



Technische  
Universität  
Braunschweig



# INTERNATIONAL BATTERY PRODUCTION CONFERENCE

1 to 3 November 2021

## CONFERENCE BROCHURE

# WELCOME



Dear participants of the IBPC,

batteries are a key driver of the global mobility revolution and the core of the energy turnaround. The last years have shown an enormous growth in demand, production capacity and technological progress in terms of energy capacity, fast charging ability and cost reduction. On top, several manufacturers have the ambition to produce the most environmentally friendly battery on earth. The end of the innovation process is not yet in sight. For example, we are experiencing a fast innovation rate regarding new materials as well as cell and pack designs that need to be transferred from the lab-scale to the industrial mass production.

When we started preparing for this year's conference, still under conditions and regulations around the Covid-19 Pandemic, we hoped that the IPBC 2021 could return as an on-site event. Thrilled with the success of the fully online IBPC 2020 with over 200 participants and 37 exciting presentations on recent advancements in battery production and six keynotes, we started to plan the 2021 conference, discussing how our society has adapted to handle the still ongoing regulations and difficulties. With the ever-growing number of vaccinated people, we are delighted to welcome you to the IBPC 2021 as on-site event in Braunschweig to discuss recent developments and research around battery production and its circular economy.

This year's plenary talks, presentations and poster sessions address the electrode, cell, module and pack production, cell and pack design as well as safety, simulation, sustainability and digitalization of the battery production. We are also happy to announce sessions from the German competence clusters ProZell and greenBattNutzung as well as European consortia as LiPLANET. In addition to liquid electrolyte lithium-ion batteries, the conference addresses the production of all-solid-state and solid polymer batteries with two exciting sessions.

We are very happy to welcome speakers and their presentations that deal with the entire battery value chain up to the battery recycling and are excited about their contribution to the battery production community. We are particularly thankful for the support of our partners, especially VDMA Battery Production. A special thank goes to our sponsors Coperion GmbH, Custom Cells Itzehoe GmbH, Bio-Logic Science Instruments SAS, EIRICH GROUP, Netzsch Feinmahltechnik GmbH and Keysight Technology GmbH. Their support enables us to maintain the high quality of the conference under these difficult circumstances. We warmly welcome you and wish you all interesting talks and exciting discussions.

Prof. Dr.-Ing. Christoph Herrmann & Prof. Dr.-Ing. Arno Kwade

# BATTERY LABFACTORY

The Battery LabFactory (BLB) stands for an open research infrastructure to investigate and develop electro-chemical storage devices from laboratory to pilot plant scale. The research spectrum covers the entire value cycle, from material, electrode and cell manufacturing, up to recycling as well as the subsequent active material resynthesis from recycled raw materials.

The BLB holds the production infrastructure and characterizing equipment to develop large-sized batteries as well as battery modules and packs. This allows the research on fundamental and application-oriented aspects. The scope of the BLB is to establish a knowledge-driven electrode and cell production to accomplish a fast transfer of R&D-based developments into technical or pilot scale production processes. For this purpose, engineers and scientists with different areas of expertise join forces in the transdisciplinary team of the BLB. In detail, 9 institutes of the TU Braunschweig, two from TU Clausthal and one from LU Hannover combine their knowledge and scientific competence. Furthermore, the Fraunhofer Institute for Surface Engineering and Thin Films IST and the Physikalisch-Technische Bundesanstalt Braunschweig (PTB) complete the joined Battery LabFactory Braunschweig.

The Battery LabFactory represents an open platform for R&D on processes, cell design, diagnostic and simulation of today's and future battery technologies.



## Sustainable Circular Production of Batteries



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# PARTNERS



BioLogic Science Instruments SAS, with its corporate office in Seyssinet-Pariset, is a French designer and manufacturer who offers powerful laboratory research instruments and software since 1983. The company is characterized by its close relationships with battery development and testing laboratories around the world. With our high precision, high-performance product range of potentiostats, galvanostats, battery cyclers and impedance analyzers we are able to cover a wide field of research: New battery technology, fuel cells, photovoltaics, corrosion, super capacitors, and bio-sensors to name a few. The modularity of our multi-channel potentiostats and our wide range of battery characterization accessories offers highest flexibility to configure the appropriate device according to your needs.

[www.bio-logic.net](http://www.bio-logic.net)



Coperion is the international market and technology leader in compounding systems, feeding technology, bulk materials handling systems and services. Coperion designs, develops, manufactures and maintains systems, machines and components for the plastics, chemicals, pharmaceuticals, food and minerals industries. Coperion has 2,500 employees, nearly 30 sales and service companies worldwide and a total of 30 extrusion systems permanently available for testings.

**Your Partner in Extrusion of Battery Materials:** Coperion is your partner for the continuous production of anode and cathode slurries, battery separator films and solid-state battery compounds. The combination of high-class Coperion ZSK twin screw extruders with high-accuracy Coperion K-Tron feeders and reliable rotary valves ensures continuous processes with high reliability, long operating times and constant high product quality.

[www.coperion.com](http://www.coperion.com)



## **CUSTOMCELLS® - Advanced battery cell development and production**

CUSTOMCELLS® is one of the leading companies in the fields of development and series production of special lithium-ion battery cells. Based upon modern process technologies, flexible production environments, access to an extend supply chain and thru a highly experienced R&D team CUSTOMCELLS® develops customer's application driven cell technologies and is handling the industrialization of the developed cell concepts. CUSTOMCELLS® automated series production is one of the most modern battery cell production facilities in Europe. Traceable cell assembly processes, a flexibility in cell chemistry and format, high precision production processes including state-of-the-art quality assurance systems and a unique, modern cell formation and testing makes CUSTOMCELLS® to a leading technology specialist and supplier for the automotive, aerial vehicles, aerospace, medical, oil and gas and for the maritime industry.

[www.customcells.org](http://www.customcells.org)



The EIRICH GROUP is a family-managed group of companies operating in the field of special mechanical engineering with its headquarters in Hardheim, Germany. As one of the world's leading manufacturers of machines and systems for processing raw materials, EIRICH develops, plans and manufactures advanced technologies for mixing, granulating, dispersing, kneading, reacting, tempering and fine grinding since 1863. Solutions for process technology and automation round the portfolio off. A staff of approx. 1,300 employees work at the 15 sales and production sites all over the world.

[www.eirich.de](http://www.eirich.de)



Keysight is the world's leading electronic measurement company, with 13,000 employees in 150 locations worldwide. We offer a full range of formation and test solutions with over 25 years of experience in cell and battery characterization. State-of-the-art software improves production quality, utilization of battery testers, along with inventory tracking. Cloud-based software provides secure access to data from anywhere in the lab.

[www.keysight.com](http://www.keysight.com)



The NETZSCH Business Unit Grinding & Dispersing offers an extensive machine program for process engineering, providing solutions for wet and dry grinding, mixing, dispersing and deaeration. Long-term experience, consistent development work, daily contact with our customers and developments with more than 100 patents ensure our technical competence and further attest to our quality-consciousness. The bundling of process-engineering expertise and the extensive machine program, ranging from laboratory to production machines to complete production lines, is unique worldwide.

[www.netzsch-grinding.com](http://www.netzsch-grinding.com)

# PARTNERS



Batterieproduktion

The VDMA Battery Production department is the partner for all questions relating to machine and plant construction in the field of battery production. The member companies of the department supply machines, plants, machine components, tools and services for the entire process chain of battery production: From raw material preparation, electrode production and cell assembly to module and packaging production. The current focus of VDMA battery production is on Li-ion technology. We research technology and market information, organize customer events and road shows, hold our own events, such as the annual conference, which has established itself as an important industry meeting place, and are in dialogue with research and science on current topics and on joint industrial research.

<http://battprod.vdma.org>



Kompetenzcluster zur Batteriezellproduktion

The aim of the competence cluster is to research and improve the entire process chain of the battery cell production and assess the influence of each individual production step on cell properties, product development costs and sustainability. In cooperation with the BMBF, the KLiB and the ProZell management board, the active ProZell network successfully creates synergies between science and industry. Together, they lay the foundation for a high-performance and cost-effective battery cell "Made in Germany".

[www.prozell-cluster.de](http://www.prozell-cluster.de)



Scientific Partner

Fraunhofer  
IST

The Fraunhofer Institute for Surface Engineering and Thin Films IST in Braunschweig is an innovative partner for research and development in surface technology, with expertise in the associated product and production systems. Together with customers from industry and research we develop customized and sustainable solutions: from prototypes, through economic production scenarios, to upscaling to industrial magnitudes - and all this whilst maintaining closed material and substance cycles. Coating and surface technology is the key to innovative products and systems: Through modification, patterning and coating of the surface, a wide range of functions and functionalities can be realized. On the basis of a broad spectrum of processes and coating materials we create the optimum process chain for the respective task.

[www.ist.fraunhofer.de](http://www.ist.fraunhofer.de)



Querschnittsinitiative Batterie-Lebenszyklus

The BMBF-funded research initiative greenBattNutzung combines the competence clusters Recycling & Green Battery (greenBatt) & Battery Utilization Concepts (Batt-Nutzung). greenBatt aims to develop, design and apply innovative technologies, methods and tools for a sustainable battery life cycle and closed material and resource loops. BattNutzung's mission is to develop, design and apply new concepts for battery system evaluation, linking findings at cell level to the level of battery technology requirements. Both clusters combine a total of 55 research institutes that collaborate in 29 research projects. The initiative is accompanied by an industrial management board ensuring an active exchange between science and industry.

[www.greenbatt-cluster.de](http://www.greenbatt-cluster.de), [www.battnutzung-cluster.de](http://www.battnutzung-cluster.de)

## GET AHEAD. BREAK THE LIMITS.



### Production of lithium-ion batteries Trailblazing process technology for perfect electrode mixes

Gigafactories for battery production place new demands on processes - especially in the processing of electrodes. The quality of the electrodes influences the performance of your storage cells. The processing of a first-class electrode mix - slurry, plastic body or even structured dry mix - is a demanding task. Trust a technology partner with in-depth material and process knowledge as well as decades of experience.

**Eirich MixSolver® with EvacMix® technology and you benefit from:**

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From lab scale to gigafactory

**Sustainable process**

Efficient and environmentally friendly

**High performance batteries**

Mixing technology for highly reproducible process

**Clean turnkey solution**

Strong capabilities for metering and handling

**Continuous coater supply**

The best of batch and conti

**Future-oriented concept**

For wet- and dry-processed electrodes



unique mixing principle



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# COPERION TECHNOLOGY FOR CONTINUOUS PRODUCTION OF BATTERY MATERIALS.

- + For solid state batteries, anode and cathode slurries and separator films
- + First-class twin screw extruders with excellent mixing behavior and consistently high product quality
- + High-accuracy and dust-tight feeders for best possible recipe control
- + Containment secure material handling



## KEYSIGHT

Keysight is the world's leading electronic measurement company, with 13,000 employees in 150 locations worldwide. We transform today's cell and battery measurement experience with innovative technologies for material research, manufacturing, and validation.

Cloud-based software is the key to keeping up with processes and tracking and managing data from anywhere in the lab.

### Cell Formation

Keysight solutions scale with cell size, production level, and process complexity. PathWave Cell Formation Manager software tracks formation ensuring quality and consistency.

### Cell/Battery Test Lab

PathWave Lab Operations for Battery Test software and Scienlab testers verify cells and battery packs—manage and fully utilize test assets from any location.



# PROGRAMME

## CONFERENCE DAY 1 | Nov. 1st

- 8:30 | Arrival of Attendees
- 9:00 | Welcome by the Conference Chairs
- 9:15 | Keynote by Dr. Tim Schulze, BMWi
- 9:50 | Keynote by Stefano Saguatti, Manz Italy  
Manz FBT-Future Battery Technologies: A sustainable strategy for Lithium battery cells Mass Production made in Europe
- 10:25 | Break
- 10:45 | **Parallel Sessions**
- |  |  |
|--|--|
| <p><b>Electrode Production (I)</b><br/><b>Room Maschinenhalle I Chair: Prof. A. Kwade</b><br/>Enabling aqueous processing of positive electrodes for lithium ion batteries – Challenges and opportunities for process and electrode design<br/><i>Markus Börner, University of Münster/MEET</i><br/>Development of a solvent reduced electrode production process integrating coating, drying and calendaring<br/><i>Eike Wiegmann, TU Braunschweig/iPAT</i><br/>Performance increase and process enhancements for capacitors and batteries production by aLITE-Technology<br/><i>Kai K. O. Bär, adphos Innovative Technologies GmbH</i></p> | <p><b>Cell Assembly</b><br/><b>Room Nimes I Chair: Prof. K. Dröder</b><br/>Inline X-ray Metrology for Lithium-Ion Battery Cells<br/><i>Markus Möller, Viscom AG</i><br/>The warmer, the faster?- An investigation of the temperature dependence on the wetting process<br/><i>Nicolaj Kaden, TU Braunschweig/IWF</i><br/>Quality assessment for laser-based contacting of lithium-ion battery cell components using inline process monitoring<br/><i>Sophie Grabmann, TU München/IWB</i></p> |
|--|--|
- 11:30 | Discussion
- 11:45 | Lunch Break
- 12:45 | **Postersession and Presentation**
- |  |   |
|--|---|
| <p><b>Room Maschinenhalle</b><br/>Inline Monitoring of Battery Electrode Lamination Processes Based on Acoustic Measurements<br/><i>Nikolas Dilger, Fraunhofer IST</i><br/>Introducing Lignin as a Bio-based Binder Material for NMC Electrode Production an Aqueous<br/><i>Silje Nornes Bryntesen, NTNU</i><br/>Fully continuous mixing technology for both conventional electrode slurry and solvent-reduced electrode masses<br/><i>Christian Hänsel, Bühler AG</i><br/>STATE-OF-THE-ART SURFACE INSPECTION SOLUTIONS- Intelligent interaction of defect detection incl. geometric measurements with web guiding systems in the electrode manufacturing process<br/><i>Klaus Hamacher, BST GmbH</i></p> | <p><b>Room Nimes</b><br/>The Effects of Ultraviolet Radiation on Inorganic Solid Electrolyte after Direct Laser Sintering<br/><i>Houssin Wehbe, TU Braunschweig/ifs</i><br/>Solvent-free Processing of Solid State Electrodes Based On A Plastic Crystal Electrolyte System<br/><i>Arnaud du Baret, Fraunhofer IKTS</i><br/>Scale-up of mechanochemical synthesis of sulfide-based solid electrolytes for all-solid-state batteries<br/><i>Michael Grube, Fraunhofer IST</i><br/>CO2 snow jet cleaning – increasing the yield in battery production<br/><i>Jonas Gude, acp systems AG</i></p> |
|--|---|

- 13:45 | Keynote by Prof. Dr.-Ing. Petr Novák, Institute of Energy and Process Systems Engineering TU Braunschweig  
Energy Storage in Batteries: Wishful Thinking, Unpleasant Facts, and the Role of Scientific Characterization Techniques
- 14:20 | **Parallel Sessions**
- |  |  |
|--|--|
| <p><b>Electrode Production (II)</b><br/><b>Room Maschinenhalle I Chair: Prof. A. Kwade</b><br/>Simultaneous two-layer slot die coating of ultra-thick LIB electrodes<br/><i>Alice Hoffmann, Center for Solar Energy and Hydrogen Research Baden-Württemberg (ZSW)</i><br/>Calendering of carbon coated nano-silicon graphite composites (Si@Gr/C)<br/><i>Jannes Müller, TU Braunschweig/iPAT</i><br/><br/>Schwarz-Primitive structured electrodes for lithium-ion battery via 3D printed wax templating<br/><i>Yige Sun, University of Oxford /Department of Materials</i></p> | <p><b>Recycling &amp; Sustainability (I)</b><br/><b>Room Nimes I Chair: Prof. C. Herrmann</b><br/>The sustainability of battery cell production in Europe<br/><i>Marlene Eisenträger, VDI/VDE Innovation + Technik GmbH</i><br/><br/>Aqueous processing of cathode slurries with nickel rich active materials and sustainable binder for green battery production<br/><i>Anton Werwein, Fraunhofer IKTS</i><br/>Characterization of spent lithium-ion batteries in different recycling process routes using automated mineralogy<br/><i>Kai Bachmann, Helmholtz Institute Freiberg for Resource Technology</i></p> |
|--|--|
- 15:05 | Discussion
- 15:20 | Break
- 15:40 | **Parallel Sessions**
- |   |   |
|---|---|
| <p><b>Electrode Production (III)</b><br/><b>Room Maschinenhalle I Chair: Prof. A. Kwade</b><br/>Laser structuring in battery production for enhancing the electrochemical performance of NMC 811 high energy electrodes for xEV energy storage systems<br/><i>Bernd Eschelmüller, AIT Austrian Institute of Technology GmbH</i><br/>Electronic pathway length manipulation and impact on conductivity networks in lithium-ion battery electrodes.<br/><i>Jake Entwistle, University of Sheffield</i><br/>Electrode structure optimization adjusting battery manufacturing parameters by applying computational EIS<br/><i>Clara Ganuza, CIDETEC</i></p> | <p><b>Module &amp; Pack Production</b><br/><b>Room Nimes I Chair: Prof. K. Dilger</b><br/>Agile Production Lines- flexible scaling for high-speed production<br/><i>Alexander Weis, SCIO Technology GmbH</i><br/>Dynamic beam shaping and forming technics for improved results in battery welding applications<br/><i>Peter Kallage, Coherent</i><br/>Automatic Machines for Leak Testing of Sealed Batteries<br/><i>Michael Klenk, MARPOSS GmbH</i></p> |
|---|---|
- 16:25 | Discussion
- 16:40 | Break



# PROGRAMME

## 17:00 | Parallel Sessions

### Battery Production 4.0 (I)

Room Maschinenhalle | Chair: Prof. C. Herrmann

Flexible scaling of future battery cells

Jan Diekmann, CUSTOMCELLS

Let's put smart to work- Future Battery Factories adapting Industry 4.0 Technologies

Klaus Eberhardt, Exyte

Digital Twin in Battery Production 4.0 – From Data Management and Traceability System to

Target-Oriented Application

Alexander Kies and Jonathan Krauß, Fraunhofer IPT

### Production of solid state batteries (I)

Room Nimes | Chair: Dr. P. Michalowski

Challenges and opportunities for solid-state players – can they be competitive on the battery market within automotive applications?

Joscha Schnell, P3 automotive GmbH

Advanced Battery Material: Challenges within the laser cutting process of lithium metal

Lars Schmidt, TU Braunschweig/ifs

Manufacturing of Composite Cathodes for Sulfide-based All-solid-state Batteries

Hans-Christoph Töpfer, Technical University of Munich

17:45 | Discussion

18:00 | Break

## 18:30 | Industry Session

Room Nimes | Chair: Prof. A. Kwade

Structuring of particle systems for the production of wet and dry electrodes with Eirich intensive mixers.

Stefan Gerl, Maschinenfabrik Gustav Eirich GmbH & Co KG

NETZSCH Helios fast and efficient dispersion system for Li-ion Battery Electrode coatings

Alberto Masi, NETZSCH-Feinmahltechnik GmbH

Coperion- Producing Battery Compounds

Anselm Lorenzoni, Coperion GmbH

BioLogic – Our Solutions to Test Your Batteries

Sandra Möller, Bio-Logic Science Instruments GmbH

19:10 | End of Day one, start of evening event



Battery Production

## VDMA Battery Production

The VDMA department is the direct contact for all questions relating machine- and plant construction. The member companies of the department supply machines, plants, machine components, tools and services for the entire process chain of battery production: From raw material preparation, electrode production and cell assembly to module and packaging production. The current focus of VDMA Battery Production is on Li-ion technology. Our activities:

- We research technology and market information: (Roadmap Battery Production Equipment 2030, Process Flyer Battery Production, business climate survey, short expert reports on employment effects)
- We organize customer events and roadshows (most recently in the USA, in China with CATL, BAK and BYD or in Korea with LG Chem and Samsung SDI but also with Automotive OEM etc.)
- We supervise fairs (Battery Japan, CIBF, Battery Show USA) and hold our own events, such as the VDMA Battery Production Annual Conference: Established itself as an important industry meeting
- We are in dialogue with research and science on current topics and on joint industrial research.
- We represent our industry in politics and the public.

If you have any questions, please do not hesitate to contact us!

Website: <https://battprod.vdma.org/en/ueber-uns>

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VDMA Battery Production

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Phone: +49 69 6603 1784





# PROGRAMME

## CONFERENCE DAY 2 | Nov. 2nd

8:30 | Keynote by Nicolas Vallin, Dassault Systèmes Industry  
Digital Twin in the Context of Next Generation Batteries

9:05 | Keynote by Patrick Bernard, SAFT  
Advanced Li-ion and beyond technologies : key materials, cells and processes

9:40 | Break

### 10:00 | Parallel Sessions

[ProZell – competence cluster for battery cell production](#)

**Room Maschinenhalle | Chair: Prof. A. Kwade**

Multilayer silicon/graphite anodes for the use in high-energy cells

*Laura Gottschalk, TU Braunschweig/iPAT*

About the vacuum post-drying process in the production of lithium-ion batteries

*Thilo Heckmann, KIT/TFT*

Reconstruction of the carbon-binder domain in Li-ion battery cathodes and its influence on the electrochemical performance

*Benedikt Prifling, Ulm University/Institute of Stochastics*

10:45 | Discussion

### 11:00 | Postersession and Presentation

**Room Maschinenhalle**

Flotation for the separation of anode and cathode materials of end-of-life-lithium-ion-batteries

*Johanna Köthe, TU Clausthal/IFAD*

Innovation in Conductive Carbon Additives for Li-ion Batteries: Improved Cell Performance and More Efficient Electrode Manufacturing

*Giulio Ferraresi, Imerys Graphite & Carbon*

Graphite Recycling from End-of-Life Lithium-ion Batteries: Processes and Applications

*Mozaffar Abdollahifar, TU Braunschweig/iPAT*

Tortuosity Investigations on Cathodes with porous nanostructured vs. bulk NMC Particles

*Luca Schneider, KIT*

[Production of solid state batteries \(II\)](#)

**Room Nimes | Chair: Dr. P. Michalowski**

Novel processing technique for making solid-state batteries with fast lithium ion transport kinetics

*Chun Huang, King's College London*

Laser sintering of ceramic-based solid-state battery materials

*Linda Hoff, Fraunhofer ILT*

Evaluation of formulation variation for scalable processing of sulfide-based all-solid-state batteries

*Mattis Batzer, TU Braunschweig/iPAT*

**Room Nimes**

Investigations of binder migration for NMC cathode slurries with compact and nanoporous structured particles

*Julian Klemens, KIT/TFT*

Optimization of edge quality in the coating of li-ion battery electrodes

*Sandro Spiegel, KIT/TFT*

The influence of formulation on the coating quality of an innovative extrusion-based coating process for li-ion electrodes

*Jann Seeba, Fraunhofer IKTS*

Exploring smart battery cell production based on a generic system architecture and an AI-enhanced process monitoring

*Xukuan Xu, Fraunhofer IPT*

### 12:00 | Parallel Sessions

[Battery Production 4.0 \(II\)](#)

**Room Maschinenhalle**

**Chair: Prof. C. Herrmann**

Towards a comprehensive semantic information structure in the battery value chain

*Simon Stier, Fraunhofer ISC*

Data-mining-based multi-criteria quality prediction in battery cell production

*Artem Turetskyy, TU Braunschweig/IWF*

Modelling of Lithium-ion Battery Electrode Calendering by Discrete Element Method (DEM)

*Ruihuan Ge, University of Sheffield*

12:45 | Discussion

13:00 | Lunch Break

14:00 | Keynote by Dr. Felix Eberle, BASF

Cathode active material synthesis: From product and process to large scale production

### 14:35 | Parallel Sessions

[Recycling & Sustainability \(II\)](#)

**Room Maschinenhalle | Chair: Prof. A. Kwade**

Recovery of spheroidized graphite from spent lithium ion batteries

*Anna Vanderbruggen, Helmholtz Zentrum Dresden Rossendorf*

Techno-economic and environmental assessment of production processes for all-solid-state batteries using the example of active material coating by atomic layer deposition

*Deidre Wolff, Fraunhofer IST*

Recycling of oxide electrolytes from all-solid-state lithium-ion batteries by acid leaching

*Kirstin Schneider, TU Clausthal/IFAD*

15:20 | Discussion

15:35 | End of Conference

[Cell formation and Quality Control](#)

**Room Nimes**

**Chair: Prof. M. Kurrat**

Optimized cell finishing through innovative plant technology and parameter-based evaluation of process and quality parameters in the virtual production system

*Nikolaus Lackner, RWTH Aachen University/TU Munich*

High Potential Test for Quality Control of Separator Defects in Battery Cell Production

*Ferry Kienberger, Keysight Labs Austria*

[Multivariable process- property relationships](#)

**Room Nimes | Chair: Prof. C. Herrmann**

The round-robin test: Towards process standardization of EU research pilot lines

*Katja Froehlich, AIT Austrian Institute of Technology GmbH*

Study of the cathode coating-drying manufacturing process by design of experiments

*Geanina Apachitei, University of Warwick*

Understanding and correcting for temperature variations when measuring self-discharge in cell production

*Manuel Moertelmaier, Keysight Technologies*

# The benchmark for energy testing devices.



## From research **VMP3/VMP-300**

- Current range from 1 pA to 800 A
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- Versatile: on-site hardware upgrades
- HPC < 10 ppm

## Via development & cycling **MPG-2xx series**

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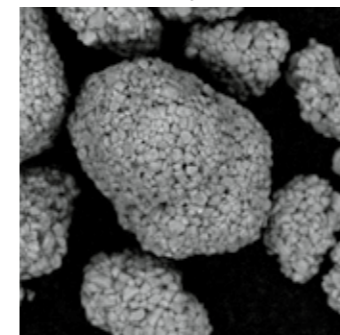
## PROCESSING OF ACTIVE BATTERY MATERIALS

### About NETZSCH

The family-owned company NETZSCH, with its Business Units Analyzing & Testing, Pumps & Systems and Grinding & Dispersing was founded in 1873. In the NETZSCH Group are currently approx. 3500 employees worldwide. The business unit Grinding & Dispersing is specialist in mechanical engineering and in supplying special machines or complete systems. The machine equipment enables the development of products on a laboratory scale just as well as the scale up to production size machines. The machines excel by their long lifetime and hereby guarantee a high reliability.

### Processing of active battery materials

NETZSCH is active with dry and wet grinding equipment. Examples for wet material preparation with agitator bead mills are: LFP (ZETA®, NEOS) and metallic anode material (ZETA®RS). After synthesis steps a gentle des-agglomeration by usage of dry working CSM-classifier mills can be performed obtaining the desired active material shape. Process examples are MNC, NCA and LMO. With ceramic machine executions metal contamination is avoided. To ensure a cost effective processing dry processing closed loop systems are in operation.



NMC after deagglomeration

### Slurries for cell production

Planetary Mixers PMH are reliable in terms of being flexible in viscosity and input materials. The high performance kneading and mixing process is supported by excellent temperature control and vacuum deaeration. In addition equipment for binder dissolution, intermediate storage and continuous deaeration is in scope of supply. In processing of conductive additives and exfoliation of carbons is obtained by Economic Dispersionizer OMEGA®. Increased battery safety was demonstrated by ceramic coated polymer films, raw materials produced in NETZSCH agitator bead mills.



Classifier Mill  
CSM CERAMIC



Agitator Bead Mill  
ALPHA® ZETA®, ceramic version



Planetary Mixer  
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# POSTER SESSION

## Electrode Production

- P1-01 Analysing the impact of particle size distribution on differently compacted graphite based electrodes via X-ray microcomputed tomography  
*Alexander Diener, TU Braunschweig | iPAT*
- P1-02 Effect of carbon additives on the flowability of cathode powder mixtures in dry electrode production  
*Andreas Gyulai, KIT*
- P1-03 Investigation of the influence of calendaring on the mechanical properties of electrodes for sodium-ion batteries  
*Ann-Kathrin Wurba, KIT*
- P1-04 Implications and opportunities for processing high solid content positive electrode pastes – a conductive additive and processing solvent approach  
*Candeniz Gercek, MEET*
- P1-05 LaserScale: Scaled laser structuring and drying of lithium-ion batteries to increase cell performance  
*Daniel Neb, RWTH Aachen University | PEM*
- P1-06 Solvent-free manufacturing of electrodes for Li-Ion batteries towards developing a continuous process  
*David Dirnbauer, Austrian Institute of Technology*
- P1-07 Incoming Goods Inspection for Electrode Production in a Battery Production Plant- A Closer Look into Key Parameters  
*Fabian Horsthemke, Westfälische Wilhelms-Universität | Meet Battery Research Center*
- P1-08 Investigation of the Drying-behaviour of granule-based battery coat-ings with low solvent content  
*Kevin Ly, KIT | TFT*
- P1-09 Preheating of Electrodes prior to the Calendaring Process – a Numerical Approach  
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- P1-10 Novel Structuring Methods to Increase the Porosity of Lithium-Ion Electrodes  
*Michael Bredekamp, TU Braunschweig | iPAT*
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- P1-12 Process Failure Mode and Effect Analysis in agile Battery Cell Production  
*Sebastian Schabel, KIT*
- P1-13 Calendaring of silicon-containing electrodes and their influence on the mechanical and electrochemical properties  
*Sören Scheffler, TU Braunschweig | iPAT*
- P1-14 Inductive drying processes for Li-ion electrode production towards higher throughput  
*Tobias Krüger, TU Braunschweig | ifs*
- P1-15 Solvent-free extrusion of cathode electrodes for high-energy Lithium-Ion Batteries (LiB)  
*Tom Sperling, Center for Solar Energy and Hydrogen Research Baden-Württemberg (ZSW)*
- P1-16 In-situ ultrasound acoustic measurement of the lithium-ion battery electrode drying process  
*Yeshui Zhang, UCL*

## Cell Assembly

- P2-01 Clamping mechanism for a high-speed stacking process  
*Christina von Boeselager, TU Braunschweig/IWF*
- P2-02 Recent advances for agile battery electrode production  
*Jonas Mohacsi, KIT | TFT*

- P2-03 FE-Model-based optimization of positioning accuracy in stack formation of lithium-ion battery cells  
*Kamal Husseini, KIT | wbk*

## Recycling & Sustainability

- P3-01 Environmental and socio-economic assessment of all-solid-state lithium-sulfur batteries for use in electric aircraft  
*Alexander Barke, TU Braunschweig | AIP*
- P3-02 Biopolymer electrolytes for zinc-based batteries  
*David Lammers, TU Braunschweig | ibvt*
- P3-03 Feasibility of water-based NMC-cathode preparation for lithium-ion-batteries with recycled cathode composite  
*Inga Landwehr, Fraunhofer IPA*
- P3-04 Towards sustainable anodes using pine wood biochar as an additive  
*Juan Carlos Espinosa, Instituto Tecnológico de la Energía*
- P3-05 Prospective LCA of a model Magnesium Battery  
*Sebastián Pinto Bautista, KIT | Helmholtz Institute Ulm*

## Module and pack design and battery safety

- P4-01 Influence of Punch Diameter and Punch Speed on the Reproducibility of Crush Tests  
*Alexander Hahn, TU Braunschweig | iPAT*
- P4-02 Inline Failure Detection Based on Process Emission for Laser Beam Welding of Copper  
*Bernhard Klier, F&K Delvotec Bontechnik GmbH*
- P4-03 Lithium plating detection method for Second-Life BMS Applications  
*Daniel Kehl, TU Braunschweig | elenia*
- P4-04 Scalability of Nail Penetration Tests to predict the Thermal Runaway behavior of LiB  
*Stefan Doose, TU Braunschweig | iPAT*

## Battery Production 4.0

- P5-01 Use Cases and Application Levels for a Guide to Digitalization of Battery Cell Production  
*Alexander Puchta, KIT*
- P5-02 Decreasing battery cell production ramp up times through scaling factors and spillover effects  
*Markus Eckstein, Fraunhofer IPT*

## Production of solid state batteries

- P6-01 Implementation of a slot die coating process for the production of thin solid electrolyte layers  
*Andrea Wiegandt, Fraunhofer IFAM*
- P6-02 Energy efficient production of ceramic electrolyte layers for solid-state batteries by FAST  
*Carsten Glanz, Fraunhofer IPA*
- P6-03 Influence of Molecular Weight and LiTFSI on the Thermal Processability of PEO for Solid-State Electrolytes  
*Katharina Platen, Fraunhofer IFAM*



# POSTER SESSION

## Housing, geometrical shapes and cell types

- P7-01 Influence analysis of large cell formats on the battery production costs  
*Natalia Soldan Cattani, Production Engineering of E-Mobility Components*
- P7-02 Reproducible Production of Various Test Cells for Academic Research  
*Paul-Martin Luc, TU Berlin*

## ProZell – competence cluster for battery cell production

- P8-01 Pilot-plant scale production of nickel-rich cathodes for lithium-ion batteries  
*Carina Heck, TU Braunschweig | iPAT*
- P8-02 The influence of infrared post-drying parameters on residual moisture and structural and electrochemical properties of electrodes  
*Fabienne Huttner, TU Braunschweig | iPAT*
- P8-03 Effects of Production Scrap Rate on a Process Chain Level: An Energetic and Economic Assessment  
*Gabriela Ventura Silva, TU Braunschweig | IWF*
- P8-04 Production-Oriented Numerical Optimization of NMC Cathodes with Maximized Energy Density  
*Hassan Karaki, TU Braunschweig | InES*
- P8-05 Towards Logical Description and Linkage of Battery Data- DigiBatMat Platform of ProZell and MaterialDigital  
*Milena Perovic, TU Braunschweig | iPAT*
- P8-06 Fast Charging Limitations during the Formation of Lithium-Ion Batteries  
*Robin Drees, TU Braunschweig | elenia*
- P8-07 The importance of passive materials in thick Li-ion battery electrodes  
*Tobias Knorr, German Aerospace Center*

## greenBatt – competence cluster for recycling and green batteries

- P9-01 DIGISORT: Digitally improved sorting of lithium-ion batteries  
*Alexandra Kaas, Technische Universität Bergakademie Freiberg*
- P9-02 Development of Design Guidelines for Battery Systems in the Context of a Circular Economy  
*Gregor Ohnemüller, University of Bayreuth*
- P9-03 Life Cycle Sustainability Assessment of battery raw material supply chains  
*Jan-Linus Popien, TU Braunschweig | AIP*
- P9-04 Polymer-SSB Recycling – How mechanical processes can be adapted for efficient recycling  
*Marco Ahuis, TU Braunschweig | iPAT*
- P9-05 Leaching of black mass from lithium-ion batteries by biodegradable leaching agents  
*Monika Keutmann, RWTH Aachen University | IME*
- P9-06 Data-Mining in the Recycling of Lithium-Ion Batteries  
*Sandro Süß, TU Braunschweig | IWF*

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# POSTER ABSTRACTS

## Electrode Production

### ■ Analysing the impact of particle size distribution on differently compacted graphite based electrodes via X-ray microcomputed tomography

Alexander Diener

TU Braunschweig | Institute for Particle Technology (iPAT)

Calendering as the common process step for electrode compaction affects the structure of the pore network as well as the particulate solid fraction and thus, has a crucial influence on the electrical and electrochemical cell behaviour. Consequently, process and machine parameters have to be changed well-founded in order to generate electrode structures optimized for a desired application. In order to meet this objective a deep understanding of how the calendering process affects the electrode structure beyond the simple reduction of porosity is needed. The focus of the presented work is to investigate the impact of the calendering on the electrode microstructure and to correlate it with electrode properties by developing process-structure-property functions regarding above mentioned criteria. Therefore, graphite based electrodes with different particle size distributions were produced and compacted to different coating densities. Changes in the electrode microstructure on active material scale were investigated using X-ray microcomputed tomography (XMT). By taking the limitations of the XMT method into account special model anodes out of spherical graphite particles were build up without carbon black. Local density distribution as well as the particle and pore network structure were analysed with XMT. The homogeneity of the electrode was examined by correlating the coating density with the coating surface density. To evaluate the quality of the XMT results in terms of the solid phase depiction, particle size distributions obtained by XMT and by laser diffraction analysis were compared. Furthermore, mercury intrusion porosimetry (MIP) was used as a reference method to analyse changes in the pore structure induced by the calendering. In summary it was found that non uniform density distribution after the coating and drying process still persist after calendering the electrodes. Consequently, a uniform electrode coating has to be ensured before calendering.

### ■ Effect of carbon additives on the flowability of cathode powder mixtures in dry electrode production

Andreas Gyulai

Karlsruhe Institute of Technology (KIT)

Dry production of battery cathodes has received a lot of attention in the past years, nevertheless the processing from powder to high quality electrodes remains challenging. The reason lies mainly in the lack of solvents that act as lubricants in traditional electrode casting processes. Therefore, the used dry materials not only have to show good electrochemical properties, but also have to be able to be processed homogeneously without liquid lubricants. This work studies the effects of different carbon additives and PVDF binders on the processibility in a dry electrode production process using a single calender. Structure and particle size and shape of carbon additives, as well as their surface chemistry are correlated to the flow properties of the powder mixture. By better understanding the influence of the additive components, tailored battery compositions can be developed to strongly improve the flow properties of the powder mixtures. This leads to significant improvements in the processibility of the powder mixtures, thus resulting in dry produced electrodes of higher quality.

### ■ Investigation of the influence of calendering on the mechanical properties of electrodes for sodium-ion batteries

Ann-Kathrin Wurba

Karlsruhe Institute of Technology (KIT)

The increasing demand for high-performance electrical energy storage is associated with the demand of the optimization of existing technologies and the development of alternative energy storage systems. An improvement of the cell performance can be achieved by improving the active material itself and by an optimal production process. Therefore the DFG-funded project

Post-Lithium Storage Cluster of Excellence (POLiS) has the overall aim to develop such promising active material alternatives and to accelerate the technology transfer from laboratory to industry scale production. Once the dry electrode is manufactured, the process step of calendering is the decisive one to set the final density of the coating. This in turn determines the energy density and finally the cell performance. During compaction the electrode is exposed to high mechanical stresses that lead to changing mechanical properties. This can even cause that the electrode is not able to be further processed. To prevent this, a deep understanding of the relation of the electrode material and the process of calendering is needed.

In this study this material-process-relation is investigated by a systematic analysis of the influence of calendering on the mechanical properties of the electrodes. Mechanical tests such as bending tests and peel tests are performed to examine the changing mechanical behaviour of both calendered and uncalendered electrodes. The achieved compaction is calculated and related to process parameters and the mechanical properties. These results will give an insight into the mechanical interactions of the electrode during calendering. Furthermore these relations will build a basis for the prediction of the further processability.

### ■ Implications and opportunities for processing high solid content positive electrode pastes – a conductive additive and processing solvent approach

Candeniz Gercek

MEET - Münster Electrochemical Energy Technology

Low cost and high energy density lithium ion batteries (LIBs) produced under the most sustainable conditions possible are amongst others strongly related with the positive electrode (cathode). For instance, the energy consumption required for cathode production can be reduced significantly by increasing the solid content (SC) during electrode paste preparation resulting in a potentially shorter drying process and less use of solvent.

In order to achieve higher energy densities on cell level, the application of thick positive electrodes is needed. However, to ensure a high lithium ion mobility and a maximized capacity utilization, it is crucial to tailor the electrode microstructure. During production of thick electrodes, negative effects like binder-migration can occur upon the drying process inducing an inhomogeneous distribution of binder and conductive agent as well as crack formation. By working with high SCs both effects can be widely suppressed. However, increasing viscosity with increasing SC limits the production of homogenous electrodes. The use of nano- and micro-scale spherical, linear and three-dimensional conductive agents facilitates achieving an appropriate paste viscosity and the formation of an electronic percolation network to ensure high electronic conductivity. Thus, tailoring of active and inactive components is crucial to enable processing of high SC electrode pastes with an appropriate viscosity as well as production of thick electrodes with good electronic and ionic properties.

In summary, a comprehensive study on the factors influencing the rheological and electrochemical properties by different processing additives is presented. The increase of the SC from 70% to 80% and the addition of a less toxic co-solvent is a first step towards the reduction of the production costs of LIBs. In future, it is essential to develop new methods to further increase the solid content.

### ■ Fully continuous mixing technology for both conventional electrode slurry and solvent-reduced electrode masses

Christian Hänsel

Bühler AG

Bühler has been pioneering the continuous mixing technology for LIB electrode slurries for already 10 years. The technology is based on twin screw mixing which combines all unit operations in a single device. The result is a very high productivity per mixing line, elimination of batch-to-batch variation and reduced operating costs.

In this presentation, we will elaborate on selected key aspects of the mixing of conventional electrode slurry. Additionally, we will demonstrate the potential of this technology for next generation manufacturing techniques (i.e. solvent-free processing).

## ■ LaserScale: Scaled laser structuring and drying of lithium-ion batteries to increase cell performance

*Daniel Neb*

*RWTH Aachen University | Production Engineering of E-Mobility Components (PEM)*

The LaserScale research project aims to implement an innovative system concept to reduce production costs while increasing product quality within the manufacturing process of lithium-ion batteries. The innovative system concept is intended to increase the energy and power density of lithium-ion batteries by introducing microstructures into the electrodes with the help of laser radiation. The two-dimensional structuring of the electrode material is to be determined using a multi-beam multiscanner module. By combining laser structuring with flexible laser drying, process efficiency is to be increased at the same time. Surface emitters ("vertical-cavity surface-emitting laser", VCSEL for short) are intended for drying the coated electrode foils. These enable dynamic adjustment and control of the temperature distribution for targeted drying of the electrodes. With the help of the integration of a data-driven process control system, essential process data will be recorded and adapted to the optimal system range. The research work also focuses on scaling the underlying laser processes towards industrial applicability.

Another innovation for more efficient production and thus cost reduction of battery cells is the process integration of a simultaneous coating on both sides in the vertical design direction. The combination of the various approaches within the LaserScale project should make a significant contribution to increasing the efficiency and sustainability of battery production in the sense of a green battery.

## ■ Solvent-free manufacturing of electrodes for Li-Ion batteries towards developing a continuous process

*David Dirnbauer*

*Austrian Institute of Technology*

The production of cathodes for Li-Ion Batteries is based on the toxic solvent n-methyl pyrrolidone (NMP), which is necessary for processing PVDF. There are economical and ecological aspects, such as high unit costs compared to other solvents flammable and toxic vapours during manufacturing and ghg emissions due to the energy intensive drying process. The use of NMP causes up to 14.5 % of overall cell manufacturing costs.

One way to eliminate NMP/PVDF is the substitution with Water/CMC/SBR. Slurry agglomerates, low wettability and corrosion of the current collector or active material, especially when focusing on high-nickel NMC, are challenges of water based processing. These can be solved via pre-processing steps and including additives. This and the nevertheless necessary drying step lead to a reduction of the ecological and economical advantages of water based processing.

Solvent free manufacturing of electrodes seem the mean of choice for reducing cost and environmental impact of battery manufacturing. The time and energy consuming drying step as well as remaining solvent after drying can be avoided.

Solvent free electrodes were manufactured using an process based on pilot scale equipment. The process is displayed and the effects of several manufacturing parameters on the characteristics of the cathodes are analysed. The electrochemical data of solvent free cathodes in coin and as well pouch cells is shown. First tests show comparable results to conventionally manufactured cathodes.

## ■ Incoming Goods Inspection for Electrode Production in a Battery Production Plant - A Closer Look into Key Parameters

*Fabian Horsthemke*

*Westfälische Wilhelms-Universität | Meet Battery Research Center*

The worldwide demand of lithium ion batteries (LIBs) is increasing steadily. Therefore, the number of different material suppliers and cell manufacturer plants is growing alongside the LIB development. Furthermore, the LIB chemistry for electrodes and inactive materials diversifies increasingly with the applications fields e.g., from the lithium cobalt oxide to a variety of different cathode materials utilized for automotive and aviation purposes. Merging this complexity of different LIB systems and the varie-

ty of suppliers and manufacturers, the analysis of incoming materials is an important factor to produce LIBs with uniform quality. Furthermore, the incoming goods inspection represents a fundamental investigation for a safety-related quality assurance as well as for process planning and control. Therefore, the parameters of interest have to be defined for the expected material classes i.e., for powders, liquids and coils.

From the huge variety of analytical methods applied on LIB materials reported in literature[1] only a limited number is applicable in the context of a LIB production plant. Analytical tools used in academia are often prototypes, expensive or specifically modified for a particular use. Therefore, the implementation of a specific method depends on its general applicability, the importance of obtainable parameters, accuracy, reliability, robustness and overall costs.

The presented work summarizes the setup of quality assurance laboratories to analyze incoming goods in a comprehensive manner for a lithium ion battery production plant. The Fraunhofer Forschungsfertigung Batteriezelle (FFB) brings together industrial scale LIB manufacturing and scientific research. This combination will form a demanding environment for analysis techniques as the variety of cell chemistries, processes and materials and additional end-of-line testing has to be accounted for.

[1]S. Nowak, M. Winter, Journal of the Electrochemical Society 2015, 162, A2500-A2508.

## ■ Innovation in Conductive Carbon Additives for Li-ion Batteries: Improved Cell Performance and More Efficient Electrode Manufacturing

*Giulio Ferraresi*

*Imerys Graphite & Carbon*

The increasing demand for advanced energy storage systems has been pushing Li-ion batteries towards optimization and decarbonization of its production chain. The automotive industry is betting at full speed on Li-ion for electrified vehicles and it is demanding significant upgrades of the actual cell manufacturing process to meet the sustainable EV cost-performance paradigm.

Taking a look at the Li-ion cell cost breakdown, it becomes clear that major improvements should be carried out at the electrode production level, which accounts for almost 30% of total manufacturing cost of the battery cell. [1] On one side, an efficient electrode production goes through the enhancement of electrode component mixing that can be obtained by reducing energy input and mixing time for carbon additive dispersions. On the other side, significant improvement in electrode coating and drying processes can be achieved by reducing/removing solvents during slurry preparation allowing to increase the production rates and cost savings. In parallel, Li-ion cell performance must be further improved by the introduction of advanced carbon conductive additives to overcome the actual performance hurdles, namely low electrical conductivity, energy and power density and cycle life.

During the presentation, we will outline how the design of high quality conductive carbon additives enables the improvement in Li-ion cell performance and electrode manufacturing. We will introduce new C-ENERGY Carbon Black products featuring stringent purity requirements and excellent compatibility with solvent-based and dry-based electrode manufacturing, while enhancing the electrode electrical conductivity. Their outstanding performance in Li-ion cells will be highlighted and the synergistic effect of our C-ENERGY Carbon Black products in combination with other additives (such as conductive graphites and carbon nanomaterials) will also be discussed.

[1] Turcheniuk et al. Materials Today, 42, 2021, 57-72

## ■ The influence of formulation on the coating quality of an innovative extrusion-based coating process for li-ion electrodes

*Jann Seeba*

*Fraunhofer Institute for Ceramic Technologies and Systems IKTS*

High-load electrodes are considered promising to improve the energy densities of Li-ion batteries (LIB) for mobile applications. Conventional casting processes for electrode coating are limited in terms of film stability and homogeneity, especially for thick,



high-load electrodes above 4 mAh/cm<sup>2</sup>. To tackle these challenges, an innovative extrusion-based coating processes is being developed.

The innovative process starts with a premixture of solid components and a binder solution. The components are homogenized in a double screw extruder. The resulting mixture is directly coated onto a current collector using a slot die at the extruder. After extrusion the foil is smoothed and dried. The process uses an increased solid content (>80 wt.%) to reduce the solvent consumption and decrease the energy consumption of the drying process. Furthermore, the process is currently scaled up to a R2R pilot plant, by optimizing the mixing process and the slot die geometry.

To achieve proper coatings in the R2R process the influence of different factors (e.g., particle size or shape, binder type, ...) must be understood. To correlate the coating quality to the formulation, in this work the rheology of different formulations of anodes or cathodes is investigated. The different formulations are characterized regarding their properties (viscosity, complex modulus) and coated with the extrusion based slot die. The electrodes are then analyzed regarding the quality, homogeneity and inner structure (e.g. SEM cross section, 2D-profile, ...) and are correlated to the mixing process and formulation. The first results show significant impact of the ratio of SBR and CMC binders for anodes, by optimizing the ratio of binder a reduction in viscosity by 50% can be reached, allowing for a better coating quality.

### ■ Flotation for the separation of anode and cathode materials of end-of-life-lithium-ion-batteries

*Johanna Köthe*

*TU Clausthal | Institute of Mineral and Waste Processing, Waste Disposal and Geomechanics (IFAD)*

A possible recycling approach for lithium-ion batteries consists of mechanical and thermal treatment followed by flotation and hydrometallurgical treatment. The recycling process, especially flotation, is negatively influenced by organic substances that originate from the liquid electrolyte and the binder. In lithium-ion batteries, the anode active material consists mainly of hydrophobic graphite and the cathode material of hydrophilic mixed metal oxides which can be separated by flotation. However, adhering organics render cathode active material hydrophobic and therefore the separation efficiency is reduced.

As part of the umbrella concept of the German Ministry of Education and Research for battery research, researchers investigate the degradation of these organic components along the recycling process in the research project "Monitoring of low-volatile electrolytes in the mechanical recycling process chain". The aim is to completely remove the organic matter through thermal treatment before entering the flotation process and hydrometallurgical treatment.

In this presentation, the flotation of thermally pre-treated black mass is optimized which is free from organics. The target is a graphite yield of 90% at a purity of 90%. To gain knowledge about their flotation behavior, the dosage of reagents such as collectors, frothers, and depressants and the conditioning time will be varied at a fixed solid-liquid ratio. The number of steps required for the required separation is then determined experimentally. Afterward, a multi-stage closed-loop-process is to be developed.

### ■ Investigations of binder migration for NMC cathode slurries with compact and nanoporous structured particles

*Julian Klemens*

*Karlsruhe Institute of Technology (KIT) | Thin Film Technology (TFT)*

Electrochemical storage is a key technology of the current century. In the future, electricity storage systems will become more important components of energy systems, especially for the upcoming electric mobility and for everyday use. Lithium-ion batteries (LIB) are currently the most important electrochemical energy storage devices. The largely mature technology is characterized by high gravimetric and volumetric energy densities.

An improvement in the fast-charging capability and the durability of lithium-ion batteries is achieved through the use of nanoporous structured NCM. This improvement is attributed to electrolyte wetting right into the core of the active material and thus shorter diffusion paths for charge transfer.

The enlarged interface results in lower charge transfer resistance during phase change, which is why higher capacities can be achieved even at lower current strengths. Due to the particle morphology, new processing challenges arise compared to the compact solid NCM.

Here, the properties of the slurries and the electrodes made of the two active materials and, in particular, the influence of the drying process on the binder distribution are comparatively investigated.

### ■ Investigation of the Drying-behaviour of granule-based battery coat-ings with low solvent content

*Kevin Ly*

*Karlsruhe Institute of Technology (KIT) | Thin Film Technology (TFT)*

Lithium-ion batteries have become an indispensable part of modern life. Due to their high energy and power density, lithium-ion batteries are expected to be used primarily in the field of electromobility in the future. The desire for higher performance, cost efficiency and safety poses various challenges for the automotive industry and battery research.

The drying process of battery electrodes has an enormous influence on the electrode quality. It determines electrode characteristics and most importantly cell performance. The major problem during drying is the migration of binder to the electrode surface, resulting in an inhomogeneous binder-distribution throughout the film.

As for state of the art electrodes, drying is one of the most cost-intensive process steps, a new approach for reducing the solvent content in electrode-processing and therefore increasing the cost-efficiency for the battery manufacturing process will be investigated in this work. By reduction of solvent content and the usage of granulates in battery-paste manufacturing, the storage stability of the produced electrode pastes is several weeks. This leads to a decoupling of paste and electrode production and a significant increase in production flexibility.

In terms of the drying step it is essential to investigate the influence of the highly-concentrated particulate granular system on the drying process. Especially the influence on pore structure, film consolidation and binder migration is crucial for understanding the drying process. For this purpose, a series of fundamental studies will be conducted. This work presents the experimental methods for the investigation of the drying behaviour under defined process conditions. These are mainly gravimetric drying tests and investigations by means of cryo-SEM for the elucidation of the pore emptying mechanism, as well as investigations with a magnetic suspension balance for the disclosure of the sorption behaviour.

### ■ STATE-OF-THE-ART SURFACE INSPECTION SOLUTIONS - Intelligent interaction of defect detection incl. geometric measurements with web guiding systems in the electrode manufacturing process

*Klaus Hamacher*

*BST GmbH*

To grant a high product quality in the production of the electrode material it is a must to have:

- a precise web positions
- a defect free material
- a coating geometry in a high quality and within the tolerance

Beside the challenge of an exact guidance and positioning of the material, also state of the art inspection systems are requested. These provide today various functions like

- reliable surface defect detection
- machine learning of defect classification
- lane management
- simultaneously geometric measurements of coating positions
- qualification of edge profiles

- data workflow
- lowest footprint

Based on this solution the BST Group goes one step beyond: smart linked network between of the inspection and high precision web guiding system allowed an intelligent communication of process data. This prevents quality deviations before they occur.

The BST Group is a global leading supplier of in-line quality assurance systems and services for the web processing production industries. We transform our decades of experience, for example from the printing & converting industry, into the cell- and assembly production processes of Lithium-Ion Batteries.

Our Mission is: less waste, increased quality, more time for production!

### ■ Tortuosity Investigations on Cathodes with porous nanostructured vs. bulk NMC Particles

Luca Schneider

Karlsruhe Institute of Technology (KIT)

Lithium-ion batteries (LIB) are the most used batteries nowadays and the most important energy storage devices regarding to electric vehicles. LIBs have a huge energy and power density compared to other battery chemistries, which is the reason for their widespread use. However, conventional particle and electrode structures do not yet fully exploit the potential of these materials.

It is already known that porous nanostructured particles show some benefits compared to bulk particles. The increased specific surface and shorter diffusion paths inside the active material enhance the rate capability and the small primary particle size prevents particle cracking and leads to an improved cycle stability. [1] However, the impact of these particle system on the electrode characteristics is not investigated up to now in every detail as ionic and electronic resistivity are determined by the structural parameters on multiple levels. Therefore, in addition to understanding and optimizing the chemistry of the active materials, it is also necessary to improve particle morphology and electrode structure. Advanced electrodes like multi layered and blended electrodes have mainly been studied in terms of the electrochemical behaviour of various active materials, but the influence of particle morphologies on the electrode structure has not been considered. Therefore, the talk wants to close this gap and gives a debate contribution for such enhanced electrode structures.

The focus of this work is the measuring of tortuosity by impedance spectroscopy on electrodes with porous nanostructured and bulk particles. The experimental set up is a symmetric cell with blocking conditions to fit the data with a transmission-line model approach. To get a deeper insight into porosity-tortuosity relation, cathode coating with different layer thicknesses and porosities will be investigated and discussed.

[1] Müller, M. ACS Appl. Energy Mater (2021) doi:10.1021/acsaem.0c03187.

### ■ Preheating of Electrodes prior to the Calendaring Process – a Numerical Approach

Mark Lippke

TU Braunschweig | Institute for Particle Technology (iPAT)

Batteries are a key technology for the energy transformation. Lithium-ion-batteries are state-of-the-art and still, their performance is constantly improving. This is, among other things, because of the perpetual optimization of the production steps.

In order to increase the energy density and electric conductivity, electrodes are usually calendared. A higher degree of compaction can be reached by heating the calendaring rolls to utilize the thermoplasticity of typical binders. Additionally, preheating devices can be employed to magnify this phenomenon. With the aid of the presented method the benefit of a preheating device can be investigated.

The poster shows a numerical approach based on the discrete element method (DEM) to simulate the heating up behavior of electrodes before and during calendaring taking a typical NMC-622-Cathode as model material. To improve the results of exist-

ing, simplified DEM simulations, which neglect the heat transfer through the carbon-black-binder matrix (CBM), an extension to the DEM-code has been implemented to consider the heat transfer through the CBM and their respective heat capacity. In consideration of process parameters like roll temperature and line speed as well as electrode parameters like formulation, thickness porosity and binder distribution, this methodology can be applied to provide an individual estimation to evaluate the need for a preheating device. A good consensus with literature data has been reached. Also, it is demonstrated that existing approaches strongly underestimate the time needed to heat up the electrode due to neglecting the inactive material. Contrary to former approaches thermally isolated particles could be avoided, which results in a more realistic numerical modeled electrode.

### ■ Novel Structuring Methods to Increase the Porosity of Lithium-Ion Electrodes

Michael Bredekamp

TU Braunschweig | Institute for Particle Technology (iPAT)

The development of battery systems for mobile and stationary applications is an essential step towards a sustainable energy industry. Therefore, the lithium-ion battery cell is the subject of current research in industry and science. The rate at which lithium ions are intercalated is largely determined by the porosity of the electrode and leads to a decrease in the specific power density, especially with increasing electrode loading (thickness). The introduction of vertically-oriented structure pores, which provide fast transport pathways for Lithium-ions can maximize the rate-performance of electrodes while holding a higher energy density. For this reason, the targeted structuring of electrodes is a promising approach to improve the high-current capability of lithium-ion batteries by reducing cell internal resistances. The current state of research is laser structuring, in which an improvement in the rate capability has been proven. However, the process leads to material loss and the theoretical capacity decreases [1, 2].

In the work described here, lithium-ion battery anodes were structured in order to create small pores locally in the coating by using novel methods/processes without loss of active material. First, the influence of pore forming agents on the structuring of conventional graphite anodes was investigated. The possibility of structuring through targeted pressure surges and injection of liquid additives was also examined. Finally, mechanical processes with structured roller calenders were tested. Experimental data from Graphite/NCM622 coin cells are used for the evaluation of the individual methods/processes and were compared with laser structured electrodes. In addition, the structured electrodes could be characterized more precisely with EIS and physical tests.

References

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### ■ Graphite Recycling from End-of-Life Lithium-ion Batteries: Processes and Applications

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In recent years, recycling and regeneration of end-of-life Li-ion batteries (EoL-LIBs) is motivating world widely and have become a critical and an urgent task for a sustainable and an environmentally friendly future. This is mainly due to the exponentially growing number of LIBs from hybrid electric vehicles (HEVs), electric vehicles (EVs), and variety of portable electronic devices every year, and might result in supply risks for excellent resources, but also increasing potential environmental pollution caused by inappropriate solid waste disposal. However, several industrial plants have developed recycling on cathode materials, transition metal oxides, but no more attentions have been dedicated to the recycling and reusing of graphite from EoL-LIBs. Herein, the current status of EoL-LIBs regenerating is summarized in light of the recycling and purifying of graphite as a state-of-the-art anode material for most of the commercial LIBs. The reported techniques of recycled graphite from the

EO-LIBs (R-Gr) in the different classifications is summarized. Three major processes for R-Gr is categorized, including (i) washing process (only washing the separated graphite with different solvents, drying and then reusing), (ii) thermal treatment process (temperatures from 300 to 3000 °C), and (iii) chemical (acid leaching) + thermal treatment processes. The environmental friendly process, cost, and importantly the next application to use, are the key criteria for selecting the recycling process. The R-Gr depending on the method of separation and purification can be reutilized as an active material in LIBs, or making graphene, and can be also used for many other applications.

### ■ Investigation of the formulation type of SBR on the processability, structure and performance of silicon-containing anodes

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A global challenge to develop rechargeable batteries like lithium-ion batteries (LIB) with advanced materials arises from the development of new electric devices and the increase in registered electric vehicles. The specific capacity of the standard anode active material graphite is approximately ten times lower than that of novel materials like silicon (370 mAh/g vs. 3579 mAh/g) [1, 2]. However, silicon anodes create new challenges in manufacturing and cycling processes, for instance a high-volume change (approx. 280 %) during cycling [3] which can lead to a continuous lithium consumption due to persistent solid electrolyte interphase (SEI) growth or delamination. New binder materials can prevent delamination, but the different surface and morphology of silicon compared to graphite pose challenges to the manufacturing process regarding processability and homogeneity.

This study focuses on the investigation of styrene-butadiene rubber (SBR) with different formulation types (emulsion, fine powder, gross powder) and their influence on homogeneity in silicon containing electrodes. Regarding comparability, parameters like recipe and manufacturing processes are kept constant. The electrodes consist of 93 wt.% active material, 1.4 wt.% carbon black, 2.8 wt.% carboxymethylcellulose (CMC) and 2.8 wt.% of the varying SBR-type as well as one electrode in which SBR is substituted by CMC. During manufacturing, the resulting structural, mechanical and electrochemical properties of the suspensions and electrodes (e.g. rheology, elastic deformation behaviour and porosity) are determined and compared. For example, the SBR particle sizes lead to different initial porosities. The particles accumulate in the electrode according to an ideal sphere packing and fill the vacant spaces. A decrease in particle size results in a decrease of the initial porosity and therefore in an increase of electrode and energy density.

### ■ Process Failure Mode and Effect Analysis in agile Battery Cell Production

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Within the framework of quality management, the Failure Mode and Effects Analysis (FMEA) is used preventively to avoid errors and increase the reliability of the production system. FMEA is used in particular in the design or development phase of new products or processes. In the AgiloBat1&2 research project, a new type of modular production system is being developed and set up that can react agilely to different quantity scenarios and the materials and formats used. In addition, the QualiZell research project is systematically investigating the influence of quality deficiencies during production on battery performance.

On the poster, a structured process FMEA of the individual production processes is carried out based on the process steps of an agile battery cell production. From the FMEA, the probability of occurrence of a defect cause can be reduced and the probability of detection of a potential defect cause can be increased, for example, by providing additional tests. The results of the FMEA serve as input for the production system to be developed in AgiloBat as well as for the in-line compatible sensor technology to be developed in QualiZell.

### ■ Optimization of edge quality in the coating of li-ion battery electrodes

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In today's production of battery cells, the slot-die coating process in large roll-to-roll systems is state of the art. In addition to the known limiting coating defects, such as air entrainment and low-flow streaks (Schmitt et al. 2013), there is a formation of elevated bulges on the edges of the coating, which can cause damage in subsequent process steps. In the industrial continuous cell production, edge elevations are removed except at the position of the current collector, resulting in high material waste. (Schmitt et al. 2014). During calendaring and rolling up electrodes, edge elevations at the current collector can lead to an inhomogeneous force distribution over the width of the coating, which causes waves and cracks at the edges of the electrode. This problem is intensified especially in the production of thick electrodes. The dimensions of the edge elevations can be minimized by a combination of suitable internal fittings in the slot die and an adjustment of relevant process parameters such as the gap between slot die and current collector and the wet film thickness of the coated electrode. To be able to minimize the edge elevations, it is important to develop an understanding of the process.

In order to reduce edge elevations, a procedure has been developed, in which the internal geometry of the slot die is adapted in a suitable manner. In this work, the influence of material properties, process parameters and the internal geometry of the slot die on the edge formation was investigated experimentally. With the knowledge gained, the development of edge formation shall be predicted.

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### ■ Calendaring of silicon-containing electrodes and their influence on the mechanical and electrochemical properties

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The growing electrical mobility market requires improvement in the energy density of battery technology as a key factor for reaching a higher range of mobility. Thus, enhanced gravimetric and volumetric capacity of lithium ion batteries (LIB) is required for manufacture of electric vehicles. There are different ways to optimize the energy density of the battery, for example, the usage of new active materials or calendaring the electrodes. In this study, these two possible methods are combined.

As an anode active material, silicon has a specific capacity that is ten times higher compared to commonly used graphite (3579 compared to 370 mAh/g). Due to this, silicon is a promising material to increase the capacity of the anodes used in LIBs. However, using silicon as an active material has a significant drawback: during lithiation and de-lithiation a large volume change of silicon takes place (approx. 280%). This disadvantageous property of silicon limits the long-term cycling stability of the electrodes, because of the large strain stress and thereby mechanical forces during the charging cycles, as demonstrated by Du et al. for silicon-alloy anodes. The aim of this study is to identify the influence of varying silicon contents on the electrode behavior during calendaring and their mechanical properties and electrochemical performance. The mechanical properties of the anodes are quantified by adhesion strength as well as elastic and plastic deformation behavior.

The anodes produced are calendared to create a variation in the coating density and mechanical properties (e.g. ability of plastic deformation) of the electrodes. After calendaring, a decrease of the adhesion strength of the electrode coating is detected, which can be explained by damage to the binder-network. The porous network is also affected by the compaction



process, resulting in decreasing ionic conductivity as well as worse electrochemical performance with increasing applied forces during calendaring.

### ■ Inductive drying processes for Li-ion electrode production towards higher throughput

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Aiming an efficient lithium-ion battery production all process steps have to be enhanced in terms of quality and performance. In this regard, the drying process of electrodes keeps great potential for improvement. The main challenge of state-of-the-art convective drying methods is the negative impact of high drying rates on electrode properties, for example; a destruction in cycle stability; due to binder and carbon black segregation. Therefore, conventional drying processes are limited to a certain drying rate, which limits the overall battery production speed.

This work focuses on the development and prototypical implementation of a new method for drying electrodes through inductive heat input. By the means of induction, the conductive material the coating and the coated current collector can be heated specifically using electromagnetic waves. This way, the coating is dried from the bottom up. Furthermore, induction allows the generation of complex heating zones with individual heat distributions, for example to prevent the formation of tensions inside the active material coating. Therefore, this method enables the effective drying of the coating through direct and targeted energy input into the metallic substrate. This in turn should lead to a significant reduction of heating time and accordingly allows a significant energy saving compared to conventional convective methods respectively.

Within this study, experiments were carried out on state-of-the-art SMG-A5 anodes. The heat distribution inside the coating was observed using an infrared thermal imaging camera. To determine optimal process parameters for a homogenous heating of the coating, the influence of various factors such as PWM (pulse width modulation), distance between inductor and electrode and inductor geometry on the generated heating zone was investigated.

### ■ Solvent-free extrusion of cathode electrodes for high-energy Lithium-Ion Batteries (LiB)

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The growing demand for automotive lithium-ion batteries (LiBs) and the strong push towards lower costs is forcing the development of lower-cost production processes for LiB that also should be more environmentally friendly. [1]

LiB anode materials based on Carbon materials are already coated in an aqueous process whereas NCM/NCA cathode materials are still using toxic NMP as solvent. The solvent-free extrusion of electrodes is now developing from a pure academic process into a valid alternative for mass-production avoiding any kind of solvent. This directly results into lower costs and electrodes that are favorable in end-of-life battery recycling.

Solvent-free and therefore highly viscous electrode pastes enable the manufacturing of ultra-thick electrode layers, which provide increased energy density. By using such a dry-extrusion process, typical shortcomings of the wet-coating such as crack formation, variation in electrode thickness, or binder migration occurring during solvent evaporation [2;3] can be avoided.

We present the development of a process for the production of solvent-free cathode composites for extrusion. A formulation containing NCM622, binder, carbon black and a superplasticizer was optimized in a small kneader imitating the extrusion equipment. The influence of process parameters as temperature or rotation speed on paste viscosity was examined. As a first result, an optimized, solvent-free paste was developed and processed in a twin-screw pilot-extruder. Pastes with up to 81 wt.-% NMC622 could be processed. The results also show good prospects to reach even higher active material contents in the pastes while still keeping their good processability.

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### ■ In-situ ultrasound acoustic measurement of the lithium-ion battery electrode drying process

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The electrode drying process is a crucial step in the manufacture of lithium-ion batteries and can significantly affect the performance of an electrode once stacked in a cell. However, there is a need to develop more informative in-situ metrologies to better understand the dynamics of the drying process. Here, ultrasound acoustic based technique has been developed as an in-situ tool to study the electrode drying process using NMC622-based cathodes and graphite-based anodes. The drying dynamic evolution for cathodes dried at 40 and 60 °C, and anodes dried at 60 °C were investigated, with the attenuation of the reflective acoustic signals used to indicate the evolution physical properties of the electrode coating film. The drying-induced acoustic signal shifts were discussed critically and correlated to the reported three-stage drying mechanism, which offering a new mode to investigate the dynamic drying process. Ultrasound acoustic based measurement has been successfully shown to be a novel in-situ metrology to acquire dynamic drying profiles. The findings would potentially fulfil the research gaps between acquiring dynamic data continuously for drying mechanism study and existing research metrology. It shows the great potential to be further developed and understand the drying process to achieve a more controllable electrode manufacturing process.

## Cell Assembly

### ■ Clamping mechanism for a high-speed stacking process

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With the emerging use of thick electrode coatings for high-energy applications, the importance of single-sheet stacking as a production technology for the electrode-separator-assembly (ESA) is increasing. Usually, this process is carried out as a robot-based handling process, in which the electrodes are picked one by one and placed into a magazine where a pair of clamps fixes the electrodes. Compared to continuous or semi-continuous processes like winding or Z-folding, the robot-based single-sheet stacking lacks productivity. The alternative to the pick-and-place process is a novel stacking technology, where a continuously rotating paddle wheel replaces the robot as a handling unit. The paddle wheel can achieve high-speed handling operations by continuous material flow. However, this high-throughput process requires specialised periphery. This work shows the design and experimental evaluation of a productivity-adapted clamping mechanism. The clamping mechanism prevents the loss of positioning accuracy of the stack in the magazine in the required process time. Above that, it can achieve a centering effect for readjusting the ESA in the magazine. Finally, the experimental evaluation shows that the clamping mechanism performs well to prohibit the damage of the active material of the electrodes.

### ■ Recent advances for agile battery electrode production

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The increasing demand for energy storage may position battery cells as one of the main players in the process of the energy transition concerning aspects like e-mobility. Current production systems for battery cells produce standardized high quality

cells, but are not specifically adaptable to customer requirements or altering production condition, such as changing material or format. Therefore, an aim is the flexibility of the manufacturing process in terms of format, material and quantity which is the scope of the project called “AgiloBat”. The subject of the project the development of a fully automated battery production in a single sheet batch process based on robot cells. Placing the production modules inside of microenvironments guarantees the production under regulated air conditions.

A crucial step in the production process of battery cells is the coating and drying of the battery electrodes. In order to achieve the desired properties such as thermal conductivity or ionic conductivity in the complex porous structure of the active material layer, a controlled and homogeneous coating and drying process of the electrodes is essential. Moreover, aspects of energy efficiency play an increasingly important role in battery production. Therefore, a main aspect in the AgiloBat project is the development and optimization of new coating and drying concepts for the production of battery electrodes of flexible format and material, which will be presented in the conference poster.

This work contributes to the research performed at CELEST (Center for Electrochemical Energy Storage Ulm Karlsruhe) and Material Research Center for Energy Systems (MZE). The authors would like to acknowledge financial support of the Ministry of Science, Research and Arts Baden-Württemberg (MWK-BW) and the Federal Ministry of Education and Research (BMBF) under project ID O3XP0369A (AgiloBat2).

### ■ FE-Model-based optimization of positioning accuracy in stack formation of lithium-ion battery cells

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Large-format lithium-ion batteries are becoming significantly important against the background of the steadily growing demand for electric vehicles. Pouch cells provide a comparatively high packing density and exhibit high flexibility with regard to their format. The efficient production of this cell type is increasingly moving into focus of research and industry. Pouch cells can be manufactured by alternating stacking of electrode and separator sheets, so called single sheet stacking. An important quality criterion of this process is the stacking accuracy of the final electrode stack. The mechanical and geometrical properties of the singulated electrode sheets influence the stacking accuracy.

In this work, a simulation and optimization methodology is presented to compensate occurring positioning inaccuracies during the stacking process. FE simulations are used to determine positioning inaccuracies of the electrode sheets during the stacking process and to derive data sets. Based on the generated data, the expected stacking inaccuracies can be calculated that occur due to the mechanical and geometric properties of the electrode sheets. Furthermore, a methodology is derived to determine an alternative and optimized stacking position based on the generated data sets. The result is a model that determines an optimized deposit position before each stacking process. This determined position serves as a reference point for the gripper system of the stacking machine to compensate the predicted inaccuracies.

## Recycling & Sustainability

### ■ Environmental and socio-economic assessment of all-solid-state lithium-sulfur batteries for use in electric aircraft

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The aviation sector is responsible for a large amount of climate-damaging greenhouse gas (GHG) emissions, which lead to global warming. While recent reductions of GHG were achieved by improving aircraft engine efficiency, further decarbonization requires technological transitions. A promising strategy is the development and deployment of battery-electric aircraft.

Due to the technical restrictions concerning aircraft weight, high specific energy of the battery is crucial. A key technology in this context is the all-solid-state battery (ASSB). Especially the lithium-sulfur-ASSB (LiS-ASSB) enables high specific energy by using sulfur as active material. By carefully selecting materials and components, LiS-ASSBs suited for air travel are achievable.

However, it is still questionable which solid electrolyte (SE) is best suited for using LiS-ASSB in aviation and how suitable battery systems are designed. In addition, while the usage of these batteries reduces GHG emissions during flight operation, the materials required for battery cells and energy-intensive production processes are associated with new environmental and socio-economic challenges.

Thus, this poster aims to provide first insights concerning the use of LiS-ASSB for electric aircraft. Based on electrochemical models, SEs for LiS-ASSB are selected, and the battery systems are designed. Subsequently, a sustainability assessment is conducted to analyze the environmental and socio-economic impacts of battery production. For this purpose, the LiS-ASSB production is modeled, including raw material extraction, component manufacturing, cell production, and pack assembly.

The results indicate that sulfides are the most suitable inorganic SEs for electric aircraft. However, comparing different configurations of LiS-ASSB show significant differences, especially regarding the environmental impacts of resource consumption and climate change, the economic indicator of production costs, and the social risk of child labor.

### ■ Biopolymer electrolytes for zinc-based batteries

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The increasing demand for portable electronics and smart grid energy storing technologies forces the needs for sustainable, low cost and safe battery chemistries. Next to dominating lithium-ion batteries (LIBs), sodium- or zinc-based batteries are investigated to reach similar electrochemical but improved safety properties. Zn batteries as one of these “beyond-lithium-ion” technologies stand out due to safe aqueous electrolytes, lower costs and specific power and energy close to LIBs [1]. To reduce Zn anode derived dendrite growth and shape change during cycling, gel polymer electrolytes (GPEs) are investigated to stabilize the Zn anode in a nickel-zinc battery (NiZnB) and pave the way for flexible and leak-proof batteries. Thereby, the focus is set on biopolymer GPEs due to their structural diversity and renewability.

The investigation of these biopolymers requires reproducible cell cycling with reference NiZnBs based on alkaline liquid electrolyte and glass fiber separators. Therefore, Zn foil and electrochemically deposited Ni(OH)<sub>2</sub> on a Ni foil served as anode and cathode, respectively. Successful cycling of NiZnBs was shown electrochemically for at least 100 cycles at 1 C and via X-ray diffraction measurements proving the oxidation of Ni(OH)<sub>2</sub> to NiOOH by specific diffraction patterns. Subsequently applied GPEs in the NiZnB are based on alginate, xanthan or bacterial cellulose and were chosen due to their polar functional groups and availability. These polymers are combined and varied in their concentration in the electrolyte to be compared in terms of mechanical stability and electrochemical behaviour. The obtained results will also be beneficial for upcoming Zn-air batteries facing similar challenges as the NiZnB regarding the anode ageing.

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### ■ Feasibility of water-based NMC-cathode preparation for lithium-ion-batteries with recycled cathode composite

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Water-based electrodes are almost essential for green battery production. Since battery cells are usually recycled only after several years, it is crucial for a closed-loop strategy that the recovered materials can also be used in this future process routes. In these routes, both anodes and cathodes could be aqueous processed.

In anode manufacturing, aqueous-based processes are well established. However, cathode production also offers potential for water-based electrode production. Not only due to the elimination of carcinogenic solvents such as NMP and related environmental aspects but also with regard to further production strategies. Nevertheless, many issues in water-based cathode paste production are still insufficiently researched: Can different aqueous binder systems be processed in the same way with various NCM types? How much virgin material can be replaced by direct recycled material in aqueous slurries and how does this influence the cell performance?

Therefore, in a first step we developed and tested a recipe for project-internal reference electrodes with  $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$  and  $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$  as well as different aqueous binder systems. In a further step, the formulation and processing were adapted for the production of aqueous electrodes containing a proportion of recycled cathode composite material. Comparative measurements on various electrodes showed the influence of the composition on cell performance.

The feasibility could be demonstrated with directly recycled cathode composite with a content of up to 10 wt.% in lab-scale.

### ■ Towards sustainable anodes using pine wood biochar as an additive

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Sustainable materials development for their integration in battery components in the frame of circular economy concept is a priority in the battery sector. Carbon materials coming from abundant carbon rich wastes could be a promising type of materials for their application in Lithium battery anodes. Previous results of carbon materials obtained from orange skin wastes have been shown to be promising to improve the cyclability of Li-ion cells. In this work, biochar obtained from pine wood have been tested as an additive in graphite based anodes.

Initially, different properties of pine wood were characterized in terms of volatile matter, ash and water content; and elemental and inorganic composition. A carbonization process was performed in order to obtain the biochar. Selected properties of it were measured, i.e., porosity by N<sub>2</sub> adsorption isotherms at -196 °C, density and ash content. And, finally, carbonaceous obtained material were tailored and slurry processed for their integration as additive in anodes (10 wt.%); and the manufactured electrode was electrochemically tested.

The obtained results have shown that composite containing 10 wt.% of the sustainable carbon coming from pine wood allows a feasible fabrication of anode without detriment of the capacity of reference pristine graphite anode, demonstrating the possibility to include waste and sustainable materials in the production of anode electrodes in total or partial substitution of other complex and more expensive additives such as carbon black or conductive graphite. However, the irreversible capacity is slightly increased in comparison to graphite anodes.

### ■ Prospective LCA of a model Magnesium Battery

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Energy-storage systems are considered as a key technology for energy and mobility transition. Because traditional batteries have many drawbacks, there are tremendous efforts to develop so-called postlithium systems. The magnesium-sulfur (MgS) battery emerges as one alternative. Previous studies of Mg-S batteries have addressed the environmental footprint of its production. However, the potential impacts of the use-phase are not considered yet, due to its premature stage of development. Herein, a first prospective look at the potential environmental performance of a theoretical Mg-S battery for different use-phase applications is given to fill this gap. By means of the life cycle assessment (LCA) methodology, an analysis of different scenarios and a comparison with other well-established technologies are conducted. The results suggest that the environmental footprint of the

Mg-S is comparable with that of the commercially available counterparts and potentially outperforms them in several impact categories. However, this can only be achieved if a series of technical challenges are first overcome.

### ■ Introducing Lignin as a Bio-based Binder Material for an Aqueous NMC Electrode Production

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A sustainable shift from internal combustion engine vehicles (ICEV's) to electric vehicles (EVs) is essential to reduce greenhouse gas emissions significantly. The  $\text{LiNi}_x\text{Mn}_{1-x-y}\text{Co}_y\text{O}_2$  (NMC) materials are widely used as cathodes in lithium-ion batteries (LIBs) for EV's due to their high capacities. By replacing the commercial N-methyl-2-pyrrolidone (NMP)/polyvinylidene fluoride (PVDF) solvent/binder based cathode-production with a water-based production, the electrode production cost and environmental impact can be reduced substantially. However, the water compatibility of Ni-containing materials has been problematic due to lithium(Li)-leaching, corrosion of the aluminium (Al) current collector, and lack of aqueous dissolvable binders. Ideally, a bio-derived industrial waste material would be preferred, such as water-soluble lignin. Lignin is inexpensive, abundant and bio-degradable, as it is a by-product from Kraft pulp mills and extracted from black liquor. For the first time, we demonstrated that a water/lignin-based  $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$  (NMC111) cathode could be formulated with comparable specific capacities to NMP/PVDF-based cathodes. Multiple different solvent/binder systems were tested, including NMP/PVDF, NMP/PVDF:lignin, water/CMC:SBR, water/lignin:CMC, and water/lignin. According to viscosity measurements, the amount of solvent could be reduced by 15 % for a water-based slurry (NMC111:CB:lignin, 80:11:9) to obtain the optimal viscosity for coating, compared to an NMP-based slurry (NMC111:CB:PVDF, 85:10:5). Scanning Electron Microscopy (SEM) was used to analyze the cathode surface's morphology and topography, and galvanostatic cycling was employed to investigate the cathode's electrochemical properties. When gradually replacing the NMP/PVDF with NMP/lignin, the initial specific discharge capacity and adhesion to Al decreased due to poor lignin dissolution and inhomogeneous particle distribution. Dispersing lignin in water enhanced the mixing, the cathode coating's surface roughness, and its electrochemical performance. Decreasing the drying temperature from 90 to 50 °C was necessary for all water-based cathodes to obtain a controlled evaporation rate and reduce cracking. By introducing lignin/water into a CMC/water-based cathode, the discharge capacity and Al-adhesion strength increased, especially at higher C-rates. Calendering pressures were also investigated to find the optimal porosity and avoid particle cracking. The calendaring magnified the cathode coating's initial mechanical strength. This was especially clear for the lignin/water-based cathode where the poor adhesion between the Al-foil and the coated layer, and the strong particle-particle cohesion resulted in complete detachment of the coated layer as a whole. To further improve adhesion between the coating and the current collector, a carbon(C)-coated Al-foil was tested. Such C-coated Al-foil enhanced the Al-adhesion for the water-based cathodes. Furthermore, as the high surface tension of water caused poor wetting to the Al-foil, the C-Al also improved the slurry wettability during the coating. Electrochemical testing in half coin-cells revealed that lignin/water-based processing provided adequate binding to the Al- or C-Al-foil to permit the aqueous processing of NMC. However, the long term cyclability and capacity retention of the waterexposed cathodes were deteriorating faster than the PVDF/NMP-based cathodes. Li-leaching occurs when NMC111 particles are exposed to water, and are known to decrease the electrochemical performance of active NMC111 particles and promote corrosion of the Al-foil. Li hydroxide (LiOH) and Li carbonate ( $\text{Li}_2\text{CO}_3$ ) form on the NMC particle surface, and the slurry's pH increases above the stability window of the Al-foil. Such Li-leaching effect was studied by measuring the pH during exposure of NMC111 to pure water- or NMP-solvents, and NMP/PVDF-based or water/lignin-based slurries. All NMP and water solutions stabilized at a pH >10 within the first minute. The rapid initial growth in pH for water-exposed NMC111 provided evidence that the reactions occurred on the NMC111 particle surfaces with a strong driving force. The Li-leaching effect decreased when utilizing phosphoric acid ( $\text{H}_3\text{PO}_4$  or PA) as an additive. The solutions with PA additive had a constant increase in pH, but remained within the stability window of Al-foil.

Keywords: Lithium-ion Battery, Battery Electrode, Electrode Drying, Convection, Solvent Chemistry



## Module and pack design and battery safety

### ■ Influence of Punch Diameter and Punch Speed on the Reproducibility of Crush Tests

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Nowadays lithium ion batteries (LIB) became one of the key technologies for energy storage in different kind of applications because of their high energy density and their advanced stage of development. Therefore, LIB's are not simply rated by their performance parameters but also by issues of safety. With respect to the interaction of electrical and chemical hazards as well as emergence of fire and explosions, the thermal runaway represents the main risk potential related to the extended use of LIB's. During thermal runaway, exothermal chemical reactions trigger further exothermal reactions, leading to the release of flammable and toxic gases plus particles. For safety studies a thermal runaway can be provoked by different events which can be analyzed via temperature and voltage monitoring, as well as measurements of gaseous products and post mortem studies.

The safety assessment is examined using various standards before the battery is approved on the respective market. The requirements of the standards for the individual tests can be very different. Therefore, it is necessary to identify the crucial parameters to unify a particular test. In order to understand the crucial parameters of a crush test, this study presents the results of different punch diameters and punch speeds in a custom-made battery cell investigation chamber. This chamber allows the determination of parameters which influence the response of battery cells to internal short circuits. The response is analyzed via measurement of cell voltage, temperatures, as well as camera recording, and FTIR spectroscopy to identify and quantify infrared active gas species. The similarities and differences of the various test procedures on the electrical, thermal and chemical response of the cells are explained and the changes in the mechanical behavior discussed. The influence on the reproducibility of the results is emphasized in order to develop a general proposal for more comparable and reliable tests.

### ■ Inline Failure Detection Based on Process Emission for Laser Beam Welding of Copper

Bernhard Klier

F&K Delvotec Bontechnik GmbH

Nowadays, a shift to electric vehicles can be observed due to CO2 targets. Therefore, fast and reliable production processes for battery systems and power electronics are demanded. The creation of the electric connections between the components in power and battery modules poses a challenge. As a solution, the Laserbonder of F & K DELVOTEC Bondtechnik GmbH represents a fast, versatile, and reliable production machine. It enables state-of-the-art laser micro welding, also called laser bonding, of copper and aluminum to join electronic components with an integrated clamping method for the weld partners. External factors such as unwanted pollution of the substrate introduce process instabilities, which significantly affect the weld quality. To ensure the quality of the finished connection in a production environment, a new method to monitor the weld seam quality was investigated. It evaluates the back reflections, which were recorded by the integrated sensor of the laser beam source. To test the method's feasibility for the implementation on the Laserbonder, connections made of pure copper were examined and typical weld defects such as spatters were investigated. To classify the recorded back reflections into the categories "no defect" or "defect present", Machine Learning methods were applied. Signal transformation techniques based on the Wavelet Analysis are used to preprocess the signal for the classification and fault detection in a shallow neural network. The presented method is capable of detecting major critical defects for the examined weld partners and can be implemented as a cost-effective alternative to commercially available systems.

### ■ Lithium plating detection method for Second-Life BMS Applications

Daniel Kehl

TU Braunschweig | elenia Institute for High Voltage Technology and Power Systems

Due to the use of battery modules in electromobility, there will be many aged Li-ion batteries in the future that can no longer meet the requirements of the automotive sector at the end of the product life cycle. The requirements include intrinsic safety of the energy storage device and consistent energy storage capability so that a reduction in capacity does not result in a shorter range. This development enables cost-effective production of stationary energy storage systems using the battery modules. For low-maintenance and intrinsically safe operation, an initial test is necessary. The initial test must allow statements to be made on the aging condition of the battery modules and prove their suitability for further use. Depending on the area of application, the energy storage system must meet different requirements. If the application is for buffering energy from the grid, e.g. in charging stations, high performance is required. For use in a home storage system to store PV power, high energy density at comparatively lower power tends to be more important. For fast charging applications, lithium plating must be prevented, since in case of dendrite growth, the separator could be irreparably damaged, which is a danger especially for aged batteries. By analyzing the cell or module voltage after a charging process, lithium plating can be detected on the basis of the relaxation behavior. By implementing the detection method in a BMS, an aging-optimized fast charging operation can be carried out.

### ■ Scalability of Nail Penetration Tests to predict the Thermal Runaway behavior of LiB

Stefan Doose, Alexander Hahn, Peter Michalowski, Arno Kwade

TU Braunschweig | Institute for Particle Technology (iPAT)

Lithium ion batteries (LIB) became one of the key technologies for energy storage. The most important application is energy supply for mobile devices as well as for the green mobility because of their high energy density and their advanced stage of development. Therefore, LIB's are not simply rated by their performance parameters but also by issues of safety.

With respect to the interaction of electrical and chemical hazards as well as emergence of fire and explosions, the thermal runaway represents the main risk potential related to the extended use of LIB's. During thermal runaway, exothermal chemical reactions trigger further exothermal reactions, leading to the release of flammable and toxic gases plus particles. For safety studies a thermal runaway can be provoked by direct heating, overcharging or short circuit events. Such events can be analyzed via temperature and voltage monitoring, as well as measurements (qualitative and quantitative) of gaseous products and post mortem studies.

This study presents results of nail penetration tests in a custom-made battery cell investigation chamber. This chamber allows the determination of parameters which influence the response of battery cells to internal short circuit tests. The response is analyzed via measurement of cell voltage, temperatures as well as camera recording. Infrared gas species are identified and analyzed in-line by Fourier transform infrared spectroscopy (FTIR). The cells investigated in this study are manufactured, formed and electrochemically characterized. Cells of different capacity and chemistry (NMC111, NMC 622, NCA) are tested using a conductive nail material to determine the minimum required capacity to trigger a thermal runaway while using constant cell parameters. This study points out the scaling possibility of nail penetration experiments in lab scale for testing with low capacity battery cells. This results enable to development of safer and more efficient batteries for various use cases.

## Battery Production 4.0

### ■ Use Cases and Application Levels for a Guide to Digitalization of Battery Cell Production

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As a result of the rapidly advancing digitalization of production and electrification of mobility, the German industry is facing new challenges and fundamental changes. One of these is the establishment of a globally competitive lithium-ion battery production through the holistic introduction of digitalization and Industry 4.0 in all areas of production. To meet this challenge, a guideline for the digitalization of battery cell production has to be developed. Based on the requirements of equipment manufacturers and cell producers, instructions to reach the desired level of digitalization and Industry 4.0 are derived.

For the creation of the digitalization guide, use cases for digitalization in battery cell production and application levels are developed. Therefore, a wide range of research projects in the field of battery production as well as examples from other industries are evaluated. The focus is on the identification of use cases covering all aspects of battery cell production to ensure full applicability. A systematic abstraction of the identified use cases is done before they are assigned to application levels. These are developed in parallel and represent Industry 4.0 levels in production covering all stages from manual process execution to fully adaptive and predictive processes.

Thus the basis for the guide to the digitalization of battery cell production is established. On the one hand, a collection of practical examples has been created, and on the other hand, a comprehensive framework has emerged from their systematic abstraction, on which further work can be carried out about the development of the guide.

### ■ CO2 snow jet cleaning – increasing the yield in battery production

Jonas Gude  
acp systems AG

CO2 snow jet cleaning is an interdisciplinary technology that can be applied in different areas of battery production to increase battery lifetime reliability and yield. The current state of the art for cleaning processes in battery manufacturing (battery cells, packs, and modules) is the use of brushes or compressed air to remove residuals from the previous manufacturing steps. Numerous production processes leave traces of often electrically conducting dust, films and other particles that are physically or chemically adhered to the parts. With brushes and compressed air, only non- or weakly adhered particles can be removed from the surface. Additionally, brushes degrade over time and then need to be replaced. Compared to conventional cleaning technologies, CO2 snow jet cleaning can remove much larger portions of particles and residues. By itself, it is residual free and works in hollow complex shaped structures as well as on sensitive surfaces. Particles are supposed to be the major cause of yield losses in battery production as well as of shunts and degradation that appear later in battery systems operation. Molecular contaminations on contact surfaces originate, among other things, from volatile organic compounds in the production environment and must be removed immediately before the bonding process to achieve contact resistances within specification.

Exemplary applications of CO2 snow jet cleaning in battery production are as follows:

- Removal of particles or laser smoke from electrode surfaces after slitting
- Removal of micro-burrs after electrode slitting
- Removal of particles from the surface of cell and module housings
- Inline cleaning of contact pads of cells and modules

This presentation contains a description of applications of the technology and the corresponding results with the aim of contributing to ecological and economical battery production through reduced scrap and lower degradation over the service life.

### ■ Decreasing battery cell production ramp up times through scaling factors and spillover effects

Markus Eckstein  
Fraunhofer Institute for Production Technology IPT

In recent years, the field of battery cell production has gained momentum, mostly vivid through the growth in installed capacity of the many battery cell production lines world wide. However, the field of ramp up management for battery cell production is yet to be analysed in more detail with many incumbent as well as new companies experiencing major struggles. Whereas discrete manufacturing and assembly lines are thoroughly analysed in literature and practice, hybrid production facilities as battery cell productions still produce many delays and unplanned costs. To close this gap, this paper proposes several hypotheses how to diminish the high material costs during ramp up and how to run first production orders after shorter ramp up times. This is done by analyzing the extant literature on ramp up management and the main characteristics of battery cell production lines. Also, organizational structures and learning are considered as moderating effects on the ramp up performance of a battery cell manufacturer. This all is important since on-time SOP of first battery cells may be crucial for the many battery cell production lines currently under construction world wide to survive.

### ■ Inline Monitoring of Battery Electrode Lamination Processes Based on Acoustic Measurements

Nikolas Dilger  
Fraunhofer Institute for Surface Engineering and Thin Films IST

Great demands are being placed on the quality of battery cells and their electrochemical properties. Through the use of innovative measurement methods over the entire life cycle of batteries, relevant product and process data can be collected and leveraged to identify correlations between e.g. the product properties defined during production and the resulting performance characteristics in the use phase. The understanding of interactions between products and processes and the implementation of quality management measures in the production phase are essential factors that require inline capable process monitoring. Although the use of acoustic measurement methods for process monitoring has already proven its usefulness in various fields of application, it has not yet been applied to battery cell production. In our work, a process monitoring system based on acoustic measurements was applied to processes in the battery cell process chain. One example is the application in battery cell lamination for an automatic detection of components by interpretation of acoustic emissions. Signal analysis and machine learning techniques were used to distinguish between processed components. As a result, a detection accuracy of up to 83 % could be achieved, proofing the general feasibility of the approach as an inline capable monitoring system. Therefore, it can contribute to minimize sources of error in the complex production chain of batteries.

### ■ Exploring smart battery cell production based on a generic system architecture and an AI-enhanced process monitoring (project KIproBatt/BMBF)

Xukuan Xu, Michael Möckel, Simon Stier, Andreas Wolf, Lukas Gold, Christoph Berger, Dominik Fischer, Christina Leinauer  
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Due to its complexity and vast economic as well as ecological impact, the Li-ion battery cell production process is subject to ongoing digitization and optimization in order to increase cell performance while reducing resource consumption and production costs.

In this context, artificial intelligence (AI) holds immense potential in leveraging manufacturing data to improve the cell production process. Hence, we aim to enhance cell production with AI-based end-to-end process monitoring, which covers all steps of the process chain. For this purpose, we develop a generic, software-implemented system architecture as reusable structure that allows us to connect the process data acquisition with a carefully constructed ontology-based semantic data space. Based on this system architecture, we attach machine learning approaches from two perspectives: In the first perspective, we apply both data and physics driven models to specific process parameters to detect and evaluate correlations and process anomalies. In the second perspective, we develop an overarching end-to-end process monitoring. For this application, we integrate the

previously developed models into a dashboarding system in order to assess their relevance for cell performance and monitor the cells' state in production constantly and as real-time as possible.

The combination of these two perspectives allows us to detect defects early in the production process, rapidly increase the quality performance and derive flexible adjustments to the process parameters in case of malfunction or defects. Thus, we expect to reduce the total cost of cell production as well as improve its carbon footprint by reductions in resource and energy consumption. Finally, the generic, semantically structured design enables an easy transfer to other research and industrial processes.

## Production of solid state batteries

### ■ Implementation of a slot die coating process for the production of thin solid electrolyte layers

*Andrea Wiegandt*

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The constantly growing requirements on energy storage make an optimization regarding costs, safety, energy and power density inevitable. Therefore, many research activities on different technologies are in progress. One of these technologies is the replacement of the liquid electrolyte by a solid electrolyte, the so-called all-solid-state batteries (ASSB). In addition to improve the safety aspect, the ASSB technology generates an increase of the energy and power density compared to a conventional lithium-ion battery (LIB). The research approach to use a solid electrolyte raises the question of the material of ASSB and new processing options. Typical materials for the use as solid electrolytes are oxides, sulphides and polymers. Processability of polymers mark significant advantages of the use as electrolyte. The good solubility of PEO and Li-salt in solvents enables slot die coating processes for the production of thin layers of solid electrolyte.

The slot die process is a well-established coating method for conventional LIB, offering the potential of adapting the production process from conventional LIBs to ASSBs. Further advantages are low material loss and the production of thin layers with a controlled layer thickness. Hence, production of thin-film solid electrolytes requires the optimization of the process parameters.

In this work, we define a suitable processing window for the production of PEO based electrolyte. Therefore, we vary the concentration of the PEO/Li-salt mixture in a solvent and determine its suitability for the slot die process. The influence of material parameters, e.g. viscosity, as well as process parameters, such as coating speed and dispense rate, on the final coating result is determined. The dried layers are evaluated with regard to their homogeneity and layer thickness.

### ■ Solvent-free Processing of Solid State Electrodes Based On A Plastic Crystal Electrolyte System

*Arnaud du Baret*

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Succinonitrile (SN) has long been known as a useful additive in polymer electrolytes and lately as an electrolyte basis for solid state Li-ion batteries. However, the processing of such a material into corresponding composite electrodes has yet to be addressed in depth. We studied here several paths, comparing an infiltration method to the more scalable and therefore industry-relevant extrusion-based processing route. The latter has the advantage of solvent-free processing as well as reducing the numbers of processing steps from the base materials to the electrode foil and improving the achievable porosity fill rate.

The electrodes consists of state of the art NCM-622 (cathode) and LTO (anode) active material mixed with an SN-based polymer mixture: a backbone polymer such as PAN and ionic salts such as Li-TFSI provide ionic conductivity. Finally, due to fiber reinforcement of the electrolyte with glassfiber nonwoven a mechanical stability during processing, reduction of short circuits and a fixed electrolyte thickness could be achieved. According to our design considerations the cell can reach an energy density of 590 Wh/L based on these components.

A benchmark infiltration process is compared to the new preparation process: the mixing of all the components into a homogeneous electrode paste using a kneading machine with high shear stress. The active material content can be maximized by

processing the material at an optimal temperature where the electrolyte mix exhibits a low-viscosity behavior. The impact of particle size is

We characterize the corresponding product from a composition and structure standpoint with SEM and EDX analyses as well as porosity measurements. We further analyze the complete cells from an electrochemical standpoint with C-rate capability and cell longevity.

### ■ Energy efficient production of ceramic electrolyte layers for solid-state batteries by FAST

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Solid-state batteries with a solid ceramic electrolyte are seen as a promising alternative for conventional lithium-ion batteries. The main advantages are more safety and potentially higher energy density with less dangerous materials. For industrialization, there is a need for research, especially for the sintering process, since classical sintering methods are not suitable for the required materials and geometries. Furthermore, the process-reliable scalability from the point of energy efficiency is still unresolved. In our poster we show the applicability of the Field Assisted Sintering Technique (FAST), as an energy efficient alternative production technology for graded ceramic solid-state electrolyte hybrid structures.

During the FAST process, the applied current flows directly through the sintering mould, which leads to rapid heating of the sintering material. The sintering chamber itself does not need any additional heating. Consequently, sintering cycles are much shorter and therefore more energy efficient compared to conventional sintering technologies. The FAST process also enables better sintering results by reducing grain growth and preventing evaporation of substances. The process is investigated using lithium iron phosphate (LFP) as cathode material and lithium aluminium titanium phosphate (LATP) as ceramic solid electrolyte. Co-sintering of these materials leads to the formation of solid electrolyte-cathode composites. The amount of electrolyte is gradually reduced from the electrolyte surface to the current collector, improving both the interfacial transfer of lithium ions and thus the interfacial resistance. The graded structure should also result in higher thermomechanical stability of the cathode-electrolyte composite and consequently better cyclability.

### ■ The Effects of Ultraviolet Radiation on Inorganic Solid Electrolyte after Direct Laser Sintering

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Next-generation batteries should exhibit significant improvements in energy density and safety compared to current battery systems. One promising attempt is the establishment of all-solid-state batteries with inorganic solid electrolytes as a non-flammable component. Combined with a solid composite cathode and lithium metal as an anode, a cell concept with high potential concerning capacity and gravimetric and volumetric energy density is expected. In this concept, the Li<sup>+</sup> conductive component requires two features: Substitution of the liquid electrolyte and physical segregation between cathode and anode.

However, the mobility of Li<sup>+</sup> in inorganic solid electrolytes, for instance, lithium aluminium titanium phosphate (LATP), is dominated by diffusion processes in grains and grain boundaries. Without a densification process, the solid electrolyte displays low ionic conductivity and large grain boundary resistance. Consequently, effective sintering of the solid electrolyte is mandatory for achieving sufficient properties. For this purpose, different sintering techniques have been employed such as conventional sintering in a furnace or spark plasma sintering (SPS). Both methods have several disadvantages e. g. enduring sintering time, contaminated dies and expensive vacuum systems.

Accordingly, to reduce these disadvantages and slim down the development period novel sintering techniques are needed. In this work, ultraviolet laser radiation as a source of heat for sintering LATP is evaluated, since this method benefits from rapid processing time and neither dies nor a vacuum system are needed. Furthermore, the processing of complex geometries is feasible. The results focus on the effects of ultraviolet radiation on the solid electrolyte LATP (e. g. roughness) generated by various laser parameters. Moreover, the temperature during the process is recorded as a quality factor.



## ■ Influence of Molecular Weight and LiTFSI on the Thermal Processability of PEO for Solid-State Electrolytes

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All-solid-state battery is the next generation battery technology, which enables the use of electrodes made of metallic lithium that have a higher capacity and cell voltage than conventional electrodes. By replacing classical liquid electrolytes with ion-conducting solids, it is possible to improve the safety of such a battery. Polymer electrolytes are one promising material group, which are non-volatile and less flammable. One of the most investigated material is PEO due to its capability to dissolve alkali salts like LiTFSI. Furthermore, PEO based solid electrolytes give a perspective to easily scale up its processing from laboratory scale to industrial series production. Due to effects of solvents on the electrochemical and mechanical properties of SE, a solvent free method is preferred. This eliminates the need for costly drying steps, which is ecologically and economically advantageous for up-scaling processes. An option is a twin-screw extruder that can thermally process semi-crystalline thermoplastic like PEO. The use of an extruder offers the possibility of continuously mixing of PEO and LiTFSI to produce homogeneous polymer based SEs. Additionally, using two feeding units eliminates a premixing step to get a homogeneous mixture of the components. The thermal processing route of PEO and its effect on the polymer properties is currently investigated. To characterize the processing behaviour of PEO, we use a laboratory kneader that measures the torque depending on the applied kneading speed and processing temperature over a defined kneading time. In this work, we determine the influence of process parameters on PEO and PEO based electrolytes. In addition, we examine the influence of the molecular weight on the processability of PEO. Temperature and speed are optimised to avoid decomposition of PEO. For characterization, we do further analytical measurements such as the determination of thermal properties and microscopic characterization of mixing behaviour.

## ■ Scale-up of mechanochemical synthesis of sulfide-based solid electrolytes for all-solid-state batteries

Michael Grube

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Over the last years all-solid-state batteries (ASSB) are increasingly considered as promising follow-up technology of conventional lithium-ion batteries due to their potentially superior properties [1,2]. In order to produce cells with competitive characteristics, such as high energy density as well as high thermal and mechanical stability, the choice of materials at the beginning of the value-added chain is a main key. Currently, there are still challenges for upscaling the synthesis and processing routes of solid electrolytes (SE) wherefore ASSB have been just established at the laboratory scale. Bearing this aspect in mind, the focus of this work was to evaluate and establish new scalable process strategies to produce SE.

Thiophosphates have attracted broad interest as SE because of their high Li<sup>+</sup> conductivity and compliant mechanical properties [3]. Within this work, their synthesis was performed via high energy ball milling. Mechanochemical syntheses are typically known as scalable but time consuming processes [4]. In this work process time for the syntheses of Li<sub>3</sub>PS<sub>4</sub> and Li<sub>6</sub>PS<sub>5</sub>Cl could be highly reduced to a few hours by the systematic variation and optimization of operational parameters. Moreover, a deeper understanding of process-structure-product-relations was gained.

In Addition, the simulation of the process with the discrete-element-method (DEM) gave insights into the stressing conditions. The coupling of the experimental and simulation results reveals a direct correlation between the existing stressing conditions for different parameter sets and the obtained product properties. In a next step, the identified stressing conditions can be transferred to DEM models of scalable mill types such as stirred media mills to get indications for required operational parameters. Based on the obtained insights, the synthesis procedure was successfully demonstrated in a upscaled mill type, which will thus facilitate the industrial production of ASSB.

## Housing, geometrical shapes and cell types

### ■ Influence analysis of large cell formats on the battery production costs

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Production Engineering of E-Mobility Components

Lithium-ion cells are used as energy storage in a wide range of applications. The battery components such as electrodes, separators and electrolyte are surrounded by a housing which is subject to various requirements. For automotive applications there are several requirements a battery must fulfil. There are the product linked factors like energy density, safety and cooling capability and there is cost on the other hand. In Battery electric vehicles (BEV), the cost of the battery makes up the largest percentage. Therefore, it is important to control and reduce the cost as far as possible in order to make BEV more affordable.

Besides benefits in energy density, large format concepts are also expected to reduce production cost by simplified cell assembly and improved module integration concepts. But there are many other aspects that also impact the cost of the battery system. For example, the choice for one of the cell formats, pouch cell or prismatic cell, which is an unanswered question to the present. Further the material choice of the cell impacts the cost, because it makes the biggest part of total battery cost. Also, the material prices are uncertain due to price fluctuations on the market for the needed resources. A possible way to reduce manufacturing cost is to increase scale of production, which is expected to occur due to higher demand.

To address these challenges in cost estimation, a bottom-up analysis is appropriate. All production steps will be analyzed with emphasis on material and manufacturing cost. In order to get a comprehensive picture, the production of the whole battery system, including battery cell, module and pack, is investigated. The aims are to identify the most cost intensive manufacturing steps and to break down the cost per kWh on the three levels of the battery system. Additionally, the comparison of state-of-the-art cells with large format cells should reveal cost saving potential.

### ■ Reproducible Production of Various Test Cells for Academic Research

Paul-Martin Luc

Technische Universität Berlin

Coin (type 2032) and Swagelok cells are often used in the research and development for lithium-ion batteries (LIB). Based on the simple structure and the possibility of manual production, both cells types enable a quick and inexpensive examination option compared to larger cell formats. The structures or even the validity of different test cell types used for LIB is often insufficiently documented in the literature. Due to the lack of standards in this field, the comparability and traceability of presented results in literature is only partially or not possible. For this reason, the presented comparison of various test cells is focused on the reproducibility of the cell performance. Optimal and comparable setups for three different cell types (Coin Cell 2032, Swagelok, L-Cell) were investigated by connecting several published results which are unfortunately not building up on each other. Gaps from the presented studies will be filled with own experimental data using the design of experiments. For each cell type various parameters like separator count (one/two) or electrolyte amount were examined. To quantify the reproducibility of the cell performance parameters like the ohmic resistance and the capacity (fade) were determined and compared to the other cell types.

To validate this comparison and the relationship between parameters, at least eight cells of the same parameter setting where assembled and tested. According to the results from more than 300 assembled full cells a optimal cell type as well as an assembly method will be presented.

## ProZell – competence cluster for battery cell production

### ■ Pilot-plant scale production of nickel-rich cathodes for lithium-ion batteries

Carina Heck, Fabienne Huttner, Julian K. Mayer, Arno Kwade  
TU Braunschweig | Institute for Particle Technology (iPAT)

Nickel-rich layered oxides like  $\text{LiNi}_0.8\text{Co}_0.1\text{Mn}_0.1\text{O}_2$  (NCM811) as cathode active material can improve energy density, capacity and reduce material costs for lithium-ion batteries. However, a higher sensibility to moisture and carbon dioxide as well as a higher delithiation degree of nickel-rich cathode active materials can result in a stronger capacity fading during cycling and lower safety. Furthermore, in contrast to the fully commercialized NCM622, a production of NCM811 at industrial scale is still not well established. In this study, the NCM811 cathode production from dry mixing and dispersing up to cell assembly is shown for two polycrystalline NCM811 materials from different manufactures. The electrode production from slurry preparation to calendaring was performed within ambient atmosphere with a dew point of  $\text{TD} \approx 0^\circ\text{C}$ . Coating and drying was successfully carried out on a pilot-scale continuous convective coating and drying machine as a first step for a production at larger scale. For the individual process steps, microstructural properties were analyzed and affirmed by scanning electron microscopy. Furthermore, the moisture adsorption during calendaring of the cathodes was measured and the electrodes were electrochemically analyzed in full monolayer pouch cells. The results of this study indicate a good electrochemical performance of NCM811 cathodes processed within ambient atmosphere and post-dried under vacuum at  $80^\circ\text{C}$  for 5 hours. Also, these first investigations are highly relevant for the establishment of nickel-rich active material processing at larger scale and path the way for future materials like  $\text{LiNi}_0.9\text{Co}_0.05\text{Mn}_0.05\text{O}_2$ .

### ■ The influence of infrared post-drying parameters on residual moisture and structural and electrochemical properties of electrodes

Fabienne Huttner  
TU Braunschweig | Institute for Particle Technology (iPAT)

As moisture represents a critical contamination in lithium-ion batteries (LIBs), electrodes and separators need to be post-dried prior to cell assembly to guarantee a low residual moisture. On industrial scale, this is most commonly conducted in continuous roll-to-roll infrared dryers or discontinuously in vacuum drying ovens. The advantage of roll-to-roll drying is that the surfaces of the cell components are freely accessible. Hence, the diffusion paths are extremely short and short post-drying times can be implemented.

Previous research results have shown that regarding the electrodes not only low residual moisture, but in particular gentle post-drying is essential to obtain a good cell performance. [Huttner et al., 2019] High intensities can damage the sensitive electrode structures and deteriorate the physical and electrochemical properties. Therefore, a systematic investigation of the influence of different infrared post-drying temperatures and hold times on residual moisture and structural and electrochemical properties of LIB electrodes was carried out. The post-dried electrodes were analyzed regarding moisture, adhesive strength and electrical resistance. To investigate the influence of different post-drying intensities on the electrochemical performance, the most concise post-drying parameter combinations were identified for anodes and cathodes. Subsequently, electrodes were post-dried with the selected parameter combinations and built into coin cells.

The analyses showed that a high moisture reduction can already be achieved by a moderate post-drying temperature of  $80^\circ\text{C}$  and very short hold times between 60 and 120 s for both anode and cathode. These parameter combinations also led to a slightly better performance than higher post-drying intensities. In addition, the experiments showed that it is important to keep the dew point as low as possible during post-drying and further processing to prevent remoistening.

### ■ Effects of Production Scrap Rate on a Process Chain Level: An Energetic and Economic Assessment

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Regulative and social changes towards sustainability are promoting a significant growth of the electromobility sector. As a consequence of this development, an increase in the demand for lithium-ion batteries (LIBs) and its global production capacity is expected for the next years. The production of LIBs is characterized by an energy-intensive process chain due to high energy demands at various processes (e.g. calendaring and formation) and to maintain adequate production conditions. In addition, the production phase is the main cost driver, with materials accounting for up to 70 % of the costs of a single battery. Therefore, a more environmentally sustainable and cost-competitive battery cell production depends on an energy and material-efficient production. Different works in the battery production context with focus on energy efficiency and cost estimation consider in their models and calculations production scrap rates based on static values derived from historical production data. Moreover, these works do not discuss the influence of production scrap on the process chain dynamics (e.g. machine states and utilization rate). Against this background, this work presents the results of a combined discrete event and agent-based simulation that enables to (i) dynamically study the effect of different scrap rates on a process chain level and (ii) provide identification of critical processes from energetic and economic perspectives. The assessment of effects as well as the process identification are based on the indirect and direct energy demand and costs associated with production scrap.

### ■ Production-Oriented Numerical Optimization of NMC Cathodes with Maximized Energy Density

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Production of Lithium-Ion batteries has ramped up significantly in the past years and will continue to do so. The relationship between electrode microstructure, which strongly depends on the production process, and cell performance has been studied extensively in literature, e.g. by Schmidt et al. [1]. Model-based analysis of electrodes is advantageous in this context because no time-consuming experiments, including electrode fabrication, need to be conducted. Doyle-Fuller-Newman (DFN) models, being pseudo-two-dimensional (P2D), are computationally less expensive than their higher dimension counterparts; however, they often apply the Bruggeman relation to compute effective parameters, such as electrode ionic conductivity. Despite its simple implementation (i.e. a correction factor to reflect effective parameters), the Bruggeman relation delivers inaccurate results (Laue et al. [2]).

In this work, we coupled a DFN model with the surrogate structure model from [2], while excluding the Bruggeman relation. The model predicts the effective transport parameters and thus the battery performance more accurately. With this coupled model, we investigated the effect of active material and conductive additive on the discharge energy density. Furthermore, we developed a numerical optimization tool, which can predict the optimal content of active material and conductive additive, the porosity, and the thickness of the electrode for maximizing the discharge energy density. The tool yields the optimal structural parameters of an electrode for different manufacturing scenarios and different discharge current densities. Our findings would allow to further optimize the manufacturing process for NMC cathodes and the production of batteries with higher energy density. This approach can assist to increase the efficiency and cost-benefit of Lithium-Ion battery manufacturing in the years to come.

[1] Schmidt et al., J. Electrochem. Soc. 2020 167 060501

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### ■ Towards Logical Description and Linkage of Battery Data - DigiBatMat Platform of ProZell and MaterialDigital

Milena Perovic  
TU Braunschweig | Institute for Particle Technology (iPAT)

Modern lithium-ion batteries are a complex system consisting of cathode and anode active materials, conductive additives, binders, and electrolytes. These materials are brought together in a process chain of electrode preparation, including dry mixing and dis-

persing, coating and drying, as well as calendaring, and cell construction. The properties of a final product – a battery – are strongly dependent on each processing step, and on the properties of materials and their interaction.

The research on battery materials and systems is nowadays higher than ever, with numerous data being collected and published daily. However, the collected data is often difficult to correlate or even compare to each other, due to their high heterogeneity, resulting in a lack of interoperability. Digital Platform for Battery Material Data, Knowledge and Their Linkage (DigiBatMat) creates a platform for interoperable management of battery materials and process parameters data that will enable predictions of quality and performance through machine learning and correlation analysis. This is accomplished with structured data management through the formation of ontologies and taxonomies, a way of representing and structuring entities and their properties according to a system of categories. The reference electrodes with well-established cell chemistry based on NMC622 and graphite are prepared at the Institute for Particle Technology, whereas further steps in the cell preparation and electrochemical measurements are accomplished together with project partners. Furthermore, the platform is tested on new cell chemistry based on LM(N)O cathodes and LTO/activated carbon anodes.

After the successful development and implementation of the DigiBatMat platform, battery data across different users can be collected and structured albeit their heterogeneity. Ontologies will be used to clearly point to the relevant challenges, and interoperability of the battery research will be achieved.

### ■ Fast Charging Limitations during the Formation of Lithium-Ion Batteries

*Robin Drees*

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The formation of lithium-ion batteries is one of the most time-consuming processes during the production. Common formation methods consist of several relatively slow constant charging and discharging cycles (up to 50 hours) as it is believed to guarantee good performance of the Solid Electrolyte Interphase (SEI) on the negative electrode. The SEI has a significant impact on the performance, aging and safety of battery cells. Aiming at faster production times, cheaper production costs and better properties of lithium-ion batteries, the optimization of common formation methods is necessary.

This contribution is focused on developing optimized fast charging formation procedures. NMC622/G cells are characterized with a three-electrode setup and tested with different formation strategies. The cells are tested with an End-Of-Line-Test (EOL-Test) in order to check the cell quality. Afterwards the cells are optically checked for lithium-plating by opening the cells under argon atmosphere. It is found that the high current CCCV charged cells during the formation show lot of lithium-plating and have significant capacity loss while the optimized formation procedure prevents lithium-plating without having higher formation times. Furthermore, the optimized formation procedure result to similar capacities, internal resistances and impedances as the slow formation procedures.

### ■ The importance of passive materials in thick Li-ion battery electrodes

*Tobias Knorr*

*German Aerospace Center*

Due to their outstanding energy and power density, Li-ion batteries are widely used. The porous composite of a Li-ion battery electrode generally consists of active material, conductive carbon and polymeric binder. The so-called carbon binder domain (CBD) is distributed in the macro-pores of the electrode and both, provides better mechanical stability as well as electronic contact. However, at the same time the CBD increases the tortuosity of Li-ion transport pathways in the electrolyte. At high current densities, this increases mass transport limitations and reduces the performance of the battery cell. It has already been shown that the production process has a significant effect on the morphology and spatial distribution of the CBD.

In our contribution we present results of pore scale simulations in Li-ion batteries, which explicitly consider the morphology and spatial distribution of the CBD. In these simulations we study 3D realizations of NMC cathodes created by a 3D stochastic microstructure generator with varying density, particle size, thickness and CBD content.

In a first step we determine effective conductivities of the virtual samples which provides insight on limiting processes during operation of the battery cell. In a second step, we simulate the electrochemical performance of the virtual electrodes with an extended version of the “Battery and Electrochemistry Simulation Tool” (BEST) considering both the transport of electrons in the conductive carbon network as well as Li-ion transport in the pore space of the CBD. In our simulations we see that insufficient contact and high electronic resistance reduces the capacity at low CBD content. Interestingly electronic conductivity is not limiting the electrode capacity at high electrode densities, which opens up new strategies for the electrode design of for high-energy density batteries.

## greenBatt – competence cluster for recycling and green batteries

### ■ DIGISORT: Digitally improved sorting of lithium-ion batteries

*Alexandra Kaas*

*Technische Universität Bergakademie Freiberg*

The DIGISORT project is part of the greenBatt research cluster which examines heuristically LIB recycling. The project itself focusses on the digitalisation of the mechanical separation via air flow sorting by implementing new sensor technology. Through separation, the components and valuables of lithium-ion batteries have to be transferred into concentrates of high qualities and high recovery rates. In detail, the feed material in air flow sorting is a mixture of electrode foils which is contaminated with other material fractions. Depending on the types of batteries processed and the previous processing steps and parameters, the properties of the mixture vary in size, mass, degree of liberation and composition. As a result, stationary conditions rarely apply in battery recycling and challenge the following separation processes. A unique sensor technology is developed in DIGISORT recording the most important properties of the material in-line and on-line considering individually each fragment (particle) discretely. Image data with spectroscopic or hyperspectral information will be combined on particle level. Due to the enormous depth of information, these data sets will be structured as information vector for each particle. The control of the air flow sorting using the information vector is intended to increase the separation efficiency and thus the product quality and yield of the aluminium and copper concentrates. The present contribution introduces the broadness of material characteristics in LIB recycling. Different model mixtures as well as real recycling materials are characterised with regard to their size, shape and composition using a SOPAT camera. These off-line generated data serve as a comparison for the sensor technology to be developed in DIGISORT. In addition to characterisation of the feed material, the camera will be used to describe the separation products and process efficiency.

### ■ Development of Design Guidelines for Battery Systems in the Context of a Circular Economy

*Gregor Ohnemüller*

*Universität Bayreuth*

To reduce greenhouse gas emissions in Europe by 55 % till 2030, the energy and mobility transition sets new demands for traction batteries. The lithium-ion battery is a particularly promising battery type for an application in electric vehicles. Currently developed and manufactured lithium-ion battery systems are especially optimized in terms of performance, energy density, and costs. At the end of their usage phase, several End-of-Life strategies, e.g. reuse, remanufacturing or material recycling, can be applied to lithium-ion battery systems. However, the recycling of battery systems on product level is subject to certain requirements. This includes, e.g. the efficient and non-destructive disassembly of battery systems to enable a remanufacturing of the whole system and its components. As a result, battery systems as a whole are generally recycled on material level, since the removal of individual components from the battery system involves high technical and organizational



efforts. Despite the growing significance of battery systems, there is still a lack of research concerning the design of cells, modules and systems concerning an efficient recycling on product and material level. The project “Redesign” aims to increase the circularity of battery systems to avoid “open loops” in the product and material cycle and to reduce negative environmental impacts. This will be accomplished by the systematic development of design guidelines for a circular design and a recycling-friendly construction of battery systems to create the conditions for a closed, efficient, and low-loss battery system life cycle.

### ■ Life Cycle Sustainability Assessment of battery raw material supply chains

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Due to the increasing demand for batteries in electric vehicles and stationary energy storages as well as the related raw materials, the recycling of waste batteries and the return of secondary raw materials are important factors for reducing the risk of shortages and for ensuring economic success, especially in resource-dependent regions such as Europe. Besides, the recycling of waste batteries could reduce the environmental impacts and social risks of a battery pack by avoiding primary raw material extraction.

However, there are different options for recycling waste batteries, such as the hydrometallurgical or pyrometallurgical route, which may have different effects on the environmental and socio-economic impacts of the battery raw material supply chain. Therefore, it is necessary to demonstrate that potential recycling routes as part of the battery raw material supply chains are beneficial in terms of their environmental and socio-economic impacts compared to the primary raw material routes. For this purpose, the Life Cycle Sustainability Assessment (LCSA) approach can be used. Furthermore, the LCSA approach can be used to identify hotspots within the battery raw material supply chain to derive recommendations for action.

Thus, this poster aims to provide a better understanding of the environmental and socio-economic impacts of battery raw material supply chains, including battery recycling. To this end, an exemplary battery raw material supply chain is assessed in terms of environmental and socio-economic indicators. The LCSA approach is implemented in the python-based Brightway2 framework. The results of the LCSA reveal hotspots such as the transportation of waste batteries in this supply chain and demonstrate improvement potentials for the supply chain by using secondary raw materials. Furthermore, the results show that trade-offs exist between the different impact categories that need to be considered in the context of decision support.

### ■ Polymer-SSB Recycling – How mechanical processes can be adapted for efficient recycling

Marco Ahuis, Stefan Doose, Peter Michalowski, Arno Kwade

TU Braunschweig | Institute for Particle Technology (iPAT)

The electrification of transport and the associated increase in the use of lithium-ion batteries is leading to growing raw material demand, which may result in raw material shortages and soaring material costs in the future.

In this context, solid-state batteries (SSBs) are promising next-gen batteries that can be used as high-performance energy storage systems and score with advantages such as increased lifetime, energy density and safety compared to today’s lithium-ion batteries. For successful development and establishment of the polymer SSB system in the market, possible supply chains have to be identified and sustainability over the life cycle has to be achieved. With this context in mind, recycling and reuse is an essential component of a climate-neutral energy system. The implementation of the targeted circular economy enables cost-effective and sustainable production, which can only be achieved with efficient recycling processes.

In this contribution, a possible adaptation of processes for early development of mechanical recycling processes for polymer-SSB is shown. The different structure of the polymer SSB requires the recombination of existing process steps but also integration of new processes.

This work presents that the use of conventional mechanical processes such as shredding and screening in combination with wet process steps allows a simple and robust separation of the comminuted battery materials. Advantageous about this pro-

cess route is the possible early recovery of lithium, which will gain great importance in the near future. Furthermore, the cathode active material can be separated in a gentle way within this process route to be reused as recycle. The results obtained are compared by means of TGA, ICP-OES and SEM, among others, and impurities caused by process design are identified.

### ■ Leaching of black mass from lithium-ion batteries by biodegradable leaching agents

Monika Keutmann, Dominik Schmitz, Bernd Friedrich

RWTH Aachen University | IME Institute for Process Metallurgy and Metal Recycling

The recycling of lithium-ion batteries is a very important point in the European circular economy to return back rare materials like lithium to the production of batteries. For recycling processes, the industry commonly uses mineral acids for their leaching process, which leads to non-biodegradable waste. In this context, the investigation of biodegradable leaching agents is essential to develop ecological and sustainable processes. These environmentally friendly processes can be considered as zero-waste processes.

There is already a number of publications on the recycling of lithium-ion batteries with organic acids. However, most of them only deal with a few selected organic acids. Additionally, the results from these publications are difficult to compare, cause there using different black mass. In this publication, a wide range of different organic acids is set in comparison, using the same black mass as a starting point, to ensure a very high comparability of the individual acids. In addition to the organic acids, deep eutectic solvents (DES) will be investigated as biological leaching agents. These DES consist of two different organic solids, such as choline chloride and urea. In certain molar ratios, these form ionic liquids that can be used to dissolve metal ions. Furthermore, the analyzation of various influences is researched. In some publication are shown that concentration, temperature, leaching and time has effects of leaching quality. So these parameters were researched for every organic acid and are parts of the research with DESs to determine the most suitable leaching agent.

### ■ Data-Mining in the Recycling of Lithium-Ion Batteries

Sandro Süß

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Closing material loops is essential for sustainable electric mobility. Therefore, efficient recycling systems are needed, which maximize the output quantity and quality while minimizing the associated costs and environmental impacts. Here, especially the battery is of interest. To support efficient recycling systems for batteries, knowledge about optimal settings for process parameters in their recycling processes is needed. This requires information about potential cause-effect relationships between recycling inputs, process and quality parameters. Information on the recycling inputs can be derived from battery life cycle data. In addition, data from sensors and control loops implemented in the recycling processes support the derivation of information on relationships between recycling process parameters and recycling quality. Both help to significantly increase the achievable recovery rate and quality while reducing energy and material consumption.

The focus within the project DiRectION is on the collection, processing, and evaluation of data from the recycling of lithium-ion batteries, as well as developing a related structured data platform. Based on this, the analysis and visualization of the overall recycling process chain and individual processes become possible. This supports the build-up of data-based and artificial-intelligence-based modeling to derive decision support for new recycling strategies and optimized process parameters. Furthermore, the information can be used to derive requirements for a recycling-oriented design of batteries.

This work presents a framework for the data-based analysis and evaluation of the recycling of lithium-ion batteries. In particular, it focuses on the involved stakeholders along the life cycle of the battery and our concept for the measurement infrastructure, which incorporates all the requirements for the sensor technology, data interfaces, data processing and data management along the mechanical recycling process chain.

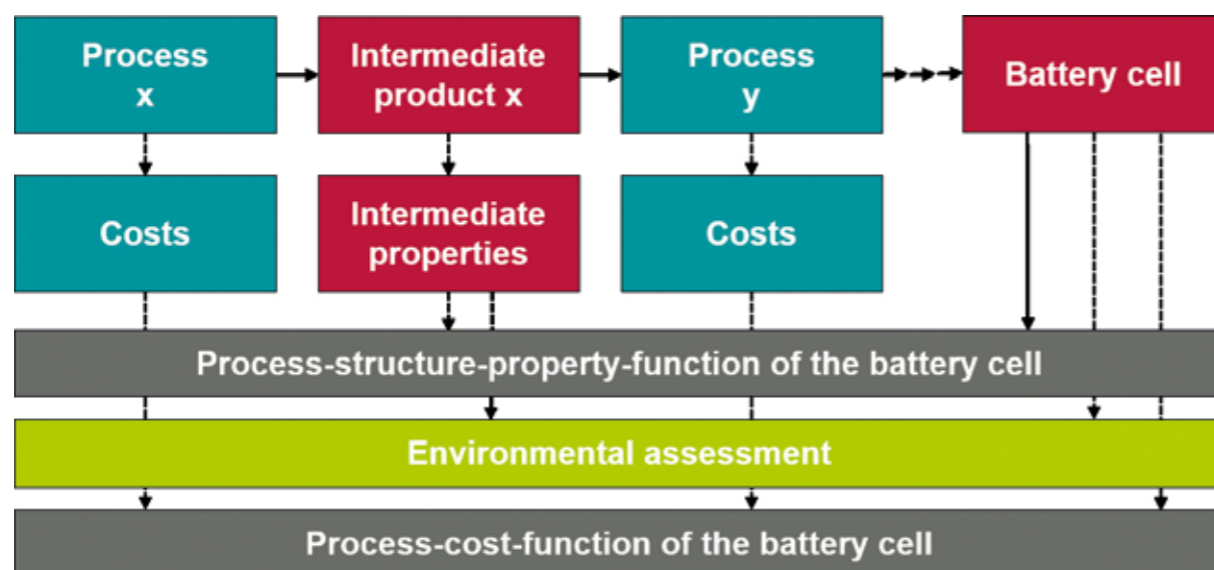
## ProZell – Competence cluster for battery cell production – Influences of production steps and parameters on cell performance and quality – Challenges and goals

In the competence cluster for battery cell production (ProZell), German research institutions join forces to strengthen the national battery cell production. The aim of the competence cluster is to research and improve the entire process chain of the battery cell production and assess the influence of each individual production step on cell properties, product development costs and sustainability. The competence cluster elaborates the scientific basis for the establishment and sustainable development of an internationally leading, competitive battery cell production in Germany. In this context, the economic efficiency of cell production and the environmental assessment are highly relevant. While aiming at reducing the energy-related cell price (€/kWh), the competence cluster pursues the simultaneous goals of increasing cell performance, especially energy density, and elaborating recycling pathways for battery cells.

The central concept of the cluster is to cross-link specific knowledge, special equipment, and various research institutions in joint projects. In 16 research projects, experts and scientists from various fields of expertise collaborate on identifying variables that cause relevant changes in intermediate product properties, as well as cell performance, quality, and costs. Process-structure-property relationships and process-cost-functions along the entire process chain of the battery cell production are developed. An accompanying project fosters cooperation and networking within the entire cluster and ensures a structured bundling of knowledge in a results database. In addition, an advisory board including representatives from industry and research advises the projects and fosters synergies between research institutions and industry.

### Contents and main areas of work

The continuous production of battery suspensions by extrusion, in addition to increasing the energy density through the targeted structuring of high-capacity electrodes and the use of silicon are areas of focus for the field of electrode production. A pre-lithiation technique adds lithium to the anode prior to cell assembly in order to increase the stability of lithium ion battery cells containing silicon during charging and discharging cycles. A novel dry-coating technology allowing for solvent-free electrode production diminishes the need for energy cost during expensive drying procedures and opens the door towards environmentally friendly electrodes. In addition, interactions between process control and product properties are investigated in detail, especially for calendaring and post-drying of electrodes. Within the field of cell production, the optimization of filling and wetting processes, taking into account all essential cell components, is topic of intensive research. The energetic



optimization of cell formation is investigated. The investigations regarding cell stack formation focus, above all, on the special requirements of high-energy electrodes. Overarching projects in the cluster focus on developing innovative quality assurance concepts to reduce fluctuations and rejection rates and to optimize the interlinked production processes with regard to an appropriate definition of production tolerances. Based on mathematical models for the entire process chain, the detailed understanding of related process-structure-property relationships is used in order to digitally describe the entire process chain and optimize it for new battery generations. Further points of interest are the comprehensive cost and environmental assessment of the process chain and the recycling of battery cells. A results database brings together key findings of the projects in a transparent manner.

### Application, use of results and contribution to energy storage

The properties of electric vehicles and systems for the electrochemical storage of energy as well as their respective customer benefits correlate directly with the properties of the battery cells used. A better understanding of influencing variables along the entire process chain, including the production environment, is therefore essential. The establishment of an economical and sustainable battery cell production is the central milestone on the way to establishing Germany as a leading market and provider of electro-mobility. The fundamental challenge for competitive battery cell production is to increase cell performance while simultaneously reducing the energy-related cell price (in € per kWh). The knowledge gained in the competence cluster should form the essential basis for the development of economically producible battery cells, i.e. battery cells with a significantly improved performance-to-cost ratio. The results provide a scientific basis for achieving and continuously expanding the sustainable, international technology and cost leadership of all German industries involved in the value chain of cell production.

### Partners and funding

Currently, the ProZell competence cluster comprises 19 partners, including the following universities and research institutions: TU Braunschweig, Karlsruhe Institute of Technology, Landshut University of Applied Sciences, TU Berlin, TU Clausthal, TU Dresden, TU Bergakademie Freiberg, Center for Solar Energy and Hydrogen Research Baden- Württemberg, German Aerospace Center via the Helmholtz Institute Ulm, Ulm University, RWTH Aachen University, TU Munich, Münster Electrochemical Energy Technology (MEET) at University of Münster, Fraunhofer-Gesellschaft, and Forschungszentrum Jülich via the Helmholtz Institute Münster.

The ProZell cluster was funded by the Federal Ministry of Education and Science (BMBF) with more than 16 million euros in the first funding period from 2016 to 2019. Due to the promising results and the good cooperation and collaboration within the ProZell competence cluster, the BMBF has been intensifying its financial support for battery research since 2019. The second funding period of ProZell started on October 1st, 2019 and comprises more than 35 million euros in total funding. The successful ProZell concept serves as a model for the establishment of further competence clusters by the BMBF, namely: InZePro – Intelligent battery cell production, greenBatt – Recycling/Green battery, BattNutzung – Battery use concepts, AQua – Analytic/Quality assurance.

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# PRESENTATION ABSTRACTS

## Electrode Production

### ■ NETZSCH Helios fast and efficient dispersion system for Li-ion Battery Electrode coatings

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The market for Lithium-Ion-Batteries (LIB) is growing significantly. Therefore, cell producers demand scalable cost efficient processes.

Planetary mixers are mature and often used as state-of-the-art. Due to limited mixer sizes, multiple planetary mixing systems are required to meet the slurry demand in large factories. Especially Gigafactories require alternative processes in order to reduce production cost of LIB cells.

The system HELIOS separates the individual mixing and dispersion steps within the slurry production. Raw materials are treated selectively and with adjusted energy input. The modular concept enables cell producers to react flexibly on materials variations in each single step of the process, and consequently process times can be always optimized. Each process step is fully controllable in order to achieve reproducible product qualities. A case study for a 16 GWh line clearly shows the benefits of System Helios compared to planetary mixing process. The system Helios reduces considerably investment and production costs by avoiding large number of planetary mixing lines.

### ■ Simultaneous two-layer slot die coating of ultra-thick LIB electrodes

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Lithium-ion batteries (LIB) have attracted much attention in the recent years. An increase of energy density which is additionally expanded to higher current densities would crucially extend their range of application possibilities. To increase energy density, electrodes can be made ultra-thick [1] to provide an increased share of active material. An approach to mitigate the accompanied limitation of rate capability is to customize the local structure of the electrode composite [2].

Such a customized structure can be realized by simultaneous two-layer slot die coating of two differently composed slurries. However, the composition of the slurries influences their rheological properties which in turn determines their behaviour in each of the involved process steps of electrode fabrication. Furthermore, the adjustment of a slurry for an optimized behaviour in one process step will additionally affect its behaviour in other process steps.

In this contribution, the interrelation between slurry formulation, solid content and rheological properties is shown and their impact on the behaviour in the process steps of mixing and dispersing, casting and drying is elucidated. At the example of ultra-thick NCM 622 electrodes, it is explained what has to be considered to reach a working process and realize a targeted ratio of the two layers with respect to their contribution to capacity.

By applying simultaneous two-layer slot die coating, the volumetric capacity of an ultra-thick cathode was increased by 50 % at a rate of 1 C. This result demonstrates the potential of this procedure and its suitability to expand the range of usage of ultra-thick electrodes to higher current densities.

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### ■ Producing Battery Compounds

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*Coperion GmbH*

Coperion provides ideal feeding and extrusion technology for producing a wide variety of battery components. The focus is on the continuous production of electrode slurries and separator films.

Ensuring consistently high product quality is one of the key advantages of the continuous production of battery compounds. Active materials, binding agents, conductive carbon and liquids are used as raw materials. These are fed either via independent Coperion K-Tron feeders or premixes. Precise adherence to the formula ensures the high quality of the end products. Certain raw materials are partly toxic and/or abrasive. For this reason, processing in the Coperion ZSK extruder takes place under controlled conditions, from feeding the raw materials through to discharging the compound. Feeding the raw materials with dust-tight equipment is just as important as avoiding metal contamination in the end product. Furthermore, ZSK extrusion systems intended for producing battery compounds are designed in compliance with stringent explosion protection regulations due to the raw materials.

### ■ Laser structuring in battery production for enhancing the electrochemical performance of NMC 811 high energy electrodes for xEV energy storage systems

*Bernd Eschelmüller*

*AIT Austrian Institute of Technology GmbH*

To fulfill the ambitious requirements for future Li-ion batteries in electric drive applications, e.g. 700 Wh/l, layered oxide cathodes, especially lithium nickel manganese cobalt oxide, NMC, thick-film electrodes are under continuous investigation. Currently, most current studies attempt to reduce the Co content with a concurrent increasing Ni-content in the NMC cathodes such as NMC 811. Unfortunately, NMC cathodes suffer from low high rate capability and corresponding low capacity retention at high C-rates. In particular, the negative impact is even higher for thick-film high energy cathodes. To counteract the negative effect, high repetition ultrafast laser ablation is applied to create appropriate 3D electrode designs. The laser structuring process provides new Li<sup>+</sup>-diffusion pathways, enhances electrolyte wettability, and reduces overpotentials at high C-rates.

It is attempted to integrate the laser structuring into a continuous roll-to-roll electrode production process. In this way, the positive properties achieved through 3D structuring can also be obtained for Li-ion batteries that are produced on a large scale. By using this novel production technology, future NMC batteries can be produced with improved performance characteristics for xEV applications. This work is performed under the frame of the RealLi! project, in which the following aspects are covered:

- 1) Development of thick film NMC 811 electrodes with high areal capacity.
- 2) Passivation approach to improve cycle stability and lifetime.
- 3) Cell Assembly and electrochemical characterization.
- 4) Evaluation of the environmental impact of the NMC 811 cells via life cycle assessment.
- 5) An experimentally validated electrochemical model to describe electrode structures and their optimization.
- 6) Improved electrochemical performance of NMC 811 electrodes on a laboratory scale by using 3D laser structuring.
- 7) Scale up of the 3D laser structuring process and improved electrochemical performance.

### ■ Electrode structure optimization adjusting battery manufacturing parameters by applying computational EIS

*Clara Ganuza*

*CIDETEC*

EIS is a nondestructive, effortless, fast and widely used technique that parametrizes battery cells. It provides information about the electrochemical system and its internal reaction mechanisms, in particular: charge transfer kinetics, mass transport and thermodynamics. This technique consists in applying small sinusoidal amplitude input either current or voltage for a wide range



of frequencies and processing the sinusoidal signal output in the frequency domain, obtaining a spectral representation of the tested cell.

We present a computational EIS that consists of a sequence of simulated tests in which the described procedure is followed. Time domain outputs are processed using discrete Fourier analysis. This method is applied to an adjusted P2D physics-based model for blocking electrolyte conditions, which is commonly used to characterize the electronic limitations of the electrode and can describe the whole cell frequency response. The aforementioned model approach can be used for determining unmeasured parameters by fitting the simulation to experimental results.

For this purpose, and in order to validate the model and the computational EIS method, several EIS experiments were made for which we manufactured some electrodes under different calendaring conditions. As the last step of the process chain of the manufacturing of the lithium-ion battery electrodes, calendaring has a major impact on electrode structure, thus enhancing the electrochemical performance of the produced cells. Understanding this compaction process is of vital importance in order to determine the performance-optimized pore structure. In this study, NMC622 cathodes are pressed against two cylindrical rolls at various speed, pressure and gap between the rolls, obtaining different coating densities and porosities, getting a full parametrized data set for the electrochemical model validation.

### ■ Development of a solvent reduced electrode production process integrating coating, drying and calendaring

*Eike Wiegmann*

*TU Braunschweig | Institute for Particle Technology (iPAT)*

Conventional electrode coating processes for Lithium-ion batteries, such as doctor blade or slot die coating, require relatively low solids contents, which entail long drying times and the associated high energy and investment costs. In this form of electrode processing, due to the high solvent content, segregation of inactive components of the electrodes, which impair their physical properties, can be detected. Therefore, a new process based on very low solvent content was developed. The process was investigated for the manufacturing of high viscous water based graphite anodes and LFP cathodes. With a mass content of less than 25% of water for the anode and cathode slurry, the solid ingredients were processed together with the solvent in a twin screw extruder combined with a strand pelletizer. Such a processing enables a highly filled paste to be produced, which is stable in storage. Afterwards, the obtained high viscous electrode granulates were coated directly within the calender on the electrode copper foil. After calender coating, the coated substrate is dried continuously using an IR radiator field. In addition, this type of coating achieves an increased initial density of the electrode so that calendaring is not unconditionally necessary. Due to the high viscosity of the electrode pastes, the individual components such as the binder and active material are exposed to high shear stresses during the processing inside the extruder and calender gap. This requires mechanical and thermal stability of the used binders and can change the morphology of active material particles. The results show, that the produced anodes show a higher cell capacity at higher C-rates due to the structural changes of active material based on the high shear stress within the twin screw extruder. Furthermore, energetic costs of this process are referred to a conventional electrode production process. It will be shown that the energetic cost for mixing and drying can be reduced to half.

### ■ Electronic pathway length manipulation and impact on conductivity networks in lithium-ion battery electrodes

*Jake Entwistle*

*University of Sheffield*

Pursuit of increased energy density has put downward pressure on the inactive constituents in lithium-ion battery electrode formulation. Energy-formulated electrodes now typically containing less than 2wt% conductive additive. This requires increasingly efficient conduction networks through the solids to maintain adequate levels of electronic conductivity for a cell to function

properly. There is increasing evidence[1] that the combination of short (1 particle) will provide an optimal conductivity balance. The exact combination of long and short and the fundamental governing principles of how to achieve both types of pathway is still an open question. However, processing the conductive additive, in particular carbon black, under a range of shear has enabled a degree of control of pathway length through fragmentation of the carbon black.

This work investigates the use of 'long' and 'short' range electrical conductivities within battery electrodes by combining a dry-processed pre-structured electrode powder with a traditional slurry casting process to control the relative number of length pathways. We present the physical and electrochemical characterisation of these networks to build a holistic understanding of electrode performance.

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### ■ Calendaring of carbon coated nano-silicon graphite composites (Si@Gr/C)

*Jannes Müller*

*TU Braunschweig | Institute for Particle Technology (iPAT)*

Silicon-graphite composites are considered the most promising future active material on the anode side for near-term industrial application. However, silicon suffers from two main drawbacks, a large volume change during lithiation [1, 2] and its poor electrical conductivity [3]. In order to address these drawbacks, a carbon-coated nano-silicon graphite composite (Si@Gr/C) with 8 wt% silicon ( $\approx 600 \text{ mAh g}^{-1}$ ) was developed in a previous study [4], which showed superior rate capability and long-term cycling stability. Based on this, within the current investigation the process route of composite manufacturing (fluidized bed granulation and carbonization) and electrode manufacturing (dispersing and coating) have been scaled up to a small pilot scale. Furthermore, a calendaring study of coated (Si@Gr/C) and uncoated (Si@Gr) composites has been carried out and its influence on electrode properties and electrochemical performance in half-cells has been evaluated.

It could be demonstrated that calendaring these silicon graphite composite electrodes had only minor influence on mechanical and electrical properties. In contrast, the rate capability and long-term cycling stability deteriorated clearly with increasing electrode density. A similar behaviour for initial discharge capacity and initial coulombic efficiency, both following a linear trend, could also be found. This behaviour might be related to the lower porosity and thus less space for the expansion of the silicon particles.

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### ■ Performance increase and process enhancements for capacitors and batteries production by aLITE-Technology

*Kai K. O. Bär*

*adphos Innovative Technologies GmbH*

The adphos developed proprietary advanced Light Initiated Thermal Emission (aLITE) – Technology consists of a high energetic photonic energy source (for energy transfer) in combination with separate adjustable air ventilation (for mass transfer) for accelerated drying and thermal treatments in a wide range of capacitors and batteries production processes.

The aLITE-process result in single digit seconds (2–8 s) drying and only requiring a fraction of energy (of today's solutions) of battery electrode coatings, depending on coating type and layer thickness. Even for 3D-multilayer integrated battery productions, proper and complete coatings (for electric conduction layers, isolation layers, electrode layers, electrolyte layers, separator coatings, ...) drying has been already successfully proven.

A competitive evaluation with traditional drying processes (hot air and IR-assisted hot air) and laser based drying technology will be outlined based on experimental studies. The extraordinary benefits of the aLITE-technology will be demonstrated on a layout for a new integrated battery production facility as presently build.

### ■ Enabling aqueous processing of positive electrodes for lithium ion batteries – Challenges and opportunities for process and electrode design

Markus Börner

University of Münster | MEET Battery Research Center

Apart from energy density, cycle life, and safety of lithium ion batteries (LIBs), the continuous reduction of costs and environmental impact is crucial for future large-scale LIB production. In this regard, replacing the toxic and costly NMP solvent in LIB processing by aqueous binder systems is highly desirable. However, aqueous processing of positive active materials is facing challenges due to side reactions.

To develop an aqueous production process for positive electrodes, an in-depth knowledge of the key reactions during processing is inevitable. Therein, the lithium-proton exchange during aqueous processing was identified to greatly influence the performance of NCM-based electrodes. While commonly mild acids are used to prevent the accompanied rise of the electrode paste pH value, it could be shown that an increased pH value of the binder solution, effectively reduces the exchange reaction leading to an improved performance. Another approach to reduce the lithium-proton exchange reaction is the introduction of Li containing processing additives. Thus, it could be shown that specific processing additives do not only reduce the exchange reaction but also become active during charge/discharge cycling improving performance and lifetime as additional conducting salt.

Apart from the challenges regarding the instability of state-of-the-art active materials towards water, the use of aqueous processing also offers a large variety of opportunities to tailor the process and resulting electrode structure. Therein, the application of surfactants can lead to an improved connectivity between electrode components resulting in superior cycling performances. Beyond that, a large variety of aqueous binders and their combination can be used to tailor the electrode paste viscosity and the mechanical stability of the composite electrode.

Finally, the developed approaches are verified by up-scaling the processes to production on a battery pilot-line.

### ■ Structuring of particle systems for the production of wet and dry electrodes with Eirich intensive mixers

Stefan Gerl

Maschinenfabrik Gustav Eirich GmbH & Co KG

In order to further increase the performance of electrodes for LIB, the particle system that makes up the cathode is increasingly being structured. This process usually goes far beyond the state-of-the-art mixing of active materials, conductive and binders. The Eirich mixer has already proven its performance here- even more powerful machine technology, such as mixing tool circumferential speeds of up to 50 m/s and efficient double jacket cooling now enable further leaps in performance in the application.

A common method for particle structuring/design in this context is the initial dry coating of the active material with nanoparticles. Depending on the dispersibility of the nanoparticles, different shear forces must be applied for this purpose. Complete disintegration of the conductive carbon agglomerates and homogeneous coating of the active materials are basic requirements for dry electrode production but open the doors for further improvement in wet electrode production as well. Experimental investigations showed significant changes in powder properties depending on operating parameters used.

The active material- additive mixtures structured in this way can now be processed in a subsequent step into applicable electrode compounds. For this the powdered polymer binder is added directly to the previously prepared powder mixture and homogenized. In wet preparation the coating slurry is prepared with the addition of NMP via the kneading and dilution phase. For the production of dry electrodes the dry mix is fibrillated under high shear stress. Depending on the polymer content, shear stress and mixing time, this produces a sticky powder mixture or even a kneadable mass. In the Eirich mixer, this can subsequently be converted back into an easily meterable granular to powdery bulk.

The Eirich mixer is the only mixer available on the market that allows the structuring of active material as well as the production of wet and dry electrode masses in a one-pot process.

### ■ Schwarz-Primitive structured electrodes for lithium-ion battery via 3D printed wax templating

Yige Sun and Patrick S. Grant

University of Oxford | Department of Materials

Despite steady progress in improving many performance aspects of Li-ion batteries, power density remains a key characteristic where further improvements are required. A common approach to preserve power performance for thicker electrodes is to engineer directional porosity through the electrode thickness to facilitate faster ion transport.

Various types of templating techniques show the benefits of pore engineering on electrode dynamic response but most suffer from insufficient control and reproducibility, difficulty in scaling etc. Nonetheless, progress continues, and the magnitude of these challenges is being progressively reduced, and in some cases, overcome.

This paper describes the application of a new approach to pore templating for LIBs, based on ink-jet printing of wax. This combination was selected for the potential for large area, low cost, easy template removal, reasonable speed, and flexibility. As an initial step, an electrode slurry with carbon nanofiber (CNF) as the active material was infiltrated into a dual-wax ink-jet printed template. After chemical removal of the wax template, leaving no residue, a 200 µm thick CNF-based Schwarz-P primitive labyrinth (bi-continuous pore and CNF network) electrode was produced. The electrode was investigated by in detail by microscopy and electrochemical testing.

Although rate performance was improved by the pore structure, volumetric capacity – as often for pore templating approaches – was low. Therefore, a further novel step was introduced that selectively filled some pores with a high-capacity cathode material. This hybrid electrode provided significant improvements in electrochemical response, including volumetric capacity while preserving power response. The advantages and disadvantages, and future potential, of this approach, are described in detail.

## Cell Assembly

### ■ Inline X-ray Metrology for Lithium-Ion Battery Cells

Markus Möller

Viscom AG

X-ray inspection plays a major role in today's quality control for the mass production of lithium-ion battery cells. Viscom X-ray metrology solutions are developed especially for the use in the battery cell industry. The product portfolio ranges from lab systems to full inline solution for 100% quality control at high speeds. With X-ray inspection from Viscom customers can ensure the product safety, long term performance, process efficiency and process learning, as well as traceable product data. Typical inspection criteria's among others are anode – cathode overhang, telescoping, bent anodes, crimping analysis and many more. Viscom is specialized on a variety of different cell formats, like coins, cylindrical, pouch cells or prismatic cells covering different sizes for consumer products over energy storage to e-mobility cells.

## ■ The warmer, the faster? - An investigation of the temperature dependence on the wetting process

*Nicolaj Kaden*

*TU Braunschweig | Institute of Machine Tools and Production Technology (IWF)*

With the increasing importance of electrical storage technologies for mobile and stationary applications, the need for improved productivity to increase throughput and reduce manufacturing costs is growing. In current cell production, the electrolyte filling of lithium-ion batteries and the subsequent wetting are essential process steps and represent the interface between cell assembly and its formation. The electrolyte filling and wetting process presents a high potential for throughput increase and cost reduction through a reduction of the otherwise usual long storage times. Best practice solutions are currently being used. However, it has hardly been systematically analyzed, which mechanisms dominate filling and wetting, and how these can be accelerated.

One of the many reciprocally influencing factors is the temperature of the cell during the wetting process. For instance, it has a major influence on the wetting-relevant material properties of the electrolyte, such as surface tension and viscosity. To gain a better understanding of the temperature depending on wetting mechanisms, the wetting behavior of different cell components has been investigated in a first step. The impregnation and wetting of the porous structure of the electrodes and separators was determined by a modified Washburn measurement with a tensiometer for different electrolyte temperatures. The material-specific findings obtained were then applied to the wetting behavior of assembled battery cells. For this purpose, the wetting behavior of battery cells with different numbers of compartments and geometric dimensions was investigated. The wetting state was characterized through electrochemical impedance spectroscopy at different storage temperatures of the battery cells during and after filling.

In further investigations, additional influencing parameters, such as the geometric dimensions and the aspect ratio of the cells, will be examined in more detail.

## ■ Quality assessment for laser-based contacting of lithium-ion battery cell components using inline process monitoring

*Sophie Grabmann*

*Technical University of Munich | Institute for Machine Tools and Industrial Management (iwb)*

One major challenge in the manufacturing of lithium-ion battery cells is the qualification of reliable and stable production processes. The cell internal contacting addresses the electrical connection of the uncoated current collector foils of the individual electrodes of the cell stack. The requirements for the joining process are high as the foils are very thin, and other components of the battery, such as the polymeric separator, are very sensitive to high temperatures. Ultrasonic welding, which is the established contacting process, leads to an increased risk of mechanical damage. In contrast, laser beam welding is a promising alternative due to its high productivity as well as the flexible and contactless process design.

Different welding strategies and the influence of process parameters on the weld seam quality were investigated. Foil stacks were welded with an arrester tab in lap joint configuration. Applicable methods and techniques for the inline monitoring of the process were evaluated to ensure a high product quality. The used quality measures were a high mechanical integrity and a low electric resistance. Different sensor systems based on process emissions and image data were compared and patterns in the signals identified. The investigations showed that quality criteria of the laser-based contacting process could be assessed using inline measurement techniques. Additionally, the integrability of this new technology into the industrial battery production lines was considered. Further investigations will be carried out to merge the processed signals with the quality criteria using machine learning techniques.

## Recycling & Sustainability

### ■ Recovery of spheroidized graphite from spent lithium ion batteries

*Anna Vanderbruggen*

*Helmholtz Zentrum Dresden Rossendorf*

Recycling of lithium ion batteries has attracted a lot of attention and is particularly focusing on the valuable metals such as cobalt, nickel and lithium. Despite the growth in graphite consumption and the fact that it is counted as a critical material in Europe, USA and Australia, there is little previous work focusing on graphite recycling. Thus, graphite is usually not recovered as a product but rather used as a reducing agent during metallurgical treatments. The aim of this research is to increase the recycling recovery of the LIBs by integrating a flotation stage. This recycling process is able to separate active electrode particles. Two valuable products, one of graphite and one with the valuable metals are recovered using a batch mechanically agitated Outotec flotation cell. The batch flotation study shows that pre-treatment, such as attritioning, improves the process. The graphite recovery from the fine fraction of the black mass (< 100 µm) is +98 % with a grade of 85 wt. %. This research aims to reach a closed-loop system for spheroidized graphite from spent LIBs.

### ■ Aqueous processing of cathode slurries with nickel rich active materials and sustainable binder for green battery production

*Anton Werwein*

*Fraunhofer Institute for Ceramic Technologies and Systems IKTS*

The cost-efficient and environmentally friendly manufacturing of battery electrodes is an important factor on the way to a sustainable Lithium-ion cell production. Today, N-Methyl-2-pyrrolidone (NMP) is mainly used as a solvent for the PVDF-binder for cathodes. Since NMP is toxic, explosive, has a high boiling point and needs to be recovered, an aqueous processing of cathode slurries is favorable to reduce material and investment costs. With water as solvent new challenges appear e.g., the demand for water soluble binders that are stable at high potentials, the lithium leaching of nickel rich materials, high pH values and surface cracking during the coating of thick films. There are several attempts to tackle these problems. One of them is the usage of alginate from brown algae as binder. In this work sodium alginate was used as binder for different cathode active materials (NCM622, NCM811, NCA). The electrode slurry was mixed in a dissolver and the pH was adjusted. Different mixing routes were carried out to investigate material process interactions. The binder enables a good dispersing of the carbon black and the active material particle. The slurries are suitable for electrode coating due to the strong shear thinning behavior and the good adhesion. The cycle stability and rate capability were tested in half cells and compared to equivalent PVDF based cathodes. An initial capacity loss due to the aqueous processing was observable. Additionally, the electrode morphologies were studied by porosity measurements and scanning electron microscopy. In this work a feasible aqueous processing route for stable cathodes were established. The sodium alginate as binder enables the formation of a homogeneous suspension and a stable coating even at high mass loadings without additional preparation steps.

### ■ Techno-economic and environmental assessment of production processes for all-solid-state batteries using the example of active material coating by atomic layer deposition

*Deidre Wolff*

*Fraunhofer Institute for Surface Engineering and Thin Films IST*

Current research on all-solid-state batteries intensively deals with various new material classes, innovative cell concepts and suitable production processes. However, there is a need for systematic evaluation and comparison of the corresponding technologies based on techno-economic and environmental assessments. Furthermore, as identified within the FestBatt cluster of competence and aligned with current research, there is a high demand for electrochemically and mechanically suitable coatings



for cathode active materials [1]. One aspect of the EProFest project is the development and comparison of innovative coating processes. This includes the investigation and evaluation of promising coating materials, such as niobates and halides, and techniques, such as spray coating, physical vapour deposition and atomic layer deposition (ALD).

The focus of this work is the exemplary investigation and evaluation of the ALD coating technique for nickel-manganese-cobalt active materials (NMC) using lithium containing phases and suitable oxides, as developed in EProFest. The main benefits of ALD include homogenous and uniform thin film deposition on complex-shaped geometries and high material efficiency compared to other techniques, such as high throughput spray coating. In order to further compare this process to other coating techniques, the energy consumption, material flows and product properties are further investigated, and the techno-economic and environmental impact assessed from a life cycle perspective. EProFest aims to support the development of promising process chains for all-solid-state batteries and materials by giving recommendations for action for comparable process steps.

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### ■ Characterization of spent lithium-ion batteries in different recycling process routes using automated mineralogy

*Kai Bachmann*

*Helmholtz Institute Freiberg for Resource Technology | Erzlabor Advanced Solutions GmbH*

The growing global demand for LIBs is accompanied by an increase in the need for Co, Mn, Ni, Li and graphite. In order to narrow the gap between supply and demand and to achieve the European sustainability goals, the recycling of LIBs has attracted a lot of attention in recent years. Due to a constant evolution of LIB chemistry and production processes, battery material remains to be a huge challenge for recycling and as well as a proper materials characterization. Hence, there is not only a need to find innovative and comprehensive LIB recycling process solutions but also to develop new analytical workflows to enhance the understanding of the recycled battery material. Here, LIBs are fed to a mechanical, thermo-mechanical and electro-mechanical recycling process route to liberate the valuable active materials from electrodes foils. In addition, to the valuable metals Co and Ni, the focus is particularly on the recovery of spheroidized graphite. The mechanical route works with an impact shear crusher, while for the thermo-mechanical tests the batteries were vacuum pyrolyzed at 500-650°C before crushing. For the electro-mechanical route, the batteries were immersed in water and opened with a shock wave fragmentation system. The so-called black mass fraction smaller than 1 mm was separated and further classified into four size fractions. For visualization and quantification of the results of the processing success, this study proposes a characterization method based on automated mineralogy (MLA), providing results of various important particle parameters such as size, composition and adhesion. While these analytical systems are well established in the primary raw materials sector, there are no dedicated databases for use in the secondary raw materials sector in order to analyse black mass material in a fast and precise manner. An analytical challenge of this study is therefore to create an understanding for the behaviour of active during the recycling processes.

### ■ Recycling of oxide electrolytes from all-solid-state lithium-ion batteries by acid leaching

*Kirstin Schneider*

*Clausthal University of Technology | Department of Mineral and Waste Processing*

All-solid-state lithium-ion batteries (ASSBs) exhibit high specific energy density and improved safety features compared to commercial lithium-ion batteries due to the lack of flammable liquid electrolyte. Currently, researchers focus on material development and processing technology to overcome barriers that hinder the widespread industrial implementation of ASSBs. However, within the European circular economy framework, recycling concepts for ASSBs need to be developed before widespread application to ensure sustainable production. As part of the competence cluster for recycling and green batteries (greenBatt) of the German Ministry of Education and Research (BMBF), researchers investigate the recyclability of ASSBs with a holistic approach in the project "Development of All-Solid-State Battery Recycling Processes".

So far, only theoretical studies for the treatment of ASSBs in hydrometallurgical battery recycling processes were published. Besides the introduction of lithium metal anodes, the solid electrolyte presents a substantial difference to traditional lithium-ion batteries with liquid electrolytes. Oxide electrolytes are a promising candidate to be applied in ASSBs. To gain fundamental knowledge about their leaching behavior, we perform an acid leaching study with lithium lanthanum zirconium oxide (LLZO). Different inorganic and organic leaching agents at varying acid concentrations and temperatures at a fixed solid-liquid ratio are investigated. Process parameters are selected accordingly to lithium-ion battery leaching studies to conclude how LLZO will behave in battery recycling processes. Finally, the paper compares agitator leaching and ultrasound-assisted leaching.

This presentation is a result of the joint research between the Department of Mineral and Waste Processing of Clausthal University of Technology and the Institute for Process Metallurgy and Metal Recycling of RWTH Aachen University.

### ■ The sustainability of battery cell production in Europe

*Marlene Eisenträger*

*VDI/VDE Innovation + Technik GmbH*

The sustainability of battery cell production is one of the core requirements regarding the European efforts to establish value creation in the field of lithium-ion batteries in the EU. The concept of sustainability is multi-faceted and often the subject of political as well as social debates. Producing "green" batteries requires a holistic view on issues such as climate, raw material governance, circular economy, employment, technology leadership/competitiveness, costs as well as subsidies for battery cell manufacturing.

In a new study, we compiled and comparatively analyzed the current debate and verified scientific findings on the individual topics. We have also included the sustainability-related aspects of the proposal of the new EU Battery Ordinance. As to regulation, it is necessary to make sustainability measurable. We have collected corresponding approaches from politics and industry on how the identified sustainability aspects can be made plausibly measurable and verifiable by means of indicators.

Some key statements of the study:

The development of cell manufacturing capacities in Europe can further strengthen the decarbonization effect, as the CO<sub>2</sub> footprint of a European cell production will be smaller due to an energy supply in Europe, which currently is significantly lower in emissions as compared to China.

New digital concepts (like the battery passport) allow the seamless and verifiable documentation of material and information flows of individual battery products in the supply chain.

The reprocessing and reuse of used batteries improves the ecological footprint of batteries.

Due to the currently very high market dynamics and falling battery prices, the economic establishment of second-life business fields is (however) challenging.

Future-oriented and sustainable battery (cell) production within Europe compensates for the decline in employment in the (European) automotive industry, as a result of advances in process technology and the market.

## Module & Pack Production

### ■ Agile Production Lines - flexible scaling for high-speed production

*Alexander Weis*

*SCIO Technology GmbH*

Agile production lines are the future of module and pack production. The flexibility in meeting customer demand for a variety of module sizes and pack designs through high-speed production enables efficient, cost-effective scaling. Key points include:

- What are the key components of agile production?
- How does this apply to battery module and pack production?

- How can an automated batterie module / pack production line become agil? What changes / steps are necessary?
- How is module / pack variant management handled in agile production?
- How can the variant management be implemented in the agile high speed production line?

With the rapidly growing demand for electrification along all product and market segments including e-mobility, the need to rapidly meet customer module & pack design demands with the appropriate production volume increases exponentially. Only agile, high-speed production lines set up with variant management ability can meet these challenges.

### ■ Automatic Machines for Leak Testing of Sealed Batteries

*Michael Klenk*

*MARPOSS GmbH*

We are facing new challenges in leak testing as we strive to deliver high-performance, long-lasting, and safe lithium batteries.

From a closer analysis of the lithium battery, we learned that humidity needs to be avoided at any cost. If humidity penetrates a battery it can cause:

- Premature battery aging: the battery reduces its capacity.
- Longer battery recharge time to full capacity.
- Possibilities of battery fires and runaway.

The other potential problem is the hazard to the environment if the electrolyte is leaking from a battery. The salts dissolve in the electrolyte, then combine with water, and then create hydrogen fluoride (HF) which is considered Toxic at a very low concentration (1 ppm) or is considered corrosive at a higher concentration (above at 24 ppm).

To solve the above concerns, modern leak test equipment should meet a series of characteristics that will ensure state of art quality standards:

- High sensitivity to sort the good batteries form the leaking ones.
- Short cycle time in order to test up to one cell per second for the button cell batteries.
- Safety verification to avoid cross-contamination of the system in the case a battery cell with a gross leak arrives into the station.
- Automatic contamination detection.
- Automatic cleaning cycles to recover from contamination.
- Sorting devices in order to send the good batteries to the next process in the production line and to segregate the leaking batteries for further analysis.

Marposs has developed a system (Patent Pending) that can test the sealed batteries in mass production, to detect if the batteries are leaking. This system is conceived to work in the automatic lines of a production plant and we have solved all the industrial challenges and have successfully addressed all the pain points associated with leak testing lithium batteries.

### ■ Dynamic beam shaping and forming technics for improved results in battery welding applications

*Peter Kallage*

*Coherent*

Many tasks in the battery manufacturing process such as busbar welding both foil and thick materials, prismatic and cylindrical cell seam sealing, pouch cell foil-to-tab welding, and hairpin welding require minimizing thermal input, spatter, cracking and porosity.

Solid-state lasers with a visible wavelength have emerged as a possible alternative for welding of delicate materials such copper, aluminium or dissimilar material, but their practical limitations lead to a higher cost of ownership. The Coherent ARM Laser brings all the cost, reliability and practical advantages of fiber laser to challenging copper welding tasks. Tests have proven that the dual-beam of the ARM Laser, with a high brightness center spot, can efficiently weld copper without the process instabilities that fiber lasers have.

In addition the SmartWeld+ plays out its advantages in battery manufacturing, when welding dissimilar materials, improved blending of melt pool, reduced spatter, low viscosity and surface tension as well as the reduction of porosity and cracks can be achieved with this processing head utilizing the advantages of precise control of energy input per unit length over the weld pattern.

## Battery Production 4.0

### ■ Digital Twin in Battery Production 4.0 – From Data Management and Traceability System to Target-Oriented Application

*Alexander Kies and Jonathan Krauß*

*Fraunhofer Institute for Production Technology IPT*

The advancing climate change necessitates a shift from the use of conventional energy sources to renewable ones, which is accompanied by the indispensability of efficient storage of electrical energy. This is resulting in a boom in demand for battery cells- according to estimates, the market for lithium-ion battery cells will grow from around 200 GWh per year in 2020 to over 2,000 GWh by 2030. However, the production of battery cells involves enormous complexity for companies and entails various hurdles. Here, cell quality proves to be a central challenge. One possible answer to this problem lies in the digitalization and networking of production associated with the fourth industrial revolution, which holds great potential for improving quality in battery cell production 4.0. In this context, the collection and useful processing of data that already is generated in large quantities in the production environment is essential. However, the generated data is often heterogeneous and a unique assignment to the produced object is rarely possible, which complicates a target-oriented use of data. In this presentation, we propose the conceptualization of a digital twin in battery cell production, taking into account the different phases of its creation. Required data describing the battery cell is initially identified by using a methodology for integrating process knowledge from different areas of expertise. Data aggregation across the various production processes can be accomplished with the help of a suitable and tailor-made traceability system. Based on the aggregated data, the derivation of a digital twin of the battery cells is made possible. The use of the digital twin is in turn realized through algorithm-based modeling, on the basis of which conclusions can be derived about the real-world equivalent. This procedure is being evaluated in the context of a battery cell production environment.

### ■ Data-mining-based multi-criteria quality prediction in battery cell production

*Artem Turetskyy*

*TU Braunschweig | Institute of Machine Tools and Production Technology (IWF)*

Batteries play a major role in the transformation of the mobility (e.g. electric cars) and the energy (e.g. storage of renewable energy) sector. The production of the state of the art technology, the lithium-ion battery (LIB), suffers from high scrap rates and high energy demand leading to a high environmental impact and high costs. Its production chain is of a complex nature and consists of various highly specialized processes (batch/ continuous process and single-unit processes) with diverging and converging material flows and a plethora of interdependencies. Small process fluctuation can accumulate over the whole production chain to strong quality deviations or even insufficient quality and therefore scrap. Quality deviation or even a quality loss can directly affect the costs and the environmental impact of the whole production. Therefore, it is of great importance to fully understand the production and its influence on the battery cell in order to achieve lower production costs and lower environmental impacts.

This work presents a data acquisition and data management strategy implemented in the Battery LabFactory Braunschweig to support data mining applications in LIB cell production to uncover potential interrelations within the production chain. The data mining approach is applied to identify and evaluate the influences of intermediate product features (particle distribution, coating layer thickness, etc.) on the final product properties of the battery cell (maximal capacity, self-discharge, etc.). The work further shows its potential deployment to support quality management in LIB cell production.

## Flexible scaling of future battery cells

Jan Diekmann  
CUSTOMCELLS

As the use of lithium-ion batteries expands, so do the requirements for new battery cells. In the automotive sector, for example, the focus is not only on gravimetric energy density, but also on high charge and discharge currents. The production of large automotive cells with future lithium-ion chemistries will be largely determined by the process technology used in addition to the ambient conditions. To meet these requirements, cell architecture and in particular battery cell components are being developed further and further. The focus here is on the electrodes as the components that determine the key performance of the cells. Nickel-rich materials are used on the cathode side, while Si compounds are often planned for the anode in conjunction with pre-lithiation. The processing of these materials results in special demands on the production environment due to their high water sensitivity. At the same time, large cell formats and high currents pose challenges for the cell design. This presentation will present a flexible process chain for the processing of all major lithium-ion battery materials as well as new material developments. The talk will describe an appropriate environment from slurry production to cell finishing. The implementation for scaling will be explained in more detail using specific examples.

## Let's put smart to work - Future Battery Factories adapting Industry 4.0 Technologies

Klaus Eberhardt  
Exyte

There is a strong Trend towards automation and data exchange in Manufacturing Technologies including IoT, cloud computing, cognitive computing and artificial intelligence. Such Technologies should be already implemented during the initial Concept development of future Battery GigaFactories.

This presentation gives an overview on current and future Industry 4.0 Technologies, and their implementation during the initial Concept development, followed by the Design, Pre-Construction, Construction until the Operation of the Battery Factory.

Advanced design of high-tech factories starts today with a 5D BIM model. This includes not only the 3D design, but adds information on the time schedule and the costs as the 4th and 5th dimension. Since the BIM architecture contains as-built documentation of the building & facility systems, it can be used during the entire fab's life cycle. In combination with a MES (Manufacturing execution System) model, this allows advanced process control, virtual site walks, life cycle analysis as well as financial modelling.

The benefits of adapting IOT (Internet of Things) during Battery Production such as faster time to market, potential cost reductions and faster expansion possibilities will be discussed and reviewed

## Modelling of Lithium-ion Battery Electrode Calendering by Discrete Element Method (DEM)

Ruihuan Ge, Denis Cumming, Rachel Smith  
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The calendering process is an important step in the manufacturing of lithium ion battery electrodes. The microstructure and end-product performance is strongly affected by this process. In this study, we investigate the evolution of electrode structure during calendering using Discrete Element Method (DEM). Simulations for the calendering of a cathode structure are performed using DEM to achieve different calendering levels. These simulated structures are then compared with the microstructure of corresponding experimentally calendered electrodes, imaged using micro-CT. An adhesive elastic-plastic model and a bond model is used to describe the mechanical response of active material particles and the inter-particle conductive additive/binder phase. The simulation results show good agreement with corresponding experiment measurement, and demonstrate the potential of DEM to inform electrode manufacturing process design.

Key words: Discrete Element Method (DEM); Lithium-ion battery; Calendering; Electrode manufacturing

## Towards a comprehensive semantic information structure in the battery value chain

Simon Stier, Simon Clark, Henning Lorrmann  
Fraunhofer Institute for Silicate Research ISC

The battery value chain stands out for its high volume throughput combined with high complexity and highest quality requirements. Due to the strong interaction between material properties, manufacturing processes and the final product performance a deep integration of materials research into the process development (and a high degree of the control thereof) is required. Furthermore, theoretical modeling is essential for understanding the electrochemical system as a whole, which is subject to continuous change over the course of its lifetime. In order to integrate batteries as an essential part of a sustainable industrial future ecological and ethical constraints need to be fulfilled.

The battery value chain is therefore a distinctly interdisciplinary field in which numerous interfaces between fields of work, ways of thinking and methods must be created in order to gain a comprehensive picture of the large-scale effects of small-scale changes. Here, a structured description of both knowledge and data is the key to a barrier-free flow of information, regardless of domain-specific characteristics. For this purpose, ontologies offer the foundation to create comprehensibility based on formalization of natural language concepts—equally for humans, algorithms and machines.

Taking up the preliminary work of the last decades in the field of Semantic Web and merging it with the manifold disciplines of the battery value chain is already the subject of numerous projects. In this talk we will present several examples including production processes (KIproBatt, BMBF), advanced battery materials (BIG-MAP under the BATTERY 2030+ umbrella, EU/H2020) and live cycle assessment (eLi+, EIT Raw Materials). In addition to the underlying theoretical concepts, we will discuss their contribution to the gradual realization of a value chain spanning information structure.

## Production of solid state batteries

### Novel processing technique for making solid-state batteries with fast lithium ion transport kinetics

Chun Huang  
King's College London

Solid-state lithium metal batteries (SSLMBs) combine improved safety and high specific energy that can surpass current lithium ion batteries (LIBs). However, the lithium ion diffusivity in the composite cathode of SSLMBs is approximately an order of magnitude lower than that of the solid-state electrolyte (SSE) membrane because of the highly tortuous ion transport pathways in the cathode. This lowers the realizable capacity and mandates relatively thin (30-300  $\mu\text{m}$ ) cathodes for SSLMBs, and hence low overall energy storage. Here, a thick (600  $\mu\text{m}$ ) composite cathode comprising vertically aligned  $\text{LiNi}_0.8\text{Mn}_0.1\text{Co}_0.1\text{O}_2$  (NMC811)-rich channels filled with a polymer electrolyte is fabricated by an innovative directional freezing and polymerization (DFP) method. X-ray micro-computed tomography, ion mobility simulations and DC depolarization show that this cathode structure made by the DFP technique improved lithium ion diffusivity in the cathode from  $4.4 \times 10^{-9}$  to  $1.4 \times 10^{-7}$   $\text{cm}^2/\text{s}$ . In a SSLMB full cell, the cathode provided gravimetric capacities of 199 and 120  $\text{mAh/g}$  at 0.05 and 1 C respectively at 25  $^\circ\text{C}$ , corresponding to ultra-high areal capacities of 16.7 and 10.1  $\text{mAh/cm}^2$ . This work demonstrates a scalable approach to realizing composite cathode structures with kinetically favorable ion transport characteristics in SSLMBs.

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## ■ Manufacturing of Composite Cathodes for Sulfide-based All-solid-state Batteries

*Hans-Christoph Töpper*

*Technical University of Munich*

The promise of high energy densities of all-solid-state batteries is fulfilled only by manufacturing composite cathodes with very low to none porosity. This talk addresses the resulting challenge for the production technology. Experimental results in the manufacturing of sulfidic all-solid-state separators and composite cathodes are shown and discussed. The discrepancy between theoretical concepts and calculations is compared to the harsh reality of production.

The experiments are composed of mixing, tape casting, and compressing different sulfidic all-solid-state separators and composite cathodes on a lab scale in a microenvironment. The successful process parameters are quantified and the resulting microstructure is analyzed regarding particle contacts and porosity by scanning electron microscope and mercury intrusion porosimetry.

As a result, parameters of compaction and material composition differently affect the minimally achieved porosity. The mechanical load results in a significant decrease in porosity even at low pressures. SEM images show that this is in particular due to the reorientation of particles and the deformation of the solid electrolyte. On the material side, the composition of the cathodes strongly influences the resulting porosity. The higher the share of solid electrolyte, the lower the cathodes' porosity. However, if this is related to the energy density on a cell level, a certain share of solid electrolyte in the composite cathode is not to exceed. Therefore, the cell and electrode design is a critical parameter to reconsider. Further boundary conditions are cracking of the active material particles and material costs. Overall, the optimum composition of a composite cathode is calculated from these aspects.

In conclusion, the research results deepen the understanding of the manufacturing of sulfide-based composite cathodes and show an iterative approach of processing, analyzing, and cell design.

## ■ Challenges and opportunities for solid-state players – can they be competitive on the battery market within automotive applications?

*Joscha Schnell*

*P3 automotive GmbH*

Increasing battery demand and requirements towards high performance cells are pushing lithium-ion technology to its limits. Recent developments in solid-state technology have led to a high level of media attention, and both start-ups as well as large cell manufacturers are intensively working on the industrialization of their next-generation technology as major challenge. In this presentation, the competitiveness of currently leading players regarding technology, scalability and costs aspects will be evaluated and discussed. P3 will introduce the audience to the latest news and current development status of solid-state technology and provide an outlook on further potential within the automotive market by comparing the performance of the next-gen technologies from leading solid-state companies.

## ■ Advanced Battery Material: Challenges within the laser cutting process of lithium metal

*Lars Schmidt*

*TU Braunschweig | Institute of Joining and Welding (ifs)*

Through a largely establishing of electric vehicles in the market, an increase of energy density is needed. All-solid-state batteries are promised to be the next generation battery systems. While on the cathode side versatile materials and mixture recipes were examined, the anode side commonly consist of lithium metal. The processing of this material is an ambitious challenge caused by both the strong adhesion behavior to other materials and the highly reactive character of lithium as an alkali metal. The commonly used separation process of lithium metal foils for battery application is a punching separation, which caused an abrasion of the cutting edge on the cutting die. A contactless separation process like laser cutting could solve this problem.

For the laser cutting process, the use of a suitable or conditioned process atmosphere is necessary. The surface of the alkali metal reacts with both, a highly water content in the atmosphere or with the gas components like oxygen and nitrogen in the air. Due to the resulting new lithium surface chemistries, a changed laser-material interaction occurs in contrast to commonly electrode materials. In order to evaluate which process atmosphere leads to the laser-material interaction with high quality cutting edges, in terms of melt formation, heat affected zone and kerf width, different process atmospheres (argon, nitrogen, synthetic air and a drying room atmosphere) were investigated. Furthermore, the electrochemical performance of the prepared lithium anodes were investigated in terms of the effect of the process atmosphere on the built battery cell.

## ■ Laser sintering of ceramic-based solid-state battery materials

*Linda Hoff*

*Fraunhofer Institute for Laser Technology ILT*

Ceramic solid-state batteries can increase both gravimetric energy density and safety compared to conventional lithium-ion batteries. Ceramic materials of high ion conductivity are lithium lanthanum zirconate (LLZ) as a solid electrolyte and lithium cobalt oxide (LCO) as a cathode active material. The materials are applied to a metallic carrier foil by means of screen printing and then thermally post treated (dried and sintered) to produce adhesive layers or layer systems with the highest possible density. The conventional heat treatment is done in an oven process. Disadvantages of oven processes are the possible diffusion of the coating materials into adjacent layers and lithium loss due to long process times (in the range of minutes) at high temperatures and the high energy consumption.

For the construction of a battery cell, the preservation of the crystal structure and thus a suitable temperature management during the drying and sintering process are of enormous importance. By means of laser processing, short interaction times within the range of seconds and below are realised. High heating and cooling rates show potential for reducing diffusion processes and preserving the crystal structure of the materials.

In this work, the laser sintering of ceramic micro particle cathode and electrolyte layers as parts of thin film ceramic solid-state batteries are presented. Laser sintering offers a selective heating process which is especially important for the electrolyte because the sinter temperature of the electrolyte is higher than for the cathode. Challenges of reaching a rather homogeneous temperature profile across the layer thickness within short processing times while preserving the materials integrity and bonding the layer to the substrate are addressed. The influence of different interaction times, scanning strategies as well as use of sintering additives on the crystal structure, the electrochemical properties and adhesion are investigated.

## ■ Evaluation of formulation variation for scalable processing of sulfide-based all-solid-state batteries

*Mattis Batzer*

*TU Braunschweig | Institute for Particle Technology (iPAT)*

Sulfide based solid state batteries are predicted to have several advantages compared to conventional lithium ion batteries, e.g. in the aspects of safety, rate performance and energy density. These advantages are faced with challenges regarding large scale processing. The focus of this publication is, therefore, on a scalable solvent-based process route, using a dissolver for the first time, to produce sulfide solid-state electrodes and separators. The formulation of the slurries was systematically varied (for example type of solid electrolyte and conductive additive content) to investigate the influence of the components on the mechanical and electrochemical properties of the coated sheets. From the investigated compositions, the best were chosen for full cell assembly with indium at the anode side and cycling experiments were performed. These revealed an active material utilization up to 87% for the first discharge and a coulombic efficiency of ~92% after 10 cycles.

## ProZell – competence cluster for battery cell production

### ■ Reconstruction of the carbon-binder domain in Li-ion battery cathodes and its influence on the electrochemical performance

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It is well known that the 3D microstructure of electrodes in lithium-ion batteries has a strong impact on the electrochemical performance. In particular, the spatial distribution of the carbon-binder domain (CBD) offers a large potential to further optimize lithium-ion batteries. However, it is challenging to reconstruct the CBD from tomographic image data obtained by synchrotron tomography. In the present talk, we consider different approaches to segment 3D image data of two cathodes into three phases, namely active material, CBD and pores. More precisely, we focus on global thresholding, a local closing approach based on EDX data, a k-means clustering method and a procedure based on a neural network that has been trained by correlative microscopy, i.e., based on data gained by synchrotron tomography and FIB-SEM data representing the same electrode. Based on the different segmentations, we quantify their impact on the resulting performance by spatially-resolved transport simulations and compare the results to experimentally determined electrochemical properties. In addition, we compare both samples, namely an ultra-thick unstructured cathode and a two-layer cathode with varying CBD content in both layers.

### ■ Multilayer silicon/graphite anodes for the use in high-energy cells

Laura Gottschalk

TU Braunschweig | Institute for Particle Technology (iPAT)

Lithium-ion battery cells with high specific capacity and energy density are one of the current research priorities in industry and science. The challenges of these "thick" electrodes are transport limitations within the electrode: lithium ions cannot reach the deeper layers of the electrode coating, which leads to a drop in performance during charging and discharging. This phenomenon is a major hurdle in the further development of future high-capacity electrodes.

One possible solution is to use multilayer electrodes with different layer properties. In this way, pathways for the Li-ions can be created that reduce the ionic resistance of the electrode and ensure an increase in fast charging capability.

The focus of this work is therefore on two-layer anodes. An active material mixture of graphite, hard carbon and silicon is used ( $\approx 575$  mAh g<sup>-1</sup>). The initial focus is on the processing of the different configurations, whereby the mechanical (adhesive strength), electrical (electrical resistance) and structural (porosity measurements, SEM images) layer properties are investigated. Promising multilayer anodes are tested for their electrochemical performance in full cells ( $\approx 8$  mAh cm<sup>2</sup>). In particular, a comprehensive evaluation of the charging performance is carried out. The addition of the active material hard carbon in the upper layer resulted in a significant increase in electrical conductivity. This can be attributed to the higher electrical conductivity of the carbon-based hard carbon. With regard to the electrochemical characterization, it was observed that the two-layer anode with the active material mixture of graphite, silicon and hard carbon in the upper layer showed a significant increase in discharge capacity, especially for low C-rates. One assumption is that the addition of the hard carbon could create an increased number of pores for the lithium ions, leading to improved ion transport and the reduction of transport limitations.

### ■ About the vacuum post-drying process in the production of lithium-ion batteries

Thilo Heckmann

Karlsruhe Institute of Technology (KIT) | Thin Film Technology (TFT)

For competitive battery cells, an important prerequisite is energy savings in the cell production. An essential cost factor in the manufacturing process of Li-ion batteries poses a subsequent post-drying process. In this process, moisture adsorbed from the air gets removed from the compounds of the battery by a high energy input. Otherwise, the adsorbed moisture reacts with the electrolyte in the operation of the battery, leading to its degeneration. Literature has revealed a connection between electrochemical performance and the remaining moisture content. In order to remove the moisture cost efficiently from the compounds of the battery, this process step needs to be understood completely. Therefore, a precise and comprehensive simulation along with knowledge of the sorption equilibrium of water and the kinetics of mass transfer reveals optimization potential of energy consumption and process acceleration.

This work investigates the water mass transport in porous electrodes during the vacuum post-drying experimentally and theoretically. The time-resolved vacuum-drying curves are measured with a modified set up of a magnetic suspension balance at variable temperatures and pressures. Furthermore, as physicochemical basis for transport phenomena, sorption equilibria of the compounds of a Li-ion battery have been ascertained as well by means of a magnetic suspension balance. This measurement data validate the simulation of the vacuum drying process, which allows an easy scale-up and transfer into industrial applications.

This work contributes to the research performed at CELEST (Center for Electrochemical Energy Storage Ulm Karlsruhe) and Material Research Center for Energy Systems (MZE). The authors would like to acknowledge financial support of the Federal ministry of Education and Research (BMBF) via the ProZell cluster-project "Epic" (Grant number: 03XP0295A).

## Cell formation and Quality Control

### ■ High Potential Test for Quality Control of Separator Defects in Battery Cell Production

Ferry Kienberger

Keysight Labs Austria

Lithium-ion batteries are a key technology for electromobility, thus quality control in cell production is a central aspect for the success and competitiveness of electric vehicles. The detection of defects and poor insulation behavior of the separator are essential for high-quality batteries. Optical quality control methods are usually applied in cell construction and assembly processes, yet they are unable to detect smaller but still relevant defects in the separator layer, e.g. pinholes or particle contaminations. This gap can be closed by executing high potential testing to analyze the isolation performance of the electrically insulating separator layer in a pouch cell format using graphite anode and NMC cathode. Here we present an experimental parameter study to identify different separator defects on dry strained cell stacks based on electric voltage stress and mechanical pressure. In addition, finite element modeling (FEM) is used to generate physical insights into the partial discharge. The FEM examines the defect structures and the corresponding electric fields including topographical electrode roughness, impurity particles and air voids in the separator. Overall, the test results show that hard discharges are associated with significant separator failures which were also imaged with electron microscopy. Based on the parameter study, a voltage of 350 to 450 V and a pressure of 0.3 to 0.6 N/mm<sup>2</sup> is identified as an optimum range for the test methodology, resulting in failure detection rates of up to 85%.

## ■ Optimized cell finishing through innovative plant technology and parameter-based evaluation of process and quality parameters in the virtual production system

*Nikolaus Lackner*

*RWTH Aachen University | TU Munich*

In terms of energy and resource savings, the production of lithium-ion batteries (LIBs) shows a high potential for improvement in comparison to other industrial sectors. High reject rates of 5-12% increase the overall production costs and decrease throughput. As LIB costs are mainly driven by material costs (70-80% of total costs), production rejects have a massive impact on overall costs. This is particularly relevant in the final process steps, referred to as cell finishing. Before the aging process, ~95% of the value-adding has already been achieved, but it will take days to weeks before the LIB passes the End-of-Line (EoL) test. At the same time state of the art cell finishing does not rely on the in-line measurement and the processing of cell and process quality parameters. In addition, there is a lack of predictive knowledge about the outcome of the quality of produced cell, which causes cell manufacturers to keep the cells in storage for the extended timespan, test and grade them before delivery.

As these problems can be addressed by equipping cell finalisation facilities with in-line measurement like online electrochemical impedance spectroscopy and analysing the different parameters the Project OptiPro was initiated by the TU Munich and the RWTH Aachen. The Project aims to improve throughput and quality as well as decrease waste and scrap in cell production. To achieve these goals both universities will integrate in-line measurement into their cell finalisation equipment on a lab scale and connect it to an IoT-Platform. In a first step the connection between the various process and quality parameters will be analysed followed by integrating this knowledge into a virtual production system that is based on machine learning algorithms. Through collection and analysis of the measured data it is possible to predict the quality of the produced LIBs at an earlier stage of the process which allows to decrease aging time per battery and reduce extensive EoL testing.

## ■ BioLogic – Our Solutions to Test Your Batteries

*Sandra Möller and Julia Berlin*

*Bio-Logic Science Instruments GmbH*

Bio-Logic Science Instruments SAS, with its corporate office in Seyssinet-Pariset, is a French designer and manufacturer who offers powerful laboratory research instruments and software since 1983. The company is characterized by its close relationships with research and testing laboratories around the world. With our high precision, high-performance product range of potentiostats, galvanostats, battery cyclers and impedance analyzers we are able to cover a wide field of research: New battery technologies, fuel cells, photovoltaics, corrosion, super capacitors, and bio-sensors to name a few. The modularity of our multi-channel potentiostats and our wide range of battery characterization accessories offers highest flexibility to configure the appropriate device according to your needs.

As one of three area sales managers here in Germany, I look forward to introducing you to Biologic. I will give you an insight into our company structure and philosophy. In the further course I will introduce our product range with deeper focus on our battery cyclers for electrochemical measurements and your battery studies. After this talk you will see that Biologic has the ideal setup for testing almost any single cell battery on the market.

## Multivariable process - property relationships

### ■ Study of the cathode coating-drying manufacturing process by design of experiments

*Geanina Apachitei*

*University of Warwick*

The understanding of traditional battery manufacturing operating conditions on electrode structure and final electrochemical performance is currently limited. In this study, the design of experiments methodology (DoE) is applied for the identification of the main operating variables (factors) of the coating-drying process in the manufacturing of cathodes at the pilot-scale. The experimental design is a saturated design that considers only two settings (levels) for each of the factors. The factors studied are: comma bar gap (80 and 140  $\mu\text{m}$ ), coating ratio (110 and 150%), web speed (0.5 and 1.5 m/min), drying temperature of the first oven zone (85 and 110  $^{\circ}\text{C}$ ) and drying air speed (5 and 15 m/s). The output variables (responses) include pre-calendered and calendered physical electrode properties (mass loading, thickness and porosity) as well as gravimetric and volumetric energy capacities. Analysis of variance and multiple linear regression determines the relationship between factors and responses and their statistical significance with a confidence level of 90% (p-value =0.1). Empirical models for the responses are obtained in terms of the statistically significant factors. Results show that comma bar gap and coating ratio are critical parameters since have a direct impact on battery electrochemical performance. The drying temperature is not statistically significant at the conditions studied and therefore is a non-critical parameter. The correlations show a good agreement between the experimental data and the models, resulting in correlation coefficients ( $R^2$ ) as high as 0.99 in some cases. The work demonstrates the applicability of DoE to the manufacturing process of li-ion batteries at a pilot-scale for the identification of important operating variables and their effect on battery performance. The obtained models are useful in the determination of operating parameters settings to achieve a robust manufacturing process, therefore reducing time and cost.

### ■ The round-robin test: Towards process standardization of EU research pilot lines

*Katja Froehlich*

*AIT Austrian Institute of Technology GmbH*

International competitiveness in battery cell production from a European perspective can score mainly in two different factors: sustainability and quality. The latter can be achieved through well-defined production standards and even more, standardization of the individual production processes. As part of the LiPLANET network activities, five attending European research pilot lines have come together to sound out this future standardization by means of a round-robin test.

For the initialisation of the round-robin test and setting up the protocol, following questions were raised:

Which process parameters are considered critical?

Which tolerances are acceptable?

How does the final prototype cell format influence those?

And last but not least another perspective:

How can we share and use the generated data on a non-competitive basis?

The focus of the round-robin test is set on widespread cell chemistries to cover as many pilot lines as possible and gain knowledge on the influence of the production processes on different cell properties on a non-competitive basis.

Within the LiPLANET network, the five attending research pilot lines have been intensively discussing, defining and revising the process parameters and, according to their experiences, accepted tolerances of critical production steps. Materials and compositions have been chosen and the test protocol was defined accordingly. For this initial run, the consortium agreed to apply NMC622 cathode and artificial graphite anode active material and defined to produce prototype cells consisting of 10 cathodes, resulting in different final cell capacities depending on the individual research pilot line facilities. Round-robin test results will be compared and shared within the network and cell manufacturing community.



## ■ Understanding and correcting for temperature variations when measuring self-discharge in cell production

*Manuel Moertelmaier*

*Keysight Technologies*

Measuring lithium-ion cell self-discharge during cell manufacturing is an important quality gate. To date, there have been 2 methods used. (1) You can use the delta-OCV method to measure the change in cell OCV during a long multi-day aging step or (2) you can directly measure self discharge using a self-discharge analyzer, which eliminates the needs for a long aging step. Direct measurement of cell self-discharge is done via a potentiostatic measurement process. While this potentiostatic method is innovative and fast, the practical application of this technique is challenged by the sensitivity of the cell's OCV to temperature.

In this presentation, we will

- Describe how to measure cell self-discharge
- Highlight the potential errors in delta-OCV caused by changing temperature, as the delta-OCV method is not immune to the impact of temperature variation.
- Overview the challenges of the potentiostatic method
- Share our research on modeling the cell's reaction temperature changes while on the manufacturing line
- Describe a post-processed algorithm for prediction of self-discharge that can eliminate the impact of temperature on the measurement.

By employing this technique, practical self-discharge measurements can be made in production in the face of temperature variations that are found on a typical cell manufacturing line. This allows direct measurement of self-discharge as a quality gate suitable for industrial scale cell production.

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