Tampere University

Aalto University

VTT

NextGenBat

Next Generation Battery Materials and Concepts

2020 – 2024

Final Report

funded by Business Finland

delfortgroup

Pulsedeon

beneq

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Background

Adoption of the next-generation solid-state technology is foreseen to chance lithium battery technology drastically. These batteries based on solid electrolytes are anticipated to offer higher energy storage capacity while improving safety. Solid-state batteries are still in research and early development and commercialization phase tough several scientist and companies are working to find solutions for the challenges and bring these batteries closer to the markets.

Major issues related to the solid-state lithium batteries include charge transfer across and between different components and their compatibility as the electrode active materials undergo dimensional and phase changes during the battery operation. One approach to solve these issues is to utilize various thin films, which function as functional interphases. A solid-state battery could be realized with combing these protected electrode components with a flexible electrolyte structure.

Manufacturing of solid-state batteries requires innovative technological solution as lack of appropriate cost-effective fabrication methods is one of the bottlenecks in solid-state betty commercialization. Finland is a forerunner in such thin film fabrication technologies as pulsed laser deposition (PLD) and atomic layer deposition (ALD), which are expected to play a major role in solid-state lithium battery manufacturing. These technologies still must be engineered to meet requirements set by the chemical nature of the battery components and the battery manufactures cost-effectiveness expectations.

The NextGenBat project aimed at finding solutions for these issues by collaborative research effort between two Finnish universities, one research organization and three companies. We anticipated that our project would strengthen Finland position in this highly competitive but potentially rewarding sector while educating specialists for the field. Solid-state batteries give high promises and are expected to be adopted to consumer electronics, electric vehicle and renewable energy storage markets in late 2020's or early 2030's while their market potential is forecasted to reach around 1,300 – 2,300 million EUR by the end of 2030.

The main outcomes of the NextGenBat project are summarized below. This report describes development of the negative and positive electrodes with the focus on interface development and also work on solid electrolytes. Additionally, insight on the related thin film manufacturing technologies is provided.

Summary of the activities

The NextGenBat project focused on advancing solid-state lithium battery technologies through the development of optimized electrode materials, protective coatings, and novel electrolyte systems.

Key achievements included using ALD or ALD-MLD to coat lithium electrodes (e.g., Al_2O_3) and nickel-rich NMC811 cathodes (e.g., LiF, LTO, TiO_2-TPA), significantly improving cycling stability by suppressing the active material side reactions and deforming. Innovations in PLD enabled precise fabrication of thin lithium films and solid electrolytes or ion conducting barrier layers. The project emphasized rigorous electrochemical testing and advanced material characterization to assess structural evolution and degradation mechanisms.

The project successfully demonstrated a functional solid-state coin cell prototype integrating coated electrodes with a cellulose separator, and optimized polymer electrolyte.

Through collaboration between universities, research institution, and industry partners, NextGenBat strengthened Finland's expertise in battery R&D. Educated new experts and produced over 10 peer-reviewed publications, more than 10 scientific presentations and several innovations, the project lays a foundation for future innovations in safer, high-performance solid-state batteries for electric vehicles and renewable energy storage.

Anode

Lithium negative electrodes

Lithium metal is considered the ultimate anode material for next-generation energy storage systems due to its exceptionally high theoretical specific capacity and low electrochemical potential. These properties make it a promising candidate for high-energy-density batteries, such as lithium-metal batteries (LMBs) and solid-state lithium batteries. However, despite its advantages, lithium metal faces several critical challenges that hinder its widespread commercialization.

Challenges: One of the major obstacles is dendrite formation, where uneven lithium plating during cycling leads to the growth of needle-like structures that can penetrate the separator, causing short circuits and potential battery failure. Additionally, unstable solid electrolyte interphase (SEI) formation due to lithium high reactivity with the electrolyte results in continuous electrolyte decomposition, excessive SEI layer growth, and increased interfacial resistance. These issues contribute to poor cycle life, capacity fade, and safety risks, limiting the practical use of lithium metal anodes.

Strategies: To address above challenges, ALD has emerged as a promising strategy to enhance the stability and performance of lithium metal electrodes. ALD enables the deposition of ultrathin, uniform, and conformal coatings that can serve as artificial SEI layers, protecting lithium from direct exposure to the electrolyte while allowing efficient lithium-ion transport.

NextGenBat focuses on the development and characterization of ALD-coated lithium electrodes, evaluating their impact on electrochemical performance, dendrite suppression, and interfacial stability. By systematically optimizing ALD coatings on lithium metal and PLD-deposited lithium thin films, this research aims to provide insights into improving lithium metal anodes for future high-energy-density batteries. The work has been carried out as a collaborative effort between the Aalto University, Pulsedeon and Beneq.

Fabrication of thin lithium film electrodes

PLD is a promising technique for fabricating thin and uniform lithium metal films, which can serve as negative electrodes for next-generation batteries. This method allows for precise control over film thickness and morphology, which is crucial for achieving **high-performance lithium-metal batteries**. PLD offers an alternative route to produce lithium electrodes with improved structural integrity and tunable thickness.

Pulsedeon, equipped with advanced PLD systems, has optimized the deposition process to fabricate high-quality thin-film lithium electrodes. In addition, Aalto University and Beneq have employed atomic layer deposition to further enhance the electrochemical performance and stability of these PLD-produced lithium electrodes. This combined approach offers a pathway toward more stable and efficient solid-state lithium batteries.

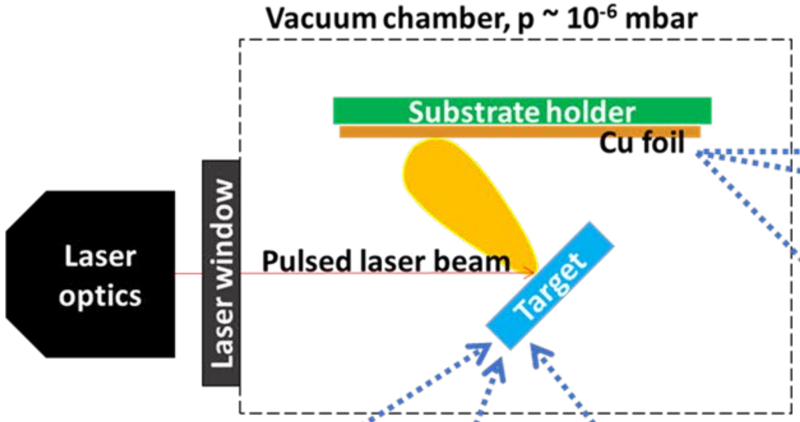
PLD is also used for the fabrication of thin solid-state lithium electrolytes, including lithium phosphorus oxynitride (LiPON), lithium lanthanum zirconium oxide (LLZO), and lithium phosphorus sulfide (LPS). Pulsedeon has utilized the PLD technology to optimize and fabricate these solid-state lithium electrolytes, further advancing their application in next-generation battery systems.

PULSEDEON

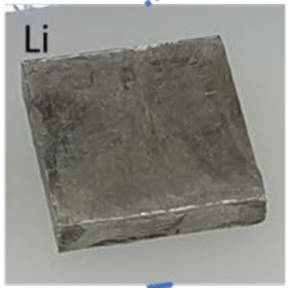
Capabilities developed in NextGenBat

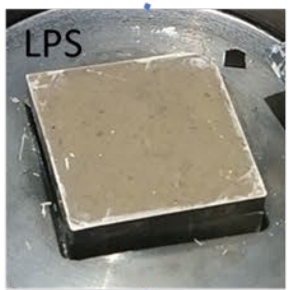
- Materials synthesis of LPS, LLZO, LiPON
- PLD targets of Li, LPS, LLZO, LiPON
    - Li pressing techniques
    - LPS hot pressing
    - LLZO, LiPON pressing and sintering techniques
- PLD process for Li, LLZO, LPS
- Metallised paper current collector
- Multi-layer PLD
- Processing, handling, and characterization of air-sensitive materials

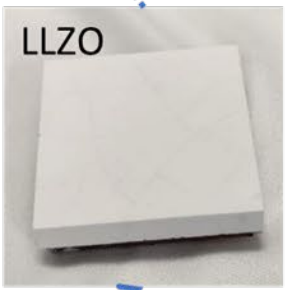
Pulsed Laser Deposition process



PLD targets

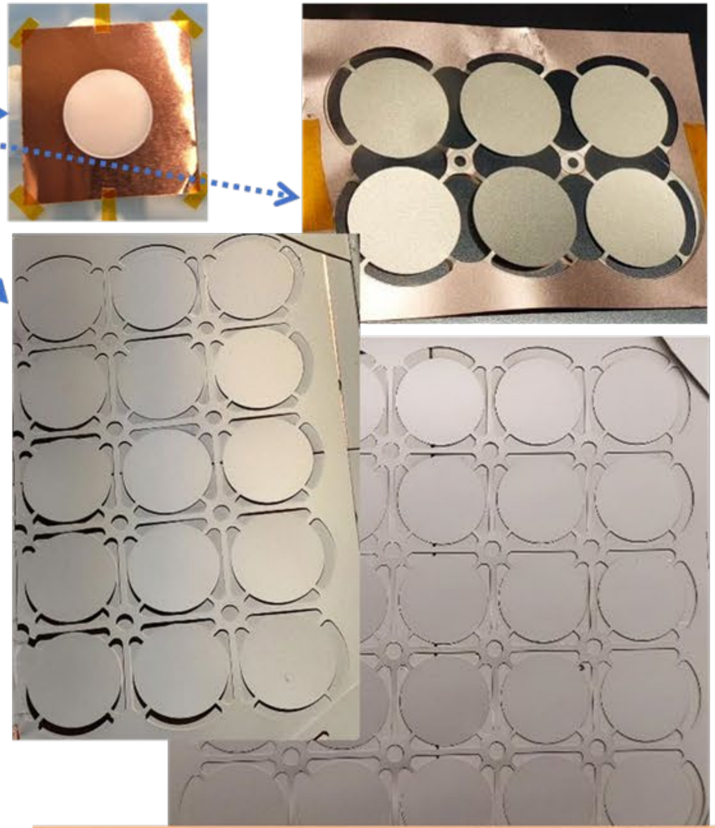


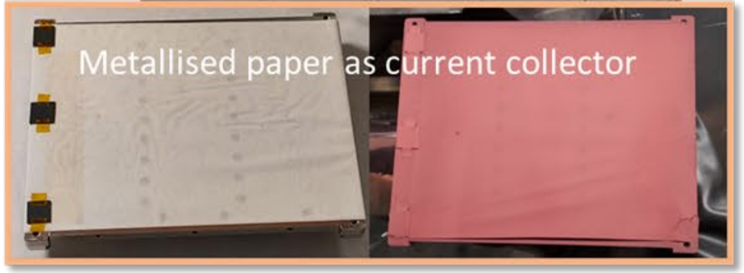




Materials synthesis and processing

Cu-Li; Cu-Li+LLZO; Cu-Li+LLZO+LPS electrodes for coin cells




Metallised paper as current collector

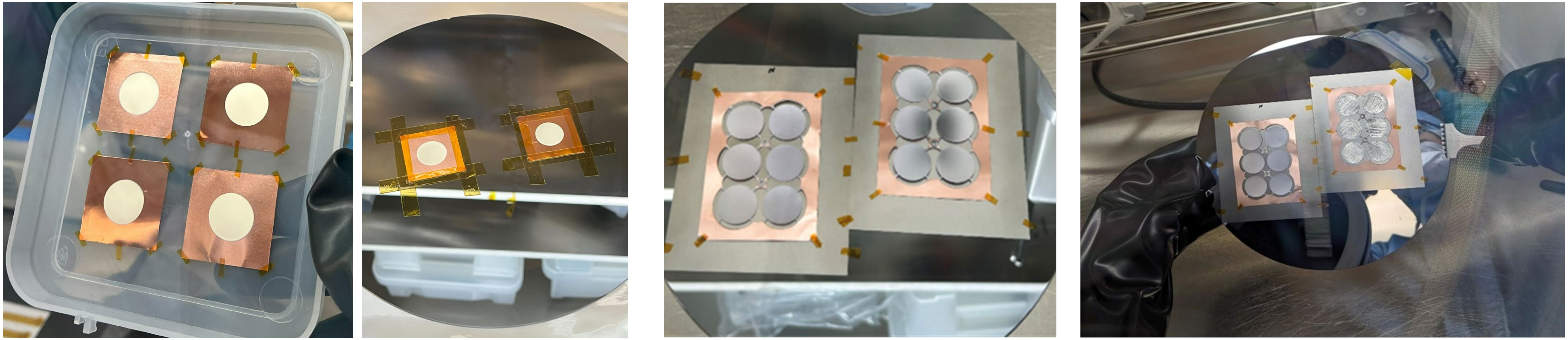
ALD coating of metallic lithium

Alumina was deposited on Li metal sheets in an ALD reactor connected to an argon-filled glovebox at the Aalto University. To evaluate the impact of these coatings on lithium electrodes, Al_2O_3 layers of varying thicknesses were deposited on metallic lithium surfaces. This study involved the characterization of coated lithium using x-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), and electrochemical measurements in both symmetric and full-cell configurations.

The Al_2O_3 coatings significantly improved the air stability of lithium, preventing rapid oxidation and making handling safer for large-scale applications. Electrochemical testing revealed that thicker Al_2O_3 coatings effectively suppressed dendrite growth and stabilized the lithium-electrolyte interface, leading to improved capacity retention and extended cycling life. However, the increased coating thickness also introduced higher interfacial resistance, which resulted in a reduction in initial capacity.

Reference:
S. Pakseresht, V. Miikkulainen, F. A. Obrezkov, A. Waleed Majeed Al-Ogaili, Jouko Lahtinen, **T. Kallio**, Atomic Layer Deposition for Protecting Lithium Metal Anodes for High-Voltage Battery Applications, submitted.

Al_2O_3 coating via ALD on Pulsedeon Lithium Samples



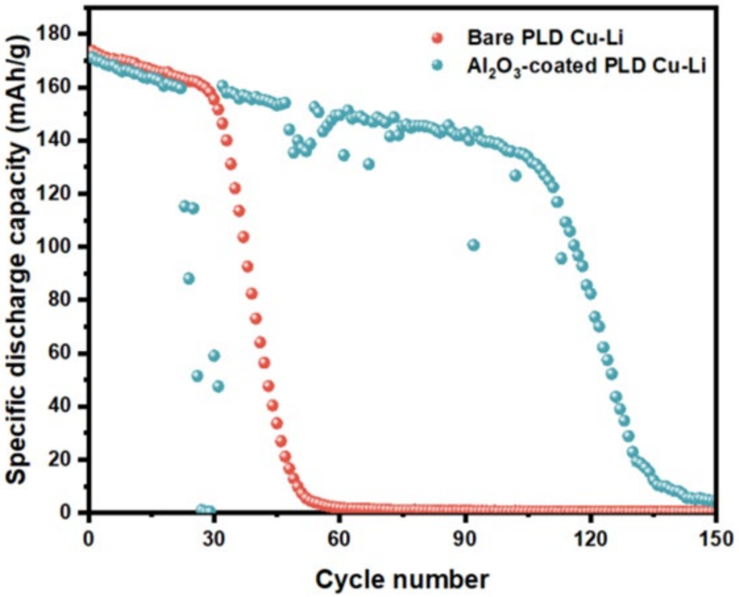
At Beneq, 2 nm and 5 nm thick Al_2O_3 coatings were deposited using ALD onto samples made utilizing PLD. These samples had lithium on a copper foil and were provided by Pulsedeon.

All sample preparation and handling was done under an inert argon atmosphere in a glovebox. Thesamples were loaded into the TFS200 ALD chamber via a loadlock from the glovebox.

CASE STUDY – performance of Al_2O_3 coated thin film lithium electrodes

Thin lithium samples (PLD Cu-Li) fabricated by Pulsedeon were coated with a 2 nm Al_2O_3 layer using ALD by Beneq. The purpose of this coating was to enhance the stability and performance of lithium electrodes by providing a protective barrier against unwanted side reactions, electrolyte decomposition, and dendrite formation.

To evaluate the impact of the Al_2O_3 coatings on electrochemical performance, full cells with Li-NMC811 configuration were assembled and tested at Aalto. The cells were cycled at 1C with the voltage range of 3.0 – 4.4 V, and the results demonstrated a notable improvement in the lifespan for the Al_2O_3 coated electrodes compared to the uncoated lithium electrodes. The enhanced cycling stability is attributed to the ultrathin Al_2O_3 layer, which likely acts as an artificial SEI, minimizing lithium consumption and reducing dendrite growth over prolonged cycling. These findings show the potential of coatings fabricated via ALD in extending the durability of lithium-metal batteries and mitigating performance degradation in high-voltage cathode applications.



Cathode

High-voltage cathode materials

NMC811 ($LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$) is a nickel-rich cathode material pivotal for advancing lithium-metal batteries (LMBs) toward achieving cell-level energy densities >400 Wh kg^{-1}. Its high nickel content (~80 %) enables exceptional specific capacity (>200 mAh g^{-1}) and energy density, outperforming lower-nickel variants like NMC111 or NMC532. By reducing cobalt usage, NMC811 lowers material costs and addresses ethical concerns tied to cobalt mining. The increased nickel content enhances the redox activity of $Ni^{2+}/Ni^{3+}/Ni^{4+}$, contributing significantly to the overall capacity. This makes NMC811 highly attractive for next-generation lithium-ion batteries and LMBs, which aim for greater energy storage capabilities. Additionally, NMC811 has a wider electrochemical window, enabling battery operation at higher voltages, which maximizes the utilized energy storage capacity and thus improves the energy density of the battery. The high energy density potential of NMC811 is crucial in addressing the growing demand for lightweight and high-performance batteries such applications as portable electronics, drones, and electric vehicles.

Challenges: Despite its high energy density, NMC811 presents several challenges that limit its broader adoption, especially under high-voltage operating conditions. These challenges stem mainly from degradation mechanisms that occur at the cathode-electrolyte interface when cycling at high voltages (above 4.3 V vs. Li/Li^+). The NMC811 based batteries suffer from severe electrolyte decomposition, cathode-electrolyte interphase instability, transition metal dissolution, irreversible phase transitions, and particle cracking, all of which degrade capacity retention and cycling performance.

Strategies: To address these challenges, several strategies have been explored at the Aalto University, including NMC811 surface coating with stable compounds to enhance cathode electrolyte interface (CEI) stability, electrolyte engineering using high-concentration or fluorinated electrolytes to minimize side reactions, and structural modifications to suppress phase transitions and mechanical degradation. These approaches collectively aim to extend cycle life, improve thermal stability, and enable the practical deployment of NMC811-based high-energy batteries. Complementary, commercialization of the ALD technique has been developed and analyzed by Beneq.

ALD for coating cathode electrodes - summary

Aalto University

Various coatings synthetized by ALD or ALD combined with molecular layer deposition (MLD) were investigated to assess their effects on high-voltage layered oxide active material ($LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, NMC811) behavior. Based on our earlier studies, the focus was on such lithium containing coatings as $LiTi_yO_x$, LiF and organaometallic TiO_2-tetraphalate. For comparison, also a solid-state coating method was investigated cover NMC811 surface with LiW_xO_y.

As a common feature, each coating was observed to suppress the electrode volume changes, which resulted in more reversible expansion and contraction upon liberating and intercalating lithium during the battery charge and discharge, respectively. The electrode volume changes were realized to reflect, in the crystal lattice level, the c-axis contraction and expansion behavior during the battery operation. This can be well understood by noting that the coatings penetrated in the NMC811 structure covering the grain boundaries between the NMC811 primary particles, in addition to the surface, and by noting that NMC811 used in the experiments has star-like morphology so that the c-axis is oriented from the material surface towards the center.

As another common feature, all the coatings favored the capacity retention of the materials. In postmortem analyses, dissolutions of the transition metals from the NMC811 structure was observed to decrease upon coating. Likewise, crack formation in the aged particles was observed to diminish. These results evidence that the NMC811 mechanochemical degradation is efficiently suppressed by thin coatings with the thickness in the range of 10 nm.

As for the differences, the lithium containing inorganic $LiTi_yO_x$ and LiF coatings were observed to protect NMC811 against the ageing in a high voltage range when cycled up to 4.4 V in full cells with graphite anodes. In the beginning of the cycling these coatings somewhat hindered the electrochemical reactions when compared to the performance of the uncoated NMC811 reference material based electrodes. However, the trend changes after cycling as the capacity decay was diminished for the coated materials.

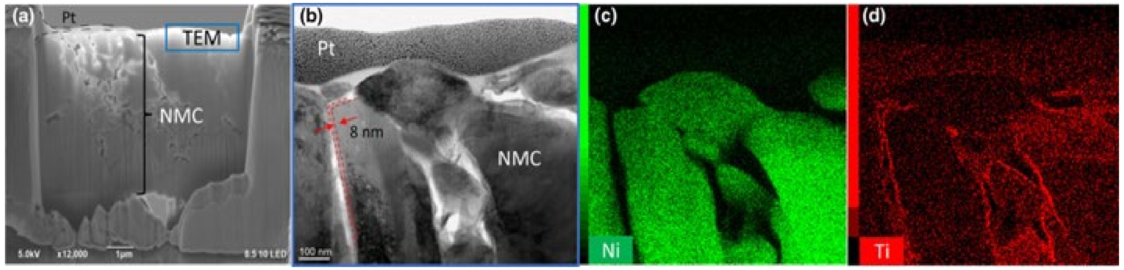
The organometallic coating allowed more facile lithium transfer than the inorganic counterparts. As a drawback, this coating suffers more from cycling in the high potential range when compared to the electrodes with inorganic coatings. However, the organometallic coated electrodes can withstand the oxidative conditions and slow down NMC811 ageing at lower voltage range i.e. when limiting the upper voltage to 4.2. V against graphite in full cell experiments.

Depositing TiO_2-titanium terephthalate thin films on a NMC811 electrode via ALD-MLD

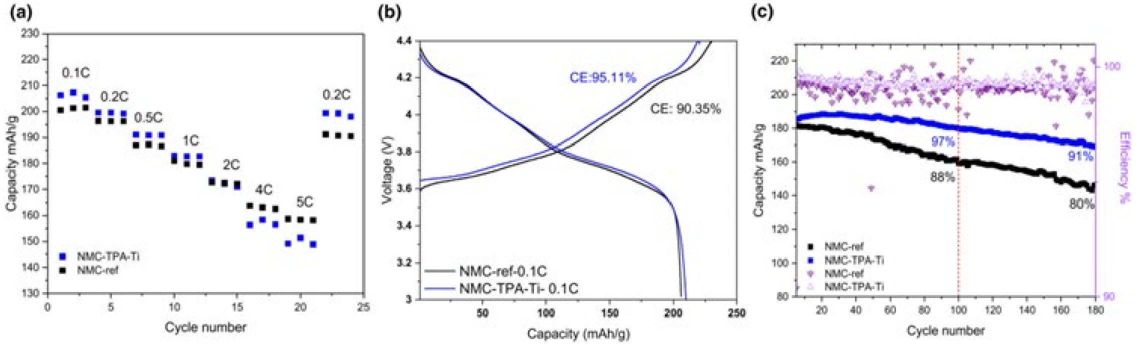
TiO_2-titanium terephthalate (TPA-Ti) as a nanoscale coating for NMC811 electrodes was explored while the coating material was synthetized using an ALD-MLD technique. Electrochemical evaluations demonstrate a significant enhancement in cycling stability, with the TPA-Ti-coated electrodes retaining 86% of their initial capacity after 500 cycles, compared to 56% for the uncoated electrodes in full-cell configurations. The TPA-Ti coating mitigated crack formation, improving structural integrity and resistance to degradation. Our findings highlight the role of coatings fabricated via ALD-MLD in enhancing performance and long-term reliability of lithium-ion batteries.

Reference

Ahaliabadeh, Z.; Miikkulainen, V.; Mäntymäki, M.; Colalongo, M.; Mousavihashemi, S.; Yao, L.; Jiang, H.; Lahtinen, J.; Kankaanpää, T.; **Kallio, T.**, Stabilized Nickel-Rich-Layered Oxide Electrodes for High-Performance Lithium-Ion Batteries, Energy and Environmental Materials, 2024. DOI:10.1002/eem2.12741.



NMC811 particle with uniform surface coating of an approximate TPA-Ti thickness of 8 nm. Wiley CC BY 3.0 license.



The long-term cycling results at 1 C for 100 cycles and 180 cycles highlight the positive effect of the TPA-Ti coating on cycling stability and capacity retention. Wiley CC BY 3.0 license.

Amorphous lithium titanate coating via ALD on a NMC811 surface

Aalto University

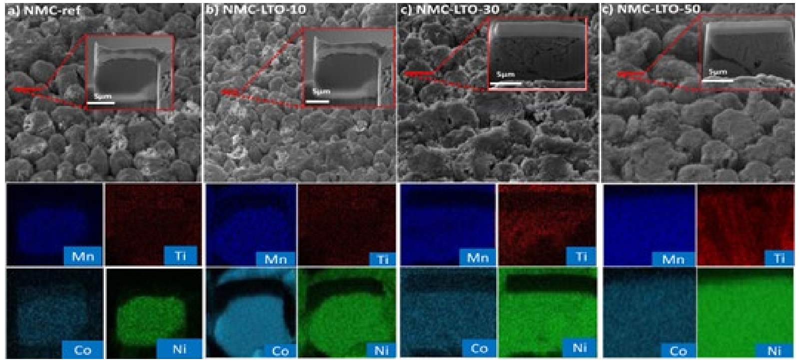
Lithium titanate (LTO) coatings were successfully applied to NMC811 electrodes using ALD to improve electrochemical stability and structural integrity. A systematic analysis was conducted to evaluate the impact of the LTO coatings utilizing scanning transmission electron microscopy, scanning electron microscopy, operando X-ray diffraction, galvanostatic charge-discharge cycling, and dilatometry techniques. The results confirm that the LTO coatings effectively mitigate NMC811 structural degradation, with coated electrodes exhibiting reduced electrode height changes compared to uncoated electrodes. Specifically, height variations of 2.62% were observed for the uncoated NMC811 electrodes, whereas the LTO-coated electrodes demonstrated significantly lower changes. These findings indicate that the LTO coatings suppress mechanochemcial degradation, intergranular crack formation, and anisotropic volume changes, leading to enhanced cycling stability.

Electrochemical performance evaluations further reveal that the LTO-coated electrodes maintain a high capacity retention of over 93% after 100 cycles, in contrast to the uncoated NMC811, which exhibited a decrease to 86%. Long-term cycling tests indicate that progressive structural changes accumulate in the uncoated electrodes, leading to degradation and capacity fading. However, the LTO-coated electrodes exhibit symmetric structural evolution, with most diffraction peaks returning to their original positions after lithiation, demonstrating improved structural reversibility.

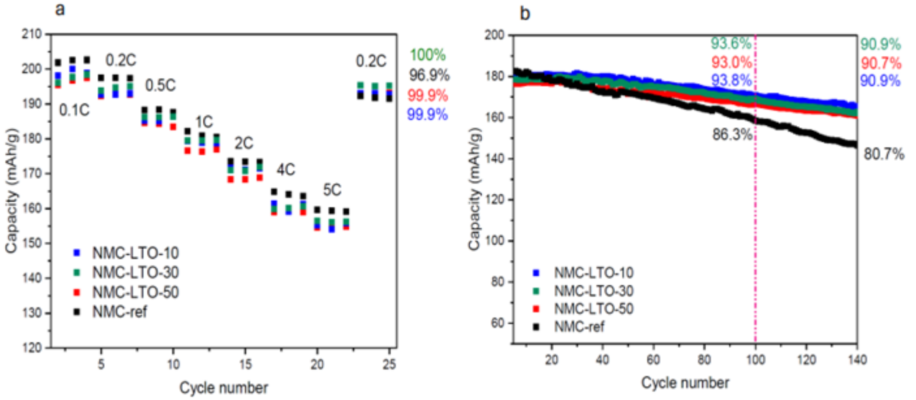
These findings highlight the critical role of grain boundary coatings in stabilizing Ni-rich electrode materials. The application of LTO as a protective layer effectively enhances structural durability and electrochemical performance, contributing to the development of high-efficiency and long-lasting LIBs.

Reference

Ahaliabadeh, Z.; Miikkulainen, V.; Mäntymäki, M.; Colalongo, M; Mousavihashemi, S.; Yao, L.; Jiang, H.; Huotari, S.; Kankaanpää, T.; **Kallio, T.** Surface and Grain Boundary Coating for Stabilizing $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ Based Electrodes, ChemSusChem. DOI: 10.1002/cssc.202400272



NMC811 electrodes are coated with LTO, which infiltrates into the NMC811 particles and cavities. Wiley-VCH CC BY 3.0 license.



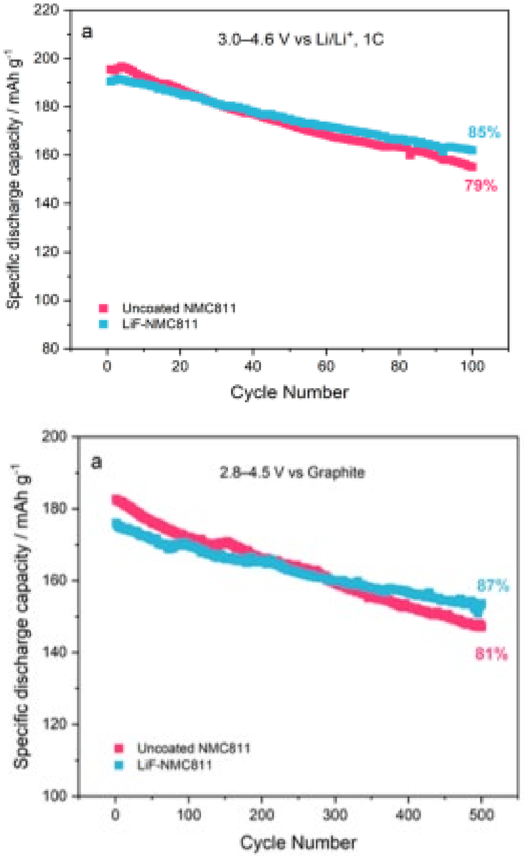
Long-term cycling results cycling up to 140 cycles reveal mitigated capacity decay and voltage fading for the coated samples (9% decrease) compared to NMC-ref (20% decrease). Wiley-VCH CC BY 3.0 license.

Improving cycling stability of NMC811 via LiF ALD coating

A lithium fluoride (LiF) coating was successfully synthetized on NMC811 electrodes using ALD to improve interfacial stability and mitigate degradation mechanisms. The structural integrity of the NMC811 electrodes remain intact following LiF deposition, as confirmed by morphological and compositional analyses. High-resolution transmission electron microscopy revealed the formation of a uniform and conformal nanoscale coating, ensuring effective coverage of the electrode surface.

The electrochemical characterization demonstrated a significant improvement in the cycling stability when comparing the performance of the LiF-coated NMC811 to uncoated electrode. In half-cell configurations, LiF-NMC811 exhibit a capacity retention of 85% after 100 cycles at 1C, while in full-cell configurations, paired with graphite, it retains 88% after 500 cycles.

The enhanced stability observed for the LiF-coated NMC811 electrodes is attributed to the suppression of parasitic electrolyte decomposition and the reduction of impedance growth, effectively minimizing active material loss over extended cycling. Despite introducing a diffusion barrier for Li^+ transport, the rate capability of LiF-NMC811 remains comparable to that of uncoated NMC811, even under high-current conditions. These findings show the potential of LiF as an artificial CEI layer for high-mass-loading and high-voltage Ni-rich cathodes, hihglighting the critical role of CEI engineering in improving long-term electrode stability.

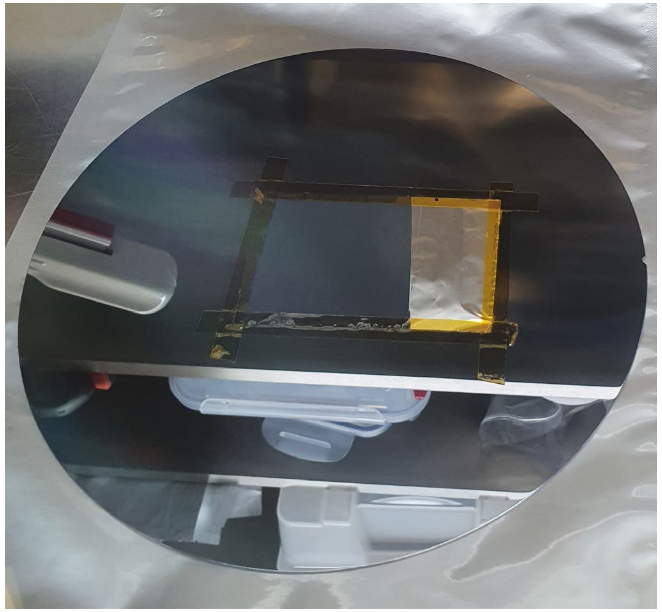


LiF-NMC811 delivers higher capacity retention compared with uncoated NMC811 in both half-cell and full cell configurations. The American Chemical Society CC BY 3.0 license.

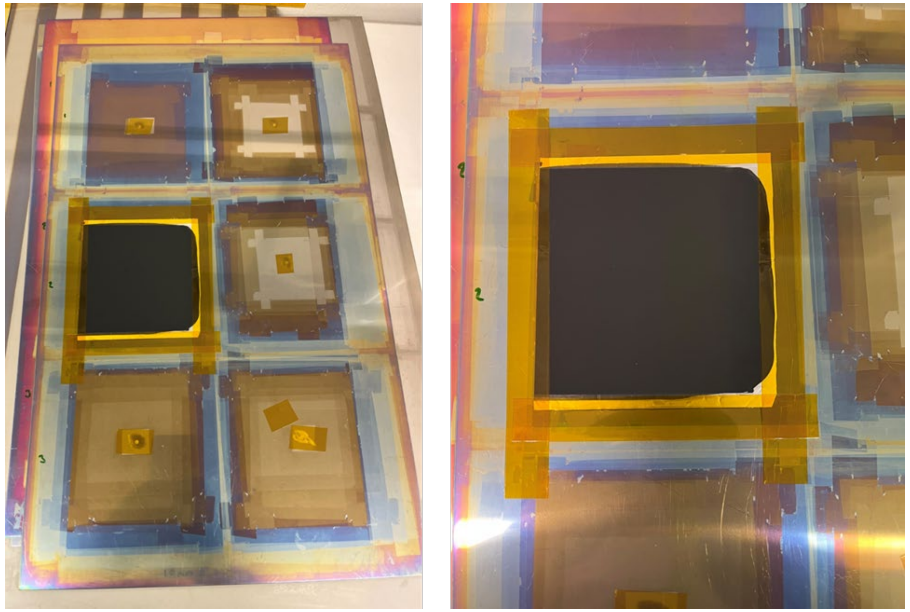
Reference

Llanos, S.; **Ahaliabadeh, Z.**; Miikkulainen, V.; Lahtinen, J.; Yao, L.; Jiang, H.; Kankaanpää, T.; **Kallio, T.**, High Voltage Cycling Stability of LiF-Coated NMC811 Electrode, ACS Applied Materials and Interfaces. 2024. DOI: 10.1021/acsami.3c14394.

ALD coating of NCM811 cathode foils



TFS200 sample carrier wafer



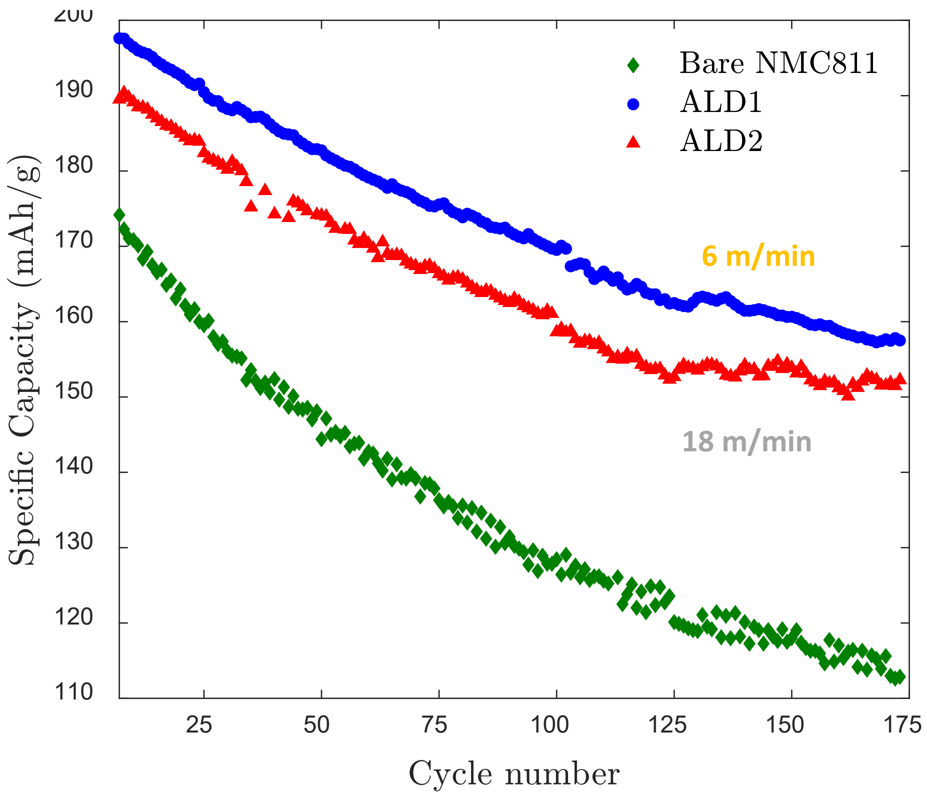
SCS1000 sample carrier plate

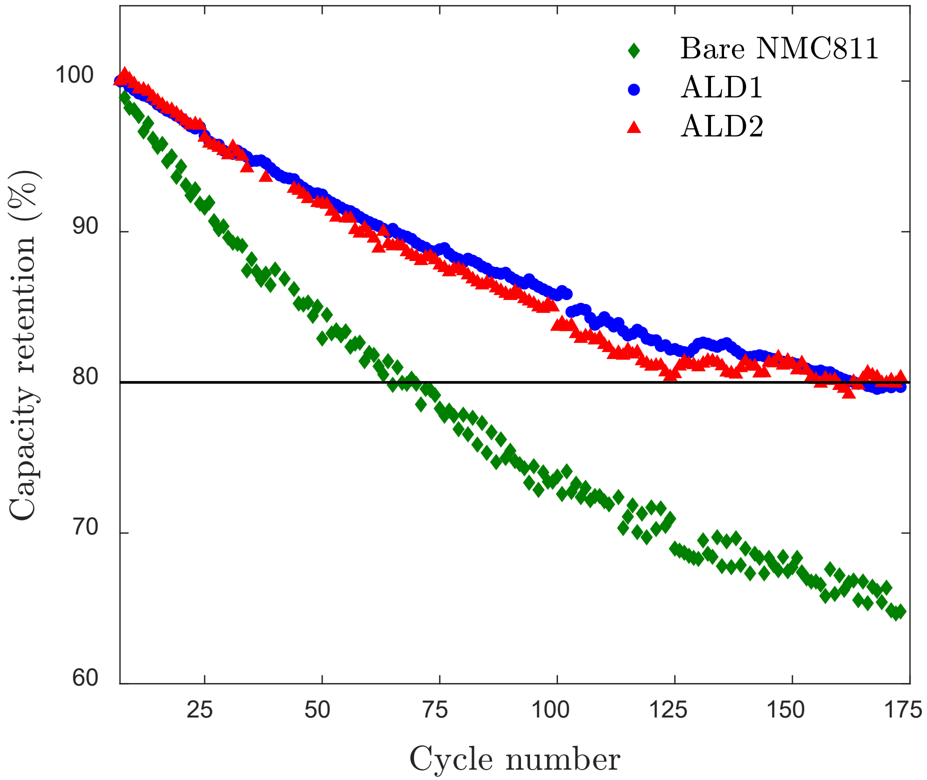
SCS1000 spatial ALD tool

- TFS200 was used to deposit 0.5, 1, 2.5 and 5 nm coating of Al_2O_3 and TiO_2 on NCM811. Most promising results were observed for the thin Al_2O_3 films.

- Larger sheets of NCM811 were produced and thin Al_2O_3 films were coated using the SCS1000 spatial ALD tool, to investigate possibility of roll to roll scale up.

CASE STUDY – ALD Al_2O_3 coating on NMC811 cathode foils





Half Cell – Coin Cell Test
Long cycling conditions:
0.5C, 3.0 – 4.4 V

- Samples were coated by Beneq using spatial ALD
- Both ALD coated samples show increased specific capacity and significant increases in cycling stability
- 18 m/min chosen for cost of ownership calculations

CASE STUDY – Cost of Ownership – Genesis ALD system

- 10 cycles of ALD Al_2O_3 on 50 µm NMC811 cathode foil
- Single side coated

Pilot Production

- 18 m/min web speed
- 0.4 m coating head width
- Estimated total cost for coating
    - ~0.23 USD/m^2
    - ~0.78 USD/kWh
- Estimated capacity
    - ~2.5 Mm^2
    - ~0.74 GWh

Large Scale Production

- 36 m/min web speed
- 1.2 m coating head width
- Estimated total cost for coating
    - ~0.06 USD/m^2
    - ~0.20 USD/kWh
- Estimated capacity
    - ~16.6 Mm^2
    - ~4.8 GWh

Other assumptions & estimations included in total cost of coating:

- CAPEX cost
- Operator costs
- Power consumption
- Annual precursor TMA usage
- Working days/year – 330
- Working hours/day - 24

Electrolyte

Solid inorganic electrolytes

Solid-state electrolytes (SSEs) have emerged as a promising alternative to conventional liquid electrolytes in lithium-ion and lithium-metal batteries, offering significant advantages in terms of safety, stability, and energy density. Unlike liquid electrolytes, which pose risks of leakage and flammability, SSEs are inherently more stable and can effectively suppress lithium dendrite growth. This enables the use of lithium-metal anodes, which have the highest theoretical capacity among all anode materials, thereby significantly increasing battery energy density.

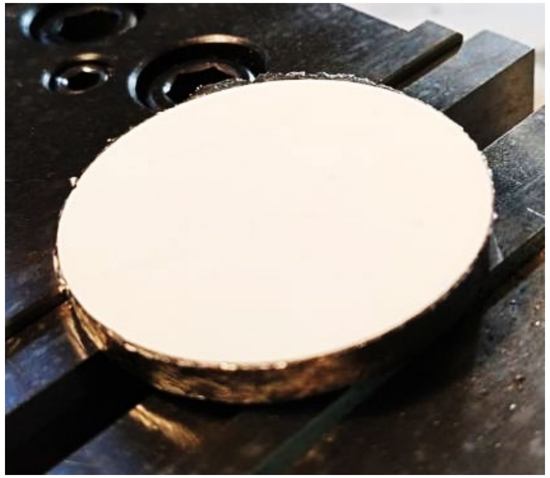
Challenges: Despite these advantages, SSEs still face challenges that hinder their widespread commercialization. Their ionic conductivities are generally lower than those of liquid electrolytes, which can limit charge and discharge rates. Additionally, interfacial stability between the solid electrolyte and the electrodes remains a key challenge, as poor contact can lead to increased resistance and reduced battery performance. To address these issues, extensive research has been dedicated to optimizing the composition, structure, and processing of solid electrolytes.

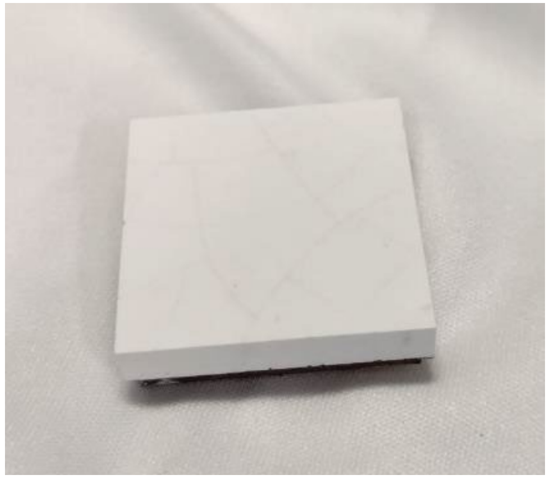
Approaches: Among various SSEs, garnet-type $Li_7La_3Zr_2O_{12}$ (LLZO) stands out as one of the most promising materials due to its high ionic conductivity, excellent chemical and thermal stability, and wide electrochemical stability window. These properties make LLZO a strong candidate for solid-state lithium-metal batteries. In addition to LLZO, other inorganic solid electrolytes such as lithium phosphorus sulfide (LPS) and lithium phosphorus oxynitride (LiPON) have been developed to further improve ionic transport and interfacial compatibility with lithium metal.

A promising approach to overcome the high resistivity issues, is to fabricate thin electrolytes, but appropriate technology should be developed. PLD enables precise control over film thickness and composition, which is crucial for achieving low overall ionic resistance and stable electrode-electrolyte interfaces. In NextGenBat, PLD was employed to fabricate thin-film electrolytes, including LLZO, LPS, and LiPON. The preparation of targets for these materials was optimized through collaboration with Tampere University and Pulsedeon to ensure high-quality deposition. Additionally, the Aalto University focused on investigating the chemical structure effect of the ionic conductivity.

Pressing of PLD targets

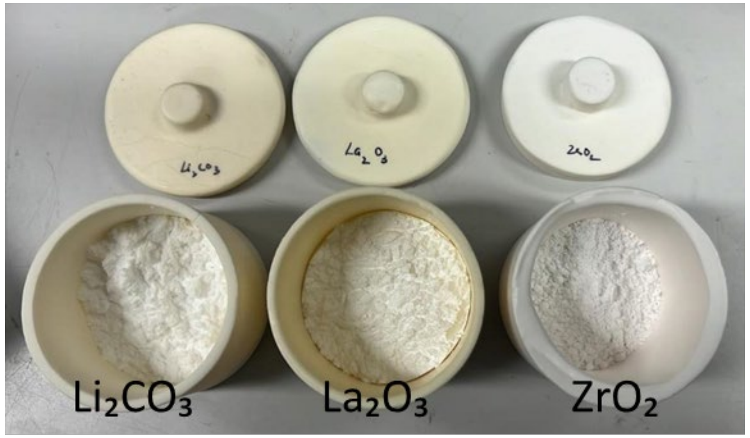

Trial and error…


Graphite cleaned from the surface of the pellet.

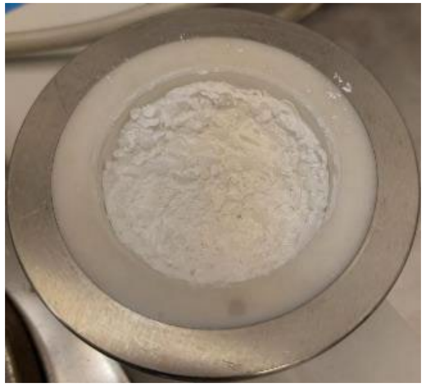

A PLD target ready for testing.

From precursors to PLD targets with different compositions

Precursor powders



Milling

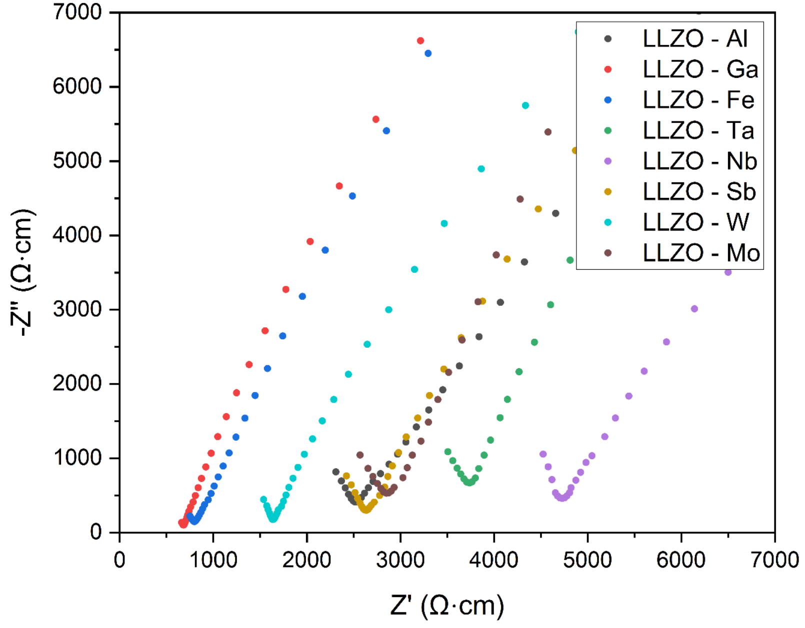


Cold pressing

Fast sintered LiPON

Heat treated LLZO

LLZO chemical composition



Impedance spectra of the doped LLZO materials for determining the ionic conductivities. The Royal Society of Chemistry CC BY 3.0 license

Reference
J. Kosir, S. Mousavihashemi, M. Suominen, A. Kobets, B. P. Wilson, E.-L. Rautama, **T. Kallio**,
Supervalent doping and its effect on the thermal, structural and electrochemical properties of $Li_7La_3Zr_2O_{12}$ solid electrolytes,
Materials Advances 5 (2024) 5260 - 5274.
DOI: 10.1039/D4MA00119B.

Effects of supervalent doping (Al^{3+},Ga^{3+}, Fe^{3+}, Ta^{5+}, Nb^{5+}, Sb^{5+}, W^{6+} and Mo^{6+}) on the properties of lithium lanthanum zirconium oxide (LLZO) solid electrolytes were investigated to fabricate an effective barrier layer. Attention was paid on the synthesis, crystal structure, morphology, and electrochemical performance.

The Ga^{3+} dopant appears to be the most promising dopant generating exceptionally high ionic conductivities for LLZO alongside with very low electronic conductivity. This is attributed to the formation of a different crystal symmetry of a $I\bar{4}3d$ space group enabling fluent lithium transfer. Ga^{3+} has also a very high affinity towards LLZO integration during the synthesis, requiring a clearly lower synthesis temperature, and demonstrated very good sinterability, which results a very dense morphology.

Fe^{3+} doping also produces similar benefits due to the same shift in the crystal symmetry. Yet, it is less suitable because of a very low affinity of Fe towards LLZO.

The other dopants generate LLZO with a $Ia\bar{3}d$ symmetry resulting in lower ionic conductivity, which appear to be related to the relative density of the LLZO grains and the size of the unit cell. This suggest that the electrochemical performance can be improved by densifying the LLZO structure.

Although the Ga doping results in LLZO with superior properties, further studies on electrochemical stability are required to understand behavior in practical applications.

Solid polymer electrolytes

Polymer-based electrolytes are particularly promising due to their flexibility, ease of processing, and compatibility with lithium metal anodes.

Challenges: The solid polymer electrolytes (SPE) comprise of several components. Therefore, optimizing the SPE composition so that fluent processing of polymer electrolytes is achieved while improving their ionic conductivity and mechanical strength is challenging.

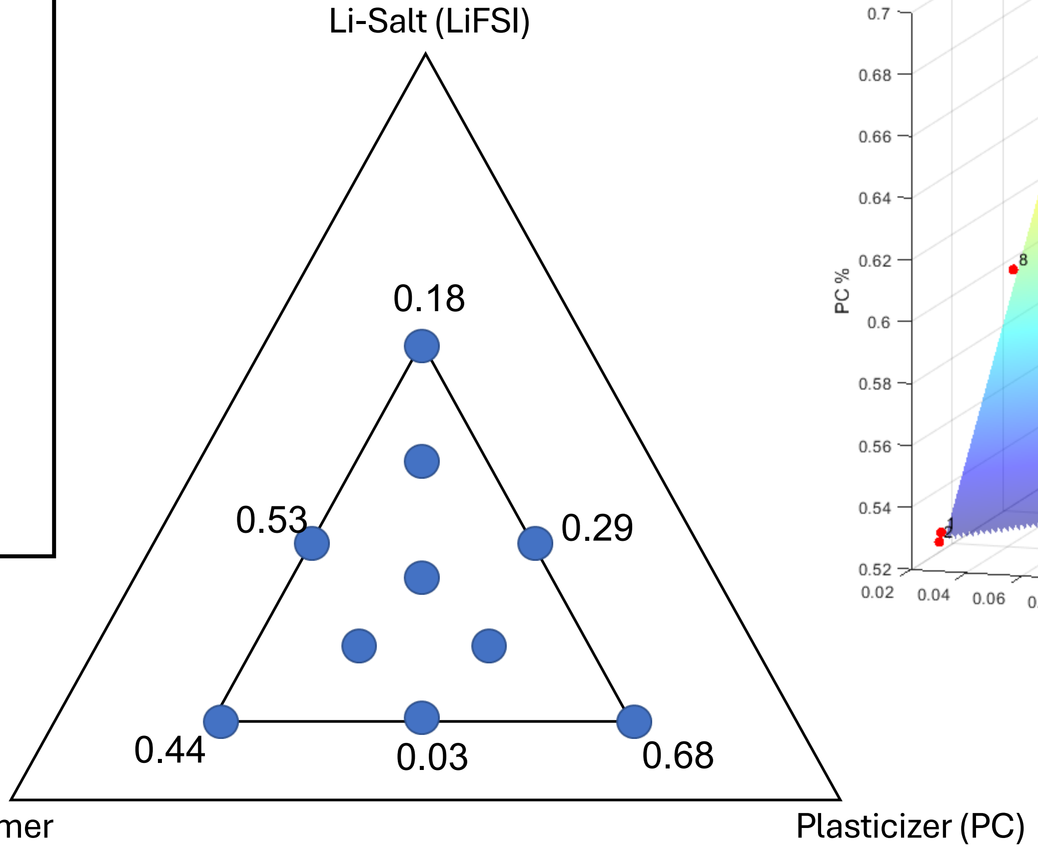
Approaches: To develop high-performance polymer electrolytes suitable for practical solid-state battery system, the formulation of various polymer electrolytes can be optimized using design of experiments (DoE). A series of electrochemical and structural characterization techniques were employed at VTT. Rheology analysis was used to study the mechanical properties of the polymer electrolytes, while electrochemical impedance spectroscopy provided insights into their ionic conductivity and interfacial stability. Thus, the optimal formulation is achieved by combining DoE to key characterization results and electrochemical performance assessments.

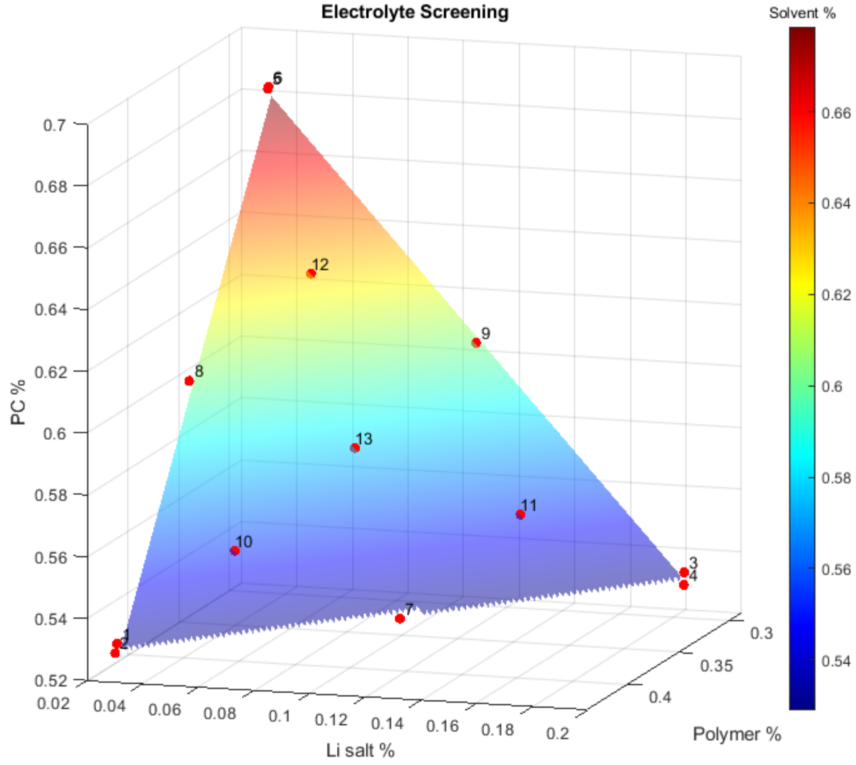
Additionally, a cellulose separator has been developed by Tervakoski by adjusting such properties as porosity and permeability for infiltrating SPE into the structure.

Design of Experiments for Polymer Electrolyte Optimization

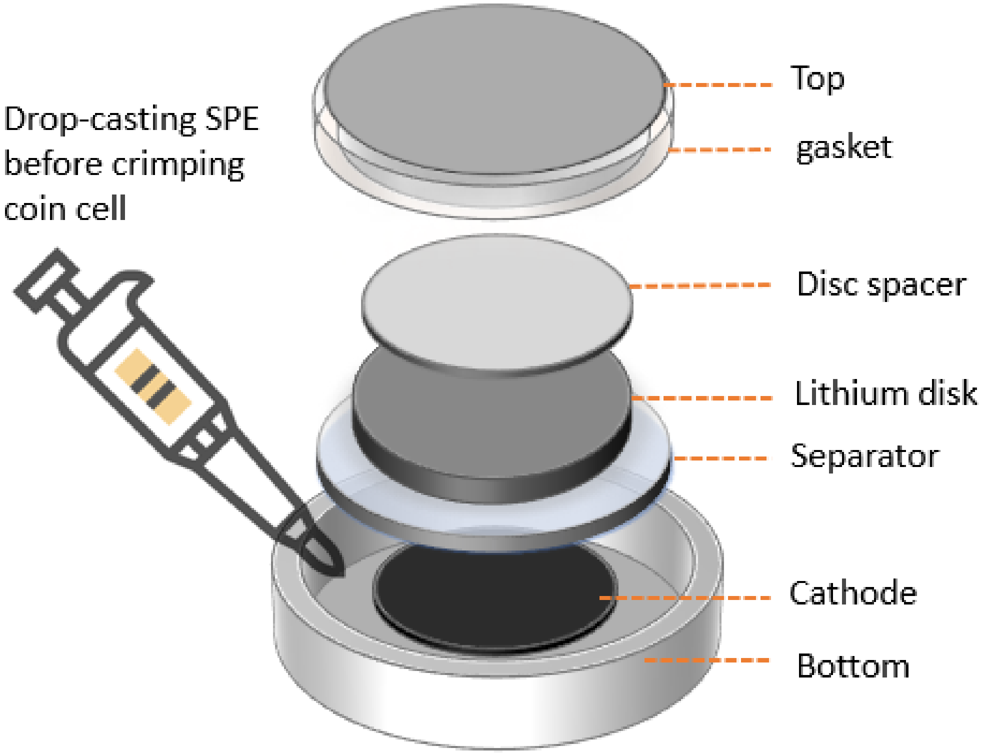
Factors:

Design
- 3 main components
  - Polymer: 0.29-0.44
  - LiFSI: 0.03-0.18
  - PC: 0.53-0.68
- 13 experiments
- 2 parameters to optimize
  - electrochemical performance
  - mechanical strength





Electrochemical Performance: Rate capability

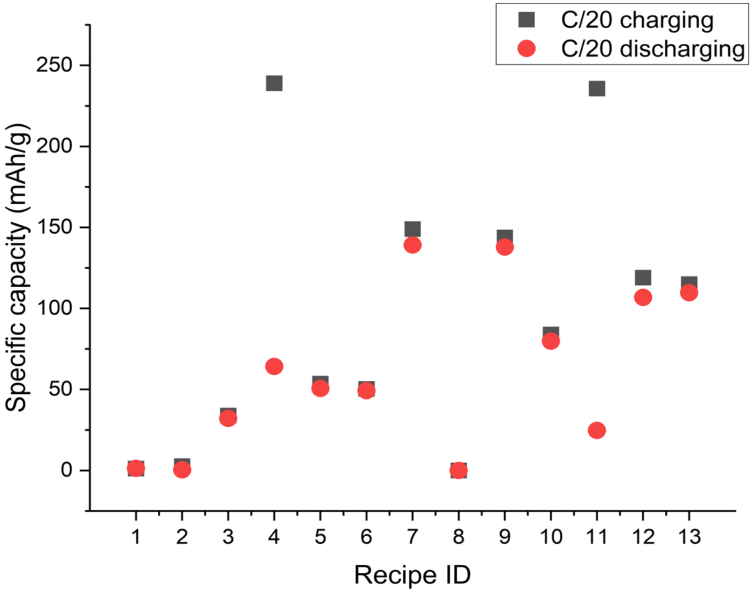


Drop-casting SPE before crimping coin cell

Curing time 16h at 70 ℃

Measurement procedure for rate capability test.

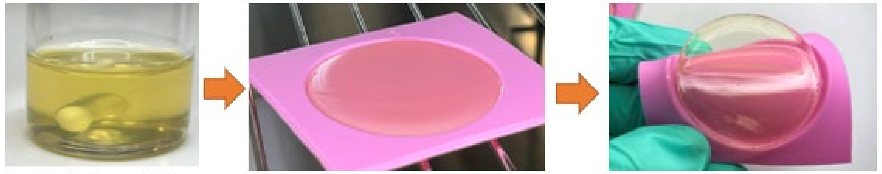
Cycle number	C-rate	Current density (mA/cm²)	Current density (mA/g)	Comments
1	C/50	0.0375	3.10	Formation cycle
2-4	C/20	0.0938	7.75	Low C-rate
5-9	C/10	0.188	15.5	High C-rate
10-14	C/5	0.375	31.0	High C-rate
15-19	C/2	0.938	77.5	Maximum C-rate
20-22	C/20	0.0375	3.10	Recovery capability after high C-rate



Mechanical Strength: Oscillatory Rheometry

Sample Preparation

a)

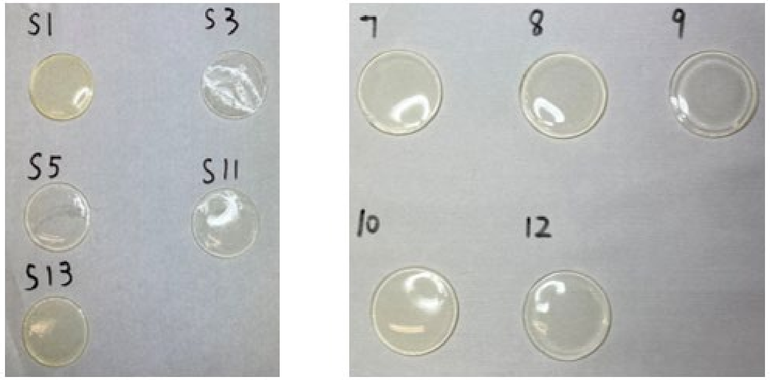


Electrolyte mixture

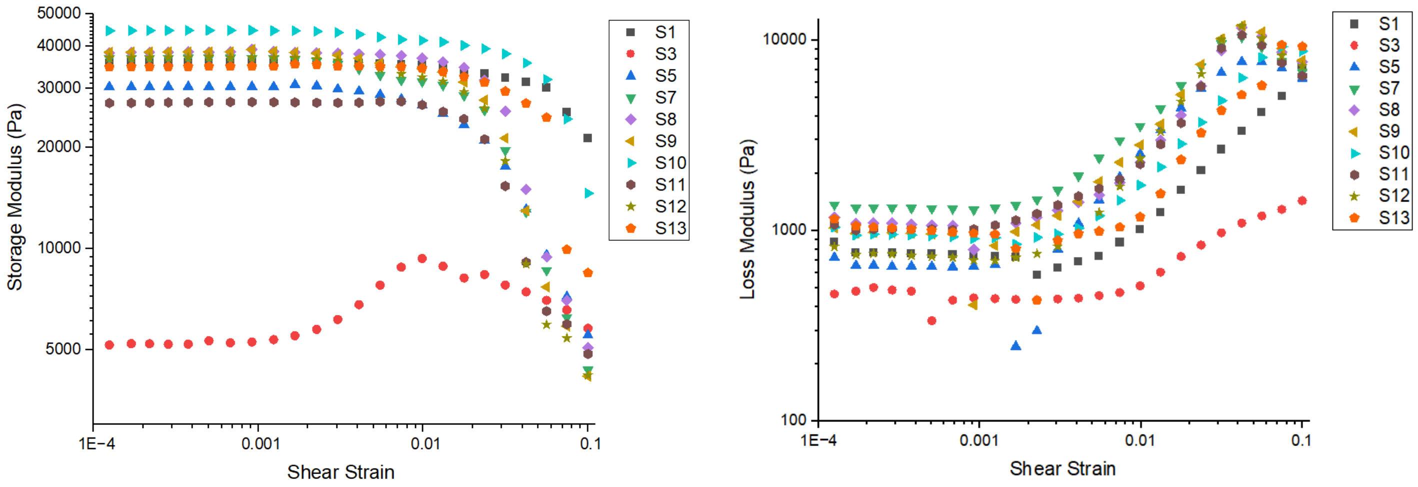
Transfer To the silicone Mould

Cure in 70°C oven for 6 hours

b)



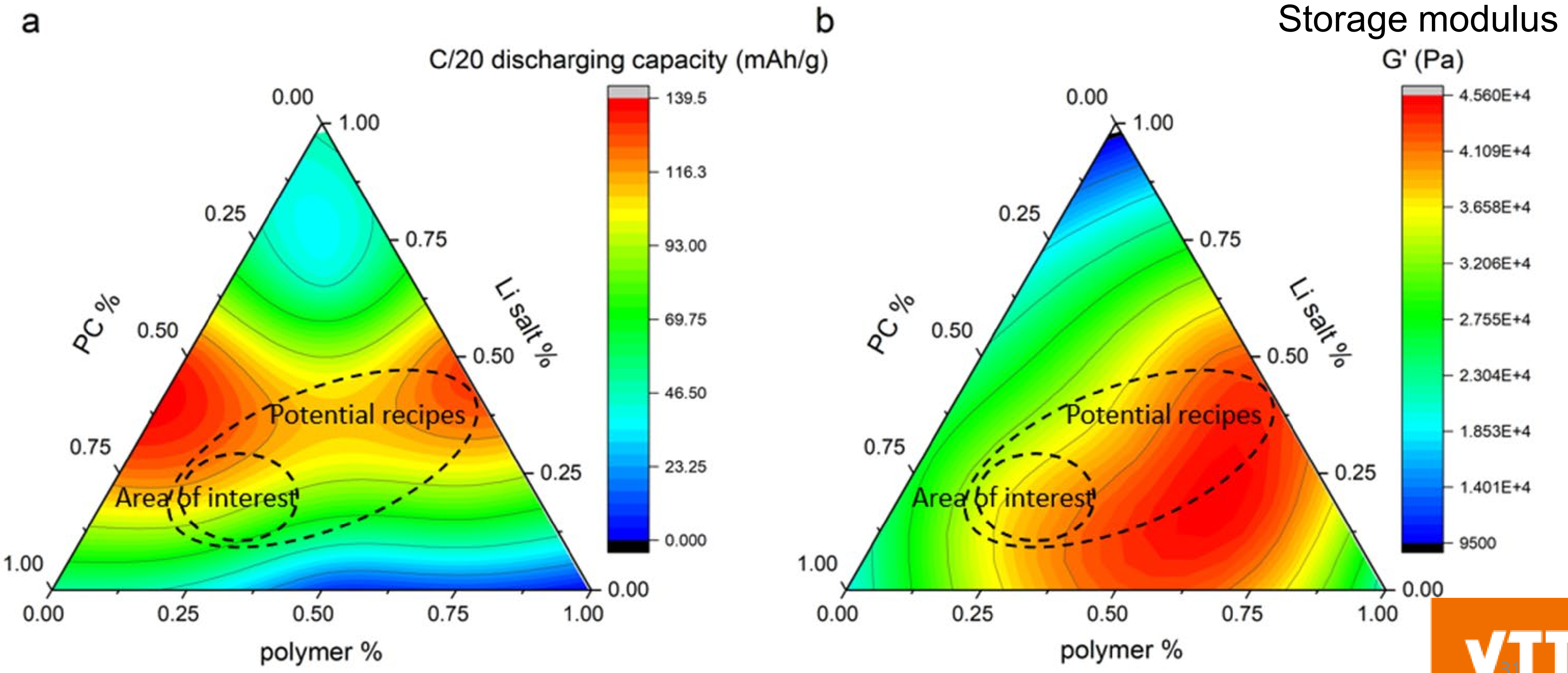
Amplitude sweeps across various samples



Linear viscoelastic region under shear strains less than 1%

Correlates to microstructure properties

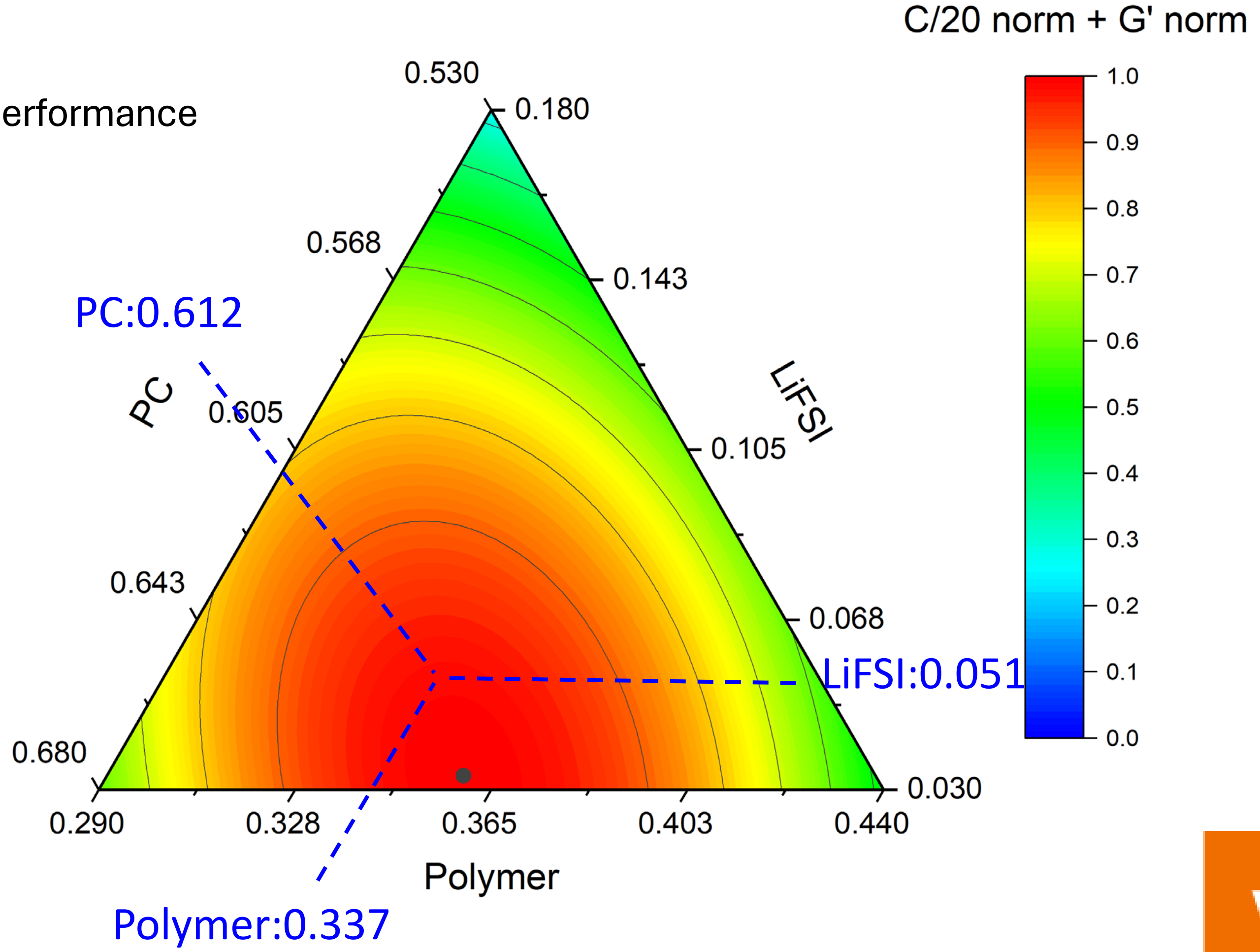
Coupling mechanical strength with electrochemical performance



Coupling mechanical strength with electrochemical performance

Sum of normalized parameters optimized

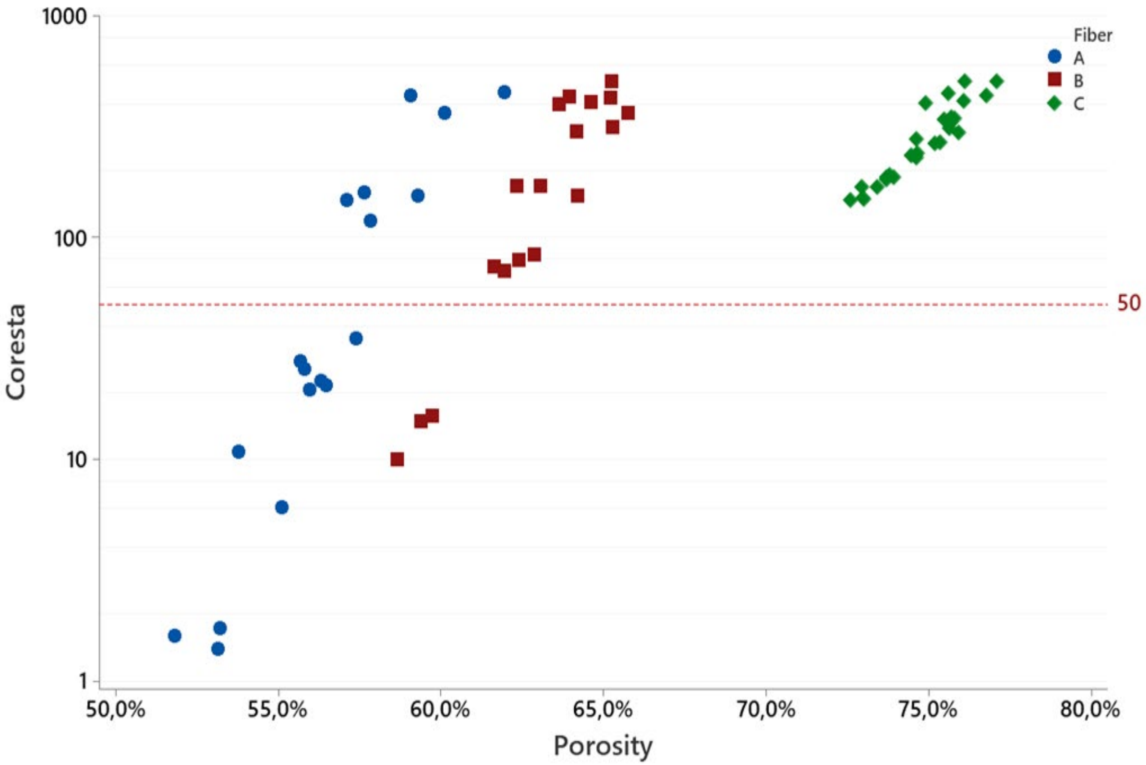
→ slightly adjusted for higher electrochemical performance



Cellulose separators

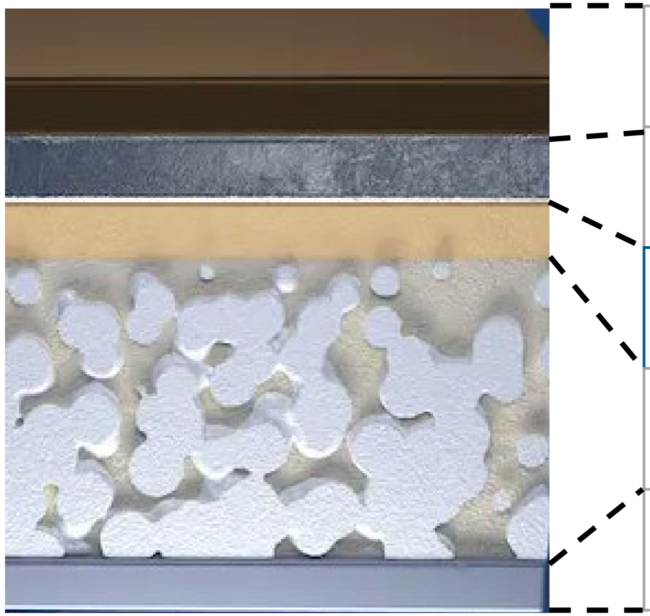
Main achievements:

- Developing a porous separator structure (up to 75 %) to work as a structural element for solid electrolytes
- Different raw-materials tested to reach desired combination of porosity and air permeability of the separators
- Feasibility confirmed by VTT coin cell tests



Full Cell

Cross-section of the NextGenBat solid-state battery



Cu current collector

Li metal PULSEDEON Tampere University

delfort separator + electrolyte VTT

VTT electrolyte + NMC cathode A! Aalto University

Al current collector

with coatings

Beneq / PULSEDEON

A! Aalto University Tampere University

The final goal was to demonstrate operation of solid state batteries assembled from the optimized components developed during the NextGenBat project.

Full cell demonstration

A demonstration was conducted by assembling coin cells with the full cell configurations. They composed of the following optimized components:

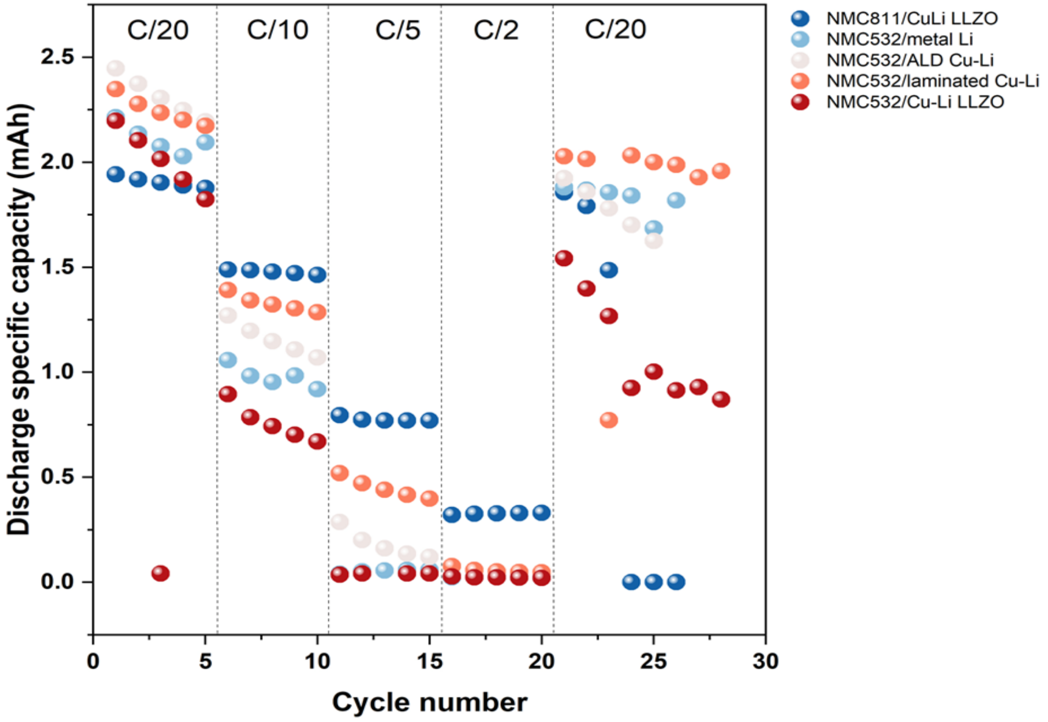
- anode: ALD-coated PLD thin-film lithium (10 μm)
- cathode: LiW_xO_y-coated NMC811
- a cellulose separator
- an injected polymer electrolyte

This demonstration is a critical step toward practical solid-state battery systems.

For reference, batteries with otherwise similar structure but a commercial NMC532 (as a reference) instead of the coated NMC811 were assembled.

The rate performance tests were performed at the voltage window of 3.0 – 4.1 V at 60 °C.

The NMC532-based cells show superior rate capability compared to NMC811. It appeared that NMC532 is more compatible with the polymer electrolyte and has better electrochemical stability in this system.



Dissemination and Communications

Publications

1) P. S. Llanos, Z. Ahaliabadeh, V. Miikkulainen, X. Kong, F. Obrezkov, J. Lahtinen, L. Yao, H. Jiang, U. Lassi, T. Kallio, Structural and Interfacial Stability of a Coated Ni-Rich Layered Oxide Cathode at High-Voltage Operation, Materials Today Energy (2025) 101862. doi.org/10.1016/j.mtener.2025.101862

2) P. S. Llanos, A. R. Bogdanova, F. Obrezkovo, N. Farrahi, T. Kallio, Impact of powder and electrode ALD coatings on the performance of cathode materials for lithium-ion batteries, Energy Advances (2025) 364-386. DOI: 10.1039/D4YA00583J

3) A. R. Bogdanova, F. A. Obrezkov, E. M. Khabushev, X. Kong, T. Kallio, Binder-free LiNi0.8Mn0.1Co0.1O2 electrode enabled by single-walled carbon nanotube coating for Li-ion batteries, Journal of Energy Storage 104 (2024) 114627. doi.org/10.1016/j.est.2024.114627.

4) B. Wang, X. Kong, F. Obrezkov, P. S. Llanos, J. Sainio, A. R. Bogdanova, A. Kobets, T. Kankaanpää, T. Kallio, Understanding of a Ni-rich O3-layered Cathode for Sodium-ion Batteries: Synthesis Mechanism and Al-gradient Doping, Small (2024) 2408072. doi.org/10.1002/smll.202408072

5) Z. Ahaliabadeh, V. Miikkulainen, M. Mäntymäki, S. Mousavihashemi, L. Yao, H. Jiang, S. Huotari, T. Kankaanpää, T. Kallio, Surface and grain boundary coating for stabilizing $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ based electrodes, ChemSusChem (2024) e202400272. DOI: 10.1002/cssc.202400272

6) S. Mousavihashemi, E. M. Khabushev, J. Lahtinen, A. R. Bogdanova, I. V. Novikov, D. V. Krasnikov, A. G. Nasibulin, T. Kallio, A binder-free Nickel-Rich cathode composite utilizing low-bundled single-walled carbon nanotubes, A binder-free Nickel-Rich cathode composite utilizing low-bundled single-walled carbon nanotubes, Advanced Materials Technologies (2024) 2301765. doi.org/10.1002/admt.202301765

7) Z. Ahaliabadeh, V. Miikkulainen, M. Mäntymäki, M. Colalongo, S. Mousavihashemi, L. Yao, H. Jiang, J. Lahtinen, T. Kankaanpää, T. Kallio, Stabilized Nickel Rich Layered Oxide Electrodes for High Performance Lithium-Ion Batteries, Energy & Environmental Materials (2024) e12741. doi.org/10.1002/eem2.12741

8) P. S. Llanos, Z. Ahaliabadeh, V. Miikkulainen, J. Lahtinen, Y. Lide, H. Jiang, T. Kallio, High Voltage Cycling Stability of LiF-coated NMC811 electrode, ACS Applied Materials & Interfaces 16 (2024) 2216. doi/10.1021/acsami.3c14394

9) J. Kosir, S. Mousavihashemi, M. Suominen, A. Kobets, B. P. Wilson, E.-L. Rautama, T. Kallio, Supervalent doping and its effect on the thermal, structural and electrochemical properties of $Li_7La_3Zr_2O_{12}$ solid electrolytes, Materials Advances 5 (2024) 5260 - 5274. DOI: 10.1039/D4MA00119B

10) Z. Ahaliabadeh, X. Kong, E. O. Fedorovskaya, T. Kallio, Extensive comparison of doping and coating strategies for Ni-rich positive electrode materials, Journal of Power Sources 540 (2022) 231633. https://doi.org/10.1016/j.jpowsour.2022.231633

Conferences and workshops

1) T. Kallio, SWNTs for Enhancing Performance and Sustainability of Electrochemical Energy Conversion Devices, Invited talk, 2024 MRS fall meeting & exhibition, 01.- 06.12.2024, Boston, USA
2) S. pakseresht, T. Kallio, Atomic Layer Deposition of Lithium Metal: A Strategy for Enhanced Stability in Lithium Batteries, 38th Topical meeting of the International Society of Electrochemistry, Oral presentation, 11.09.2024 Manchester, UK.
3) T. Kallio, Characterizing materials for electrochemical energy conversion applications, Synchrotron Light Finland 2023 Conference, Invited Talk, 30.11. – 01.12.2023 Espoo, Finland.
4) T. Kallio, Engineering $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ 2 Properties by Artificial Cathode Electrolyte Interface, ITICAT 2023, Invited Talk, 24.-25.8.2023, Espoo, Finland.
5) T. Kallio, Coatings for Engineering $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ Properties, 74th Annual Meeting of the International Society of Electrochemistry, oral presentation, 03.- 08.09.2023 Lyon, France.
6) P. S. Llanos, Z Ahaliabadeh, T. Kallio, High Voltage Cycling Stability of LiF-coated NMC811, 74th Annual Meeting of the International Society of Electrochemistry, poster presentation, 03.-08.09.2023, Lyon, France.
7) B. Ali, M. Colalongo, T. Schulli, T. Kallio, J. Drnec, T. Kankaanpää, Mechanistic Investigation of Zr coating in NMC811 Cathode Material for Li Batteries, 74th Annual Meeting of the International Society of Electrochemistry, poster presentation, 03.-08.09.2023 Lyon, France.
8) T. Kallio, Influence of coatings on nickel rich positive electrode material properties, NanoOstrava, Invited Talk, 15.-18.5.2023, Ostrava, Czech Republic.
9) J. Kosir, T Kallio, Supervalent doping and its effect on the thermal, structural and electrochemical properties of $Li_7La_3Zr_2O_{12}$ solid electrolytes, 15th International Symposium on Systems with Fast Ionic Transport (ISSFIT15), oral presentation, 18. – 22.04.2023, Dubrovnik, Croatia.
10) T. Kallio, Coatings for Engineering $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ Properties, The ISE Regional Meeting, 16.-19.8.2022, Prague, Czech.
11) T. Kallio, Structural Engineering for Enhancing Electrode Active Material Durability, Workshop on Advances in Physics and Chemistry of Energy Conversion and Storage, Invited Talk, 06.-08.07.2022, München, Germnany.
12) Z. Ahalibadeh, Surface Coating and Grain Boundary Engineering of NMC811 Materials for Next-Generation Li-Ion Batteries, oral talk, ALD 2022 (International Conference on Atomic Layer Deposition) 26.-29.6.2022, Ghent, Belgium.
13) A. Cook, “Roll-to-roll ALD Coatings for Battery Cell Interfaces”, ALD 2022 (International Conference on Atomic Layer Deposition) 26th -29th June 2022, Ghent, Belgium – Invited speaker.

Fairs

1) T. Kallio, New Materials for Electrochemical Energy Conversion & Storage, kutsuttu puhe, 08.-09.06.2022, Chem Bio Fairs, Helsinki, Suomi.T. Kallio, Coatings for Engineering LiNi0.8Mn0.1Co0.1O2 Properties, The ISE Regional Meeting, 16.-19.8.2022, Praha, Tsekki
2) A. Cook, S, Sneck, The Battery Show Europe, Stuttgart, Germany, 30th November – 2nd December 2021. – Booth at exhibition.
3) A. Cook, H. Velasco, T. Kääriäinen, 39th International Battery Seminar and Exhibition, Orlando, Florida, USA, 28th – 31st March 2022 – Booth at exhibition
4) A. Cook, T. Nishiyama, "Roll-to-roll ALD Coatings for Battery Cell Interfaces", 2nd World Smart Energy Week – Battery Japan, Makuhari Messe, Tokyo, Japan, 31st August – 2nd September 2022. – Showcase talk and booth at exhibition
5) A. Cook, Battery Show North America, Detriot, USA, 13th – 15th September 2022. – Exhibition attendee.
6) A. Cook, H. Velasco, T. Kääriäinen, "Roll-to-roll ALD Coatings for Battery Cell Interfaces", 40th International Battery Seminar and Exhibition, Orlando, Florida, USA, 20th – 23rd March 2023 – Presentation and booth at exhibition
7) A. Cook, T. Nishiyama, "Roll-to-roll ALD Coatings for Battery Cell Interfaces", Smart Energy Week – Battery Japan, Makuhari Messe, Tokyo, Japan, 13th – 15th September 2023. – Showcase talk and booth at exhibition
8) A. Cook, "Roll-to-roll ALD Coatings for Battery Cell Interfaces", 41st International Battery Seminar and Exhibition, Orlando, Florida, USA, 12th – 15th March 2024 – Presentation at seminar.

Other activities

1) Z. Ahaliabadeh, Stabilized Positive Electrodes for High Performance Lithium-Ion Batteries, doctoral thesis, 25.10.2024, Espoo, Finland
2) Z. Wu, Processing of polymer electrolyte in solid-state lithium-Ion batteries, master thesis, 29.08.2023, Espoo, Finland.
3) M. Riikonen , The Impact of Atomic Layer Deposition on the Cycle Life of Metallic Lithium Anodes, Bachelor thesis, 12. 05. 2024, Espoo, Finland.
4) T. Kallio, Litiumakun toiminta, Invited Talk, Suomen Tiedeseuran symposio, 12.10.2023, Helsinki, Finland.
5) T. Kallio, visit to Otaniemi lukio to introduce battery related research activities, 16.02.2023, Espoo, Finland.
6) T. Kallio, Tulevaisuuden näkymiä materiaalinäkökulmasta, invited talk, Eduskunta, Tulevaisuusvaliokunta, 24.05.2022, Helsinki, Finland.

Dissemination / “Greetings from”


2021


2022


2023


2022


2022


2023


2024

Final remarks

Conclusion

The NextGenBat initiative successfully validated a combination of protective thin-film coatings, improved electrode materials, and novel solid-state electrolytes, laying the groundwork for safer and higher energy batteries. Through advanced thin-film techniques like ALD and PLD, the project produced lithium electrodes with stable interfaces and demonstrated that nickel-rich NMC811 cathodes could be strengthened by nanocoatings, and explored polymer and ceramic electrolytes as viable alternatives to liquid-based cells. This comprehensive approach culminated in the assembly of pilot-scale cells that showed promise both in terms of performance and handling safety. A key takeaway is that carefully engineered electrode–electrolyte interfaces — particularly with ultrathin film coatings — are critical to improving cycle life and preventing the dendrite growth that can lead to battery failures.

Key Insights and Next Steps

- Strengthening collaboration across universities, research institutes, and industry partners has significantly enhanced Finland's capabilities in advanced battery materials processing and testing. This collaborative effort paved the way for new cell concepts and provided a clear route toward pilot-scale manufacturing. Beyond generating important scientific insights, the shared infrastructure and knowledge transfer set the stage for future projects that will expand on these breakthroughs.
- Creating robust platforms for future projects — by expanding shared testing facilities, open-access data, and practical training — will encourage ongoing research, attract new talent, and help Finland maintain its leadership position in next-generation battery technologies.