (**Figure 1a**). The structure of the obtained materials was studied by scanning electron microscope (SEM), particle size distribution (PSD), inductively coupled plasma-optical emission spectrometry (ICP-OES), X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS). Furthermore, the electrochemical performance was investigated by charge-discharge tests of the coin cells (**Figure 1b**). LNMO_110 sample showed good rate performance at room temperature comparing with other three samples.

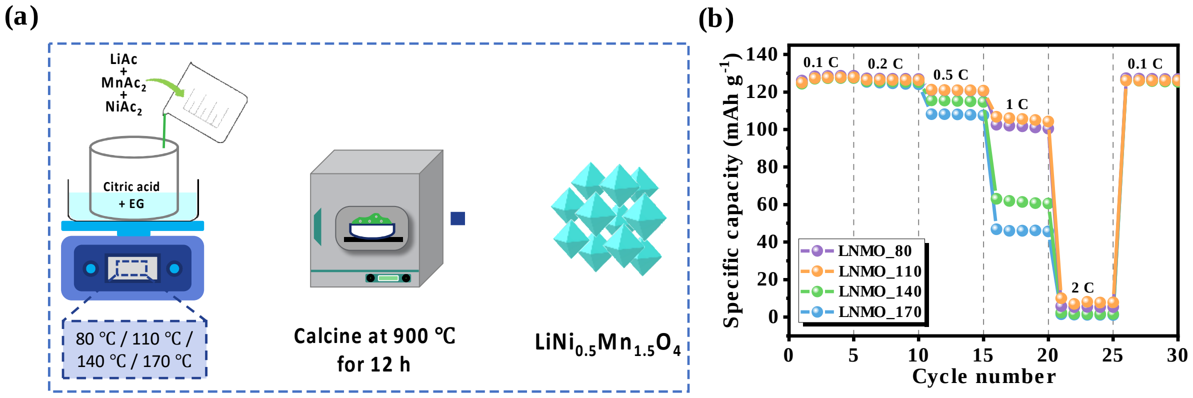

***Figure 1.*** *(a) Illustration of the synthesized process and (b) Rate performance of LNMO_x samples.*

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# Novel Electrochemical Approaches to Understand Electrolyte and Binder Degradation in LNMO-based Cells

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The spinel material $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) with an operating voltage of 4.7 V *vs.* $Li/Li^+$ is a potential candidate for high-energy and high-power density lithium-ion battery applications.[1] However, challenges such as electrolyte oxidation at higher potentials, dissolution of the transition metals and a resulting severe capacity fade in full-cells, particularly at higher temperatures, need to be addressed before commercial implementation.

The Linear Sweep Voltammetry (LSV) methodology, which is commonly used to determine the electrochemical stability of any electrolyte, is generally found to overestimate these stability limits. Here, we introduce an alternative technique termed Synthetic Charge-discharge Profile Voltammetry (SCPV) to explore the oxidative stability of some of the commonly used electrolytes in lithium-ion batteries including high voltage cells like LNMO.[2] Using SCPV, the potential is varied according to the charge-discharge curve of the active material. This approach proves to be a useful way for evaluating the electrolyte's anodic stability in a more realistically representative manner, with passivation kinetics and changes in the electrode potential at the electrode-electrolyte interface being more accurately represented. This method forms a basis for the screening of electrolytes used for the LNMO research.

Furthermore, the SCPV technique is also explored to investigate binder stability. Polyacrylonitrile (PAN), well-known for its oxidative stability, is compared with the standard polyvinylidene fluoride (PVdF) binder. While numerous studies have shown that tailoring the binder system or application of polymer coatings onto the active material LNMO particles provides an efficient strategy for mitigating capacity fade and improving the surface chemistry,[3] the PAN-based system displays faster capacity degradation and development of resistive layers on the cathode surface. By combining SCPV with characterisation techniques such as Intermittent Current Interruption (ICI) and On-line Electrochemical Mass Spectrometry (OEMS), these degradation processes could be better understood. Increased resistance and extra parasitic processes, such as gas evolution due to PAN degradation, as well as PAN dissolution into the electrolyte system, are then identified as the key drivers of quicker capacity fade.

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